Microflocculation-ultrafiltration hybrid process for High-turbidity drinking water treatment of Dahuofang Reservoir

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Abstract
The turbidity of Dahuofang reservoir reached to 982NTU when the heavy rainfall suddenly happened to Fushun city. On this occasion, the conventional treatment processes of the water plants were unable to cope with the pollution of high turbidity. The microflocculation followed by the ultrafiltration process was studied to replace the conventional filtration for the future water process. By comparing the removal efficiency of the turbidity of the conventional treatment processes and the microflocculation followed by the ultrafiltration process, the effluent quality of the later process was satisfied with the standard of the national water quality. Chemical cleaning of maintainability adopt 400 mg/L of sodium hypochlorite solution, with the cleaning time being set to 80 min, the off-line chemical cleaning being divided into alkaline cleaning and pickling, using sodium hydroxide and hydrochloric acid respectively. The operation process can effectively alleviate the microflocculation-ultrafiltration membrane pollution. The results will provide technical guidance for upgrading and re-constructing Fushun Water Plant.

Keywords
Microflocculation; ultrafiltration; Dahuofang reservoir; turbidity

INTRODUCTION

The Dahuofang Reservoir is an important water resource in China, bearing the industrial and living water supply task of seven cities in the south-central area of Liaoning province including Fushun, Shenyang, Liaoyang, Anshan, Yingkou, Panjin, and Dalian. It can achieve 2.1 billion cubic meters water supply capacity annually, serving 23 million people. Dahuofang reservoir is located in the middle-upper of the Hun River. The reservoir and its middle-upper drainage basin are all included in Fushun area, which are distributed in Qingyuan, Xinbin and Fushun separately. It is collected together by the Hun River, Suzi River and Du River. It is a large-scale water-control project integrated with water supply, flood protection, electricity generation and etc. The length, maximum width, maximum depth and maximum storage capacity of Dahuofang Reservoir is 35km, 4km, 37km and 2.187 billion m^3 respectively.

Dahuofang and its upstream basin are located at the eastern part of Liaoning province, which are more affected by the East Asian monsoon, with temperate and semi-arid and sub-humid monsoon climate. It has obvious continental climate, with the average temperature between 5 ℃ ~ 7 ℃, the highest temperature up to 36.5 ℃,and the lowest temperature below -37.6 ℃. In spring and summer, it always has southwestern wind. The wind power is generally 3 to 4 grades, with the maximum up to 7 to 8 grades. The freeze period of the reservoir is in mid-December, and the thaw period is in late February. And its annual average runoff depth is 289mm, runoff coefficient is 0.356, and average annual rainfall is 812 mm which is more concentrated in July and August of summer. Both month rainfalls can reach up to 50% of the whole year, and the annual average humidity can reach up to 68.2% of the whole year.

Affected by the sustained rainfall, Dahuofang reservoir frequently experiences the high turbidity pollution problems. Affected by heavy rainfall in the summer of 2001, 2005, 2010 and 2013, the water turbidity increased dramatically. The water turbidity increase is caused by the large-scale rainfall carrying a mass of sediment, and the fallout floats again from the bottom of the reservoir because of the violent stir when the water yield of the reservoir increasing urgently. But after the sudden increase in the turbidity of rainfall, the reservoir water turbidity reduce very slowly, mainly because the sediment floating cannot sink smoothly and successfully. This effect usually lasts until the next spring, and it is difficult to use traditional treatment processes to make the effluent water quality reach the standard in this period.

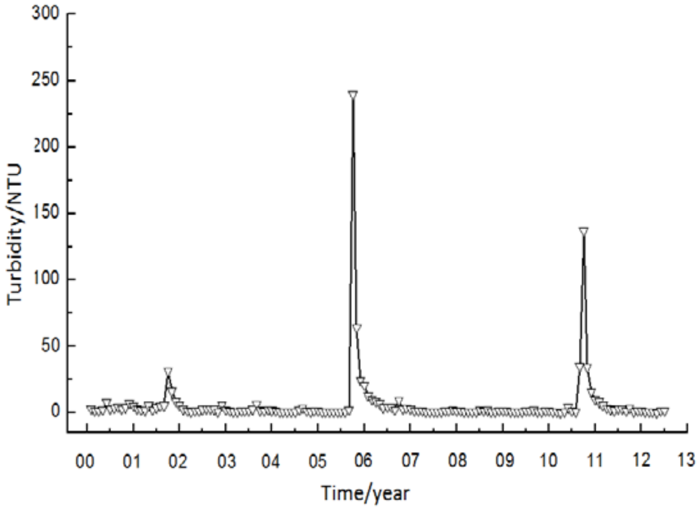

Figure 1. The turbidity of Dahuofang reservoir during the past 10 years.

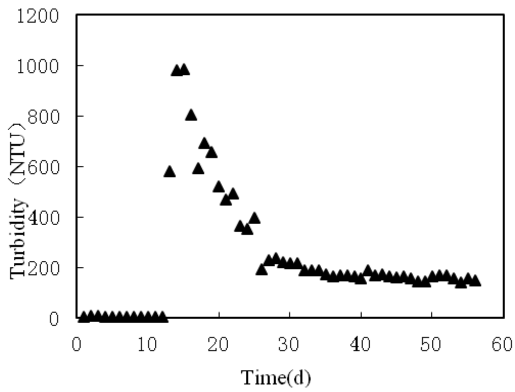

Figure 2. The turbidity of Dahuofang reservoir from Aug. to Sep. 2013.

Fushun is located in 3 km from Dahuofang, so the water quality fluctuation will obviously affect the water supply of Fushun. Fushun residents have nine drinking water supply plants, with a total design capacity of 1.11 million tons of water per day, which can supply water to the city's four districts. Its water supply area can reach up to 95 square kilometers, water supply population can reach up to 1.27 million, and the total number of water users is 473,000.

On August 16th, 2013(11:00 pm), heavy rainfall happened in the area of Dahuofang reservoir and brought high turbidity. The average rainfall of Fushun city on the day was 127.3mm. This rainfall mainly presented to the upper drainage basin of the Hun River. According to the statistics, the average rainfall amount can reach up to 156mm of the upper drainage basin of the Hun River, and the average rainfall amount can reach up to 285mm of the Qianshuikou and its upper drainage basin. The heavy rainfall caused an urgent deterioration of Dahuofang's water quality, and the high turbidity pollution brought severe influence to each water supply plants of Fushun. The turbidity of the water plant was increased from 4 NTU to 583NTU in early morning, and then rapidly increased to 982 NTU in the subsequent 10 hour, which put forward rigorous test to the existing conventional treatment technology of Fushun water plant. The water plants tried to reduce the water turbidity by increasing coagulant, and made the PAC's dosage increasing from 25mg/L to 50mg/L, but the effluent quality still cannot reach the standard and it always had filter blocking problems. In order to solve the filter blocking problems, the water plants just could wash the filter frequently. The traditional conventional process cannot effectively deal with unexpected high turbidity contamination of raw water, thus it is urgently required to improve the technology of each plants in Fushun, to meet the water quality's standards and requirements of residents in future.

Ultrafiltration has been recognized as a promising technology in drinking water treatment, due to the development of innovative materials and the decreasing costs in recent years. The common strategy of conventional drinking waterworks on high-turbidity raw waters is gradually reducing water supply capacity if turbidity of raw water exceeds 600NTU. The use of coagulants for drinking water treatment, is not able to generate water with high portability standards, which leads to the necessity of the simultaneous use of other techniques (Lin et al., 2012）. Membrane filtration technique is already widely recognized and can be implemented in combination with coagulation processes(Bergamasco et al., 2011）. In this paper, the microflocculation process hybriding with ultrafiltration process were studied for the high turbidity water treatment.

MATERIALS AND METHODS

Source water

From the investigation of Dahuofang reservoir from 2000 to 2012, it was easily to find that the range of temperature, chromaticity, turbidity, COD_{Mn} and total bacterial count is large, especially for the turbidity. According to the "Surface Water environment Quality Standard" (GB3838-2002), COD_{Mn} maximum meets the Ⅳ class water, ammonia nitrogen maximum meets the Ⅱ class water, total bacterial count and coliform bacteria satisfy Ⅰ class water quality standard.

The temperature of Dahuofang reservoir presented a cyclical change, with the minimum value at 2℃, the maximum value at 28℃. This was mainly caused by different seasons. In addition, there was around half a year when the water temperature was below 10℃ each year. The low-temperature period was relatively long. At most time, the turbidity of source water was below 10NTU. Usually, the high-turbidity period happened in July and August, because of abundant rain can carry a lot of sediment to the reservoir. Totally, the COD_{Mn} is below 3mg/L, which can be used as drinking water simply by traditional treatment process. However, when the rainfall intensity was excessively large, the COD_{Mn} increased sharply, failing to meet the standard.

Table 1. Index of raw-water quality in different period.

Index of raw-water quality	Low temperature and low turbidity period	Normal turbidity period	High turbidity period
Range of turbidity	0.37～1.56	1.25～1.87	1.20～20.10
Average turbidity	0.70	1.45	4.12
Range of COD_{Mn}	2.29～3.84	2.29～3.79	2.54～4.40
Average COD_{Mn}	2.98	3.04	3.32

Experiment set-up

The experiment included microflocculation process following with ultrafiltration process(MF-UF). The capability of the experimental apparatus was 2-3m^3/h. The raw water was pumped from sedimentation process of the water plant, then mixed fast with coagulant by the pipe static mixer. The aluminium chloride (PACl) was chosen as flocculant and the mixing time was 15min (Campinas et al., 2010 ; Hu et al., 2013）. After completely mixing and precipitation, the effluent water was pumped to the following ultrafiltration membrane process. The water flowed from the outside to inside membrane under pressure of 0.1Mpa. The membrane cleaning process was: forward-flushing 10s, up back-flushing 15s, down back-flushing 15s, forward-flushing 10s.

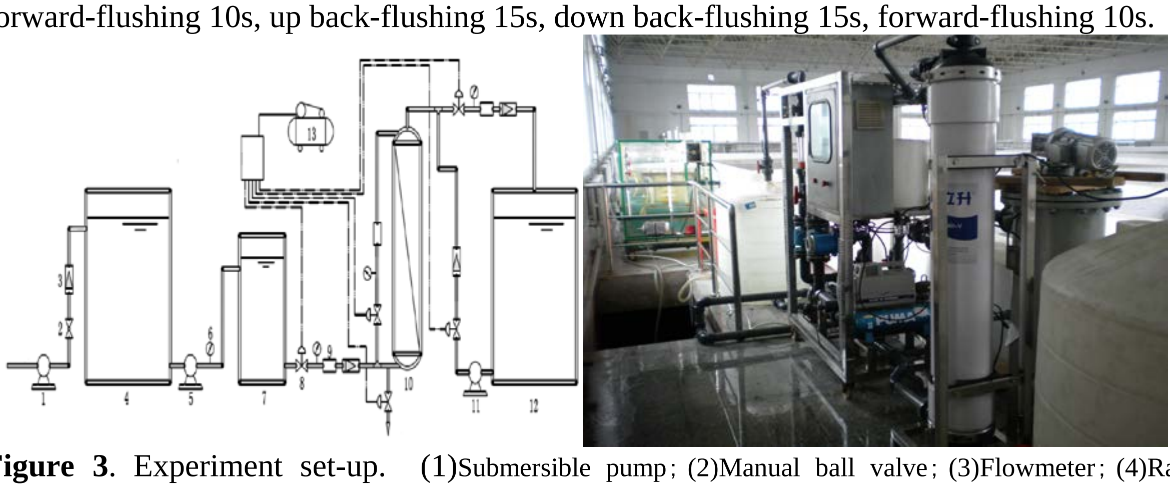

Figure 3. Experiment set-up. (1)Submersible pump; (2)Manual ball valve; (3)Flowmeter; (4)Raw water tank; (5)Booster pump; (6)Pressure gauge; (7)Self-cleaning brush filter; (8)pneumatic valve ; (9)Pressure Sensor; (10)hollow fiber membrane module;(11)Backwash pump; (12)Clean water tank; (13)Air Compressor

The raw water was lifted into the raw water tank by submersible pump, and then was pressurized by booster pump and filtered through self-cleaning brush filter to get rid of larger particles of suspended matter. After filtration, the raw water reached the lumen of ultrafiltration membrane ,was filtered from the inside out and into clean water tank. The membrane module of the experiment was produced by Hainan Lisheng Company. The model is LH3-1060V Internal Pressure Type Hollow Fiber Ultrafiltration Membrane, which takes PVC as the main material. The followings are the main technical parameters.

Table 2. Parameters of UF membrane technology.

Index	Technical parameters	Index	Technical parameters
Available membrane area/m^2	40	Material of membrane shell	ABS
Nominal scale /μm	0.01	TMP/MPa	0.02～0.08
Molecular weight cutoff/Da	50,000	Turbidity of effluent/NTU	＜0.1
pH range	1～13	Maximum operating temperature/℃	40
Membrane inner and outside diameter/mm	1.0/1.6	Filter type	Dead-end flitration and cross-flow filtration

Analysis methods

The experiment mainly used the transform membrane pressure (TMP) and specific flux (SF) to reflect situation of membrane fouling(Liu et al., 2014). The correlation formulas are listed below respectively:

$TMP=P_2-P_1$

TMP----- transform membrane pressure (kPa)

P_1----- pressure of effluent (kPa)

P_2----- pressure of inflow (kPa)

$SF= Q\times e^{-0.0239\times(T-20)}/(TMP\times A)$

$Q= Q_0\times e^{-0.0239\times(T-20)}$

SF----- specific flux L/（㎡·h·mH_2O）

Q_0----- water production measured （L/h）

Q------ water production corrected （L/h）

T------ water temperature measured （℃）

RESULTS

Turbidity removal

Comparing the effluent turbidity of water plant conventional treatment process and the MC-UF process in the flood season respectively, according to the data from August 6 to September 30, 2013, it is shown that the flood season caused a rapid increase in the turbidity of the raw water (raw water turbidity up to 982NTU). So what the water plants mostly do was discharging the turbidity by increasing the coagulant to improve coagulation. Fig.4 shows the effluent turbidity of two treatment processes.By increasing the PAC dosage to 50mg/L from August 17th, the conventional treatment process has caused the effect on removing the turbidity. But the effluent turbidity still cannot meet the requirements of ‘Standards of Drinking Water Quality’ (GB5749-2006), especially when there is the heavy rainfall, the effluent turbidity just reached up to 3NTU. With the turbidity reduction of the raw water, the average of effluent turbidity was up to 1.5NTU, and by 40 days after the flood season, the turbidity was reduced to less than 1NTU. Compared with the conventional processes, the MC-UF process has showed better turbidity removal performance. Before the flood season, without flocculating agent, directly using the ultrafiltration after coagulation and sedimentation, the effluent turbidity of the membrane process was stabilized at less than 0.1NTU. And during the heavy rainfall of flood season, use MC-UF process after coagulation and sedimentation, add 5mg/L flocculating agent, although the turbidity of raw water had significant changes, but the effluent turbidity of membrane technology was still stabilized at less than 0.1NTU. When the turbidity of raw water was reduced to an average of 165NTU, adjust the flocculating agent dosage to 1.5mg/L. Although the effluent turbidity of membrane technology had some increase, it was still stabilized at less than 0.2NTU. It shows that on the basis of water plant's coagulation and sedimentation technology, MC-UF process is more effective than original filter.

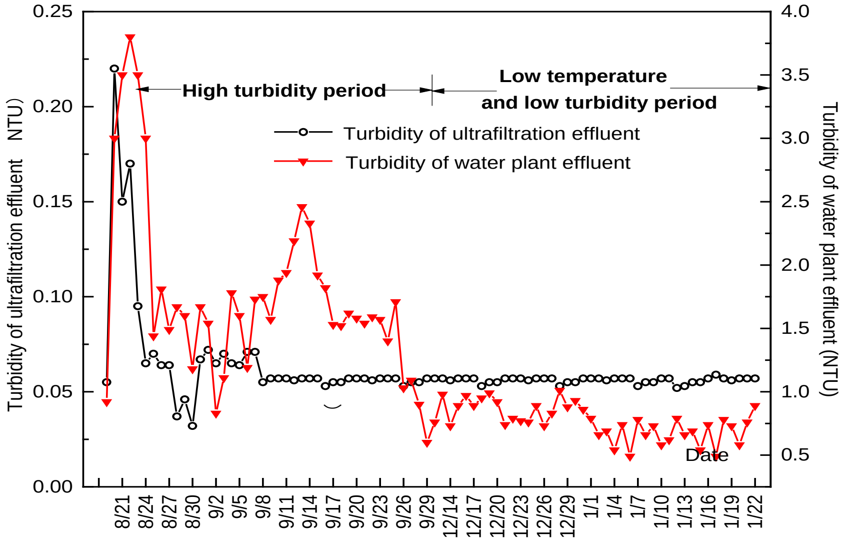

Figure 4. Turbidity removal of two treatment processes.

The following table listed the different turbidity value of raw-water turbidity, traditional treatment process effluent and MC-UF process effluent in different period. The MC-UF effluent turbidity was not affected by raw water turbidity. The membrane effluent turbidity during each period was all stabilized below 0.1 NTU. The results provided that MC-UF processes were superior to the conventional treatment process for high turbidity removal.

Table 3. The comparison of the turbidity removal during different period.

		Low temperature and low turbidity period	Normal turbidity period	High turbidity period
Raw water	Range/NTU	0.37 ～1.56	1.25 ～1.87	1.20 ～20.1
	Average/NTU	0.70	1.45	4.12
Water Plant Effluent	Range/NTU	0.15 ～0.54	0.31 ～0.44	0.32 ～2.76
	Average/NTU	0.24	0.38	0.67
	Removal rate/%	63.64	73.49	76.46
Ultrafiltration effluent	Range/NTU	0.039 ～0.1	0.044 ～0.07	0.045 ～0.08
	Average /NTU	0.059	0.059	0.062
	Removal rate /%	90.05	95.93	97.36

COD_{Mn} removal

Fig.5 showed COD_{Mn} variation tendency for the raw water of Dahuofang Reservoir in the flood period, and it also presented the variation tendency of the conventional process effluent and the microflocculation-ultrafiltration effluent. The average value of raw water for COD_{Mn} in Dahuofang was 3.37mg/L before the flood period, and the COD_{Mn} increased rapidly to 8.56mg/L during the heavy rainfall. The conventional process effluent had been in more than 3mg/L of COD_{Mn} during this time, so it can't meet the requirements of COD_{Mn}<3mg/L from 'Standards of Drinking Water Quality' (GB5749-2006). Although the water plant increased the amount of coagulant dosage, it still can't remove the organic matter effectively. And compared with the conventional process, the microflocculation process after coagulation and sedimentation can cope with the situation of sudden flood water effectively. During the whole flood season, the effluent value COD_{Mn} of microflocculation-ultrafiltration was less than 3mg/L , the average value was 1.93mg/L, and the treatment efficiency can meet the requirements of national standard. In addition, due to the raw water turbidity was gradually reduced after 20 days of heavy rain, we adjusted the dosage of secondary flocculation to 1.5mg/L. The result displays the average value of effluent COD_{Mn} was 1.91mg/L, which expressed that this process was not affected by such changes. Under the action of flocculant, microflocculation made water colloidal and dissolved organic matter transferred from the liquid to flocculation particles by adsorption, electrolysis and other physical and chemical reaction. And the flocculation particles were conductive to remove ultrafiltration membrane. Thus it can improve the removal efficiency of the process and ensure the safety of water quality effectively.

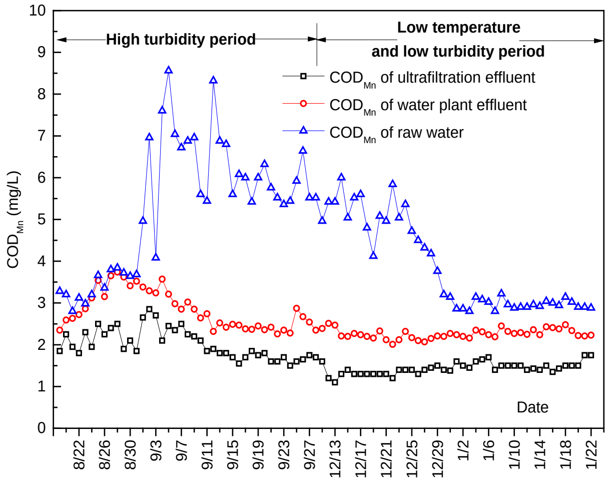

Figure 5. COD_{Mn} removal of two treatment processes from Aug.2013 to Jan.2014.

Table 4 showed that the COD_{Mn} of raw water increased with temperature rising, the same as turbidity changes. Also, the COD_{Mn} of water plant effluent was influenced greatly by raw water. Nevertheless, COD_{Mn} of ultrafiltration could still remains below 3mg/L, which was under the limit of the standard.

Table 4. The comparison of the COD_{Mn} removal effect at different period.

		Low temperature and low turbidity period	Normal turbidity period	High turbidity period
Raw water	Range/mg/L	2.29 ～3.84	2.29 ～3.79	2.54 ～4.40
	Average/mg/L	2.98	3.04	3.32
Water Plant Effluent	Range/mg/L	1.52 ～2.99	1.80 ～2.95	1.80 ～2.96
	Average /mg/L	2.16	2.27	2.51
	Removal rate/%	27.32	25.32	24.16
Ultrafiltration effluent	range/mg/L	1.23 ～2.55	1.31 ～2.51	1.38 ～2.48
	Average /mg/L	2.06	1.80	1.94
	Removal rate/%	30.66	40.66	41.30

Bacterial removal

Table.5 showed the result of determination for total bacterial count in raw water, the effluent of conventional process, and the microflocculation-ultrafiltration process during the flood season. The average number of bacteria per milliliter of water was less than 100CFU in Dahuofang Reservoir when there was no rain. But the total bacterial count was up to 2400CFU/ml which is more than dozens of times as usual during the heavy rainfall. In addition, the number of coil group was as high as 158CFU/ml above the usual. With the water turbidity decreasing gradually, the total number of bacteria was still up to 240-480CFU/ml despite the gradual decreasing of corresponding date of bacteria at the time of intense rainfall. Due to the increased number of microbes in the raw water, the operation load of conventional process was improved. And the dosage of disinfectant needs to be increased to ensure the total bacterial count of effluent to meet the requirements of standard. But this approach will lead to the increased production of disinfection by-products. No bacteria were found in the effluent of microflocculation-ultrafiltration process. The membrane can intercept all bacteria and colibacillus, so the membrane's nominal pore size was less than 0.01μm. Therefore, the process can meet the biological safety of effluent water quality, lower the dosage of disinfectant, and reduce the production of disinfection by products.

Table 5. The total number of bacteria in raw water ,the conventional plant effluent and the membrane process effluent in different period.

Test date (month,day,year)	The total bacterial counts (CFU/ml)		
	Raw water	Conventional plant effluent	Membrane process effluent
8.7.13	92	0	0
8.15.13	65	4	0
8.19.13	2400	2	0
8.23.13	2124	12	0
8.27.13	1280	2	0
8.31.13	769	0	0
9.4.13	480	2	0
9.11.13	420	0	0
9.17.13	320	3	0
9.24.13	240	1	0
10.15.13	183	1	0
11.15.13	135	0	0
12.5.13	87	0	0

Efficient cleaning effects on membrane fouling

Efficient cleaning was a significant method to control pollution, including physical cleaning and chemical cleaning(Sun et al., 2015). Physical cleaning was aimed at reversing pollution, and chemical cleaning was mainly used for irreversible pollution .The physical cleaning object was membrane pollution material such as cake layer on the surface of the pollutants and in the fenestra. The recovery rate of membrane pollution and water should be considered when determine the cleaning cycle and leaning impact, so as to ensure the good cleaning effect and the appropriate cleaning water, which was reasonable in economy and protection of resources. (Rong et al., 2013）

The physical cleaning method of the experiment, was implemented first by rushing high-speed flow of water, with amount of 8 m^3/h in 20 s, and then by the back flush, with the recoil water of 8.5 m^3/h in 15s, and lastly by rushing, with the rinse water of 7.5 m /h in 15s, and the opening and closing time should be considered. The whole cleaning process took about 1 min. The experiment was compared by transmembrane pressure(TMP) at different cleaning cycle which was 15, 20, 25, 30, 35 and 40 min. TMP difference increased by 2.5, 2.9, 2.9, 3.0, 5.2, 3.0kPa with the extension of cleaning cycle. When the cleaning cycle increased to more than 30 min, membrane pollution deteriorate rapidly. So using the 30 min for membrane cleaning cycle was more appropriate.

Chemical cleaning phase was divided into maintenance cleaning and off-line cleaning. Maintenance cleaning was usually once four days, which stands for 220 physical cleaning cycle. (Gao et al., 2011）Maintenance cleaning included back flush, soaking and physical cleaning. For backwash, sodium hypochlorite and sodium hydroxide were chosen as potions respectively, with concentration of 400 mg/L, to conduct three chemical backwash parallel experiment respectively, and observing transmembrane pressure difference changed after two reagents cleaning, with the sodium hypochlorite cleaning TMP of 3.0, 3.0, 3.4kPa and sodium hydroxide cleaning TMP was changed to 4.3, 4.6, 4.8kPa. The three test results showed that the effect of sodium hypochlorite was superior to sodium hydroxide. This was because the sodium hypochlorite had a dual role in alkali and strong oxidizing, so it can effectively remove the organic pollutants and microorganisms, and reduce membrane pollution. (Lu et al., 2015）

Under the same circumstance of sodium hypochlorite, six different experiments were set in order to develop the best soak time of maintenance chemical cleaning. The soak time were 20min，40min，60min，80min，100min and 120min respectively. As shown in figure 7, the specific flux was used to reflect the situation of membrane fouling. The higher recovery rate was, the better cleaning effect will be attained. As time goes by, the recovery rate of specific flux increased continuously. It means longer soak time can make better effect of maintenance cleaning. However, when the soak time was 100min, the recovery rate achieved 97.5％. Compared with the effect of 80min, there was small growth of 0.78％，but with 25% extra time cost. Therefore, according to the principle of economy and efficiency, it was suggested that the optimal time of maintenance cleaning was 80min.

The cycle of the off-line chemical cleaning was set to 4 months. The cleaning procedure was divided into alkaline cleaning and pickling, using sodium hydroxide and hydrochloric acid respectively. (Feng et al., 2015） In order to ensure the best cleaning agent and cleaning time, 4 experiments were implemented. The first and second adopt sodium hydroxide and hydrochloric acid. Firstly, the membrane component was steeped in sodium hydroxide solution, and the time was 1h and 2h respectively, then use hydrochloric acid for 5h cleaning. The third and fourth experiment concluded alkaline cleaning by sodium hydroxide for 2h, then use oxalic acid to immerse the membrane for 5h and 10h respectively. The concentration of sodium hydroxide was 2000mg/L, the hydrochloric acid and oxalic acid was 5000mg/L.

As shown in table 6, comparing with different soak time of sodium hydroxide solution, 2 hours can make better effect than 1 hour, resulting in higher decrease rate of TMP, and improved effect of pickling. Compare with the second time of experiment, there was the same condition of alkaline cleaning in the third one. According to the record, it was easily to find that, hydrochloric acid does better than oxalic acid in the same time. In the fourth, time of soak time is extended to 10 hours, with the similar cleaning effect to the second cycle. Based the analysis above, it can be concluded that using sodium hydroxide (2000mg/L) with soak time for 2 hours, and hydrochloric acid with cleaning time for 5h is the optimal choice for off-line chemical cleaning process.

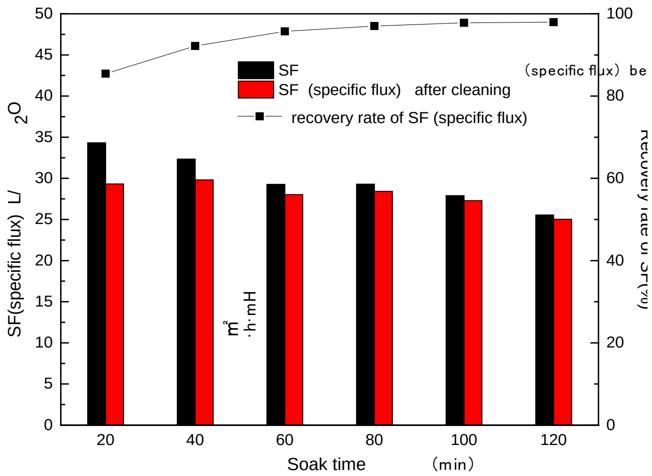

Figure 6. Conditions of long-periodic running of coagulation-ultrafiltration short flow process.

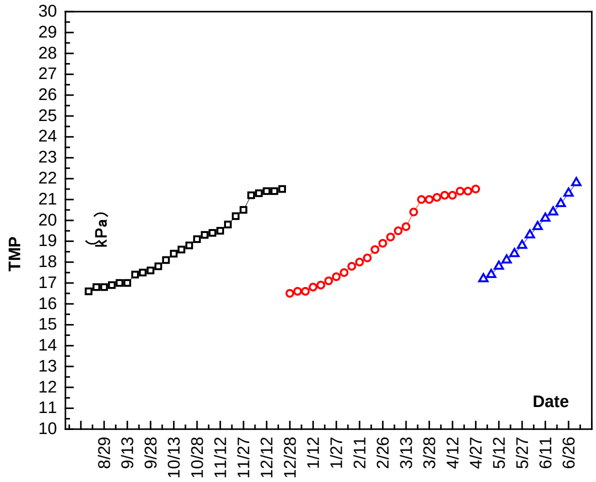

Figure 7. Changes of SF under circumstance with different soak time.

Table 6. Reduction rates of TMP after off-line chemical cleaning.

Cycle of Off-line cleaning	Cleaning process	Reduction rate of TMP after alkaline cleaning (%)	Reduction rate of TMP after pickling (%)	Reduction rate of total TMP (%)
1	NaOH(1h) →HCl(5h)	24.56	54.55	79.11
2	NaOH(2h) →HCl(5h)	26.48	57.06	83.54
3	NaOH(2h) →$C_2H_2O_4$(5h)	21.18	48.36	69.54
4	NaOH(2h) →$C_2H_2O_4$(10h)	25.30	57.69	82.99

TMP of MC-UF process during long running

After selecting technologies and parameters for water treatment and membrane cleaning, a research based on long-term running was made, aiming at the microflocculation-ultrafiltration hybrid process. Through a series of different phases, we investigated the changes of membrane fouling.

In figure 7, successively, since August 16th, 2013 to June 30th, 2014, the TMP experienced different changes. During high turbidity time, the raw-water turbidity once reached from 50 to 100 NTU. Through the adoption of microflocculation-ultrafiltration process, there was no severe fouling problem happening to the ultrafiltration membrane which was reflected by index of TMP. After that, the raw-water turbidity in low-temperature and low-turbidity period was always kept between 5 to 10 NTU. The water temperature ranged from 2 to 4℃. And we found that the increment of TMP was 4.5kPa with running MC-UF short flow processes. When it comes to the next year, it was the same with the former about changes on TMP. As results of above, a conclusion can be summarized that pretreatment of ultrafiltration membrane can effectively relieve problem of membrane fouling caused by different water quality in different seasons.

CONCLUSION

By changing the secondary microflocculation-ultrafiltration dose in the flood season, effectively economic treatment effect can be achieved. When the turbidity average of raw water was up to or above 165NTU, the secondary microflocculation-ultrafiltration dose is 5mg/L; When the turbidity average of raw water less than 165NTU, the microflocculation-ultrafiltration dose is 1.5mg/L. Chemical cleaning of maintainability adopt 400 mg/L of sodium hypochlorite solution. The cleaning time was set to 80 min. The off-line chemical cleaning was divided into alkaline cleaning and pickling, using sodium hydroxide and hydrochloric acid respectively. The operation process can effectively alleviate the microflocculation-ultrafiltration membrane pollution. The results would provide technical guidance for upgrading and re-constructing of Fushun Water Plant.

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