Pilot studies on the river water treatment using coagulation-UF membrane filtration and direct UF membrane filtration

H Nakaniishi¹, Y Yabuno¹, T Nishikawa¹, T Yasui¹, A T Pham², V -A Nguyen³

¹ Membrane and Module Production Technology Development Department, Kuraray Co., Ltd., Okayama, Japan
² Sales & Marketing Div, Kuraray Trading Vietnam Co., Ltd. Hanoi Branch, Hanoi, Vietnam
³ Institute of Environmental Science and Engineering, Hanoi University of Civil Engineering, Hanoi, Vietnam

E-mail: anhnv@nuce.edu.vn

Abstract. The pilot tests of UF membrane filtration for river surface water were carried out with coagulation as a pre-treatment (test 1) and without coagulation (test 2). Both UF membrane filtrate water of the test 1 and the test 2 met drinking water quality standard in Vietnam. Additionally, the TMP and the filtration flow rate were kept stably during test periods without changes in operation conditions although the river water turbidity was varied due to heavy rain. It was indicated that the water treatment system can be simple by skipping coagulation and/or sedimentation as pre-treatments. The filtration flux of the test 1 and the test 2 were 138 L/m²h and 93 L/m²h, respectively. The flux was higher than the reported reference value indicating that the module number of modules required for the water treatment plant can be reduced by using the FG module. The test data of the test 2 was theoretically analysed by the equations based on Darcy’s low. The estimated TMP correlated highly with measured value ($R^2=0.71$). The optimized parameters were used for the simulation of the operation. The simulation results indicated that the TMP before backwashing is increased around 35 kPa when the river water turbidity increased up to 500 NTU. It was indicated that the CIP interval should take into account the TMP increasing by turbidity variation.

1. Introduction

A combination of coagulation and flocculation, sedimentation, rapid sand filtration, and disinfection is widely used as a major water purification method in municipal and industrial water treatment facilities today. Recently, membrane filtration using microfiltration (MF) membrane or ultrafiltration (UF) membrane has been recognized as an alternative to rapid sand filtration because MF membrane and UF membrane have a high removal performance of particles, bacteria, protozoa, and so on [1]. Especially, Cryptosporidium and Giardia, which are resistant to chlorine, are the main reason to use membrane filtration for drinking water production. However, capital expenditure (CAPEX) and operation expenses (OPEX) are the concern of applying membrane filtration. Especially, processes of river surface water treatment can be complicated because the water quality can be affected by heavy rain and/or human activities. Therefore, simplification of river surface water treatment processes is important to reduce CAPEX and OPEX of water treatment facility using membrane filtration technology. Additionally, the filtration flux of the membrane filtration is important for CAPEX and
OPEX. If the system is operated under high flux, the system requires less membrane modules, less equipment, and less space. However, the operation under the high flux accelerates membrane fouling [2]. Various technologies such as pre-treatments [3, 4, 5], membrane cleaning methods [6, 7], and membrane modification [8] have been studied to improve anti-fouling properties.

In this study, we carried out pilot-scale tests for river water treatment in Vietnam without pre-treatment of coagulation, and/or flocculation and sedimentation using Kuraray’s UF membrane module which can be applicable to high turbidity water [9]. Objective of this study was to evaluate the water quality and stability of UF membrane filtration without pre-treatments.

2. Materials and Methods

2.1. Materials and chemicals

The FG module with model code FG-0101-S4 (US02-125) supplied by Kuraray was used for pilot tests. The FG module is outside-in and dead-end type UF membrane module which is consist of hydrophilic Polyvinylidene fluoride (PVDF) hollow fiber membrane with 40 m² effective surface membrane area.

Polyaluminum chloride (PACl, Goshu Kohsan supplier), sodium hypochlorite (NaClO, Viet Tri Chemicals supplier) and sulfuric acid (H₂SO₄, Lam Thao Fertilizers and Chemicals supplier) were used as chemicals for pre-treatment and chemical cleaning of membrane module.

2.2. Characteristics of the FG module

The FG module has been certified by NSF as suitable for drinking water production [10]. Figure 1 shows the structure of the FG module. The element is installed into the housing prior to use. One end of the hollow fiber membranes is potted by polyurethane resin. The hollow fibers at opposite side are sealed separately. The one end free structure enables particle removal easily during backwashing and air scrubbing. The element has a piping structure called center distributor with holes on its surface. Water and the air can be fed through the center distributor so that the particles on the hollow fibers especially around the potting side and the center of the element are removed easily.

As shown in Figure 2, the hollow fiber membrane has fine pores on its outside (filtration layer) and bigger pores on its inside (support layer) continuously connected and gradually widening. Therefore, hollow fiber membrane has higher pure water permeability compared with other MF and UF membranes. A combination of the one end free structure, the center distributor, and the high-flux hollow fiber membrane, enables operation with higher flux for higher turbidity water.
2.3. Operation procedures
Figure 3 shows the flow diagram of the testing system. River water was taken by submerged pump and fed to the 1-m³ reaction tank and mixed with chemicals. Chemical dosing rates for pre-treatment of UF membrane filtration were summarized in Table 1. Feed water was pressurized by feed pump, and filtrated by outside-in and dead-end filtration. Backwashing and air scrubbing were performed periodically. After stopping the filtration, compressed air was fed to the filtrate side pipe for the backwashing. After the pressure was increased to 0.19 MPa, air feed to filtrate side pipe was stopped. 5-Nm³/h air flow was fed to the air diffuser located at the bottom of the module, and hollow fibers were scrubbed for 36 seconds. After stopping the air feed to the air diffuser, 10-Nm³/h airflow was fed to the center distributor of the module, and hollow fibers were scrubbed for 30 seconds. Water remaining inside the module after air scrubbing was drained. The module was filled with feed water, and filtration was restarted. The test equipment is operated automatically by using programmable logic controller (PLC). Operation conditions are summarized in Table 2.

2.4. Automatic chemical cleaning (ACC)
The module was maintained periodically by ACC to avoid bacterial growth and coagulant fouling. NaClO or H₂SO₄ was diluted by feed water and fed to the module. After the module was filled with chemical, hollow fibers were immerged in the chemical for 10 minutes. 5-Nm³/h air flow was fed to the air diffuser of the module and hollow fibers were scrubbed for 30 seconds. After the chemical inside the module was discharged, hollow fibers were washed by repeating feed water filling, air scrubbing by air diffuser for 30 seconds, draining the water 5 times. ACC interval of NaClO cleaning and H₂SO₄ cleaning was 1 day and 7 days, respectively. After washing, the module was filled with feed water, and filtration restarted. ACC was performed automatically by using PLC. ACC conditions are summarized in Table 2.

2.5. Analysis
Pressure of feed water side and filtration side, feed water temperature, filtrate flow rate, raw water turbidity, and filtrate turbidity were measured and recorded continuously by using pressure sensor (Yokogawa), resistance thermometer sensor (Okazaki), electromagnetic flow meter (Keyence), turbidity meter (Microtech), and turbidity meter (Optex), respectively. Raw water and filtrate water samples haven been taken at least once a week, and analyzed at laboratory using standards methods.

2.6. Location
2 tests were carried out at different locations in Vietnam as described in Figure 4. Surface water of Da river was taken for test 1 in which the river surface water was treated by coagulation and UF membrane filtration. The test 1 was carried out continuously from September 22, 2018 to January 25,
Surface water of Lo river was taken for test 2 in which river surface water was treated by only UF membrane filtration. The test 2 was carried out continuously from May 28, 2019 to August 7, 2019.

**Table 1. Chemical dosing rates for pre-treatment of UF membrane filtration**

| Run   | River   | PACl (mg L$^{-1}$)$^a$ | NaClO (mg L$^{-1}$)$^b$ |
|-------|---------|------------------------|--------------------------|
| Test 1| Da river| 5.0                    | 1.0                      |
| Test 2| Lo river| 0.0                    | 0.0                      |

$^a$ concentration as powdered PACl equivalent to 30 % Al$_2$O$_3$

$^b$ concentration as effective chlorine

**Table 2. Conditions of operation and ACC**

| Run   | Filtration conditions | Chemical concentration for ACC |
|-------|-----------------------|-------------------------------|
|       | Flow rate (m$^3$ h$^{-1}$) | Backwash interval (min) | NaClO (mg L$^{-1}$)$^a$ | H$_2$SO$_4$ (%) |
| Test 1| 5.5                   | 30                            | 500                        | 1               |
| Test 2| 3.7                   | 30                            | 500                        | not performed   |

$^a$ concentration as effective chlorine

2.7. *Theoretical analysis*

The permeate flux $J$ (m s$^{-1}$) of a fluid with viscosity $\mu$ (Pa s) during filtration under a transmembrane pressure (TMP) $\Delta P$ (Pa) is given by Darcy’s law [11] as follows.

$$J = \frac{\Delta P}{\mu (R_m + R_c + R_p + R_o)}$$  \hspace{1cm} (1)$$

where $R_m$ (m$^{-1}$) is the membrane resistance, $R_c$ (m$^{-1}$) is the resistance of the foulant which can be recovered by backwashing and air scrubbing, $R_p$ (m$^{-1}$) is the resistance of the foulant which can be recovered by ACC, $R_o$ (m$^{-1}$) is the resistance of the foulant which cannot be recovered by backwashing, air scrubbing, and ACC. Each resistance is related to the specific resistance $\alpha$ (m s$^{-2}$), membrane area $A_m$ (m$^2$) and the mass of the foulant $M$ by the formula:

$$R_{c,p,o} = \frac{\alpha_{c,p,o} M_{c,p,o}}{A_m} = \alpha_{c,p,o} c f_{c,p,o}$$  \hspace{1cm} (2)$$
where \( c \) (kg m\(^{-3}\)) is the concentration of foulant, \( t_c \) (s) is elapsed filtration time after latest backwashing was performed, \( t_d \) (s) is elapsed filtration time after latest ACC was performed, and \( t_{st} \) (s) is elapsed filtration time after starting operation. In this study, \( c \) was assumed as suspended solids (kg m\(^{-3}\)) for simplification.

Obtained data for test 2 were analysed by equation (1) and (2). \( R_{m}, \alpha_c, \alpha_p, \) and \( \alpha_n \), were estimated by minimizing the root mean squared error (RMSE) of the calculated \( \Delta P \) using the SOLVER program in Microsoft Excel 2010.

3. Results and Discussions

3.1. Filtration test with coagulation as a pre-treatment (test 1)

Table 3 shows the water quality of the river surface water of Da river and the UF membrane filtrate water with coagulation as a pre-treatment (test 1). Some water analytical results shown that color, turbidity, iron, and aluminum in river water did not meet Vietnamese drinking water quality standard QCVN 01:2009/BYT, while all samples from UF membrane filtrate met this standard for all parameters. Therefore, it indicated that a combination of coagulation and UF membrane filtration with disinfection as a post-treatment can be applicable for drinking water production.

![Table 3. Water quality analytical results for test 1 with coagulation as a pre-treatment: minimum, average, and maximum values (n=17)](image)

Figure 5 shows the variation of the transmembrane pressure (TMP) and filtration flow rate during test period. TMP was kept from 20 to 30 kPa stably from the beginning of the test. The stable TMP indicates that foulant removed by hollow fiber membranes from river water was effectively removed by backwashing, air scrubbing, and ACC. Filtration flow rate was also kept stably at 5.5 m\(^3\)/h which is equivalent to 138 L/m\(^2\) h as the filtration flux. XIA Sheng-ji et al. carried out the filtration test using cross flow type UF membrane module with 65-90 L/m\(^2\) h for river water treatment without sedimentation as pre-treatment [12]. The higher flux of this study is indicating that number of modules required for water treatment plant can be reduced compared with the lower flux. Furthermore, cross flow filtration requires higher electric than dead end filtration because feed water should be circulated.
to remove particles from the module. Therefore, test results in this study shows CAPEX and OPEX can be reduced by using the FG module filtration without sedimentation as pre-treatment.

Figure 6 shows the variation of the river water turbidity during the test period. The river water turbidity was varied from 6 to 26 NTU on average in a day. It seems that the turbidity increasing up to 461 NTU shown as a maximum turbidity in a day in Figure 6 is caused by the ship observed around once a day. Therefore, no seasonal effects on the turbidity were found. In fact, the test period was mainly in dry season around northern area in Vietnam. Furthermore, the upstream dam might stabilize the river water turbidity around the test location. UF membrane filtrate turbidity was kept less than 0.02 NTU even when river water turbidity is increased up to 461 NTU as shown in Figure 7. Hollow fibers of FG module with 0.02 μm of fine pores enables less and more stable treated water turbidity. Furthermore, as shown in Table 3, the FG module has high removal performance of bacteria. Therefore, it is indicated that the UF membrane filtration is suitable for drinking water production.

Figure 5. TMP and filtration flow rate during the test 1. ACC cleaning of H₂SO₄ was performed at the time indicated as arrows in the figure.

Figure 6. Average and Maximum turbidity of the river water in a day during the test period

Figure 7. Filtrate turbidity during the test period.

3.2. Filtration test without coagulation as a pre-treatment (test 2)
Table 4 shows the water quality of the Lo river water and the UF membrane filtrate without coagulation as a pre-treatment (test 2). Again, some river water samples had color, turbidity, iron, aluminum, and arsenic not meeting Vietnamese drinking water quality standard. All samples of UF membrane filtrate met Vietnamese standard QCVN 01:2009/BYT with values far bellows standard except maximum value of the color which was closed to the standard value (13 vs. 15 TCU). Table 5 shows water quality analytical results of river water, filtrate without coagulation, and filtrate with coagulation on July 27 when the color of filtrate without coagulation was highest during the test. PACI dosing rate was 1.5 mg/L as powdered PACI for the coagulation. The color was effectively reduced by
combination of coagulation and UF membrane filtration. The cause of color in filtrate seemed to be the humic substances in river water. Although the UF membrane could remove the humic substances partially, it was assumed that the coagulant enhanced humic substances removal by forming small particles with humic substances bigger than membrane pores. Therefore, coagulation as a pre-treatment was preferred for raw water with high color. The coagulation can be used for continuous dosing as well as for the “emergency” when color increases in the river water.

Table 4. Water quality analytical results for test 2 without coagulation as a pre-treatment: Minimum, average, and maximum values (n=54)

| Parameters                      | Methods               | QCVN 01: 2009/BYT | Minimum River water | Average River water | Maximum River water | Minimum Filtrate water | Average Filtrate water | Maximum Filtrate water |
|---------------------------------|-----------------------|-------------------|---------------------|---------------------|---------------------|------------------------|------------------------|------------------------|
| pH                              | SMEWW 4500 H* B: 2017 | 6.5 - 8.5         | 7.6                 | 7.6                 | 8.0                 | 8.0                    | 8.2                    | 8.4                    |
| Color (TCU)                     | SMEWW 2120 B: 2017    | 15                | 4                   | 2                   | 10                  | 6                      | 32                     | 13                     |
| Turbidity (NTU)                 | SMEWW 2130 B: 2017    | 2                 | 11                  | <0.1                | 37                  | <0.1                   | 132                    | <0.1                   |
| Electric conductivity (μS/cm)   | SMEWW 2510 B: 2017    | -                 | 154.3               | 154.2               | 206.8               | 206.4                  | 270.0                  | 249.0                  |
| Iron (mg/L, Fe)                 | SMEWW 3111 B: 2017    | 0.30              | 0.59                | 0.02                | 1.83                | 0.03                   | 6.23                   | 0.04                   |
| Manganese (mg/L, Mn)            | SMEWW 3111 B: 2017    | 0.30              | <0.01               | <0.01               | 0.05                | <0.01                  | 0.17                   | <0.01                  |
| Aluminum (mg/L, Al)             | SMEWW 3111 D: 2017    | 0.006             | 0.001               | 0.015               | 0.005               | 0.005                  | 1.010                  | 0.043                  |
| Ammonia (mg/L, N)               | SMEWW 4500 NH3 C: 2017| 3                 | <1.0                | <1.0                | <1.0                | <1.0                   | <1.0                   | <1.0                   |
| TOC (mg/L, C)                   | SMEWW 5310 B: 2017    | -                 | 0.0                 | 0.0                 | 0.0                 | 0.0                     | 0.0                    | 0.0                     |
| Total dissolved solids (mg/L)   | SMEWW 2540 C: 2017    | 1000              | 80                  | 77                  | 116                | 110                    | 172                    | 176                    |
| Total hardness (mg/L, CaCO₃)    | SMEWW 2340 C: 2017    | 300               | 76                  | 74                  | 102                | 101                    | 128                    | 122                    |
| Nitrate (mg/L, N)               | EPA 352.1: 1971       | 50.00             | 0.30                | 0.32                | 0.68                | 0.67                   | 1.72                   | 1.53                   |
| Nitrite (mg/L, N)               | TCVN 6178: 1996       | 3.00              | <0.01               | <0.01               | 0.03                | <0.01                  | 0.14                   | 0.02                   |
| Arsenic                         | ASTM D 2972 - 15      | 0.010             | <0.001              | <0.001              | 0.005              | 0.002                  | 0.017                  | 0.003                  |

* Drinking water standard in Vietnam

Table 5. Water quality analytical results of the river water, filtrate without coagulation as a pre-treatment, and filtrate with coagulation as a pre-treatment on July 27, 2019

| Parameters          | River water | Filtrate without coagulation | Filtrate with coagulation |
|---------------------|-------------|------------------------------|---------------------------|
| pH                  | 7.7         | 7.8                          | 7.7                       |
| Color (TCU)         | 17          | 13                           | 9                         |
| Turbidity (NTU)     | 64          | <0.1                         | <0.1                      |
| Electric conductivity (μS/cm) | 193.1     | 193.1                        | 197.6                     |
| Iron (mg/L, Fe)     | 2.66        | <0.02                        | <0.02                     |
| Manganese (mg/L, Mn)| 0.06        | <0.01                        | <0.01                     |
| Ammonia (mg/L, N)   | <1.0        | <1.0                         | <1.0                      |
| TOC (mg/L, C)       | 1.3         | 1.0                          | 0.9                       |
| Total dissolved solids (mg/L) | 128      | 128                          | 127                       |
| Total hardness (mg/L, CaCO₃) | 93       | 93                           | 92                        |
| Nitrate (mg/L, N)   | 0.93        | 0.64                         | 0.72                      |
| Nitrite (mg/L, N)   | <0.01       | <0.01                        | <0.01                     |

Figure 8 shows the variation of the TMP and filtration flow rate during test period. TMP was kept from 20 to 50 kPa from the beginning of the test. The TMP was slightly affected by the turbidity of river water as shown in Figure 8 and Figure 9. It seemed that the increasing in river water turbidity was caused by the heavy rains. In fact, the test period was in rainy season in the northern area in Vietnam. Although the river water turbidity was increasing up to around 250 NTU, the TMP was kept
at a stably low level in the varying operation conditions such as filtration flow rate and backwash interval. It indicated that application of FG module for the water treatment in such high turbidity water was practically possible. By using the conventional coagulation, flocculation, and sedimentation, coagulant dosing rate should be strictly adjusted to keep operation stable, especially in the “emergency cases” when raw water turbidity suddenly increases, or, the water treatment facility has to be stopped and wait until the raw water turbidity decreases to acceptable value. Filtration flow rate was also kept stably at 3.7 m$^3$/h equivalent to 93 L/m$^2$h as the filtration flux. The value of filtration flux was higher than other UF membrane modules for river water treatment with coagulation as a pre-treatment [12].

![Figure 8. TMP and filtration flow rate during the test 2](image1)

![Figure 9. River water turbidity during the test 2](image2)

![Figure 10. Filtrate turbidity during the test 2](image3)

Equation 1 and 2 were used to analyze the operation data of the test 2. Figure 11 shows the measured TMP and the estimated TMP by equation 1 and 2. The measured TMP was well estimated by the parameters summarized in Table 6 including the influence of turbidity variation ($R^2=0.71$). When the turbidity increased twice from 0 to 10 days after operation started, the TMP recovery of estimated value was faster than that of measured value. It seems to be caused because ACC of NaClO could not remove all foulants from hollow fiber membranes. Therefore, hollow fiber membranes accumulated the foulants during the high turbidity condition. After the turbidity was decreased, the TMP was recovered by repeating ACC. The equation could not consider such partial recovery for the estimation.
By using the optimized parameters, it can be possible to simulate and estimate the operation TMP. Figure 12 shows the one example of the simulation. When the turbidity was increased to up to 500 NTU, the TMP before backwashing was increased around 35 kPa which was recovered immediately after the turbidity decreased to 50 NTU. Additionally, the TMP after ACC increased gradually. When the TMP increased to around 100 to 150 NTU, Cleaning in Place (CIP) using chemicals other than NaClO is necessary to remove foulants from the hollow fiber membranes. Therefore, CIP interval should take into account the TMP increasing by turbidity variation.

### Table 6. Optimized parameters and the RMSE

| $R_n$ (m$^{-1}$) | $\alpha_c$ (m s$^{-2}$) | $\alpha_q$ (m s$^{-2}$) | $\alpha_o$ (m s$^{-2}$) | RMSE (kPa) |
|-----------------|---------------------|---------------------|---------------------|-------------|
| $8.24 \times 10^3$ | $7.66 \times 10^1$ | $3.27 \times 10^2$ | $9.43 \times 10^4$ | 3.21         |

3.3. **Combination of the membrane filtration with other pre-treatments or post-treatments**

Although the water quality of filtrate water in test 1 and test 2 met drinking water standard in Vietnam, some parameters are difficult to remove by UF membrane filtration. Especially, dissolved manganese, color, and ammonia are the most concerning parameters as well as sand filtration. Effective pre-treatment and post-treatment are summarized in Table 7. Manganese can be removed as particles by UF membrane filtration after the contact oxidation as a pre-treatment. Therefore, the pre-treatment
should be considered for the water like deep well water in which manganese is not oxidized. In the test 1 and the test 2, manganese is removed because manganese is contained as oxidized particles. Color can be partially removed by combination of coagulation and UF membrane filtration. If the cause of color is soluble after coagulation in river water, color cannot be removed by UF membrane filtration as well as sand filtration. In such cases, activated carbon is effective to remove organic compound which causes color. Ammonia should be oxidized to nitrate before UF membrane filtration. The Moving bed biofilm reactor (MBBR) and the biological activated carbon are effective for nitrification of ammonia. Because ammonia is oxidized by bacteria, the oxidation should be considered as pre-treatment for safety.

Table 7. Effective pre-treatment and post-treatment for manganese, color, and ammonia removal.

| Parameter | Effective pre-treatment | Effective post-treatment |
|-----------|-------------------------|--------------------------|
| Manganese | Contact oxidation        |                          |
| Color     | Coagulation              | Activated carbon         |
| Ammonia   | MBBR<sup>a</sup> and biological activated carbon |

<sup>a</sup> Moving bed biofilm reactor

4. Conclusions
The pilot tests of UF membrane filtration for river surface water were carried out with coagulation as a pre-treatment (test 1) and without coagulation (test 2). Both UF membrane filtrate water of the test 1 and the test 2 met Vietnamese drinking water quality standard QCVN 01:2009/BYT. Additionally, the TMP and the filtration flow rate were kept stably during test periods without changes in operation conditions although the river water turbidity was varying due to heavy rains. It indicated that this method of water treatment could allow to avoid coagulation and/or sedimentation as pre-treatment steps. The filtration flux of the test 1 and the test 2 were 138 L/m<sup>2</sup> h and 93 L/m<sup>2</sup> h, respectively. The flux was higher than the reported reference values indicating that the number of modules required for the water treatment plant could be reduced by using the FG modules. The data of the test 2 was analyzed, compared with the theoretical equations based on Darcy’s low. The estimated TMP correlated highly with measured values (R=0.71). The optimized parameters were used for the operation simulation which shown that the TMP before backwashing was increased around 35 kPa when the river water turbidity increased up to 500 NTU. It indicated that the CIP interval should take into account the increase of TMP values due to turbidity variations.

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