

BUBBLE NUCLEATION, PRELIMINARY CAVITATION EXPERIMENTS IN WATER AND MASS TRANSPORT MODELLING IN THE RAYLEIGH-PLESSET EQUATION

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***Summary:** The paper presents theoretical and preliminary experimental results attempting to validate the modified nucleation theory for mixtures (published by Maršík et al. (2002)) for the case of cavitation. The non-equilibrium (dissipative) processes involved in the origination of a nucleus are expressed using an experimentally obtained coefficient. The value of the coefficient is estimated from the cavitation experiment in a convergent-divergent nozzle. It is assumed that the energy of thermodynamic fluctuations near the breaking tension is comparable to the minimum work of formation of a bubble. In addition, the effect of mass transport across the bubble wall is discussed in numerical experiments using the previously published modified Rayleigh-Plesset equation.*

1 Introduction

Cavitation is one of the critical engineering challenges for hydrodynamic machinery designers. Currently, the problem of prediction of cavitation regions in the liquid flow often relies on the “heterogeneous” view of the problem, i.e. it is assumed that a given (usually experimentally obtained or empirical) population of nuclei is convected to the region of low pressure within the flow where the nuclei become activated and cavitate. Bubble nucleation is disregarded assuming that pre-existing microbubbles or other particle or chemical heterogeneities (or even ions) serve as sole origins for cavitating bubbles. Although practical observations are often in good agreement with solutions based on the above assumptions, the origination of microbubbles and the role of other factors (such as gas contamination) have not been described satisfactorily mainly due to the fact that the classical theory of homogeneous nucleation fails to predict the experimentally observed nucleation rates for boiling and cavitation at temperatures far from the critical point.

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2 Revision of the nucleation theory

This work relies on the theoretical extension of the classical theory of homogenous nucleation suitable for mixtures published by Maršík et al. (2002). In this extension, the discrepancy between the nucleation rates obtained from the classical theory and the experiments is explained by the effect of non-equilibrium (dissipative) processes on the work of formation of a nucleus of the new phase. The change of the nucleation barrier due to these dissipative phenomena (caused for example by bubble dynamics, departure of the liquid from purity, and even some heterogeneous effects) is expressed using a substance dependent correction coefficient which must be determined experimentally.

In the classical nucleation theory the nucleation rate J is expressed using the Döring-Volmer formula:

$$J = \rho_L \sqrt{\frac{2\sigma}{\pi m_1^3}} \exp\left(-\frac{W}{kT}\right) \quad (1)$$

In the above, ρ_L is the liquid density, k is the Boltzmann constant ($1.38 \cdot 10^{-23}$ J/K), T is the liquid temperature, σ is surface tension, and m_1 is the mass of one molecule. In this extension, the nucleation work W is split into the equilibrium portion W_{eq} and the non-equilibrium portion F_{noneq} :

$$W(\Delta\mu_{iVL}, \sigma(\Delta\mu_{iVL}, T, p)) = W_{eq}(\Delta\mu_{iVL}, \sigma) + F_{noneq}(\Delta\mu_{iVL}, T, p) = W_{eq}(\Delta\mu_{iVL}, \sigma)(1 - 2\alpha) \quad (2)$$

In Eq. (2), $\Delta\mu_{iVL}$ denotes the change of the chemical potential of component i due to evaporation (the difference between the chemical potential of the liquid and the vapor phase, $\Delta\mu_{iVL} = \mu_{iL} - \mu_{iV}$), p is pressure, and α is the coefficient capturing the dissipative processes. For a one-component system ($i = 1$, $\Delta\mu_{iVL} \rightarrow \Delta\mu_{VL}$), the coefficient is defined as follows:

$$\alpha = -\frac{F_{noneq}(\Delta\mu_{VL}, T, p)}{2W_{eq}(\Delta\mu_{VL}, \sigma)}, \text{ where } 0 \leq \alpha \leq 1/2 \quad (3)$$

The coefficient α expresses dissipative effects involved in the origination of the nucleus such as inertial and damping effects coupled with evaporation and diffusion of contaminant gas (described by Eq. (6) in Chapter 5).

In order to express the equilibrium portion of the nucleation work the theory of thermodynamic fluctuations has been engaged (Maršík et al. (2003)) yielding the following formula for the dimensionless equilibrium nucleation work (Gibbs number) $Gb_{eq} = W_{eq} / kT$:

$$Gb_{eq} = \sigma R_B(T) \left(-\left(\frac{4\pi}{3kT}\right)^2 v^{-1} \left(\frac{\partial p}{\partial v}\right)_T^{-1} \right)^{1/3}, \text{ where the bubble radius } R_B = \left(\frac{-3kT}{4\pi v (\partial p / \partial v)_T} \right)^{1/3} \quad (4)$$

It has been assumed that the energy of thermodynamic fluctuations near the maximum superheat or maximum breaking tension is comparable to the minimum work of formation of a bubble. The key factors in formula (4) are the surface tension σ and the value of $(\partial p / \partial v)_T$ at the saturation line (v is specific volume). The latter quantity represents the ability of the liquid to withstand tension at phase transition and is shown in Fig. 1a in for water in reduced coordi-

nates ($\bar{p} = p / p_{crit}$, $\bar{v} = v / v_{crit}$, $\bar{T} = T / T_{crit}$, where p_{crit} , v_{crit} and T_{crit} are pressure, specific volume, and temperature at the critical point, respectively).

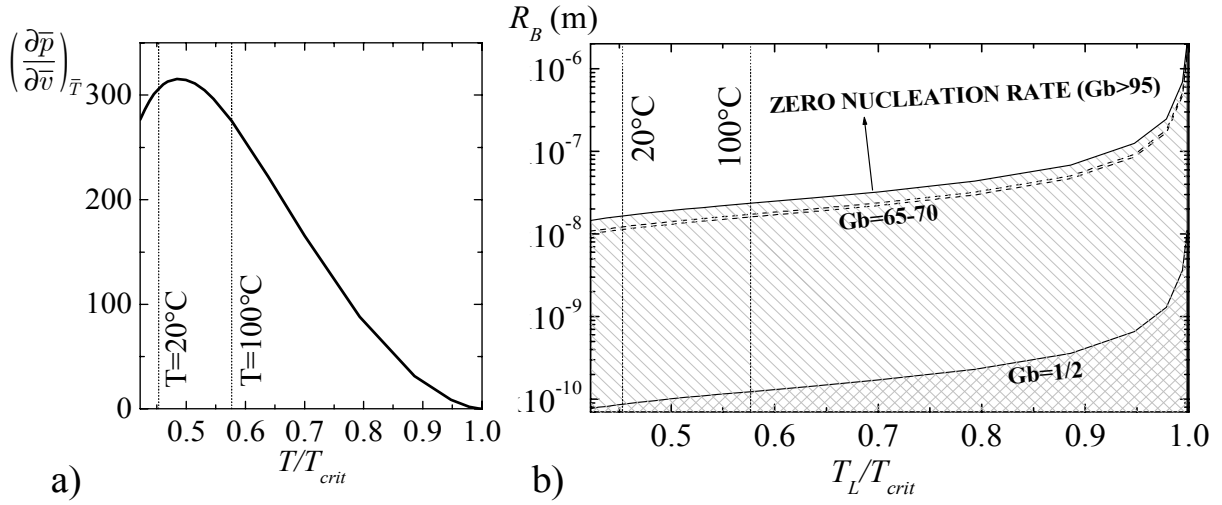


Fig. 1 a) The dependence of $-(\partial\bar{p}/\partial\bar{v})_{\bar{T}}$ on the reduced temperature for water at the saturation line (from IAPWS 95 data). b) The variation of the critical nucleus radius (the most probable fluctuated volume) with temperature for different assumed values of the Gibbs number. Calculated from Eq. (4) and IAPWS 95 data.

The nucleation rate for cavitation takes the final form,

$$J = \rho_L \sqrt{\frac{2\sigma}{\pi m_1^3}} \exp(-Gb), \text{ where } Gb = W / kT = (1 - 2\alpha)Gb_{eq} \quad (5)$$

3 Theoretical estimation of α for water

Table 1 contains the theoretical estimation of the value of α based on the theory described in Maršík et al. (2003). It has been assumed that the pressure fluctuations at the saturation line are equal to the tensile strength of water. The value of the Gibbs number has been set to typical values $Gb = 60 \div 70$ observed by most experimentators. However, the corresponding volume fluctuations for spinodal breakdown theory (i.e. pressure fluctuation Δp equal to the breaking tension 10^8 Pa at 20°C) correspond to very small values of nucleus radii R which are comparable to the size of a water molecule. The associated non-equilibrium portion of the nucleation work F_{noneq} is about 20 times larger than the equilibrium work W_{eq} .

It has been demonstrated in Zima (2003) that the theory predicts very high nucleation rates at any temperature. In order to obtain nucleation rates in the desired region $Gb = 65 \div 70$, the corresponding fluctuation must be in a narrow range about 10^{-8} m (see Fig. 1b). It can be speculated that in a pure liquid, the most probable volume fluctuations would always produce explosive nucleation rates. In a real fluid contaminated with gases and heterogeneous impurities, however, only bubble volumes greater than a certain value (for example $V \approx 10^{-25}$ m³)

serve as origins of the new phase. The probability of thermodynamic fluctuations of this magnitude is practically zero at the given temperature. Since the corresponding portion of the non-equilibrium work is immense (as will be shown in the next chapter) this is a demonstration of the expected fact that other significant non-equilibrium processes are responsible for nucleation in water at normal temperatures. It also implies that the search for an improved formula determining the equilibrium work of formation of a nucleus has to be continued in order to provide more acceptable values in the range of observed nucleation rates and critical radii.

Table 1. Estimation of α for cavitating water for various assumed values of the Gibbs number. Fitted to the theoretical breaking tension at 20°C.

Temperature $T = 293.15$ K		Pressure fluctuation $\Delta p = 10^8$ Pa	
Calculation from the theory			
Bubble radius R [m]	Gb_{eq}	Correction α	
		$Gb=65$ ($J=2 \cdot 10^{12}$ [m ⁻³ s ⁻¹])	$Gb=70$ ($J=2 \cdot 10^{10}$ [m ⁻³ s ⁻¹])
$2.12 \cdot 10^{-10}$	3	-9.10	-9.84
		Ratio of non-eq. and eq. work $ F_{noneq}/W_{eq} $	
		18	20

4 Experimental estimation of α for tap water

The value of α for tap water has been estimated experimentally using the experimental setup based on the convergent-divergent nozzle principle and described previously in Zima & Maršík (2001). It is assumed that bubbles are nucleated before entering the convergent section of the nozzle where the nucleation is ceased and the number of nucleate bubbles remains constant. The bubbles then grow in the convergent section and they reach the throat where the void fraction is estimated from the measured throat pressure (given the choked flow conditions have been achieved in the nozzle). The growth of bubbles in the convergent section is calculated using the Rayleigh-Plesset equation and the initial (critical) bubble radius at the entrance to the convergent section is calculated from the revised nucleation theory described in Chapter 2.

The value of α for tap water at 15.8°C has been estimated to be $\alpha = 0.4994$ for the assumed value of the nucleation rate $J = 10^{12}$ nuclei m⁻³ s⁻¹. This corresponds to non-equilibrium portion of the nucleation work equal in magnitude to the equilibrium portion work ($|F_{noneq}/W_{eq}| \sim 1$) and the critical radius $R = 6.5 \cdot 10^{-7}$ m.

5 Mass transport across the bubble wall

The present-day modeling of mass transport across the surface of a bubble in a liquid follows two basic schemes: full single bubble models and simplified solutions to the mass diffusion

equation tailored to specific problems (such as rectified diffusion). The first scheme is associated with substantial computational difficulties with the little prospect of implementing into the practical models of cavitating flows, whereas the second scheme provides useful insight into the mass transfer effects, however, generalization to complex problems is not possible because the bubble dynamics is disregarded. Recently, Preston et al. (2001) have combined the full single bubble model with the simple Rayleigh–Plesset equation to identify areas of possible simplifications to the full single bubble model. Our approach is based on using the revised Rayleigh–Plesset equation presented in Zima & Maršík (2000). This equation (unlike the widely used simple R–P equation) includes the effects of mass transport across the bubble wall via the mass flux per unit area, j_B :

$$\frac{p_B - p_\infty}{\rho_L} = R\ddot{R} + \frac{3}{2}\dot{R}^2 + \left(4\frac{\mu_L}{\rho_L} - 2R\frac{j_B}{\rho_B}\right)\frac{\dot{R}}{R} + \frac{2\sigma}{\rho_L R} + R\frac{\dot{j}_B}{\rho_L} \quad (6)$$

R is the bubble radius, μ_L , ρ_L , σ are the dynamic viscosity, density, and surface tension of the liquid, respectively, and p_B , p_∞ are the bubble and ambient pressures, respectively. Eq. (6) has been engaged in two numerical experiments after showing encouraging results for the problem of bubble dissolution by gas diffusion. First, the mass flux j_B across the bubble surface has been modeled using the assumption of steady gas diffusion:

$$j_B = -D\rho_L(c_{LB\infty} - c_{LB})/R \quad (7)$$

D is the diffusivity for the given gas–liquid combination, $c_{LB\infty}$ and c_{LB} are mass concentrations of gas in infinity and at the surface, respectively. The application to rectified diffusion (Fig 2a) shows good adherence to the theory found in Fyrrillas & Szeri (1994). In the second experiment, the mass transport due to phase transition at the bubble wall has been modeled by

$$j_B = -\alpha\rho_V^2 h_{LV}^2 R^2 \dot{R} \left(\lambda T_\infty^{3/2} R_0 \sqrt{2\pi\tilde{r}_V}\right)^{-1} \quad (8)$$

where α is the accommodation coefficient, h_{LV} is the evaporation heat, T_∞ is the ambient liquid temperature, R_0 is the initial bubble radius, \tilde{r}_V is the gas constant of vapor, λ is the liquid thermal conductivity, and ρ_V is the vapor density. Fig. 2b shows the effect of the phase transition on bubble growth which is in accordance with Preston et al. (2001).

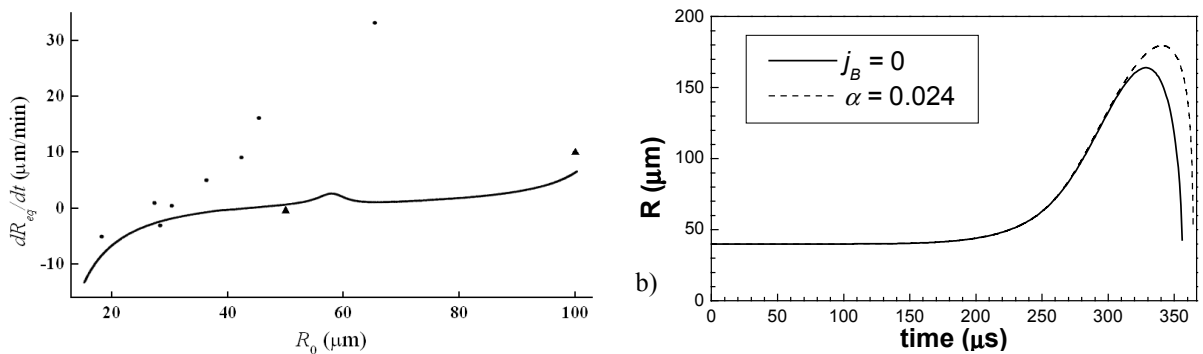


Fig. 2 a) The bubble growth rate vs. equilibrium radius for 0.2 bar sinusoidal pressure forcing at 1 bar (Fyrrillas & Szeri (1994) (theory = solid curve, round points = numerical experiment). Numerical solution by Eqs. (6) and (7) represented by triangles. b) A bubble growth–collapse cycle with phase transition (dashed curve) and without phase transition (solid curve).

6 Conclusion

One of the motivations for this research are very low nucleation rates in cavitation predicted by the classical theory of homogenous nucleation. On the other hand, the nucleation rates predicted by the revised theory presented in Maršík et al. (2002) and (2003) are very high indicating that substantial non-equilibrium effects are responsible for lowering the nucleation rates. The energy dissipation accompanying bubble formation must be studied further. Possible areas for investigation are the effect of temperature fluctuations and the dependence of surface tension on curvature and chemical composition (concentration and type of contaminants).

The modified Rayleigh-Plesset equation (with the inclusion of mass transport across the bubble boundary) improves the accuracy of modeling of bubble dynamics and proves that gas diffusion can be neglected for macroscopic bubbles and short time scales whereas the effect of phase transition should be considered in many cases.

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