Optimization of ultrasound-induced inactivation of model bacterial mixture using response surface methodology

Zhiwei Zhou, Yanling Yang and Xing Li

ABSTRACT

Ultrasound (US)-based disinfection involves the disruption of cell membranes, oxidation of active free radicals, as well as hotspot heating, either individually or in combination. Various factors can affect US efficiency for inactivating microbes. However, only a few studies have discussed the use of US for microbial inactivation via response surface methodology. Here, we evaluated the potential of US for the disinfection of water supply or the elimination of drinking water residues. Moreover, the effects of US inactivation parameters, such as energy density, sonication time, and US device duty cycle on reduction in total bacterial (TB) count and total coliform (TC) count were investigated and optimized. The results indicated that the optimal inactivation condition was achieved at an energy density of 8.30 W/mL, a sonication time of 950 s, and a duty cycle of 0.7:0.3. Under optimal conditions, the experimental values of TB and TC inactivation efficiency were 47.26% ± 4.35% and 39.23% ± 2.27%, respectively, while the predicted values were 46.57% and 38.65%, respectively. The models developed here helped to predict the effectiveness of inactivation efficiency to a 'sufficiently applicable' extent. Under the optimized conditions, US has high potential as an effective disinfection method, as shown by energy efficiency analysis.

Key words | bacterial inactivation, optimization, power ultrasound, response surface methodology

Zhiwei Zhou
Yanling Yang (corresponding author)
Xing Li
The College of Architecture and Civil Engineering,
Beijing University of Technology,
Beijing 100124,
China
E-mail: *yangyanling@bjut.edu.cn*

INTRODUCTION

Disinfection is an important stage in water and wastewater treatment. Different disinfection technologies have been tested, with each possessing unique advantages and disadvantages (Drakopoulou *et al.* 2009). For example, the use of chemical oxidation (e.g., chlor(am)ination) leads to the formation of by-products with increased toxicity for aquatic organisms and ecosystems, especially after prolonged exposure (Liu *et al.* 2006). In addition, traditional methods of disinfection come with many more disadvantages, with some microorganisms developing resistance or undergoing resuscitation after the application of biocides, ultraviolet light, chlorine, or antibiotic and heat treatments (Malley *et al.* 1995; Richardson 2003). To overcome these limitations, research has focused on developing alternative disinfection methods. Recently, the use of ultrasound (US) as a standalone method (Gao *et al.* 2014a), and combined with ultraviolet irradiation and hydrogen peroxide (Joyce *et al.* 2006), as well as US combined with ozone (Jyoti & Pandit 2004), has been studied in detail.

US usually has a frequency of ≥20 kHz; low-frequency US (20–100 kHz) is also termed high-power US. Sonication can cause a series of compression and rarefaction cycles, leading to the generation of cavitation bubbles. Millions of these bubbles implode, yielding localized temperatures as high as 5,000 °C, pressures as high as 100 MPa, and free radicals such as •OH, HO_2•, and O• (Pilli *et al.* 2011). The disinfection capacity of sonication in water stems from acoustic cavitation, which is the formation and collapse of micro-bubbles occurring in milliseconds, thus producing extreme temperature and pressure gradients. To date, the bactericidal mechanisms of US have not been fully proven. However, it is widely believed that the elevated temperature,

doi: 10.2166/aqua.2015.045

pressure, and subsequent free radical actions are responsible for microbial inactivation (Joyce *et al.* 2003; Herceg *et al.* 2012; Gao *et al.* 2014b).

Indeed, numerous experimental variables, such as US-related parameters (frequency, energy density, sonication time, duty cycle, and reactor configuration), medium characteristics (pH, temperature, concentration of the solids, content and property of organic matter, manosonication, thermosonication, and manothermosonication), as well as bacterial properties can strongly influence efficacy of microbial inactivation. For instance, Joyce *et al.* (2003) found significantly higher inactivation in the *Bacillus* species when exposed to US at low frequencies of 20 and 38 kHz, as compared to that at higher frequencies (512 and 850 kHz). Microorganisms such as Gram-positive and Gram-negative bacteria also have dissimilar membrane structures, and thus respond differently when exposed to ultrasonic waves. Drakopoulou *et al.* (2009) found that the presence of 5 g/L titanium dioxide particles generally enhanced the destruction of Gram-negative bacteria (total coliforms and fecal coliforms); however, the relatively weak sonochemical inactivation of Gram-positive bacteria (*Clostridium perfringens* and fecal streptococci) was only slightly affected. Gao *et al.* (2014b) chose to study *Enterobacter aerogenes*, *Bacillus subtilis*, *Staphylococcus epidermidis*, *S. epidermidis* SK, and *S. pseudintermedius* due to differences in their physical and biological properties, and found that microbes with a thicker and 'soft' capsule were highly resistant to ultrasonic deactivation.

Response surface methodology (RSM) with Box–Behnken design (BBD) was used to statistically evaluate multiple parameters in order to optimize US conditions for extraction of phenolic compounds (Wang *et al.* 2013) and bacterial inactivation in clinical solid wastes (Hossain *et al.* 2015). This approach can reduce the overall number of experimental trials. By establishing a mathematical model, RSM evaluates multiple parameters and their interactions using quantitative data, thereby effectively optimizing complex extraction procedures in a statistical manner. We selected this method in the present study to optimize the inactivation procedure, with the goal of achieving higher inactivation efficiency while maintaining low-energy consumption. US energy density, sonication time, and duty cycle (the ratio of working time to pause time in an irradiation cycle) were independent variables examined in the study. The inactivation rates of total bacteria (TB) and total coliforms (TC) were the measured response values. Although RSM has been used to optimize inactivation conditions in many studies, we focused on the US parameters required for the inactivation of bacterial mixture in aqueous solution. The findings of this study could support implications for the disinfection of water supply or the elimination of drinking water residues.

MATERIALS AND METHODS

Preparation of simulated water with microbial load

Pre-determined amounts of bacterial mixture were added to tap water, which was left overnight to decay residual chlorine. Prior to sonication, this water sample was thoroughly mixed to achieve a homogenous mixture. For all experiments, the characteristics of tap water were as follows: turbidity, 0.401 NTU; UV_{254}, 0.0120 cm^{-1}; pH, 8.23; and temperature, 20 °C. The microbial density levels in the simulated water were as follows: 1.2×10^5colony-forming units (CFU)/mL for TB and 1.61×10^6 CFU/100 mL for TC.

Ultrasonic trial

A probe-type sonicator (XH-2008DE; Xianghu Ultrasonic Instrument Co., Beijing, China) equipped with a digital timer and temperature controller, operating at fixed frequencies of 25 and 40 kHz and a nominal power output of up to 1,500 W, was used in the study (Figure 1). Samples (50 mL)

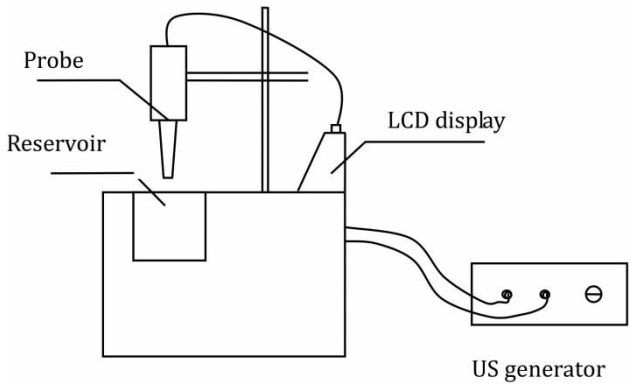

Figure 1 | Schematic diagram of the ultrasonic treatment system.

were placed in a double-walled, jacketed glass container, and subjected to continuous US irradiation via a 7-mm-diameter tip at maximum nominal power. During the experiment, temperature was controlled with a water bath coupled to a circulator, and was kept at 20 ± 1 °C. To minimize sample contamination, the probe was immersed in the solution through a silicon rubber plug. The water level inside the container was 4 cm and the probe (total length, 10 cm) was positioned in the middle, with its tip 2 cm from the bottom of the container.

Experimental design

RSM was used to investigate the effect of three independent variables on TB and TC inactivation. The main factors, including energy density (W/mL, X_1), sonication time (s, X_2), and duty cycle (X_3), were selected as independent variables that needed optimization for microbial inactivation analysis. Experiments were performed on the BBD based on results from a single-factor test. The coded values of the experimental factors and their levels for the BBD are shown in Table 1. The code number 0 for X_1 and X_2 represents the optimum condition in single-factor test, while the code number 0 for X_3 was 06:04, because inactivation rates of 04:06 and 08:02 in the single-factor test corresponded to the maximum and minimum values. In order to achieve high TB and TC inactivation efficiency while maintaining low energy consumption, these parameters were selected. The complete design was in random order, and consisted of 17 combinations, including five replicates at the central point. Data from BBD were analyzed by multiple regression tests to fit the quadratic polynomial model shown in Equation (1), where Y is the predicted response; γ_0 is a constant; and α_i, α_{ii}, and α_{ij} are the linear, quadratic, and interactive coefficients of the model, respectively. Accordingly, X_i and X_j represent the levels of the independent variables, respectively:

$$Y = \gamma_0 + \sum_{i=1}^{3} \alpha_i X_i + \sum_{i=1}^{3} \alpha_{ii} {X_i}^2 + \sum_{i \neq j=1}^{3} \alpha_{ij} X_i X_j \quad (1)$$

Experimental results from the response surface design were analyzed using the Design-Expert 9.0 software (Trial Version; State-Ease, Inc., Minneapolis, MN, USA). Model terms were selected or rejected based on P values (probability) with a 95% confidence interval. To assess the fit of the model, results obtained were analyzed by analysis of variance (ANOVA) using the Fisher's statistical method. A high R^2 value, close to 1 is desirable, and a reasonable agreement with $Adj.\ R^2$ is necessary. In addition, adequate precision (AP) was used to compare the range of the predicted values at designated points to the average prediction error. A ratio of >4 suggests adequate model discrimination. The coefficient of variance (CV), as the ratio of the standard error of the estimate to the mean value of the observed response, defines the reproducibility of the model. A model normally can be considered reproducible if its CV is not >10% (Ghafari *et al.* 2009).

Table 1 | Levels of variables for the experimental design

Symbols	Independent variables	−1	0	+1
X_1	Energy density (W/mL)	4	8	12
X_2	Sonication time (s)	300	900	1,500
X_3	Duty cycle	08:02	06:04	04:06

Inactivation efficiency

The inactivation efficiencies (IE) of TB and TC under different sonication conditions were calculated according to Equation (2), where N_0 and N_t are the numbers of TB and TC before and after ultrasonic treatment at time t (min), respectively:

$$IE = (1 - \frac{N_t}{N_0}) \times 100\% \quad (2)$$

Energy efficiency analysis

Energy efficiencies (EE) under different reaction conditions were calculated using Equation (3) based on the calculation by Jyoti & Pandit (2004). Here, C_0 and C_t are the concentrations of TB and TC before and after sonication at a given time, respectively. V is the volume of sample (mL). P_{diss} is the ultrasonic power dissipated into the samples

(W), and t is the time of sonication (s). EE is expressed as CFU/kJ:

$$EE = 10^{-3} \times \frac{(c_0 - c_t) \bullet V}{P_{diss} \bullet t} = 10^{-3} \times \frac{(c_0 - c_t)}{\text{Density} \bullet t} \quad (3)$$

Analytical methods

All analyses were carried out using chemicals of analytical grade. pH was determined by Thermo (Shanghai, China) pH meter, which was calibrated daily using pH buffer solutions. Measurements of turbidity, UV_{254}, and plate counts for TB and TC were performed in accordance with *Standard Methods* (APHA 1998). Specifically, samples taken from the container following different US exposures were transferred to saline/0.8% Ringer solutions at various sample dilutions. Approximately 1 mL of the serially diluted sample was transferred to a nutrient agar plate. Samples were incubated for 24 h at 35 °C, followed by quantitative analysis of colony formation. TC density was determined by the membrane filter technique, and was measured following a 48 h incubation period at 37 °C; red colonies with a metallic (golden) sheen on the membrane were counted. All experiments were carried out in triplicates, and the results are represented as the mean values.

RESULTS AND DISCUSSION

We first investigated the effect of energy density, sonication time, and duty cycle on the TB and TC inactivation rates by single-factor experiments (Figure 2). With a 900 s sonication pulses and a duty cycle of 10:00 (continuous model), inactivation rates of TB and TC were initially observed to increase, and then decrease with increasing energy density (ranging from 2 to 13 W/mL). The highest inactivation rates for TB and TC were 50.00% and 50.29%, with energy densities of 10 and 8 W/mL, respectively. At fixed US frequency, sonication time, and duty cycle, higher energy density produces more cavitation bubbles, which, in turn, leads to higher pressure, increased hydroxyl radical formation, and higher probability of cytoplasm release from the cell wall (Gonze *et al.* 2003). Therefore, higher ultrasonic energy density enhanced the inactivation efficiency of TB and TC. However, energy densities that are too high (>10 W/mL) can reduce energy efficiency (Figure 2(a)). Our results showed that the acoustic cavitation effect tended to be limited or even declined when energy density exceeded the cavitation threshold.

In addition, when US was performed at different inactivation times ranging from 120 to 1,500 s with an energy density of 8 W/mL and a duty cycle of 10:00, higher TB inactivation rates were observed with increased sonication time. TC inactivation rate sharply increased at the beginning of 300 s, then slowed in the rate of increase with longer sonication time until 1,500 s. TB inactivation rate was, on average, consistently higher than that of TC; thus, the TC was comparably less susceptible to US inactivation. It has been suggested that Gram-positive bacteria are more resistant to sonication than Gram-negative bacteria. Our results differed from previous findings (Herceg *et al.* 2012) where the inactivation rate of Gram-negative bacteria was higher compared to that of Gram-positive bacteria. This may be attributed to the lower concentration of TC than TB used in this model bacterial mixture.

We also found that the highest TB and TC inactivation rates were 46.32% and 45.76%, respectively, with a duty cycle of 04:06 for 900 s and at 8 W/mL. The inactivation rate in the impulse working model was nearly the same as that of the continuous model; this could be due to structural stability of the cavitation bubbles. In the case of the impulse working model, stability of the cavitation bubbles with more activated regions and 'nuclei' was higher than that of the continuous model, resulting in reduced cavitation effects (Yao *et al.* 2011). Ashokkumar *et al.* (2003) also reported that the deactivation rate of *Cryptosporidium* oocysts was determined based on the effective sonication time, excluding the pause in a US cycle. Here, when the working time was nearly equal to the pause time (04:06), the lifetime of the bubble–water interface increased, and therefore, could provide more sites for inactivation by shear force, hydroxyl radicals, acoustic streaming, and pyrolysis. In addition, unstable bubbles generated in a former cycle had to collapse before the next cycle. Both phenomena may promote the cavitation process. More importantly, this operation model achieved higher inactivation efficacy, while reducing electricity consumption.

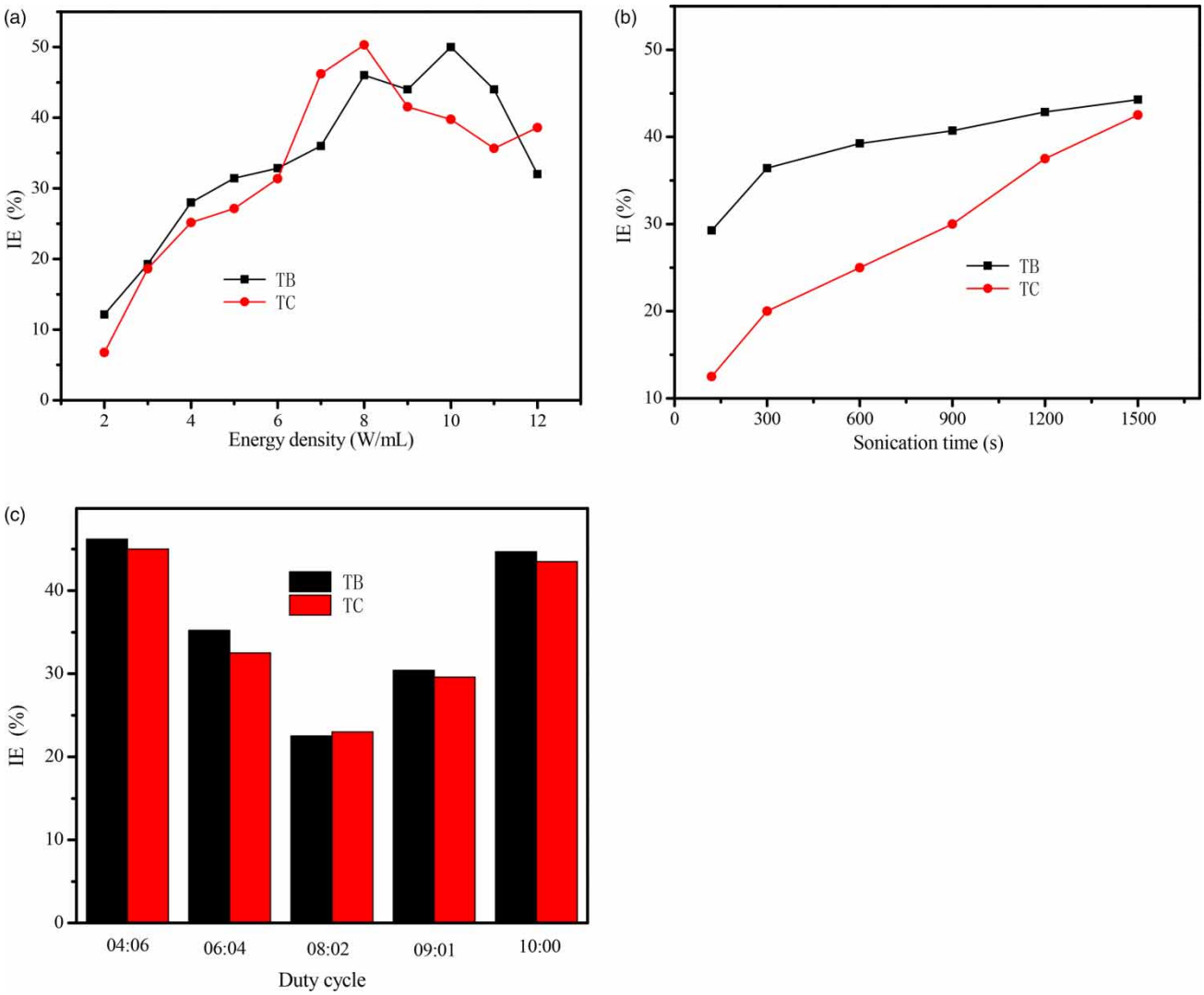

Figure 2 | Inactivation rate of TB and TC using single-factor experiments: (a) energy density, (b) sonication time, and (c) duty cycle.

Fitting the response surface models

Table 2 shows the inactivation rates of TB and TC under different conditions of US in all experiments. Multiple regression analyses using the quadratic polynomial model (Equation (1)) were performed based on the results listed in Table 2. Table 3 represents the results of ANOVA and regression coefficients, suggesting that the contribution of the quadratic model was significant ($p < 0.05$). The factors influencing inactivation efficacy for both TB and TC are as follows: duty cycle > sonication time > energy density. From the perspective of lack-of-fit test, however, the 'fitness' of the model for TB was not acceptable ($p < 0.05$), which indicates that other principal factors also affect inactivation efficacy. In contrast, the 'fitness' of the model for TC was very good ($p > 0.05$), indicating the suitability of models to accurately predict the variation (Prasad *et al.* 2011). The data shown in Table 2 indicated that TC inactivation rates and the US parameters were quadratic, with a good regression coefficient ($R^2 = 0.9580$), while TB inactivation rates did not fit the model ($R^2 = 0.8259$).

Effect of US parameters on TB and TC inactivation rates

Response surface and contour plots showing the influence of inactivation parameters on TC are presented in Figure 3(a)–3(c). Figure 3(a) shows the interaction of energy density and

Table 2 | BBD and experimental data

				IE $_{TB}$ (%)		IE $_{TC}$ (%)	
Run	X_1	X_2	X_3	Actual	Predicted	Actual	Predicted
1	1 (12)	0 (900)	1 (04:06)	30.43	25.51	23.90	22.47
2	− 1 (4)	0 (900)	1 (04:06)	32.00	26.65	28.60	26.47
3	0 (8)	1 (1,500)	1 (04:06)	23.91	29.09	22.15	24.68
4	0 (8)	1 (1,500)	1 (08:02)	44.29	39.20	29.50	28.47
5	0 (8)	0 (900)	0 (06:04)	44.35	45.46	39.44	38.82
6	− 1 (4)	1 (1,500)	0 (06:04)	27.15	27.32	22.18	21.78
7	− 1 (4)	0 (900)	1 (08:02)	28.00	32.92	23.15	24.58
8	0 (8)	− 1 (300)	1 (04:06)	22.17	27.26	21.31	22.34
9	1 (12)	0 (900)	1 (08:02)	32.00	37.35	28.60	26.47
10	− 1 (4)	− 1 (300)	0 (06:04)	27.56	27.82	18.75	19.84
11	0 (8)	0 (900)	0 (06:04)	45.65	45.46	36.63	38.82
12	1 (12)	1 (1,500)	0 (06:04)	32.61	32.35	24.95	23.85
13	0 (8)	0 (900)	0 (06:04)	46.87	45.46	39.07	38.82
14	1 (12)	− 1 (300)	0 (06:04)	26.25	26.08	19.51	19.91
15	0 (8)	− 1 (300)	1 (08:02)	40.43	35.25	27.45	24.92
16	0 (8)	0 (900)	0 (06:04)	44.25	45.46	38.63	42.02
17	0 (8)	0 (900)	0 (06:04)	46.20	45.46	40.34	38.82

Table 3 | Results of ANOVA and regression coefficients

	TB			TC		
Source	Coefficient	*F*-value	*P*-value	Coefficient	*F*-value	*P*-value
γ_0	+45.46	–	–	+38.82	–	–
X_1	+0.82	0.17	0.6883	+0.54	0.42	0.5387
X_2	+1.44	0.54	0.4866	+1.47	3.15	0.1190
X_3	−4.53	5.30	0.0548	−1.59	3.70	0.0958
X_1^2	−9.58	12.50	0.0095	−8.26	52.37	0.0002
X_2^2	−7.49	7.64	0.0280	−9.22	65.26	<0.0001
X_3^2	−5.27	3.79	0.0927	−4.50	15.57	0.0056
$X_1 X_2$	+1.69	0.37	0.37	+0.50	0.18	0.6806
$X_1 X_3$	−1.39	0.25	0.25	−2.54	4.70	0.0669
$X_2 X_3$	−0.53	0.036	0.036	−0.30	0.067	0.8035
Model	–	3.69	0.0496 (significant)	–	17.72	0.0005 (significant)
R^2	0.8259			0.9580		
R^2(adj)	0.6020			0.9039		
Lack-of-fit	–	53.46	0.0011 (significant)	–	5.41	0.0683 (insignificant)

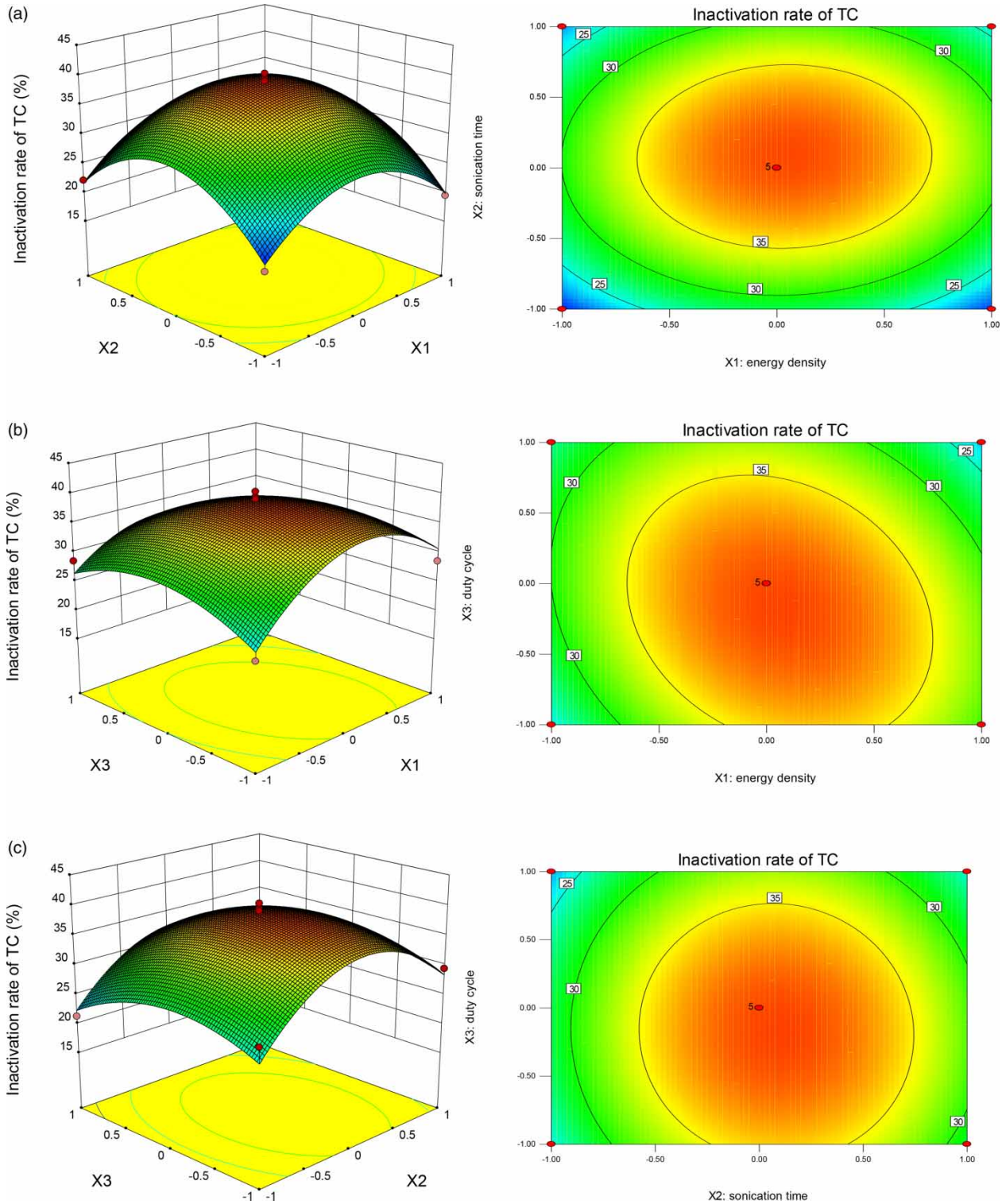

Figure 3 | Response surface and contour plots for the effect of independent variables on TC inactivation rate: (a) X_1-X_2; (b) X_1-X_3; (c) X_2-X_3.

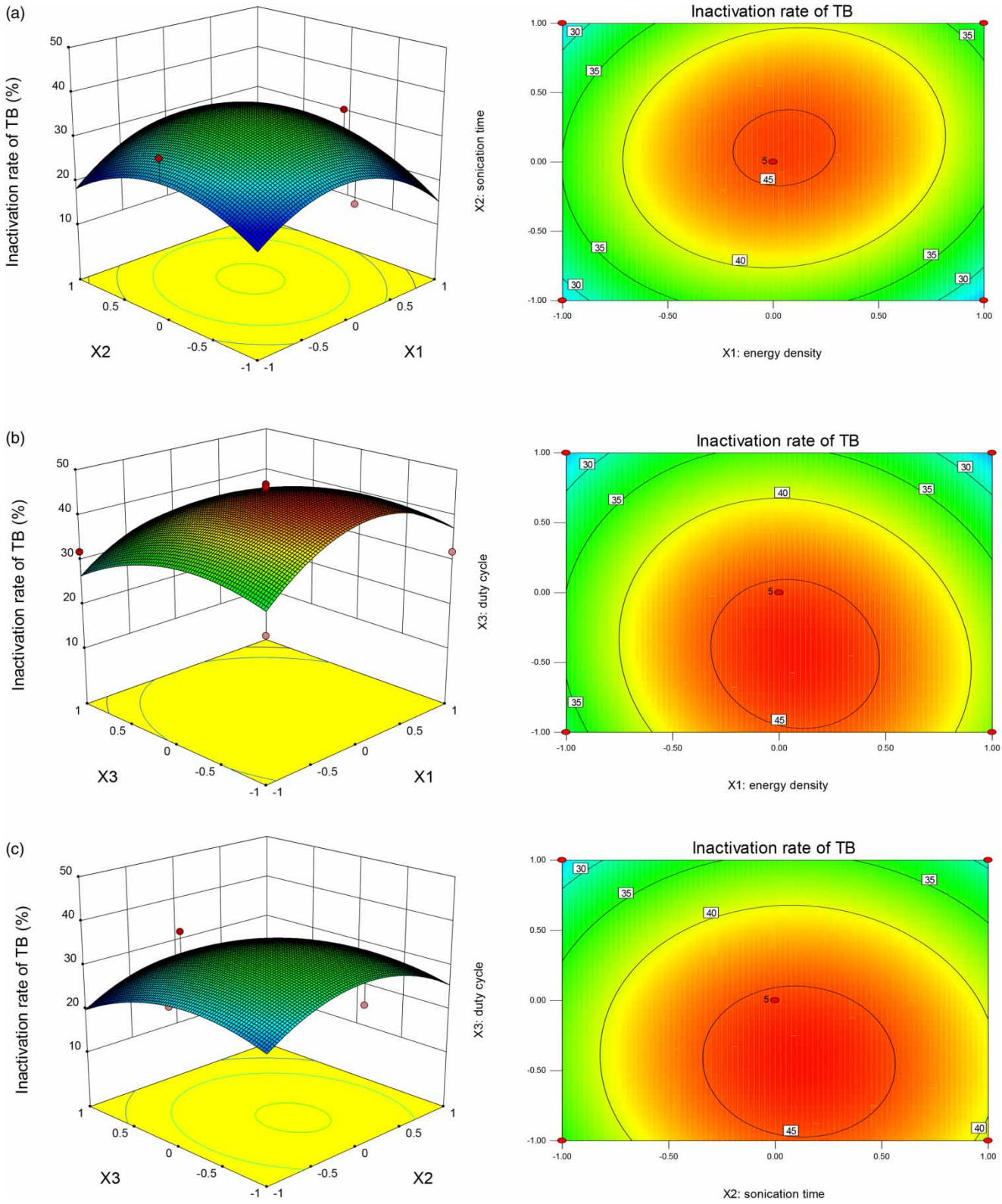

Figure 4 | Response surface and contour plots for the effect of independent variables on TB inactivation rate: (a) X_1-X_2; (b) X_1-X_3; (c) X_2-X_3.

sonication time; it can be concluded that the increased TC inactivation rate is correlated with the increase of sonication time from 300 s ($X_2 = -1$) to 950 s ($X_2 = 0.084$). With further increase in sonication time, a decline in the rate of inactivation of TC was observed. It is possible that extended inactivation time favored TC inactivation, as indicated in Figure 2(b). As shown in Figure 3(b), duty cycle played a relatively less important role than energy density, as the former had a flat inactivation rate curve, while the latter had a steep one. This was not the same as the case displayed in Table 3, wherein duty cycle was the most critical variable, as indicated by the *p*-values of the coefficients X_1, X_2, and X_3. The maximum TC inactivation efficiency was achieved at an energy density and a duty cycle of 8.26 W/mL ($X_1 = 0.0657$) and 0.64:0.36 ($X_3 = -0.198$), respectively. The interaction of sonication time and duty cycle is shown in Figure 3(c). The relationship between these two variables showed the same trend as did energy density and duty cycle. Maximum inactivation rate was achieved when sonication time was 950 s ($X_2 = 0.084$) and duty cycle was 0.64:0.36 ($X_3 = -0.198$).

Response surfaces and contour plots of TB are shown in Figure 4. Figure 4(a) shows the interaction between energy density and sonication time, suggesting that an increase in energy density and sonication time enhanced TB inactivation up to a certain threshold, and further increase in both energy density and sonication time led to a decline in inactivation rates. This trend was consistent with that observed for TC. The highest inactivation rate was achieved when the energy density and the sonication time were 8.34 W/mL ($X_1 = 0.0860$) and 973 s ($X_2 = 0.122$), respectively. A duty cycle of 0.69:0.31 ($X_3 = -0.447$) resulted in the maximal inactivation rate of TB (Figure 4(b) and 4(c)).

Verification of predictive models

In the present study, an optimization experiment was performed to evaluate the optimal inactivation parameters for TB and TC. Our goal was to obtain high bacterial inactivation rates. Two optimal inactivation parameters were established: for TB, an energy density of 8.34 W/mL ($X_1 = 0.0860$), sonication time of 973 s ($X_2 = 0.122$), and duty cycle of 0.69:0.31 ($X_3 = -0.447$); for TC, an energy density of 8.26 W/mL ($X_1 = 0.0657$), sonication time of 950 s ($X_2 = 0.084$), and duty cycle of 0.64:0.36 ($X_3 = -0.198$). As TB and TC were in the same aqueous sample, we determined the optimum US conditions to be as follows: energy density of 8.30 W/mL ($X_1 = 0.075$), sonication time of 950 s ($X_2 = 0.084$), and duty cycle of 0.7:0.3 ($X_3 = -0.5$). Under optimal conditions, the experimental values of TB and TC inactivation rates were 47.26% ± 4.35% and 39.23% ± 2.27%, respectively, while the predicted values were 46.57% and 38.65%. No significant differences were observed between the experimental and predicted values in both TB and TC inactivation rates. Therefore, the model can be used to optimize the process of US-induced inactivation of bacterial mixture.

In this study, the energy efficiency analysis for TB and TC was conducted according to Equation (3) under optimum US conditions. For TB and TC with average inactivation rates of 47.26% and 39.23%, respectively, the corresponding EEs were 8.0 and 1.2 CFU/J, respectively. Therefore, when we treated 1-m^3 water sample, and used the inactivation rate of TC as the 'treatment goal', the electricity used was only 0.34 RMB, that is, under 1.0 RMB/kWh. This clearly indicated that optimized US conditions are a very important process.

CONCLUSIONS

RSM was successfully employed to optimize inactivation conditions for the model bacterial mixture, in particular, for TC. Two sets of optimal inactivation conditions were established for the different behaviors of TB and TC under US. We finally determined the optimum US condition to be as follows: an energy density of 8.30 W/mL, sonication time of 950 s, and duty cycle of 0.7:0.3. Under such conditions, the experimental values for TB and TC were 47.26% ± 4.35% and 39.23% ± 2.27%, respectively, while the predicted values were 46.57% and 38.65%, respectively. No significant differences were observed between the experimental and predicted values; this indicates that the model can be used to optimize the process of US-induced inactivation in a bacterial mixture.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support extended by the National Natural Science Foundation (Grant No. 51278005) and Beijing Natural Science Foundation (8132007) of China and Doctoral Fund of Innovation of Beijing University of Technology, as well as Shanghai Tongji Gao Tingyao Environmental Science and Technology Development Foundation (STGEF). The kind suggestions from the anonymous reviewers are highly appreciated.

REFERENCES

APHA 1998 *Standard Methods for the Examination of Water and Wastewater*, 20th edn. American Public Health Association/ Water Environment Federation, Washington, DC, USA.

Ashokkumar, M., Vu, T., Grieser, F., Weerawardena, A., Anderson, N., Pilkington, N. & Dixon, D. R. 2003 Ultrasonic treatment of *Cryptosporidium* oocysts. *Water Sci. Technol.* **47** (3), 173–177.

Drakopoulou, S., Terzakis, S., Fountoulakis, M. S., Mantzavinos, D. & Manios, T. 2009 Ultrasound-induced inactivation of gram-negative and gram-positive bacteria in secondary treated municipal wastewater. *Ultrasonics Sonochem.* **16** (5), 629–634.

Gao, S. P., Lewis, G. D., Ashokkumar, M. & Hemar, Y. 2014a Inactivation of microorganisms by low-frequency high-power ultrasound: a simple model for the inactivation mechanism. *Ultrasononics Sonochem.* **21** (1), 454–460.

Gao, S. P., Lewis, G. D., Ashokkumar, M. & Hemar, Y. 2014b Inactivation of microorganisms by low-frequency high-power ultrasound: effect of growth phase and capsule properties of the bacteria. *Ultrasonics Sonochem.* **21** (1), 446–453.

Ghafari, S., Aziz, H. A., Isa, M. H. & Zinatizadeh Ali, A. 2009 Application of response surface methodology (RSM) to optimize coagulation–flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum. *J. Hazard. Mater.* **163** (2–3), 650–656.

Gonze, E., Pillot, S., Valette, E., Gonthier, S. & Bernis, A. 2003 Ultrasonic treatment of aerobic activated sludge in batch reactor. *Chem. Eng. Process.* **42** (12), 965–975.

Herceg, Z., Jambrak, A. R., Lelas, V. & Thagard, S. M. 2012 The effect of high intensity ultrasound treatment on the amount of *Staphylococcus aureus* and *Escherichia coli* in milk. *Food Technol. Biotechnol.* **50** (1), 46–52.

Hossain, Md. S., Nik Ab Rahman, N. N., Balakrishnan, V., Alkarkhi Abbas, F. M., Rajion, Z. A. & Kadir Mohd, O. A. 2015 Optimizing supercritical carbon dioxide in the inactivation of bacteria in clinical solid waste by using response surface methodology. *Waste Manage.* **38**, 462–473.

Joyce, E., Phull, S. S., Lorimer, J. P. & Mason, T. J. 2003 The development and evaluation of ultrasound for the treatment of bacterial suspensions. A study of frequency, power and sonication time on cultured *Bacillus* species. *Ultrasonics Sonochem.* **10** (6), 315–318.

Joyce, E. M., Mason, T. J. & Lorimer, J. P. 2006 Application of UV radiation or electrochemistry in conjunction with power ultrasound for the disinfection of water. *Int. J. Environ. Pollut.* **27** (1–3), 222–230.

Jyoti, K. K. & Pandit, A. B. 2004 Effect of cavitation on chemical disinfection efficiency. *Water Res.* **38** (9), 2248–2257.

Liu, W., Cheung, L. M., Yang, X. & Shang, C. 2006 THM, HAA and CNCl formation from UV irradiation and chlor(am)-ination of selected organic waters. *Water Res.* **40** (10), 2033–2043.

Malley, J. P., Shaw, J. P. & Ropp, J. R. 1995 *Evaluation of by-products produced by treatment of groundwaters with ultraviolet irradiation*. AWWARF, Denver, CO, USA.

Pilli, S., Bhunia, P., Yan, S., LeBlanc, R. J., Tyagi, R. D. & Surampalli, R. Y. 2011 Ultrasonic pretreatment of sludge: a review. *Ultrasononics Sonochem.* **18** (1), 1–18.

Prasad, K. N., Hassan, F. A., Yang, B., Kong, K. W., Ramanan, R. N., Azlan, A. & Ismail, A. 2011 Response surface optimisation for the extraction of phenolic compounds and antioxidant capacities of underutilized *Mangifera pajang* Kosterm. *Peels. Food Chem.* **128** (1), 1121–1127.

Richardson, S. D. 2003 Disinfection by-products and other emerging contaminants in drinking water. *TrAC Trends Anal. Chem.* **22** (10), 666–684.

Wang, X. S., Wu, Y. F., Chen, G. Y., Yue, W., Liang, Q. L. & Wu, Q. N. 2013 Optimisation of ultrasound assisted extraction of phenolic compounds from *Sparganii rhizoma* with response surface methodology. *Ultrasonics Sonochem.* **20** (3), 846–854.

Yao, J. J., Michael, R. H., Gao, N. Y., Zhang, Z. & Li, L. 2011 Sonolytic degradation of dimethoate: kinetics, mechanisms and toxic intermediates controlling. *Water Res.* **45** (18), 5886–5894.

First received 10 April 2015; accepted in revised form 18 September 2015. Available online 17 October 2015