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Evaluation of mechanical membrane cleaning with moving beads in MBR using Box–Behnken response surface methodology

Se Na Shim, Sang-Ryoung Kim, Sung Jun Jo, Kyung-Min Yeon, Chung-Hak Lee*

School of Chemical and Biological Engineering, Seoul National University, Seoul 151-744, Korea, Tel. +82 2 874 0896; emails: senaraha@hanmail.net (S.N. Shim), super3376@naver.com (S.-R. Kim), josungjun@snu.ac.kr (S.J. Jo), yeonkm76@hanmail.net (K.-M. Yeon), Tel. +82 2 880 7075; email: leech@snu.ac.kr (C.-H. Lee),

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ABSTRACT

Incorporation of moving beads into membrane bioreactor (MBR) has been suggested as an effective membrane fouling control because moving beads can mechanically remove bio-cakes on the membrane surface without additional equipment and energy input. As the efficiency of fouling control is dependent on factors associated with moving beads, however, the design of experiment was applied to find optimum condition for the effective mechanical cleaning with moving beads in MBR. Bead diameter (mm), bead number, and aeration rate (m^3/h) were selected as independent design parameters. Based on batch test results, the correlation between the detachment efficiency of bio-cakes and three design parameters was established using Box–Behnken methodology. When all three design parameters at their optimal conditions ($bead_{opt}$) were extended to the continuous lab-scale MBR, membrane filterability increased by three times, compared with that in the control MBR without bead. On the other hand, each of five experimental sets with two optimal and one random parameter ($bead_{random}$) showed less membrane filterability by 9–80%, respectively, compared with $bead_{opt}$. The parameter *C* (aeration rate) affected most significantly as it is associated with not only bead movement but also shear induced by air bubbles.

Keywords: MBR; Moving bead; Mechanical cleaning; Box-Behnken design; Response surface methodology (RSM); Wastewater treatment

1. Introduction

Membrane bioreactor (MBR) has become one of the key technologies for advanced wastewater treatment and reuse [1]. However, fouling caused by the inevitable formation of bio-cake layer on the membrane surface remains the main bottleneck that limits its widespread use [2]. For example, one case study of a commercial MBR with capacity of 5 MLD (mega liter per day) has reported that operating cost consisted of fouling related factors such as aeration energy for fouling control (37%), cleaning chemicals (8%), and membrane replacement (15%) [3]. Many researchers have attempted to mitigate membrane fouling in various ways such as process configuration [4,5], fouling resistance membrane materials [6–8], and chemicals targeting main foulants [9,10].

Moving beads have been used in a submerged-type MBR for wastewater treatment to remove bio-cakes on membrane surface through physical friction [11] and/or to provide carriers for functional

*Corresponding author.

microorganisms (e.g. Nitrifying bacteria) [12]. Incorporation of moving beads in MBR can be one of the effective physical fouling control methods because it requires neither additional equipment nor energy input [11,13]. It can also reduce dosage of harsh cleaning chemicals such as sodium hypochlorite which may affect the span of membrane life and the microbial activity [14]. Furthermore, recently Kim et al. [15] added into MBR the beads entrapping quorum quenching bacteria and revealed that those moving beads greatly enhanced membrane permeability through a synergic effect of biological quorum quenching and physical washing. On the other hand, the moving beads can damage the membrane surface. So, the form of moving beads should be regular (lens or sphere) without sharp edges and high elasticity to avoid membrane damage [13].

Mechanical cleaning with moving beads may be affected by various factors such as bead size, bead packing density, and aeration intensity, etc. These factors have reciprocal action, which means that independent design parameters connected with one another determine the optimal design point. Therefore, the conventional one-variable-at-a-time method, which tests one parameter at a time holding other parameters constant, is not an efficient optimization approach because the interaction or quadric effect of parameters could be ignored [16,17].

Response surface methodology (RSM) is a collection of mathematical and statistical techniques that are useful for modeling and analyzing engineering problems [18,19]. The application of RSM to design optimization is aimed at reducing the cost of expensive analysis methods and their associated numerical noise. While the proper choice of an experimental design is very important for fitting and analyzing response surfaces, Box–Behnken experimental design (BBD) which is formed by three-level designs for fitting response surfaces is known to be very efficient in terms of the number of required runs [20,21]. However, there is little information available on the effects of bead diameter/number and aeration rate on the accumulation of bio-cake on the membrane surface in MBR with moving beads using a combination of operating parameters through BBD.

The purpose of this study was to model the optimal condition for moving beads in MBR to make the most use of them. Three-level, three-factorial BBD was used to find the optimal condition for the batch test sets in terms of three parameters (bead diameter, bead number, and aeration rate). The optimal value of each parameter was determined and the validity of optimum parameters was examined in a continuous lab-scale MBR fed with synthetic wastewater.

2. Materials and methods

2.1. Preparation of moving beads

Ca-alginate beads were adopted as model moving beads and prepared according to procedures described by previous studies [15,22]. Four percentage (w/v) of sodium alginate solution was dripped into well stirred 3% (w/v) $CaCl_2$ solution using a peristaltic pump through a single nozzle at a constant injection rate of 2 mL/min. The size of alginate beads was controlled by changing the nozzle diameter. After being hardened in the $CaCl_2$ solution for 10 h, the alginate beads were washed twice with distilled water and dried for 1 h at room temperature.

2.2. Box–Behnken design

The influence of bead diameter, bead number, and aeration flow on the detachment of bio-cake from membrane was investigated at high (+1), middle (0), and low (−1) levels. Independent variables, their levels and symbols are presented in Table 1. The 15 experiments of the Box–Behnken design are presented in Table 2. A total of 15 experiments were required to obtain a quadratic model consisting of 12 trials plus 3-center points. The 3-center point (Run order 13–15) runs were added for the measurements of process stability and inherent variability [20,21]. The experiments were performed randomly to avoid systematic errors. Minitab statistical software was used to analyze the experimental data. A following quadratic polynomial model was defined to fit the response [23]:

$$Y = b_0 + \sum b_i X_i + \sum b_{ii} X_i^2 + \sum b_{ij} X_i X_j$$

where Y: predicted response, b_0: constant coefficient, b_i: linear effect coefficient, b_{ij}: interaction effect coefficient, b_{ii}: quadratic effect coefficient.

The fitting quality of the polynomial model equation was expressed by the coefficient of determination R^2.

2.3. Batch test for the bio-cake detachment

The efficiency of mechanical cleaning by moving beads was evaluated as the percentage of bio-cake detachment in batch reactor (Fig. 1(a)). For this purpose, lab-scale MBR with working volume of 20 L operated. When trans-membrane pressure (TMP) reached 60 kPa, the used hollow fiber membrane module was taken out of each MBR and immersed into an aeration tank filled with 2.6 L of distilled water and

Table 1
Independent variables, their levels and symbols for Box–Behnken design

Variable	Symbol	Variable levels		
		Low (−1)	Middle (0)	High (+1)
Bead size (mm)	*A*	2.5	3.5	4.5
Number of bead	*B*	50	125	200
Aeration rate (m^3/h)	*C*	0.024	0.042	0.06

Table 2
Experimental set of Box–Behnken design for batch test

Run order	Coded factors			Uncoded factors		
	A	*B*	*C*	*A*	*B*	*C*
1	−1	−1	0	2.5	50	0.042
2	+1	−1	0	4.5	50	0.042
3	−1	+1	0	2.5	200	0.042
4	+1	+1	0	4.5	200	0.042
5	−1	0	−1	2.5	125	0.024
6	+1	0	−1	4.5	125	0.024
7	−1	0	+1	2.5	125	0.06
8	+1	0	+1	4.5	125	0.06
9	0	−1	−1	3.5	50	0.024
10	0	−1	+1	3.5	50	0.06
11	0	+1	−1	3.5	200	0.024
12	0	+1	+1	3.5	200	0.06
13	0	0	0	3.5	125	0.042
14	0	0	0	3.5	125	0.042
15	0	0	0	3.5	125	0.042

moving beads. Then, aeration was conducted with moving beads for 3 h to detach bio-cakes from the membrane module and then the weight of detached biocake was measured. As a next step, aeration tank was further sonicated for 30 min without bead to further remove the remaining bio-cakes from the membrane surface. Finally, the percentage of the detached bio-cakes by moving beads was calculated by the ratio of detached bio-cake to total attached bio-cake.

2.4. MBR operation

Two lab-scale MBRs with working volume of 2.6 L (Fig. 1(b)) were run in parallel with synthetic wastewater whose composition was as following: glucose 306.75 mg/L, peptone 115 mg/L, yeast extract 14 mg/L, $(NH_4)_2SO_4$ 104.75 mg/L, KH_2PO_4 21.75 mg/L, $MgSO_4{\cdot}7H_2O$ 32 mg/L, $MnSO_4{\cdot}5H_2O$ 2.88 mg/L, $FeCl_3{\cdot}6H_2O$ 0.13 mg/L, $CoCl_2{\cdot}6H_2O$ 1.25 mg/L, $CaCl_2{\cdot}H_2O$ 3.25 mg/L, and $NaHCO_3$ 255.5 mg/L. The effective area of hollow fiber membrane module (GE-Zenon, US) was 160 cm^2. The membrane material was hydrophilic polyvinylidene fluoride (PVDF) with a pore size of 0.04 μm. Constant permeate flux and hydraulic retention time (HRT) were set to 27 L/m^2/h and 8 h, respectively. Mixed liquor suspended solids (MLSS) were measured to be in the range of 8,000–8,500 mg/L and 87 mL of sludge was withdrawn daily from the mixed liquor in MBR to adjust solids retention time (SRT) to 30 d.

3. Results and discussion

3.1. Responsible model for mechanical cleaning by alginate moving beads

Both experimental and predicted results of 15 biomass detachment batch test set designed by Box–Behnken method were summarized in Table 3. The following second-order polynomial equation was established to explain the relationship between biocake detached from membrane a fouling state and design parameter group:

$$Y(\%) = -165.7 + 75.8A + 0.4B + 1.7 \times 10^3 C - 9.9A^2 - 1.5 \times 10^{-3} B^2 - 1.5 \times 10^4 C^2 \quad (1)$$

where *Y*: percentage of detached bio-cake by moving beads, *A*: diameter of alginate bead (mm), *B*: number of alginate bead (beads), *C*: aeration flow (m^3/h).

In this model equation, the adjusted correlation coefficient was determined to be 0.903, which indicates reliable predictive capability of the model equation [24]. Simultaneously, statistical significances of regression equation were checked. Analysis of variance and effectiveness of each input feature was determined using backward elimination method. As shown in Table 4, interaction variables of *AB*, *BC*, and *CA* were determined to be insignificant when compared to pure error [16]. On the other hand, both of linear and 2nd order variables of bead diameter (*A*), number (*B*) and aeration flow (*C*) were determined to be significant. *p*-Value for "lack of fit" was calculated to be 0.237, which revealed that this model was highly significant ($p > 0.05$) [24,25].

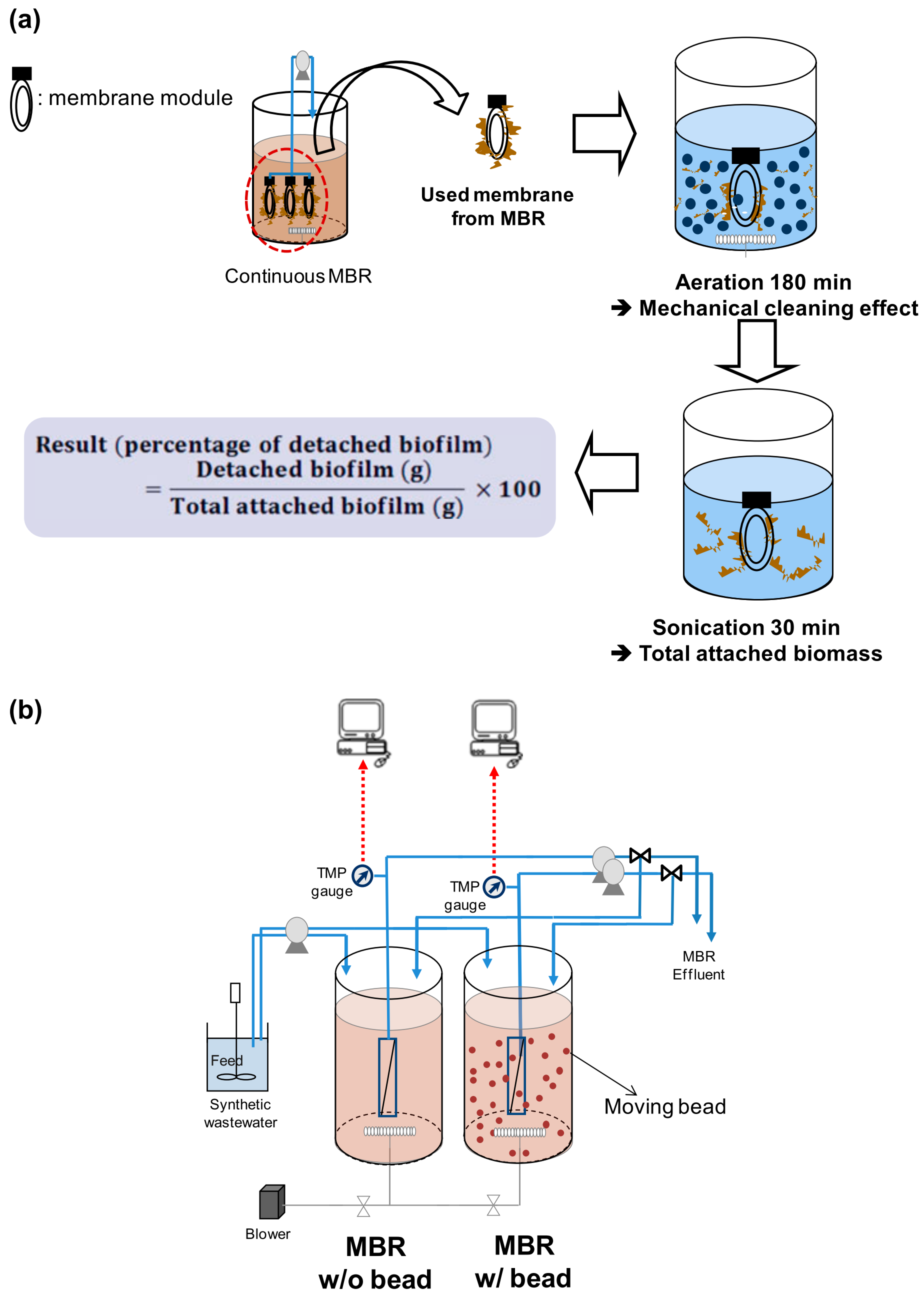

Fig. 1. Schematic diagram of (a) batch reactor for bio-cake detachment and (b) parallel operation of lab-scale MBRs.

3.2. Residual analysis

Residual was defined as the difference between the experimental value and the predicted value. Residual was analyzed to assess the appropriateness of the overall model equation established by Box–Behnken surface method. Normal probability plot (Fig. 2) clearly shows the linearly, which means the error terms are normally distributed. The scatter plot of residuals vs. fitted values and the scatter plot of residuals vs. order of data were confirmed. The residuals had equal variance through the scatter plot of residuals vs. fitted values. The scatter plot of residuals vs. order of data suggested that the error term was independent from order during the experimental process. Consequently, the assumptions of the residual fulfilled the normality, homoscedasticity, and time independence, which validate the model fitness.

Table 3
Experimental and predicted values of batch test for bio-cake detachment

Experiment	Bead size (mm)	Number of bead	Aeration rate (m^3/h)	Result (%)	
				Experimental	Predicted
1	2.5	50	0.042	26.8	22.9
2	4.5	50	0.042	33.8	35.6
3	2.5	200	0.042	30.4	29.6
4	4.5	200	0.042	40.0	42.4
5	2.5	125	0.024	19.9	21.9
6	4.5	125	0.024	35.7	34.6
7	2.5	125	0.060	35.3	37.5
8	4.5	125	0.060	53.8	50.3
9	3.5	50	0.024	28.5	26.6
10	3.5	50	0.060	33.2	42.2
11	3.5	200	0.024	37.7	33.3
12	3.5	200	0.06	52.2	49.0
Center point	3.5	125	0.042	50.7	50.8
Center point	3.5	125	0.042	49.0	50.8
Center point	3.5	125	0.042	52.9	50.8

Table 4
Regression analysis of variables of (a) raw data-set and (b) data-set after backward elimination of variables

Variable	Coefficient	*p*-value
(a)		
Constant	−146.88	0.000
A	73.15	0.004
B	0.31	0.033
C	1.31×10^3	0.002
A^2	−9.92	0.003
B^2	-1.46×10^{-3}	0.006
C^2	-1.47×10^4	0.047
AB	8.67×10^{-3}	0.726
AC	37.5	0.716
BC	1.81·	0.221
Model	–	0.005
Lack of fit	–	0.181
$R^2 = 0.893$		
(b)		
Constant	−165.71	0.000
A	75.80	0.001
B	0.41	0.015
C	1.67×10^3	0.000
A^2	−9.92	0.000
B^2	-1.46×10^{-3}	0.001
C^2	-1.47×10^4	0.025
Model	–	0.000
Lack of fit	–	0.237
$R^2 = 0.903$		

3.3. Determination of optimal condition

Optimal range of each parameter (diameter, number, and aeration flow) which can achieve more than 50% of bio-cake detachment was determined using two-dimensional contour plots (Fig. 3). Considering that the regression model had three independent variables, one variable was fixed as constant at the central level for each plot.

At a constant aeration flow of 0.042 m^3/h, bead diameter and numbers had the optimal range of 3.3–4.3 mm and 102–181 beads, respectively (Fig. 3(a)). When bead number was set to 125, diameter and aeration rate was shown to be in the range of 3.5–4.1 mm and 0.048–0.060 m^3/h, respectively (Fig. 3(b)). For alginate bead with the diameter of 3.5 mm, effective range of each number and aeration was determined to be 87–195 beads and 0.04–0.06 m^3/h, respectively (Fig. 3(c)). Simultaneously, three-dimensional response surface plots for each fixed parameter value was constructed based on the model equation herein [26]. Response surface plot at each holing parameter value clearly displayed that there was the stationary point of each design variable within range of interest: diameter (2.5–4.5 mm), number (50–200 beads), and aeration rate (0.025–0.060 m^3/h).

Finally, the optimal condition of each of the three parameters was obtained using Minitab optimizer (Fig. 4). As a result, the optimum value of each bead parameter was determined to be 3.8 mm (diameter),

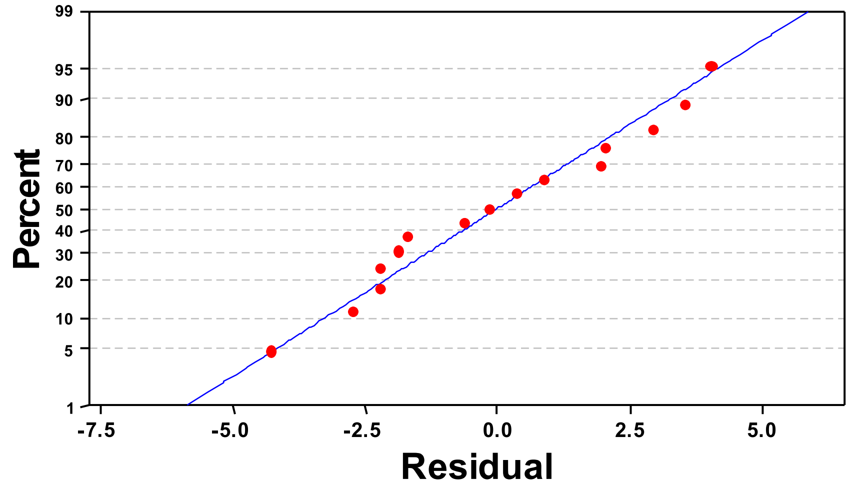

Fig. 2. Residual plot of model for error values: normal probability plot of residuals.

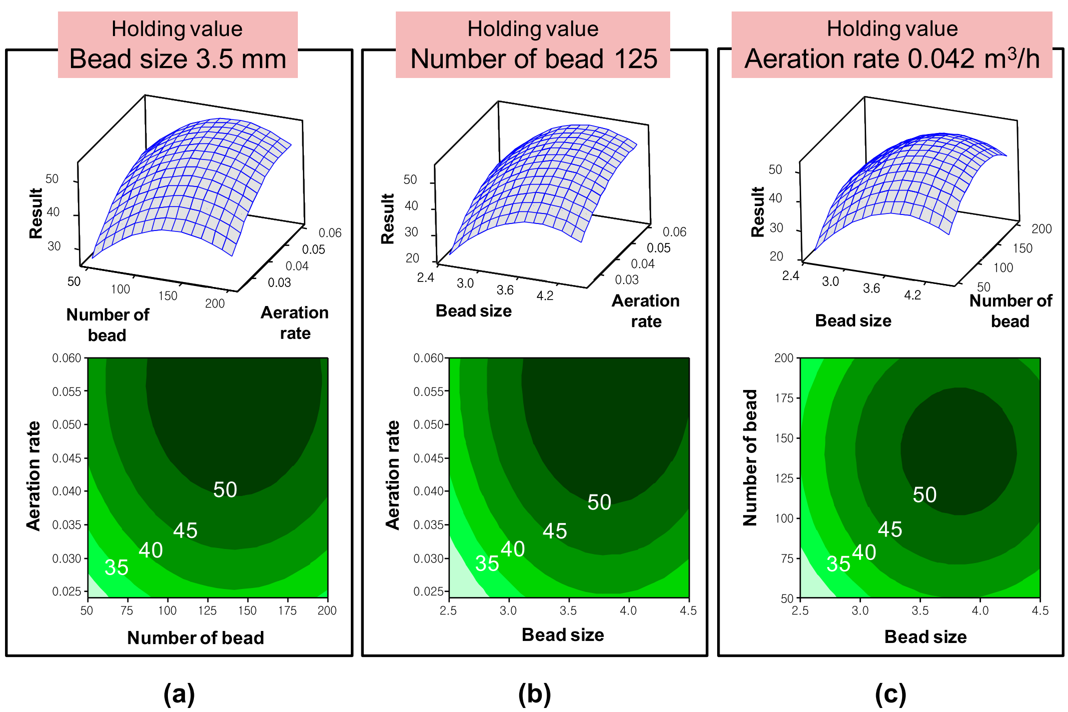

Fig. 3. 2D contour and 3D responsive surface plots for the effect of two parameters with one fixed value of (a) bead diameter, (b) number, and (c) aeration rate.

140 beads (number), and 0.057 m^3/h (aeration rate) for the maximum cleaning efficiency.

3.4. Model feasibility test in continuous MBR operation

As a final step of this study, feasibility of Box–Behnken responsive surface method for optimum operation was checked in the continuous MBR system. At first, all three bead parameters were set to their optimal value ($bead_{opt}$) and the permeability of MBR with moving beads was compared with that of conventional MBR without moving bead (Fig. 5). In control MBR, it took about 22 h for TMP to reach 40 kPa, at which module should be replaced due to severe fouling. However, MBR with moving bead showed that the filtration time to the same TMP point was extended by about three folds. This indicates that the

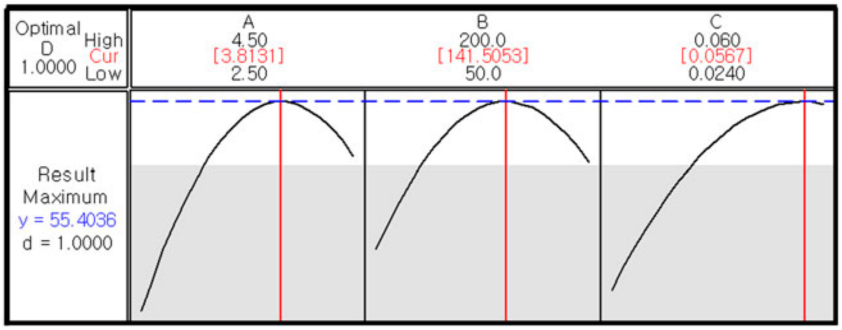

Fig. 4. Responsive optimization of each parameter of diameter (*A*), number (*B*), and aeration rate (*C*). Each symbol of "*y*" and "*d*" indicates response at the determined condition and composite desirability.

moving bead technology could effectively alleviate fouling during continuous MBR operation. As a next step, five experiments with two optimal and only one random parameter change was conducted and its filtration performance was compared with that of MBR with bead$_{opt}$. Briefly, the effect of bead diameter variance was checked through two MBR operation runs where diameter values were set to 3.0 and 4.5 mm, respectively. In case of bead number, two MBR runs with 70 and 200 beads was conducted. For aeration rate, only one MBR run with aeration of 0.030 m^3/h was compared with MBR run with bead$_{opt}$ condition because the optimal aeration rate of 0.057 m^3/h was close to the designed high level (0.060 m^3/h). As a result, Fig. 6 clearly shows that filterability of all cases decrease by 9–80% compared to that of bead$_{opt}$ condition, which confirms that optimal points determined by Box–Behnken method can be directly applied to mechanical cleaning by moving beads in continuous MBR operation. One main concern about this optimization frame is that the basic correlation was established based on not continuous but batch reactor. To confirm the degree of this discrepancy between batch and continuous system, all six experimental sets for continuous MBR run were checked in terms of their predicted bio-cake detachment efficiency and rate of TMP delay (Table 5). Bio-cake detachment efficiency of five random experimental conditions was in the range of 42–47%, which is slightly lower that 52% of bead$_{opt}$ condition. Furthermore, TMP delay ratio of each MBR runs displayed similar tendency with that of relative bio-cake detachment except for the random aeration condition of 0.030 m^3/h. That is to say, MBR runs with this partial aeration flow deviation showed TMP delay of only 55%, which is much lower than expected relative bio-cake detachment efficiency of 81%. One of the main reasons for this difference was that aeration

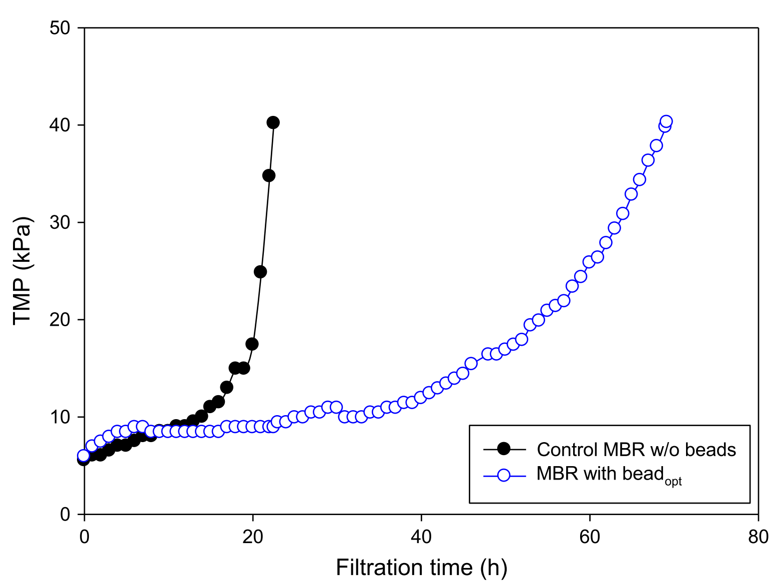

Fig. 5. Effect of fully optimized bead condition (bead$_{opt}$) on the MBR performance.

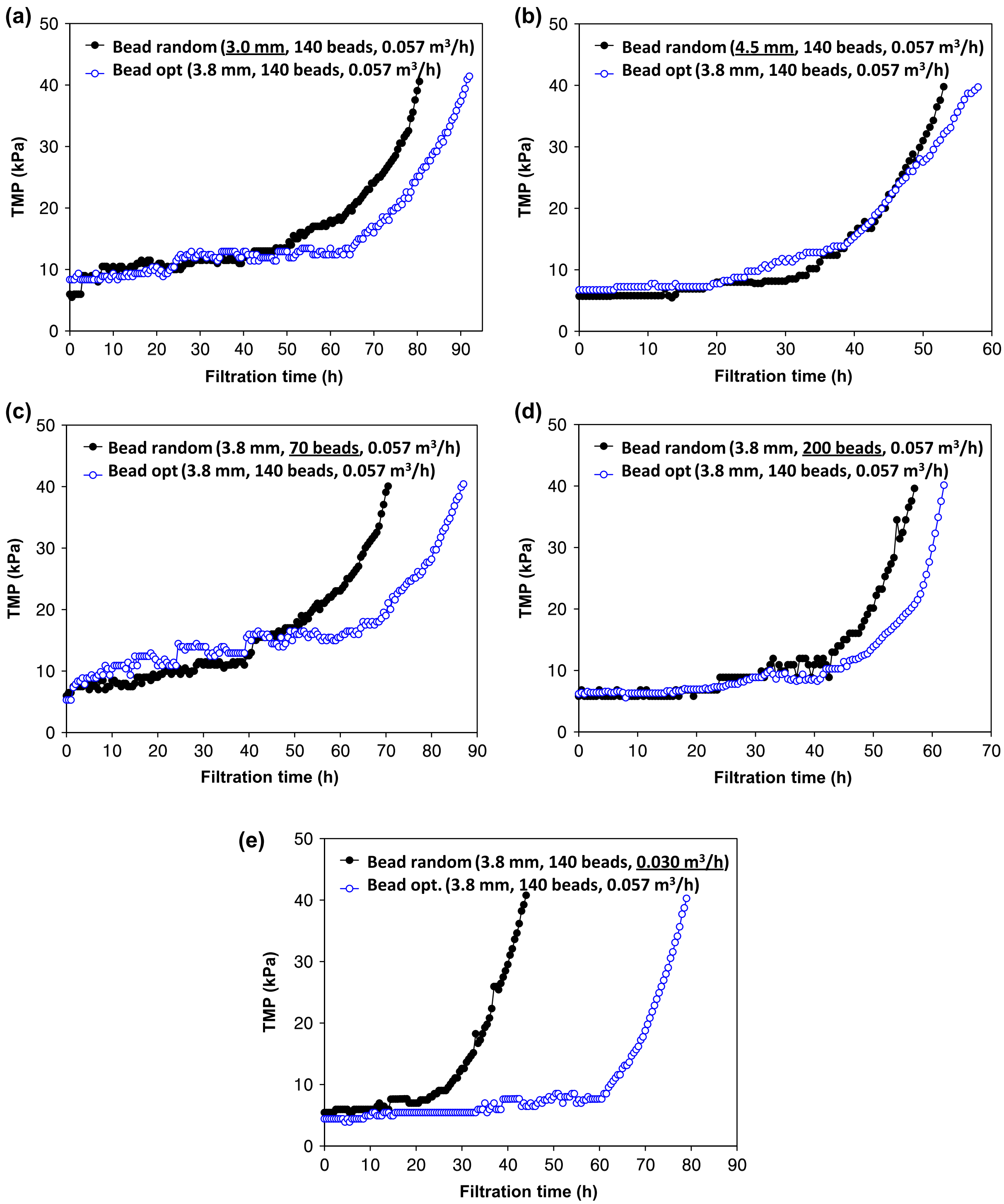

Fig. 6. Effect of partial variance of single parameter on the MBR performance. (a) Low diameter level (3.0 mm), (b) high diameter level (4.5 mm), (c) low number level (70 beads), (d) high number level (200 beads), and (e) low aeration level (0.030 m^3/h).

affects not only the movement of alginate bead but also shear force, all of which can mechanically remove the surface biomass. This implies that aeration rate is more significant than other two parameters. All of these results clearly support that optimization through simple batch tests designed by Box–Behnken method can be successfully extended to the continuous MBR operation.

Table 5
Predicted bio-cake detachment efficiency and rate of TMP delay at each continuous MBR operation set

	Parameters			Bio-cake detachment (%)	Relative bio-cake detachment (%)	TMP delays compared to $bead_{opt}$ (%)
	Diameter	Number	Aeration			
$Bead_{opt}$	3.8	140	0.057	52.17	–	–
Diameter	*3.0*	140	0.057	45.38	86	88
	4.5	140	0.057	47.71	91	91
Number	3.8	*70*	0.057	46.22	88	81
	3.8	*200*	0.057	45.57	87	93
Aeration	3.8	140	*0.030*	42.44	81	55

4. Conclusions

In this study, design of experiment (DOE) was used to optimize the engineering parameters of polymeric moving beads for the effective physical cleaning in MBR system. Optimal value of three parameters including bead diameter, number, and aeration rate could be obtained with highly significant level through 15 batch tests designed by Box–Behnken responsive surface method. The validity of these optimal design conditions was confirmed in terms of an efficient fouling alleviation in a continuous MBR. Aeration flow was turned out to be more significant parameter on the fouling control than the other two (bead number and bead diameter). All these results clearly show the potential of DOE as a simple and effective optimization tool in MBR system with moving beads for mechanical cleaning.

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