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Bubble dynamics on the evolving bubble formed from the droplet at the superheat limit

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Abstract—The violent oscillation of the bubble formed from the evaporated droplet at the superheat limit has been investigated analytically and numerically. In this study, we have formulated a general bubble dynamics model, which is suitable for the oscillating bubble in an incompressible liquid medium. One of distinct features of this model is that the velocity and temperature distribution of the gas inside the bubble are obtained by solving continuity and energy equations for the gas analytically. With uniform density and temperature distribution approximation, the calculated values of the far field pressure signal from the evolving bubble formed from the fully evaporated droplet are in good agreement with experimental results.

1. INTRODUCTION

It is well known that one may heat a liquid held at 1 atm to a temperature far above its boiling point without the occurrence of boiling. The maximum temperature limit at which the liquid boils explosively is called the superheat limit of liquid. It has been verified experimentally that, when the temperature of a liquid droplet in an immiscible medium reaches its superheat limit at the ambient pressure of 1 atm, the droplet vaporizes explosively and becomes a bubble [1]. Since the pressure inside the bubble formed from the fully evaporated droplet is greater than the ambient pressure in this case [2], the bubble expands rapidly so that it overshoots the mechanical equilibrium condition and its size oscillates. Even though such rapid evaporation of droplets at the superheat limit and subsequent violent bubble evolution phenomena have been investigated in the pioneering work of previous researchers [1, 3], many aspects of the phenomena are virtually unknown. The detailed nature of the evaporation process at this limit may lead to the understanding of 'vapor explosion', which poses a potential hazard in the transport of liquid gas and in the operation of liquid metal fast breeder reactors [4, 5].

Using the Rayleigh equation and the functional relationship between the pressure inside the bubble (P_b) and the bubble volume (V), $P_b V^{n'} = \text{constant}$, which denotes the polytropic process, Kwak and Cho [6] calculate the far field pressure signal from the compact point source, i.e. the growing and collapsing bubble formed from the fully evaporated droplet at the superheat limit. The polytropic index n' can range in the interval from 1 (isothermal) to the ratio of specific heats γ (adiabatic). This polytropic approximation fails to account for the thermal damping effect on the far field signal because $P_b\,dV$ is a perfect differential. On the other hand, Prosperetti *et al.* [7] solved the energy equation for the gas inside bubble numerically to obtain the instantaneous temperature distribution for the gas in their bubble dynamics study. However, it is hard to tell whether the evolution process of the bubble formed form the droplet at its superheat limit is isothermal or adiabatic. Furthermore, it has never been discussed for which case the non-uniform temperature approximation is appropriate, while the polytropic approximation intrinsically assumes uniform density and temperature inside the bubble.

The thermal damping mechanism due to finite heat transfer between a bubble and the surrounding liquid was first considered by Moody [8]. He realized that this damping is simply a consequence of the loss of availability during a process in which there is entropy production. With this concept, he explained the ring-out mechanism clearly [9].

In this study, we have formulated a general bubble dynamics model, which is suitable for the oscillating bubble in an incompressible liquid medium. The velocity and temperature distribution of the gas inside the bubble was obtained by solving the continuity and energy equation for the gas with spatially uniform density analytically, which provides the temperature of the gas at the bubble wall and the heat flux inside the bubble. With this model, the polytropic approximation is no longer required to calculate the internal pressure of the evolving bubble. From this formulation uniform and nonuniform temperature distribution cases have been discussed.

With uniform density and temperature distribution approximation, the violent oscillation of the bubble formed from the evaporated butane droplet at its superheat limit (105°C) in hot ethylene glycol has been investigated. Calculations show that the bubble

NOMENCLATURE

C_g	sound speed in gas medium
C_p	heat capacity of vapor at constant pressure
$C_{p,\infty}$	heat capacity of liquid
C_v	heat capacity of vapor at constant volume
C_∞	sound speed in liquid medium
d_m	average distance between molecules
d_w	van der Waals diameter of molecules
E	internal energy
e	internal energy per mass
F_n	free energy needed to form n-mer cluster
h	enthalpy per mass
k_g	conductivity of gas (vapor)
k_l	conductivity of liquid
k_0	normalized constant of conductivity
m_b	vapor mass inside bubble
n	number of molecules in a cluster
n'	polytropic index
N	number density
P	pressure
P'	pressure wave signal
P_n	pressure of the evaporated molecules
P_0	gas pressure at bubble center
P_v	vapor pressure
Q	amount of heat transfer
q	heat flux
r	distance from the bubble center
R_b	radius of bubble
R_d	radius of droplet
R_g	gas constant
r_d	distance between observer and bubble center
S	entropy
S_g	entropy generation
T	temperature
T_{bl}	temperature at bubble–liquid interface
T_{b0}	temperature at the bubble center
t_0	normalized time constant
u_b	bubble wall velocity
u_0	normalized velocity constant
u_g	gas velocity inside bubble
u_r	radial velocity of liquid
V	bubble volume
V_m	effective molecular volume of liquid
z	coordination number.

Greek symbols

α_g	thermal diffusivity of gas
α_l	thermal diffusivity of liquid
α_0	normalized thermal diffusivity constant
γ	specific heat ratio of vapor
δ	thermal boundary layer thickness
η	loss factor by thermal damping
η_s	loss factor by sound radiation damping
ε_0	potential parameter of London dispersion attraction
ε_m	energy needed to separate a pair of molecule
ρ_c	critical density of liquid
ρ_g	density of gas inside bubble
ρ_m	density of metastable liquid phase
ρ_∞	density of ambient liquid
τ_b	relaxation time associated with rarefaction
τ_d	characteristic time of heat diffusion
Φ_v	viscous dissipation rate per unit volume
χ	dissipation coefficient of damping
ω	angular frequency of bubble oscillation.

Subscripts

b	bubble
c	critical cluster
l	liquid
non	starting point of nonlinear growing of bubble
∞	ambient liquid medium.

evolution process is neither isothermal nor adiabatic, and that the thermal damping due to finite heat exchange between vapor inside the bubble and the surrounding liquid plays a crucial role in bubble oscillation. Also the calculated values of the far field pressure signal from the evolving bubble are in good agreement with observed values.

2. BASIC EQUATIONS FOR THE ANALYSIS OF NONLINEAR BUBBLE EVOLUTION

(a) *Model with thermal boundary layer*

Consider a vapor bubble formed from the fully evaporated butane droplet at its superheat limit. Figure 1 shows the butane bubble of radius $R_b(t)$ in hot thermally conducting and incompressible liquid (ethylene glycol) at temperature T_∞ and distant pressure P_∞. Mass transfer through the bubble interface is negligible because evaporation of host liquid molecules and condensation of butane molecules hardly occur. In fact, the temperature of the surrounding liquid (105°C), which is the superheat limit of butane, is very high compared to the boiling point of butane (−0.5°C) and very low compared to the boiling point of glycerin (197°C). Only heat transfer due to the temperature difference between the vapor inside the bubble (T_b) and the surrounding liquid is considered in this analysis.

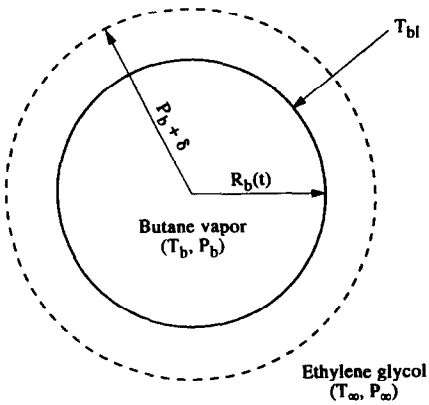

Fig. 1. Physical model for spherical bubble oscillation in liquid medium.

Heat transfer is assumed to occur through the thin boundary layer adjacent to the bubble wall. It is also assumed that the temperature profile in this layer is quadratic [10]:

$$\frac{T-T_\infty}{T_{bl}-T_\infty} = (1-\xi)^2 \tag{1}$$

where

$$\xi = \frac{r-R_b}{\delta}.$$

The boundary layer thickness, δ is, of course, a time-dependent quantity. Such a second-order curve satisfies the following boundary conditions used in this model:

$$T(R_b, t) = T_{bl} \quad T(R_b+\delta, t) = T_\infty \tag{2}$$

and

$$\left(\frac{\partial T}{\partial r}\right)_{r=R_b+\delta} = 0.$$

(b) *Overall energy equation for bubble*

The overall energy conservation for bubble in liquid medium may be written as [9]

$$\frac{dE}{dt} + P_b\frac{dV}{dt} - \frac{dQ}{dt} = 0. \tag{3}$$

Assuming that the vapor contents of the bubble obey the ideal gas law, the internal energy of the vapor can be expressed as

$$E = m_b C_{v,b} T_b = \frac{C_{v,b} P_b V}{R_g} = \frac{P_b V}{\gamma - 1}. \tag{4}$$

This equation implies that the gas is calorifically perfect at equilibrium [11] or the temperature of the gas is spatially uniform, in which the equation of state for an ideal gas, $P_g = \rho_g R_g T_b$, is valid.

The heat transfer conducted through the boundary layer can be obtained by applying the Fourier law at the bubble wall, or

$$\dot{Q} = k_l 4\pi R_b^2 \left.\frac{\partial T}{\partial r}\right|_{r=R_b} = -\frac{8\pi R_b^2 k_l (T_{bl}-T_\infty)}{\delta}. \tag{5}$$

Substituting equations (4) and (5) into equation (3), we have the time-dependent pressure inside the bubble, such as

$$\frac{dP_b}{dt} = -\frac{3\gamma P_b}{R_b}\frac{dR_b}{dt} - \frac{6(\gamma-1)k_l(T_{bl}-T_\infty)}{\delta R_b}. \tag{6}$$

(c) *Conservation equations for the gas inside the bubble*

If temperature distribution exists inside the bubble one cannot use the equilibrium relation, equation (4). In this section, conservation equations for gas inside the bubble are considered to obtain the gas pressure inside the bubble for the case where nonequilibrium phenomena exist.

The continuity equation for the gas with spatially uniform density may be written as

$$\frac{D\rho_g}{Dt} = \frac{d\rho_g}{dt} = -\rho_g \nabla \cdot u_g \tag{7}$$

where the notation of the total derivative used here is

$$\frac{D}{Dt} = \frac{d}{dt} + \mathbf{u}_g \cdot \frac{d}{dr}.$$

With equation (7) and the mass conservation for the gas inside the bubble, $D(\rho_g V)/Dt = 0$, which does not allow mass transfer, one may obtain the following radial-dependent gas velocity u_g as

$$u_g = \frac{\dot{R}_b}{R_b} r. \tag{8}$$

The gas density inside the bubble may be spatially uniform, provided that the period of bubble oscillation is much longer than the relaxation time which is required for the rarefaction due to bubble wall motion to propagate across the bubble, or $\tau_b = R_b/C_g$ [12]. In fact, the relaxation time for the bubble in this study is about 10 μs, which is much less than the period of bubble oscillation (ms).

The momentum equation for the gas inside the bubble can be written as follows, since the viscous force term vanishes with the velocity profile given in equation (8):

$$\rho_g \frac{D\mathbf{u}_g}{Dt} = -\nabla P_b. \tag{9}$$

The velocity profile given in equation (8) provides the pressure for the gas inside the bubble. That is

$$P_b = -\frac{\rho_g \ddot{R}_b}{2R_b} r^2 + P_0(t). \tag{10}$$

The velocity and pressure profiles given in equations (8) and (10) also satisfy the kinetic energy equation for the gas. Since the first term of the RHS in equation (10) is negligible compared to $P_0(t)$, the pressure inside the bubble can be regarded as uniform spatially. This is permissible until the characteristic time of bubble oscillation is much less than the relaxation time of the translational motion of gas molecules, which is about 10^{-9}–10^{-10} s, because the pressure is always related to the translational motion of molecules [11].

Assuming that the internal energy and enthalpy for the gas inside the bubble are functions of the temperature of the gas, or $de = C_{v,b}\,dT$ and $dh = C_{p,b}\,dT$, the energy equation for the gas can be written as [13]

$$\rho_g \frac{De}{Dt} = \rho_g C_{v,b} \frac{DT_b}{Dt} = -P_b \nabla \cdot \mathbf{u}_g - \nabla \cdot \mathbf{q} + \Phi_v. \quad (11)$$

Using the definition of enthalpy, the energy equation becomes

$$\rho_g \frac{Dh}{Dt} = \rho_g C_{p,b} \frac{DT_b}{Dt} = \frac{dP_b}{dt} - \nabla \cdot \mathbf{q} + \Phi_v. \quad (12)$$

Eliminating DT_b/Dt from equations (11) and (12), one may obtain the following equation:

$$\frac{dP_b}{dt} = -\gamma P_b \nabla \cdot \mathbf{u}_g - (\gamma - 1)\nabla \cdot \mathbf{q} + (\gamma - 1)\Phi_v. \quad (13)$$

Without the viscous dissipation term, this equation is the same one as equation (6), providing that

$$\nabla \cdot \mathbf{q} = \frac{6k_l(T_{bl} - T_\infty)}{\delta R_b} = -\frac{\left\{\dfrac{dP_b}{dt} + \dfrac{3\gamma P_b}{R_b}\dfrac{dR_b}{dt}\right\}}{(\gamma - 1)}. \quad (14)$$

Certainly this equation holds at the bubble wall as well as inside the bubble. With the velocity profile given in equation (8) and this heat flow rate per unit volume, the pressure inside the bubble is dependent on time only, as verified in equation (10).

(d) *Temperature distribution for gas inside the bubble*

The temperature distribution of the gas inside the bubble may be obtained by using the Fourier law and equation (14) by assuming that the conductivity for gas is constant. That is

$$T_b = \frac{r^2}{6(\gamma - 1)k_g}\left[\frac{dP_b}{dt} + 3\gamma P_b \frac{\dot{R}_b}{R_b}\right] + T_{b0}(t). \quad (15)$$

The above equation can be written, with the help of equation (14), as follows:

$$T_b = (T_{b0} - T_\infty)\left\{1 - \frac{\left(\dfrac{r}{R_b}\right)^2}{1 + \dfrac{k_g}{k_l}\dfrac{\delta}{R_b}}\right\} + T_\infty \quad (16)$$

which shows that the temperature distribution of the gas inside the bubble is also quadratic w.r.t. r. The temperature at the bubble wall can be obtained easily from equation (14) or equation (16).

$$T_{bl} = \frac{T_\infty + \dfrac{k_g}{k_l}\dfrac{\delta}{R_b}T_{b0}}{1 + \dfrac{k_g}{k_l}\dfrac{\delta}{R_b}}. \quad (17)$$

This relation shows how the bubble wall temperature is related to the temperature at the bubble center and the ambient temperature. Assigning an arbitrary value T_{bl} with a boundary condition is not permitted.

(e) *Uniform and nonuniform temperature distribution*

Using the property relation, $T\,ds = de + P\,d(1/\rho)$ and equation (11), the rate change of entropy for gas particles inside the bubble is given by

$$\rho_g T_b \frac{DS_b}{Dt} = -\nabla \cdot \mathbf{q} + \Phi_v. \quad (18)$$

If the characteristic time of the bubble evolution is long compared to the relaxation time τ_b, the viscous dissipation in equation (18) may be negligible [12]. Further, if there is no heat transport at the bubble wall (adiabatic wall), one may obtain the well-known relation $P_b V^\gamma$ = constant, which guarantees uniform temperature distribution inside the bubble as verified in equation (15).

The uniform temperature distribution also occurs when there is no heat flux or $\mathbf{q} = 0$ inside the bubble. This can be achieved when the bubble oscillating period is much shorter than the characteristic time of heat diffusion, $t_d = R_b^2/\alpha_g$, so that the gas distribution function depends only on the peculiar or thermal velocity (thermal equilibrium case). In this limit, we may obtain the gas temperature inside the bubble by taking the value of the gas conductivity as infinity in equation (17). That is $T_b = T_{bl} = T_{b0}(t)$, which validates the bubble dynamics formulation with the assumption of uniform gas temperature inside the bubble [14]. Heat transfer through the thermal boundary layer adjacent to the bubble wall only determines the heat exchange between bubble and medium in this case.

However, the temperature gradient inside bubble should exist, provided that thermal equilibrium does not follow mechanical equilibrium. This situation is realizable when the characteristic time of bubble evolution is much shorter than the relaxation time of vibrational motion, which is of the order of 10^{-6} s for high gas temperature. If a temperature gradient exists inside the bubble, the heat transfer through the bubble wall depends on the properties of the bubble and the thermal boundary layer medium. In this case, one may rewrite equation (5), the heat exchange between bubble and medium, with the help of equation (17). That is

$$\dot{Q} = -\frac{8\pi R_b^2 k_g (T_{b0} - T_\infty)}{\dfrac{R_b}{\left(1 + \dfrac{k_g}{k_l}\dfrac{\delta}{R_b}\right)}}. \quad (5')$$

As long as the value of $(k_g\delta)/(k_l R_b)$ is finite, there exists a temperature distribution inside the bubble (nonuniform temperature limit). For a very small value of $(k_g\delta)/(k_l R_b)$, the heat flow rate from the bubble is solely determined from the temperature gradient of the gas inside the bubble [7]. Or

$$\dot{Q} = -\frac{8\pi R_b^2 k_g (T_{b0} - T_\infty)}{R_b}. \quad (19)$$

(f) *Properties at the bubble center*

Now the only parameter undetermined is the bubble center temperature T_{b0}. At the bubble center, where the state equation for an ideal gas, $P_b = \rho_0 R_g T_{b0}$, holds, the energy equation without viscous dissipation, equation (12), becomes, with the help of equation (14)

$$\rho_0 C_{p,b} \frac{dT_{b0}}{dt} = \frac{\gamma}{\gamma - 1} \frac{dP_b}{dt} + \frac{3\gamma P_b}{\gamma - 1} \frac{\dot{R}_b}{R_b}. \quad (20)$$

After rearranging the above equation with the ideal gas law, we have

$$\frac{d\left\{\ln\left(\frac{P_b R_b^3}{T_{b0}}\right)\right\}}{dt} = 0$$

or

$$\frac{P_b R_b^3}{T_{b0}} = \text{constant} \quad (21)$$

which is another representation of the ideal gas relation. The time dependence of the temperature of the gas at the bubble center is readily obtained from equations (21) and (6) or (13):

$$\frac{dT_{b0}}{dt} = -\frac{3(\gamma - 1) T_{b0}}{R_b} \frac{dR_b}{dt} - \frac{6(\gamma - 1) k_l T_{b0} (T_{bl} - T_\infty)}{\delta R_b P_b}. \quad (22)$$

Now, all the gas properties at the bubble center can be calculated by using equations (13), (21) and (22). The above relations, equations (21) and (22), are valid for the whole region inside the bubble, provided that the temperature of gas is spatially uniform, as discussed in Section 2(b).

Note that the material derivative of temperature inside the bubble is invariant for spatially uniform density, or $DT_b/Dt = DT_{b0}/Dt$.

(g) *Bubble dynamics*

Compared to the vapor, the liquid is virtually incompressible because the bubble wall velocity concerned in this analysis is much smaller than the sound speed of liquid. With this assumption, the conservation of mass and momentum for the liquid results in the well-known Rayleigh equation governing the radial motion of a spherical bubble in an unbounded liquid [15]. That is

$$R_b \frac{du_b}{dt} + \frac{3}{2} u_b^2 = \frac{1}{\rho_\infty} (P_b - P_\infty). \quad (23)$$

Effects of surface tension and viscosity on the momentum equation have been neglected. This assumption is valid for the mm-size bubble considered in this analysis. Indeed, the pressure terms due to surface tension and viscosity in equation (23) are very much less than the internal pressure of the bubble, P_b, for such bubble dimensions.

The bubble wall velocity, u_b, is the time derivative of the bubble radius:

$$\frac{dR_b}{dt} = u_b. \quad (24)$$

(h) *Liquid energy equation*

The energy conservation equation for liquid under the influence of bubble wall motion is expressed by

$$\frac{\partial T}{\partial t} + u_r \frac{\partial T}{\partial r} = \frac{\alpha_l}{r^2} \frac{\partial\left(r^2 \frac{\partial T}{\partial r}\right)}{\partial r} \quad (25)$$

where $\alpha_l = k_l/\rho_\infty C_{p,\infty}$ is the thermal diffusivity, and the radial velocity of liquid due to bubble motion can be obtained from mass conservation for an incompressible liquid, $\nabla \cdot \mathbf{u}_r = 0$. That is

$$u_r = \left(\frac{R_b}{r}\right)^2 u_b.$$

Integrating equation (25) from $r = R_b$, to $r = R_b + \delta$, yields

$$\int_{R_b}^{R_b+\delta} r^2 \frac{\partial T}{\partial t} dr + \int_{R_b}^{R_b+\delta} u_r r^2 \frac{\partial T}{\partial r} dr = \int_{R_b}^{R_b+\delta} \alpha_l \frac{\partial\left(r^2 \frac{\partial T}{\partial r}\right)}{\partial r} dr. \quad (26)$$

With the temperature profile and boundary conditions given in equations (1) and (2), and the velocity profile in the liquid, the above equation becomes [16]

$$\left[1 + \frac{\delta}{R_b} + \frac{3}{10}\left(\frac{\delta}{R_b}\right)^2\right] \frac{d\delta}{dt} = \frac{6\alpha_l}{\delta} - \left[\frac{2\delta}{R_b} + \frac{1}{2}\left(\frac{\delta}{R_b}\right)^2\right] \frac{dR_b}{dt} - \delta\left[1 + \frac{\delta}{2R_b} + \frac{1}{10}\left(\frac{\delta}{R_b}\right)^2\right] \frac{1}{T_{bl} - T_\infty} \frac{dT_{bl}}{dt}. \quad (27)$$

Such an ordinary differential equation obtained from the integral one is very useful to compute the chaotic behavior of bubble [16] and sonoluminescence phenomena [17], which need enormous computing time.

Now we have six equations, (6) or (13), (17), (22)–(24) and (27) to solve six unknowns, P_b, T_{bl}, T_{b0}, u_b, R_b and δ.

(i) *Bubble pressure field and sound transimission*

During the nonlinear evolution, the bubble emits a pressure wave. The far field pressure signal from this evolving bubble at distance r_d from the bubble center can be written in terms of the volume acceleration of the bubble, $\ddot{V}$, where $\dot{V} = 4\pi R_b^2 \dot{R}_b$ [18]:

$$P'_{non}(t) = \frac{\rho_\infty \ddot{V}}{4\pi r_d} = \frac{\rho_\infty}{r_d} (2R_b \dot{R}_b^2 + R_b^2 \ddot{R}_b). \quad (28)$$

The damping mechanism of the pressure wave due to bubble pulsation in this case is mainly due to sound radiation and heat transfer between vapor in the bubble and the surrounding liquid, because damping due to viscous losses in the liquid is negligible.

If the bubble dimension is much less than the characteristic wavelength of bubble oscillation, the loss factor η_s due to sound radiation by the compact point source is given by [18]

$$\eta_s = \frac{1}{C_\infty}\sqrt{\frac{3\gamma P_\infty}{\rho_\infty}}. \tag{29}$$

Then the resultant expression for the acoustic pressure from bubble pulsation becomes approximately

$$P'_{non}(t) = \frac{\rho_\infty}{r_d}(2R_b\dot{R}_b^2 + R_b^2\ddot{R}_b)\exp\left(-\frac{\eta_s}{2}\omega t\right). \tag{30}$$

(j) *Bubble entropy production*

Using the thermal boundary layer with constant thickness at the bubble–liquid interface, the thermal damping due to finite heat exchange with the surrounding liquid was first treated by Moody [8]. Even though the damped bubble oscillation closely resembles damping of a mechanical system by friction, neither frictional dissipation nor energy lost in such a bubble–liquid system is present. The damped bubble oscillation due to finite heat transfer through the bubble wall simply displays an available power loss where there is entropy production. The entropy generation rate in such an oscillating bubble–liquid system is the combination of the rate change of entropy for vapor inside the bubble due to bubble pulsation and the net entropy flow out of the bubble as the result of heat exchange [19]. That is,

$$\dot{S}_g = \frac{DS_b}{Dt} - \frac{1}{T_\infty}\frac{dQ}{dt}. \tag{31}$$

Rewriting the above equation for the uniform temperature case, we have

$$\dot{S}_g = m_b C_p \frac{1}{T_{b0}}\frac{dT_{b0}}{dt} - m_b R_g \frac{1}{P_b}\frac{dP_b}{dt} + \frac{2k_l}{\delta}\left(\frac{T_{b0}}{T_\infty} - 1\right)$$

$$4\pi R_b^2 = 4\pi R_b^2 k_l \frac{(T_{b0} - T_\infty)}{\delta}\left(-\frac{1}{T_{b0}} + \frac{1}{T_\infty}\right) \tag{32}$$

which guarantees that the entropy generation is always positive.

3. INITIAL CONDITIONS FOR THE EVOLVING BUBBLE FORMED FROM THE DROPLET AT THE SUPERHEAT LIMIT

A fully evaporated droplet which maintains its saturated liquid volume may be considered as an assemblage of a tremendous number of critical clusters. The internal pressure of the agglomeration of the critical cluster or the fully evaporated droplet may be obtained from the vapor bubble formation model proposed by Kwak and Panton [20]. A crucial assumption of this modified bubble nucleation model based on molecular interaction is that the bubble formation process is initiated by the birth of a critical molecular cluster [2] rather than a critical bubble [4].

A stability condition for a cluster and the corresponding minimum free energy for the cluster can be obtained by assuming that the liquid has a face-centered cubic (FCC) lattice structure and that the London dispersion force is the only important interaction between molecules in their metastable state [20]. These are

$$-(P_\infty - P_v)n_c^{1/3} = \frac{z\varepsilon_m}{3}/V_m \tag{33-1}$$

$$F_{nc} = \frac{z\varepsilon_m}{6}n_c^{2/3} = \frac{1}{2}(P_v - P_\infty)V_m n_c. \tag{33-2}$$

If any cluster, an aggregate of the activated molecules in the metastable state, meets the condition (33-1), the liquid molecules in the cluster vaporize spontaneously by breaking the interaction between molecules. Thus, equation (33-1) can be regarded as the equation of vapor state of the critical cluster. The evaporated state, retaining the molecular volume of the saturated liquid state, V_m, is characterized by its very high pressure, P_n:

$$P_n = -(P_\infty - P_v)n_c^{1/3} = \frac{z\varepsilon_m}{3}/V_m. \tag{33-1'}$$

For example, the value of P_n is about 138 bar for butane at its supherheat limit. In the above equations, ε_m is the energy required to separate a pair of molecules from the given liquid state to the critical state. This is given approximately by [20]

$$\varepsilon_m = 4\varepsilon_0\left[1 - \left(\frac{\rho_c}{\rho_m}\right)^2\right]\left[\left(\frac{d_w}{d_m}\right)^6 - \left(\frac{d_w}{d_m}\right)^{12}\right]. \tag{34}$$

The average distance between molecules d_m in equation (34) and the effective molecular volume V_m in equation (33) can be found from the number density of liquid, N. The relation is

$$\frac{\pi}{6}d_m^3 N = V_m N = 0.7405 \tag{35}$$

where 0.7405 is the packing fraction of the FCC lattice structure. Certainly the equation of state for the vapor from the fully evaporated droplet, which is given in equation (33-1'), is not the one for an ideal gas.

Since the internal pressure of the fully evaporated droplet is very large, as noted before, it is expected that the droplet expands spontaneously. At the initial stage of this process, the bubble expands linearly with time. However its linear growing fashion slows down near the point where the nonlinear growing starts [1]. At the starting point of nonlinear growing, the bubble wall velocity vanishes. Thus, it is reasonable to choose this time as the starting point for calculation of non-

linear bubble evolution. The pressure wave signal from the bubble at this point is given by

$$P'_{non} = \frac{R_{non}}{r_d}[P_b(R_{non}) - P_\infty]. \quad (36)$$

The pressure inside the bubble at this time may be expressed by the droplet radius, R_d, and its internal pressure, P_n. Then equation (36) may be written as

$$P'_{non} = \frac{R_{non}}{r_d}\left[\left(\frac{R_d}{R_{non}}\right)^3 P_n - P_\infty\right]. \quad (36')$$

The far field pressure signal at this point is strongly dependent on the droplet volume, R_d^3, and its internal pressure of evaporated state, P_n. If one knows the magnitude of the far field pressure signal, P'_{non}, and the droplet radius, R_d, one can estimate the radius of the bubble and the pressure inside the bubble at this point, which may be used as initial conditions for studying subsequent nonlinear bubble evolution. The other possibility is that one may calculate the pressure inside the bubble if the values of far field pressure and the bubble radius at this point are provided. The temperature at the starting point of nonlinear growing was taken as just the superheat limit of liquid. In this study, we used the available experimental data obtained by Shepherd and Sturtevant [1] and Frost [21] to estimate the initial conditions needed for numerical calculations.

4. NUMERICAL CALCULATIONS

In numerical analysis, a normalization of the governing equations was carried out as follows. The radius and the boundary layer thickness are compared to the initial radius R_{non}, the temperature is related to the ambient temperature T_∞ and the velocity and pressure are related to constants, namely $u_0 = (P_\infty/\rho_\infty)^{1/2}$ and P_∞, respectively. For the other physical quantities such as time, thermal conductivity and thermal diffusivity, the normalization constants are obtained from the condition that the governing equations are invariant after the normalization procedure, which are given as:

time

$$t_0 = \frac{R_{non}}{u_0}$$

thermal conductivity

$$k_0 = \frac{P_\infty u_0 R_{non}}{T_\infty}$$

thermal diffusivity

$$\alpha_0 = u_0 R_{non}.$$

Using these normalized parameters, we may obtain the following nondimensional governing equations:

$$\frac{du}{dt} = \frac{1}{R}(P - 1 - \tfrac{3}{2}u^2) \quad (23')$$

$$\frac{dR}{dt} = u \quad (24')$$

$$\frac{dP}{dt} = -\frac{3\gamma}{R}Pu - \frac{6(\gamma-1)k_l(T_{bl}-1)}{\delta R} \quad (6') \text{ or } (13')$$

$$T_{bl} = \frac{1+\zeta T_{b0}}{1+\zeta} \quad (17')$$

$$\frac{dT_{b0}}{dt} = -\frac{3(\gamma-1)T_{b0}u}{R} - \frac{6(\gamma-1)k_l T_{b0}(T_{bl}-1)}{\delta RP} \quad (22')$$

$$\frac{d\delta}{dt} = \frac{1}{(1+\beta+0.3\beta^2)}\left[\frac{6\alpha}{\delta} - 2\beta u(1+0.25\beta) - \delta(1+0.5\beta+0.1\beta^2)\frac{1}{T_{bl}-1}\frac{dT_{bl}}{dt}\right] \quad (27')$$

and

$$\frac{dT_{bl}}{dt} = \frac{(T_{bl}-1)\left(\frac{\dot\delta}{\delta} - \frac{\dot R}{R}\right) + \zeta\frac{dT_{b0}}{dt}}{1+\zeta}$$

where

$$\zeta = \frac{k_g}{k_l}\frac{\delta}{R} \quad \text{and} \quad \beta = \frac{\delta}{R}.$$

With known values of P, T_{b0}, T_{bl} and R, which are state variables of the vapor inside the bubble at the starting point of nonlinear bubble evolution, initial bubble wall velocity and approximate value of δ, we solved the foregoing six equations by using the Runge–Kutta numerical method to obtain the next time step values of u, R, P, T_{b0}, T_{bl} and δ. This calculation was repeated up to the desired time step. In this calculation, we let $d\delta/dt = 0$ to avoid an infinite value of the boundary layer thickness, provided that the absolute value of $(T_{bl}-1)$ is less than 0.0005, which corresponds to $|T_{bl} - T_\infty| < 0.21°C$. The uniform temperature distribution case can be obtained by replacing T_{bl} with T_{b0}.

5. CALCULATION RESULTS AND DISCUSSIONS

Bubble dynamics formulated in this study contain a general thermal behavior for the gas inside the bubble in oscillation. For numerical calculations, we used the 'uniform temperature limit', which is appropriate for the bubble evolution investigated in this study, since the temperature of the vapor inside the bubble is so low that vibrational motion of the vapor molecules is not excited and the characteristic time of bubble evolution (ms) is much longer than the relaxation time of translational motion of vapor molecules.

The calculated pressure wave signal from the evolving butane bubble in ethylene glycol at the ambient pressure of 1.013 bar and at a temperature of 378 K is

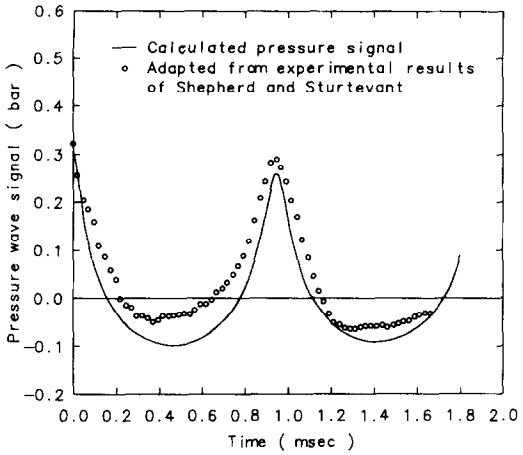

Fig. 2. Pressure wave signal from the oscillating butane bubble in ethylene glycol at 1.013 bar. The initial conditions are $R_b = 1.37$ mm, $P_b = 6.67$ bar, $T_b = 378$ K.

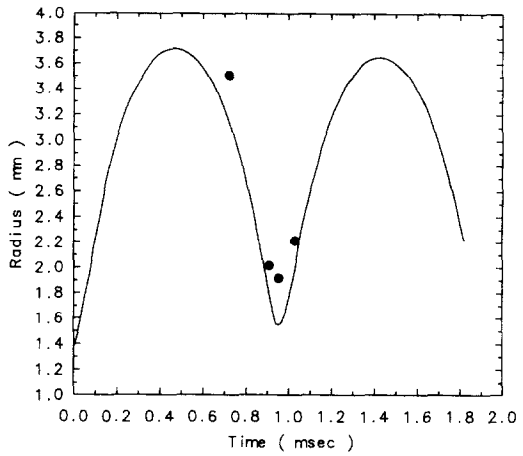

Fig. 3. Radius–time curve for the butane bubble with same initial conditions as used Fig. 2. The solid circles represent the values observed from the different sequences of similar bubbles.

shown in Fig. 2, together with observed data [1]. The radius of the bubble and the pressure inside the bubble at the starting point of the calculation are 1.37 mm and 6.67 bar, respectively. Using these values, which were chosen to produce the same value of observed pressure wave signal at the starting point, the droplet radius estimated from equation (34) is about 0.50 mm. The agreement between calculation and observation is exceptionally good, which supports the validity of the bubble dynamics model presented in this paper.

The calculated value of the loss factor due to thermal and sound radiation damping combined is about 0.053, while the loss factor due to sound radiation is only 0.011. So the thermal damping turns out to be the more important factor for controling the oscillation for such a size of bubble. The loss factor due to thermal and sound radiation damping may be obtained from the relation to the dissipation coefficient, χ, by $\eta = 2\chi/\omega$ [18]. The loss factor was calculated from the following relation:

$$\chi = \ln(P'_1/P'_2)/2\pi/\omega \tag{37}$$

where P'_1 and P'_2 are the peak values in the far field pressure signal observed during one oscillation period. The theoretical radius–time curve, along with the approximate bubble diameters at several points, which was measured from the different sequences of similar bubbles [1], is also shown in Fig. 3. A reasonable agreement between calculated values and observed ones can also seen in this figure.

Figure 4 shows the time rate change of gas temperature during bubble evolution with the same initial conditions as used in Fig. 2. As expected, the process of bubble evolution is neither isothermal nor adiabatic. As can be seen in Fig. 4, the lowest temperature experienced by the vapor during bubble oscillation is about 86°C, which eliminates the possibility of condensation of vapor inside the bubble, as assumed. As shown in Fig. 5, the entropy generation during this

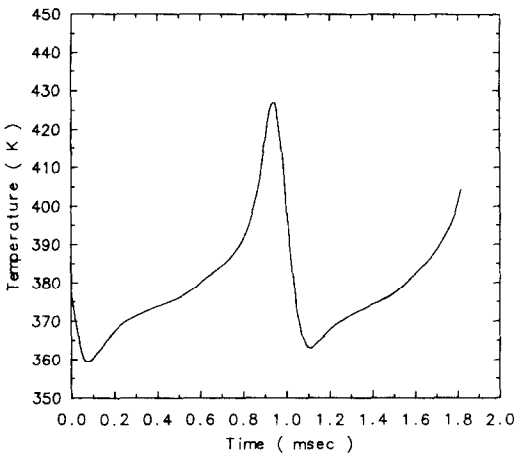

Fig. 4. Time dependence of the temperature inside bubble with same initial conditions as used in Fig. 2.

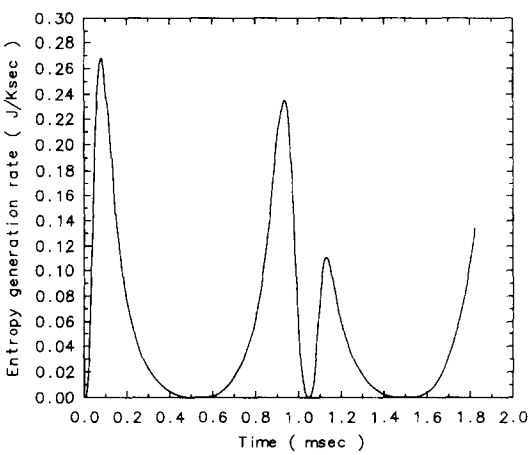

Fig. 5. Time dependence of the entropy generation rate of the oscillating butane bubble with same initial conditions as used in Fig. 2.

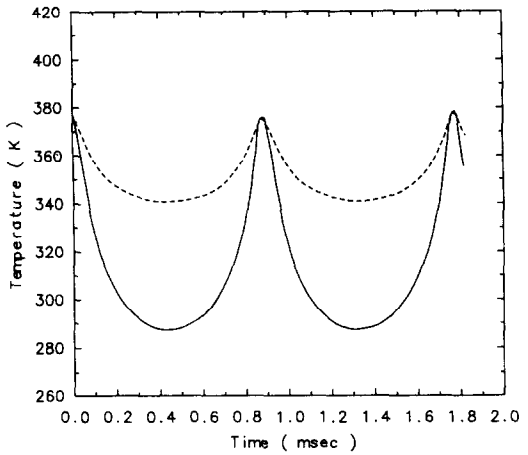

Fig. 6. Time dependence of the temperature at the bubble center (—) and the average temperature (···) of the oscillating butane bubble with nonuniform temperature limit.

bubble oscillation turns out to be always positive, as expected.

For the case that the temperature of the gas inside the bubble is nonuniform (nonuniform temperature limit), the bubble has a drastically different temperature excursion from the one obtained from the uniform temperature limit, as shown in Fig. 6. The change of gas temperature at the bubble center behaves almost adiabatically. In this case, the loss factor due to thermal and sound radiation damping is about 0.013, which shows that thermal damping effect on bubble oscillation is negligible. This is because the temperature difference between the bubble wall and the liquid medium is small. Consequently less heat exchange between the bubble and liquid occurs.

Another calculated pressure trace obtained from the oscillating ether bubble in glycerin at 3 bar with uniform temperature assumption is shown in Fig. 7. The initial conditions used were 2.45 mm for bubble radius and 5.2 bar for the corresponding bubble pressure. The bubble oscillation pattern, determined from the bubble's maximum radius and difference in pressure between the vapor in the bubble and the ambient liquid [22], is quite different from the case of ambient pressure of 1 bar. The oscillating behavior at 3 bar is similar to the typical simple harmonic motion with damping rather than impulsive oscillation with greater amplitude, which occurs in the case of 1 bar. The calculated loss factor during the second period is about 0.04, which is much smaller than the experimental observation, 0.135 [21]. However this figure clearly shows the thermal damping effect on bubble oscillation. An adiabatic and isothermal bubble yields undamped bubble oscillation [9]. A high value of the loss factor observed [21] may be due to catastrophic deformation of the bubble surface, which enhances the heat transfer area. In this case, the calculated oscillation period of 0.64 ms is in good agreement with the observed value of 0.67 ms. This may be due to the fact that the pressure oscillation frequency remains constant even after the bubble break-up [21]. The value of the specific heat ratio used for ether was calculated from the specific heat data in Weast [23].

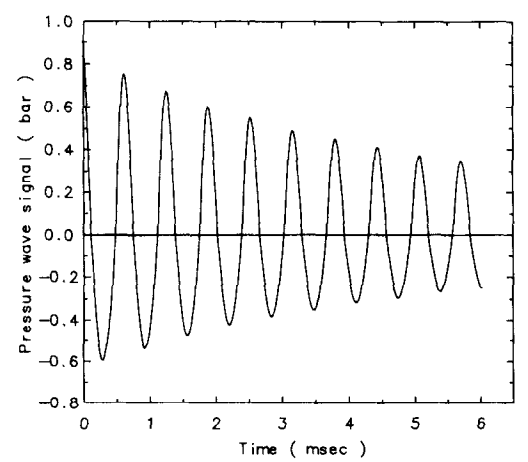

Fig. 7. Calculated pressure wave signal from the oscillating ether bubble in glycerin at 3 bar with initial conditions, $R_b = 2.45$ mm, $P_b = 5.2$ bar, $T_b = 420.0$ K ($r_d = 6$ mm).

At high ambient pressure, the pressure difference between the vapor inside the bubble and the ambient liquid, which may be considered as the driving force for nonlinear bubble growth, decreases substantially. With a small driving force, impulsive growth does not happen and the bubble growth is followed by a slight oscillating motion, approaching mechanical equilibrium. For high ambient pressure, the driving force virtually vanishes, which guarantees equilibrium phase transition [24].

With the bubble dynamics model with nonuniform temperature distribution, which is valid for the case where the bubble compression time is much shorter than the relaxation time of vibrational motion of molecules, one may predict the chaotic behavior of the bubble under an ultrasonic field [16] and sonoluminescence phenomena from the oscillating bubble in the ultrasonic field [17].

6. CONCLUSIONS

A general model for bubble dynamics has been formulated and applied to study the nonlinear evolution of vapor bubble formed from the evaporated droplet at the superheat limit. A distinct feature of this model is that the velocity and temperature profiles for the gas inside the bubble have been obtained analytically. With this model, the polytropic approximation is no longer required to calculate the internal pressure of the evolving bubble. With uniform density and temperature approximation, it has been found that the thermal damping due to finite heat exchange between vapor inside the bubble and the surrounding liquid plays a crucial role in the oscillation of the bubble, and that the bubble evolution process is neither isothermal nor adiabatic. Also the calculated values of the far field pressure signal from the evolving bubble are in good agreement with experimental results.

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