

Impact of Operational Condition on the Membrane Permeate Flux and Quality of Domestic Wastewater Treatment using Hollow Fiber Membrane Module in Cross-Flow Mode

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Abstract: In this paper, the impacts of process conditions on the membrane permeate flux (J) and the quality of effluent in domestic wastewater treated by using 0.1 μm hollow fiber membrane module in cross-flow mode were quantitatively analyzed with a statistical method. The results showed that (1) trans-membrane pressure (TMP), cross-flow velocity (u), mixed liquor suspended solids ($MLSS$), dissolved oxygen concentration (DO), pH , temperature (T), sludge retention time (SRT) and hydraulic retention time (HRT) were all the influencing factors of the permeate flux. Among them, $MLSS$, T , SRT and HRT are negative contributors to the permeate flux while TMP , u , DO and pH are positive contributors. In addition, the quantitative relationship between the permeate flux and process conditions is established. (2) TMP and u had no effect on the quality of effluent COD while other operating conditions were the influencing factors. Only HRT had a negative effect on the quality of effluent NH_3-N . The quantitative relationships between COD , TOC , NH_3-N and process conditions were also established. These mathematical expressions, to some extent, could be used to optimize operational conditions, predict the permeate flux and efficiency of domestic wastewater treatment using hollow fiber membrane module in cross flow mode.

Keywords: Membrane bioreactor, Operational condition, Multivariate linear regression.

1. INTRODUCTION

The membrane bioreactor (MBR), which has many advantages in comparison with the conventional wastewater treatment technology [3, 4], is considered as advanced membrane technology for wastewater treatment [1, 2]. In principle, the performance of the MBR is determined by the synergistic effect of the membrane property, membrane module structure, operational condition and bioreactor parameter [5]. However, for specified membrane module, the membrane and membrane module are fixed and the process condition (operation conditions and bioreactor process conditions) plays a critical role in process optimization.

Membrane fouling behavior in a real membrane bioreactor is the result of the synergy of operation conditions and bioreactor process conditions. The bioreactor parameters, such as sludge concentration ($MLSS$) [6-10], dissolved oxygen (DO) [11, 12], pH [13], temperature [13, 14], sludge residence time (SRT) [15-19], hydraulic residence time (HRT) and F/M [20], determine the composition of the main pollutant, such as extracellular polymeric substance (EPS), soluble microbial products (SMP) and colloids in bioreactor

[6, 15-17]. These pollutants directly affect the degree of the membrane fouling in membrane bioreactor [21-24]. Moreover, the operating conditions of the membrane module, such as operating pressure (TMP), velocity (u) [9, 25-26], concentration (C) and temperature (T) are important factors for the operation efficiency of the membrane module. Many researchers have been studying with regard to operational conditions. For example, Jianying *et al.* explored that removal of NH_3-N can be made cost effective by using negative pressure steam-stripping pretreatment, which makes MBR technology more effective [27]. Tardieu *et al.* determined that higher cross-flow velocity improves the membrane fouling [28, 29]. Muhammad *et al.* reported that integration of MBR with hybrid approach could be effective in order to reduce bio-fouling and improve MBR performance [30]. Kui *et al.* investigated the stress-state of the membrane to examine the membrane mechanical responses under similar loading conditions, and presented the advanced mechanical testing approaches [31]. Chin *et al.* explored that MBRs integrated with enzymes can decompose micro pollutants that cannot be decomposed in the simple MBR system by bacteria [32]. Wang *et al.* studied the impacts of operating conditions on the efficiency of domestic wastewater treatment, and devised the mathematic expressions between COD , NH_3-N and operating conditions [5]. Wang *et al.* also respectively studied the influence of operational conditions on the flux [33], specific cake resistance [34], flux changing rate [35] and the filtration behavior [36]. Meanwhile, Bai

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et al. [37] studied the impact of operational conditions on the collected filtrate volume (V) and water quality (COD, $\text{NH}_3\text{-N}$) in re-circulated membrane bioreactor. Gui *et al.* investigated the impacts of operational conditions on the membrane fouling [38]. Junwon *et al.* studied the influence of polyaluminium chloride and chitosan on the removal of $\text{NH}_3\text{-N}$ into the MBR [39]. However, impacts of process condition on the permeate flux and the quality of effluent of domestic wastewater treated by using hollow fiber membrane module in cross flow mode has not been reported yet. The purpose of present paper is to investigate the impacts of operating conditions on the permeate flux and the quality of effluent of domestic wastewater treated by using hollow fiber membrane module in cross-flow mode. The obtained mathematic

expressions could provide the basis for process optimization and modeling in MBR.

2. MATERIALS AND METHODS

2.1. Experimental Set-up and Conditions

As presented in Figure 1, the test system consists of an activated sludge bioreactor with the effective volume of 25 L and 0.1 μm hydrophilic polyvinylidene fluoride (PVDF) hollow fiber microfiltration membrane module (Tianjin MOTIAN Co., Ltd) was used. 11 different bioreactors (Table 1) were used in this experiment.

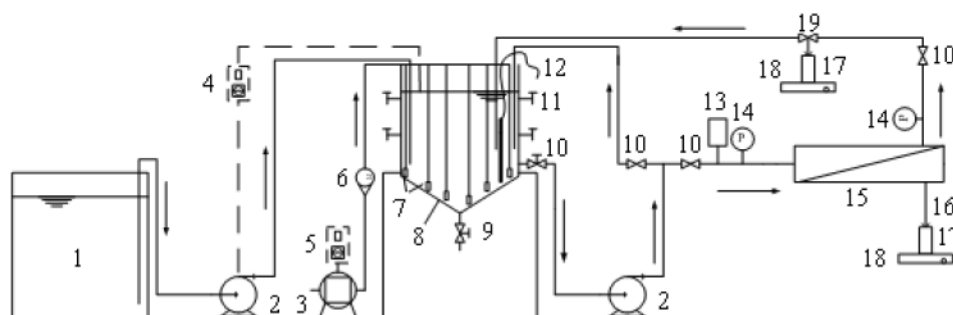


Figure1: Schematic diagram of the experimental system.

1.Raw water tank; 2. Pump; 3. Air compressor; 4. Water level sensor; 5.Timing relay; 6. rotameter; 7. Air blower; 8.MBR; 9.Valve; 10.Valve; 11.Sample point; 12.Heating rod; 13.Buffer tank; 14.Pressure gage;15. Hollow fiber membrane model; 16.Exit; 17 Volumetric cylinder; 18. Electronic balance and 19. Sample point.

Table 1: Operating Conditions, Influencing Factors and Analysis of Experimental Data of 11 Different Bioreactors

Bioreactor	MLSS ($\text{g}\cdot\text{L}^{-1}$)	Temp ($^{\circ}\text{C}$)	pH	DO	SRT	HRT	TMP	u	J	COD	TOC	$\text{NH}_3\text{-N}$
				($\text{mg}\cdot\text{L}^{-1}$)	(d)	(h)	(MPa)	($\text{m}\cdot\text{s}^{-1}$)	($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$)	($\text{mg}\cdot\text{L}^{-1}$)	($\text{mg}\cdot\text{L}^{-1}$)	($\text{mg}\cdot\text{L}^{-1}$)
1	1.8	15	7.5	3	100	24	0.16	0.18	30.4	17.56	7.72	0.66
2	2.7	20	7	4	60	18	0.21	0.04	16.62	16.56	10.01	1.86
3	4.1	25	8	5.5	200	15	0.15	0.28	8.77	23.58	11.75	1.69
4	6.6	30	8.5	5	30	13	0.08	0.23	26.44	32.61	13.81	3.54
5	9.9	35	9	7.5	15	10	0.1	0.15	12.12	39.13	17.36	11.48
6	3	28	6.5	2	50	9	0.06	0.41	63.28	23.08	14.9	56.24
7	5.4	27	7.2	4.5	90	14	0.09	0.35	28.7	34.11	14.16	13.62
8	3.3	31	6.2	2.5	160	11	0.13	0.48	33.6	38.63	10.78	33.16
9	3.5	23	6.7	3.5	80	12	0.07	0.37	54.28	24.08	14.35	53.36
10	1.5	24	6	1.5	180	8	0.12	0.39	35.76	23.08	13.29	70.8
11	5.7	17	7.7	5	120	16	0.17	0.1	10.8	23.08	13.89	1.03

Note : The bioreactor runs for 8 hours (aeration and de-nitrification time is 6 and 2 hours) every day (aeration for 10 minutes and then stop for 2 minutes).

2.2. Experimental Steps

The experimental process includes following steps: (1) inoculate sludge in 11 different bioreactors and run the system to stabilize; (2) activated sludge suspension was filtrated by using filter with 40 mesh to remove impurities, which block the membrane module, then the viscosity and particle size distribution of activated sludge suspensions were determined; (3) cross-flow filtration under different TMPs and cross flow velocities in the membrane module using 11 kinds of activated sludge suspensions was carried out (Figure 2) and corresponding quality of effluent (COD , TOC and NH_3-N) was measured; (4) the effects of TMP , u , $MLSS$, DO , pH , T , SRT , HRT , COD , TOC and NH_3-N on the permeate flux and the quality of effluent were studied by multiple linear regression method, and the quantitative relationship between the operating condition and the permeate flux or the quality of effluent (COD , TOC and NH_3-N) was established. Multiple linear regression method is a statistical linear approach to modeling the relationship between dependent variable and more than one independent variables [40]. So in multivariate linear regression correlated dependent variables are predicted instead of a single variable [41]. In this method, known model parameters are estimated from data by using linear predictor functions to model a relationships between dependent and independent variables [42]. Linear regression was the first type of regression analysis with extensive practical applications [43]. This is due to the fact that models which depend linearly on their unknown parameters are easier to fit as compared to the models which are related to their parameters non-linearly. In case of linear regression statistical properties of the resulting estimators can also be determined easily.

The $MLSS$ concentration and pH were measured by using weight method and pHS-3C acidity meter, respectively. COD , TOC and NH_3-N , as the items of the influent and effluent of the membrane module, were measured by adopting the Chinese SEPA standard methods [45].

2.3. Analytical Model of Influencing Degree of the Process Conditions on the Membrane Permeate Flux or the Quality of Effluent (COD , TOC and NH_3-N)

In order to study the impact of the operating conditions on the membrane flux or COD , TOC and NH_3-N , a multivariate linear regression model is used.

The experimental data is processed according to the method reported in literature [5].

3. RESULTS AND DISCUSSION

3.1. MBR Treatment Efficiency

TMP , u , $MLSS$, DO , pH , T , SRT and HRT were selected as the influencing factors for the membrane flux and COD , TOC , NH_3-N . The experimental results are shown in Table 1. Figure 2 qualitatively describes the removal of organic matter and ammonia nitrogen in 5 bioreactors under different process conditions (T , $MLSS$ and pH) using 0.1 μm PVDF hollow fiber membrane module. The contents of COD and NH_3-N in effluent are up to the reuse standard of CJ 25 and 1-89 water quality standards. COD , TOC and NH_3-N in effluent are relatively low (in low concentration zone (1.800-4.1g/L), due to the fact that the sludge concentration is appropriate and the temperature range (15-25°C) is in the optimum temperature range for aerobic decomposition of microorganisms to remove organic matter. Therefore, the removal rate of COD and TOC is high. In contrast, in higher concentration zone (6.6-9.9 g /L), the activity of activated sludge is reduced because the concentration is too high and bigger viscosity of the sludge makes it difficult to move in the reactor; Moreover, the higher temperature is also not suitable for aerobic decomposition of microorganisms (the optimum temperature is 15-25°C), and weakens the physiological activities of microorganisms. Therefore, COD and TOC are relatively high due to little change in pH value. According to microorganisms requirement optimum pH value (ranging from 6.5 to 8.5) was used, which has little effect on the pollutants' removal.

In practice, effluent COD , TOC and NH_3-N are affected by many factors. 8 factors, such as TMP , u , $MLSS$, DO , pH , T , SRT and HRT are selected to study their effects on the membrane permeate flux (J) and the quality of effluent (COD , TOC and NH_3-N). Multivariate linear regression analysis is carried out and the corresponding results are shown in Tables 2, 3, 4 and 5. Finally, the quantitative relationship between these indexes and the influencing factors is also established.

The absolute values of TMP , u , $MLSS$, DO , pH , T , SRT and HRT were all influencing factors on J . A greater impact of $|F_7| > |F_3| > |F_6| > |F_2| > |F_8| > |F_5| > |F_1| > |F_4|$, $MLSS$ and SRT on J was observed while the effect of TMP , DO

and P on J was same. The impact of HRT on J is relatively small. The contribution of these 8 factors to J

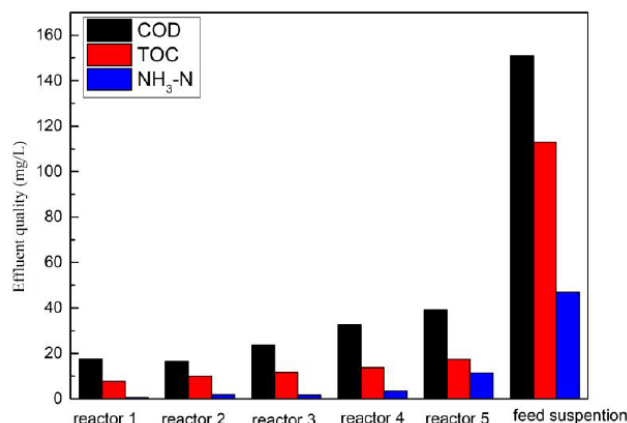


Figure 2: Treatment results for 5 different bioreactors.

were 7.6%, 13.6%, 16.9%, 6.4%, 9.3%, 14.3%, 18.4% and 13.4%, respectively. In addition, the regression coefficients b_3 , b_6 , b_7 and b_8 are all negative, which indicates that $MLSS$, T , SRT and HRT are negative contributors. The increase of $MLSS$, T , SRT and HRT leads to the decrease of J . The increase of $MLSS$ will produce more microbial metabolites (e.g. EPS), which will lead to the increase of sludge suspension viscosity. At the same time, the higher $MLSS$ will lead to more serious concentration polarization on the membrane surface and the thickening of cake layer, which will lead to the decrease of J [44]. Generally, J increases with the increase of temperature, but further increase in temperature (30–45°C), causes increase of EPS and sludge suspension viscosity.

3.2. Influence Degree and Establishment of Quantitative Relationship Formula

Due to high experimental temperature (30°C, 35°C), the impact of temperature on J will be negative.

The change of SRT and HRT resulted in the change of sludge characteristics in the reactor, which correspondingly led to the variation of membrane fouling state. Prolonging SRT will increase the concentration of $MLSS$, which will further effect the concentration of EPS in the feed suspension, and it will promote the occurrence of the membrane fouling. The variation of HRT caused the variation of $MLSS$ and organic load, and indirectly affected the membrane fouling. In contrast, the rest regression coefficients of other influencing factors were all positive, which indicates that the relationship between them and J is a positive correlation. This shows that with the increase of TMP , u , DO and pH , J will also increase. J increases linearly with the increase of TMP and u , in which u effects the migration of particles from the membrane surface by increasing shear force. This will affect the thickness of the cake layer. When DO is higher, due to smaller particles' size, higher porosity of the deposited layer on the membrane surface and smaller cake specific resistance, J becomes bigger [11]. The variation of pH effects J by changing the characteristics of activated sludge suspension and the composition of pollutants in the activated sludge mixture.

Based on the above analysis, the data of J and the process conditions presented in Table 1 were analyzed by multiple linear regression, and the following quantitative mathematical relationship between J and the process conditions was obtained:

$$J = -117.86 + 773.03TMP + 516.83u - 0.007MLSS + 7.63DO + 30.67pH - 6.47T - 0.72SRT - 5.77HRT \quad (1)$$

where, J is the membrane permeate flux, $L \cdot m^{-2} \cdot h^{-1}$; TMP is operating pressure, MPa; u is cross-flow velocity, $m \cdot s^{-1}$; $MLSS$ is sludge concentration, $mg \cdot L^{-1}$; pH is pH value; T is temperature; DO is dissolved

Table 2: Values of Regression Coefficients (F_j and D_j) for the Permeate flux (J)

j	b_j	σ_j	F_j	D_j (%)
1	2.048	0.166	12.341	7.60
2	4.113	0.250	16.463	13.6
3	-0.949	0.052	-18.351	16.9
4	0.761	0.067	11.310	6.40
5	1.663	0.122	13.599	9.30
6	-2.239	0.132	-16.901	14.3
7	-2.502	0.131	-19.134	18.4
8	-1.512	0.093	-16.342	13.4

oxygen, mg.L^{-1} ; SRT is sludge residence time, d and HRT is hydraulic residence time, d.

3.3. Influence of the process condition on effluent COD

The results of the first regression showed that the absolute value of F_1 and F_2 was less than 1 while the absolute value of F for the other parameters was more than 1. This indicates that TMP and u had no effect on the removal efficiency of COD while other operating conditions are the factors affecting the removal efficiency of COD . This is due to fact that the removal efficiency of COD is mainly dependent on the biological treatment of activated sludge. For example, due to vital role of biological treatment in this process, only a few organic pollutants are removed by the cake layer or gel layer on the membrane surface. Therefore, the influence of TMP and u on the removal efficiency of COD is very small.

Ignoring the impact of TMP and u , the results of the second regression analysis of the remaining data after standardization showed that $|F_6| > |F_3| > |F_8| > |F_7| > |F_4| > |F_5| > i.e., T > MLSS > HRT > SRT > DO > pH$ and their contribution to the removal efficiency of COD were 29.9 %, 28 %, 16.8 %, 13.1 %, 6.4 % and 5.8 %, respectively. In addition, the negative value of F_4, F_5, b_4 and b_5 indicate that DO and pH have

a negative effect on the removal efficiency of COD . It shows that the increase of DO and pH will lead to the decrease of COD due to the fact that higher DO enhances the physiological activities of microorganisms and is beneficial for the decomposition of organic matter. Most of the selected pH values were in the optimum pH range of 6.0-8.0, which was beneficial for the aerobic decomposition of microorganisms. On the other hand at higher pH value, the metabolic function of microorganism will be higher and physiological activity of microorganism will be stronger. In contrast, $MLSS$, T , SRT and HRT have a positive effect on the removal efficiency of COD . The higher sludge concentration, the longer SRT and HRT will make the sludge remain in endogenous respiration stage for longer time and reduce the organic load. Consequently, it will cause the mass death of microorganisms, while microbial activity and metabolic capacity will decrease. Therefore, low COD removal efficiency and poor processing efficiency was observed. At same time, higher temperature will inhibit the activity of microorganisms and microbial metabolism process becomes slower, which results in poor effluent quality [14].

The quantitative relationship between COD and process conditions under reliability $\alpha=0.05$ was obtained by analyzing multivariate linear regression as follows:

Table 3: Values of Regression Coefficient (F_j and D_j) for COD

Regression times	j	b_j	σ_j	F_j	D_j (%)
1st regression	1	-0.814	1.168	-0.697	---
	2	-1.087	1.758	-0.619	---
	3	1.787	0.364	4.915	---
	4	-0.903	0.473	-1.909	---
	5	-1.144	0.860	-1.330	---
	6	1.427	0.932	1.531	---
	7	0.994	0.920	1.080	---
	8	1.164	0.651	1.788	---
2nd regression	1	---	---	---	---
	2	---	---	---	---
	3	1.695	0.265	6.384	28.0
	4	-0.760	0.249	-3.051	6.4
	5	-0.589	0.203	-2.892	5.8
	6	0.875	0.133	6.593	29.9
	7	0.410	0.094	4.371	13.1
	8	0.736	0.149	4.945	16.8

$$\text{COD} = 1.60 + 0.005\text{MLSS} - 3.45\text{DO} - 4.91\text{pH} + 1.14\text{T} + 0.053\text{SRT} + 1.27\text{HRT} \quad (2)$$

3.4. Influence of the process conditions on effluent TOC

The regression results of experimental data of TOC and process conditions are shown in Table 4. The absolute values of TMP , u , DO , pH , T and SRT were all less than 1, indicating that TMP , u , DO , pH , T and SRT had no effect on removal efficiency of TOC , and they were not influence factors or there were multiple linear relationships between influence factors. As it can be clearly seen from Figure 3, that there is a multiple linear relationship between temperature (T) and removal efficiency of TOC , as is the case with other factors. The absolute value of F_3 and F_8 is greater than 1. Hence, F_3 is positive, which indicates that $MLSS$ has a positive effect on removal efficiency of TOC , while the negative effect of F_8 indicates that HRT has a negative effect on the removal efficiency of TOC . Moreover, their contributions to the removal efficiency of TOC are 44.7% and 55.3% respectively. In fact, SMP is the main component of TOC in biological treatment. When the sludge concentration is high, the corresponding organic load is low, which makes the sludge in endogenous respiration stage for a long time, and the metabolic ability of microorganisms becomes worse. At the same time, a large amount of SMP is released, which results in higher TOC . The longer HRT means the longer hydraulic retention time of the refractory macromolecular substances in the bioreactor, which strengthens the removal efficiency of refractory substances in the system. At the same time, some SMPs are degraded by microorganisms, which

will cause reduction in TOC [46].

In similar way, the quantitative relationship between TOC and process conditions under reliability $\alpha=0.05$ was obtained as follows:

$$\text{TOC} = 15.17 + 0.001\text{MLSS} - 0.36\text{HRT} \quad (3)$$

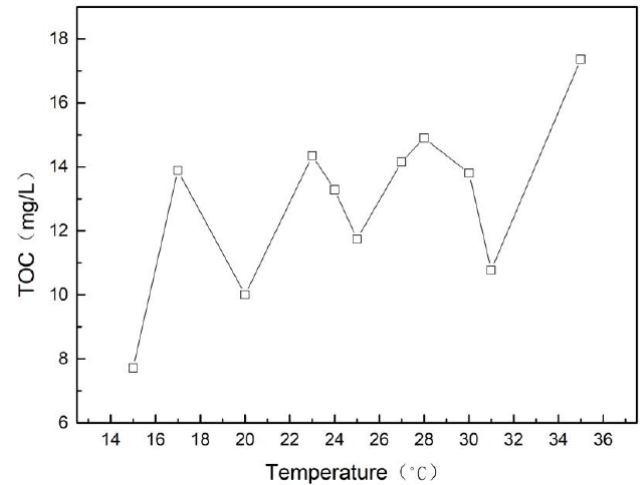


Figure 3: Relation of TOC in effluent and T.

3.5. Influence of Process Conditions on NH_3-N

Multivariate linear regression analysis was used to analyze the experimental data of NH_3-N and operation conditions after standardization. The corresponding results are shown in Table 5. According to the absolute value of F_j , only HRT effects the NH_3-N removal while other factors do not have any effect on NH_3-N removal. As it can be seen from Figure 4, that there is a multiple linear relationship between sludge concentration and effluent NH_3-N , as is the case with other factors. The

Table 4: Values of Regression Coefficient F_j and D_j for TOC

Regression Times	j	b_j	σ_j	F_j	D_j (%)
1st regression	1	-0.742	1.120	-0.663	---
	2	-0.666	1.686	-0.395	---
	3	0.488	0.349	1.398	---
	4	0.265	0.454	0.584	---
	5	-0.429	0.825	-0.520	---
	6	-0.261	0.894	-0.292	---
	7	0.183	0.882	0.207	---
	8	-0.685	0.624	-1.097	---
2nd regression	3	0.552	0.139	3.979	44.7
	8	-0.614	0.139	-4.424	55.3

Table 5: Values of Regression Coefficient (F_j and D_j) for NH_3-H

j	b_j	σ_j	F_j	D_j (%)
1	0.620	2.161	0.287	---
2	1.335	3.253	0.411	---
3	-0.576	0.673	-0.856	---
4	0.264	0.876	0.302	---
5	0.485	1.592	0.304	---
6	-1.004	1.725	-0.582	---
7	-0.797	1.702	-0.468	---
8	-1.304	1.205	-1.083	---

longer HRT will reduce the organic load, but the nitrifying bacteria are autotrophic bacteria, and the longer HRT will make the longer hydraulic residence time of NH_3-N in the reactor and thorough nitrification will take place, so the NH_3-N concentration in effluent decreases with the increase of HRT .

The quantitative relationship between NH_3-N and process conditions under reliability $\alpha=0.05$ was obtained by multivariate linear regression as follows:

$$NH_3-N = 77.79 - 4.06HRT \tag{4}$$

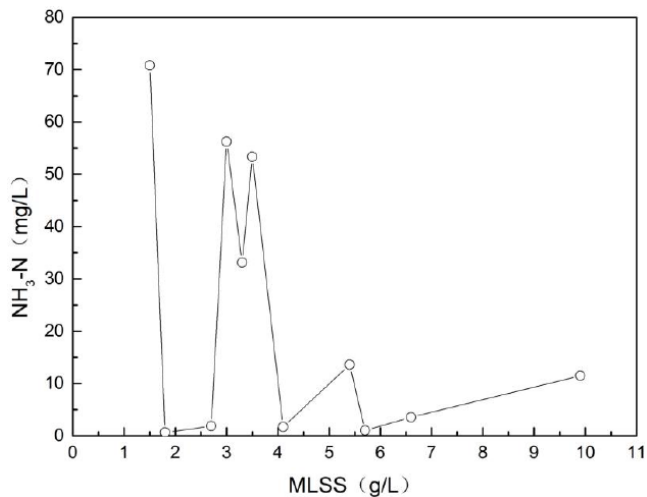


Figure 4: Relation of NH_3-N in effluent and $MLSS$.

4. CONCLUSION

The combined effects of 8 operating conditions and process conditions (trans-membrane pressure (TMP), cross flow velocity (u), mixed liquor suspended solids ($MLSS$), dissolved oxygen concentration (DO), pH , temperature (T), sludge retention time (SRT) and hydraulic retention time (HRT)) on membrane

permeate flux (J) and effluent quality indexes (COD , TOC and NH_3-N) in cross flow hollow fiber membrane module were quantitatively analyzed by using multiple linear regression model. According to the regression results, the effects of various influencing factors on J , effluent quality indexes (COD , TOC and NH_3-N) were analyzed, and the corresponding quantitative relationships were obtained. The obtained results are as follows:

- ❖ TMP , u , $MLSS$, DO , pH , T , SRT and HRT were all the influencing factors of the membrane permeate flux (J), and $MLSS$ and SRT had the greater influence on J and their contributions to J were 16.9% and 18.4%, respectively; u , T and HRT had the same effect with the contributions of 13.6%, 14.3% and 13.4%, respectively; while TMP , DO and pH had the relative influence on J and the corresponding contribution are 7.6%, 6.4% and 9.3%, respectively. Among them $MLSS$, T , SRT and HRT are negative contributors to J while TMP , u , DO and pH are positive contributors. The quantitative relationship between the permeate flux (J) and process conditions can be expressed as follows:

$$J = -117.86 + 773.03TMP + 516.83u - 0.007MLSS + 7.63DO + 30.67pH - 6.47T - 0.72SRT - 5.77HRT$$

- ❖ TMP and u had no effect on COD and other process conditions were the influencing factors. $MLSS$ and T had the greatest influence. $MLSS$, T , SRT and HRT had a positive effect on COD and their contributions were 28%, 29.9%, 13.1% and 16.8%, respectively while DO and pH had a negative effect on effluent COD and their contributions were 6.4% and 5.8%, respectively.

- ❖ The influence of *TMP*, *u*, *DO*, *pH*, *T* and *SRT* on *TOC* is multiple linear. *MLSS* and *HRT* had influence on effluent *TOC* and their respective proportions were 44.7% and 55.3% respectively.
- ❖ Only *HRT* had a negative effect on *NH₃-N*. The quantitative relationships between *COD*, *TOC* and *NH₃-N* and process conditions were as follows:

$$\text{COD} = 1.60 + 0.005\text{MLSS} - 3.45\text{DO} - 4.91\text{pH} + 1.14\text{T} + 0.053\text{SRT} + 1.27\text{HRT}$$

$$\text{TOC} = 15.17 + 0.001\text{MLSS} - 0.36\text{HRT}$$

$$\text{NH}_3 - \text{N} = 77.79 - 4.06\text{HRT}$$

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