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A simple universal equation for bubble growth in pure liquids and binary solutions with a non-volatile solute

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Abstract—A simple equation suitable for predicting the growth rate of a vapor bubble in uniformly-superheated pure liquids and in binary solutions with a non-volatile solute was developed. The equation also improves on the popular pure-liquid bubble growth expression of Mikić *et al.* (*International Journal of Heat and Mass Transfer*, 1992, **35**, 1711–1722) in that it is valid throughout the bubble growth history, i.e. in the surface-tension-, inertia-, and heat-transfer-controlled regimes, it accounts for bubble growth acceleration effects, and uses correctly-related and variable fluid properties. It was found to agree very well with experimental data for pure water and for aqueous NaCl solutions. As the bubble growth in superheated solutions with a non-volatile solute was found to be quite insensitive to diffusion and nonequilibrium effects in a broad range of common solution properties, this equation is likely to be universally valid for many liquids and solutions. Copyright © 1996 Elsevier Science Ltd.

1. INTRODUCTION

Bubble growth in superheated fluids is of key interest in boiling phenomena in general and in flash evaporation in particular. Most of the large amount of research on such bubble growth has been conducted for *pure* liquids (see reviews in refs. [1–3]), and very little is therefore known about bubble growth in superheated solutions with a non-volatile solute, a topic of both fundamental and practical importance, with applications including a wide variety of separation processes such as water desalination, and energy conversion processes such as ocean-thermal energy conversion, geothermal power generation, and nuclear reactor safety.

Amongst the small number of papers on bubble growth in superheated binary solutions, Scriven [4] (with corrections [5, 6]) has described the general approach to modeling uniformly-heated spherically-symmetric bubble growth of both pure liquids and binary mixtures, and has derived approximate asymptotic solutions in the heat and mass-transfer-controlled regime; subsequent studies are cited in refs. [7–12].

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The past research shows that bubble growth in superheated liquids can be characterized as progressing in three consecutive regimes (as depicted in Fig. 1): at first, just when the bubble has nucleated (with radius just larger than the critical), surface tension is dominant, impeding significant growth for a certain "delay period". After the nucleus grew somewhat, say doubled its diameter, inertia forces become dominant and the bubble grows primarily due to the difference between the vapor pressure inside the bubble (p_v) and the exterior pressure (p_∞). During that period bubble growth is a linear function of time, $R \sim t$. As the bubble grows further and its wall temperature consequently drops, causing an increased temperature difference between the surrounding liquid and the bubble wall, its growth rate becomes dominated by heat transfer from the surrounding liquid which causes addition of vapor to the bubble by evaporation at the interface. During that period bubble growth is characterized by $R \sim t^{1/2}$.

The primary objective of this paper is to introduce a simple yet rather accurate universal equation for bubble growth rates in *either* pure, *or* binary solution liquids with a non-volatile solute, which is valid throughout all of the bubble growth regimes.

Mikić *et al.* [13] have developed a simple general

NOMENCLATURE

A^+	parameter defined by equation (3) [m s^{-1}]
A^*	parameter defined by equation (10) [m s^{-1}]
B^+	parameter defined by equation (4) [m s$^{-1/2}$]
B^*	parameter defined by equation (11) [m s$^{-1/2}$]
c_l	specific heat of pure liquid or of solvent [J kg^{-1} K^{-1}]
D	mass diffusivity [m^2 s^{-1}]
h_{fg}	latent heat of vaporization [J kg^{-1}]
P	vapor pressure of pure liquid or of solution [Pa]
p_v	vapor pressure inside the bubble [Pa]
p_∞	pressure in pure liquid or solution far from the bubble [Pa]
R	bubble radius [m]
R_c	critical bubble radius expressed by equation (13) [m]
R^+	dimensionless bubble radius defined by equation (2), dimensionless
R^*	dimensionless bubble radius defined by equation (9), dimensionless
$\bar{R}$	gas constant [J kg^{-1} K^{-1}]
t	time [s]
t_d	bubble growth delay period, defined as the time at the intersection of $R = R_c$ and the tangent to the R vs t relation curve at the point $d^2R/dt^2 = 0$ [s]
t_u	upper limit of time during the period concerned [s]
t^+	dimensionless time defined by equation (5), dimensionless
t^*	dimensionless time defined by equation (12), dimensionless
t_r^*	dimensionless time for estimating T_r, dimensionless
T	temperature [°C]
$\hat{T}$	absolute temperature [K]
T_e	equilibrium temperature of solution corresponding to p_∞ [°C]
T_i	bubble wall temperature [°C]
T_r	reference temperature at which ρ_v is evaluated [°C]
T_s	saturation temperature [°C]
$\hat{T}_i$	bubble wall absolute temperature [K]
$\hat{T}_s$	saturation absolute temperature [K]
T_∞	temperature of pure liquid or of solution far from the bubble.

Greek symbols

α_l	thermal diffusivity of pure liquid or of solvent [m^2 s^{-1}]
ΔP_0	initial pressure difference between interior and exterior of the bubble, expressed by equation (17) for pure liquid and by equation (22) for a solution [Pa]
ΔT_s	superheat defined by equation (7) for pure liquid and by equation (20) for a solution [°C], [K]
ρ_l	density of pure liquid or of solvent [kg m^{-3}]
ρ_v	density of pure liquid vapor or of solvent vapor [kg m^{-3}]
σ	surface tension [N m^{-1}]
σ_e	evaporation coefficient, dimensionless
ω_∞	mass fraction of solute in solution far from the bubble, dimensionless.

equation for calculating bubble growth rates in *pure* liquids, starting with a bubble radius of zero, only in the inertia- and heat-transfer-controlled regimes, viz.

$$R^+ = \tfrac{2}{3}[(t^+ + 1)^{3/2} - (t^+)^{3/2} - 1] \quad (1)$$

where

$$R^+ = \frac{A^+}{(B^+)^2} R \quad (2)$$

$$A^+ = \left(\frac{2}{3} \frac{h_{fg}\rho_v \Delta T_s}{\rho_l \hat{T}_s}\right)^{1/2} \quad (3)$$

$$B^+ = \left(\frac{12}{\pi}\alpha_l\right)^{1/2} \frac{c_l \rho_l \Delta T_s}{h_{fg}\rho_v} \quad (4)$$

and

$$t^+ = \left(\frac{A^+}{B^+}\right)^2 t. \quad (5)$$

The properties of the vapor and liquid in the above equations are based on the saturation temperature (T_s) of the liquid, which corresponds to the pressure (p_∞) in the liquid far from the bubble, viz.

$$p_\infty = (P)_{T_s} \quad (6)$$

and the superheat (ΔT_s) is defined as

$$\Delta T_s = T_\infty - T_s. \quad (7)$$

In the development of this bubble-growth, Mikić *et al.* [13] have assumed that the relationship between the vapor pressure and temperature can be expressed by the linearized Clausius–Clapeyron equation, and

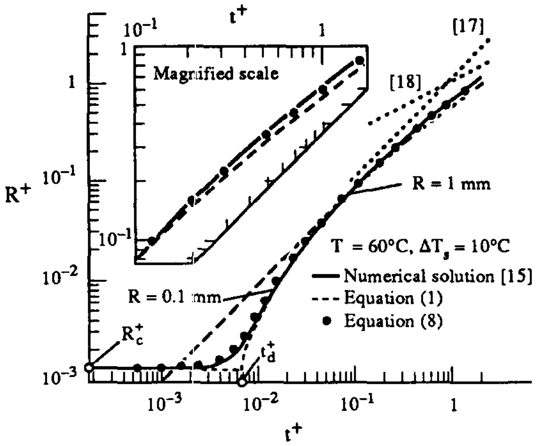

Fig. 1. Bubble growth history in water and comparison of the pure-liquid simple bubble growth equation (● symbols) and numerical solution (solid line) of Miyatake and Tanaka [15] with the equation by Mikić *et al.* [13] (dashed line) and the limiting-case equations (dotted lines) for the inertia-controlled regime (Rayleigh [17]) and the heat-transfer-controlled regime (Plesset and Zwick [18]). The dimensionless bubble radius R^+ and time t^+ are defined by equations (2) and (5), respectively. The bubble growth delay time (t_d^+) here is at the intersection of the tangent at the solution inflection point $(d^2R^+/(dt^+)^2 = 0)$ with the critical radius ($R^+ = R_c^+$).

that the vapor density is constant. Theofanous and Patel [14] have shown that these assumptions may lead to large errors for large initial superheats, when the vapor density changes during the process significantly, and have modified the Mikić *et al.* [13] bubble growth equation to correct this deficiency by using a more realistic dependence of vapor density on temperature.

2. AN IMPROVED BUBBLE GROWTH EQUATION FOR PURE LIQUIDS

In an experimentally-validated numerical study, Miyatake and Tanaka [15, 16] have developed an improved simple equation for bubble growth in pure liquids, which has more generality and reflects reality more closely, by including the following effects:

(1) The initial, surface-tension-controlled bubble growth regime, which occurs immediately after the nucleation of a bubble, and which causes an initial lag in bubble growth (the 'delay period', t_d, see refs. [1]–[4] and Fig. 1 where $t_d^+ = (A^+/B^+)^2 t_d$) was added to the inertia- and heat-transfer-controlled regimes taken into account in the equation by Mikić *et al.* [13]. Consequently, the new equation now covers the entire bubble life span.

(2) Consistently with improvement 1 above, growth was considered to start when the bubble radius was just larger than the critical radius R_c (at which the bubble nucleus is sustained as a result of equilibrium between surface tension and the pressure difference across the bubble wall), specifically here at $R(0) = 1.0001R_c$. In ref. [13] bubble growth was considered to start from $R(0) = 0$, which is a physical impossibility.

(3) The correct, non-linear relationship between the vapor pressure and temperature, obtained from the steam tables, was used, eliminating the linear relationship assumption used in ref. [13].

(4) The effect of the bubble growth acceleration term d^2R/dt^2, neglected in ref. [13], was included.

The new general equation for bubble growth *in pure liquids* [15], between a dimensionless bubble radius (R^*) and dimensionless time (t^*), which was shown in ref. [16] to represent experimental data very well, is

$$R^* = \frac{2}{3}\left\{1+\frac{t^*}{3}\exp\left[-(t^*+1)^{1/2}\right]\right\} \times \left[(t^*+1)^{3/2}-(t^*)^{3/2}-1\right] \quad (8)$$

where

$$R^* = \frac{A^*}{(B^*)^2}(R-R_c) \quad (9)$$

$$A^* = \left[\frac{2}{3}\frac{\Delta P_0}{(\rho_l)_{T_\infty}}\right]^{1/2} \quad (10)$$

$$B^* = \left(\frac{12}{\pi}\right)^{1/2}\left[\frac{\Delta T_s}{(\rho_v)_{T_r}}\right]\left(\frac{\alpha_l^{1/2}c_l\rho_l}{h_{fg}}\right)_{T_s} \quad (11)$$

and

$$t^* = \left(\frac{A^*}{B^*}\right)^2\left\{t-t_d\left[1-\exp\left[-\left(\frac{t}{t_d}\right)^2\right]\right]\right\} \quad (12)$$

where

$$R_c = 2(\sigma)_{T_\infty}/\Delta P_0 \quad (13)$$

and

$$t_d = 6R_c/A^*. \quad (14)$$

In the above equations the properties are based on T_∞, T_s and T_r of the liquid. T_r is a reference temperature at which the temperature-sensitive saturation density (ρ_v) of the vapor is evaluated, and is defined as

$$T_r = T_s+(T_\infty-T_s)\{1-2(t_r^*)^{1/2}[(t_r^*+1)^{1/2}-(t_r^*)^{1/2}]\} \quad (15)$$

where

$$t_r^* = (1/2)(A^*/B^*)^2(t_u-t_d) \quad (16)$$

and where t_u is the upper limit of the time period during which the bubble growth is investigated by these equations.

The initial pressure difference (ΔP_0) between the bubble interior and exterior is expressed by

$$\Delta P_0 = (P)_{T_\infty} - p_\infty \quad (17)$$

in which P is the vapor pressure of the liquid, and the subscript is indicating the temperature T_∞ at which the vapor pressure is evaluated.

Examination of the bubble growth equations by Mikić *et al.*, equations (1)–(5), show that $R_{t\to 0} = A^+ t$, i.e. A^+ is the dominant coefficient in the inertia-controlled regime, and $R_{t\to\infty} = B^+ t^{1/2}$, i.e. B^+ is the dominant coefficient in the heat-transfer-controlled regime. The same is true for the equation of Miyatake and Tanaka, equations (8)–(12), where the corresponding coefficients A^* which depends on ΔP_0, and B^* which depends on ΔT_s, play the same roles in these regimes.

The comparison of the simple general bubble growth equation of Miyatake and Tanaka {[15], equations (8)–(12)} with the equation by Mikić *et al.* [13], with the experimentally-validated numerical solution of Miyatake and Tanaka [15, 16], and with the Rayleigh [17] and Plesset and Zwick [18] equations which represent bubble growth in the limiting cases of the inertia- and heat-transfer-controlled regimes, respectively, is shown in Fig. 1 [15]. The agreement between the numerical solution and this equation is excellent, and the capability of the equation to predict bubble growth from its inception at the critical radius and through the surface-tension-, inertia-, and heat-transfer-controlled regimes is clearly demonstrated. The equation by Mikić *et al.* [13] over-predicts the radius in the early stage (by up to about three-fold for the case shown in Fig. 1) because it does not consider the delay period and its effects on subsequent growth; and increasingly under-predicts it (up to about 15% for the case and range of growth time shown in Fig. 1) due to the omission of the acceleration term.

All of these simplified bubble growth equations [13, 14] have been developed assuming equilibrium at the evaporating interface, i.e.

$$p_{v(t)} = (P)_{T_i(t)}. \tag{18}$$

Real evaporation is, however, a non-equilibrium process, characterized by an equation such as [19]

$$p_v(t) = (P)_{T_i(t)} - \left(\frac{2-\sigma_e}{2\sigma_e}\right)\frac{\mathrm{d}R(t)}{\mathrm{d}t}\,\rho_v(t)[2\pi\bar{R}\hat{T}_i(t)]^{1/2} \tag{19}$$

where σ_e is the evaporation coefficient and $\bar{R}$ is the gas constant. The evaporation coefficient has a value between 0 and 1.0, depending on the liquid and its purity, and is especially sensitive to the presence of surfactants on the evaporating interface. Increasing values of σ_e imply higher vapor pressures and closer approach to equilibrium, and lead to higher evaporation and bubble growth rates. The studies by Miyatake and Tanaka [15, 16] have shown that bubble growth rates decrease, as expected, with a decrease in σ_e, but that this decrease is negligible for $0.5 < \sigma_e < 1.0$, the range currently believed to be pertaining to evaporation of water, and becomes significant only for approximately $\sigma_e \leqslant 0.1$.

3. DERIVATION OF A SIMPLIFIED EQUATION FOR THE RATE OF BUBBLE GROWTH IN A BINARY SOLUTION CONTAINING A NON-VOLATILE SOLUTE

In contrast to the above-discussed bubble growth in *pure liquids*, bubble growth in uniformly superheated *binary solutions* with a non-volatile solute is determined not only by the temperature (T_∞) and pressure (p_∞) of the solution, but also by the mass fraction (ω_∞) of the solute. It was found in the authors' previous study [20] that the concentration (ω_∞) has a significant effect on the bubble growth rate when the far-field solution pressure (p_∞) is held constant. The bubble growth rates were observed to decrease with increasing concentration in this case because of the consequent boiling-point elevation, i.e. the reduction of the vapor pressure inside the bubble and the increase of the equilibrium temperature. This effect of the concentration was much larger for higher p_∞, where a change in concentration larger at a given T_∞ creates larger differences between (a) the magnitudes of the bubble-growth driving forces ($p_v - p_\infty$) dominating the inertia-controlled regime, and (b) the magnitudes of the bubble-growth driving forces ($T_\infty - T_i$) representing the actual driving force $(\partial T/\partial r)_{\text{bubble wall}}$ dominating the heat-transfer-controlled regime.

As shown in Fig. 2(a), the effect of concentration became, however, very small when ΔT_s was the parameter held constant, especially in the heat-transfer-controlled bubble growth regime, or, as shown in Fig. 2(b), when ΔP_0 was held constant, especially in the inertia-controlled regime. This is consistent with experiments, as an examination of the bubble-growth driving force ($p_v - p_\infty$) dominating the inertia-controlled regime, and the driving force ($T_\infty - T_i$) dominating the heat-transfer-controlled regime, can demonstrate. When p_∞ is fixed (at constant T_∞) both driving forces increase when ω_∞ is lower, the first one due to an increase in p_v and the second one due to the decrease in the equilibrium temperature (which is also the temperature T_i).

If the superheat (ΔT_s) defined as

$$\Delta T_s = T_\infty - T_e \tag{20}$$

where T_e is the equilibrium temperature satisfying the relation

$$p_\infty = (P)_{T_e,\omega_\infty} \tag{21}$$

in which P is the vapor pressure of the solution, and the subscripts are indicating the temperature T and mass fraction ω at which the vapor pressure is evaluated, is fixed, then both p_∞ and p_v are inversely proportional to ω_∞. The dominant bubble-growth driving force ($p_v - p_\infty$) in the inertia-controlled regime consequently becomes almost constant, affected only by the relatively-weak dependence of $p_v(T)$ on ω_∞ for the conditions examined in this study.

The dominant driving force ($T_\infty - T_i$) in the heat-transfer-controlled regime also remains almost con-

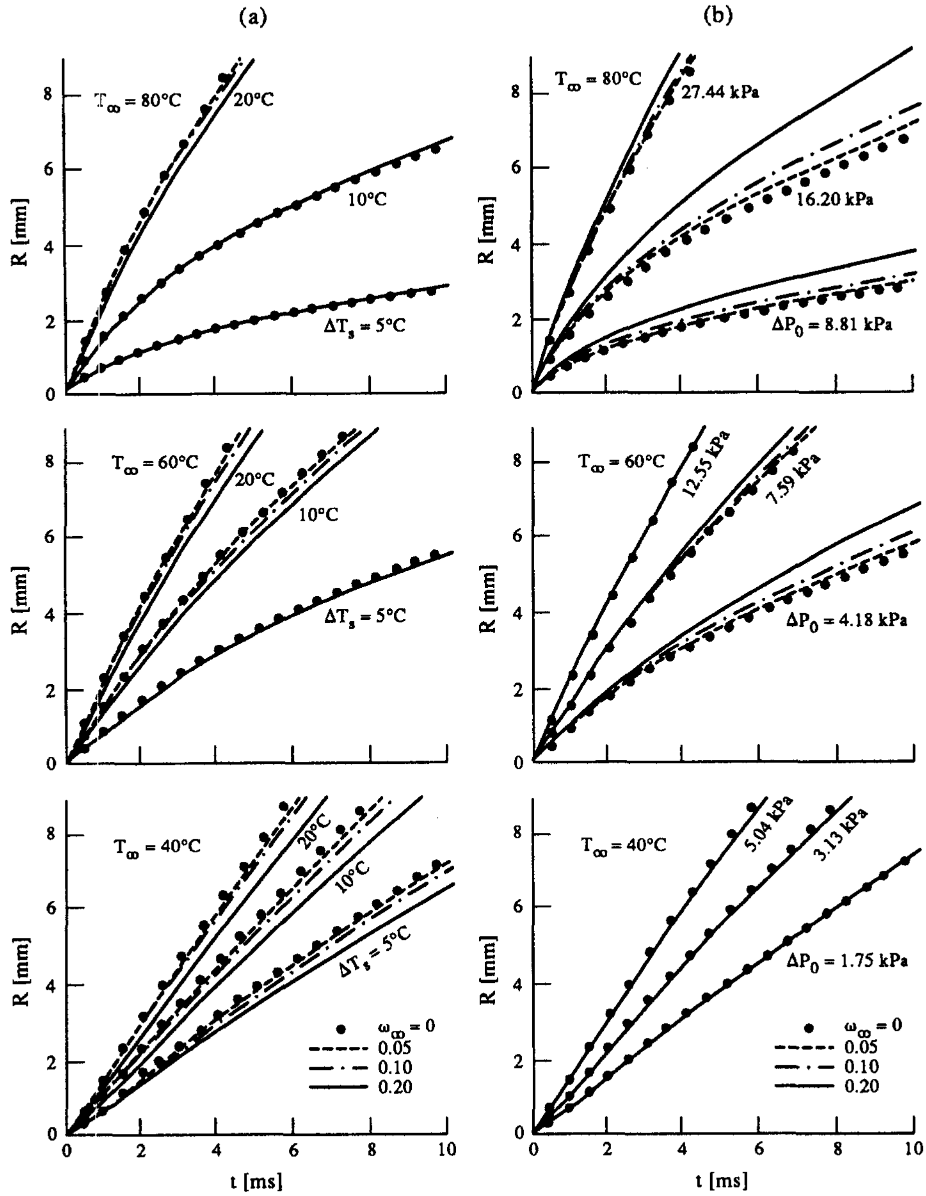

Fig. 2. The bubble growth histories in aqueous NaCl solutions at four solute mass fractions (ω_∞), at three given solution temperatures (T_∞), from ref. [20], for three fixed values of (a) the superheat (ΔT_s); (b) the initial pressure difference (ΔP_0).

stant because T_i can vary only within the fixed range from T_∞ to T_e, and consequently the effect of ω_∞ becomes practically negligible for a specified ΔT_s in the heat-transfer-controlled regime.

If ΔP_0 is specified, defined as

$$\Delta P_0 = (P)_{T_\infty,\omega_\infty} - p_\infty \tag{22}$$

ω_∞ has, by definition, no effect on bubble growth in the inertia-controlled regime.

Since diffusion is present in binary solutions and not in pure liquids, the effect of the mass diffusivity was examined to determine the importance of its inclusion in the bubble growth equation. Evaporation at the bubble interface raises the solute concentration there, and thus causes its diffusion away from the interface into the solution. The diffusion has counteracting effects in bubble growth. Decreasing the mass diffusivity reduces the migration of solute from the bubble interface, consequently increasing its concentration and the boiling point elevation there. This, in turn, results in: (1) a decrease of p_v and increase of T_e, with consequent decreases in the bubble-growth driving forces ΔP_0 and ΔT_s, respectively, and an overall tendency to diminish R; (2) an increase of the superheat of the generated vapor with consequent decrease of ρ_v, which tends to increase R. Large mass diffusivity obviously has a small effect on bubble growth.

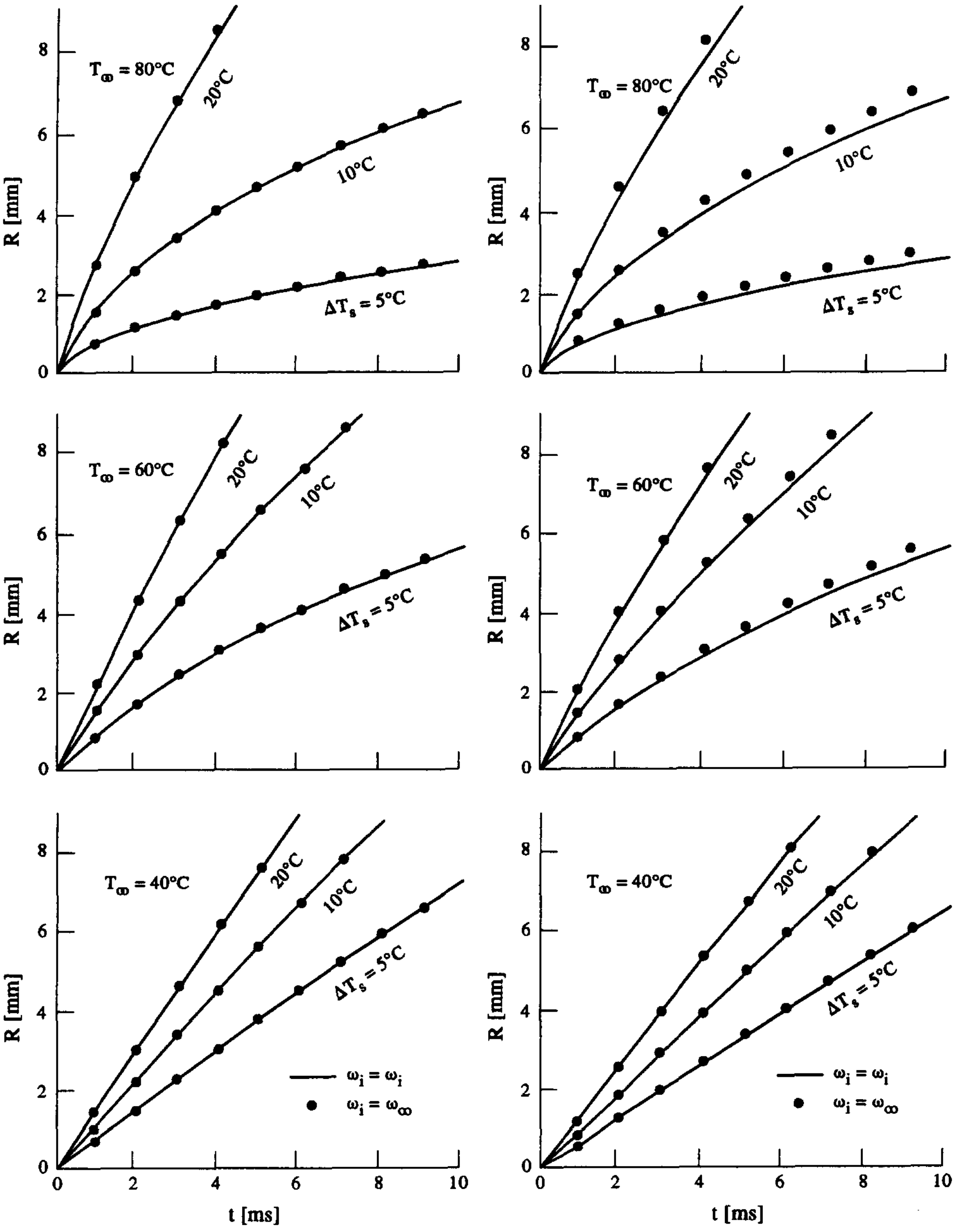

Fig. 3. Examination of the effects of the solute concentration at the bubble wall on bubble growth, at several values of T_∞ and ΔT_s. The solid lines represent the solutions under physically-correct formulation for the bubble-wall concentration, $\omega_i = \omega_i$. The symbols ● correspond to the simplifying assumption $\omega_i = \omega_\infty$, where it is assumed that the concentration at the bubble wall does not change during evaporation and remains constant at ω_∞.

Using the authors' experimentally validated numerical model for bubble growth in binary solutions [20], computations of bubble growth were made throughout the range of T_∞ and ΔT_s for two limiting cases of the interfacial concentration of NaCl: one being real ($\omega_i = \omega_i$), and the other where it is assumed to equal the far-field concentration ($\omega_i = \omega_\infty$). As seen in Fig. 3, the results are indistinguishable for lower concentrations and start differing somewhat when the concentration rises to $\omega_\infty = 0.20$ at temperatures above 60°C. This indicates that NaCl diffusion has negligible effect on bubble growth in the examined range of variables because the above-described counter-acting effects cancel each other.

Based on the above conclusions, it seems reasonable to assume that the bubble growth equation for a superheated pure liquid, equation (8)–(12), may thus also be applicable for a superheated solution containing a non-volatile solute, if the superheat ΔT_s defined by equation (7) is replaced by that defined by equation (20), the initial pressure difference ΔP_0 between the bubble interior and exterior defined by

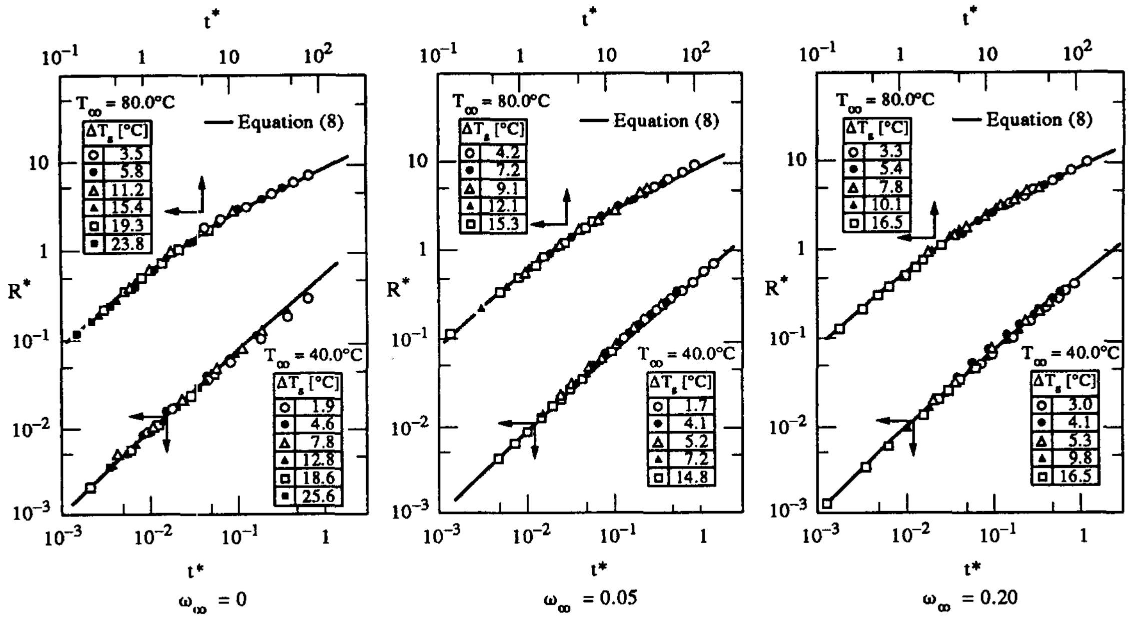

Fig. 4. Comparison of the bubble growth equations (8)–(12), with the authors' experimental data [20], for bubble growth in aqueous solutions of NaCl.

equation (17) is replaced by that defined by equation (22) and the physical properties of the liquid are taken as those of the solvent. Shown in Fig. 4, the simple general bubble growth equation defined by equations (8)–(12) was thus compared with experimental results obtained from the growth-measurement of bubbles in uniformly superheated aqueous NaCl solutions as described in the authors' previous paper [20]. The agreement between the predictions of the simple general bubble growth equation and the experimental results is seen to be very good.

The equation was developed for any binary solution with a non-volatile solute; the only consideration specific to the experimentally-examined aqueous NaCl solution was the neglection of diffusion effects as discussed above and shown in Fig. 3. The applicability of the equation to other solutions was examined by noting that the mass diffusivity D of 34 common inorganic solutes in water are close to that of NaCl, D_{NaCl}, viz. $0.5 < D/D_{NaCl} < 2.1$. A numerical study conducted to examine the effect of the magnitude of D on bubble radius has shown that the deviation is within 4% even for the low diffusivity value of $D/D_{NaCl} = 1/3$. This result extends the validity of the equation to many other aqueous inorganic solutions.

4. CONCLUSIONS

(1) A simple universal equation suitable for predicting the growth rate of a vapor bubbles in uniformly-superheated pure liquids and in binary solutions with a non-volatile solute was developed.

(2) The equation is valid throughout the bubble growth history, i.e. in the surface-tension-, inertia-, and heat-transfer-controlled regimes, it represents reality significantly better than the earlier equation of Mikić *et al.* [13], and was found to agree well with experimental data for pure water and aqueous NaCl solutions.

(3) As the bubble growth in superheated solutions with a non-volatile solute was found to be quite insensitive to diffusion and non-equilibrium effects in a broad range of common solution properties, this equation is likely to be universally valid for many liquids and solutions.

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