
ABSTRACT

In this study, a cavitation effect was used to remove H₂S which is one of the main causes behind complaints of offensive odors. The result of the optimization gas flow rate and water flow rate to remove H₂S using the central composite method showed that the removal rate of H₂S increases as the gas flow rate decreases and the water flow rate increases at the gas flow rate of 1.39 LPM and water flow rate of 2.16 LPM, the maximum removal rate of H₂S through reaction optimization was estimated to be 96.1 %.

KEYWORDS: H₂S, Hydrodynamic Cavitation Effect, Odor.

INTRODUCTION

The rapid increase in the emission of atmospheric pollutants recently has caused serious environment problems, while public awareness of consequent unpleasant odors has also been increasing markedly as a result of improving living standards. One of the causes of the foul odors is the presence of hydrogen sulfide (H₂S). H₂S is a colorless substance that smells like rotten eggs and can cause suffocation, lung disease and neural paralysis

The technologies in handling offensive odors caused by emissions include absorption, adsorption, combustion, washing, and the use of catalysts and bio-filters. Each of these methods has positive and negative effects, and there are active ongoing studies into their relative effectiveness. The use of catalyst methods, for example, has proven to be highly efficient but it may be toxic and there are also other problems such as high reaction temperatures, high installation and maintenance costs, degradation of efficiency at higher concentrations, and the need for regular replacement. The adsorption method, such as the use of conventional activated carbon, is widely applied in commercial processes, but it's low, and the adsorbent must be recharged in large amounts, thus the development of low cost and high performance adsorbents is needed. The bio-filter method is a technology that works by passing polluted gas through biologically active materials such as soil or compost. Although it has the strength of having low capital and operating costs than physical or chemical processes and also discharges no secondary pollutants, it is vulnerable to pollution load, and the removal efficiency varies according to the type of carrier and microorganism (Hodge et al., 1993; Hodge et al., 1994; Leson et al., 1991; Zilli et al., 1993).

In South Korea, absorption is used to process the offensive odors generated from most industrial complexes. However, the absorption method is problematic because of the low removal efficiency for some substances and though not focusing particularly on offensive odors, studies aimed at solving environmental issues by generating cavitation using hydrodynamic venturi tubes are currently ongoing (Muller et al., 1998). When fluid passes through the neck part of the venturi tube, i.e. the central part of the tube where it becomes constricted, air bubbles called cavitation bubbles are generated. The offensive odors can then be removed by high temperature shock waves as these cavitation bubbles are destroyed as they pass into area of the tube beyond the constriction where the pressure has recovered (Pandit and Joshi, 1993).

This study intends to conduct a test to discover the optimum gas input to remove H₂S and water input to generate cavitation in order to maximize the removal rate of the offensive odor substance H₂S and to present a mathematical model using the central composite design method which is one of the response surface methodologies.

MATERIALS AND METHODS

Figure 1 shows the schematic diagram of the experimental apparatus used in this study. The apparatus consists of a polluting gas (H_2S) supply unit, cavitation generation unit, water tank, circulation pump, and measurement unit to analyze the polluting gas before and after the reaction. The internal diameter of the inlet and outlet parts of the venturi tube used for generating cavitation was 10mm while the constriction at the neck area was 1mm. The angle of the declining diameter from the inlet to the neck was 20° while the angle of the increasing diameter from the neck to the outlet was 12° . The angles were the optimum values calculated for the lab environment of Bae et al (Kim et al., 2006). Two water tanks, one for the cavitation and another one for the role of water circulation, were serially installed. The 100mm diameter and 500mm height water tanks were fabricated with acryl material to allow observation of the inside. Two circulation pumps were mounted to adjust the amount of water flowing into the water tank, and the liquid levels in the tanks were maintained at 250mm during the test. To measure the H_2S removal rate by cavitation, the H_2S concentration was measured using an automatic analyzer before and after the reaction.

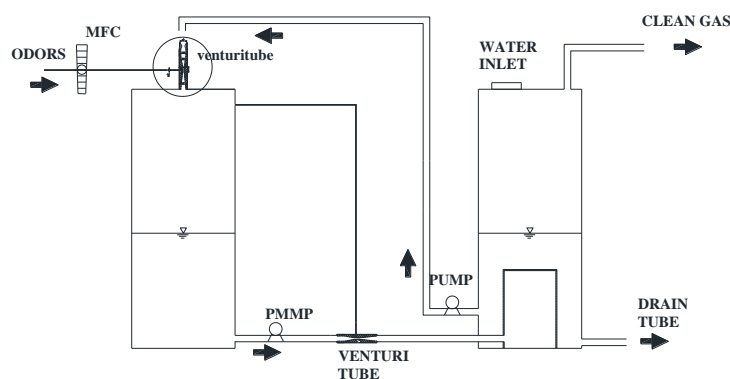


Fig. 1. Schematic diagram of experimental test apparatus.

To observe the characteristics of the H_2S gas removal rate by cavitation, the operating conditions for the test were altered as shown in Table 1. In the gas supply unit, a device to inject the H_2S gas and pure air for dilution was installed, and the concentration of H_2S gas injected into the cavitation generation unit was set to 1000ppb. The flow rate of H_2S gas into the reactor was then operated, ranging from 1.4 to 2.1 L/min. The flow rate of water into the cavitation system was adjusted to a 0.8 to 2.2 L/min range, which was adjusted by the rotation of the water circulation pump. The flow rate at the inlet of the venturi tube where cavitation occurs ranged from 0.04 m/sec to 0.11 m/sec, and the flow rate at the neck part ranged from 4 m/sec to 11 m/sec.

Table 1. Design and operation parameters

Parameters	Conditions in the experiment
Initial concentration of H_2S (ppb)	1000
H_2S gas flow rate (L/min)	1.4 ~ 2.1
Water flow rate (L/min)	0.8 ~ 2.2

To review the characteristics of H_2S removal and the optimum inlet flow using cavitation in the reactor fabricated for this study, the H_2S concentration was measured using the automatic H_2S analyzer (model: S5065 by SIR S.A. (Spain)) before and after the reaction. Pure air for dilution was used to maintain the concentration of H_2S gas going into the reactor at 1000ppb. The inlet concentration was measured after the initial concentration became stabilized for about

30 minutes, and then the H₂S was injected into the reactor. After the H₂S removal, the operation was continued until the concentration of the H₂S remained constant and the concentration was then recorded. The analysis instrument used in this study was an automatic continuous analyzer which can relay data every 5 second and uses UV fluorescence for measurement.

RESULTS AND DISCUSSION

Characteristics of H₂S removal

To observe the characteristics of H₂S removal using cavitation according to the inlet flow rate, the H₂S inlet concentration was fixed at 1000ppb and the flow rate was set at from 0.5LPM to 2LPM. Figure 2 shows the removal efficiency of H₂S according to the flow rate of water injected to generate cavitation. Although the graph shows that the H₂S removal rate increased as the injected water flow rate increased, this study conducted further tests to analyze the optimum amount of water that should be injected.

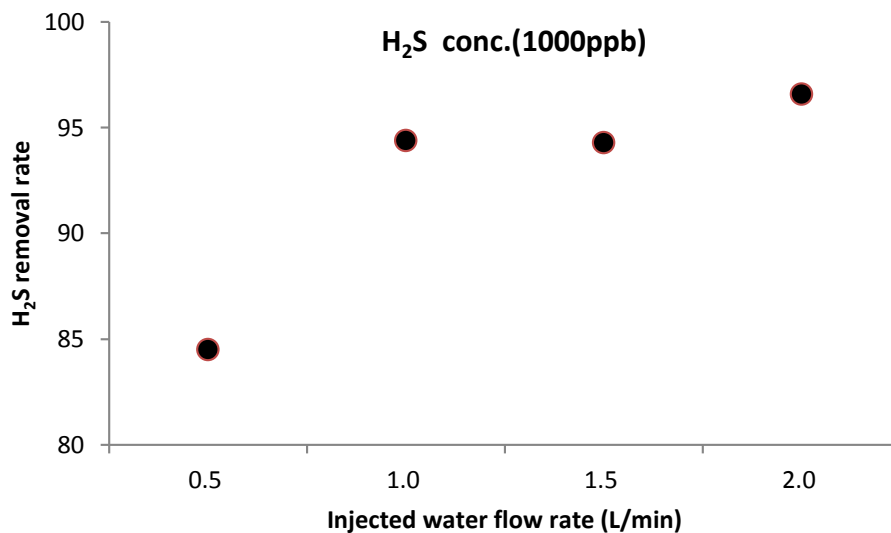


Fig. 2. Efficiency of H₂S Processing according to Injected Water Flow Rate

Reaction surface analysis

The reaction surface analysis method was used to optimize H₂S removal using cavitation. To understand the process of maximizing the removal rate of H₂S, the central composite design method including inlet and water flow rate, one of the response surface methodologies, was used. The central composite design method is generally used in reaction surface design and widely used to efficiently estimate the primary and secondary parameters and to model curvature response variables through an additional test after the factorial design test. Since the reaction surface equation is generally not known, an approximate model is assumed, and its suitability is evaluated through lack-of-fit (LOF) test. Equation (1) shows the secondary model by the central composite design method (Box and Behnken, 1959; Box and Behnken, 1960).

$$Y = \beta_0 + \beta_1 x_{1i} + \beta_2 x_{2i} + \beta_{11} x_{1i}^2 + \beta_{22} x_{2i}^2 + \beta_{12} x_{1i} x_{2i} + \epsilon \quad \epsilon \sim N(0, \sigma^2) \quad (1)$$

This consists of the factorial design points (vertices) including the central point and axial points. If the number of independent variables is k, the number of factorial design points will be 2^k. Since the number of axial points is 2k, and if the repetition counts at the central point is n_c, the total number of tests will be 2^k + 2k + n_c. Figure 3 shows the removal rate of H₂S according to the analysis condition of the central composite design. A total of 5 repeated tests were conducted at the central point using the flow rates of the inlet gas and inlet water as independent variables and 4 factorial design test points (vertices) and 4 axial points as the analysis condition.

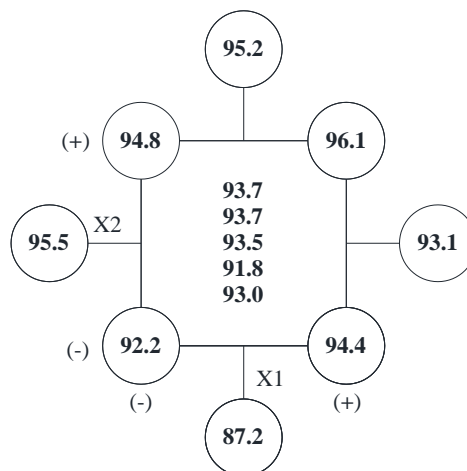


Fig. 3. Experimental levels and results of central composite design (2^2+2+5 , X1: gas flow rate, X2: water flow rate)

Table 2. Coefficients of the model [Eq. (1)] for H_2S degradation using the soil biofilter.

Coefficient	Value	Standard Error	T-value	P-value
Constant	93.6391	0.4066	230.271	0.000
α_1	-0.5243	0.3981	-1.317	0.220
α_2	2.4892	0.3981	6.253	0.000
α_1^2	-0.9261	0.4233	-2.188	0.056
S = 1.126		$R^2 = 83.5\%$		$R^2(\text{revision}) = 78.0\%$

Table 2 shows the result of T analysis of the model coefficients. The analysis showed that the secondary parameters of the gas inlet vs. gas inlet and water inlet vs. water inlet were not significant, thus they were excluded from the model. Although the P-value (0.220) of the gas inlet seems insignificant, which was higher than the substantial level 5%, the main parameter cannot be removed if the interaction parameter or secondary parameter containing the main parameter was significant. As a result, the following equation can be obtained for the secondary reaction surface model estimated by the analysis. It proves that there is a curvature effect between the amount of gas injected for oxidation of H_2S and the reaction parameter.

$$\text{Removal rate (\%)} = 81.5066 - 2.09706 x_1 + 16.915 x_2 - 3.70435 x_1^2 \quad (2)$$

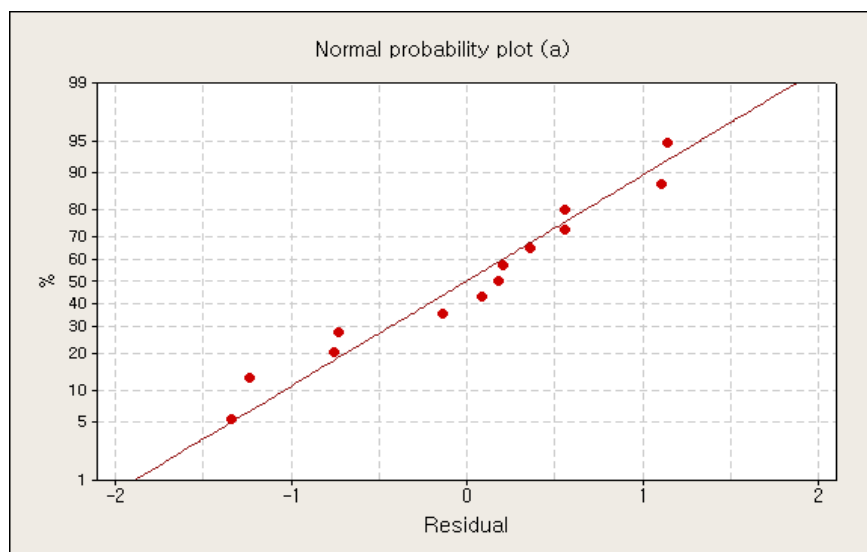
Table 3. Regression analysis and response surface model fitting (ANOVA) for degradation of H_2S .

Source	Degree of freedom	Sum of squares	Mean squares	F-value	P-value
Regression analysis	3	57.8377	19.2792	15.21	0.001
Linear	2	51.7683	25.8841	20.42	0.000
Square	1	6.0694	6.0694	4.79	0.056
Residual error	9	11.4100	1.2678		
Lack of fit (LOF)	5	8.8380	1.7676	2.75	0.174
Pure error	4	2.5720	0.6430		
Total	12	69.2477			

Table 3 shows the result of a regression analysis and a variance analysis to review the feasibility of the estimation model to calculate the optimum gas inlet and water inlet. A mathematical model is considered not to be feasible if the P-value of LOF in the ANOVA is less than the significant level. Since the P-value of the LOF of the result was 0.174, and is considered to be higher than the 5% significant level, the mathematical secondary model can be considered to be suitable.

To further verify whether the model was applicable the assumptions of the error terms of the model were checked using a residual normal probability plot, residual vs. adequacy, and residual vs. data order graphs as shown in Figure 3.

Fig. 4(a) shows the residual normal probability plot, this is to check if the residuals significantly deviate from normal distribution. If the residuals are within normal distribution, the points will be in near linear form, and that this graph proves that the data residuals of reaction values do not deviate significantly. The residuals vs. adequacy plot in Fig. 4(b) shows that the residuals are evenly distributed above and below the 0 line and thus proves that there is no evidence of a violation of the assumption. Fig. 4(c) shows the residuals vs. data order, this is whether to check if the residuals are independent and random. If the residuals are not affected by data order, they will be scattered randomly around 0, and the plot shows that there is no evidence that the reaction values are not affected by time and order.



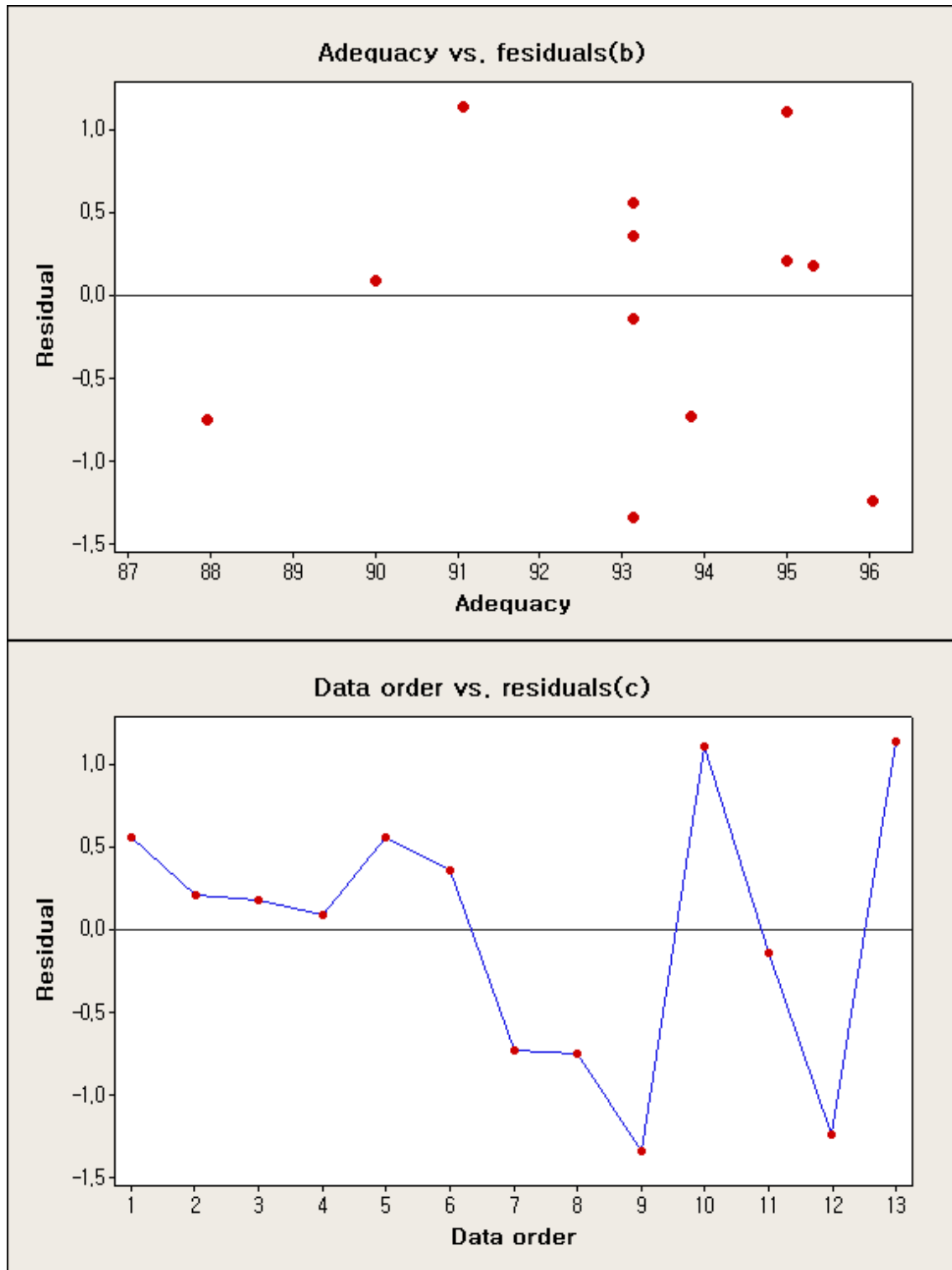


Fig. 4. Model adequacy checking plot

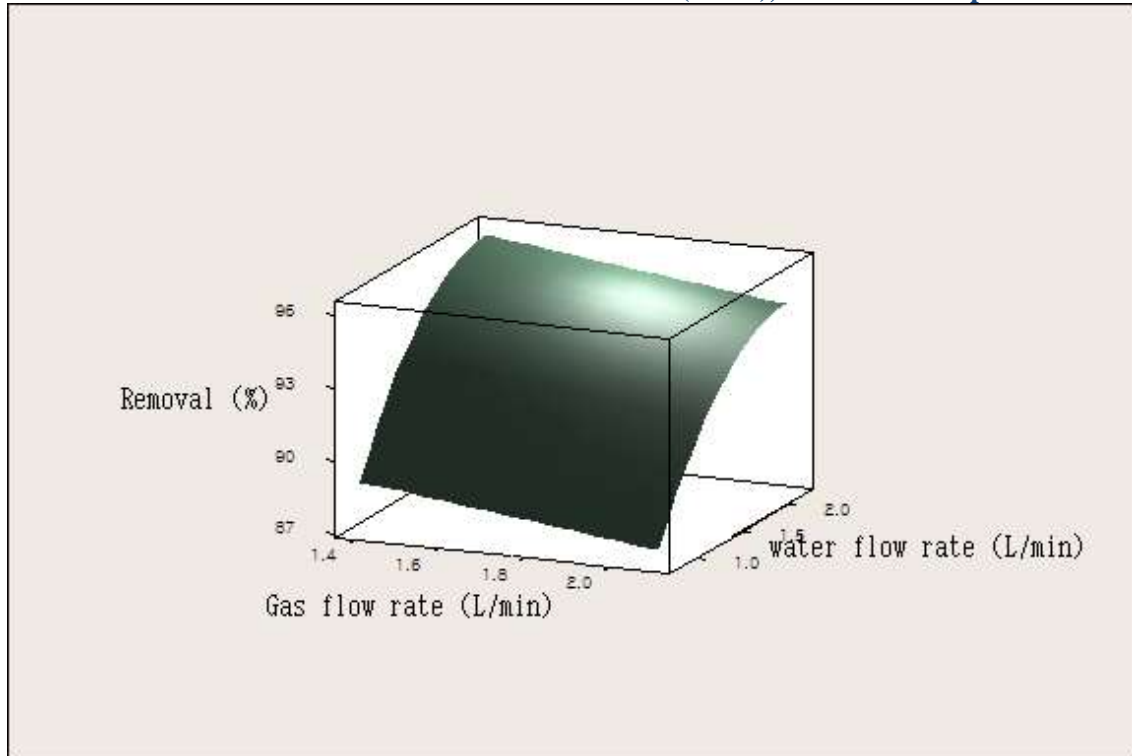


Fig. 5. Response surface optimization of H₂S degradation vs. gas flow rate and water flow rate

Fig. 5 shows the optimized reaction diagram according to the optimum gas flow rate and water flow rate for removing H₂S using cavitation. As shown by the figure, the removal rate of H₂S increases if the gas flow rate decreases and the water flow rate increases. At the gas flow rate of 1.3964LPM and water flow rate of 2.1641LPM, the maximum removal rate of H₂S through reaction optimization is estimated to be 96.05%.

CONCLUSIONS

This study used cavitation effect to remove H₂S which is one of the main causes behind complaints of offensive odors. To deduce the optimum conditions between the H₂S gas flow rate and water flow rate injected to generate cavitation, the central composite method of reaction surface analysis was used, and the mathematical model for optimization was presented.

The central composite method of reaction surface analysis for optimization showed that the gas flow rate, water flow rate and curvature of water flow rate affected the removal rate of H₂S, and the factor that had the largest effect on the removal rate, which was the reactivity value, was the flow rate of the water injected to generate cavitation.

The result of the optimization gas flow rate and water flow rate to remove H₂S using the central composite method showed that the removal rate of H₂S increases as the gas flow rate decreases and the water flow rate increases. At the gas flow rate of 1.3964LPM and water flow rate of 2.1641LPM, the maximum removal rate of H₂S through reaction optimization was estimated to be 96.05%, and the following mathematical model was obtained.

$$\text{Removal rate (\%)} = 81.5066 - 2.09706 x_1 + 16.915 x_2 - 3.70435 x_1^2 \quad (3)$$

To confirm the validation of this estimated value, when the replication test was performed under the above same conditions, the removal rate by the experiment was shown to be 96.3% which is similar to the estimated value by mathematical model. Therefore we can conclude that the mathematical model used in this research is valid.

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