ARCHIVES OF ACOUSTICS Vol. 47, No. 4, pp. 513–518 (2022), doi: 10.24425/aoa.2022.142898

Research Paper

Energy Analysis of Cavitation Bubbles Under Dual-Frequency Acoustic Excitation

Liang LV[(1)], Kai HU[(2)], Fei LIU[(2)*], Yawei LI[(2)], Bing CUI[(1)]

[(1)] *School of Mechano-Electronic Engineering, Suzhou Vocational University*
Suzhou, China

[(2)] *Department of Sports Health and Art Education, Hebei Petroleum University of Technology*
Chengde, China

*Corresponding Author e-mail: liufei_123321@163.com

(*received January 28, 2022; accepted July 26, 2022*)

Cavitation has been widely used in wastewater degradation, material synthesis and biomedical field under dual-frequency acoustic excitation. The applications of cavitation are closely related to the power (i.e. the rate of internal energy accumulation) during bubble collapse. The Keller–Miksis equation considering liquid viscosity, surface tension and liquid compressibility is used to describe the radial motion of the bubble. The model is built in predicting the power during bubble collapse under dual-frequency acoustic excitation. The influences of parameters (i.e. phase difference, frequency difference, and amplitude ratio) on the power are investigated numerically. With the increase of phase difference, the power can be fluctuated in a wide range at all conditions. Three typical characteristics of the power appear under the effects of frequency difference and amplitude ratio. With the increase of amplitude ratio, if the frequency difference is small, the power has two maximum values; and if the frequency difference is medium, there is a maximum value. Otherwise, the power monotonously decreases. The results can provide theoretical references for the selections of experimental parameters of sonoluminescence and sonochemistry in the dual-frequency acoustic field.

Keywords: dual-frequency acoustic excitation; power; sonoluminescence; sonochemistry.

1. Introduction

Under irradiation of acoustic waves, the bubble in the liquid grows and may collapse violently, which is termed as "acoustic cavitation". The intense collapse of the bubble can produce extremely high temperature and pressure around it, and this environment has been applied in many fields, such as ultrasonic images (LV *et al.*, 2020), water treatment (ZUPANC *et al.*, 2016), non-contact therapy (COUSSIOS *et al.*, 2008), ultrasonic cleaning (MASON, 2016) and viscosity reduction for residual oil (HUANG *et al.*, 2018). Especially, in the dual-frequency sound field, the active cavitation volume and the utilization rate of acoustic energy are all implemented.

Dual-frequency acoustic system is often used to increase the intensity of sonoluminescence, which is an indicator of cavitation activity in acoustic reactor. Based on the theoretical and numerical results in the paper written by KREFTING *et al.* (2002), compared to single-frequency excitation, the bubble driven by dual-frequency was reported to increase the maximum light output by 3 times. With the same energy, the sonoluminescence intensity was enhanced when the energy of low frequency was fixed and the energy of high frequency was low (KANTHALE *et al.*, 2008). The rate of chemical reaction is increased and the structure of acoustic chemical reactor can be improved in the dual-frequency acoustic field. MOHOLKAR (2009) analyzed the effect of two major parameters (i.e. frequency and relative phase) in dual-frequency system on the maximum and most balanced energy dissipation of the acoustic chemical reactor. The results showed that the free radical production under the most uniform energy dissipation was twice as high as that under the maximum energy dissipation. The influence of relative phase was greater than that of frequency ratio. Waldo and Vecitis (2018) experimentally designed a variety of multi-frequency piezoelectric crystals and reactor components, and found that some component structures could increase the efficiency of chemical reactions by three times. Dual-frequency system has great applications in biological field, such as improving the efficiency of non-contact therapy and the accuracy of image. Yeh *et al.* (2008) found that when the envelope frequency was close to the microbubble resonance frequency, the dual-frequency system could improve the signal ratio and significantly cause nonlinear scattering. A dual-piezoelectric device was used to crush stones by Loske *et al.* (2002), and the efficiency of lithotripsy was improved while tissue damage was reduced.

Recently, bubble dynamics under acoustic excitation with dual-frequency have been investigated by many researchers, in order to enhance the effects of application in the fields of sonoluminescence, sonochemistry and non-contact therapy, etc. Compared to the single-frequency system, the cavitation in the dual-frequency system can be enhanced due to some special phenomena, such as appearing combination resonance (Zhang *et al.*, 2017), reducing the critical radius of the bubble instability (Zhang *et al.*, 2015) and inertial cavitation thresholds (Suo *et al.*, 2018), enhancing the maximum bubble response (Guédra *et al.*, 2017) and improving mass transfer across the bubble interface (Zhang *et al.*, 2015). As far as we know, the internal energy of the bubble in dual-frequency excitation sound field has not been revealed, which is closely related to bubble dynamics, e.g. radial oscillations, collapse time, temperature and pressure inside the bubble.

In the present paper, a model to predict the internal energy of a bubble in dual-frequency excitation sound field is established, and the influence of key parameters on bubble oscillations from the perspective of energy is analyzed. The paper is organized as follows. In Sec. 2, the basic equations and the calculation method are provided. In Sec. 3, the results and limitations of the present paper are discussed. Section 4 concludes the main findings of the present work.

2. Equations and calculation method

Cavitation occurs under ultrasonic radiation in the liquid. The equilibrium radius of the bubble is generally within 100 μm, and the time scales of expansion and compression are of the orders of microseconds. Therefore, the calculation model adopted in this paper has the following assumptions: (1) the bubble keeps spherical symmetry during its oscillations; (2) the surrounding liquid is infinite and uniform Newtonian fluid; (3) the influence of gravity and buoyancy are ignored; (4) the effect of gas diffusion during bubble oscillations is ignored, i.e. the equilibrium radius remains constant; (5) the bubble expansion and compression processes are assumed to be isothermal and adiabatic, respectively. The bubble radial motion is predicted by Keller–Miksis equation, which considers the influence of liquid viscosity, surface tension and compressibility (Zhang *et al.*, 2017; Guédra *et al.*, 2017; Suo *et al.*, 2018):

$$\rho\left(1-\frac{\dot{R}}{c}\right)R\ddot{R}+\frac{3}{2}\rho\dot{R}^2\left(1-\frac{1}{3}\frac{\dot{R}}{c}\right) = \left(1+\frac{\dot{R}}{c}\right)\left[p_{\text{ext}}\left(R,t\right)-p_s\left(t\right)\right] + \frac{R}{c}\frac{\mathrm{d}}{\mathrm{d}t}\left[p_{\text{ext}}\left(R,t\right)-p_s(t)\right], \tag{1}$$

$$p_{\text{ext}}(R,t)=\left(P_0+\frac{2\sigma}{R_0}\right)\left(\frac{R_0}{R}\right)^{3\gamma}-\frac{2\sigma}{R}-\frac{4\mu}{R}\dot{R}, \tag{2}$$

$$p_s(t)=P_0+P_{A1}\cos\left(2\pi f_1 t\right)+P_{A2}\cos\left(2\pi f_2 t+\varphi\right), \tag{3}$$

where R is the instantaneous bubble radius; the over-dot denotes the time derivative; ρ is the density of the liquid; c is the speed of sound in the liquid; σ is the surface tension coefficient of the liquid; μ is the viscosity of the liquid; P_0 is the ambient pressure; γ is the polytropic exponent; R_0 is the equilibrium bubble radius; P_{A1} and P_{A2} are the amplitudes of the external acoustic excitation, and the common setting is $P_e^2=P_{A1}^2+P_{A2}^2$; f_1 and f_2 are the frequencies of the external acoustic excitation, and ϕ is the phase difference between them.

In this paper, the internal energy during the bubble oscillations under dual-frequency acoustic excitation is analyzed, and the energy transfer inside and outside the bubble is ignored. The expression of internal energy is calculated by Eq. (4), and the detailed derivation is referred to Merouani *et al.* (2014):

$$\Delta E=\frac{p(t)V(t)-p_{\min}V_{\max}}{\gamma-1}, \tag{4}$$

where $p(t)$ and $V(t)$ are the internal pressure and bubble volume as a function of time, and $p_{\min}$ and $V_{\max}$, respectively, are the minimum internal pressure and the maximum bubble volume.

In order to compare the efficiency of the cavitation under different operating conditions, it is necessary to analyze the time scale of the internal energy accumulation. Therefore, the expression of the bubble collapse power w, which is defined as the internal energy divided by the duration of the bubble collapse τ_c, is as follows:

$$w=\frac{\Delta E}{\tau_c}, \tag{5}$$

where τ_c is defined as the time when the bubble compresses to 7% of its maximum radius (Merouani *et al.*, 2014). For the sake of discussions, the non-dimensional parameter is employed as below:

$$N = \frac{P_{A2}}{P_{A1}}. \tag{6}$$

The model is numerically solved using the method of 4th–5th order Runge–Kutta formula with variable step length. To obtain the radius-time curve satisfying the precision requirement, the absolute error and relative error are 10^{-12} and 10^{-7}, respectively. If not specified, the following parameters are used in calculations: T_∞ = 293.15 K; P_0 = 1 bar; ρ = 1000 kg/m^3; σ = 0.072 N/m; c = 1480 m/s; γ = 1.4; μ = 0.001 Pa·s; R_0 = 5 μm; ϕ = 0.

3. Results and discussion

When two acoustic waves with different frequencies are superimposed on each other, there are some different characteristics under the superimposed wave excitation, and the radial motion of the bubble will be changed first. The change trends of the instantaneous bubble radius with time under single-frequency and dual-frequency excitation are shown in Fig. 1. The maximum bubble radii are $15.11R_0$, $9.01R_0$, and $7.81R_0$ for single frequencies of 20 kHz, 35 kHz, and 140 kHz, respectively. While in the cases of the dual frequency, there are $21.21R_0$ and $8.72R_0$ for 20 + 35 kHz, and 20+140 kHz, respectively. It can be seen that when the difference between two acoustic frequencies is small, the maximum bubble radius is greater than that of the two excitation waves acting alone. However, when the frequency difference is larger, the maximum size of the bubble expansion under dual-frequency excitation is between the maximum radius of two excitation waves acting alone. In the study of Suo *et al.* (2018), when the difference between two excitation frequencies is small, the threshold of the internal cavitation decreases greatly, which is consistent with the above results.

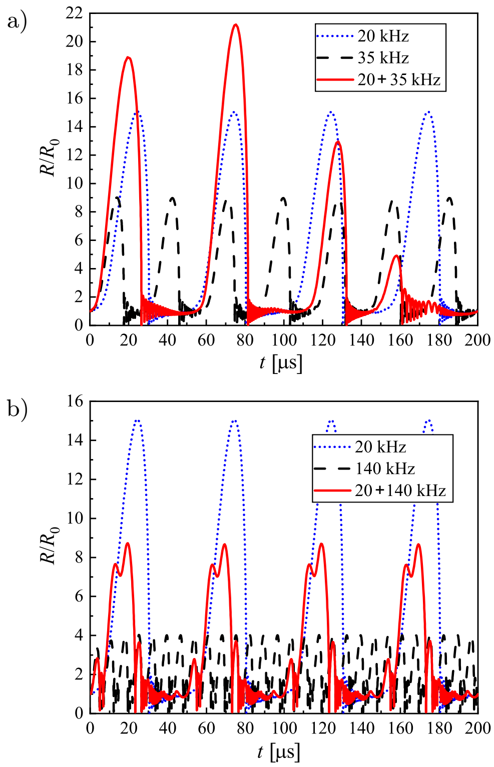

Fig. 1. Instantaneous radius of a single bubble in the acoustic field with the frequencies of: a) 20 kHz, 35 kHz, and 20 + 35 kHz; b) 20 kHz, 140 kHz, and 20 + 140 kHz (P_e = 1.5 bar, N = 1).

According to the theory in (Tinguely *et al.*, 2018, i.e. Eq. (9)), it can be found that the initial energy of the bubble, which is equal to the sum of the potential energy of the rebound, the work done by the non-condensable gas and the compression energy of shock wave, is proportional to the cubic power of the maximum bubble radius. Therefore, the capacity of the bubble to gather energy can be reflected by the maximum bubble radius. During the bubble collapse, some of the energy is converted to the chemical energy. The time of the bubble collapse plays a significant role in the concentration of the free radicals inside the bubble. The shorter the collapse time is, the greater the concentration of free radicals is. In (Tatake, Pandit, 2002), the dynamic characteristics of the bubble was predicted by the parameter R^3_{max}/t_c, which was employed to describe the acoustic cavitation and hydraulic cavitation well. The authors have concluded that the pressure during the bubble collapse increased monotonously with this parameter. Although this parameter can reflect the cavitation intensity and indirectly predict the collapse pressure of the bubble, it can not estimate the internal energy inside the bubble, which is related to the efficiency of sonoluminescence and sonochemical reactions. Merouani *et al.* (2014) derived an expression to calculate the internal energy considering the time effect of the bubble collapse, namely, the bubble collapse power w, and analyzed the relationship between acoustic amplitude, frequency, static pressure, liquid temperature and the internal energy under single frequency excitation. The influence of the acoustic parameters (i.e. phase difference, frequency difference and amplitude ratio) in the dual-frequency acoustic system on the bubble collapse power will be analyzed below.

The effect of phase difference ϕ on bubble collapse power w is shown in Fig. 2, where ϕ increases from 0 to 2π with an internal of $1/36\pi$ (5°) between two adjacent steps. Under dual-frequency acoustic excitation, with the phase difference increase, the sensitivity of w variation increases first and then deceases. As observed in the figure, the maximum values are 1.54,

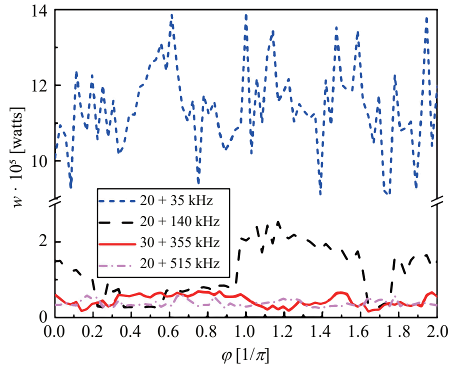

Fig. 2. Calculated power for one collapse of the bubble as the function of phase difference under dual-frequency acoustic excitation (P_e = 1.5 bar, N = 1).

9.50, 4.35, and 2.39 times of the minimum values in the excitation frequencies of 20 + 35 kHz, 20 + 140 kHz, 20 + 355 kHz, and 20 + 515 kHz, respectively. In the experimental and numerical studies of the dual-frequency excitation, 0 phase is generally adopted. Under this condition, the internal energy of the bubble is not the maximum, which reduces the energy utilization rate. Some current researches have also obtained the same conclusions. For example, Holzfuss *et al.* (1998) analyzed the sonoluminescence intensity under dual-frequency excitation, and found that there were multiple maxima and minima during the phase changing between 0 and 2π. Krefting *et al.* (2002) obtained consistent results. Moholkar (2009) found that the phase difference had a great influence on sonochemical reactions, which was higher than the degree of frequency difference.

The influence of frequency difference on bubble collapse power w under dual-frequency acoustic excitation with different driving pressure amplitudes is shown in Fig. 3. The pressure amplitude is a certain value, and w fluctuates continuously with the increase of frequency difference. The maximum w appears in the field of 20 + 20 kHz. There is a diversity of frequency difference corresponding to the minimum w. When the pressure amplitudes are 1.5 bar, 2.0 bar, 2.5 bar, and 3.0 bar, the minimum w occurs in the acoustic field with frequencies of 20 + 380 kHz, 20 + 340 kHz, 20 + 80 kHz, and 20 + 140 kHz. Meanwhile, the sensitivity of w variation to frequency difference decreases, and the maximum values are 307.28, 19.84, 11.15, and 8.77 times of the minimum values, respectively. When the two excitation frequencies are constant, the increase of pressure amplitude makes w enhance continuously. From the above analysis, it can be found that w is closely related to the driving pressure and frequency difference. Koda *et al.* (2003) found in the experimental studies that the sonochemical intensity and H_2O_2 production enhanced with the increase of acoustic power. The similar conclusions were obtained by Kanthale *et al.* (2008).

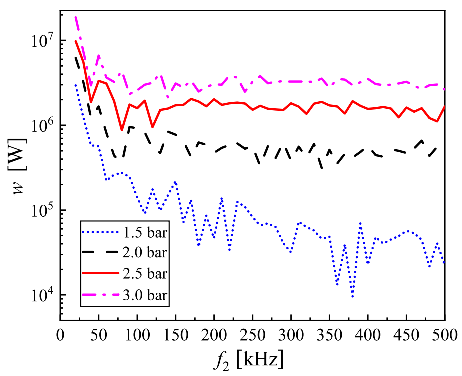

Fig. 3. Calculated power for one collapse of the bubble as the function of frequency difference under dual-frequency acoustic excitation (f_1 = 20 kHz, N = 1).

The effect of amplitude ratio N on bubble collapse power w under dual-frequency acoustic excitation with different frequencies is shown in Fig. 4. It can be found that when the frequency difference is small (20 + 35 kHz), w reaches the maximum at N = 0.5 and deceases when N is away from 0.5. When the frequency difference is large (20 + 140 kHz, 20 + 355 kHz, and 20 + 515 kHz), with increasing N, w is drastically reduced during N changing between 0 and 1. Nevertheless, N from 1 to 10, w fluctuates slightly. The sonochemical efficiency in the filed under dual-frequency acoustic excitation of 20 + 355 kHz with different powers was compared in Brotchie *et al.* (2010). It was found that the highest sonochemical efficiency was obtained in the acoustic field excited by the frequency of 20 kHz with the input power of 5 W together with the frequency of 355 kHz with the input power of 10 W.

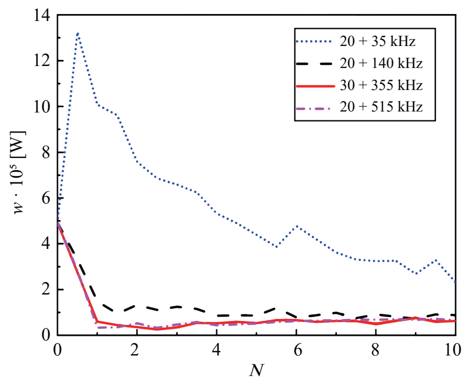

Fig. 4. Calculated power for one collapse of the bubble as the function of N under dual-frequency acoustic excitation (P_e = 1.5 bar).

The frequency difference and amplitude ratio N play significant roles in the bubble collapse power w under dual-frequency acoustic excitation as shown in Fig. 4. Therefore, Fig. 5 shows in detail the variation of w with f_2 and N when f_1 is a fixed value of 20 kHz. With the increase of N, the effect of the frequency difference

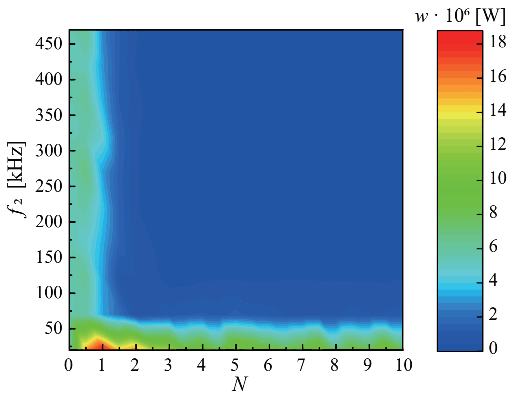

Fig. 5. Calculated power for one collapse of the bubble versus N and frequency difference under dual-frequency acoustic excitation (P_e = 1.5 bar, f_1 = 20 kHz).

on w can be divided into three typical regions. When f_2 is between 20 kHz and 32 kHz, there are two maximum points of w, which are near N = 1 and N = 2. As f_2 changing from 32 kHz to 64 kHz, w has a maximum value corresponding to N in the range of 0.5–1. When f_2 is larger than 64 kHz, w decreases gradually.

The paper focuses on the effects of the control parameters (phase difference, frequency difference, and amplitude ratio) in the dual-frequency acoustic system on sonoluminescence and sonochemistry. The results of internal energy inside the bubble variation with environment can be obtained in (Merouani *et al.*, 2014). The present model does not include rectified mass diffusion (Zhang *et al.*, 2015) (i.e. the gas diffuses into or out of bubble interface), which process is rather slow. The internal energy variation inside the bubble is a transient process, hence, the rectified mass diffusion can be safely ignored. According to the literature (Mettin *et al.*, 1997; Moshaii, Sadighi-Bonabi, 2004; Yang, Church, 2005), introducing bubble-bubble interaction, liquid compressional viscosity and the viscoelastic dissipation into Keller–Miksis will weaken the bubble collapse intensity, nonetheless, the variation trends of radius, pressure and temperature with time are consistent. Therefore, the present model can provide good references for the selection of sonoluminescence and sonochemistry parameters qualitatively. In order to obtain the quantitative suggestions, establishing a more accurate model will be the main work of our research group.

4. Conclusions

In this paper, a model for predicting the collapse power inside the bubble under dual-frequency acoustic excitation has been established. The effects of paramount parameters (e.g. frequency difference) have been discussed within a wide range. They are consistent with the experimental and numerical results of sonoluminescence and sonochemistry. The main findings are as follows. The variation of phase difference causes the continuous change of collapse power with multiple maximum and minimum values. With the increase of frequency difference, the sensitivity of collapse power variation to phase difference first increases and then decreases. The f_1 is fixed, and amplitude ratio increases. When f_2 is between 20 and 32 kHz, there are two maximum values of collapse power; when f_2 is between 32 and 64 kHz, there is a maximum value of collapse power; when f_2 is larger than 64 kHz, the collapse power decreases continuously.

Acknowledgment

The authors would like to acknowledge S&T Program of Hebei (Project No.: 20475704D).

References

1. Brotchie A., Grieser F., Ashokkumar M. (2010), Characterization of acoustic cavitation bubbles in different sound fields, *Journal of Physical Chemistry B*, **114**(34): 11010–11016, doi: 10.1021/jp105618q.
2. Coussios C.C., Roy R.A. (2008), Applications of acoustics and cavitation to noninvasive therapy and drug delivery, *Annual Review of Fluid Mechanics*, 2008, **40**(1): 395–420, doi: 10.1146/annurev.fluid.40.111406.102116.
3. Guédra M., Inserra C., Gilles B. (2017), Accompanying the frequency shift of the nonlinear resonance of a gas bubble using a dual-frequency excitation, *Ultrasonics Sonochemistry*, **38**(1): 298–305, doi: 10.1016/j.ultsonch.2017.03.028.
4. Holzfuss J., Rüggeberg M., Mettin R. (1998), Boosting sonoluminescence, *Physical Review Letters*, **81**(9): 1961–1964, doi: 10.1103/PhysRevLett.81.1961.
5. Huang X.T., Zhou C.H., Suo Q.Y., Zhang L.T., Wang S.H. (2018), Experimental study on viscosity reduction for residual oil by ultrasonic, *Ultrasonics Sonochemistry*, **41**(1): 661–669, doi: 10.1016/j.ultsonch.2017.09.021.
6. Kanthale P.M., Brotchie A., Ashokkumar M., Grieser F. (2008), Experimental and theoretical investigations on sonoluminescence under dual frequency conditions, *Ultrasonics Sonochemistry*, **15**(4): 629–635, doi: 10.1016/j.ultsonch.2007.08.006.
7. Koda S., Kimura T., Kondo T., Mitome H. (2003), A standard method to calibrate sonochemical efficiency of an individual reaction system, *Ultrasonics Sonochemistry*, **10**(3): 149–156, doi: 10.1016/S1350-4177(03)00084-1.
8. Krefting D., Mettin R., Lauterborn W. (2002), Two-frequency driven single-bubble sonoluminescence, *Journal of the Acoustical Society of America*, **112**(5): 1918–1927, doi: 10.1121/1.1509427.
9. Loske A.M., Prieto F.E., Fernández F., Cauwelaert J.V. (2002), Tandem shock wave cavitation enhancement for extracorporeal lithotripsy, *Physics in*

Medicine and Biology, **47**(22): 3945-3957, doi: 10.1088/0031-9155/47/22/303.

10. Lv L., Zhang Y. X., Wang L.Y. (2020), Effects of liquid compressibility on the dynamics of ultrasound contrast agent microbubbles, *Fluid Dynamics Research*, **52**(5): 1–17, doi: 10.1088/1873-7005/abb09b.

11. Mason T.J. (2016), Ultrasonic cleaning: An historical perspective, *Ultrasonics Sonochemistry*, **29**: 519–523, doi: 10.1016/j.ultsonch.2015.05.004.

12. Merouani S., Hamdaoui O., Rezgui Y., Guemini M. (2014), Energy analysis during acoustic bubble oscillations: Relationship between bubble energy and sonochemical parameters, *Ultrasonics*, **54**(1): 227–232, doi: 10.1016/j.ultras.2013.04.014.

13. Mettin R., Akhatov I., Parlitz U., Ohl C.D., Lauterborn W. (1997), Bjerknes forces between small cavitation bubbles in a strong acoustic field, *Physical Review E*, **56**(3): 2924–2931, doi: 10.1103/PhysRevE.56.2924.

14. Moholkar V.S. (2009), Mechanistic optimization of a dual frequency sonochemical reactor, *Chemical Engineering Science*, **64**(24): 5255–5267, doi: 10.1016/j.ces.2009.08.037.

15. Moshaii A., Sadighi-Bonabi R. (2004), Role of liquid compressional viscosity in the dynamics of a sonoluminescing bubble, *Physical Review E*, **70**: 016304, doi: 10.1103/physreve.70.016304.

16. Suo D.J., Govind B., Zhang S.Q., Jing Y. (2018), Numerical investigation of the inertial cavitation threshold under multi-frequency ultrasound, *Ultrasonics Sonochemistry*, **41**: 419–426, doi: 10.1016/j.ultsonch.2017.10.004.

17. Tatake P.A., Pandit A.B. (2002), Modelling and experimental investigation into cavity dynamics and cavitational yield: Influence of dual frequency ultrasound sources, *Chemical Engineering Science*, **57**(22): 4987–4995, doi: 10.1016/S0009-2509(02)00271-3.

18. Tinguely M., Obreschkow D., Kobel P., Dorsaz N., Bosset A., Farhat M. (2012), Energy partition at the collapse of spherical cavitation bubbles, *Physical Review E*, **86**: 046315, doi: 10.1103/PhysRevE.86.046315.

19. Waldo N.B., Vecitis C.D. (2018), Combined effects of phase-shift and power distribution on efficiency of dual high-frequency sonochemistry, *Ultrasonics Sonochemistry*, **41**(1): 100–108, doi: 10.1016/j.ultsonch.2017.09.010.

20. Yang X., Church C.C. (2005), A model for the dynamics of gas bubbles in soft tissue, *The Journal of the Acoustical Society of America*, **118**(6): 3595–3606, doi: 10.1121/1.2118307.

21. Yeh C.K., Su S.Y., Shen C.C., Li M.L. (2008), Dual high-frequency difference excitation for contrast detection, *IEEE Transactions on Ultrasonics Ferroelectrics and Frequency Control*, **55**(10): 2164–2175, doi: 10.1109/TUFFC.916.

22. Zhang Y.N., Billson D., Li S.C. (2015), Influences of pressure amplitudes and frequencies of dual-frequency acoustic excitation on the mass transfer across interfaces of gas bubbles, *International Communications in Heat and Mass Transfer*, **66**: 167–171, doi: 10.1016/j.icheatmasstransfer.2015.05.026.

23. Zhang Y.N., Du X.Z., Xian H.Z., Wu Y.L. (2015), Instability of interfaces of gas bubbles in liquids under acoustic excitation with dual frequency, *Ultrasonics Sonochemistry*, **23**(1): 16–20, doi: 10.1016/j.ultsonch.2014.07.021.

24. Zhang Y.N., Zhang Y.N., Li S.C. (2017), Combination and simultaneous resonances of gas bubbles oscillating in liquids under dual-frequency acoustic excitation, *Ultrasonics Sonochemistry*, **35**(Part A): 431–439, doi: 10.1016/j.ultsonch.2016.10.022.

25. Zupanc M., Pandur Ž., Perdih T.S., Stopar D., Petkovšek M., Dular M. (2016), Effects of cavitation on different microorganisms: The current understanding of the mechanisms taking place behind the phenomenon. A review and proposals for further research, *Ultrasonics Sonochemistry*, **57**: 147–165, doi: 10.1016/j.ultsonch.2019.05.009.