Parameter Optimization of Ultrasound Technology for Algae Removal and its Application in Pengxi River of Three Gorges Reservoir

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The effects on algae removal imposed by ultrasound technology employing different combinations of parameters, including frequency, power and irradiation time, were estimated using response surface methodology. This technology was applied in the backwater area of Pengxi river in the three Gorges Reservoirs. Results showed that, the algae removal rate by ultrasound was 64.1 %, at an optimal frequency of 45 kHz, input power of 60 W and irradiation time of 48 s. After 2 h of ultrasound irradiation on the 500 m^2 study field, the algae density dropped from $(1.10 \pm 0.02) \times 10^7$ to $(2.1 \pm 0.1) \times 10^6$ cells/mL. Chlorophyll-a concentration dropped to 13.8 % of its initial amount algae were effectively controlled. Ultrasound technology is concluded to be effective for algae removal and can thus be applied to algal blooms.

Keywords: Algae removal, Response surface design, Ultrasound, Water bloom.

INTRODUCTION

Eutrophication is currently one of the major water pollution problems in the world[1]. In eutrophic water, the "water bloom" formed by rapid algal growth, results in the deterioration of water quality, reduction of aquatic species, as well as other ecological system disorders. Even worse, a number of algae species (*Microcystis* sp., *Anabaena* sp., *etc.*) produce algal toxins. These toxins affect a variety of aquatic and semi-aquatic animals[2] and can threaten human health through the food chain[3]. Other algae species also directly or indirectly threaten human health by causing rashes, gastrointestinal diseases, pneumonia[4], allergic reactions, headache, fever[5] and liver cancer[6]. The outbreak of blue-green algae in Taihu Lake in May 2007 imposed serious effects on the lives of the people residing near the area[7]. Therefore, the control of eutrophication has emerged as a pressing and challenging issue.

Water blooms are currently controlled *via* chemical[8,9], biological[10] and physical methods[11,12], among others. Chemical methods require the addition of pharmaceutical products onto the water, which easily results in secondary pollution. Moreover, the cost for such methods is relatively high. Biological methods entail low cost, but their effects are not immediately evident. Hence, biological methods are unsuitable for emergency treatment of algal blooms.

Ultrasound technology has recently been used in environmental applications. The principle behind this technology lies in the use of various effects of ultrasound to inhibit the growth of algae[13,14]. The use of this technology has a number of advantages. First, ultrasound is easy to control and does not involve the use of chemicals. Second, the technology has mild reaction conditions and quick reaction speed. Thus, the ultrasound technology is considered environment friendly, which gives it a broad development prospect.

The use of ultrasound technology for algae removal currently focuses on control parameters, mechanisms and practical applications[13-15]. However, the control parameters and the mechanism of this technology remain indefinite. Moreover, this technology has limited practical applications and its manner of operation is not clear. Various studies on the optimal frequency for ultrasound technology have reported largely different values, including 580[16], 150[17], 1700[18-20], 200[21] and 80[22] kHz. Similarly, different studies have reported largely different values for optimal power, including 30[17], 60[23], 80[22], 120[24] and 1200[21] W, as well as 0.6[20], 47.2[25] and 0.0018[16] W/cm^3. Optimal values for irradiation time include 5[23] s, 5[22] and 10[21] min. These differences can primarily be attributed to the interaction among the control parameters. The optimal values for the control parameters can be obtained through actual optimization.

In this paper, we designed and optimized the control parameters for ultrasound technology by conducting a response surface experiment. The designed and optimized parameters include ultrasonic frequency, ultrasonic power and ultrasonic

irradiation time. Through this experiment, we achieved an optimal combination of the ultrasound parameters, which we used in a field study on the backwater area of Pengxi river in the three Gorges Reservoir. This study aims at providing an effective and scientific basis for the application of ultrasound technology and for the effective prevention of eutrophication.

EXPERIMENTAL

The algae species used in this laboratory research were taken below the Double River Bridge spanning Pengxi river. After identification, we ascertained that the predominant algae species in the collection site were *Microcystis sp.* and *Anabaena sp.*, which belong to *Cyanobacteria* and *Scenedesmus*, respectively. We placed the algae in a constant-light incubator to inactivate the culture. The temperature was set to 25 °C, light intensity to 2000 lx and the light-dark cycle to 14 h:10 h. The algae were initially cultivated in the prepared monoculture BG11, which was provided by the Freshwater Algae Culture Collection of the Institute of Hydrobiology. The composition of BG11 was 1.5 g/L $NaNO_3$, 0.04 g/L $K_2HPO_4 \cdot 3H_2O$, 0.075 g/L $MgSO_4 \cdot 7H_2O$, 0.036 g/L $CaCl_2 \cdot 2H_2O$, 0.006 g/L citric acid, 0.006 g/L ferric ammonium citrate, 0.001 g/L EDTA, 0.02 g/L Na_2CO_3 and 1 mL/L A_5 + Co solution. The composition of the A_5 + Co solution was 2.86 g/L H_3BO_3, 1.86 g/L $MnCl_2 \cdot 4H_2O$, 0.22 g/L $ZnSO_4 \cdot 7H_2O$, 0.39 g/L $Na_2MoO_4 \cdot 2H_2O$, 0.08 g/L $CuSO_4 \cdot 5H_2O$ and 0.05 g/L $Co(NO_3)_2 \cdot 6H_2O$. The pH value of the BG11 medium was 7.1, which was regulated using NaOH or HCl. The algae were cultivated until their density reached 10^7 cells/mL. At this concentration, the algae liquid can be used for the ultrasonic experiment.

Ultrasonic algae removal device: The device used in this study was developed by the Institute of Acoustics of the Chinese Academy of Sciences. The other parts of reactor were homemade. The experimental setup is shown in Fig. 1. The ultrasound source was an ATANA AT3020 ultrasonic signal generator. The power amplifier with model number HFVA-62 was from Nanjing Foneng Co., Ltd. The oscilloscope with model number was UT2025C was produced by Hong Kong Unit Co.
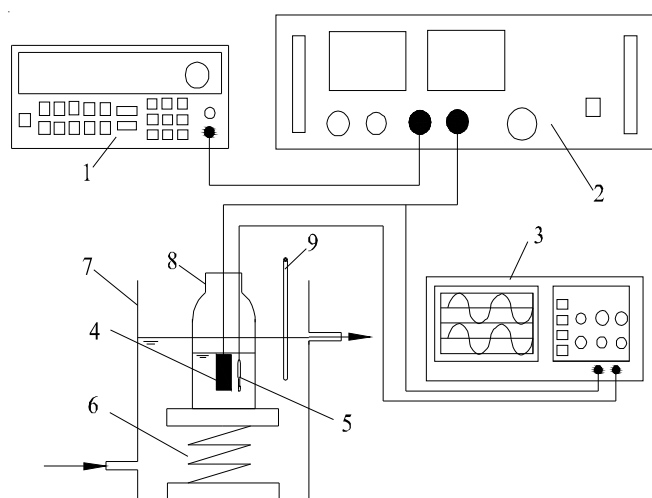

Fig. 1. Experimental ultrasonic apparatus (1. signal generator; 2. power amplifier; 3. oscilloscope; 4. transducer; 5. hydrophone; 6. telescopic stand; 7. cooling device; 8. glass flask 9. thermometer)

Detection of experimental parameters: In this study, the detected parameters were *in vivo* chlorophyll-a (*in vivo* Chl-a) content of the algae and algal density, which were measured during both laboratory testing and field testing using the AquaFluor Handheld Fluorometer produced by Turner Designs (USA). This instrument requires calibration before use. Water samples were taken and the steps listed in the "Analysis Editorial Board. Water and wastewater monitoring analysis method (4th Edition) (add Edition)" were followed for testing. The resulting value from the test and the water sample were used in the calibration. The instrument was only used for experimental testing when the correlation between the results of the standard and the experimental test groups was 99.9 %. In the detection of *in vivo* chlorophyll-a, blue light with a wavelength of 430 nm was chosen; the detection range was 0.3 to 3 μg/L. Yellow light with a wavelength of 590 nm was chosen in the determination of algae density. The detection range was 150 cells/mL to 150000 cells/mL. When the actual density exceeded the maximum range for the instrument, the sample was diluted. The immediate chlorophyll-a removal rate used in this experiment was calculated using eqn. (1).

$$\text{Removal of chlorophyll - a} = \frac{\text{Chl - a}_0 - \text{Chl - a}_1}{\text{Chl - a}_0} \times 100\% \quad (1)$$

where Chl-a_0 is the *in vivo* Chl-a concentration (μg/L) in the algae before ultrasonic treatment and Chl-a_1 is the *in vivo* Chl-a concentration (μg/L) in the algae after ultrasonic treatment.

Response-surface experimental design for ultrasonic parameters optimization: Ultrasonic power, ultrasonic frequency and ultrasonic irradiation time are the three main parameters of ultrasonic algae removal. Based on comprehensive pre-testing and considering energy consumption and costs, we chose the following parameter values for this experiment: frequency range, 20 to 120 kHz; power range, 20 to 60 W; and irradiation time, 10 s to 50 s. Based on the principles of the response surface central composite design, we designed a test program that includes three factors and three levels, totaling 20 test points. The assignment of factors and levels are shown in Table-1. The response surface experimental design is shown in Table-2. Data were processed by the design-expert software. For each experimental group, an 800 mL sample was used. The ultrasonic transducer was submerged 30 mm below the water surface. Ultrasound with different frequencies and power was applied to the samples at various lengths of irradiation time. The supernatant of the samples was subsequently used to determine the *in vivo* Chl-a concentration. The algal density and *vivo* Chl-a concentration were detected after ultrasonic after 2 h stationary sampling.

Field experiment

Experimental site: In this study, the testing site was a 500 m^2 backwater area near the double river bridge spanning Pengxi river in the three Gorges Reservoir. In this area, the water flow is slow. Hence, the effect of water flow can be disregarded. The temperature during the experiment ranged from 23.5 to 31.2 °C, with an average of 27 °C. Light conditions were good and suitable for algal growth.

TABLE-1
FACTORS AND LEVELS OF THE SMALL-SCALE LABORATORY EXPERIMENTAL DESIGN

Factor	Variable	Units	Level		
			-1(low)	0	+1(high)
Ultrasonic Frequency	X_1	kHz	20.00	70.00	120.00
Ultrasonic Power	X_2	W	20.00	40.00	60.00
Irradiation Time	X_3	min	10	30	50

Pengxi river is an important tributary of the Yangtze river and is located in the hinterland of the three Gorges Reservoir area. The river is in within the territory of Yunyang County and Kaixian and has a basin area of 5172.5 km^2, main river length of 182.4 km and annual average runoff of 3.41 billion m^3.

The catchment area of Pengxi river in Yunyang County was initially 609.2 km^2 with a length of 50.4 m. After the three Gorges Reservoir was formed, the flood area of Pengxi river decreased to 92 km^2, the migrant population in the area reached 221.5 thousand and a fluctuation zone with an area of approximately 65 km^2 formed. Among the tributaries of the three Gorges Reservoir, Pengxi river has the largest flood area, the largest number of immigrants and the most widely fluctuating area. From 2005 to 2007, the water quality of the Pengxi river section of the reservoir was Class IV. Currently, the number of months during which the water quality of this section is Class IV is increasing. Water bloom occurs almost every year with a worsening trend. Therefore, emergency measures for bloom control are desperately needed.

Mobile boat ultrasonic algae removal device: In this test, 20 ultrasonic transducers were used. The transducers were installed on both sides of a boat spaced at 1.4 m. The optimal parameters of the mobile boat, such as power and frequency, were obtained by optimizing the experimental design. The launch direction of the ultrasonic transducers was perpendicular to the boat. The launcher was installed 25 cm below the water surface to ensure that the ultrasound emission was horizontal. The elevation and the plan of the mobile boat with ultrasonic transducer. The full length of the boat was 11.5 m, full width was 3.2 m, draft was 0.3 m to 0.5 m and speed was 16 km/h. The boat was made of glass fiber-reinforced plastic. Treatment and control groups were established. Treatment group samples were taken upstream, whereas control group samples were taken approximately 500 m from the treatment group area. During testing, the boat with ultrasonic transducer sailed a round trip in the water for 2 h each day. Boat speed was maintained at 50 m/h when ultrasonic transducers were on.

In the area of the treatment groups, the boat maintained the same speed and travelled for 2 h while the ultrasonic transducers were off. Samples were collected immediately in the middle of the water and were tested in the field for algae density and *in vivo* Chl-a concentration for six times. For the control group, the same procedure was performed six times in the area 500 m upstream of the test area.

RESULTS AND DISCUSSION

The result of the response surface experiment for the optimized parameters of ultrasonic algae removal is presented in Table-2.

The experimental data were used for polynomial regression analysis. The response design was suitable for the second-order model, which is an experience model that can describe the relationship between response variables (dependent variable) and independent variables (operating conditions), as shown in eqn. (2):

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_{ii}^2 + \sum_{1 \le i \le j \le k} \beta_{ij} X_i X_j \qquad (2)$$

where y is the predicted response value; β_0, β_i and β_{ii} are the offset term and second-order linear offset and offset coefficients, respectively; β_{ij} is the interaction coefficient; X_i and is the coded value of the independent variable. The conversion equation between X_i and the actual value of the variable tested is shown in eqn. (3):

$$X_i = \frac{2[x_i - x_i(\text{at level } 0)]}{x_i(\text{at high level}) - x_i(\text{at log level})} \qquad (3)$$

According to the mathematical model and the results of the variance analysis shown in Table-3, individually, all of the factors, interaction terms and their squares significantly affect on the removal rate of chlorophyll-a. The regression equation is:

$$\begin{aligned} Y = 18.853 &+ 0.822X_1 + 1.7185X_2 + 0.537X_3 \\ &- 7.319 \times 10^{-3}X_1X_2 - 1.984X_1X_3 + 0.011X_2X_3 \\ &- 3.242 \times 10^{-3}X_1^2 - 0.016X_2^2 - 0.013 X_3^2 \end{aligned} \qquad (4)$$

where Y is the response value of the removal rate of Chl-a; and X_1, X_2 and X_3 stand for ultrasonic frequency, power and irradiation time, respectively.

A large F-value and a small P-value represent significance of the correlation coefficient of variables[26]. From the analysis of variance shown in Table-3, the F-value of the model was 152.01, Prob > F < 0.0001 (Prob' > F' < 0.05, models are significant) and lack of fit was insignificant (Prob > F value was 0.3798, which is greater than 0.05). All the values above indicate that the model was significant. That is, the model fits well in the entire regression area. The multiple correlation coefficient obtained was $R^2 = 0.9927$, indicating a good correlation. $RA_{dj}^2 - R_{Pred}^2 = 0.9862 - 0.9520 = 0.0342 < 0.2$, CV = 2.58 % < 10 %, showing that the credibility and accuracy of the experiment are both high (Table-4). A_{deq} precision is a ratio of effective signal to noise and its value was 37.414, which can be considered reasonable given the values above 4.

The regression equation provides a suitable model for the removal of algae using ultrasonic technology. The test for significance using the regression coefficients of the quadratic regression model showed that the factors X_1, X_2 and X_3, are linearly correlated to *in vivo* Chl-a removal rate. X_1, X_2, X_1, X_3, X_2 and X_3 have clear effects on the removal rate of

TABLE-2
CENTRAL COMPOSITE DESIGN TEST SCHEME AND RESULTS

No.	Factors			Response	No.	Factors			Response
	X_1	X_2	X_3	Y		X_1	X_2	X_3	Y
1	20	20	10	26.17	11	70	20	30	49.79
2	120	20	10	48.1	12	70	60	30	59.5
3	20	60	10	43.33	13	70	40	10	54.15
4	120	60	10	33.68	14	70	40	50	58.6
5	20	20	50	28.45	15	70	40	30	59.85
6	120	20	50	40.5	16	70	40	30	59.05
7	20	60	50	61.45	17	70	40	30	61.1
8	120	60	50	46.53	18	70	40	30	61.92
9	20	40	30	52.84	19	70	40	30	59.16
10	120	40	30	53.13	20	70	40	30	59.07

TABLE-3
ANALYSIS OF VARIANCE OF THE REGRESSION EQUATION

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	Note
Model	2359.19	9	262.13	152.01	< 0.0001	significant
X_1	9.41	1	9.41	5.46	0.0416	significant
X_2	265.02	1	265.02	153.68	< 0.0001	significant
X_3	90.60	1	90.60	52.54	< 0.0001	significant
X_1X_2	428.51	1	428.51	248.49	< 0.0001	significant
X_1X_3	28.69	1	28.69	16.64	0.0022	significant
X_2X_3	164.62	1	164.62	95.46	< 0.0001	significant
X_1^2	180.65	1	180.65	104.76	< 0.0001	significant
X_2^2	114.23	1	114.23	66.24	< 0.0001	significant
X_3^2	61.14	1	61.14	35.45	0.0001	significant
Residual	17.24	10	1.72			
Lack of Fit	9.86	5	1.97	1.33	0.3798	not significant
Pure Error	7.39	5	1.48			
Cor Total	2376.44	19				

TABLE 4
MODEL RELIABILITY ANALYSIS

Index	Std. Dev.	Mean	C.V (%)	PRESS*	R^2	Adj R^2	Pred R^2	Adeq precision
Results	1.31	50.82	2.58	114.07	0.9927	0.9862	0.9520	37.414

*Press: Predicted residual error sum of squares

chlorophyll-a. X_{12}, X_{22} and X_{32} have a clear curved surface effect on the removal rate of *in vivo* Chl-a.

Based on the analysis of data using eqn. (4), with dimensionality reduction, the effect of the two factors on the removal rate of chlorophyll-a when the conditions of the other factors are unchanged can be observed. The response surface plots and contour maps from the design-expert analysis are shown in Fig. 2.

Fig. 2a and 2b show the effect of ultrasonic power and ultrasonic frequency on removal rate of chlorophyll-a when ultrasonic irradiation time was unchanged (center value). When the shape of the contour line is oval, the interaction between the factors is significant. When the shape is circular, the interaction between factors is insignificant. Therefore, the interaction between the ultrasonic power and ultrasonic frequency was significant, as reflected by the contour map. From the response surface graph, a high removal rate of chlorophyll-a can be achieved with low frequency ultrasound (20 to 80 kHz) and high ultrasonic power (40 to 60 W).

Figs. 2c and 2d show the effect of ultrasonic power and ultrasonic irradiation time on the removal rate of chlorophyll-a when the ultrasonic frequency was unchanged (center value). Research shows that increasing the ultrasonic power and ultrasonic irradiation time can help improve the removal rate

of chlorophyll-a. To obtain the desired removal rate, the ultrasonic power and ultrasonic irradiation time should be in the within 40 to 60 W and 30 to 50 s, respectively.

Figs. 2e and 2f show the effect of ultrasonic frequency and ultrasonic irradiation time on the removal rate of chlorophyll-a when ultrasonic power was unchanged (center value). The contour map shows that the interaction between these two parameters is insignificant. When the ultrasonic frequency increased from 20 to 70 kHz, the removal rate of chlorophyll-a increased. However, when the ultrasonic frequency continued to increase, the removal rate decreased. This phenomenon indicated that, the optimal value of ultrasonic frequency is approximately 70 kHz. Given that the interaction between ultrasonic frequency and irradiation time was insignificant, the irradiation time should be kept at 30 to 50 s to ensure high algae removal rate.

A stable point exists in the regression equation and this point represents the maximum value. By solving the inverse model, the maximum values for the major factors were obtained: $X_1 = 45$, $X_2 = 60$ and $X_3 = 48$. Hence, the optimal conditions were 45 kHz for ultrasound frequency, 60 W for ultrasonic power and 48 s for ultrasonic irradiation time. The removal rate of chlorophyll-a can reach 64.1 %.
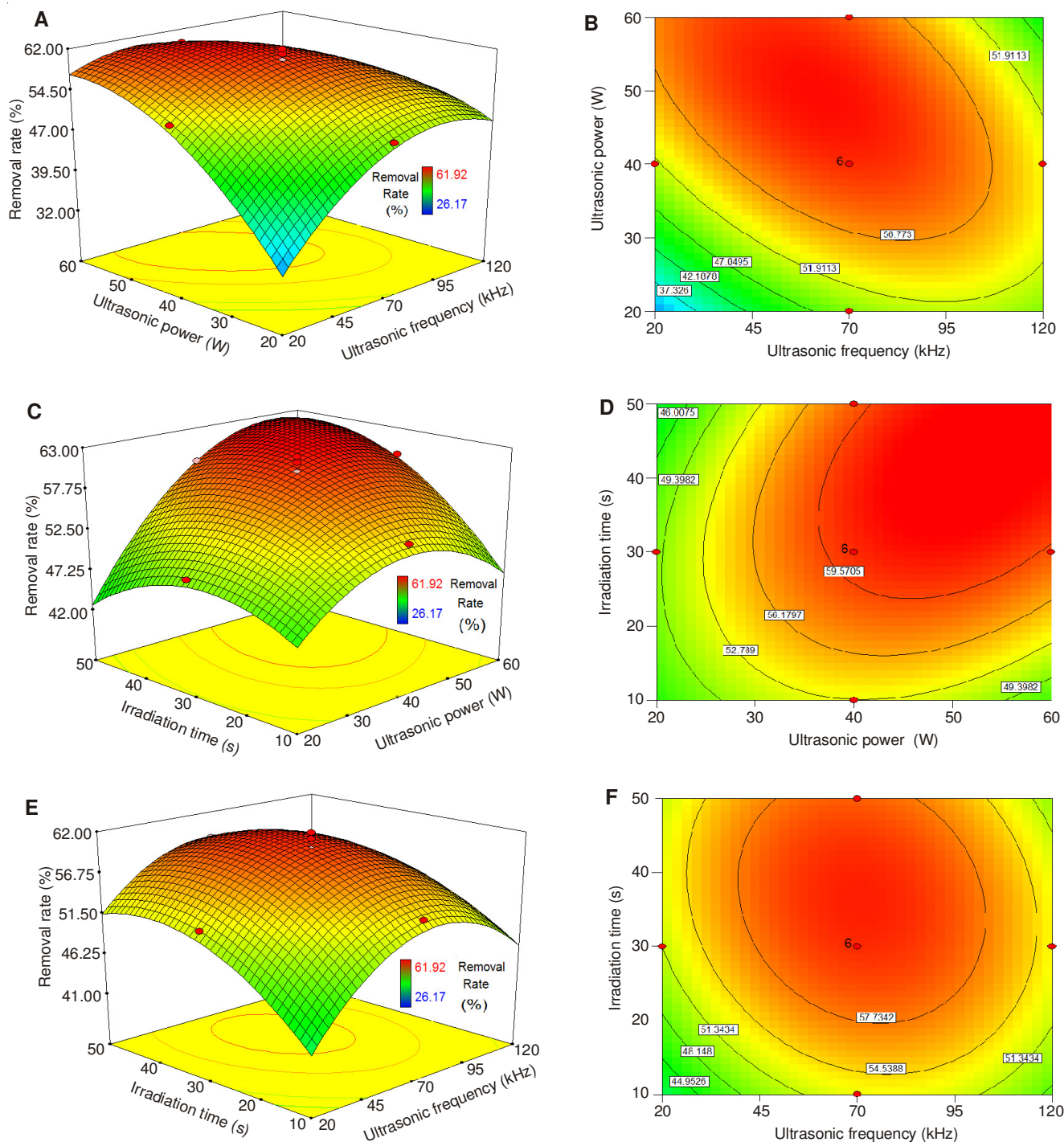
Fig. 3.   Response surface plot of removal rate as a function of ratio of (a) frequency and ultrasonic power 3D; (b) frequency and ultrasonic power 2D; (c) irradiation time and ultrasonic power 3D; (d) irradiation time and ultrasonic power 2D; (e) irradiation time and ultrasonic frequency 3D; and (f) irradiation time and ultrasonic frequency 2D

To verify the predicted results, the calculated optimum conditions of (45 kHz, 60 W and 48 s) were used in the experiment. The initial concentration of algae in the verification test was also used in the experiment using the optimal conditions to eliminate the effects on the model. In this experiment, three test groups and a control group were prepared and an average removal rate of 64.4 % was obtained. The removal rate obtained from the regression equation is similar to this result and the error is only 0.47 %. This finding indicated that the test value and the predicted value were consistent and proved that the

model is accurate and reliable for analyzing and predicting the parameters of ultrasonic algae removal.

Results and analysis of the field test: Based on the conclusion of the optimal experiment, the optimal parameters of the ultrasonic transducer were a power of 60 W and frequency of 45 kHz. As the distance from the transducer increased, ultrasonic intensity decreased; the effective functional range of the transducer was 50 cm. To ensure the algae were irradiated for 48 s, we sailed the boat at a speed of 50 m/h.

Changes in algal density: Before testing, a large number of algae were seen floating on the water. Through microscopic examination, the major algae species present in the site were *Microcystis* sp. and *Anabaena sp*. On the first day, after treating the water with ultrasound for 2 h, the floating algae sank and the density of algae in the water decreased from $(1.10 \pm 0.02) \times 10^7$ to $(2.1 \pm 0.1) \times 10^6$ cells/mL. The immediate removal rate obtained reached 80 %. After 10 d of treatment, the algal density in the treatment group was only $(4.40 \pm 0.17) \times 10^5$ cells /mL, whereas in the control group was $(2.01 \pm 0.03) \times 10^7$ cells/mL. The algal density of treatment group was only 2.3 % of that of the control group. The changes in the algal density in the water of the test and control groups are shown in Fig. 3a.

Changes in concentration of *in vivo* Chl-a: Chlorophyll-a is an important evaluation factor for eutrophication. After 2 h of the ultrasonic treatment in the first day, the concentration of *in vivo* Chl-a decreased from (3.01 ± 0.03) to (0.41 ± 0.01) mg/L, which was 13.8 % of the pre-test value. After 10 d of treatment, the concentration of *in vivo* Chl-a in the treatment group was only (0.089 ± 0.004) mg/L, whereas that in the control group was (4.3 ± 0.02) mg/L. The concentration of *in vivo* Chl-a in the treatment group was only 2.3 % of that of the control group. The changes in the concentration of *in vivo* Chl-a in the water of the test and control groups are shown in Fig. 3b. Thus, the frequency and the input power we used were rather low, no negative effect on zooplankton was observed.
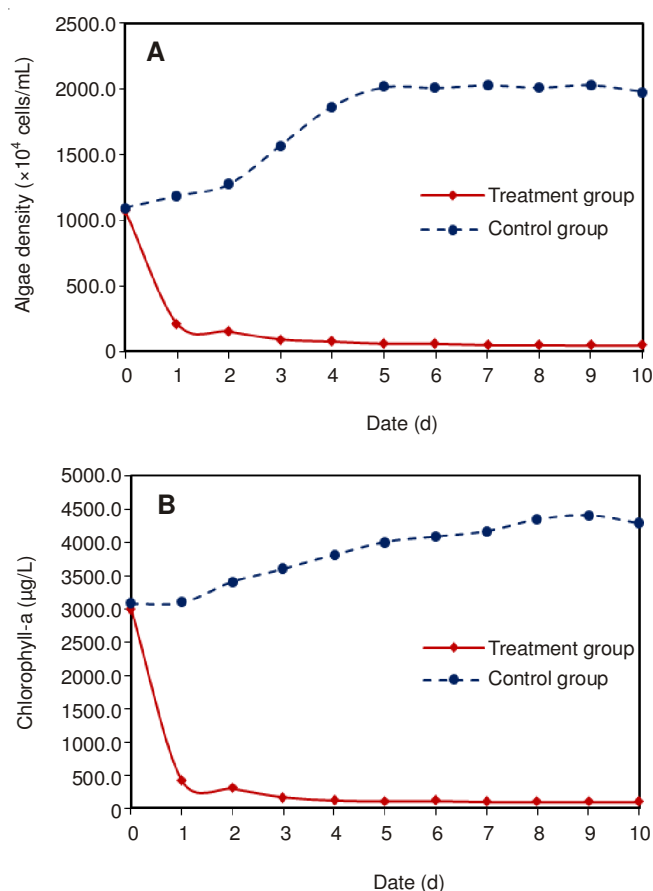

Fig. 4 . Changes in quality of control and treated water during the experiment: (a) algae density (b) Chlorophyll-a

Conclusion

The optimized combination of ultrasound parameters obtained were 45 kHz for ultrasonic frequency, 60 W for ultrasonic power and 48 s for ultrasonic irradiation time. The field test confirmed that after the ultrasonic treatment, a large amount of algae in the water sank. After 2 h of treatment in the first day, algal density decreased from $(1.10 \pm 0.02) \times 10^7$ to $(2.1 \pm 0.1) \times 10^6$ cells/mL and the concentration of *in vivo* Chl-a decreased to 13.8 % of the initial concentration. All results indicate that ultrasound technology can be applied for the emergency treatment of algal blooms.

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