Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2020.25758

192 (2020) 54–60
July

The purification of rainwater with nanofiltration membrane

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Received 2 September 2019; Accepted 28 February 2020

ABSTRACT

The harvest and reuse of rainwater as a way to help relieve the water shortage is getting increased attention globally, especially in the countries facing water shortage or in rural areas without access to centralized water supply systems. As rainwater generally contains substantial amounts of contaminants including particles, microorganisms, heavy metals, and organics, further treatment of the collected rainwater is necessary. The removal efficiency of contaminants under different conditions was studied in the nanofiltration (NF) process. The results indicated that the optimum conditions in rainwater purification were as follows: the transmembrane pressure 1.0 MPa, the flow rate 30 L/min. The removal rate of chemical oxygen demand was more than 76.5%, turbidity was beyond 95%, the hardness was over 72.4%, and the total nitrogen and phosphorus were in the ranges of 32.5%–33.3% and 17.4%–18.5%, respectively. NF technology effectively removed organic substances and inorganic ions could ensure water safety. After purification, the effluent can meet the drinking water standard.

Keywords: Nanofiltration membrane; Rainwater; Removal rate; Treatment

1. Introduction

Water is an indispensable resource for human survival. With continuing growth of population and overexploitation of water bodies, the world is facing a serious challenge to satisfy the water demands for humans. The shortage of water resources is becoming a serious problem all over the world. It encourages demand for secure water supplies and promotes the search for new strategies for the sustainable use of water. Therefore, explore new water sources, which includes recycle and reuse wastewater through appropriate treatment processes is one of the useful ways to address the culprit behind the global water crises. The use of rainwater is not a novel concept but a powerful tool for solving the problems of water shortage [1–3]. Rainwater has a low concentration of pollutants. Therefore, it would require less treatment. However, the quantity of rainwater which highly depends on the weather conditions and climate has seasonal variations. Thus, it is necessary to utilize secure rainwater purification technology in order to gain safety useful water.

To mitigate water scarcity and improve its quality, many studies and researches have been focusing on improving and finding new and efficient water treatment technologies that allow achieving the expectations. Membrane technology opens up new possibilities in the exploitation of water sources, which were difficult to use previously due to technical or economic reasons [4]. Membrane process is a highly efficient separation technology developed rapidly over

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the past decades and is expected to be the prevalent water treatment technology for the 21st century [5]. Membrane technologies such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) are applied worldwide to satisfy freshwater demand [6,7]. Specifically, NF has been increasingly investigated in recent decades. NF considered as an intermediate process between RO and UF are used generally for the retention of solutes with a molecular weight cut off (MWCO) in the range of 100–1,000 Da. NF membrane can remove various organic and inorganic impurities and provide high water fluxes at relatively low applied pressure. It is also characterized by low retention of monovalent ions (K^+, Na^+) and high retention of di- and multi-valent ions (such as Ca^{2+}, Mg^{2+}), as well as organics with a molecular weight above 200–500 Da [8]. Previous studies have investigated the practicability of NF as wastewater and potable water treatment technology. Very high retentions of sulfates by two types of NF membranes were examined during the NF treatment of the artificial water and the real water [9]. NF rejected almost 100% of copper ions at low concentrations. The rejection was 99%, 89%, and 74% for cadmium, manganese, and lead respectively, using 1,000 mg/L concentration level, pH 1.5, and 4 bar [8]. NF membrane separation process had been applied fully or partly removing the pharmaceuticals. For its high selectivity to norfloxacin, the rejection of this antibiotic remained between 87% and 99.5%. The highest norfloxacin rejection was reached at pH 6.5 [10]. The rejections of neutral pharmaceutical active compounds (carbamazepine) and ionic pharmaceutical active compounds (diclofenac and ibuprofen) from drinking water resources were respectively 31%–39% and 55%–61% [11]. With the advantages of high water flux, high retention of organic molecules, high retention of multivalent anionic salts, and low cost of operation and maintenance, NF can be obtained large-scale application [12–17].

A major challenge to all membrane-based water treatment systems is membrane fouling [18–20]. Membrane fouling is inevitable, resulting in a drop in permeability and changes in separation performance. There were many ways to control membrane fouling, such as pretreatment, physical and chemical cleaning, optimized operating conditions. A robust and inexpensive pretreatment is one of the useful ways to alleviate membrane fouling. The pretreatment not only lightens the load of foulant but also improves the treatment efficiency. Although it does not eliminate fouling entirely. Ozonation has been found to be effective in reducing NF membrane fouling. After ozonation, feed waters differed significantly in terms of foulant charge, hydrophilicity, and concentration [21]. These differences were important for the NF membrane cleaning procedure. Coagulation is another useful pretreatment process. A previous study had reported that hybrid coagulation-NF membrane was a potential technique in effectively removing humic acid and reducing bromate and bromide, compared with using coagulation or NF membrane alone. And at the same time, it can maintain and even improve the membrane permeate flux [22]. Ultrafiltration and microfiltration, which are also reliable methods for the removal of suspended substances, some organic and microbiological contaminants, are frequently applied as pretreatment methods for the NF membrane [23].

In this study, the roof rainwater was purified by an integrated pretreatment–NF process in order to obtain safety potable water. Therefore, the main objective of this study was to evaluate the effect of the NF membrane on rainwater purification. For this purpose, the rejections of chemical oxygen demand (COD_{Mn}), turbidity, total hardness, total nitrogen and phosphorus during the NF membrane at different conditions such as transmembrane pressure (TMP), flow rate and operation time were investigated. The flux recoveries were also investigated using different chemical cleaning agents.

2. Experiments

2.1. NF membrane

The membrane module was spiral-wound (GE Osmonics Co., USA). The membrane was cross-linked thin-film composite membrane with polysulfone (PS) support substrate over polyamide (PA). It had a filtration area of 2.6 m^2, with the MWCO was 200 Da.

2.2. Experimental procedures

A schematic diagram of the NF system is demonstrated in Fig. 1. The experimental temperature was 25°C ± 1°C, operation pressure was from 0.4 to 1.4 MPa, and the feed flow was in the range of 15–40 L/min. Roof rainwater was collected by the pipes. The water was treated by sedimentation, ozone, biological activated carbon, and secondary filter in order to remove the particles and a part of organics. Then, the water flowed into the NF membrane. And the membrane system was operated at a constant TMP. An additional chemical cleaning was carried out. After NF membrane filtration, ultraviolet (UV) disinfection was used. COD_{Mn}, turbidity, total hardness, and other water quality indexes were analyzed. In addition, the energy was provided by a solar power generation system, which consisted of a highly efficient crystalline silicon solar cell module array, a controller, a battery, and an inverter power box.

2.3. Analytical methods

The acid potassium permanganate method was used to determine COD. Firstly, sulfuric acid (H_2SO_4) and excessive potassium permanganate ($KMnO_4$) solution were added in the water sample. The color of the solution was aubergine. In order to speed up the reaction, the sample was heated. Then, excess sodium oxalate ($Na_2C_2O_4$) solution was added to consume the residual $KMnO_4$. The color of the solution fades by degrees, from aubergine to colorless. And finally, a surplus of $Na_2C_2O_4$ was titrated with the $KMnO_4$ solution. When the tipping point came, the color of the solution became reddish. According to the consumption of $KMnO_4$, the COD of the sample can be calculated. Turbidity was measured by a turbidimeter (HACH, TL23, USA). Total phosphorus and nitrogen were determined by spectrophotometry. Ethylenediamine tetraacetic acid complexometric method was used to analyze the total hardness. Furthermore, the color was determined according to the national standard GB11903–89. The others (total bacterial

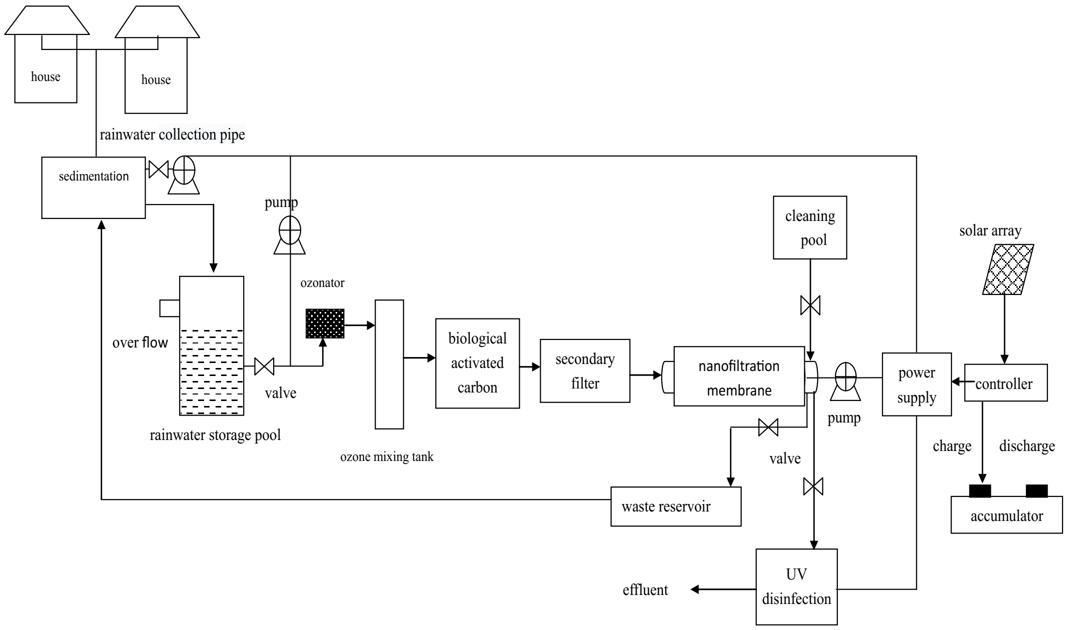

Fig. 1. Schematic diagram of the experimental equipment.

count, total dissolved solids, conductivity, and so on) were measured based on the national standards.

3. Results and discussion

3.1. Pretreatment

Roof rainwater was collected by pipes, and then the water flowed into the sedimentation basin. The characteristics of the rainwater after sedimentation are displayed in Table 1. After sedimentation, turbidity, $COD_{Mn,}$ and color significantly decreased to 87.95 NTU, 78.9 mg/L, 2^6 to 65.27 NTU, 34.2 mg/L, 2^3, respectively. However, there were no obvious change in total hardness, total bacterial count, total dissolved solids, conductivity, total phosphorus, and nitrogen. Sedimentation had a good effect on the removal of organic matters, a little influence on ions. Ozone which can oxidize the macromolecular organic matters and provide dissolved oxygen for activated carbon improved the activity of activated carbon. Biological activated carbon can remove the organic matter, reduce the color and turbidity in water. Secondary filter removed the insoluble substances and colloids in water, can reduce the fouling of the NF membrane, and extend membrane lifetime. After Ozone, biological activated carbon, and secondary filter, all the water indexes except hardness and pH were decreased remarkably. The removal rates of COD_{Mn}, turbidity, color, bacterial count, total dissolved solids, conductivity, total phosphorus and nitrogen were 73.34%, 68.84%, 87.5%, 95.3%–96.5%, 15.13%, 18.56%, 94.85%, and 78.99%, as displayed in Table 1.

3.2. Effect of different factors on membrane filtration properties

3.2.1. Transmembrane pressure

The removals of COD_{Mn}, turbidity, hardness, total bacterial count, total phosphorus, and nitrogen with a different TMP were investigated under a flow rate of 30 L/min.

Table 1
Characteristics of the rainwater after sedimentation

	Rainwater	After sedimentation	Before membrane
Turbidity (NTU)	87.95	65.27	20.34
Chemical oxygen demand (COD_{Mn}, mg/L)	78.9	34.2	9.02
Total hardness ($CaCO_3$, mg/L)	99.4	98.2	97.9
Total bacterial count (CFU, mL)	15–20 × 10^4	15–20 × 14	7 × 10^3
pH	7.41	7.34	7.32
Color	2^6	2^3	2^0
Total phosphorus (mg/L)	0.14	0.13	0.0067
Total nitrogen (mg/L)	13.3	12.9	2.71
Conductivity (μS/cm)	183	181	149
Total dissolved solids (mg/L)	119	117	101

The experimental results are shown in Fig. 2. The removal rates were increased with the operating pressure increasing from 0.4 to 1.4 MPa. The removals of turbidity, COD_{Mn}, total hardness, total phosphorus, and nitrogen were respectively in the ranges of 93.5%–97.4%, 51.7%–85.6%, 53.3%–74.5%, 12.6%–25.9%, and 28.5%–37.5%. It had significant growth in the removals of COD_{Mn} and hardness. The removal of COD_{Mn} was increased with the operating pressure. The phenomenon could be explained by the fine-pore model [23–25]. A membrane retention rate of the neutral molecular was regarded as unchanged. The thickness and concentration of the stagnant layer would increase with TMP. And the penetration of COD had little change when its concentration in the stagnant layer was within the fine-pore model allowed range. Therefore, an increase in membrane flux caused the increasing removal of COD. The membrane flux increased linearly with membrane operating pressure as Fig. 3 shows. Therefore the removal of COD was increased with the TMP. Previous studies had introduced a parameter $J \times R_{COD}$, which was the product of flux and COD removal rate. It can be an index that investigated the NF membrane properties. The maximum value of $J \times R_{COD}$ corresponding to the pressure was regarded as the theoretical optimal operating pressure. Meanwhile, membrane pressure should be minimized when it satisfied the flux and removal rate. Hence, the optimum operating pressure was chosen considering all factors.

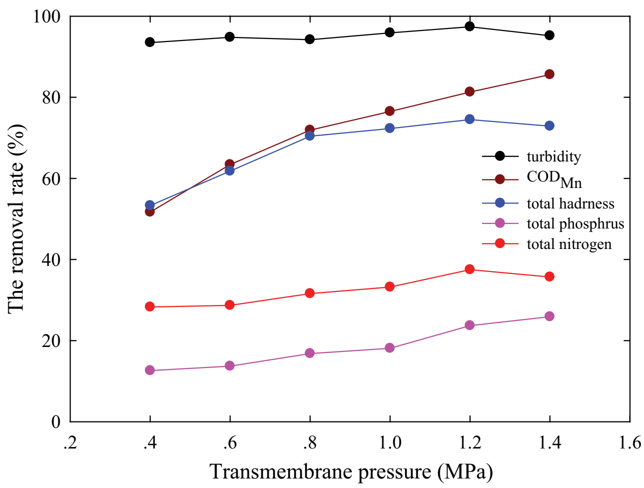

Fig. 2. Removal rate with transmembrane pressure (flow rate 30 L/min, 25°C ± 1°C, operation time 24 h).

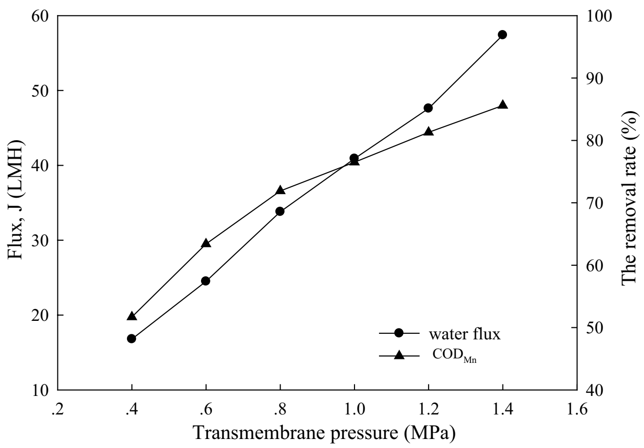

Fig. 3. Variation of flux and COD_{Mn} removal rate with transmembrane pressure (flow rate 30 L/min, 25°C ± 1°C, operation time 24 h).

3.2.2. Flow rate

At the constant pressure with 1.0 MPa and 1.2 MPa, the removal rates under different flow rates were measured (Figs. 4a and b). Fig. 4a showed that both of the turbidity removals at the TMP 1.0 MPa and 1.2 MPa were over 90% when the feed flow rate was between 15 to 40 L/min. More than 80% of COD_{Mn}, 72% of hardness, 21% of phosphorus, and 35% nitrogen were removed under the pressure of 1.0 MPa in the NF membrane process. And the removal rates of COD_{Mn}, total hardness, total phosphorus, and nitrogen were about 68%, 72%, 17%, and 32% respectively under the pressure of 1.2 MPa (Fig. 4b). The results illustrated that

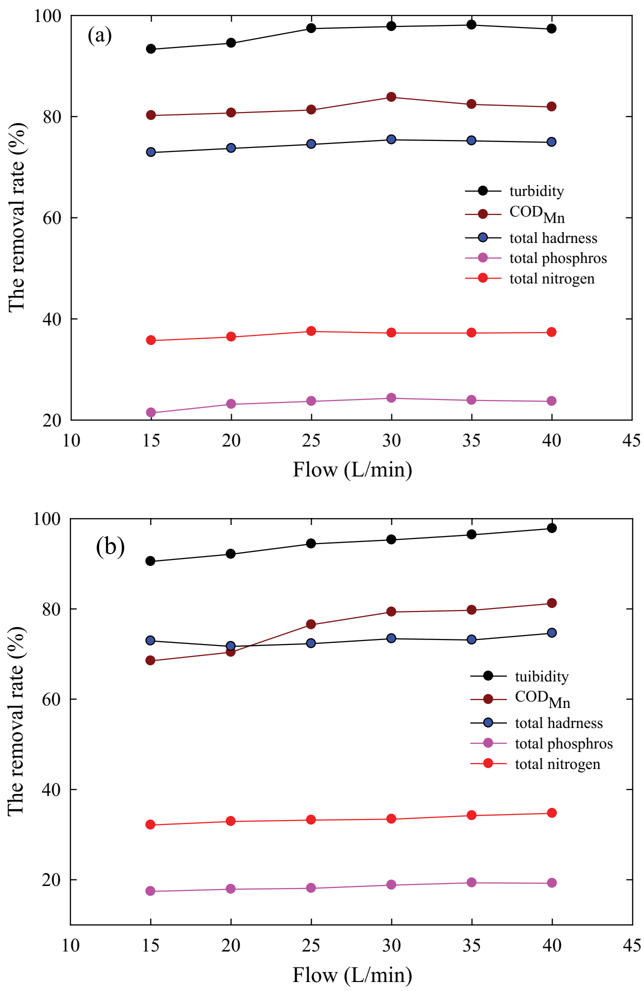

Fig. 4. Removal rates of contaminants under different flows at 25°C ± 1°C with 24 h (a) ΔP = 1.0 MPa and (b) ΔP = 1.2 MPa.

although the rejections were increased with the increased flow rate, the variations were small. In addition, membrane flux showed a small increase with the flow as displayed in Fig. 5. The increased flow accelerated the water velocity, made the liquid fluidity increase, which caused membrane surface fouling alleviated. However, the fluid flush on the membrane surface with excessive water flow can shorten membrane lifetime. Under small water flow, membrane fouling was easy to be formed [26]. Therefore, the optimum flow rate of 30 L/min was determined according to the experimental results.

3.2.3. Operation period

Time is an important factor affecting membrane properties. Normalized flux values of experiments with operation time are shown in Fig. 6. Flux declined with operation time, which illustrated the formation of membrane fouling. However, the trend of flux decline rapidly in the beginning and then a constant permeability loss. This result revealed different fouling mechanisms. Foulant-membrane interactions determined fouling behavior in the beginning. Organics caused pore plugging or absorbed on the membrane surface, which resulted in a rapid reduction of the effective permeate area. And foulant-foulant interactions affected the performance in the later stage. A cake layer was formed and accumulated on the membrane surface, leading to a further decrease in flux. As displayed in Fig. 6, little flux decreased and the effluent quality was stable. After pretreatment, the good water quality had little effect on membrane fouling. Therefore, there was not serious membrane fouling formation. The removal of COD_{Mn} was over 70% with operation time as shown in Fig. 7. And the removal of turbidity was beyond 95%, the hardness was in the range of 72.4%–73%. The removal rates of phosphorus and nitrogen were between 17% and 18.5%, 27.5%, and 33.3%, respectively.

3.3. Membrane cleaning

Membrane fouling caused flux decrease. Therefore, it is necessary to clean the membrane. Hence, pure water cleaning and an additional chemical cleaning with sodium hydroxide (NaOH, 0.5%), hydrochloric acid (HCl, 0.5%) and hydrogen peroxide (H_2O_2, 0.5%) were carried out in order to investigate the recovery of flux under 1.0 MPa and flow rate 30 L/min. The results were displayed in Fig. 8. After cleaning, the recovery rate of flux was 65.78% (pure water), 64.25% (HCl), 84.8% (NaOH), and 89.2% (H_2O_2), respectively. The recovery rate using H_2O_2 as chemicals was the highest. The main foulants in rainwater contribution to membrane fouling were organic matters. H_2O_2 which possessed powerful oxidization had a good effect on removing membrane organic fouling.

3.4. Discussion

After sedimentation, COD_{Mn} and turbidity of the rainwater were decreased. However water hardness, pH, and the other indexes were no obvious changes. Ozone which can oxidize the macromolecular organic matters and provide

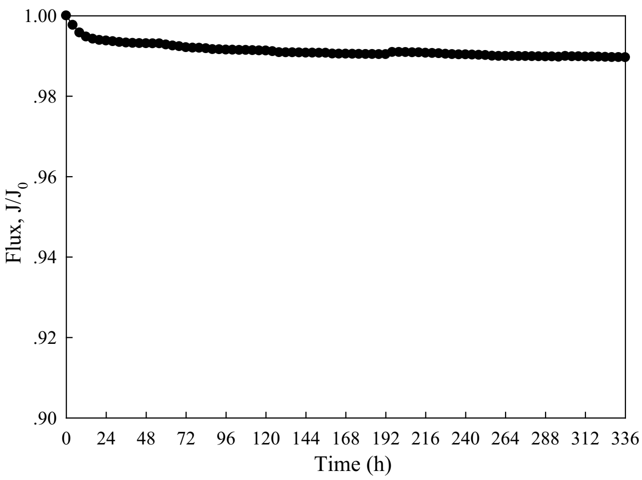

Fig. 6. Variation of flux with operation time (ΔP = 1.0 MPa, flow rate 30 L/min, 25°C ± 1°C).

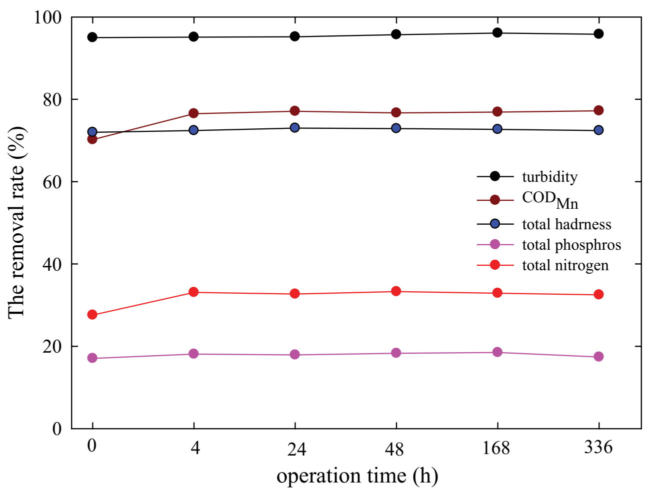

Fig. 7. Removal variation with operation time (ΔP = 1.0 MPa, flow rate 30 L/min, 25°C ± 1°C).

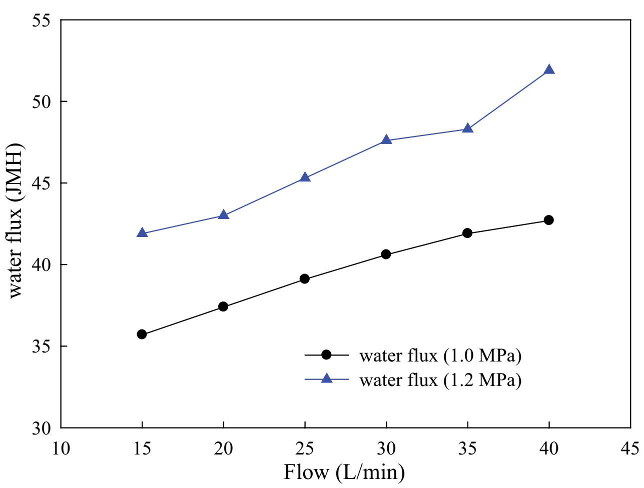

Fig. 5. Variation of flux with the different flow (25°C ± 1°C).

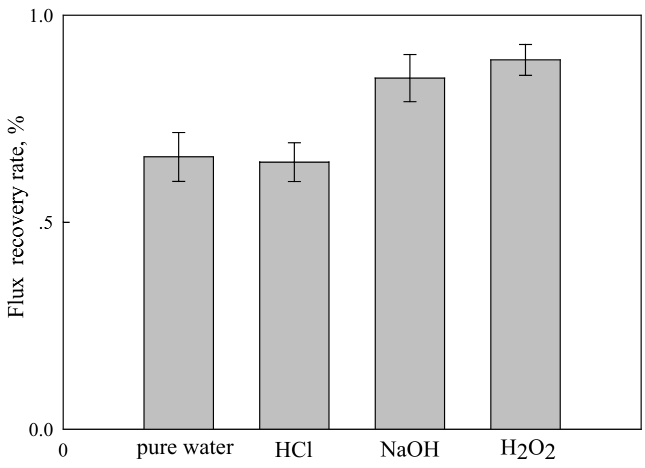

Fig. 8. Membrane flux recovery after chemical cleaning (ΔP = 1.0 MPa, flow rate 30 L/min, 25°C ± 1°C).

dissolved oxygen improved the activity of activated carbon. Biological activated carbon can remove the organic matter, reduce the color and turbidity in water. After the treatment of ozone, biological activated carbon, and secondary filter, the indexes including COD_{Mn}, turbidity, color, total hardness, total bacterial count, total phosphorus, and nitrogen were decreased rapidly. It ensured the normal operation of the NF membrane process. And rainwater was further purified by the NF membrane. Ultimately, COD_{Mn} of the membrane effluent was less than 1.5 mg/L, turbidity was about 0.44 NTU, the total hardness was below 24.1 mg/L ($CaCO_3$), the total phosphorus and nitrogen were 0.0051 and 1.7 mg/L, respectively. Moreover, bacteria were not detected. And the water pH was about 7.29. The effluent quality can meet the drinking water standard. Therefore, the NF membrane was an excellent technology for rainwater purification. Moreover, a solar power generation system consisted of a highly efficient crystalline silicon solar cell module array, a controller, a battery, and an inverter power box that was equipped to provide electrical.

4. Conclusion

The effect of TMP, flow rate, and operation time on the removal efficiency of COD_{Mn}, turbidity, total hardness, total phosphorus, and nitrogen were investigated during the NF membrane process. And an additional chemical cleaning with different chemicals was carried out at the end of filtration. The results were shown as follows:

- According to the experimental results, the optimum TMP was 1.0 MPa, the flow rate was 30 L/min respectively.
- Under the optimum conditions, COD_{Mn} of the effluent was less than 1.5 mg/L, turbidity was about 0.44 NTU, the total hardness was below 24.1 mg/L ($CaCO_3$), and phosphorus and nitrogen were 0.0051 and 1.7 mg/L, respectively. Bacteria cannot be detected. It indicated safe and high-quality potable water can be obtained after NF membrane treatment. It was one of the useful ways of rainwater reuse, provided an available way to solve the water.
- NaOH and H_2O_2 had an excellent effect on removing membrane fouling. The recovery rate of flux was about 84.8% and 89.2%, respectively.

Acknowledgments

This research was supported by grants from the National Natural Science Foundation of China (no. 51709007) and the Fundamental Research Funds for the Central Universities (No. 2019MS033).

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