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Optimization of chemical cleaning condition for microfiltration process using response surface methodology

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ABSTRACT

Microfiltration (MF) processes are used in a variety of separation and concentration applications. Since membrane fouling is inevitable, membranes must be regularly cleaned to remove both organic and inorganic material deposited on the surface and/or into the membrane bulk. Optimization of the cleaning conditions for MF membranes is especially important. If the dose of cleaning chemical or cleaning time is inadequate, the membrane permeability is not recovered. If a dose of the chemical or the time is excess, irreversible damages in membrane properties occur. However, it is difficult to find the optimum conditions for chemical cleaning of MF. In this study, response surface methodology (RSM), a facile tool for optimization, was employed to determine the optimum conditions for chemical cleaning of MF. The Box–Behnken center-united experimental design was used to quantify the effects of respective chemicals (citric acid and sodium hypochlorite) dose and treatment time on fouling control and organic removal. The dose of citric acid and sodium hypochlorite ranged from 1,600 to 2,400 ppm and from 200 to 1,400 ppm, respectively. The treatment time was also conducted from 1 to 5 h. After the chemical cleaning treatment, transmembrane pressure data of distilled water were compared with them before the chemical cleaning treatment. Experimental results indicated that the efficiency of chemical cleaning is sensitive to the concentration of cleaning chemicals (citric acid and sodium hypochlorite) as well as cleaning time. Nevertheless, the dependency of cleaning efficiency on these parameters was different. The cleaning efficiency, which is expressed as the recovery of membrane permeability after cleaning, varies from 0 to 72%. The RSM analysis could suggest the optimum conditions for membrane cleaning.

Keywords: Microfiltration; Coagulation; Water treatment; Response surface methodology; Optimization

1. Introduction

Membrane filtration, including microfiltration (MF) or ultrafiltration (UF), is gaining popularity as a feasible option for advanced water and wastewater treatment [1,2]. The global use of MF and UF systems for drinking water treatment has drastically increased since the mid-1990s [3]. One reason for the increase is their ability to help meet regulatory requirements for lower filtered water turbidity and for reliably

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removing pathogens such as Giardia cysts and Cryptosporidium oocysts. Another reason is that continual advances in membrane technologies have led to comparable or lower costs for membrane filtration vs. conventional filtration systems [4].

However, membrane systems inherently have problems associated with fouling [5,6]. Fouling, which is common to all types of membrane separation methods, arises from a combination of chemical and physical interactions [7]. The constituents in the feed can attach to the membrane surface though chemical binding and/or the interaction of surface properties, such as the degree of hydrophilic or charge effects [7]. Also, membrane fouling is a process where solute or particles deposit onto a membrane surface or into membrane pores in a way that degrades the membrane's original performance. This leads to increase in transmembrane pressure (TMP) to produce water, thereby increasing the cost for water production [8,9].

Accordingly, many researchers and operators have studied a physical and chemical cleaning method and operation condition to reject membrane fouling on the membrane surface. Especially, since membrane fouling is inevitable, the membranes must be regularly cleaned to remove both organic and inorganic material deposited on the surface and/or into the membrane bulk [9,10]. So the general approach to restore hydraulic cleanliness of membranes exposed to biological suspensions is the use of strong oxidants (sodium hypochlorite, NaOCl) to remove organic matter followed by an acidic cleaning step (e.g. citric acid coupled with mineral acid) to remove metal hydroxides [11].

However, optimization of the cleaning conditions for MF membranes is especially important. If the dose of cleaning chemical or cleaning time is inadequate, the membrane permeability is not recovered. If a dose of the chemical or the time is excess, irreversible damages in membrane properties occur [12]. Also, frequent chemical cleaning not only shortens membrane lifetime, but it also consumes additional energy and produces concentrated waste streams, thus decreasing sustainability. However, it is difficult to find the optimum conditions for chemical cleaning of MF [13,14].

In this study, response surface methodology (RSM), a facile tool for optimization, was employed to determine the optimum conditions for chemical cleaning of MF. This technique allows the derivation of empirical equation for predicting the effectiveness of coagulation process [15]. Accordingly, it can be also used for system control and may have a lot of applications. Of the analysis method, The Box–Behnken center-united experimental design was used to quantify the effects of respective chemical (citric acid and sodium hypochlorite) dose on fouling control and organic removal.

2. Materials and methods

2.1. Experimental methods

2.1.1. MF hollow fiber membrane

Membranes used from pilot-scale plant in Kwangam water treatment plant ($Q = 1{,}000\ m^3/d$) were examined. After fouling occurred in pilot plant, the membrane fibers were obtained from a full-scale submerged membrane module and used for the chemical cleaning experiment. Prior to the cleaning experiments, the permeabilities of these membranes were analyzed by measuring TMP profile using deionized water. As shown in Fig. 1, the TMP of the membranes from the pilot-plant was more than three times than that of new membrane, suggesting that the fouling was significant.

The used MF membrane was made of polyvinylidene fluoride and manufactured by the SDI Samsung, Korea. It has a nominal pore size of 0.03 μm, an internal diameter of 1.2 mm, and an external diameter of 2.1 mm. A single submerged membrane was, respectively, adopted and has 0.0132 m^2 of surface area.

2.1.2. Laboratory operation of submerged membrane system

A schematic diagram of the submerged hollow fiber membrane system in laboratory-scale is shown in Fig. 2. This system has 10 filtration units of submerged hollow fiber membranes and can simultaneously measure TMP data from all the units. The system was operated in the total recycle mode. Accordingly, feed

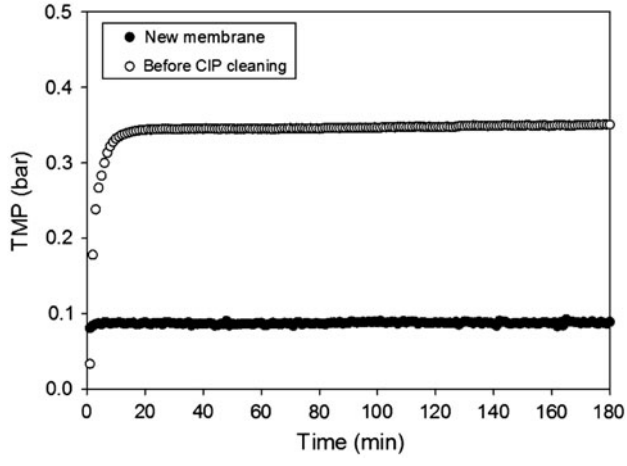

Fig. 1. Comparison of TMP profiles between new and fouled membranes.

water in a tank continuously is returned to the same tank after the filtration. The permeate from the hollow fiber membrane was pulled by a multi-channel cartridge peristaltic pump (EW-07551-00, Cole-Parmer, USA). The TMP was continuously measured by a pressure transducer (ISE40A-01-R, SMC, Japan) and a data logger (usb-6008, NI, USA) that was connected to a computer for data analysis.

2.1.3. Membrane surface analysis

Surfaces of MF membrane fibers before, after the chemical cleaning, and of non-use (named "new") were analyzed by a field emission scanning electron microscope (FE-SEM, S-4700, Hitachi, Japan). Samples were coated with platinum for 2 min before they were taken picture of by FE-SEM. Also, operation conditions of the FE-SEM were as follows: magnification = 3,000, accelerating voltage = 10,000 V, emission current = 15,500 nA, working distance = 1 4,200 um, micron marker = 10,000, specimen bias = 1, condenser 1 = 5,000, scan speed = slow 3, calibration scan speed = 25). It can help to visually confirm a cleaning ability of the chemical cleaning optimum condition.

2.2. Chemical cleaning

2.2.1. Chemicals for the chemical cleaning

To remove and control the membrane fouling caused by inorganic material or organic material from feed water, the chemicals (1. Acid: citric acid (99.5%), 2. Base: sodium hypochlorite (10–15 %)) for the chemical cleaning were used.

2.2.2. Chemical cleaning method

Used membranes from a pilot-scale water treatment plant (Q = 1,000 m^3/d) were examined. After fouling occurred, the membrane fibers were obtained from a full-scale submerged membrane module. They were stored in a refrigerator and used for the cleaning experiments. Before the cleaning was applied, the permeability of each fiber was measured using a flux step method (30, 60, 90, 120 L/m^2 h) through the submerged hollow fiber membrane system. A flux step method was consisted of four steps gradually increasing 30 L/m^2 h for 3 h. At the same time, TMP data was recorded to analyze a fouling rate which a membrane has. It helps to easily analyze previous and later fouling rate for each membrane. A explanation for the method is specifically stated as stated below Section 2.3. Then, the experiment was conducted according to the experimental conditions from RSM experimental design in Table 2. At this time, a standard condition of chemical cleaning consulted the operation method from a pilot-scale water treatment plant (Q = 1,000 m^3/d). The standard condition was citric acid 2000 ppm and sodium hypochlorite 800 ppm for 3 h. Since the results were intended to be applied to full-scale membrane modules, we had to follow certain criteria. We also tried to simulate the situations in the MF pilot plant. Accordingly, the conditions were selected based on the cleaning conditions of the pilot-scale MF process.

Citric acid and sodium hypochlorite were ready to clean an MF membrane following the cleaning conditions such as concentration and time. Treatment time for chemical cleaning was, respectively, implemented under the same condition in acid and base solutions. After chemical cleaning, a treated MF membrane

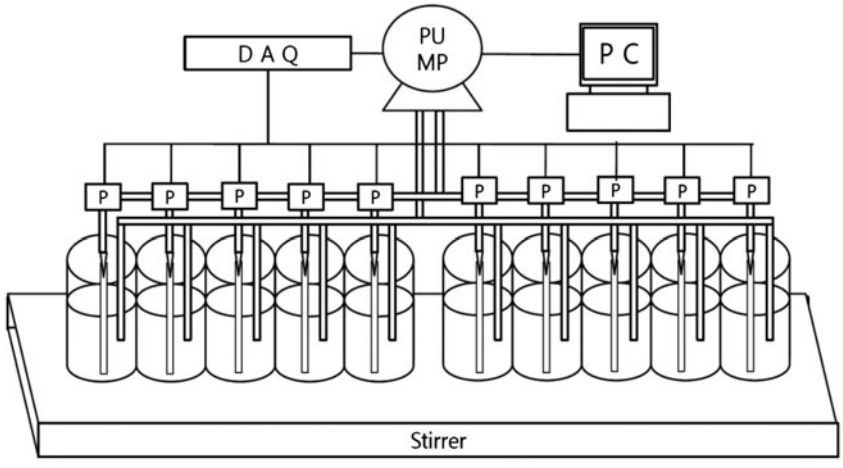

Fig. 2. Schematic diagram of a device for TMP measurement of MF hollow fiber.

was washed by dilute water three times and was conducted in a submerged membrane system to re-measure the permeability of the fibers.

2.3. Accelerated testing

This testing is alternatively called flux step method. Accelerated testing is an approach for obtaining more information from a given test time than would normally be possible. It is done using a test environment that is more severe than that experienced during normal use of equipment. Since higher stresses are used, accelerated testing must be approached with caution to avoid introducing failure modes that will not be encountered in normal use.

Accelerating factors for testing membrane modules or systems, which are either single or in combination, include high concentration of foulants, high flux, less frequent backwashing or cleaning, more severe chemical conditions, and high recovery. Accelerated testing falls into two main categories, each with specific two purposes. One is to accelerate the life of a membrane. The other is to conduct accelerated stress testing. It gives us identification information of problem or weakness on any membrane.

2.4. Response surface method

RSM explores the relationships between several explanatory variables and one or more response variables. The main idea of RSM is to use a sequence of designed experiments to obtain an optimal response. An easy way to estimate a first-level polynomial model is to use a factorial experiment or a fractional factor design. This is sufficient to determine which explanatory variables have an impact on the response variable(s) of interest. Once it is suspected that only significant explanatory variables are left, then a more complicated design, such as a central composite design (CCD) can be implemented to estimate a second-degree polynomial model, which is still only an approximation at best. However, the second-degree model can be used to optimize (maximize, minimize, or attain a specific target for). This approach is being used efficiently in optimizing the surface roughness response [13].

2.4.1. Experimental design

Based on the results of our preliminary studies from a pilot-scale water treatment plant, a CCD with four variables and five levels (i.e. −1.6817, −1, 0, 1 and 1.6817) was employed. The three independent variables are a dose of citric acid (X_1), a dose of NaOCl (X_2) and treatment time (X_3). The response is the recovery rate (Y_1, Y_2, Y_3, Y_4) on flux of 30–120 L/m^2 h, which can indicate fouling control and rejection efficiency of organic and inorganic materials, respectively.

The acid dose was ranged from 1,600 to 2,400 ppm and gradually increased by 200 ppm. The base dose was ranged from 200 to 1,400 ppm and gradually increased by 300 ppm. Also, the reaction time was conducted from 1 to 5 h. The experimental design is shown in Table 1. All experiments were conducted in the order as shown in Table 2.

2.4.2. Statistical analysis and regression analysis

A second-order polynomial model, as shown below, was used for regression analysis between the experimental data using the Minitab® 16.2.0 (Minitab, USA).

$$Y_k = \beta_{k0} + \sum_{i=1}^{3} \beta_{ki} X_i + \sum_{i=1}^{3} \sum_{j=i+1}^{4} \beta_{kij} X_i X_j + \sum_{i=1}^{3} \beta_{kii} X_i^2 \quad (1)$$

where Y_k are the responses, namely Y_1 for the recovery rate on 30 L/m^2 h, Y_2 for the recovery rate on 60 L/m^2 h, Y_3 for the recovery rate on 90 L/m^2 h, Y_4 for the recovery rate on 120 L/m^2 h; β_{k0}, β_{ki}, β_{kii} and β_{kij} are the regression coefficients; and X_i are the coded independent variables. The R^2 and the lack-of-fit are evaluated for the fitness of the model.

3. Results and discussion

3.1. The results of recovery rate after chemical cleaning

As shown in Fig. 3, they have similar graph forms at the same condition. Four bar graphs indicated that they have similar tendency and optimum conditions: citric acid—1,600–1,800 ppm, sodium hypochlorite—500–800 ppm and treatment time 3–4 h. Also, experimental results in Fig. 3 indicate that the efficiency of chemical cleaning is sensitive to the concentration of cleaning chemicals (citric acid and sodium hypochlorite) as well as cleaning time.

However, membrane cleaning is a complex process and difficult to predict. This is why current membrane systems adopt cleaning methods based on trial-and-error approaches. The reason why RSM was applied was to predict the cleaning efficiency as a function of operating parameters. Unfortunately, RSM analysis cannot reveal the mechanisms. Accordingly, RSM analysis cannot explain why run number 5 and 9 showed higher cleaning efficiencies.

Table 1
Box–Behnken center-united experimental design for chemical cleaning condition

	−α (−1.6817)	−1	0	1	α (1.6817)
X_1 (citric acid), ppm	1,600	1,800	2000	2,200	2,400
X_2 (NaOCl), ppm	200	500	800	1,100	1,400
X_3 (treatment time), h	1	2	3	4	5

Table 2
Box–Behnken center-united experimental design for chemical cleaning condition

Run	X_1 (citric acid), ppm	X_2 (NaOCl), ppm	X_3 (time), h	Y (recovery rate), %			
1	−1 (1,800)	−1 (500)	−1 (2)	41	50	49	46
2	1 (2,200)	−1 (500)	−1 (2)	38	37	32	26
3	−1 (1,800)	1 (1,100)	−1 (2)	46	44	35	25
4	1 (2,200)	1 (1,100)	−1 (2)	47	44	41	34
5	−1 (1,800)	−1 (500)	1 (4)	62	70	70	64
6	1 (2,200)	−1 (500)	1 (4)	49	45	38	29
7	−1 (1,800)	1 (1,100)	1 (4)	47	46	40	32
8	1 (2,200)	1 (1,100)	1 (4)	0	2	5	10
9	−1.6817 (1,600)	0 (800)	0 (3)	67	69	66	61
10	1.6817 (2,400)	0 (800)	0 (3)	51	51	45	36
11	0 (2,000)	−1.6817 (200)	0 (3)	14	13	12	10
12	0 (2,000)	1.6817 (1,400)	0 (3)	48	44	38	32
13	0 (2,000)	0 (800)	−1.6817 (1)	29	23	11	10
14	0 (2,000)	0 (800)	1.6817 (5)	50	50	43	37
15	0 (2,000)	0 (800)	0 (3)	54	52	46	37
16	0 (2,000)	0 (800)	0 (3)	46	36	26	15
17	0 (2,000)	0 (800)	0 (3)	44	36	29	18
18	0 (2,000)	0 (800)	0 (3)	56	42	28	16
19	0 (2,000)	0 (800)	0 (3)	45	36	32	28
20	0 (2,000)	0 (800)	0 (3)	48	38	29	19

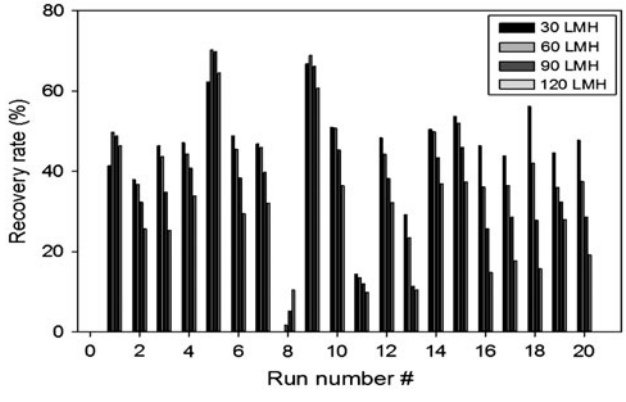

Fig. 3. The recovery rate of permeability after chemical cleaning.

One of the hypotheses is that there are optimum concentration for NaOCl and citric acid. Increasing the NaOCl concentration above a certain value (~800 ppm) seems to decrease cleaning efficiency. Increasing the citric acid concentration can result in a decrease in cleaning efficiency. This may be attributed to possible modifications of foulant properties due to an exposure to excessive cleaning agents.

Furthermore, the dependency of cleaning efficiency on these parameters was also different. As shown in Fig. 3, the cleaning efficiency, which is expressed as the recovery of membrane permeability after cleaning, varies from 0 to 72%. Especially, the highest result of recovery rate was far different than a data of other literature reviews which reached above 90%.

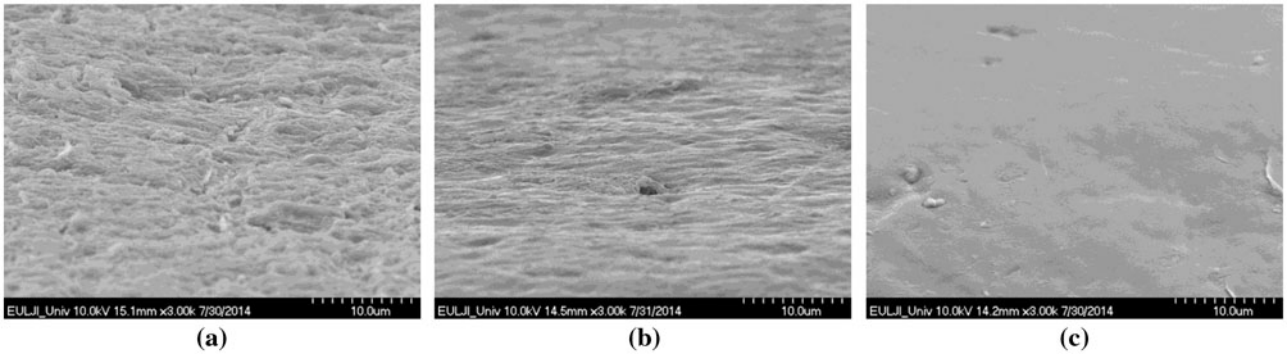

Fig. 4. SEM images of (a) used membrane before treatment, (b) used membrane after chemical cleaning, and (c) new membrane.

In our case, we took the samples from a pilot plant (1,000 m^3/d) and the recovery of permeability was less than 70%. This was actually the motivation of this study, which aims to optimize the cleaning efficiency. Since the recovery ratios were similar in the lab-scale and pilot-scale tests, the results are reliable. There are several possible reasons why the recovery of membrane permeability after chemical cleaning was relatively low.

First, the membranes used in this study were obtained from a pilot plant, which was operated for more than a year. Accordingly, the membranes are likely to be irreversibly fouled by the long-term operations. The SEM images in Fig. 4 confirm that foulants still remained on the membrane surface even after the chemical cleaning.

Second, the test conditions in the pilot plant were more severe than those in conventional membrane processes. The membrane filtration was carried out without aeration, which may lead to high propensity of membrane fouling. Accordingly, it appears that the efficiency of membrane cleaning was relatively low.

3.2. Prediction of membrane fouling using the response surface method

In this study, RSM, a facile tool for optimization, was employed to determine the optimum chemical cleaning conditions for fouling control of MF membrane. The center-united experimental design was used to quantify the effects of the dose of citric acid and sodium hypochlorite on fouling control and organic removal. The results obtained from the experiments were evaluated by multiple regression analysis method and empirical relationship between the response and independent variables has been expressed by a multi-order polynomial equation. Empirical models were developed to understand the interactive correlation between the responses and process variables.

Table 3
Coefficients of the fitted polynomial models for responses

	Y_1(30 L/m^2 h)		Y_2(60 L/m^2 h)		Y_3(90 L/m^2 h)		Y_4(120 L/m^2 h)	
	Coef	P	Coef	P	Coef	P	Coef	P
B_0 (Constant)	48.884	0.000	39.984	0.000	31.581	0.000	22.111	0.001
B_1(X_1, Conc)	−6.510	0.073	−8.221	0.043	−8.297	0.037	−8.058	0.026
B_2(X_2, pH)	0.526	0.875	−1.015	0.781	−1.777	0.617	−1.977	0.537
B_2(X_2, pH)	1.561	0.641	2.446	0.506	3.648	0.314	3.618	0.269
B_{11} (Conc × Conc)	3.263	0.327	7.173	0.065	8.989	0.023	9.672	0.009
B_{22} (pH × pH)	−6.637	0.062	−3.964	0.278	−1.795	0.604	−0.051	0.987
B_{22} (pH × pH)	−3.632	0.278	−1.136	0.749	−1.088	0.752	0.833	0.787
B_{12}(X_{12}, Conc × pH)	−3.750	0.398	−0.750	0.875	2.500	0.591	5.250	0.223
B_{12}(X_{12}, Conc × pH)	−7.250	0.118	−7.000	0.162	−7.000	0.151	−5.750	0.185
B_{12}(X_{12}, Conc × pH)	−9.750	0.044	−8.500	0.097	−7.250	0.138	−4.750	0.267

Table 4
ANOVA for the polynomial models

Response	Source	Degrees of freedom	Sum of square	*F*-value	*p*-Value
Y_1	Model	9	2,930.43	2.26	0.110
	Residual	10	1,441.37		
	Lack of fit	5	1,316.54	10.55	0.011
	Pure error	5	124.83		
	R^2	19	4,371.80		
Y_2	Model	9	3,082.84	1.99	0.149
	Residual	10	1,719.96		
	Lack of fit	5	1,519.96	7.60	0.022
	Pure error	5	200.00		
	R^2	19	4,802.80		
Y_3	Model	9	3,341.37	2.29	0.106
	Residual	10	1,618.38		
	Lack of fit	5	1,353.05	5.10	0.049
	Pure error	5	265.33		
	R^2	19	4,959.75		
Y_4	Model	9	3,147.39	2.68	0.070
	Residual	10	1,304.36		
	Lack of fit	5	933.53	2.52	0.167
	Pure error	5	3,370.83		
	R^2	19	4,451.75		

The experimental design and four resulted responses (Y_1, Y_2, Y_3, Y_4) are shown in Table 3. Two second-order polynomial regression models were established and tested for adequacy and fitness by the analysis of variance (ANOVA). The regression coefficients (coded factors) of the models for Y_1, Y_2, Y_3, and Y_4 are listed in Table 3. The significance of each coefficient was tested by *p*-value at 0.10 level. The *p*-value is a criterion for determining the significance of the results. Although 0.05 is generally used, other values may be used. In our case, due to the quality of the experimental results, we applied 0.10 instead of 0.05. All terms which are not significant at $p > 0.10$ level were removed from the models and the reduced forms of the full polynomial models are as follows:

$$Y_{1,30\mathrm{LMH}} = 48.8841 - 6.5102X_1 - 6.6370X_2^2 - 9.75X_2 \cdot X_3 \quad (2)$$

$$Y_{2,60\mathrm{LMH}} = 39.9844 - 8.2209X_1 + 7.1728X_1^2 - 8.5X_2 \cdot X_3 \quad (3)$$

$$Y_{3,90LMH} = 31.581 - 8.297X_1 + 8.989X_1^2 \quad (4)$$

$$Y_{4,120LMH} = 22.1113 - 8.0578X_1 + 9.672X_1^2 \quad (5)$$

where X_1, X_2, and X_3 take the coded values of the independent variables.

According to the results of ANOVA, the main factors for Y_1 and Y_2 were included but almost main factor for Y_3 and Y_4 such as X_2 and X_3 were except. The linear terms are significant for both Y_1 and Y_2 because the equations of Y_1 and Y_2 have the linear term of X_1, X_2, and X_3, respectively. The significant quadratic terms for Y_1 are X_1 and X_2; the significant quadratic term for Y_1 is X_2 only.

However, the significant quadratic terms for the others have X_1. Also, X_1 as the interaction term for Y_1 and Y_2 is significant for overall. The results of ANOVA are shown in Table 4. The R^2 of Y_1, Y_2, Y_3, and Y_4 models were 67.03, 64.19, 67.37, and 70.70, respectively, indicating that the model calculations are reasonable. Moreover, both the lack-of-fits were not significant at $p > 0.10$ level. This indicates that both established models are in good agreement and they are appropriate for representing the relationship between independent variables and responses.

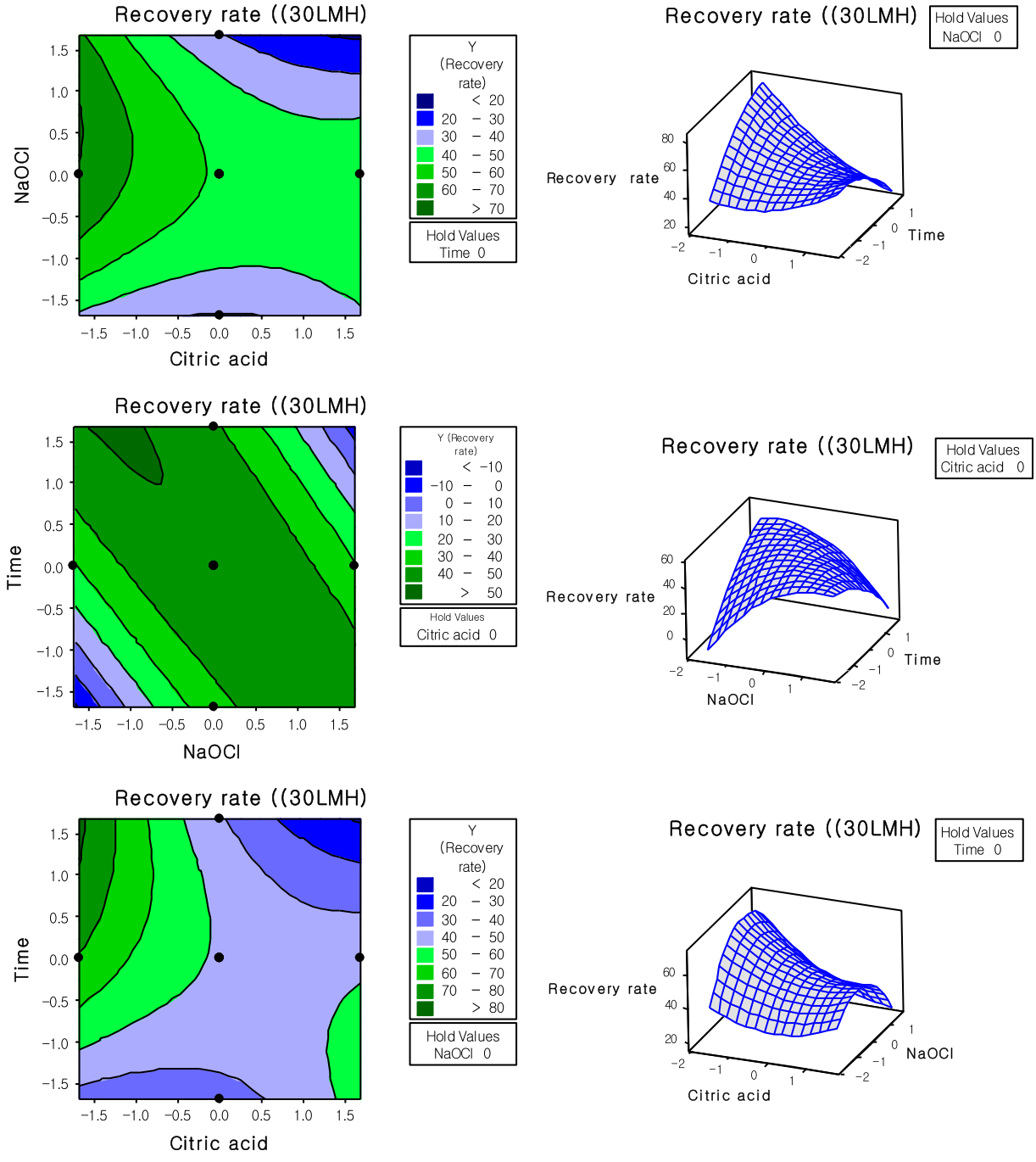

Fig. 5. Response surface of the effects of three independent variables for recovery rate on flux at 30 L/m^2 h; Y_1.

3.3. *Effect and interaction of variables*

The effects of the independent variables and their interaction on Y_1, Y_2, Y_3, and Y_4 are illustrated as response surfaces in Figs. 5–8. The RSM data show how the recovery rate changes with the variables.

Based on the response surface plots, it is likely that the cleaning efficiency increases with decreasing the concentration of citric acid and increasing the treatment time. On the other hand, the cleaning efficiency increases with an increase in the concentration of sodium hypochlorite when it ranges from 300 to 700

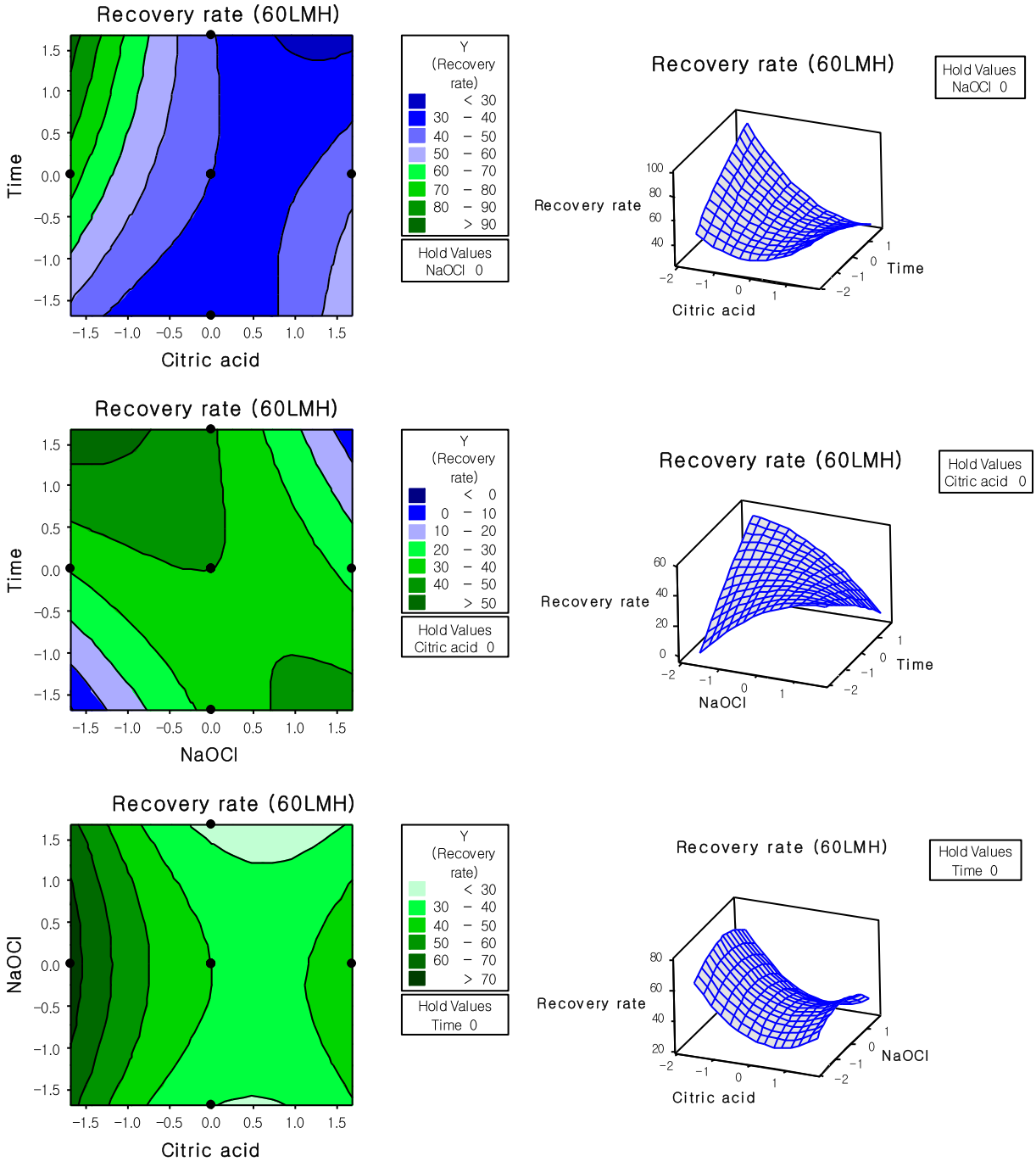

Fig. 6. Response surface of the effects of three independent variables for recovery rate on flux at 60 L/m^2 h; Y_2.

ppm. It is interesting to note that the cleaning efficiency is not simply proportional to the concentrations of cleaning chemicals. In conclusion, it appears that the interactions among three variables are not simple and have non-linear correlations.

Organic fouling seem to be more important that inorganic fouling because sodium hypochlorite was more effective than citric acid. It is evident from the response surface plots that these complex interactions could be successfully predicted. Accordingly, the

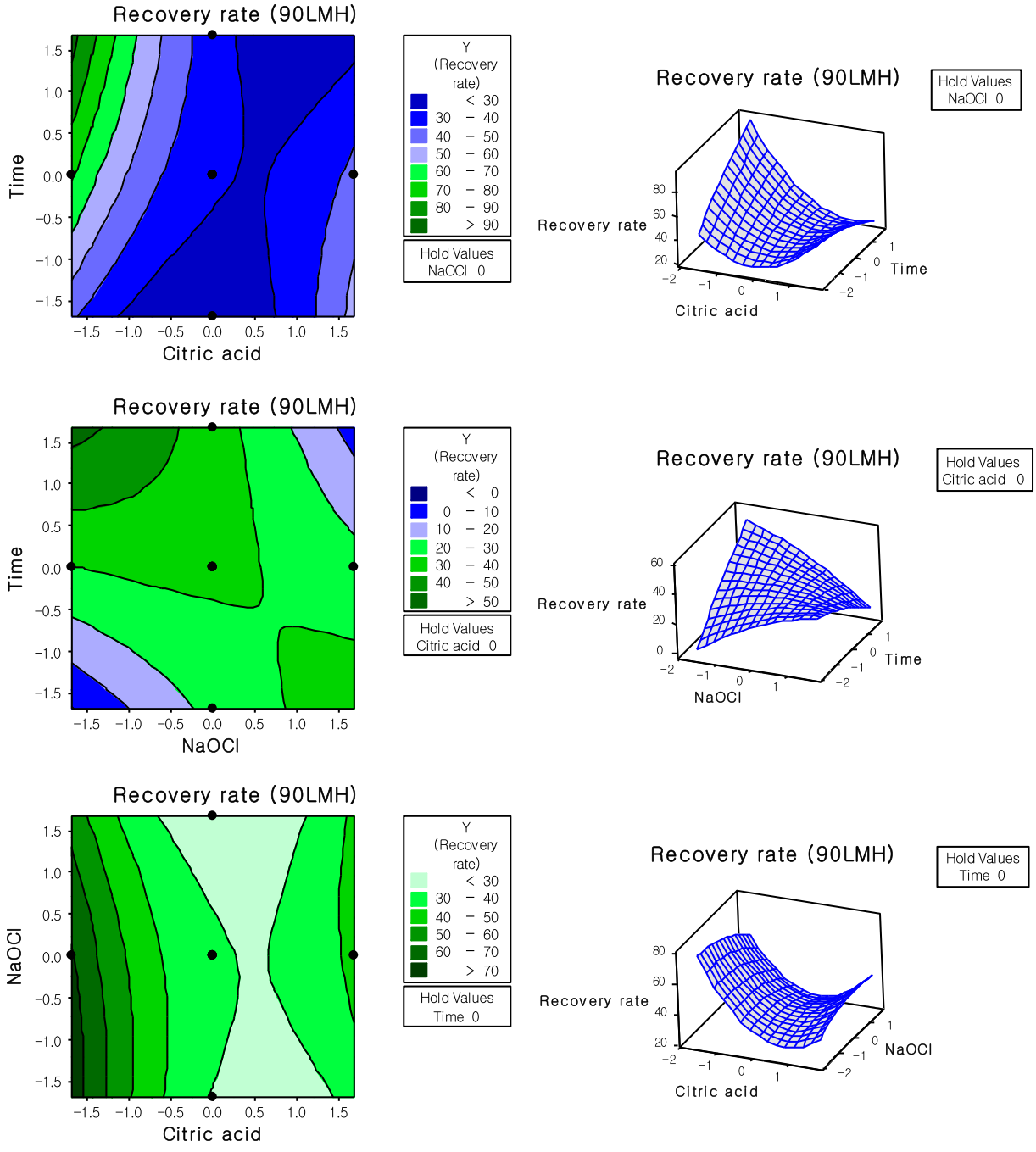

Fig. 7. Response surface of the effects of three independent variables for recovery rate on flux at 90 L/m^2 h; Y_3.

optimum conditions for the recovery rate could be easily found by RSM analysis.

3.4. Optimization of the conditions for chemical cleaning

Fig. 9 shows the analysis of the effects of three independent variables for the cleaning efficiency by the response optimizer. Based on the results, the optimum condition for citric acid concentration, NaOCl concentration, and cleaning time was identified as 1,600 ppm (−1.68179), 580 ppm (−0.73076) and 5 h (1.68179), respectively, on flux at 30 L/m^2 h. Under this condition, the recovery ratio was predicted to be 85.3 %, which were also experimentally confirmed. This result was similar with the

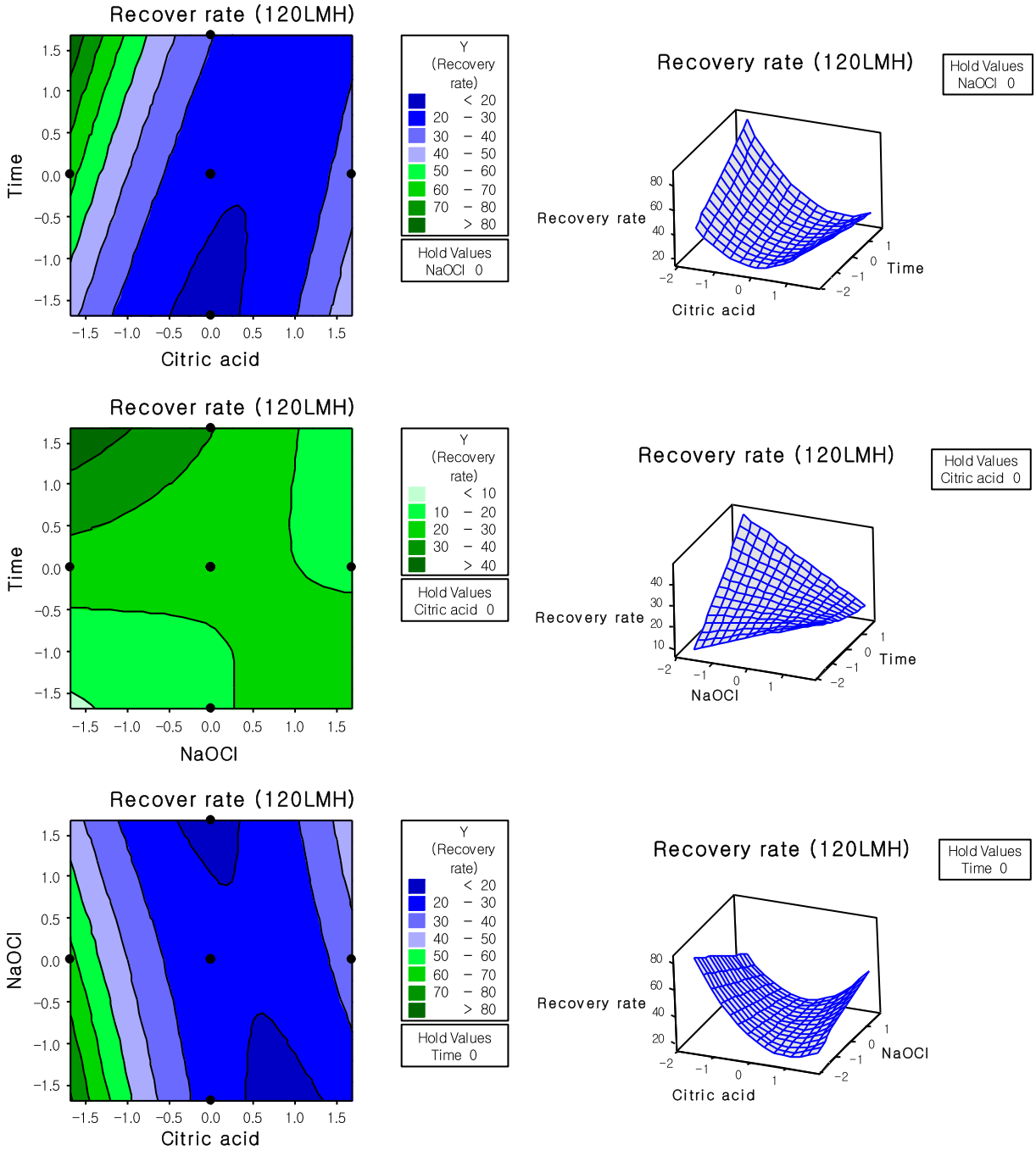

Fig. 8. Response surface of the effects of three independent variables for recovery rate on flux at 120 L/m^2 h; Y_4.

operation condition of chemical cleaning in a pilot plant. At this time, since the results were intended to be applied to full-scale membrane modules, we had to follow certain criteria. This is why we could not reduce the citric acid concentration.

An increase in cleaning time may result in a better result. However, we would like to compare the cleaning efficiency within a reasonable time range. Increasing cleaning time over 5 h may not be very practical because it increases the plant downtime.

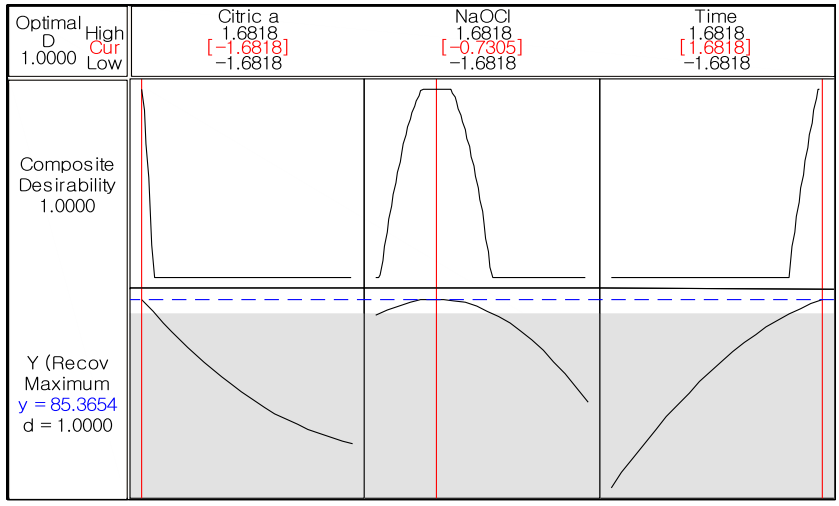

Fig. 9. Response optimizer of the effects of three independent variables for recovery rate on flux at 120 L/m^2 h.

4. Conclusions

To optimize the cleaning condition for MF membranes, a set of experiments based on RSM was conducted. The results suggested that this method is effective to obtain the conditions to minimize chemical dose and cleaning time without scarifying the cleaning efficiency. The following conclusions can be drawn:

(1) Experimental results indicated that the efficiency of chemical cleaning is sensitive to the concentration of cleaning chemicals (citric acid and NaOCl) as well as cleaning time. Nevertheless, the dependency of cleaning efficiency on these parameters was different. The cleaning efficiency which is expressed as the recovery of membrane permeability after recovery rate varied from 0 to 72%.
(2) RSM was used to examine the fouling control and the rejection rate as a function of chemical cleaning. The fouling rates were successfully predicted by the second-order polynomial equations. This technique could also be used to investigate the interactions with effect factor such as dose of acid, dose of base, treatment time, and so on.
(3) The RSM analysis could suggest the optimum conditions for membrane cleaning. The optimum conditions were determined as follows: The conditions of three independent variables (citric acid, sodium hypochlorite, treatment time) were 1,600 ppm (−1.68179), 580 ppm (−0.73076), and 5 h (1.68179), respectively, on flux at 30 L/m^2 h. According to the RSM prediction, the recovery rate under this condition is 85.3%.
(4) Moreover, this approach may have potential to be used for process control of pilot or full-scale membrane plant.

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