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Interactions of Factors for Effluent Quality in Membrane Bioreactor

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ABSTRACT. A new-type membrane module was designed for this project. The permanent hydrophilic macromolecular polymer was adopted as membrane material in the membrane bioreactor (MBR). This MBR combined structural advantages of both hollow fiber membranes and flat sheet membranes. These advantages were large membrane flux, no aeration blind area, and extremly high loading density to allow backflushing. The membrane does not break easily. It is able to swing freely in the water under aeration to reduce membrane fouling. The experimental design was chosen to estimate the influence of several variables. The dyed wastewater was disposed as delegate of refractory industrial effluent with batch operation in this work. The four variables were influent loading, duration of effluent, duration of anoxia and duration of aeration. Then the influence of interaction of factors was studied on the condition that removal rates of COD, TN and membrane flux were considered as response variables. The results showed that influent loading was the most obvious factor affecting the removal rate of COD, and duration of anoxia had the most obvious impact on the removal rate of TN. The removal rate of COD and TN were 94.3% and 81.9%, respectively. The other two factors were more effective for the membrane flux.

Keywords: membrane bioreactor, factorial design, interaction, dyeing wastewater

1. Introduction

Reclamation and recycling of wastewater have become increasingly important in current treatment practice. The Membrane Bioreactor (MBR) for dealing with wastewater is superior to traditional biological wastewater treatment. MBR technology is a promising method for water and wastewater treatment because of its ability to produce high-quality effluent that meets water quality regulations (Yoon and Collins, 2006; Liu et al., 2010). MBR has the advantages of compact structure, simple operation and management. It has the potential to be one of the most effective wastewater treatment technologies, with a very promising future. Membrane bioreactors (MBRs) can be broadly defined as systems integrating biological degradation of waste products with membrane filtration (Vuković et al., 2006). In addition to enriching microbes, the membrane filtration could also intercept contaminant particles efficaciously (Baek and Pagilla, 2009).

The external and the submerged configurations are the two main types of configurations for the membrane array (Marrot et al., 2004; Liang and song, 2007). The submerged one is

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adopted in this work owing to its lower energy consumption. There are also two types for the membrane module: hollow fiber membrane and flat sheet membrane (Busch et al., 2007). The main advantage of the hollow fiber membrane is that it could take back flushing because of its high loading density. But the membrane filaments jam easily due to solid particles in the wastewater (Galil and Jacob, 2009; Gil et al., 2010). Membrane filament breakage and aeration blind area are the insuperable weaknesses of the hollow fiber membrane (Frechen et al., 2008). Although these shortages do not exist for the flat sheet membrane, this module cannot carry out back flushing because of its low loading density. One must take into account the fact that the limiting factors for further process development are membrane fouling and cleaning (Ng and Kim, 2007; Zhou et al., 2008). These factors should be considered to keep membrane equipment in high-efficiency condition.

Factorial design gives information on the importance of each impact factor and the interaction between them with fewer experiments (Sousa and Mateus, 2003; Callahan et al., 2006). Factorial designs allow for the simultaneous study of the effects that several factors may have on a process (Tovar et al., 2010). They are widely used to investigate the effects of experimental factors and the interactions between them, that is, how the effect of one factor varies with the level of the other factors in a response (Aoki and Takemurab, 2009; Bingol et al., 2010). The advantages of factorial design are the relative-

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Table 1.	Pro	perties	of	Sludge	and	Superna	tant
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Index	Mean
pH	7.72~7.64
DO (mg/L)	4.53~4.21
T (°C)	25.1~26.7
SV(ml/L)	343~372
MLSS (mg/L)	4.23~4.19
SVI	81~89
COD (mg/L)	104~138
TN (mg/L)	28.9~27.3
HRT (h)	20

*DO: Dissolved oxygen concentration; SV: Settling Velocity; MLSS: Mixed liquor suspended solids; SVI: Sludge volume index; TN: Total nitrogen; HRT: Hydraulic retention time.

ly low cost, a reduced number of experiments, and increased possibilities to evaluate interactions among the variables (Ch-akraborty et al., 2009).

The aim of this work is to develop a new-type membrane module combining hollow fiber membrane and flat sheet membrane. The factorial design is introduced into design of experimental program to optimize the operating parameters. Four impact factors were contained in a full 2⁴ factorial design in this work. The functional mechanism of their interactions was studied thoroughly and accurately. The action conditions and physical and chemical character will be concluded in further research.

2. Materials and Methods

2.1. Materials

The wastewater and sludge were both obtained from HuaLun Printing and Dyeing Mill in Xiamen. It was unnecessary to take sludge acclimatization. The COD, BOD₅, and TN of wastewater were $2000 \sim 6000, 400 \sim 700$ and $65.9 \sim 125.3$ mg/L, respectively. The index of sludge and its supernatant are exhibited in Table 1. The membrane was developed using permanent hydrophilic macromolecular polymer as material with the cooperation of Xiamen University of Technology and Xiamen Tianquanxin Membrane Technology Corp., Ltd. The total area of the membrane was 0.8 m².

2.2. Methods

2.2.1. System Setup and Operation

The new-type membrane module in this work was designed to assimilate structural advantages of both hollow fiber membrane and flat sheet membrane. Two selective membranes in both sides of module and dialyzate collection layer in the middle composed the new-type membrane module. The dialyzate collection layer was made of special textile fabrics. The inner bracing structure formed a very thin membrane whose thickness was only 2 mm. Therefore the loading density was extremely high parallel with hollow fiber membrane. In addition, the back flush pressure was up to $1 \sim 1.5$ bar to ensure the strength of the membrane.

Table 2. Factors and Values Used in the Factorial Design*

Factors	Low level (-)	High level (+)
A: ICL (mg/L)	500	3000
B: DE (min)	30	90
C: DAO (min)	0	120
D: DAE (min)	10	30

*ICL: Influent COD loading; DE: Duration of effluent; DAO: Duration of anoxia; DAE: Duration of aeration.

The whole module consisted of membrane, aeration foundation and frame trestle. As the filtration part, the membrane submerged in the wastewater without any frame. Due to this structure, the membrane could swing freely in the water under aeration like a hollow fiber membrane. Then membrane fouling could be brought under control to a certain degree.

The other advantage of the module was that the dialyzate collection channel was set in the centre to minimize the pressure loss. It was helpful for the pressure distribution homogeneously to avoid partial fouling. The dialyzate collection channel of traditional flat sheet membrane module was distributed in one side of membrane which results in higher pressure loss. The individual aeration pipe was applied in the equipment, generating small homogeneous bubbles to flush the membrane. So the membrane fouling could be minimized.

Dyed wastewater was disposed as a batch operation in this experiment. Continuous influent and effluent, anoxia stage, and aeration stage were the four stages in the experiment. After a fixed duration of effluent, all the operations would cease to the anoxia stage. Then the aeration device could be started to clean membrane.

2.2.2. Factorial Design

The factorial design was introduced to formulate experiment programming. This design could cover the main and interaction effects of the factors within the whole range of selected parameters (Butler, 2008; Nejad et al., 2011). There were four impact factors in this work. The low and high levels of each factor are listed in Table 2. Removal rate of COD and TN were statistically analyzed as response variables. The full 2^4 factorial design matrix and response variables measured in each factorial experiment are shown in Table 3. The order in which the experiments were made was randomized to avoid systematic errors.

3. Results and Discussion

3.1. Main Effects of Each Factor on Response Variables

The main effects of each parameter on the response variables (removal rate of COD and TN) are shown in Figure 1. Compared with the slopes of the mean connect lines, it was concluded that the effects of ICL (the influent COD loading) and DAO (the duration of anoxia) on the response variables were positive. The effects of DE (the duration of effluent) on the RoC (removal rate of COD) and RoN (removal rate

Run	Factor				Run	RoC	RoN
Number	Α	В	С	D	Label	(%)	(%)
1	-	-	-	-	i	87.3	69.2
2	+	-	-	-	а	94.3	70.3
3	-	+	-	-	b	86.8	67.7
4	+	+	-	-	ab	92.5	70.9
5	-	-	+	-	c	89.8	77.8
6	+	-	+	-	ac	93.2	79.1
7	-	+	+	-	bc	89.5	81.3
8	+	+	+	-	abc	92.2	80.8
9	-	-	-	+	d	88.0	66.4
10	+	-	-	+	ad	93.8	67.7
11	-	+	-	+	bd	90.1	67.0
12	+	+	-	+	abd	94.0	70.1
13	-	-	+	+	cd	91.5	80.7
14	+	-	+	+	acd	93.1	81.9
15	-	+	+	+	bcd	91.9	81.2
16	+	+	+	+	abcd	92.5	81.8

Table 3. Analysis Matrix for the Full 2⁴ Factorial Design and Trail Results^{*}

*RoC: Removal rate of COD; RoN: Removal rate of TN.

of TN) were negative and positive, respectively. Meanwhile, the effect of DAE (the duration of aeration) on the RoC was positive and there was negligible effect on the RoN., Response variables increase as the factor changes from low to high levels when the effect of a factor is positive (Bingol et al., 2010). In contrast, if the effect is negative, a reduction in RoC (or RoN) occurs for high level of the same factor. It could be pointed out that the influent COD loading had the greatest effect on RoC and so did the duration of anoxia on RoN. This may be attributed to the direct impact of influent COD loading on the activity of sludge and the removal rate of COD. The activity of sludge would improve in a certain range following the increase of the COD loading. In the meantime, the removal rate of nitrogen reflected the strength of nitration and denitrification. The anoxia section had a direct connection with the denitrification so the duration of anoxia had a most evident effect on RoN. However, the effects of the duration of effluent and aeration were very weak, especially the duration of aeration on the RoN which had a negligible effect. The reason might be the fact that these two factors mainly worked not on biochemistry side, but the physical side, where dramatic fluctuation of membrane flux was observed in the analysis of membrane flux. Though the hydraulic retention time (HRT) and sludge biochemistry character were influenced by these two factors in a certain extent, the effect was insignificant and can be ignored.

3.2. The Interaction Effect of Factors on RoC and RoN

Interaction plots are used herein to visualize the interaction of two factors on the response and to compare the relative strength of the effects. An increasing response variable RoC was observed when the factor B changed from low to high at factor A's low level. For the high level, the opposite was observed. As shown in the Figure 2(a), two mean connect lines did not run parallel, therefore, there was a significant interaction between factors A and B. Meanwhile, two mean connect lines were parallel and their slope was relatively small. It could be clearly inferred that the effective on the RoN of factors A and B and their interaction were extraordinary weak.



Figure 1. Main effects for removal rates of COD (a) and TN (b).

According to the analysis above, the relationship of two mean connect lines (A and C) was not parallel in the Figure 2(a). A significant interaction was detected in accordance with the reverse signs of their slope. These plots clearly indicate that the interaction between factors A and C was stronger than that between A and B. An approximate parallel relation was shown in Figure 2(b) through their respective steep slope (A and C). It leads to the conclusion that the single actions of two factors were strong but there is hardly any interaction between them. The interaction between A and D was extraordinarily similar to that between A and B. In addition, the interaction between them on the RoN was negligible. An increase of RoC was obtained following the increase of DAO in spite of low or high levels of factor B. Yet, the growth of RoC would be higher at the low level of DE. There was little effect between DE and DAO on RoN which could be similar with factors A and C.

It is clearly indicated from the plot (Figure 2a) that with the increase of DAE, the increase of RoC is higher when DE was at a low level compared to the high level of DE. The two



Figure 2. Interactive effects for removal rates of COD (a) and TN (b).



Figure 3. Normal probability plots for the effects of RoC (a) and RoN (b).

connect lines of factors B and D in Figure 2(b) were almost coincident, which illustrated that there was no significant interaction existing between DE and DAE on RoN. Similarly,



Figure 4. Pareto chart of the effects for removal rates of COD (a) and TN (b) (Alpha = 0.05, Lenth's PSE = 0.28125).

due to the parallel relation between the two connected lines of factors C and D in Figure 2(a), their interactions were very weak and could be negligible. In contrast, the interaction of CD on RoN was extremely obvious in Figure 2(b). Decreasing RoN would take place when the DAE changed from low to high level. The opposite was also true.

3.3. The Statistical Significance of Main and Interaction Effects

Both the main effects plot and the interaction plot reveal the individual effect of each factor. Thus, combined with the normal plot of the standardized effects, the effect form of each factor with significant effect and strong interaction between factors are introduced intuitively. It is used to compare the relative magnitude and the statistical significance of both main and interaction effects.

As shown in Figure 3, a line is drawn to indicate where the points would be expected to fall if all effects were zero. Points that do not fall near the line usually signify significant effects. Such effects are larger and generally further from the fitted line than unimportant effects. A α -level of 0.05 is adopted herein and the significance effect is labeled at the same time.

Therefore, factors A, C, D, AC, and AD had significant effects on RoC among all the factors. It could be observed that factor A had the strongest impact on RoC which was si-



Figure 5. Probability plots for residual for removal rates of COD (a) and TN (b) (Normal-95% Cl).

milar to that in the main effects plot. The effects of factors A, C, and D were positive and proved that RoC would increase when these factors changed from low level to high level. Meanwhile, factors AC and AD had negative effects on RoC.

Similarly, factor C (DAO) had the greatest impact on RoN. The effects of factors A, B, C, and CD were positive and the effects of factors AC, ABC, and BCD were negative. The greatest effect of factor C might be ascribed to the biochemistry effect. The nitrification and denitrification of sludge were influenced by the duration of anoxia. Then in a certain range, the longer the DAO, the more thorough the denitrifycation. So the RoN would be higher.

A Pareto chart of the effects is used to compare the relative magnitude and the statistical significance of both main and interaction effects. The chart is plotted in the decreasing order of the absolute value of the standardized effects. A reference line is drawn on the chart. Compared with the normal plot of the standardized effects, it could lead to the same conclusion. The sequence of these impact factors of RoC and RoN is in descending order: A > AC > D > AD > C and C > CD > A > BCD > B > ABC > AC.

The residual and expected values were exhibited when the distribution was normal in Figure 5. No evidence of nonnormality, skewness, exception value, and indeterminacy variable existed because residual was presented as a straight line.



Figure 6. Contour plots for removal rates of COD with (a) factor A/C (-), hold value B (-)/D (-) and (b) factor A/C (+), hold value B (+)/D (+).

3.4. The Contour Plots and Surface Plots

The contour plot shows how a response variable relates to two factors based on a model equation. Points possessing the same response are connected to produce contour lines of constant responses. A contour plot shows only two factors at a time holding any other factors and covariates at a constant level, thus, the contour plots are only valid for fixed levels of the extra factors. If the holding levels change, the response surface changes as well, sometimes drastically.

The contour plot of RoC and factors A and C is shown in Figure 6. The other two factors (B and D) are kept constant and were introduced as (-) (+) in the figure title. When factors B and D are kept at low level (-), the greater the gray level, the higher the RoC (>94%). The ICL and DAO should be chosen in the bottom right corner of the contour plot to maximize the RoC. Meanwhile, the bottom right corner area would also be the best choice for ICL and DAO when the other two factors are kept at high level. Figure 7 showed how factors A and D were related to response variable RoC. As mentioned above, the bottom right corner of the contour plot is chosen for ICL and DAE to maximize RoC when factors B and C kept at low level. In the high level, the top right corner area would be the optimal setting.



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Figure 7. Contour plots for removal rates of COD with (a) factor A/D(-), hold value B (-)/C(-) and (b) factor A/D(+), hold value B (+)/C(+).

Observed from the four contour plots of RoN, the significant factors were AC and CD. This could easily lead to the conclusion that the top right corner area would be the best choice for the significant factors regardless of whether the other factors are kept at low or high level. The best way to visualize the effect of the independent variables on the dependent ones is to draw surface response plots of the model. These were done by changing two variables within the experimental range and holding the other constant. Surface plots are useful for establishing desirable response values and operating conditions.

RoC could be promoted by increasing ICL and decreesing DAO, Whether factors B and D are held at low or high level, the operating conditions of ICL and DAO should be kept at high level and low level respectively to maximize RoC. Factor A was proportional to RoC and factor D and was reciprocal to RoC when the other two factors are kept at low level. The response variable RoC had a direct ratio with factor A and inverse ratio with factor D when the other two factors were kept at the high level.

These results were identical with those in the contour plots analysis. When the hold values of factors B and D are kept at low level, RoN had the same changing patterns with



Figure 8. Contour plots for removal rate of TN with (a) factor A/C (-), hold value B (-)/D (-) and (b) factor A/C(+), hold value B (+)/D (+).

factors A and C. Then keeping A and C at high level could maximize RoN. An identical variation is noted in Figure 12 (b) when factors B and D are held at high level. It could be inferred that factors C and D at high level could maximize RoN whenever the other two factors are low or high.

3.5. Analysis of Membrane Flux

The membrane flux variation behavior is shown in Figure 14a. Mean fluxes in every two sets of experiments were approximately the same while dissimilar visibly with the next two. The similarity was displayed in the contiguous two sets of experimental schemes through analyzing changes between them. In runs 1 and 2, values of all other three factors were identical except factor A. Therefore, it led to the conclusion that the influence of factor A to the membrane flux was too tiny and can be ignored. Eliminating factor A, the only difference between run 1, 2 and run 3, 4 was the value of factor B. Consequently, factor B, the duration of effluent, impacted distinctly on membrane flux due to the significant difference between them. The explanation could be this: the longer the duration of effluence was, the more serious the temporal membrane fouling. When the duration of effluence took lower



Figure 9. Contour plots for removal rates of TN with (a) factor C/D (-), hold Value A (-)/B (-) and (b) factor C/D (+), hold Value A (+)/B (+).

bound like runs 1, 2, 5, 6 etc., compared with runs 3, 4, 7, 8 etc., the mean membrane flux was higher up to $9.81 \text{ L/(h \cdot m^2)}$. Thus it could be seen that there is a direct effect of the duration of effluence on the membrane flux.

There were six periods in every run. Two membrane fluxes were recorded in every period. The two membrane fluxes in each run would decrease as the experiment proceeded. In comparison with the previous period, the membrane flux had been resumed after aeration flush at the end of the period. This suggested that aeration played a vital role in the flux recovery. The fluctuation of membrane flux was inspected by the standard deviation of 12 sets of membrane flux in each run as shown in the Figure 14(b). In the scatter diagram, there was a strong disparity between every four adjacent runs. After integrating the mean scatter and the experimental scheme, the main distinction of them was the value of factor C, duration of anoxia. It illustrated that factor C was not so effective on the membrane flux. This corresponded with the variation tendency of factor D, whose standard deviation increased in the meantime. The cleaning effect would be more impressive when the duration of aeration got longer, then the flux recovery would be much better. In other words, the fluctuation of membrane flux reflected the alteration of standard deviation in the statistical data. This led to a conclusion that the dura-



Figure 10. Surface plots for removal rates of COD with (a) factor A/C (-), hold Value B (-)/D (-) and (b) factor A/C (+), hold Value B (+)/D (+).



Figure 11. Surface plots for removal rate of COD with (a) factor A/D (-), hold Value B (-)/C (-) and (b) factor A/D (+), hold Value B (+)/C (+).

tion of aeration was also very effective on the membrane flux besides the duration of effluence.

On the basis of the analyses above, the duration of effluence and aeration were the effective factors of membrane flux, as shown in the scatter of mean flux and standard devia-



Figure 12. Surface plots for removal rates of TN with (a) factor A/C(-), hold Value B(-)/D(-) and (b) factor A/C(+), hold Value B(+)/D(+).



Figure 13 Surface plots for removal rate of TN with (a) factor C/D(-), hold Value A(-)/B(-) and (b) factor C/D(+), hold Value A(+)/B(+).

tion. This could explain that the impact of the two factors was from the physical part, while the two other factors (influent COD loading and duration of anoxia) took effect on the effluent quality in terms of biochemistry.



Figure 14. Scatter plots of (a) mean flux and (b) standard deviation.

4. Conclusions

The main effect of each factor and interaction between them were integrally, systematically, and veraciously revealed through factorial design and statistical methods. These approaches were applied by optimizing the conditions of maximum removal rate of COD and TN. Factor A had a dramatic positive effect on removal rate of COD (RoC). There were negative interactions between AC and AD. Then maximum RoC would be obtained in the condition that influent COD loading was 3000 mg/L, duration of anoxia was 120 min, and duration of aeration was 30 min. Similarly, the optimization of operating conditions for removal rate of TN (RoN) were that influent COD loading was 3000 mg/L, duration of effluent was 90 min and duration of aeration was 30 min. Factor C had the most significant effect on RoN and there were also interactions existing between AC and CD. Obviously, factors B (duration of effluent) and D (duration of aeration) had the most dramatic effects on membrane flux.

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