Available online at www.sciencedirect.com
ScienceDirect
Journal of Hydrodynamics
2016,28(2):325-328
DOI: 10.1016/S1001-6058(16)60635-2
www.sciencedirect.com/science/journal/l0016058

Improved formulas for thermal behavior of oscillating nanobubbles*

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(Received October 3, 2015, Revised January 14, 2016)

Abstract: The study of nanobubbles (with sizes of the order of tens to hundreds of nanometers) is currently a hot spot of cavitation and bubble dynamics. In the literature, classical formulas are widely employed for the predictions of the thermal behavior of oscillating macro-bubbles. However, for modelling nanobubbles, the classical formulas may not be adequate due to the effects of the surface tension. In the present paper, a formula with the effects of surface tension fully considered is proposed for the predictions of thermal behavior. The predictions based on the classical formula are also presented for comparisons to show the advantages of the present formula.

Key words: nanobubbles, cavitation, thermal behavior, surface tension

Nanobubbles are bubbles with sub-micro sizes and gas inside in the aqueous solution. Recently, the presence of stable surface nanobubbles has attracted intensive studies[1,2]. Those surface nanobubbles have many unique characteristics, leading to plenty of applications in the industry[1]. For example, the surface nanobubbles could have a long lifetime up to several days[1]. The production of surface nanobubbles can be easily achieved through the so-called standard solvent exchange procedure (i.e., exchange of short-chain alcohol with water on solid substrate). The gaseous nature of surface nanobubbles has also been confirmed by experimental studies[1]. Lohse and Zhang[1] gave a full review of surface nanobubbles.

Owing to the small size of nanobubbles, many classical formulas may not be valid. For example, the classical diffusion theory predicts that the nanobubbles should be totally dissolved into the solution through the mass diffusion within tens of microseconds. However, the air nanobubbles could persist for more than four days, suggesting a new mechanism beyond the classical theory[1]. Similarly, the thermal behavior of nanobubbles has its features. The thermal damping mechanism is one of the important aspects for accurate modeling of bubbles. The thermal damping mechanism of macro-bubbles has been investigated by many researchers over several decades. Assuming a uniform pressure inside the gas bubbles, Devin[3] derived an analytical formula for the predictions of the thermal damping mechanism, which is currently highly cited in the literature and has been widely quoted by many textbooks and research papers. Formulas related with the thermal damping mechanism are widely employed for revealing underlying physics in the bubble phenomenon, e.g., the rectified mass diffusion, the wave propagation in bubbly liquids, the acoustical scattering, the bubble instability, the bubble-bubble interaction, and the sonoluminescence. Zhang[4] gave a brief review of the models for thermal effects.

In the present paper, the thermal behavior of spherical nanobubbles in the liquids are theoretically investigated. After a close re-examination of the classical formula, it is found that the effects of the surface tension are not fully considered in the aforementioned formula, leading to a less accurate prediction of the nanobubble behaviors.

In this section, the classical work by Devin[3] on

* Project supported by the National Natural Science Foundation of China (Grant No. 51506051), the Open Research Fund Program of State Key Laboratory of Water Resources and Hydropower Engineering Science, Wuhan University (Grant No. 2014SDG01).
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the spherical bulk bubbles will be introduced with details. The following assumptions are used[3]:

(1) The liquid temperature adjacent the bubble interface does not change so the liquid behaves as a heat reservoir[3]. Therefore, the equation of the energy conservation in the liquid was not solved in Ref.[3].

(2) The density and the specific heats of the gas are regarded as constants[3].

(3) The pressure in the gas bubbles is uniformly distributed. Therefore, the pressure in the gas is only a function of the time but not the radial coordinate[3].

(4) The boundary conditions on the bubble interface and at the bubble center are given as follows[3]: at the center of the bubble, the changes of the temperature must be finite and the gradient of the change of the temperature must be zero, on the bubble-liquid interface, the changes of the temperature must be zero and the gradient of the change of the temperature must be finite. If we define θ_1 as the change of the temperature inside the gas bubbles from the equilibrium absolute temperature, the boundary conditions at the center of the bubble are $\theta_1 \to \infty$ and $\mathrm{d}\theta_1/\mathrm{d}t = 0$ while the boundary conditions on the bubble-liquid interface are $\theta_1 = 0$ and $\mathrm{d}\theta_1/\mathrm{d}t \to \infty$.

(5) The oscillations of the pressure, the bubble volume and the temperature are assumed to be small[3].

In this section, the key points of the derivation process of Devin[3] is summarized. Here, most notations of Devin[3] are retained.

By differentiating the first law of thermodynamics, one obtains,

$$\frac{\mathrm{d}U}{\mathrm{d}t} = \frac{\mathrm{d}q}{\mathrm{d}t} + \frac{\mathrm{d}W}{\mathrm{d}t} \tag{1}$$

with

$$\frac{\mathrm{d}U}{\mathrm{d}t} = \rho_1 s_{v1}\frac{\mathrm{d}\theta_1}{\mathrm{d}t},\quad \frac{\mathrm{d}q}{\mathrm{d}t} = K_1\nabla^2\theta_1 = \frac{K_1}{r}\frac{\partial^2(r\theta_1)}{\partial r^2},$$

$$\frac{\mathrm{d}W}{\mathrm{d}t} = -\frac{P_2'}{v'}\frac{\partial v'}{\partial t}$$

Here, U is the internal energy of the gas bubble, q is the amount of heat transferred, W is the work done on the gas bubbles, t is the time, ρ_1 is the gas density, s_{v1} is the specific heat of the gas at a constant volume, θ_1 is the change of the temperature inside the gas bubbles from the equilibrium absolute temperature, K_1 is the thermal conductivity of the gas, r is the radial coordinate, P_2' is the pressure on a infinitesimal spherical shell with volume v'. Using the ideal gas law, v' in Eq.(1) can be eliminated, resulting in a differential equation of θ_1. Then the solution of θ_1 can be obtained through solving this differential equation with related boundary conditions.

Using the ideal gas law again, the dynamic volume of the oscillating gas bubble can be obtained. By employing the equation of the bubble motion, the stiffness and the energy loss caused by the thermal damping mechanism can both be obtained.

For convenience, the dissipation of the energy through the thermal damping mechanism is represented by a term related with the "effective thermal viscosity (μ_{th})" as done in Ref.[3]. The non-dimensional thermal damping constant (δ_{th}) is defined by Devin[3] as

$$\delta_{th} = \frac{4\mu_{th}\omega}{\rho_l R_0^2\omega_0^2} = \frac{2\beta_{th}\omega}{\omega_0^2}$$

Here, ω is the angular frequency of the driving sound field, μ_{th} is the effective thermal viscosity, ρ_l is the liquid density, R_0 is the equilibrium bubble radius, ω_0 is the natural frequency of the oscillating gas bubbles, and β_{th} is the thermal damping constant. Devin[3] gave the expression of δ_{th} as follows:

$$\delta_{th} = \frac{\mathrm{Im}(\Phi)}{\mathrm{Re}(\Phi)} \tag{2}$$

where

$$\Phi = \frac{3\gamma}{1-3(\gamma-1)\mathrm{i}\chi\left[\left(\frac{\mathrm{i}}{\chi}\right)^{1/2}\coth\left(\frac{\mathrm{i}}{\chi}\right)^{1/2}-1\right]} \tag{3}$$

$$\chi = \frac{D_{g,p}}{\omega R_0^2} \tag{4}$$

Here, Im and Re denote the imaginary and real parts of the function, respectively, $D_{g,p}$ is the thermal diffusivity of the gas at a constant pressure, γ is the ratio of the specific heats of the gas. Eqs.(2)-(4) is the widely cited formulas for the predictions of the thermal damping behavior of the oscillating bubbles. In Eqs.(3) and (4), the notations of Prosperetti are employed.

After the derivation of Devin's formulas as shown in the last section, it is found that the effects of the surface tension has not been fully considered. Hence, in this section, a correction is made following the framework of Devin[3] with most of his notations retained.

In Eq.(4) of Devin[3], the instantaneous pressure on the bubble interface (P_2') can be represented by the sum of two terms as follows

$$P_2' = P'\exp(\mathrm{i}\omega t) + P_0$$