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Study of supernatant of SBR in dead-end microfiltration

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ABSTRACT

The activated sludge supernatant of sequencing batch reactor was filtrated in dead-end mode by a 0.1 μm microfiltration hydrophilic polyvinylidene fluoride (PVDF) hollow-fiber membrane module, and a multiregression model was used to study the influence of operating conditions on flux of the membrane module and the effluent quality (TOC and NH_3-N) quantificationally. The impact of the following seven parameters—(1) transmembrane pressure (TMP), (2) the sludge concentration of the mixed liquor suspended solids (MLSS), (3) dissolved oxygen concentration (DO), (4) hydraulic retention time (HRT), (5) sludge retention time (SRT), (6) temperature (T), and (7) pH value (pH) on the membrane permeate flux (J) and water quality (TOC, NH_3-N)—were investigated. The results show that: (i) The effluent quality from this microfiltration membrane module could meet the quality standard for reused wastewater issued by the Chinese Ministry of Construction (CJ25.1–89); (ii) TMP, T and pH are the influence factors of the membrane permeate flux and the relative influence degree are 50.71, 45.08 and 4.21%, respectively. The mathematic expression obtained by the multivariate linear regression model between the membrane permeate flux and operating conditions was: $J = 3.35 \times 10^{-5} - 1.3 \times 10^{-7}T - 3.4 \times 10^{-6}\mathrm{pH} + 1.1 \times 10^{-4}\mathrm{TMP}$; (iii) TMP has no effect on TOC and NH_3-N, the relative influence degree of other operational conditions on TOC as following: MLSS (49.36%) > pH (32.52%) > HRT (9.87%) > SRT (4.60%) > DO (2.21%) > T (1.44%), and the relative influence degree of them on NH_3-N as following: MLSS (61.39%) > pH (21.38%) > HRT (8.82%) > SRT (3.51%) > T (4.11%) > DO (0.79%): The relationship between TOC and operational conditions was the following: $\mathrm{TOC} = -394.553 - 48.38\mathrm{MISS} + 2.81\mathrm{T} - 13.79\mathrm{DO} + 106.52\mathrm{pH} - 0.60\mathrm{SRT} - 11.12\mathrm{HRT}$. The relationship between NH_3-N and operational condition was: $NH_3\text{-N} = 390.648 + 63.84\mathrm{MISS} - 5.61T + 9.73\mathrm{DO} - 102.20\mathrm{pH} + 0.62\mathrm{SRT} + 12.44\mathrm{HRT}$.

Keywords: SBR; Supernatant; Hollow-fiber microfiltration membrane; Linear multi-regression

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1. Introduction

Nowadays, the water shortage has become a worldwide problem. Water pollution caused by industrial wastewater and domestic sewage has seriously effect on the economic development, ecological environment, and human health, and it becomes more strained on water resources utilization. In China, during the last three decades, with the deterioration of water ecological environment and the growing water pollution, the water shortage and inadequate supply have had a tremendous impact on the rapid development of economy and society. It is imperative to treat sewage and make it resources. It has been widely used in sewage and industrial wastewater treatment by traditional activated sludge treatment process thatcan remove dissolved and colloidal, biodegradable organic matter effectively [1,2].

However, the traditional activated sludge treatment technology is not ideal and not stable enough on high-quality water production. In recent years, the membrane bioreactor (MBR) as a new technology for wastewater treatment has opened up a new direction for traditional water treatment technology by adding membrane separation technology. Compared with the traditional activated sludge process, it has many advantages, such as a small footprint, high-quality effluent, low sludge production rate, a highly retentive activated sludge concentration, and easy management due to the combination of biological treatment with membrane separation [3,4]. This technology saves the secondary settling tank process and improves the system ability of solid-liquid separation significantly. Finally, it has been greatly improved on water quality and volume load of the system.

Although MBR is widely used in industrial wastewater and domestic sewage treatment process [5], at current period in China, the price of the product water by MBR technology is higher than that by the traditional activated sludge. The wastewater is still cotreated by the combined traditional activated sludge process with other treatment methods. Therefore, it is very excellent method to treat the supernatant directly in the traditional activated sludge process deeply for different requirements. There are many ways for this purpose, and one of the simple and practical ways is the supernatant is filtered directly by the membrane to separate and remove bacteria or some organic matter.

In industrial process, the performance of the MBR is affected by membrane property, membrane module structure, operating condition, and bioreactor parameter. Nevertheless, under certain membranes and membrane modules, the flux of the membrane and MBR performance are importantly different according to operating conditions and process conditions. Therefore, it is very significant to optimize operating conditions for the high-quality water. Some researchers had carried out studies with regard to operating condition. For example, Kenji Kawasakiet et al. [6] investigated the effect of initial MLSS of submerged membrane activated sludge process. It was found that the best moderate initial MLSS was about 3,000–5,000 $mg\,L^{-1}$ for a long-term stable operation, and low initial MLSS caused organic matter unmetabolized completely while high initial MLSS might decrease the efficiency of treatment system [7]. Paula van den Brink et al. [8] analyzed the effect of temperature shocks on membrane fouling in MBRs. Sludge flocs could release polysaccharides and/or submicron particles at low temperatures and then it would increase membrane fouling at actual operation of MBR. DO concentration was one of the critical operating parameters and the two air flow rates of 0.15 and 2.33 $L\,min^{-1}$ leading to the different DO concentrations was investigated [2]. Compared with high DO concentration, the activated sludge with low DO concentration exhibited a significant flux decline. Moreover, the flux variations and resistances were analyzed under different sludge retention times (SRT) [9]. Different SRT conditions caused the variation of sludge condition and particle size distribution in the supernatants, which displayed dissimilar fouling characteristics. In our previous study, the influences of operating conditions on accumulated volume of membrane permeate and effluent qualities (COD and NH_3-N concentrations) were discussed [10]. However, during these previous studies, the effects of operating conditions were analyzed qualitatively or quantitatively, unconsidering the interrelation between target factors and operating conditions. Therefore, it is very significant to quantify and study the influence of operating conditions on wastewater treatment performance, and this can provide the based guidance for process optimization or modeling of MBR. The mathematic relationships were obtained by multivariate linear regression method and these could possibly be used to optimize and predict the efficiency of wastewater treatment in bioreactor parameters. However, the characteristics of supernatant are different under different process conditions and the supernatant requires to study in depth.

In this study, the hollow fiber microfiltration membrane is used to separate of the supernatant of sequencing batch reactor (SBR) in dead-end filtration, and the effluent water quality is surveyed. Moreover, the quantitative relationship between flux of the membrane and operating conditions are investigated. The goal is to discover the role of water quality and process conditions in order to optimize membrane filtration.

2. Materials and methods

2.1. Materials

A hydrophilic polyvinylidene fluoride (PVDF) hollow-fiber microfiltration membrane module with a nominal pore size of 0.1 μm was purchased from the Tianjin Motian Membrane Separation Eng. and Tech. Co. (Tianjin, China). The feed solution (raw waste water) was obtained from the storage tank of domestic sewage with quality shown in Table 1. Different operating parameters of the bioreactors, such as MLSS, *T*, DO, pH, SRT and HRT, are shown in Table 2, in which the values of process parameters are measured during the experiments.

2.2. Experimental setup and conditions

The laboratory-scale experimental system consisted of two parts: the first part is seven parallel intermittent mode bioreactor systems with each effective volume of 25 L (shown as in Fig. 1). Raw wastewater is stored in tank and updated every day. Aeration was done through an air compressor installed directly beneath the bioreactor for transferring oxygen to microorganisms. The second part is a hollow fiber dead-end microfiltration cell with an effective filtration area of 0.2 m^2 (shown as in Fig. 2). The permeate weight is measured during filtration process with an electronic balance (accuracy of 0.1 g). The weights are converted to volumes using density correlations. The temperature is controlled at 25 ± 1 ℃.

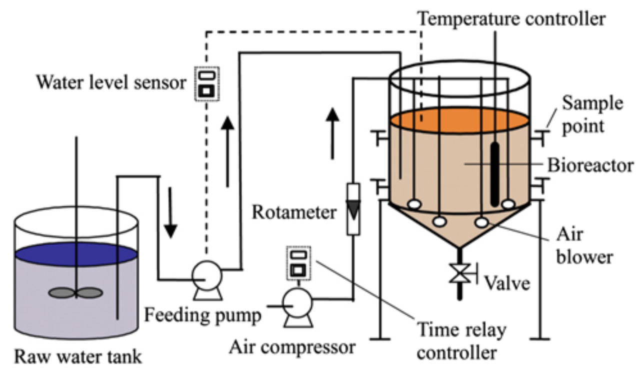

Fig. 1. Schematic diagram of the intermittent mode bioreactor system.

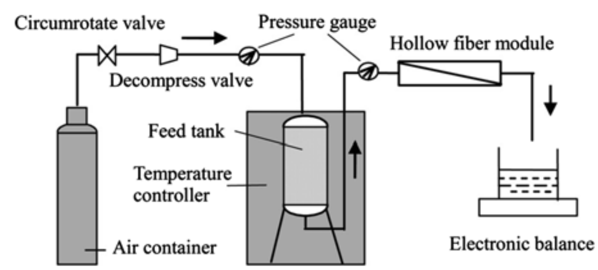

Fig. 2. Schematic of the filtration set-up.

Table 1
Quality of raw wastewater used in the experiment

COD (mg L^{-1})	180.6–225.8
NH_3-N (mg L^{-1})	45.9–73.6
TOC (mg L^{-1})	86.5–115.5
pH	7.5–8.0
Turbidity (NTU)	20–26

Table 2
Operating conditions of different bioreactors

Reactors	MLSS (g L^{-1})	*T* (℃)	DO (mg L^{-1})	pH	SRT (d)	HRT (h)
1	1.3	15	3	7.5	100	24
2	2.2	20	4	7.0	60	18
3	3.2	25	5.5	8.0	200	15
4	6.6	30	5	8.5	30	13
5	8.3	35	7.5	9.0	15	10
6	5.0	21	7	9.2	70	20
7	7.5	33	8	9.5	110	21

2.3. Analytical items and methods

The MLSS concentration and pH were measured by using a weight method and pHS-3C acidity meter, respectively. Total organic carbon (TOC) was measured using a TOC analyzer (TOC–V_{CPH}, Shimadzu, Japan). The values of NH_3-N, as the items of the influent and membrane effluent, were measured by adopting the Chinese SEPA standard methods [11]. The membrane permeability of the mixed liquor in the bioreactor was investigated using a hollow fiber module. Mixed liquor samples of about 2.5 L were taken from the different bioreactors, and experimental data were recorded in the interval of 15 s for 15 min.

2.4. Analytical method

In order to study influencing degree of operating conditions on target object, a multivariate linear regression model is used. The basic multivariate linear regression model is described as follows,

$$BY_k = \sum_{j=1}^{m} b_j BX_j \quad (1)$$

where b_j is regression coefficients of process conditions or operating conditions to water quality or flux of the membrane. $j = 1, 2, 3, \ldots, m$ is the number of impact factors.

If the regression coefficient divides the relative standard error, the F_j is obtained by:

$$F_j = b_j / \sigma_j \quad (2)$$

where σ is the relative standard error. If $|F_j|>1$, the factor is considered as an influencing factor, and the bigger the absolute value of F_j is, the larger the effect of factor on target object is. Otherwise, the factor has no effect on model parameter or there is a very small influence.

In addition, when all of b_j is influencing factors, the effect can be expressed quantificationlly by percentage as follows:

$$D_j = \left(F_j^2 \Big/ \sum_{j=1}^{m} F_j^2 \right) \times 100\% \quad (3)$$

Under the assumption that all of b_j is the influential factors, according to the experimental data of process conditions and operating conditions for the flux of the membrane and water quality, the multi-linear regression model was designed as follows:

$$Y_k = d_0 + \sum_{j=1}^{m} d_j X_j \quad (4)$$

where d_0 is constant term, d_j is the regression coefficient of the operating conditions or process conditions.

3. Results and discussion

3.1. Analysis of water quality indicators

In order to evaluate the performance of bioreactor and membrane filtration, TOC and NH_3-N contents of raw water, supernatant and membrane permeate were measured for seven reactors, and the results were shown in Figs. 3 and 4.

Fig. 3 shows that most of the raw water of TOC has been removed after activated sludge treatment in 1–3 reactors. However, 4–6 reactors show high contents after activated sludge treatment. It is because that there are some shortages of the traditional activated sludge treatment technology, such as, sludge with deep color, instability of activated sludge, difficult separation of sludge and liquid, and finally the water quality is so poor that it is not reuse directly. It can be seen from Fig. 4, the NH_3-N of raw water has been removed largely through the traditional activated sludge decomposition. At the same time, the NH_3-N of supernatant is instability of different reactor. But after membrane filtration, it has satisfied the standards of water reuse basically (CJ25.1-89). Therefore, it can play an effective protection of water quality by applying membrane technology.

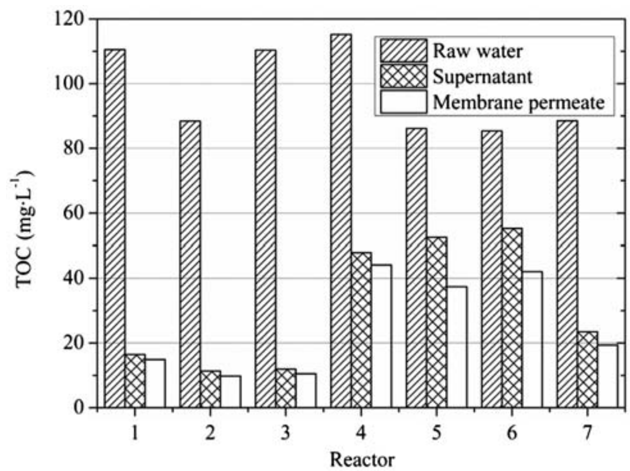

Fig. 3. Comparison of TOC concentration of raw water, supernatant and membrane permeate.

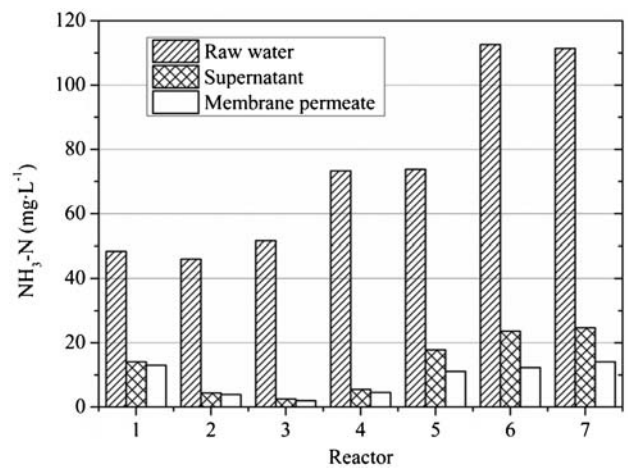

Fig. 4. Comparison of NH_3-N concentration raw water, supernatant and membrane permeate.

3.2. Effect of operating conditions on flux of the membrane

The flux of the membrane decline caused by membrane fouling in the microfiltration process is an

Table 3
Values of regression coefficient, F_j and D_j for flux of the membrane

j	1	2	3
b_j	−0.1685	−0.5513	0.5847
σ_j	0.1081	0.1081	0.0787
F_j	−1.558	−5.098	7.426
D_j (%)	4.210	45.08	50.71

inevitable phenomenon in the membrane filtration process, which will lead to increase in the cost of process operation. Operating conditions (TMP, T, MISS and pH) have significant impacts on flux of the membrane. In actual operation, it not only can reduce the membrane fouling and maintain a high flux of the membrane, but also can reduce unnecessary energy consumption under the optimized operating conditions. Therefore, the study of the impact of operating conditions on flux of the membrane has important practical significance.

In order to compare the influence degree of operating conditions on flux of the membrane, the experimental data were analyzed using multiple linear regression method of Eqs. (1)–(4). The flux of the membrane for a certain temperature and pH were in the TMP range of 0.03–0.15 MPa, and the results were showed in Table 3. It can be seen from Table 3 the absolute values of F_1 (T), F_2 (pH) and F_3 (TMP) exceed one, and so these factors can be considered as the impact factors with significant roles on flux of the membrane. The sequence of effect is TMP (50.71%) > pH (45.08%) > T(4.21%). In addition, analysis of variance was showed in Table 4. Comparison of $|F_1| < t_{0.05}(31)$ and $F > F_{0.05}(3.32)$, the temperature is not significantly impact factors. It is obvious that the flux of the membrane is found to increase with increasing pressure, and it will rise to a critical value due to forming a dense deposition layer [12]. Moreover, it is causes the different ionization states on the membrane surface under different pH, and then the adsorbance on membrane surface is also changed as different pH causing a different structural fouling layer [13]. So, TMP and pH have serious impacts on flux of the membrane.

Table 4
Analysis of variance

n	35
m	3
$t_{0.05}(31)$	1.6955
F	41.75
$F_{0.05}(3.32)$	2.904

According to Eq. (4), the quantitative relationship between the flux of the membrane and operating conditions (confidence $\alpha = 0.05$):

$$J = 3.35 \times 10^{-5} - 1.3 \times 10^{-7}T - 3.4 \times 10^{-6}\mathrm{pH} + 1.1 \times 10^{-4}\mathrm{TMP} \tag{5}$$

where J is the flux of the membrane, $m^3/(m^2\ s)$; TMP is the transmembrane pressure, MPa; T is the temperature, ℃ and pH is the pH value.

Eq. (5) shows that J increases with TMP increasing. In contrast, it decreases with the increase of pH and T. From Table 2, the process conditions of pH and T increases in bioreactors, and the concentration of the sludge is rising. The increasing sludge concentration will cause the concentration polarization, concentration boundary layer increase, and thus the flux of the membrane will continue to decline. However, in actual operation, it is unfavorable to increase TMP for the increase of the flux of the membrane. The significance guiding of this equation Eq. (5) is limited to the actual operation conditions.

3.3. Influence of processing conditions on the water quality

3.3.1. Influence of TOC

The experimental data were calculated between TOC and process conditions by regression analysis as shown in Table 5. The regression results shows that the regression value of TMP is much less than one for the first regression and this illustrates TMP have a tiny effect on the TOC removal. Therefore, the rest of the process condition data without TMP were analyzed again. The sequence of influence degree of various process conditions on TOC is: MISS (49.36%) > pH (32.52%) > HRT (9.87%) > SRT (4.60%) > DO (2.21%) > T (1.44%). In addition, Analysis of variance shows in Table 6, $|F_2| < t_{0.05}(28)$, the temperature is not a significant influencing factors.

According to the Eq. (4), the quantitative relationship (confidence $\alpha = 0.05$) between the TOC and the operating conditions as follows:

$$\mathrm{TOC} = -394.553 - 48.38\mathrm{MISS} + 2.81T - 13.79\mathrm{DO} + 106.52\mathrm{pH} - 0.60\mathrm{SRT} - 11.12\mathrm{HRT} \tag{6}$$

where MISS is the sludge concentration of the mixed liquor suspended solids, $g\,L^{-1}$; T is the temperature, ℃; DO is dissolved oxygen concentration, $mg\,L^{-1}$; pH is the pH value; SRT is sludge retention time, d; and HRT is the hydraulic retention time, h.

Table 5
Values of regression coefficient, F_j and D_j for TOC

Regression time	j	b_j	σ_j	F_j	D_j (%)
1	1	−7.691	3.234	−2.378	–
	2	1.314	0.831	1.581	–
	3	−1.629	0.382	−4.267	–
	4	6.243	1.963	3.180	–
	5	−2.348	0.819	−2.867	–
	6	−3.439	1.164	−2.954	–
	7	0.002	0.052	0.030	–
2	1	−7.691	3.178	−2.420	49.36
	2	1.314	0.817	1.609	1.440
	3	−1.629	0.375	−4.343	2.210
	4	6.243	1.929	3.236	32.52
	5	−2.348	0.805	−2.918	4.600
	6	−3.439	1.144	−3.007	9.870
	7	–	–	–	–

It can be seen from Eq. (6) MISS, DO, SRT, and HRT have positive influence on TOC removal; however, T and pH have negative effects on it within the scope of the experiment. MISS is an important parameter of the organic removal efficiency. It is because increasing sludge concentration can increase the number of microorganisms in a certain concentration range, and the more organic matter is metabolized, the better the TOC is removed. Low DO contents will lead to microbial aerobic inadequate; SRT is the average residence time of microorganisms in the system; with the increase of HRT, the average organic removal of the reactor increased gradually; the high temperature will be inhibit microbial metabolism; pH will affect the part of the organic matter removal owing to the activity of biological species.

3.3.2. Influence of NH_3-N

The experimental data were calculated between NH_3-N and process conditions by regression analysis shown in Table 7. Comparing the value F_j, the regression value of TMP is much less than one for the first time, and TMP have a tiny effect on NH_3-N removal. Therefore, the rest of the process condition data without TMP were analyzed again. Thus, MISS and pH have a larger impact on NH_3-N removal, other factors smaller. The sequence of influence degree of various process conditions on NH_3-N is: MISS (61.39%) > pH (21.38%) > HRT (8.82%) > T (4.11%) > SRT (3.51%) > DO (0.79%). In addition, Analysis of variance shows in Table 6. $|F| > F_{0.05}(28)$, the factors are significant influencing factors with a significant regression effect.

Table 6
Analysis of variance

n	35
m	6
$t_{0.05}(28)$	1.7011
F	57.41
$F_{0.05}(6.29)$	2.43

Table 7
Values of regression coefficient, F_j and D_j for NH_3-N

Regression time	j	b_j	σ_j	F_j	D_j (%)
1	1	32.44	1.0020	32.36	–
	2	−8.389	0.2577	−32.56	–
	3	3.675	0.1183	31.06	–
	4	−19.14	0.6086	−31.46	–
	5	7.762	0.2538	30.58	–
	6	12.30	0.3608	34.09	–
	7	0.007	0.0161	0.453	–
2	1	32.44	0.9886	32.82	61.39
	2	−8.389	0.2541	−33.01	4.110
	3	3.675	0.1167	31.50	0.790
	4	−19.14	0.6002	−31.90	21.38
	5	7.762	0.2503	31.01	3.510
	6	12.30	0.3558	34.56	8.820
	7	–	–	–	–

According to the Eq. (4), the quantitative relationship (confidence $\alpha = 0.05$) between the NH_3-N and the process conditions as follows:

$$NH_3\text{-N} = 390.648 + 63.84\text{MISS} - 5.61T + 9.73\text{DO} - 102.20\text{pH} + 0.62\text{SRT} + 12.44\text{HRT} \quad (7)$$

where MISS is the sludge concentration of the mixed liquor suspended solids, $g\,L^{-1}$; T is the temperature, ℃; DO is dissolved oxygen concentration, $mg\,L^{-1}$; H is the pH value; SRT is sludge retention time, d; and HRT is the hydraulic retention time, h.

It can be seen from Eq. (7) MISS, DO, SRT, and HRT have negative influence on NH_3-N removal. However, T and pH have positive effects on it within the scope of the experiment. MISS and pH have the opposite effect on the TOC and NH_3-N removal rate. A proper MISS and pH contribute to obtain high-quality water of TOC and NH_3-N removal. SRT has a lesser impact on the metabolic activity of sludge, while it is found that the opposite trend with other literature reported [14]. However, NH_3-N is removed by activated sludge treatment and membrane separation technology.

4. Conclusions

In this study, the 0.1 μm PVDF hollow fiber microfiltration membrane is used to separate of the supernatant of SBR in dead-end filtration. The quantitative relationships between flux of the membrane and operating conditions (TMP, *T* and pH) and between water quality and process conditions (MISS, TMP, *T*, DO, pH, SRT and HRT) were obtained using multiple linear regressions method. The results show that:

(1) After the supernatant of SBR by microfiltration, the water quality achieves to the standards for water reuse (CJ25.1–89).
(2) TMP and pH have significant effect on the flux of the membrane. *T* also affects the flux of the membrane, but not a significant factor. The sequence of influence degree is: TMP (50.71%), pH (45.08%) and *T* (4.21%). The quantitative relationship between the flux of membrane and operating conditions is: $J = 3.35 \times 10^{-5} - 1.3 \times 10^{-7}T - 3.4 \times 10^{-6}\text{pH} + 1.1 \times 10^{-4}\text{TMP}$.
(3) MISS, pH, HRT, SRT, DO are influence factors on TOC and NH_3-N, while TMP has no effect on water quality indicators. MISS and pH are significant influence factors, and the others have relatively small effects on it. The sequence of influence degree of various process conditions on TOC is: MISS (49.36%) > pH (32.52%) > HRT (9.87%) > SRT (4.60%) > DO (2.21%) > *T* (1.44%). The sequence of influence degree of various process conditions on NH_3-N is: MISS (61.39%) > pH (21.38%) > HRT (8.82%) > *T* (4.11%) > SRT (3.51%) > DO (0.79%). The quantitative relationship between TOC and NH_3-N and the process conditions as follows:

$$\begin{aligned}\text{TOC} = {} & -394.553 - 48.38\text{MISS} + 2.81T - 13.79\text{DO} \\ & + 106.52\text{pH} - 0.60\text{SRT} - 11.12\text{HRT};\end{aligned}$$

$$\begin{aligned}\text{NH}_3\text{-N} = {} & 390.648 + 63.84\text{MISS} - 5.61T + 9.73\text{DO} \\ & - 102.20\text{pH} + 0.62\text{SRT} + 12.44\text{HRT}\end{aligned}$$

These relationships have significant functions that can clearly express the relationship between the parameters and each factor and it can be used to achieve process optimization and forecasting.

At present, the MBR cannot completely replace traditional wastewater treatment technology. It can use the membrane technology to improve the drawbacks of traditional wastewater treatment methods. In this study, the effluent quality reaches the miscellaneous water standards ultimately using the dead-end microfiltration membrane, and sewage resources can be used sufficiently.

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