Study on Ultrafiltration of Hospital Wastewater Treatment Effluent

M Mahirullah \(^1\) and I N Widiasa \(^1\)

\(^1\) Department of Chemical Engineering, Faculty of Engineering, Diponegoro University, Semarang, Central Java, 50275, Indonesia.

Email: mahirullah@students.undip.ac.id

Abstract. The Ultrafiltration (UF) membrane is beneficial in the wastewater treatment plant (WWTP), including the hospital WWTP. The formation of fouling deposits is often encountered on the membrane surface. Pretreatment and cleaning strategy are needed to maximize flux recovery for the fouled membrane. Therefore, the investigation were conducted for the membrane UF in terms of performance and the cleaning of the UF membrane in terms of cleaning efficiency. Pretreatment of sedimentation and cloth media filtration (CMF) is recommended because the flux reduction rate of 3.3\% - 8.2\%. The cleaning procedure with NaOCl was carried out under various concentrations with pH 12.6. The cleaning duration was determined as 4 h. The best result of membrane cleaning for the experiment with pretreatment required 100 ppm NaOCl to achieve 96.43\% flux recovery, 77.77\% fouling removal and hydraulic cleanliness criterion 0.037. Meanwhile, the experiment without pretreatment required 500 ppm NaOCl to achieve 80.59\% flux recovery, 51.85\% fouling removal and hydraulic cleanliness criterion 0.24.

1. Introduction

The hospital wastewater treatment plant (WWTP) effluent contains many emission of chemicals such as pharmaceuticals, iodinated contrast media (ICMs), estrogen and other chemicals [16]. The hospital WWTP commonly uses conventional ways to reduce its hazardousness into the environment. It can be grouped as follows: (1) Physicochemical processing through coagulation-flocculation with Powdered Activated Carbon (PAC) or Granular Activated Carbon (GAC), (2) Biological processing with Conventional Activated Sludge (CAS) and Membrane Biological Reactor (MBR) processes, (3) Membrane technology with Reverse Osmosis (RO), Nanofiltration (NF) and Ultrafiltration (UF), (4) Disinfection by Ozonation, Advanced Oxidation Processes (AOP), chlorination, etc [16, 18].

The effluent from the hospital WWTP is mostly discharged into the environment. However, The need to reuse it will be the next problem in the future. The technology recommended for the treatment is Ultrafiltration. The UF membrane is the most widely used membrane technology today. It has many benefits because of its use in wide applications (ultrapure water production, applications in the food industry, biotechnology and liquid waste in the chemical industry) [6].

According to research by [1], UF successfully handled one type of HWWs, namely laundry wastewater. The research found that UF was able to provide more than 87\% rejection results for parameters such as COD, TSS, and surfactant concentration.
However, the major obstacle for the widespread implementation of the membrane process is still membrane fouling [5] and it is also a disadvantage of the UF membrane. Membrane fouling needs to be mitigated to maintain an acceptable flux over a long-term operation [8]. Pretreatment and membrane cleaning are recognized as good ways to mitigate membrane fouling. The treatment commonly used as pretreatment is sedimentation. The sedimentation process application was very effective as the pretreatment of the UF membrane due to extends the life of the membrane from the high molecular weight humic substances [14]. The new solution as pretreatment in wastewater treatment is cloth media filtration (CMF). The CMF technology provides a high-quality effluent, easy operation and major operating saving in reduced energy consumption in the treatment facility [3]. Meanwhile, the membrane cleaning process can be conducted using physical and chemical methods. Chemical cleaning is generally applied to hydraulically irreversible foulants and chemical agents are involved in the process. One of the common chemical agents used as the strong cleaner is sodium hypochlorite (NaOCl) [17]. The previous study investigated the effects of hydraulic and chemical cleaning on the fouled membrane by algae-rich reservoir water. The hydraulic cleaning strategy of backwashing followed by forward flushing was recommended. It also showed the combined use of NaOH (0.02 N) and NaClO (100 mg/L) was the better flux recovery strategy for the UF membrane [7].

Considering the facts mentioned, the aims of this study to investigate the effects of pretreatment sedimentation and CMF on the UF membrane fouled by the hospital WWTP effluent. It also focuses to investigate the effects of NaOCl on the UF membrane fouled by the effluent of the hospital WWTP.

2. Materials and methods

2.1. Characteristics of effluent

The feed water used in this study was the WWTP effluent of Diponegoro National Hospital, the water quality is presented in table 1.

| Parameter          | Range     | Standard I         | Standard II        | Standard III       |
|--------------------|-----------|--------------------|--------------------|--------------------|
| pH                 | 7-7.7     | 5.5-9<sup>a</sup>  | 6-9<sup>b</sup>     | 6.5 - 8.5<sup>g</sup> |
| TDS (mg/L)         | 375-393   | 1000<sup>a</sup>   | 1000<sup>d</sup>    | 1000<sup>h</sup>   |
| Color (TCU)        | 32-50     | 20-150<sup>a</sup> | 50<sup>d</sup>      | 15<sup>e</sup>     |
| COD (mg/L)         | 19-48     | 40<sup>b</sup>     | 0.2-4<sup>a</sup>, 0.1<sup>b</sup> | 6<sup>e</sup> |
| Ammonia (mg/L)     | 0.46-0.53 | 0.2-4<sup>a</sup>, 0.1<sup>b</sup> | 0.2<sup>e</sup>      |                  |

*<sup>a</sup> EPA Standards for discharge into the environment<br>*<sup>b</sup> MENLH-RI Standards for water hygiene sanitation<br>*<sup>c</sup> No standard for this parameter.<br>*<sup>d</sup> MENKES-RI Standards for drinking water<br>*<sup>e</sup> WHO Standards for drinking water

[13]. Meanwhile, standard for water hygiene sanitation according to Ministry of Health of the Republic of Indonesia [12] and standard for drinking water according to World Health Organization (WHO) [19].
2.2. Membrane characteristics and chemical agents
The membrane was a hollow-fiber UF membrane made of polysulfone (PS) (GDP Filter, Indonesia). The molecular weight cut-off of the membrane was 50 kDa. The effective membrane area was 0.5 m². A schematic of the UF membrane filtration set-up is shown in figure 1.

In all experiments, NaOCl has been used as the cleaning agent and NaOH has been used as the pH regulator. The specifications of both chemicals are technical grade.

2.3. Experimental procedure
In this study, the performance membrane and cleaning efficiency were investigated through an experimental procedure as shown in figure 2.

![Figure 1. Schematic of the UF membrane filtration set-up.](image)

The feed water was treated with two different treatments, with pretreatment and without pretreatment. Pretreatments were sedimentation and filtration with CMF. The transmembrane pressure (TMP) across the membrane was adjusted using a valve installed at the inlet and the retentate outlet. The TMP was set to 0.7 bar taking into account the pressure drop. The processing of feed water was held for 4 h. After fouling occurred, the UF membrane was rinsed with hydraulic flushing. Two kinds of combined hydraulic flushing were used, backwashing and forward flushing. The permeate of UF was used for backwashing and the clean water was used for forward flushing. The duration of each rinse was 5 min and the TMP used was 1 bar. Hydraulic cleaning has done for 20 min.
The UF membrane was further cleaned with chemical cleaning. NaOCl was used as the cleaning agent. In this chemical cleaning, cleaning in place (CIP) was chosen as a cleaning method for rotating NaOCl solution continuously in the membrane. The concentrations of chemical cleaning experiments were investigated as 100 ppm, 500 ppm and 800 ppm. The pH, cleaning duration and TMP during cleaning were 12.6, 4 h and 1 bar respectively.

![Figure 2. Experimental procedure of the UF membrane to process of feed water.](image)

2.4. Methods of analysis

2.4.1. Chemical oxygen demand (COD)
Measurements were made with the Multiparameter Bench Photometer HI 83200 according to the instruction manual for the instrument. Tests used COD reagents HI 93754A-25 LR for COD concentrations of 0 to 150 mg/ L (ppm).

2.4.2. Ammonia
Measurements were made with the Multiparameter Bench Photometer HI 83200 according to the instruction manual for the instrument. Tests used ammonia reagents HI 93700A-0 and HI 93700B-0 LR for ammonia concentrations of 0.00 to 10.00 mg / L.

2.4.3. Total dissolved solids (TDS)
Measurements were made with the HI 99300 EC / TDS meter according to the instruction manual for the instrument.

2.4.4 pH
Measurements were made with pHep® pH Tester HI 98107 according to SNI 06-6989.11-2004 and the instruction manual for the instrument.

2.4.5. Color
Measurements were made with the Multiparameter Bench Photometer HI 83200 according to the instruction manual for the instrument.

2.5. Cleaning efficiency
This can be explained by Darcy's law by producing equation (1) and (2).

\[
J = \frac{dV}{(A \cdot dt)} = \frac{\Delta P}{\mu \sum R_t}
\]  

(1)

where J is the permeate volumetric flux, V is the accumulation of permeate volume, A is the membrane surface area, \(\Delta P\) is the pressure drop applied to the impurities and membrane layers, \(\mu\) is the permeate viscosity.
The equation for calculating the total resistance ($R_t$) is given by:

$$R_t = R_m + R_{rev} + R_{irre}$$  \hspace{1cm} (2)

where $R_m$ is the membrane resistance of the initial water flux ($J_{wo}$) and calculated according to equation (3),

$$R_m = \frac{\Delta P}{\mu J_{wo}}$$  \hspace{1cm} (3)

$R_t$ is resistance after fouling from water flux after fouling ($J_{wf}$) and calculated according to equation (4),

$$R_t = \frac{\Delta P}{\mu J_{wf}}$$  \hspace{1cm} (4)

$R_{irre}$ is resistance after rinsing water with water flux after rinsing water ($J_{ww}$) in equation (5) and calculated according to equation (2) [8, 10].

$$R_{irre} = (\frac{\Delta P}{\mu J_{ww}}) - R_m$$  \hspace{1cm} (5)

Cleaning efficiency is determined by restoring permeability to the membrane after cleaning using three cleaning parameters, namely: 1) Flux Recovery (FR) percent calculated in equation (6); Removal Fouling (RF) percent in equation (7); and Hydraulic Cleanliness Criterion (HCC) in equation (9) [2, 10].

$$FR(\%) = \left(\frac{R_m}{R_c}\right) \times 100\%$$  \hspace{1cm} (6)

$$RF(\%) = \left[\frac{(R_f - (R_c - R_m))}{R_f}\right] \times 100$$  \hspace{1cm} (7)

where $R_c$ is the resistance after cleaning of the water flux after cleaning ($J_{wc}$) and calculated according to equation (8).

$$R_c = \frac{\Delta P}{\mu J_{wc}}$$  \hspace{1cm} (8)

The HCC parameter represents the proportion of residual fouling resistance that remains after cleaning. Cleaning is considered efficient from a hydraulic point of view when HCC < 0.05 and is defined as:

$$HCC = \frac{R_{res}}{R_m}$$  \hspace{1cm} (9)

where $R_{res}$ refers to the membrane resistance remaining after cleaning compared to $R_m$ and . This is defined by equation (10).

$$R_{res} = R_c - R_m$$  \hspace{1cm} (10)

3. Results and discussion

3.1. Permeate fluxes

The fouling process has been carried out for four different treatments to the UF membrane fouled by the hospital WWTP effluent. Figure 3 show that the flux decline occurred to four experiments during 4 h. It is due to the accumulation of concentration polarization at the UF membrane surface which can reduce the permeate flux [11].
As shown in figure 3, the experiment without pretreatment shows the flux decline rate by 28.9%. The figure 3 also shows the experiment with CMF alone, which presents the flux decline rate by 12.4%. Then, the experiment with sedimentation 7 h and CMF shows the flux decline rate by 3.3%. It is better than the experiments with sedimentation 15 h and CMF to the Hospital WWTP effluent, which gives the flux decline rate by 8.2%.

Therefore, the smallest flux decline rate is the experiment with sedimentation 7 h and CMF. This does not mean that the optimal time for sedimentation is 7 h. It needs further research for a longer time than 15 h to prove it. However, all of these indicate that the pretreatment with sedimentation and CMF is efficient to controlling the flux decline during the process. This is due to the flux decline rate of 3.3% - 8.2% exclusively.

3.2. Removal of color, COD, and ammonia

The removal of feed water from the hospital WWTP effluent by the UF membrane experiments is shown in table 2, which presents color, COD, and ammonia reduction results.

| Treatment                     | Color (%) | COD (%) | Ammonia (%) |
|-------------------------------|-----------|---------|-------------|
| Without pretreatment         | 10        | 13.79   | 2.17        |
| CMF                           | 12.5      | 55.88   | 3.77        |
| Sedimentation 7 h + CMF      | 14        | 57.89   | 5.66        |
| Sedimentation 15 h + CMF     | 38.77     | 62.5    | 6.52        |

The color removal in table 2, which shows that the UF membrane could remove color by 10%. Then, CMF only adds removal by 2.5% higher than the experiment without pretreatment. The presence of sedimentation 7 h also only increases removal by 4%. Meanwhile, the sedimentation time for 15 h actually results in the removal which is almost 39%. However, this does not indicate that the longer sedimentation time obviously will give the biggest effect on the color removal in the hospital WWTP effluent. It needs further research for a longer time than 15 h to prove it.

Table 2 shows the removal of COD concentration, which also presents the result in different treatments. The UF membrane without pretreatment during the whole filtration process could remove COD concentration by 14%. On the other hand, the experiment with CMF alone could increase its value from 14% to 56%. The addition of sedimentation for 7 h and 15 h before CMF respectively increase 2% and 6.5% higher than the experiment with CMF alone. This is due to the potential for CMF which has high removal for other organic materials [3], thereby the experiment with CMF could be efficient to reduce COD in the effluent of the hospital WWTP.
As can be seen from Table 2, there is an increase in the removal of ammonia with an average of 1% for every treatment addition. This result is not really higher than the other feed water contents. This is also due to the amount of ammonia which is only 0.46-0.53 mg/L in the feed. The concentration of ammonia is so low that treatments are not suitable to handle it.

3.3. Membrane cleaning
After the membrane was fouled by the effluent of the hospital WWTP, it was cleaned by hydraulic cleaning to achieve optimum recovery. Hydraulic cleaning used the strategy of backwashing followed by forward flushing. This strategy is more effective than backwashing or forward flushing alone and forward flushing followed by backwashing [7]. The time for each rinse was 5 min and the total washing time recommended was 20 min. It is enough time and quite effective in the previous study [7, 15].

In this experiment, the solution of NaOCl at pH 12.6 was used to wash the UF membrane for 4 h. The mixing of NaOCl and NaOH is better than NaOCl alone. It is due to NaOH attacked the Natural Organic Matter (NOM) fouling layer, which could provide an easier chance for NaClO to destroy the gel and reaching the inner layer of fouling materials [7]. We had first investigated the effect of pretreatment on chemical cleaning with 100 ppm NaOCl before we investigated the effect of concentrations NaOCl at pH 12.6 on cleaning efficiency.

| Table 3. The effect of pretreatment to cleaning efficiency. |
|-------------------------------------------------------------|
| Without pretreatment                                        |
| Flux recovery (%)                                           |
| Removal fouling (%)                                         |
| Hydraulic cleanliness criterion (<0.05)                     |
| 69.23                                                       |
| 11.11                                                       |
| 0.44                                                        |
| Sedimentation 15 h + CMF                                    |
| 96.43                                                       |
| 77.77                                                       |
| 0.037                                                       |

Table 3 shows the membrane UF with pretreatment gives better results than without pretreatment. It achieves almost 100% flux recovery, more than 70% fouling removal and hydraulic cleanliness criterion less than 0.05. Meanwhile, the membrane UF without pretreatment only achieves 70% flux recovery, 11% removal fouling and hydraulic cleanliness criterion more than 0.05. It proves that pretreatment could maintain longer permeability of the UF membrane and get better cleaning efficiency. Therefore, it can be said that the UF membrane with pretreatment does not need further research for chemical cleaning in various concentrations of NaOCl because 100 ppm NaOCl has given quite good results.

| Table 4. The effect of concentration NaOCl on cleaning efficiency. |
|----------------------------------------------------------------------|
| Concentration (ppm)                           |
| Flux recovery (%) | Removal fouling (%) | Hydraulic cleanliness criterion (<0.05) |
| 100  | 69.23 | 11.11 | 0.44 |
| 500  | 80.59 | 51.85 | 0.24 |
| 800  | 66.66 | 47    | 0.5  |

The UF membrane without pretreatment required further research for chemical cleaning in various concentrations of NaOCl (100 ppm; 500 ppm; 800 ppm), which is shown in table 4. The chemical cleaning exhibit better recovery by 500 ppm NaOCl than the other concentrations. The concentration changes of NaClO from 100 ppm to 500 ppm has a real effect on cleaning efficiency, which could increase recovery flux by 10% and increase removal fouling by 40%. It also gives a better hydraulic cleanliness criterion from 0.44 to be 0.24. According to [9], the combination of caustic and NaOCl
could oxidize the fouling material and create more ionic functional groups. These ionic functional groups increase charge density and make cleaning more efficient. Meanwhile, the NaOCl concentration 800 ppm has the opposite effect on cleaning efficiency, which could reduce efficiency from recovery flux, removal fouling and hydraulic hygiene criteria. It was found that the NaOCl concentration threshold was obtained between 500 ppm and 800 ppm. Further research is needed to determine the true threshold.

We need to underline, the UF membrane fouled by the effluent of the hospital WWTP at low pressure needs cleaning such as physical and chemical cleaning. Physical cleaning could overcome reversible fouling and chemical cleaning could help to overcome irreversible fouling. The change of flux on the UF membrane into the fouled membrane until cleaned membrane could be used as parameters to fulfill the cleaning efficiency equation and find the best cleaning result.

4. Conclusion
The UF membrane during processing was fouled by the hospital WWTP effluent. The addition of sedimentation and CMF is effective to control the flux reduction rate caused by fouling of the hospital WWTP effluent.

As we mentioned in the discussion before, we could assume the presence of pretreatment during filtration could limit excessive fouling interactions with the membrane so that the membrane could be cleaned easier than without pretreatment as shown in table 3. In this experiment, the application of sedimentation 15 h and CMF as a pretreatment was recommended to handle fouling from the hospital WWTP effluent.

And the concentration of NaOCl used has the threshold at a certain concentration to get effectiveness cleaning when it mix with the caustic cleaning agent such as NaOH. The chemical solution of NaOCl (100 ppm) at pH 12.6 could achieve satisfying cleaning efficiency for cleaning the UF membrane with pretreatment of sedimentation for 15 h and CMF. Whereas the UF membrane without pretreatment, it required chemical solution NaOCl (500 ppm) at pH 12.6.

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References
[1] Ashfaq M Y and Qiblawey H 2018 AIP Conf. Proc. (Malta) vol 2022 (United States: American Institute of Physics) p 02002-7.
[2] Blanpain-Avet P, Migdal J F and Bénézech T 2009 Chemical cleaning of a tubular ceramic microfiltration membrane fouled with a whey protein concentrate suspension-Characterization of hydraulic and chemical cleanliness J. Membrane Sci. 337 153–174.
[3] Dyson J D 2018 New Solution for primary wastewater treatment: Cloth media filtration. FWRJ 69 11.
[4] EPA 2001 Parameters Of Water Quality: Interpretation and Standards (Wexford: Environmental Protection Agency) p 47-100.
[5] Guo W, Ngo H H and Li J 2012 A mini-review on membrane fouling Bioresour. Technol. 122 27–34.
[6] Jönsson A S and Trägårdh G 1990 Ultrafiltration applications Desalination 77 135–179.
[7] Liang H, Gong W, Chen J and Li G 2008 Cleaning of fouled ultrafiltration (UF) membrane by algae during reservoir water treatment Desalination 220 267–272.
[8] Lin J C Te, Lee D J and Huang C 2010 Membrane fouling mitigation: Membrane cleaning. Sep. Sci. Technol. 45 858–872.
[9] Liu C, Caothien S, Hayes J, Caothuy T, Otoy T and Ogawa T 2001 Membrane chemical cleaning : From art to science J. Am. Water Works Ass. 25 18.
[10] Luján-Facundo M J, Mendoza-Roca J A, Cuartas-Uribe B and Álvarez-Blanco S 2015 Evaluation of cleaning efficiency of ultrafiltration membranes fouled by BSA using FTIR-ATR as a tool J. Food Eng. 163 1–8.

[11] Matthiasson E and Sivik B 1980 Concentration polarization and fouling Desalination 35 59–103.

[12] Menteri Kesehatan Republik Indonesia 2017 Peraturan Menteri Kesehatan Republik Indonesia Nomor 32 Tahun 2017 tentang Standar Baku Mutu Kesehatan Lingkungan Dan Persyaratan Kesehatan Air Untuk Keperluan Higienis, Kolam Renang, Solus Per Aqua dan Pemandian Umum (Regulation of the Minister of Health of the Republic of Indonesia Number 32 of 2017 concerning Environmental Health Quality Standards and Water Health Requirements for Sanitation Hygiene Needs, Swimming Pools, Solus Per Aqua and Public Baths) (Jakarta: Kementerian Kesehatan Republik Indonesia) pp 10-11.

[13] Menteri Lingkungan Hidup Republik Indonesia 1995 Keputusan Menteri Negara Lingkungan Hidup No. 58 Tahun 1995 tentang Baku Mutu Limbah Cair Bagi Kegiatan Rumah Sakit (Decree of the State Minister for the Environment No. 58 of 1995 concerning Quality Standards for Liquid Waste for Hospital Activities) (Jakarta: Kementerian Lingkungan Hidup Republik Indonesia) p 9.

[14] Minegishi S, Jang N Y, Watanabe Y, Hirata S and Ozawa, G 2001 Fouling mechanism of hollow fiber ultrafiltration membrane with pretreatment by coagulation/sedimentation process Water Sci. Tech.-W Sup. 1 49–56.

[15] Mohammadi T, Madaeni S S and Moghadam M K 2003 Investigation of membrane fouling Desalination 153 155–160.

[16] Pauwels B and Verstraete W 2006 The Treatment of hospital wastewater: An appraisal. J. Water and Health 4 405–416.

[17] Shi X, Tal G, Hankins N P and Gitis V 2014 Fouling and cleaning of ultrafiltration membranes: A Review J. Water Process Eng. 1 121–138.

[18] Verlicchi P, Galletti A, Petrovic M and BarcelÓ, D 2010 Hospital effluents as a source of emerging pollutants: An overview of micropollutants and sustainable treatment options J. Hydrol 389 416–428.

[19] WHO 2018 A global overview of national regulations and standards for drinking-water quality (Geneva: World Health Organization) pp 64-74.