Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2013.768441

51 (2013) 5222–5230
July

Taylor & Francis
Taylor & Francis Group

Effects of water temperature on fouling and flux of ceramic membranes for wastewater reuse

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Received 15 June 2012; Accepted 25 September 2012

ABSTRACT

Membrane filtration is one of the promising technologies to reclaim water, but fouling is a barrier to overcome to be operated economically. Irreversible fouling makes membrane life time short than reversible fouling, because pollutants inside of pores are difficult to remove by physical cleaning and can be eliminated using chemical such as acids and bases. Ceramic membranes have strong heat resistant, so it is possible to treat thermal wastewater. High water temperature can make the viscosity of water down and thus increase permeability. There are controversies between theoretical and real viscosities. The results showed that high water temperature caused initial flux rise the flux became similar to that of low temperature water after 2 h of operation. It also led R_{irr} ratios bigger than R_{rev}. High temperature of water can increase irreversible fouling of ceramic membrane probably caused by inorganic scale formation in the membrane pores.

Keywords: Ceramic membrane; Thermal water reuse; Irreversible fouling; Reduce chemical cleaning

1. Introduction

Global warming and climate changes may cause water shortage in all over the world, and many countries promote development of alternative water resources such as reclaimed water. The effluent of wastewater is a source of water reclamation/reuse, there are no fluctuation of raw water qualities, and it can down the cost of delivering water to each areas because of its accessibility.

In membrane filtration, especially, microfiltration/ultrafiltration membrane is considered as a one of the promising technologies to reclaim water. Main process of membrane filtration is physically separation of pollutants in the water by its size and pore size of membrane, so it can produce better water quality than

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7th Aseanian Membrane Society Conference (AMS7), 4–7 July 2012, Busan, Korea

Table 1
Comparison COD of wastewater effluent and surface water

Parameters	Wastewater effluent		Surface water	
	Wastewater treatment of P city	Wastewater treatment of I city	N river of P city	H river of S city
COD_{cr} (mg/L)	20–32	8.8	2.8	3.8

other conventional processes. And this process can reduce the scale of water treatment system than conventional treatments; coagulation, sedimentation, sand filtration and so on. But they have some limitations to overcome in water treatment, and the main barrier of process is fouling. During filtration, many pollutants such as particles, organic, inorganic etc. are accumulated on membrane surface and some enter pores of membrane, stuck to wall and make its passages narrow.

To operate it for longer period, it is necessary to eliminate this fouling, so physical and chemical cleaning is applied. Fouling is classified into two forms, one is reversible fouling and the other is irreversible fouling by foulants is eliminated by physical cleaning or not. Reversible fouling is easily removed by backwash or additional water flushing, and it was mainly due to the formation of cake layers. And the foulants, which is not removed, are called irreversible fouling,

Table 2
Feed water characteristics

Parameter	Value
Turbidity (NTU)	5.11
TOC (mg/L)	6.26
UV254 (/cm)	0.115

Table 3
Theoretical and experimental water viscosities

Temperature (℃)	Theoretical viscosity (μ, N s/m^2 $\times 10^{-3}$)	Estimated flux (L/m^2 h)	Experimental flux (L/m^2 h)	Calculated viscosity (μ, N s/m^2 $\times 10^{-3}$)
Criterion temperature of calculate: 15℃				
10	1.3080	480.80	481.27	1.3067
15	1.1390	552.14	552.14	1.1390
20	1.0026	627.26	556.59	1.1299
25	0.8907	706.06	625.77	1.0050
30	0.7978	788.28	666.32	0.9438
38	0.6785	926.88	744.50	0.8447
Criterion temperature of calculate: 20℃				
10	1.3080	426.63	481.27	1.1595
15	1.1390	489.94	552.14	1.0107
20	1.0026	556.59	556.59	1.0026
25	0.8907	626.52	625.77	0.8918
30	0.7978	699.47	666.32	0.8375
38	0.6785	822.46	744.50	0.7495
Criterion temperature of calculate: 25℃				
10	1.3080	426.13	481.27	1.1581
15	1.1390	489.35	552.14	1.0095
20	1.0026	555.93	556.59	1.0014
25	0.8907	625.77	625.77	0.8918
30	0.7978	698.64	666.32	0.8365
38	0.6785	821.48	744.50	0.7487

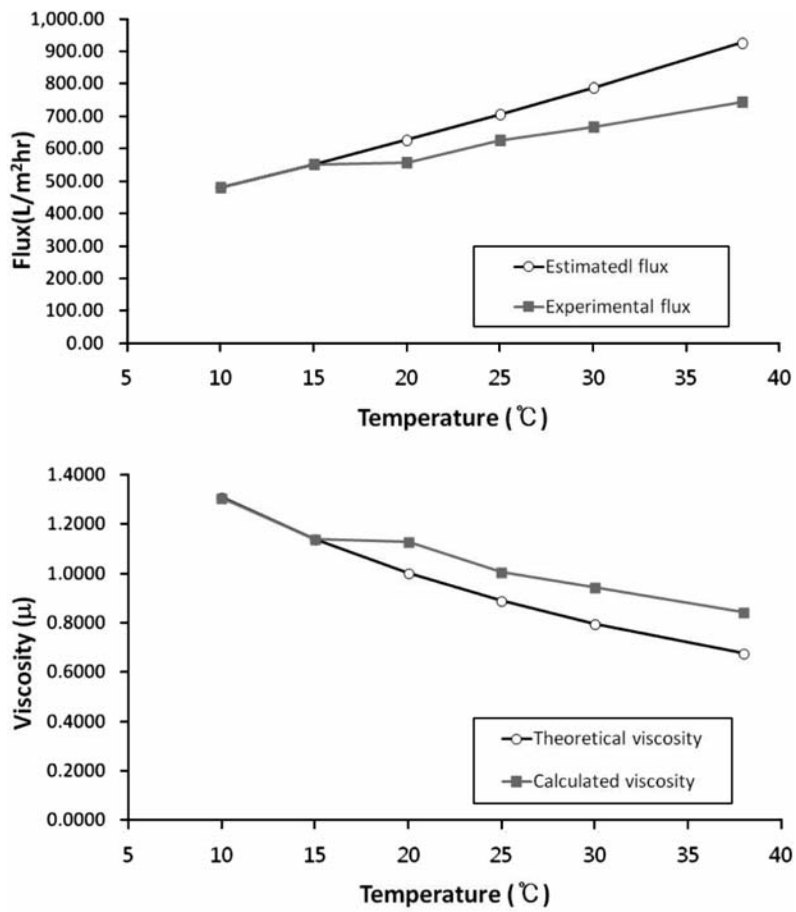

Fig. 1. Theoretical and experimental water viscosities. (a) Criterion temperature of calculate: 15℃, (b) criterion temperature of calculate: 20℃, and (c) criterion temperature of calculate: 25℃.

and it is only eliminated by chemical cleaning using base or acid solution because of it is occurred inside of pores. But chemical cleaning is method avoided because it can destroy polymer membrane and be harmful by discharging chemical maters to river water or human bodies.

The main fouling causing matters are known as natural organic matter and especially humic acid. Wastewater effluent as raw water from water reclamation process has organics as high as 2–10 times than surface water (Table 1). Therefore it is easy to be obstructed by fouling than case of drinking water treatment, so it must be effective method to control this fouling.

Recently, many researchers have paid attention to ceramic membrane as an alternative to conventional polymeric membrane. Major advantages of ceramic membrane are their durability of strong chemical oxidation and operation pressure [1,2]. And its thermal resistance is appropriate to apply on high temperature wastewater from leather, food plants, or thermal power plant [3,4]. High temperature of feed water can make a high flux in membrane system by decreasing of its viscosity. Commercial polymer membrane is easily damaged by high temperature of water, so it is hard to use on wastewater like that. But ceramic membrane is possible to apply on them.

In this study, how distribution ratio of reversible and irreversible fouling by effect of high-temperature feed water on ceramic membrane system. And it also studied coagulation effect on fouling distribution ratio.

2. Materials and methods

2.1. Water viscosity and fouling resistance calculation

The resistances in series model are employed to know about fouling resistance and raw water viscosity.

$$J = \frac{\Delta p}{\mu \times R_t} = \frac{\Delta p}{\mu \times (R_m + R_{rev} + R_{irr})} \quad (1)$$

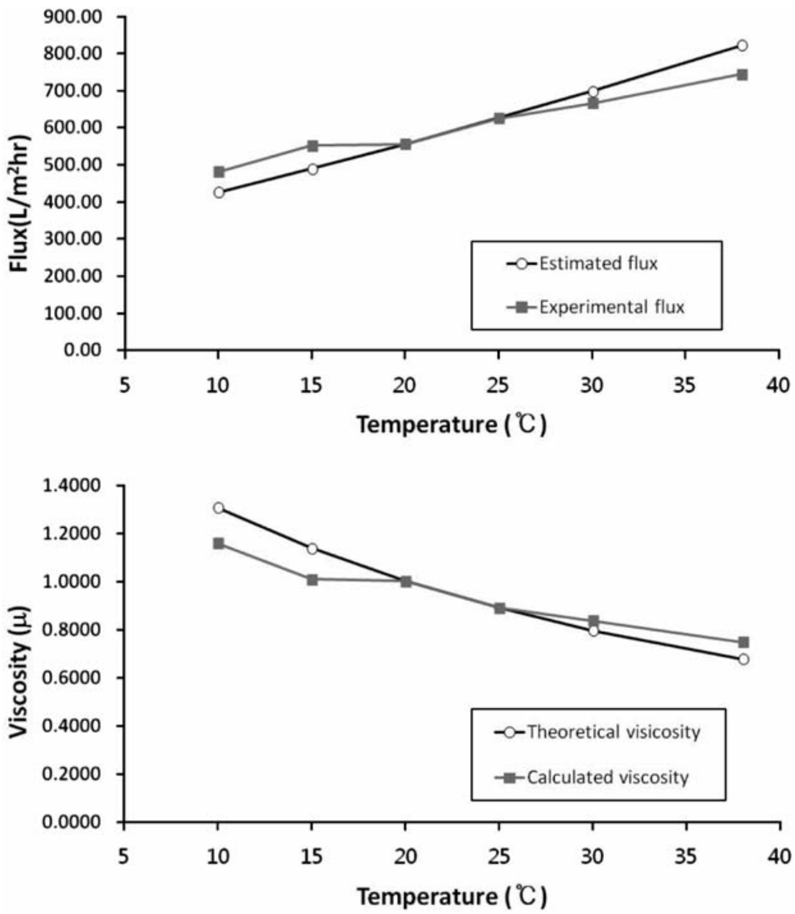

Fig. 1. (*Continued*)

where J is the permeate flux, ΔP is the TMP(trans membrane pressure), μ is the viscosity of raw water, R_t is the total resistance of filtration system, R_m is the resistance of membrane, R_{rev} is the reversible fouling resistance, and R_{irr} is the irreversible fouling resistance. During filtration, reversible and irreversible fouling resistance is gradually increase and it makes total resistance rise. Thereby membrane flux is reduced, and it means membrane efficiency is decreased. It is possible to control R_{rev} by simple backwash, but it cannot reduce R_{irr} [5–8]. And the viscosity of feed water varies with temperatures, and a higher temperature causes decrease in viscosity and vice versa. Reducing viscosity also make the membrane permeability improve.

2.2. Ceramic membrane filtration system

Feed water of this study was secondary effluent of wastewater treatment in Korea. Table 2. shows water quality having potential to make fouling. Pore size of ceramic membrane (INSIDE CeRAM, TAMI Industries, France) was 300 kD MWCO and length was 250 mm, width was 10 mm and specific surface area of 132 cm^2. The shape was monolith type and had seven channels and consisted with ZnO_3–TiO_2 as active layer and TiO_2 as support layer. Membrane system operated two hours with steady pressure of 1 bar each filtration cycle.

2.3. Water temperature increasing method

Temperature of feed water was controlled low and high to get different of water viscosities. Chiller (DH-003BH, Daehocooler, Korea) was used to make water cool or hot and maintain temperature of water. To obtain low viscosity, it was fixed on 40℃ and compared with normal temperature of effluent WWTP at 17℃. This instrument can fix the temperature continuously during filtration.

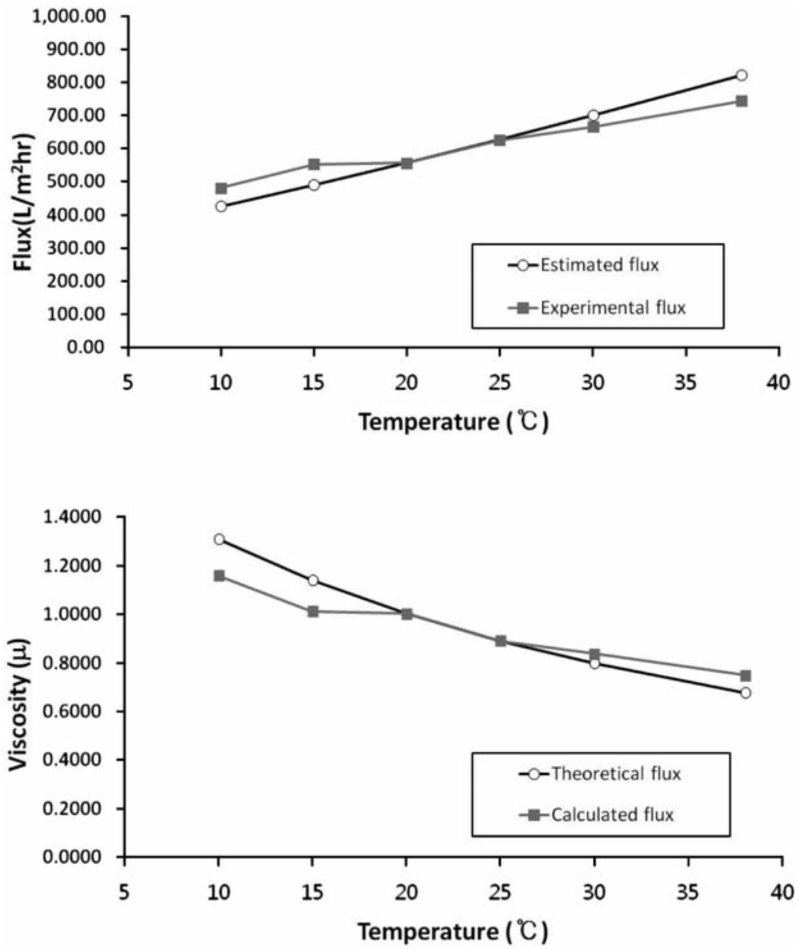

Fig. 1. (*Continued*)

2.4. Coagulation to induce increase particle size

Coagulation was performed ahead of ceramic membrane filtration to build up contaminants to flocs. Making flocs to bigger than pore size can reduce foulants which can go into membrane pores. And then irreversible fouling would be reduced and reversible

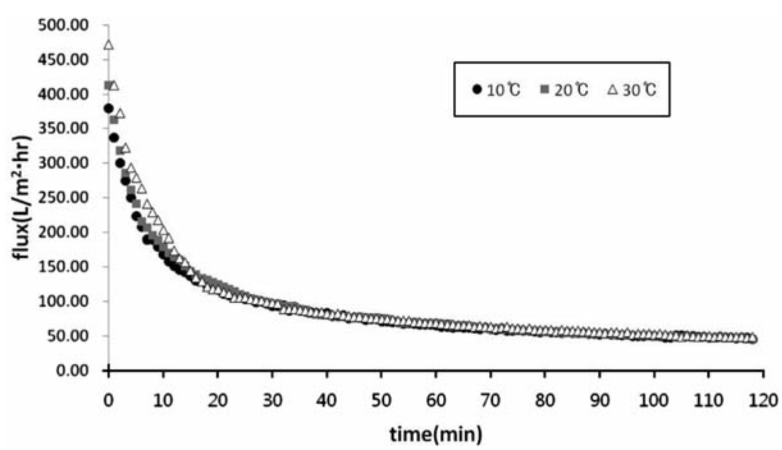

Fig. 2. Water permeability during filtration.

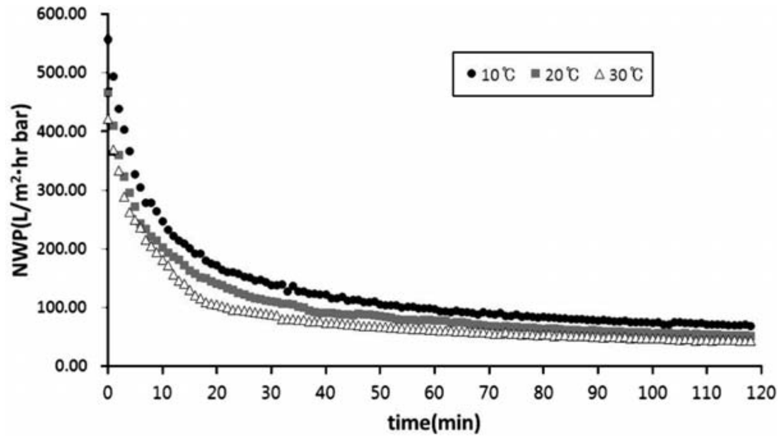

Fig. 3. Normalized water permeability during filtration.

Table 4
Calculated NWP_0 and b value result with Eq. (3)

NWP	10℃	20℃	30℃
NWP_0	704.10	627.28	595.02
b	0.47	0.50	0.54

fouling would be rose up. PAC (poly-aluminum chloride) was used as a coagulant and its optimum dosage was determined by minimum turbidity generally used.

3. Result and discussion

3.1. Experimental water viscosity according to change of temperature

Ceramic membrane filtration was performed with different water temperatures, it make water permeability grow up by increase in viscosity. Results are shown on Table 3. It is estimated what flux may have at each temperature and calculation based on each three criterion temperature's viscosities.

Although some differences was occurred on estimated flux according to change in criterion temperatures, there are similar tendency of increase flux and viscosity of each temperature. It is shown dissimilar tendency of no decrease and increase on flux and viscosity at 15–20℃. It is possible that normal temperature as approximately 20℃ could effect on this difference. Because it can make membrane have same normal temperature itself and there are no gap with water temperature, so it is easy to pass away through the membrane. But some errors exist with theoretical and experiment viscosities and flux. It may be caused by control errors on chiller setting to main same temperature or some differences of distilled water qualities on each experiment set (see Fig. 1).

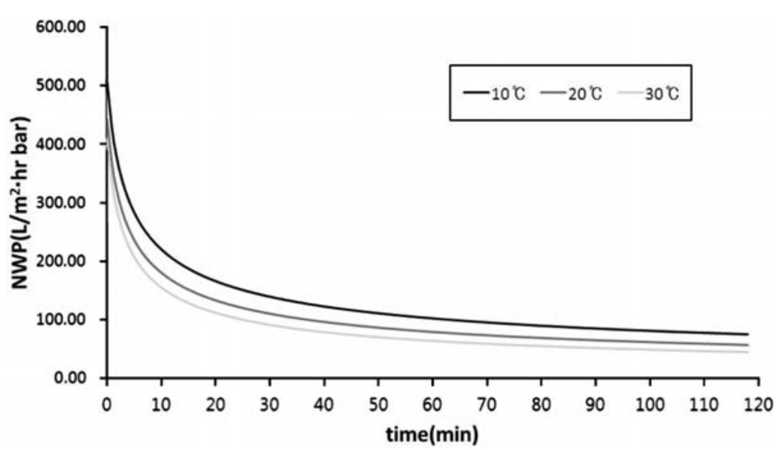

Fig. 4. Normalized water permeability during filtration with Eq. (3).

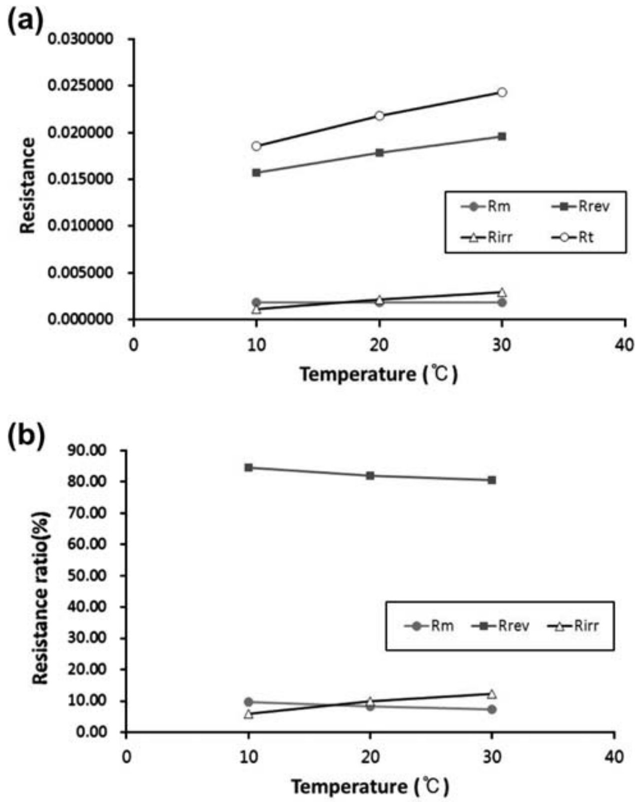

Fig. 5. Filtration resistances change in each temperature.

3.2. *Water permeability on different temperature with 2nd effluent of WWTP*

It is performed ceramic membrane filtration with raw water in different temperatures of 10, 20, and 30℃. Initial flux was 380.36 LMH at 10℃, 413.86 LMH at 20℃ and 472.55 LMH at 30℃.after 30 s from start filtration, it is observed increase on initial flux versus water temperature grow up. But after 15–20 min, they became to have same permeability as approximately 120–130 LMH and end of filtration, at 2 h later from start, these three set of experiments have 40–50 LMH of flux. It indicated that there are no benefits of low viscosity on permeability (see Fig. 2).

Graph for normalized water permeability (NWP) shows different aspect of flux declines (Fig. 3). NWP is described by the following equation:

$$\mathrm{NWP} = \frac{q_v \times K_t}{A \times \Delta p} \tag{2}$$

where q_v is permeate flow rate(L/h), K_t is temperature coefficient, A is surface area(m^2), Δp is trans membrane pressure drop (bar). It can convert flux to condition of 1 bar and 25℃. Temperature coefficient of each temperature is, 1.463 at 10℃, 1.127 at 20℃, and 0.893 at 30℃. The line of 30℃ has small flux than other lines for low temperatures. This result indicates if the low water have low viscosity at high temperature, resistances of filtration could grow up and water permeability fall.

NWP is also expressed like this:

$$\mathrm{NWP} = \mathrm{NWP}_0 \times t^{-b} \tag{3}$$

where t is time, NWP_0 is initial normalized water permeability and b is fouling rate constant. By comparing fouling rate constants at each temperature with others, it makes to know what circumstance of temperature is best to have low effects of fouling during filtration (see Table 4).

The b values correlated with membrane fouling and higher b value means its filtration time will be short than others have low value, so membrane life time will be dwindled. It is observed NWP at 30℃ has bigger b value. And it is estimated flux recovery

Table 5
Filtration resistances in each temperature (a)

Temp (℃)	μ	Order	Flux		R_t	R_m	R_{rev}	R_{irr}	$R_{rev}+R_{irr}$	R_m+R_{irr}	$R_m+R_{rev}+R_{irr}$
10	1.1595	1st	Initial	481.27	0.001792	0.001792					
			After 2h	46.50	0.018547				0.016755		0.018547
		2nd	Initial	299.64	0.002878			0.001086		0.002878	
							0.015669				
20	1.0026	1st	Initial	556.59	0.001792	0.001792					
			After 2h	45.73	0.021811				0.020019		0.021811
		2nd	Initial	253.23	0.003939			0.002147		0.003939	
							0.017872				
30	0.8375	1st	Initial	666.32	0.001792	0.001792					
			After 2h	49.05	0.024343				0.022551		0.024343
		2nd	Initial	251.36	0.004750			0.002958		0.004750	
							0.019593				

Table 6
Filtration resistances in each temperature (b)

Temp (°C)	R_t				Ratio (%)			
		R_m	R_{rev}	R_{irr}	R_t	R_m	R_{rev}	R_{irr}
10	0.018547	0.001792	0.015669	0.001086	100.00	9.66	84.48	5.86
20	0.021811	0.001792	0.017872	0.002147	100.00	8.22	81.94	9.84
30	0.024343	0.001792	0.019593	0.002958	100.00	7.36	80.49	12.15

Table 7
Water quality of specific water

Parameter	Raw water	10°C			20°C			30°C		
		Permeate	B.W	Flush	Permeate	B.W	Flush	Permeate	B.W	Flush
Turbidity (NTU)	5.113	0.104	21.833	2.975	0.102	22.100	1.757	0.118	22.000	3.500
UV_{254} (/cm)	0.1145	0.107	0.027	0.001	0.098	0.021	0.002	0.107	0.018	0.006
TOC (mg/L)	6.2550	5.223	3.239	0.699	4.628	3.673	0.486	5.059	3.090	0.745

after only physical washing as 62.3% at 10°C, 45.5% at 20°C, and 37.7% at 30°C. It is corresponded with upper results. (See Fig. 4).

3.3. Change in reversible and irreversible fouling resistances

To know how filtration resistances changed with variation of water viscosity, it is calculated with Eq. (1) based on criterion temperature of 20°C (Tables 5 and 6). It is observed that total membrane fouling resistance is related with R_{rev}, it means cake formation is dominant factor of fouling resistance (Fig. 5). It seems like R_{rev} and R_{irr} increase as temperature get higher in Fig. 5(a). But its ratio changes are different (Fig. 5(b)). Ratio of fouling resistance indicates which resistance more influenced on total fouling in same filtration time. As temperature gets higher from 10 to 30 °C, R_{irr} ratio also grows up, on the other hand, R_{rev} ratio decreases. It shows clearly that high viscosity of raw water has makes less cake layers in membrane surface. So when performing filtration with high temperature, it is easy to clean with just physical cleaning and less chemical cleaning.

Although total fouling resistance became higher as temperature increases, but it is natural because filtration with high temperature can treat the more quantity of water. It makes the membrane contacts more pollutant of water.

There are no differences as temperature getting higher (especially in B.W part, B.W is effluent after backwashing) (Table 7). It seems like there are some growing or reducing tendencies, but it is small for total values. It indicates that the position fouling grown is unrelated with what ingredients it is.

3.4. Coagulation-no sedimentation with rapid mixing

With optimum PAC dosage of 25 mg/L and 30°C water temperature, pre-coagulation and ceramic membrane filtration was conducted. It had a higher flux of 654.68 than 472.55 LMH with no coagulation after 30 s from starting filtration, and this is similar value of pure water flux as 666.32 LMH. It is clear that coagulation make permeability improve. But after 60 min, it is occurred fluctuation of TMP hard to control of steady pressure. It is because floc generated after rapid mixing flowed to monolith ceramic membrane and stuck on inner side of channels. So when backwashed into permeate port, flocs seem like long stick as same size as channel came out and hard to clear with common intensity backwashing and flushing and chemical cleaning. It is not proper to ceramic membrane which almost consisted with general monolith type with channels and operated with inside-out direction filtration flow, while polymer membrane are outside-in direction shape. So if it is necessary to do pre-coagulation, sedimentation must be existed front of the membrane.

4. Conclusions

To know that influence of viscosity to ceramic membrane system with 2nd effluent from WWTP, it is conducted with three water temperatures of 10, 20, 30°C. Water permeability did not get better with low viscosity of high temperature and fouling rate b value of 30℃ water is higher than others because of its fast flow rate. It is same result when compared R_{rev} and R_{irr} at each temperature that R_{rev} increases and R_{irr} decreases in high water temperature. Although total fouling resistance is influenced with R_{rev} a lot of part, it can make just only backwash recovers membrane ability perfectly. The results of flux recovery in 3.2 chapter prove this. Coagulation on monolith shape of ceramic membrane is not suitable because of its narrow channels and inside-out direction operation. It is necessary to do sedimentation. Therefore, it is better for ceramic membrane system to operate with low water temperature because of its small R_{rev} and b value, it makes membrane lifetime longer with only backwashing process and diminish the number and intensity of chemical cleaning. It is need to study further with lower than 10℃ to confirm that high viscosity always makes the reversible fouling increase.

Acknowledgments

This research was supported by a grant from a Strategic Research Project (Water Reuse Technology for Smart Water Micro-Grids, Code# 2012-0016-1-1) funded by the Korea Institute of Construction Technology.

Symbols

J	—	permeate flux ($L/m^2 h$)
ΔP	—	TMP(trans membrane pressure) (bar)
μ	—	viscosity of raw water (N s/m2) 10^{-3}
R_t	—	total resistance
R_m	—	resistance of membrane
R_{rev}	—	reversible fouling resistance
R_{irr}	—	irreversible fouling resistance

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