Membrane Water Treatment, *Vol. 8, No. 4 (2017) 311-321*
DOI: https://doi.org/10.12989/mwt.2017.8.4.311

PPTA/PVDF blend membrane integrated process for treatment of spunlace nonwoven wastewater

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(*Received August 25, 2016, Revised December 18, 2016, Accepted December 19, 2016*)

Abstract. Hydrophilic and high modulus PPTA molecules were incorporated into PVDF matrix via the *in situ* polymerization of PPD and TPC in PVDF solution. PPTA/PVDF/NWF blend membrane was prepared through the immersion precipitation phase inversion method and nonwoven coating technique. The membrane integrated technology including PPTA/PVDF/*NWF* blend membrane and reverse osmosis (RO) membrane was employed to treat the polyester/viscose spunlace nonwoven process wastewater. During the consecutive running of six months, the effects of membrane integrated technology on the COD, ammonia nitrogen, suspended substance and pH value of water were studied. The results showed that the removal rate of COD, ammonia nitrogen and suspended substance filtered by PPTA/PVDF blend membrane was kept above 90%. The pH value of the permeate water was about 7.1 and the relative water flux of blend membrane remained above 90%. After the deep treatment of RO membrane, the permeate water quality can meet the water circulation requirement of spunlace process.

Keywords: spunlace nonwoven wastewater; *in situ* polymerization; PVDF; PPTA; fouling; water flux

1. Introduction

Nonwoven spunlace process utilizes high speed and high pressure water needle to punch the bulk fiber network. This promotes the mutual entanglement of fibers as a result of the consolidation of a nonwoven production form. The product has the advantages of soft handle, drape and good hygroscopicity which are widely used for the medical hygiene products, such as surgical clothes, dressing, wet wipes, diapers, lens wipers, and etc (Midha *et al*. 2013, Chen *et al*. 2010, Kalebek *et al*. 2010). The core components of spunlace nonwoven technology is the high speed and high pressure water needle which has a diameter of only micron level. Thus, a demanding water quality is necessary to guarantee the water can continuously, stably and quickly perforate the pinhole on the water needle plate and form the water needle. Inorganic compounds

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 ISSN: 2005-8624 (Print), 2092-7037 (Online)

such as calcium and magnesium ions in water would be scaling and blocking the pinhole. Besides, the presence of organic matter and microorganisms can result in the instability of high pressure water needle and some short fluff would induce serious pinhole blockage (Hajiani *et al*. 2010, Martin *et al*. 2016). A daily output for 10-15 t of spunlace nonwoven production line needs water use of 150-250 m^3/h. In order to ensure the product quality, reduce production costs and enhance environmental protection, spunlace wastewater must be treated to recycle.

Currently, the treatment of spunlace circulating water mostly uses the dissolving air floatation (DAF) process. The wastewater of spunlace nonwoven process could contain the following components: fiber oil agent, all kinds of short fiber chips and the dissolved short fluff under high pressure, microorganism and inorganic matters, dust and other substances. The dissolved gas in water can well produced through the ejector in DAF process. The water contamination can be adhered to the surface of micro bubbles, rapidly rising and be removed by the slag discharge device. DAF process requires a larger floor area, a huge processing equipment and also needs sedimentation, sand filtration and other physicochemical treatments after the air floatation. The process and operation maintenance is complex which needs a larger investment (Chen *et al*. 2008, Jankowski *et al*. 2009).

Membrane separation technology has the function of separation, concentration and purification as well as the characteristics of high efficiency, energy saving and environmental protection. It has become one of the important technology of water treatment. As one kind of polymer material with excellent performance, polyvinylidene fluoride (PVDF) possesses good resistance to chemical corrosion, high temperature, oxidation, weather and radiation, and film-formation ability, which has become the most widely used materials currently in micro/ultrafiltration (MF/UF) membrane preparation (Miguel *et al*. 2016, An *et al*. 2016). However, the surface energy of PVDF is very high which results in its strong hydrophobicity. In addition, the lower glass transition temperature would induce the great shrinkage after drying process. These results contribute to the flux decline in water processing and need frequent membrane fouling. Moreover, the poor mechanical strength of PVDF membrane could cause the cleavage and even rupture of membrane filaments or sheets in the membrane bioreactor (MBR) system (Liu *et al*. 2011, Ishizaki *et al*. 2016). Even through the improvements of sealing component and process, it is difficult to guarantee the maintainance of excellent performance fundamentally during the long-term aeration operation of PVDF membrane in MBR system.

In our previous study, poly(p-phenylene terephthalamide) (PPTA) molecules which have high modulus and high strength but are difficult to dissolve and molten were incorporated into the PVDF membrane matrix via in situ polymerization of p-phenylene diamine (PPD) and terephthaloyl chloride (TPC) in PVDF solution and the porous blend separation membranes were prepared through the immersion precipitation phase inversion method. Compared with pure PVDF membrane, the hydrophilicity, mechanical strength and antifouling properties of the prepared blend membranes were obviously improved (Li *et al*. 2015a). In addition, the membrane formation mechanism of PPTA/PVDF blend membranes has been well investigated through the kinetics and thermodynamics in our previous reports (Li *et al*. 2015b).

In this study, PPTA/PVDF blend membranes were prepared through the in situ polymerization method. Their membrane modules were fabricated and applied in the spunlace wastewater treatment in membrane bioreactor (MBR) system. The membrane integrated process including PPTA/PVDF blend membrane and reverse osmosis (RO) membrane technology was used to treat the spunlace nonwoven wastewater. During the continuous operation of six months, the effects of membrane integrated process on the removal of COD, ammonia and suspended solids in wastewater, and the variation of pH were studied.

2. Experimental

2.1 Materials

Poly(vinylidene fluoride) (PVDF, FR-904, η=1.4-1.9, Mn=38000) powder was obtained by Shanghai 3F New Materials Co., Ltd., China. p-Phenylene diamine (PPD, ≥99%) and terephthaloyl dichloride(TPC, ≥99%), *N*-methyl-2-pyrrolidone (NMP, ≥99%) and anhydrous LiCl (≥99%) were all supplied from Aladdin Co. Ltd., China. Polyethylene glycol (PEG, 100 kDa) was analytical reagent and supplied by Tianjin Kermel Chemical Reagent Co. Ltd., China. Spunlace nonwoven wastewater was supplied by Zhengzhou Fenglin Non-Wovens Co., Ltd., China. Polyethylene terephthalate (PET) spunbonded NWF with the grammage of 75 $g \cdot m^{-2}$ was supplied from the Shijiazhuang Tianlue Advanced Textile CO. Ltd.

2.2 Synthesis of membrane casting solution

The detailed synthesis process of PPTA/PVDF casting solution was described in our previous study (Li *et al*. 2015a). PVDF powder was placed at 80℃ for 24 h in vacuum oven before use to remove the free water molecules. 1.6 wt% LiCl and a certain volume of N-methyl-2-pyrrolidone (NMP) were added into the three-necked flask. Stir fully until the complete dissolution of LiCl. Then PVDF powder was added to the system at 65℃ followed by fast mixing to obtain homogeneous and transparent solution. Then system temperature was declined to room temperature and a certain amount of p-Phenylene diamine (PPD, 0.4 mol/L) was introduced. After its complete dissolution, the system temperature was dropped to 5℃. Afterwards, terephthaloyl dichloride (TPC) powder (TPC/PPD=1.007/1, mole ratio) was added into the system rapidly and accurately with a rigorous stirring of 30 min. Then, the system temperature rose up to 65℃ and agitate the system for 2 h. Finally, a uniform light yellow solution was obtained. The total polymer concentration was fixed at 16 wt% and the weight ratio PVDF and PPTA was kept at 85:15.

2.3 Preparation of flat sheet membranes

The fabrication of flat sheet membrane was completed through the coating of PPTA/PVDF casting solution onto PET nonwoven fabric surface and the immersion precipitation phase inversion method. PET nonwoven fabric was pasted onto the clean and smooth glass plate. Then, the dope solution synthesized above was cast onto the PET nonwoven fabric surface at ambient temperature using a stainless steel casting knife with a gap distance of 250 m. The glass plate was immersed immediately into the coagulation of pure water at ambient temperature. Nascent membranes were rinsed with pure water several times and stored in pure water for use.

2.4 Fabrication of membrane modules

PPTA/PVDF blend flat sheet membrane modules were used in the MBR process to treat spunlace wastewater. Membrane modules were fabricated according to Fig. 1. PET nonwoven

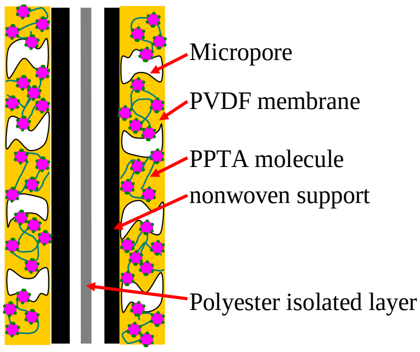


Fig. 1 The schematic diagram of the fabrication of membrane modules

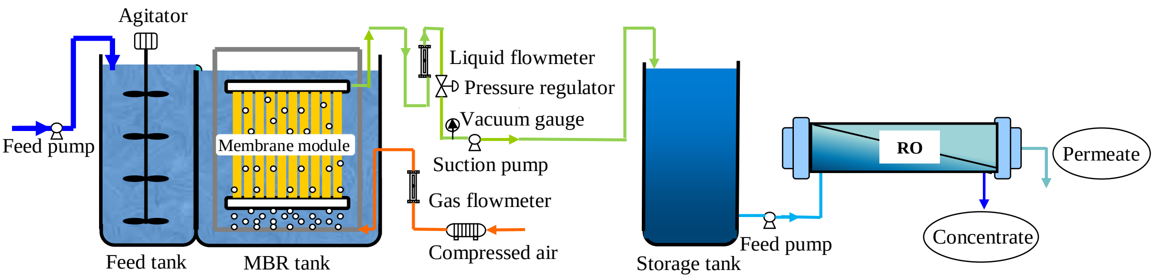


Fig. 2 The flow scheme of membrane integrated process for spunlace wastewater treatment

fabric was used as the coating support with an effective membrane area of 0.06 m^2. Two pieces of PPTA/PVDF flat sheet membranes with one isolated layer in the middle were bonded on the ultrasonic welding machine. Then, the two ends of membranes were sealed with ABS shell using curing glue and the curtain-type membrane modules were obtained. Three modules were installed on the stainless steel frame in MBR system.

2.5 Membrane integrated process for spunlace wastewater treatment

The membrane integrated process including PPTA/PVDF blend membrane and reverse osmosis (RO) membrane technology was used to treat the spunlace nonwoven wastewater as illustrated in Fig. 2. The aerator in MBR system replaced the traditional flotation process to some extent. The spunlace wastewater pre-filtrated by PP coarse filter flew into the feed tank and then overflew into the MBR tank where the curtain-type membrane modules were well placed. Underneath the aerator, dense air could easily and uniformly flow upwards. The wastewater treated by PPTA/PVDF blend membranes was drained into membrane collecting pipe through the suction pump and metered via the liquid flowmeter and then flew into the storage tank. The production water of blend membranes was used as the feed water of RO membrane (type 1812, vontron Technology CO. LTD.)

PPTA/PVDF blend membrane modules in MBR system and RO membrane modules were washed using physically backflushing method and forward flushing method, respectively. These two kinds of membrane modules were cleaned every 15 days.

2.6 Characterization of membrane performance and structure parameter

Membrane porosity (ε) was evaluated via the ratio of pore volume to membrane geometrical volume. Membrane samples were immersed in pure water for at least 12 h. Then, the samples were taken out and drained off water with filter papers. Afterwards, membrane samples were dried until a constant mass could be obtained. Five samples at least of each kind of membrane were tested to get an average value.

$$\varepsilon = \frac{(W_w - W_d)}{Al\rho} \times 100\% \quad (1)$$

where W_w and W_d were the weights of wet and dry membranes (g), respectively. A, l, and ρ.

Membrane surface hydrophilicity was characterized through the water contact angle (WCA) measurement using a Kruss Instrument (CM3250-DS3210, Germany) at ambient temperature. Five samples at least of each kind of membrane were tested to get an average value.

2.7 Characterization of membrane water flux and rejection

The permeation tests of membranes were carried out on line through the data records of liquid flow meter. The operating pressure is 0.1 MPa. In order to ensure a reliable data, each data was recorded when a steady flux was obtained. Water flux was calculated through the following equation.

$$J = \frac{V}{At} \quad (2)$$

where F is the water permeate flux ($L \cdot m^{-2} \cdot h^{-1}$). V is the water permeation volume (L). A is the effective membrane area (m^2) and t is the filtration time (h).

In the first 30 days of the test, the water flux data was record once a day, and then once every 2 days. In order to facilitate the comparative analysis, the experiment used the relative water flux (F_R) to characterize the change of water flux over time. It could be calculated by Eq. (3).

$$F_R = \frac{F_t}{F_0} \times 100\% \quad (3)$$

where F_0 and F_t were the virgin permeate flux at the beginning and the permeate flux during filtration, respectively.

The PEG (100 kDa) concentration in the feed and the permeate solution was tested via a UV-vis spectrophotometer (TU-1901, Purkinje General Instrument Co. Ltd., China) at a wavelength of 515 nm. The rejection (R) is calculated as

$$R = 1 - \frac{C_p}{C_f} \times 100\% \quad (4)$$

where C_p and C_f are the concentration of PEG in the permeate and the feed solution, respectively.

2.8 Measurement of water quality index

Chemical oxygen demand (COD) was tested using the COD measuring instrument (COD-571 type Shanghai Leici Co. Ltd. China) according to the GB/T 11914-1989. Suspended substance (SS) content was measured using gravimetric method according to GB/T 11901-1989. Ammonia nitrogen (AD) content was tested according to GB/T 11901-1989 using spectrophotometer (721S type, Shanghai Instrument Electric Co. Ltd. China). The pH value were measured according to GB/T 6920-1986. The water salinity was characterized by electrical conductivity using the conductivity meter (DDS-307A, Shanghai Leici Co. Ltd. China).

3. Results and discussion

3.1 Membrane performance and structure parameters

Membrane performance and structure parameters were listed in Table 1. It could be seen that with PPTA/PVDF blend membrane had a smaller water contact angle (WCA) value than that of PVDF membrane which suggested the blend membrane surface was more hydrophilic. The introduction of PPTA in PVDF membrane matrix increased the porosity and pure water flux of PVDF membrane. Membrane PEG-100 kDa rejection showed a slight decline which was still above 95%. These results suggested the incorporation of PPTA could effectively increase the number of membrane micropores and thus enhance membrane permeating water flux as shown in Fig. 3.

Table 1 Performance parameters of PVDF/PPTA blend membrane

Membrane sample	Porosity (%)	Water contact angle (°)	Pure water flux ($L \cdot m^{-2} \cdot h^{-1}$)	PEG-100 kDa rejection (%)
PVDF	64.2±1.0	82.1±0.8	61.5±1.1	99.2±0.5
PPTA/PVDF	69±0.8	45.6±0.5	89.2±0.9	96.5±0.6

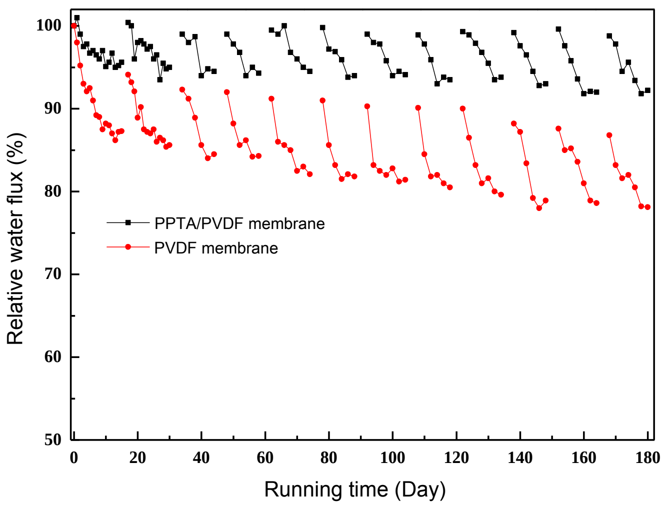


Fig. 3 The relationship between the relative water flux and the running time of different membranes

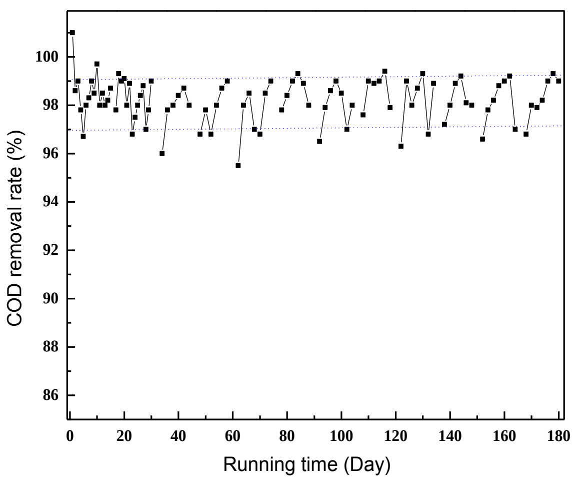
3.2 The relative water flux of different membranes

Fig. 3 showed the variation of relative water flux of different membranes with the running time. The initial water flux of PVDF and PPTA/PVDF blend membranes is 58 and 86 $L \cdot m^{-2} \cdot h^{-1}$ at 0.1 MPa. It could be seen from Fig. 3 that both the relative water flux of PVDF and blend membranes showed a decreasing trend in the filtration cycle of 15 days. The flux data of these two kinds of membranes were kept above 75% and 90%, respectively. By comparison, the relative water flux of blend membrane during the whole measurement process was higher than that of PVDF membrane which suggested PPTA/PVDF membrane had improved antifouling properties. Throughout the running time of 6 months, the water flux of blend membrane was generally maintained between 93-100%. After each backwashing, membrane relative water flux could be recovered to approximately 100%. These results indicated that the prepared blend membrane suffered the unavoidable fouling in the long-term operation. This novel modified PVDF membrane had good antifouling properties.

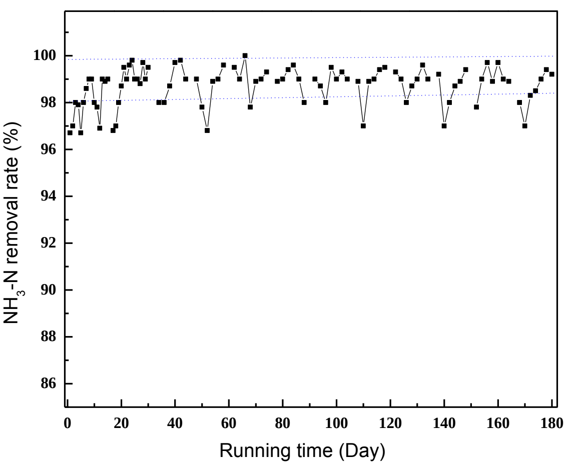
3.3 The quality index of permeate water filtrated by PPTA/PVDF blend membrane

Fig. 4 showed the relationship of different water quality index (including COD removal rate, ammonia nitrogen (NH_3-N) removal rate and suspended substance (SS) removal rate) with running time. As described in the introduction section, the wastewater of spunlace nonwoven process could contain the following components: fiber oil agent, all kinds of short fiber chips and the dissolved short fluff under high pressure, microorganism and inorganic matters, dust and other substances. Some fiber oil agents composed of complex compositions, microorganism and the dissolved short fluff under high pressure were the main constituent material of COD and ammonia nitrogen in spunlace wastewater.

It could be seen from Fig. 4(a) and (b) that the removal rate of COD and NH_3-N remained at 97-99% and nearly 100%, respectively. These results indicated that PPTA/PVDF blend membrane with a pore size in the range of ultrafiltration (UF) membrane can effectively remove various

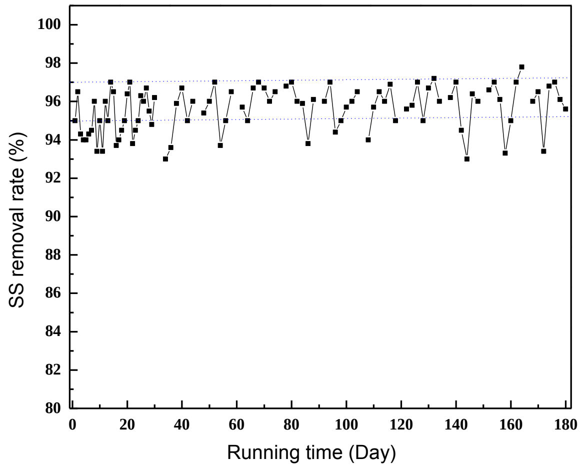


(a) COD removal rate



(b) NH_3-N removal rate

Fig. 4 The different quality index of permeate water filtrated by PPTA/PVDF blend membrane



(c) SS removal rate

Fig. 4 Continued

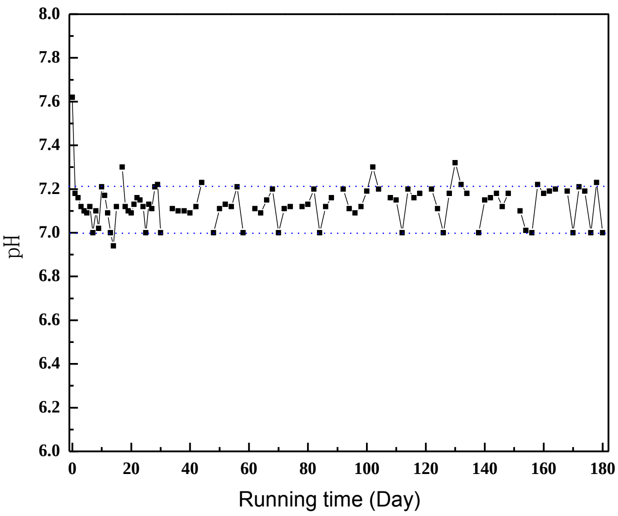


Fig. 5 The variation of pH of permeate water filtrated by PPTA/PVDF blend membrane

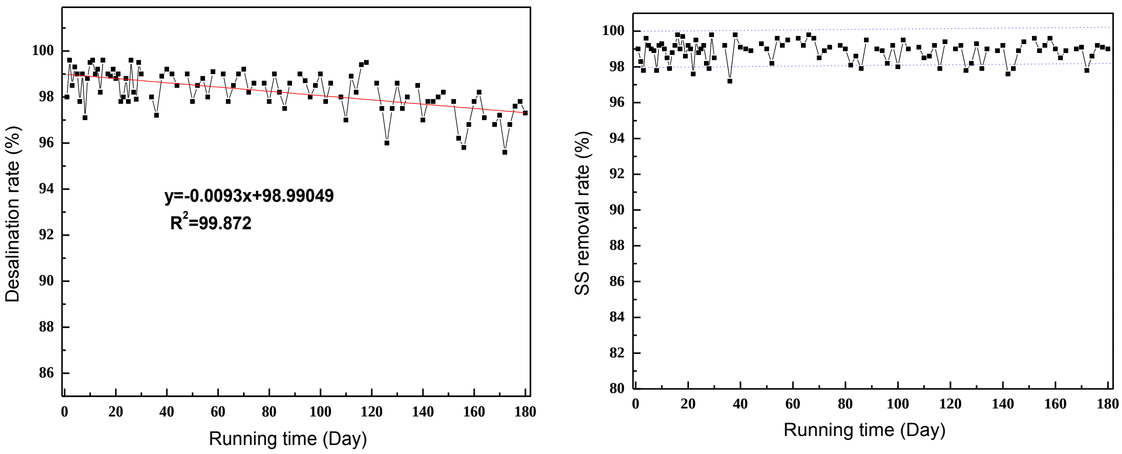
organic matters and microorganisms in spunlace wastewater. Besides, some insoluble matters such as inorganic compounds, organic compounds, dust and microorganism in water can also reach a higher removal after blend membrane filtration. As shown in Fig. 4(c), blend membrane exhibited a high removal rate of suspended substance (SS) between 95-97%.

Fig. 5 showed the variation of pH value of the water filtrated by PPTA/PVDF blend membrane. It could be seen that pH value of the original wastewater was slightly alkaline with the pH value of 7.60. This may be due to the use of cationic surfactants in fiber spinning process. After blend membrane treatment, the pH value decreased to about 7.10, which was close to neutral. These results suggested that blend membrane has a certain favorable function on the complex alkaline substances. It could be found that after the RO membrane filtration as described below, the pH

Table 2 Water quality index after the treatment of PPTA/PVDF blend membrane

Quality index	COD (mg/L)	NH_3-N (mg/L)	SS (mg/L)	pH
Limit value of national first level	100	15	70	6-9
Spunlace wastewater	57.8	7.84	37	7.60
Production water	N. D.*	0.159	7	7.19

*Not detected



(a) Desalination rate (b) SS removal rate

Fig. 6 The variation of desalination rate and the SS removal rate with running time of RO membrane

value of the production water in basically maintained between 7.0-7.1 which was close to neutral.

Combined with Figs. 4 and 5, a comprehensive conclusion can be obtained that the developed PPTA/PVDF blend membrane showed a high and stable removal rate of COD, ammonia nitrogen and suspended substance, and other pollutants in spunlaced wastewater. The quality index (pH value, SS, COD and NH_3-N) of the permeate water treated by the PPTA/PVDF blend membrane exceed the national first level of discharging standard (8978-1996 GB) (see Table 2).

3.4 The quality index of permeate water filtrated by RO membrane

Nonwoven fabric surface had more or less fiber auxiliary such as surfactants, softening agent and brightener. These substances probably contained inorganic material. The deposition of these inorganic compounds especially calcium and magnesium ions will scale and block the pinhole during the long-term production and hence induce adverse effect on the product quality of spunlace nonwoven fabric.

In addition, the prepared PPTA/PVDF blend UF membrane had no selectivity to inorganic salts. Therefore, it should be combined with reverse osmosis (RO) membrane for further treatment. It can be believed that the integrated process of UF MBR system and RO membrane technique could more effectively purify the spunlace wastewater. The inorganic salts and the residual suspended substance after the treatment of PPTA/PVDF blend membrane can be further removed by the RO membrane so as to fully achieve the water recycling use requirements of spunlace process.

Fig. 6 showed the change of desalination rate of inorganic salts and the removal rate of suspended substances with running time of RO membrane. It could be seen that the desalination rate of RO membrane and the removal rate of suspended substance in the water during the whole running time maintained at between 97-99% and about 99%, respectively. During the whole testing process, the desalination rate had a slight decrease. This was due to the negative charge surface of RO membrane used in this study. The excess inorganic salt cations were prone to adsorb on the surface of the RO membrane, which led to the weaken of the Donnan and the steric effects. Therefore, a small decrease of desalination rate of RO membrane emerged (Chan *et al.* 2016, Shang *et al.* 2014).

These results also indicated that after running a certain period of time, the simple physical washing can not effectively recover the desalination rate of RO membrane and the off-line chemical cleaning was necessary. After the filtration of PPTA/PVDF blend membrane, the content of suspended substance was very low. However, its removal rate can still reach 99% after the RO membrane deep filtration and its concentration remained in water reached the lowest. In summary, after the deep filtration of RO membrane, the water quality can meet the requirements of spunlace process water recycling.

4. Conclusions

PPTA/PVDF blend membrane was prepared via the *in situ* polymerization followed by the immersion precipitation phase inversion method. The integrated membrane technology including PPTA/PVDF blend membrane and reverse osmosis (RO) membrane was used to treat the polyester/viscose spunlace nonwoven process wastewater. During the consecutive running of six months, the high relative water flux indicated PPTA/PVDF blend membrane had good antifouling properties. The prepared blend membrane suffered the unavoidable fouling in the long-term operation. The removal rate of COD, ammonia nitrogen and suspended substance filtered by PPTA/PVDF blend membrane was always above 90 %. The deep treatment of RO membrane can further purify the water quality to meet the water circulation requirement of spunlaced process.

Acknowledgments

The authors gratefully acknowledge the funding for the project supported by the National Natural Science Foundation of China (No. 51403052), the Science and Technology Development Project of Zhengzhou (No. 153PKJGG134), the Open Fund Program of Henan Engineering Laboratory of New Textiles Development (No. GCSYS201603), the Doctoral Fund of Henan Institute of Engineering (No. D2015027), the Key Research Project of Higher Education of Henan Province (No. 18A540001) and the Program for Innovative Research Team (in Science and Technology) in University of Henan Province (13IRTSTHN024/15IRTSTHN011).

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