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Effect of Filtration Modes and Pretreatment Strategies on MF Membrane

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Abstract: Coagulation is popularly used in water treatment plants as pretreatment of filtration. The advantages of rich experiences and cost-efficiency also make it become one of the competitive pretreatment of membrane filtration. However, diverse coagulation conditions including the type, dose of coagulants and operating conditions (pH, temperature, flux) influence the performance of membrane filtration. The first objective of this study is to find out the optimal coagulation pretreatment condition by comparisons of three types of coagulants at different doses. The second objective of this study is to identify the sedimentation potential under different operating conditions. The third objective of this study is to evaluate effects of two different filtration modes, which include “pressured” mode and “submerged” mode, on membrane performance. The results imply that coagulant types have different effects on water quality improvement at different doses. The fouling is reduced at the under-dose and the maximum UV_{254} removal conditions. Sedimentation helped to improve the membrane performance. The submerged mode perform better than pressured mode under all conditions.

Keywords: Pretreatment, sedimentation, optimization, fouling, filtration mode

INTRODUCTION

Recently, membrane technologies especially, microfiltration (MF) and ultrafiltration (UF) have been used progressively in water treatment process. MF/UF is good at removing suspended solids or turbidity causing materials

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but less capable of removing dissolved organic matter (DOM) (1). On the other hand, membrane fouling is still a main obstacle that limits the further development of membrane technology. To deal with this problem, many studies have been conducted such as improving the influent quality via pre-coagulation (2–23), powdered activated carbon (PAC), granular activated carbon (GAC) (24, 25), and ozonation (26, 27) process, operating at low flux (critical flux), and supporting surface shear to prevent the cake formation using cross-flow circulation, vibration of membrane fibers, and supporting vigorous bubbles, etc. Among these strategies, pre-coagulation is one of the most popular methods due to its rich experience and high efficiencies for removal of natural organic matter (NOM). Furthermore, since coagulation facilities have already existed at all conventional treatment plants, the pre-coagulation become attractive.

Concerning the pre-coagulation, Kabsch-Korbutowicz (21) studied effects of different aluminum-based coagulants types on the removal efficiency of NOM in a coagulation-UF (C-UF) process. The best results for TOC removal were observed when alum or prehydrolized coagulant was used in the pH range 6–8. Choo et al. (22) compared the efficiency of several coagulants such as polyamine, alum, polyaluminum chloride (PACl), and ferric salts based on a hybrid C-UF process to treat textile wastewater. They concluded that PACl can control the fouling most effectively. The use of polyamine as a coagulant seemed to worsen the membrane performance because the residual polymer might cause clogging of the membranes. The effects of different coagulants types on membrane performance were also investigated by Pikkarainen et al. (4). They reported that there were slight differences in the cake formation trends between ferric- and aluminum-based coagulants and chloride and sulphate counterion. Ferric sulphate was slightly superior for DOC removal, and caused slightly lower specific resistance of filter cake. According to these studies, effects of coagulant type on the hybrid coagulation-membrane filtration has not been completely identified due to complicated chemical reaction, various raw water characteristics, and different operating conditions. On the other hand, effects of coagulant dose on the hybrid C-MF/UF process are one of the most popular topics among researchers. Generally, these studies attempted to find out the optimal dose based on the maximum removal of turbidity, TOC/DOC (5), and/or the lowest increase of specific cake resistance. However, recently the interest has been rising to effects of under-dose condition. The term of "under-dose" means the coagulant dose under which coagulation cannot help to improve water quality most effectively in the unit of conventional coagulation process. But it may be helpful in the hybrid C-MF/UF process to reduce fouling if the under-dose coagulant still can change the characteristics of particles and NOM in raw water. Choi and Dempsey (19) presented that acidic under-dosed condition improved UF performance in both water quality and fouling control based on an investigation of in-line coagulation with UF process for a range of conditions such as acidic under-dosed, alkaline under-dosed, charge neutralization, and sweep-floc. Furthermore, hydraulic cleaning of the membrane resulted in better recovery of membrane performance for the under-dosed conditions. But in contrast, Judd and Hillis (5) noted that a low coagulant dose seemed to have a slightly detrimental effect on membrane performance.

Due to these incomplete and inconsistent results, the first objective of this study is to confirm effects of different coagulant types on a hybrid C-MF process to treat natural surface water and the second objective of this study is to confirm the effects of both the under-dose and the optimal dose conditions. Furthermore, the third objective is to investigate the different efficiency between in-line coagulation and conventional coagulation combined at membrane filtration process and identify the sedimentation potential under different coagulant conditions. Finally, this study tries to evaluate the effects of two different filtration modes on membrane performance, which include "pressured" mode and "submerged" mode. The term of "pressured" mode means that membrane filtration operates by pressuring the feed solution to a membrane module. The term of "submerged" mode means that membrane filtration operates by the suction force.

MATERIALS AND METHODS

The feed water (Table 1) in this study is collected from the Wolyoung lake. It is characterized by pH (pH meter 410A), UV_{254} (Shimadzu UV-2101 UV/Vis Spectrophotometer), DOC (Dissolve organic carbon analyzer, Multi N/C 300), turbidity (ASI-5000A Autosampler, Dr.mini DMT 110). As an indication of hydrophobicity, SUVA (the specific ultraviolet absorbance divided by the concentration of DOC) is also calculated. According to the results, this raw water shows low turbidity and the organic level is also lower than that of ordinary surface water.

Polyvinylidene fluoride hollow fiber membrane (PVDF) (AsahiKASEI, Co. Japan) with 0.1 μm pore size and 207 cm^2 effective filtrated area is used throughout. Table 2 shows the detailed characteristics and the filtration mode of this membrane module.

Table 1. Raw water characteristics used in this study

Parameter	Mean value
pH	7.8
Turbidity, NTU	0.9
UV_{254}, cm^{-1}	0.022
DOC, mg/L	0.81
SUVA	2.7

Table 2. The characteristics of the membrane module used in this study

Membrane type	Hollow-fiber
Membrane material	PVDF
Filtration mode	Submerged & Pressured
Nominal pore size, μm	0.1
Operating mode	Dead-end flow
Filtration flow direction	outside-in
Number of hollow-fiber	27
Surface area, cm^2	207
Permeate flux, $l/m^2 \cdot h$	80
Length of the module, cm	20

The MF module is installed vertically (Fig. 1). T the experiments are conducted mainly in two stages. Firstly, the performance of different types of coagulants is compared via the jar tests in order to determine the optimal dose. Three coagulants such as ferric chloride, aluminum sulphate, poly-aluminum chloride (PACS) are used for this purpose. Jar tests are conducted in the standard six-position gang stirrer (Programmable Jartester, Phipps and Bird, Richmond, VA) with 2-L square acrylic Gator jars. During coagulation, rapid mixing is given for 2 minutes at 150 rpm, which is followed by slowly mixing for 15 minutes at 30 rpm. Samples are taken

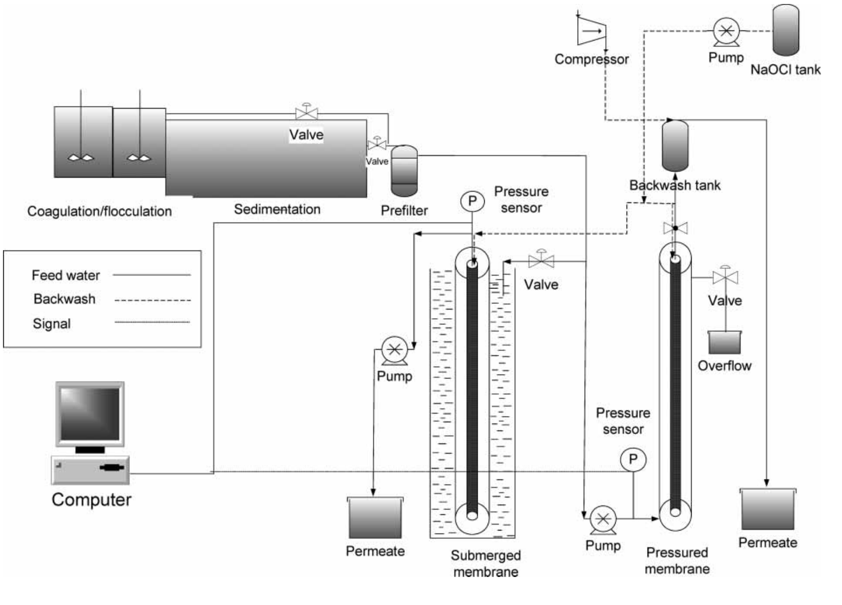

Figure 1. Schematic diagram of the hybrid C-MF apparatus used in this study.

from 5 cm above the jar bottom after addition of coagulant and after 60 minutes of quiescent setting, respectively. Secondly, the performance of the hybrid C-MF system is evaluated under various operating conditions. The effects of coagulant dose, sedimentation, and filtration mode are estimated by the TMP increasing rate and water quality.

RESULTS AND DISCUSSION

Comparison of Different Coagulant Types

The efficiency of three types of coagulants is estimated in the jar tests by the percentage removal of turbidity and UV_{254}. Fig. 2a and Fig. 2b show the residual of turbidity and UV_{254} as a function of dose for different coagulants. From Fig. 2a and Fig. 2b, it can be seen that $FeCl_3$ is effective for the removal of the turbidity when the dose is higher than 20 mg/L. Alum is effective for removal of UV_{254} when the dose is higher than 60 mg/L, and PACS is effective for removal of both turbidity and UV_{254} when the dose is lower than 18 mg/L. The maximum removal of turbidity (50%) occurs at 20 mg/L for PACS and $Al_2(SO_4)_3$ and further addition deteriorates the turbidity removal performance. The maximum removal of turbidity (60%) occurs at 60 mg/L for $FeCl_3$ and further addition fails to improve the turbidity removal.

The maximum removal of UV_{254} (43.2% and 50.9%) occurs at the dose of 20 mg/L, 60 mg/L for $FeCl_3$ and PACS, respectively. For alum, the UV_{254} removal improves with increasing dose even at 80 mg/L. Considering the large quantity of sludge production with further addition of coagulant, it is neither efficient nor economical. So, experiment with alum dose higher than 80 mg/L is not conducted.

Based on the above result, the reagent-grade commercial $FeCl_3$ is selected as coagulant in the subsequent C-MF tests. This coagulant is chosen because

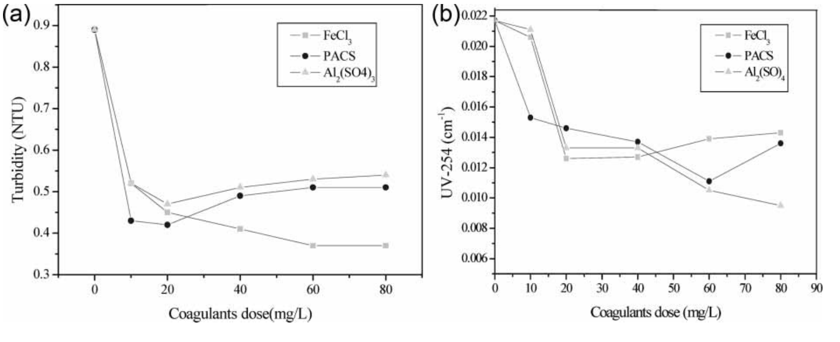

Figure 2. (a) Turbidity removal as a function of coagulant dose under three types of coagulant condition ($FeCl_3$, PACS, and $Al_2(SO_4)_3$); (b) UV_{254} removal as a function of coagulant dose under three types of coagulant condition ($FeCl_3$, PACS, and $Al_2(SO_4)_3$).

$FeCl_3$ has comparable efficiency for removal of turbidity and good efficiency for UV_{254} removal. In addition, according to work done by Moon (28), the C-MF shows the higher backwash efficiency when using $FeCl_3$ as coagulant instead of alum.

Effects of Coagulant Dose

The hybrid C-MF process is tested at doses of 0 mg/L, 5 mg/L, and 20 mg/L in order to examine the effects of the under-dose (5 mg/L) and the maximum UV_{254} (20 mg/L) conditions on membrane performance. Experiments are conducted at two different filtration modes (pressured and submerged) simultaneously to compare effects of different hydrodynamic condition on membrane performance. The filtration system is designed to stop the operation automatically when the TMP reaches 100 kPa for the pressured mode or −100 kPa for the submerged mode. Figure 3a shows the TMP as a function of time at different doses of coagulants for the pressured mode. From Fig. 3a, it can be seen that the TMP reaches 100 kPa after 20 hours of operation without the addition of the coagulant. The pressure increasing rate is 3.85 kPa/hr. The operating duration extends to 50 hours and 110 hours before the TMP reaching 100 kPa for both the under-dose (5 mg/L) and the maximum UV_{254} removal condition (20 mg/L), respectively. The pressure increasing rate are 1.56 kPa/hr and 0.71 kPa/hr, respectively.

The operation of the submerged mode, due to the limitation of system, stops simultaneously with the pressured module in each experiment even its TMP less than −100 kPa. The pressure increasing rate are 1.20 kPa/hr, 0.42 kPa/hr, and 0.19 kPa/hr for the conditions of without coagulation, the under-dose, and the maximum UV_{254} removal, respectively.

The improvement of membrane performance with coagulation is believed due to the melioration of feed water quality by adding of the coagulant. The detailed water quality analysis is put into the latter section. It should be noted that the improvement under the under-dose (5 mg/L) condition mainly due to the changes of the colloids and particles characteristics rather than reducing the turbidity or organic concentration.

The Effects of Sedimentation

Sedimentation, as a solid–liquid separation process driven by gravity is widely applied in practices. It is one of the indispensable processes in conventional water treatment plant. Whether it has significant effects on the performance of the advanced membrane filtration process is evaluated in this study. For this purpose, a series of experiments with and without sedimentation are conducted under two coagulation conditions (the under-dose and the maximum UV_{254} removal conditions) in both the pressured and the

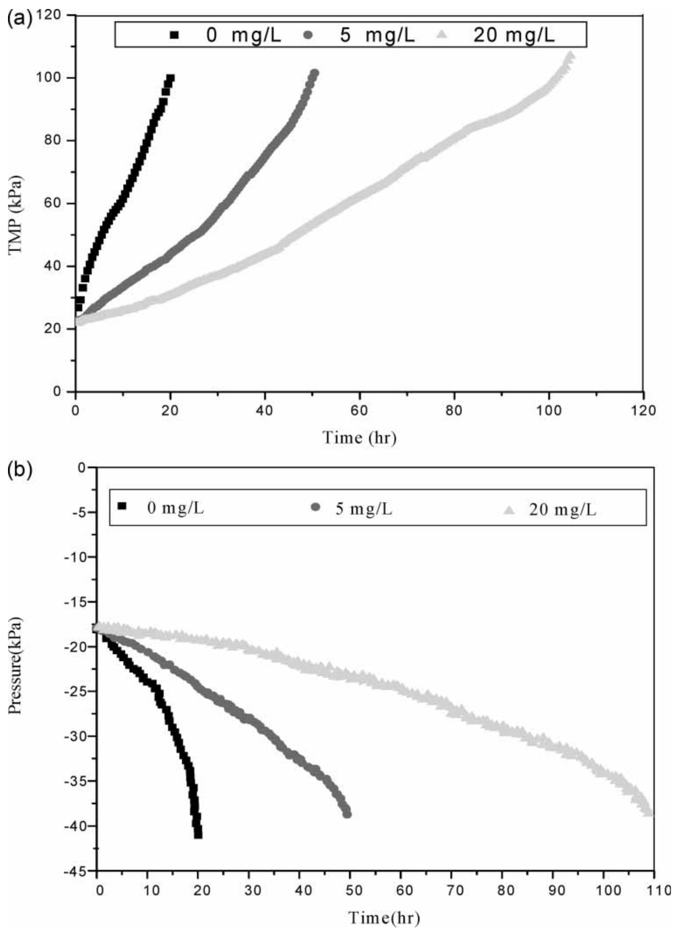

Figure 3. (a) TMP as a function of operating time at different doses of coagulant for the pressured mode; (b) MP as a function of operating time at different doses of coagulant for the submerged mode.

submerged filtration modes. As shown in Fig. 4 and Fig. 5, sedimentation slightly improves the membrane performance under the under-dose condition (5 mg/L). But the performance sharply improves after sedimentation under the maximum UV_{254} removal condition (20 mg/L) regardless of the filtration modes. The under-dose condition can cause the change of the colloids and particles characteristics, but it is unable to cause the formation of precipitation. However, the maximum UV_{254} removal condition is able to cause the formation of precipitates and most of the formed precipitates are removed through sedimentation. So, these results indicate that the effects of sedimentation are dependent on the coagulation

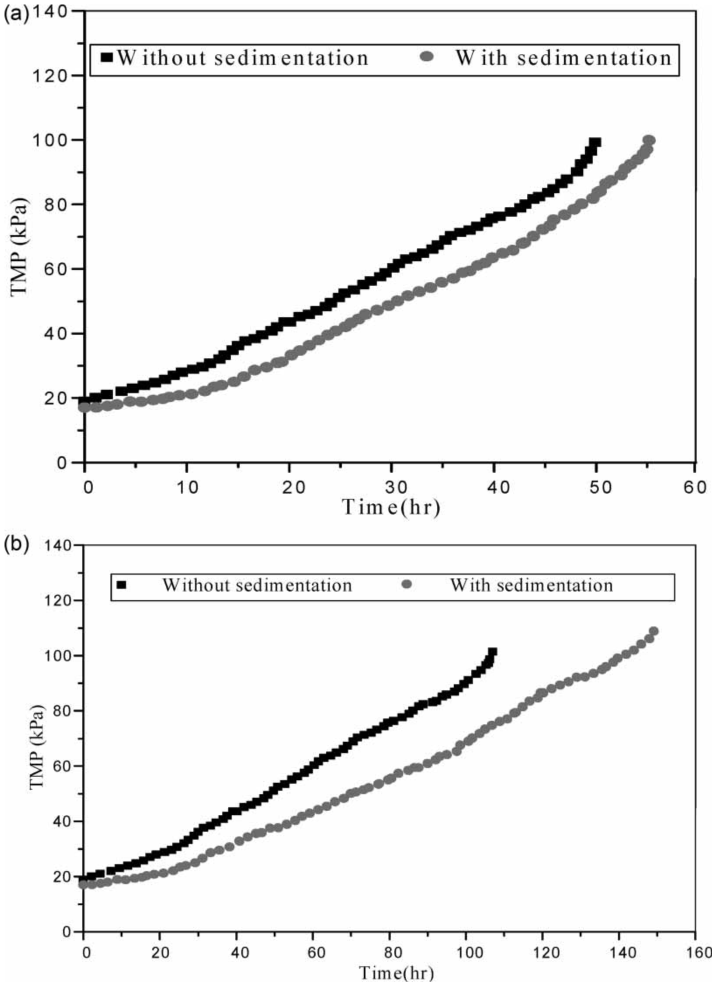

Figure 4. (a) TMP as a function of time for the pressured mode at coagulant dose of 5 mg/L with sedimentation and without sedimentation conditions (b) IMP as a function of time for the pressured mode at coagulant dose of 20 mg/L with sedimentation and without sedimentation conditions.

condition. In this study, the significant improvement of membrane performance due to sedimentation is observed under the maximum UV_{254} removal condition.

Effects of Filtration Modes

From the above figures (Fig. 3, 4, 5), it can be seen that the submerged mode performs better than the pressured mode under all kinds of conditions

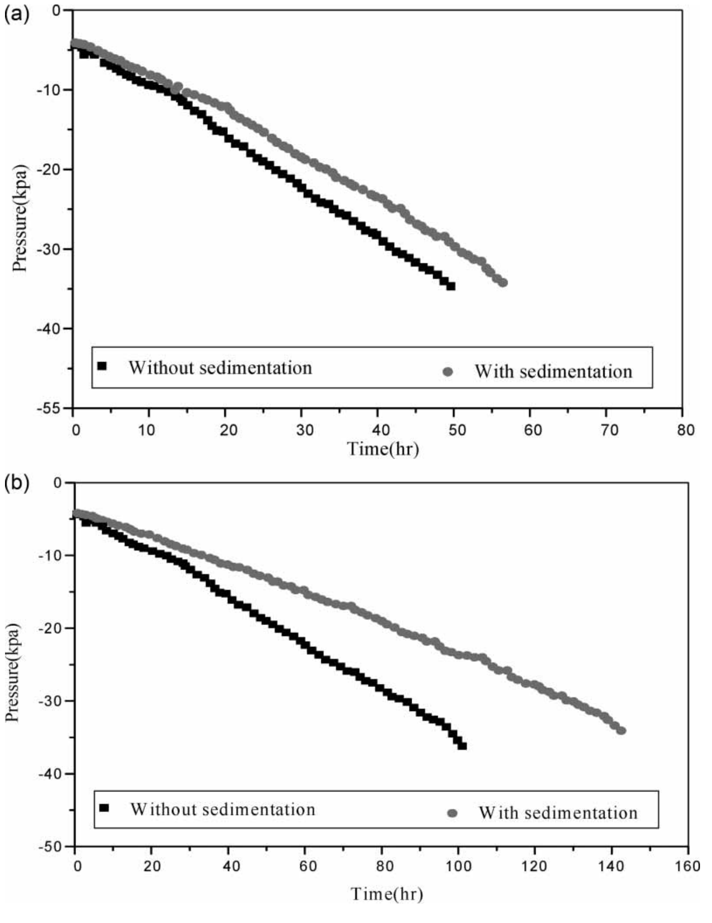

Figure 5. (a) TMP as a function of time for the submerged mode at coagulant dose of 5 mg/L with sedimentation and without sedimentation conditions; (b) TMP as a function of time for the submerged mode at coagulant dose of 20 mg/L with sedimentation and without sedimentation conditions.

according to the pressure increasing rate. This improvement can be mainly attributed to the different hydrodynamic operating conditions that the feed pumps (Masterflex peristaltic pump) are located at different places for two modes. For the pressured mode, it is located at the inlet flow. For the submerged mode, it is located at the outlet flow. It is suspected that different locations of the feed pump can cause the change in particle distribution. Therefore, the particle distributions of both modes are analyzed via the particle counter PC240PS (Chemtrac systems Inc, USA). The results are shown in Table 3. According to Table 3, the number of small particles in the feed water of pressured mode is higher than that in the feed water of the

Table 3. Particle distribution in the feed water for the submerged and the pressured filtration modes

Module	Particle size/number						
	2–4 (μm)	4–7 (μm)	7–11 (μm)	11–20 (μm)	20–50 (μm)	50–100 (μm)	100–200 (μm)
Feed water of submerged module	1100	440	88	252	170	0	0
Feed water of pressured module	1231	690	91	27	11	0	0

submerged mode. These small particles are supposed to be the main fouling materials. This different distribution of particles is caused by disaggregation of particles by the outside force coming from the feed pump, when the feed water is sent through the pump.

Permeated Water Quality

Residual turbidity and UV_{254} are analyzed as the representation of water quality for the permeated water and the water sample after coagulation. From Fig. 6, it can be seen that the raw water turbidity is around 0.9 NTU.

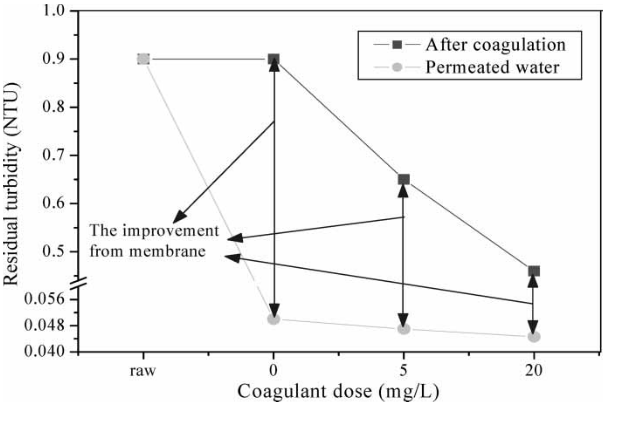

Figure 6. Residual turbidity as a function of coagulant doses for the coagulated water and the permeated water under three operating conditions: the unit of membrane filtration; C-MF process under the under-dose and the maximum turbidity removal conditions.

The turbidity removal is 27.8% and 50% for the samples after coagulation under the under-dose (5 mg/L) and the maximum UV_{254} removal (20 mg/L) conditions, respectively. For the permeated water, the turbidity removal reaches 94.4%. After the unit membrane process, coagulation slightly improves the turbidity removal. These results confirm that the membrane is effective for turbidity control even operating without coagulation but at the cost of losing permeability (see Fig. 4a and Fig. 4b).

From Fig. 7, it can be seen that the UV_{254} of the raw water is around 0.2018 cm^{-1}. The UV_{254} removal is 5.3% and 43.2% for the samples after coagulation under the under-dose (5 mg/L) and maximum UV_{254} removal (20 mg/L) conditions, respectively. The UV_{254} removal is only 3.6% for the permeated water without coagulation and its increases to 60.1% and 86.2% for the both hybrid C-MF processes under the under-dose and the maximum UV_{254} removal conditions, respectively. These results imply that the unit process of membrane filtration is ineffective to control organic compounds and the pre-coagulation has a substantial effect to improve UV_{254} removal efficiency even under the under-dose condition. This improvement is attributable to the change of characteristics of colloids or small particles by the cationic polymers from coagulant. The coagulant destabilizes a part of solvent organics and makes them to form bigger polymer-humate precipitates that can be separated from water through gravity or removed via attaching or being adsorbed by other previously formed precipitates.

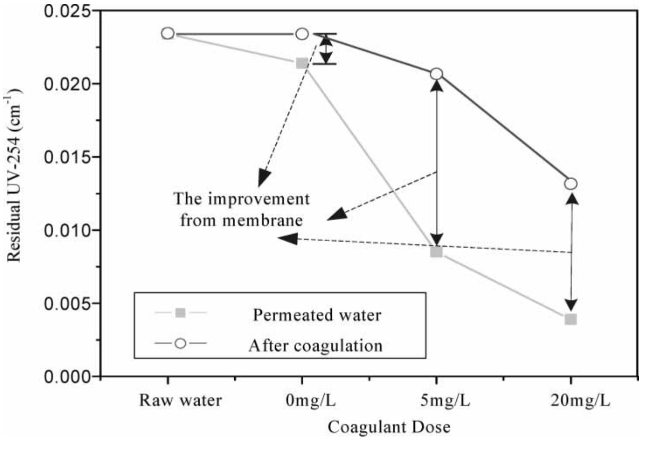

Figure 7. Residual UV-254 as a function of coagulant doses for the coagulated water and the permeated water under three operating conditions: the unit of membrane filtration; the C-MF processes under the under-dose and the maximum UV_{254} removal condition.

CONCLUSIONS

The effects of coagulant type, dose, and different filtration modes on membrane performance are investigated in this study. Based on the experimental results the subsequent conclusions are drawn:

a) $FeCl_3$ is effective for removal of turbidity when the dose is higher than 20 mg/L, alum is effective for removal of UV_{254} when the dose is higher than 60 mg/L, and PACS is effective for removal of both turbidity and UV_{254} when the dose is lower than 18 mg/L.
b) Both the under-dose and the maximum UV_{254} removal conditions help to reduce fouling.
c) Sedimentation improves the membrane performance under the under-dose and the maximum UV_{254} removal conditions, and the improvement is more significant for the maximum UV_{254} removal condition.
d) The submerged mode performs better than the pressured mode under all conditions.
e) Membrane is excellent to control turbidity but less able to control organic compounds. Pre-coagulation is able to compensate for this deficiency.

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