Validation of Bubble Dynamics Equation for a Nano-scale Bubble via Molecular Dynamics Simulation

S Tsuda, H Hyodo, S Watanabe
1Department of Mechanical Engineering, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan
2Department of Mechanical Engineering, Graduate School of Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan

E-mail: tsudashin@mech.kyushu-u.ac.jp

Abstract. For a validation of the application of conventional bubble dynamics to a nano-scale bubble behaviour, we simulated a nano-scale bubble collapsing or vibration by Molecular Dynamics (MD) method and compared the result with the solution of Rayleigh-Plesset (RP) equation and that of Confined RP (CRP) equation, whose boundary condition was corrected to be consistent with that of MD simulation. As a result, a good coincidence was obtained between MD, RP, and CRP in the case of one-component fluid. In addition, also a good correspondence was obtained particularly in the comparison between MD and CRP in the case of two-component fluid containing non-condensable gas. The present results indicate that conventional bubble dynamics equation can be applied even to a nano-scale tiny bubble.

1. Introduction
We sometimes encounter the problem of cavitation in the development of fluid machinery such as pump and water turbine, and it causes deterioration of performance, unsteady vibration, noise, and erosion, all of which are not ignorable. Above all, preventing the fluid machinery from erosion, which originates from collapse of cavitation bubbles, is important for keeping the life time of the fluid machinery longer, and appropriate prediction of erosion in advance is required in the design phase. Such prediction method has progressed from continuum mechanics, such as fluid dynamics including bubble dynamics or CFD simulation, and also by solid mechanics. Many important and useful results have been obtained, but one of the essential questions is remained against the conventional method particularly for its application to the final process of collapsing bubble: To what extent can conventional bubble dynamics derived from fluid mechanics in the framework of continuum mechanics express the behaviour of a tiny bubble (just before the collapse) quantitatively? The clear answer against this question has not been obtained, and if conventional bubble dynamics were inappropriate against the description of collapsing bubble, some parts of the conventional prediction method for erosion might be revisited.

In this paper, we conducted a validation of bubble dynamics equations such as Rayleigh-Plesset (RP) equation throughout comparison with a result by Molecular Dynamics (MD) simulation, in which all molecular motion is numerically calculated based on a momentum equation for each molecule.
Bubble dynamics equations derived from fluid dynamics as continuum mechanics, which assume local equilibrium, might have some limitations for such small bubble dynamics because a tiny bubble just before collapse within very short time scale would not be necessarily in local equilibrium state, in which all molecular velocities must be Maxwell-Boltzmann distribution. On the other hand, at least theoretically, MD can rigorously describe both microscopic and macroscopic phenomena because it does not need any assumption such as local equilibrium. In this point of view, MD can principally offer more reliable results compared with continuum mechanics, and we can investigate whether macroscopic formulation such as RP can express the microscopic bubble dynamics or not using MD simulation. However, in the validation process of the applicability of conventional bubble dynamics equations, we must also deliberate items below carefully: (I) difference of boundary condition between bubble dynamics equation and MD, (II) influence of the feature of computational domain (e.g., spherical domain or cubic domain), (III) evaluation accuracy of thermophysical properties in MD, and so on. A few studies [1-3] already conducted comparison between bubble dynamics equations and MD results for a nano-scale bubble and the above points (II) and (III) were directly or indirectly investigated. As a result, the previous studies claim that bubble dynamics equations can fairly well describe the dynamic behaviour of a nano-scale bubble through the investigations. However, the influence of the point (I) has hardly been discussed. Point (I) is very important to obtain a clear answer against the question of “To what extent can bubble dynamics derived from the framework of fluid mechanics as continuum mechanics express the behaviour of a tiny bubble quantitatively?” because boundary condition of bubble dynamics equations such as RP and that of general MD are absolutely different as mentioned later in the subsection 2.2.

In this paper, we simulated a collapsing or vibrating bubble whose diameter is several nanometers using MD simulation, and the radius change was compared with not only RP equation but also a Confined RP (CRP) equation, which was derived in order to deliberate the influence of boundary condition. Throughout the comparison, we have reached closer answer against that question.

2. Method

2.1. Molecular Dynamics Simulation

All molecular interactions were given by Lennard-Jones (LJ) potential [4] in a cubic computational domain on which a periodic boundary condition is imposed on all directions. Here, two kinds of fluid were simulated in this study; one is pure liquid oxygen, and the other is liquid oxygen including helium as non-condensable gas with the molar concentration of 0.05. Namely, the former is one-component fluid, and the latter is two-component. In both cases, 702,464 molecules were used to simulate, and the length on a side of the computational domain was around 30nm. The cut-off radius [4] was set to 3.5 in LJ unit as a standard value, and we have imposed an isothermal condition to keep the temperature constant based on so called \(NVT\)-constant MD method [4].

In the MD simulations, we first reproduced saturated liquid oxygen or the liquid oxygen containing helium as non-condensable gas with the molar concentration of 0.05. Namely, the former is one-component fluid, and the latter is two-component. In both cases, 702,464 molecules were used to simulate, and the length on a side of the computational domain was around 30nm. The cut-off radius [4] was set to 3.5 in LJ unit as a standard value, and we have imposed an isothermal condition to keep the temperature constant based on so called \(NVT\)-constant MD method [4].

In the MD simulations, we first reproduced saturated liquid oxygen or the liquid oxygen containing helium gas at 90K in the computational domain, and slightly expanded the domain uniformly and isotropically in order to decrease the liquid density from the saturation liquid state. If we choose an appropriate density after the expansion, homogeneous nucleation occurs and finally one stable bubble is formed. This state is regarded as an initial condition, and the computational domain was next slightly compressed in order to cause the bubble collapsing. In this study, we focused on the bubble radius change with time. In the evaluation of the bubble radius, bubble region is recognized as “a cluster constituted by void points” and we extracted a “cavity cluster” as a bubble [5]. Namely, the bubble radius was calculated from the volume of that cavity cluster.

Spherical shape of the bubble and incompressibility of liquid are the two important assumptions in the derivation process of RP. In advance, we confirmed that the bubble shape was sufficiently spherical at least in the present simulation condition. Also, we confirmed in advance that the bubble wall speeds observed in the MD simulations were clearly less than the Mach number of 0.3. Note that the present MD simulations were conducted so as to satisfy these two points.
2.2. Rayleigh-Plesset (RP) and Confined Rayleigh-Plesset (CRP) Equations

RP equation describes a spherical bubble motion which is placed in an incompressible liquid. In this equation, pressure as one of the boundary conditions is given at a place sufficiently far from the bubble. On the other hand, the computational region in MD is finite and a periodic condition is imposed. Namely, the liquid region surrounding the bubble in MD is very small and confined. To eliminate the influence of the difference in boundary condition between RP and MD as much as possible, we have derived a Confined RP (CRP) equation, whose boundary condition matches that in the present MD simulation. The derivation process is as follows.

In the derivation of RP, incompressible Navier-Stoke equation in a spherical polar coordinate is integrated from a position infinitely far from centre of a bubble to the bubble wall corresponding to the bubble radius \( R \). In the derivation of CRP, we have limited the integration range from a finite \( R_{\text{ref}} \) whose radius equivalently corresponds to the volume of the computational domain in MD, to the bubble radius \( R \). Here, fluid velocity is set to be zero at a place infinitely far from the bubble in RP while it was set to be zero at \( R_{\text{ref}} \) in CRP. Rigorously speaking, the latter boundary condition for the velocity is inconsistent to that in MD, but the region corresponding to \( R_{\text{ref}} \) is fairly close to the boundary of the computational domain in which periodic boundary condition is imposed. Taking into account that the periodic boundary condition keeps the number of molecules in the domain constant, the net velocity around the region is expected to be close to zero. In addition, pressure in RP is given at the position infinitely far from the centre of the bubble while the pressure in CRP is given as \( p_{\text{ave}} \) which denotes the spatially averaged pressure of liquid field at each time \( t \). Note that pressure in MD is generally obtained in spatially averaged manner [4] while pressure in the original RP is given at sufficiently far from the bubble. This inconsistency could be also the possible reason of the difference between RP solution and MD result. Therefore, CRP was formulated so as to eliminate those differences as much as possible. The final form of CRP derived in this paper is given as follows.

\[
\rho \left\{ a(z)R\ddot{R} + b(z)R^2 \right\} = p(R) - p_{\text{ave}}(t) - \frac{2\gamma}{R} - 4\mu \dddot{R}/R \\
(1)
\]

Here, \( z \) is defined as \( z = R_{\text{ref}}/R \) (note that \( z \) as the ratio of equivalent domain radius \( R_{\text{ref}} \) and bubble radius \( R \) must be larger than 1 because \( R_{\text{ref}} \) is of course larger than \( R \)) and

\[
a(z) = \frac{(z-1)(5z^3 + 6z^2 + 3z + 1)}{5(z^2 + z + 1)^2}, \quad b(z) = \frac{1}{2} \left\{ \frac{(5z^3 + 6z^2 + 3z + 1)(4z^3 + 5)}{5(z^2 + z + 1)^2} - 1 \right\} \\
(2)
\]

In Eq. (1), \( R \) is the bubble radius, \( \rho \) the liquid density, \( p(R) \) the pressure at the bubble wall, \( \gamma \) the surface tension, and \( \mu \) the liquid viscosity coefficient. In L.H.S. of Eq. (1) as the inertial term, we can easily confirm the coefficients \( a(z) \) and \( b(z) \) become 1 and 3/2 respectively as same as those in original RP if \( R_{\text{ref}} \) or \( z \) becomes infinity (This situation almost corresponds to the boundary condition of original RP). Note that \( a(z) \) is less than 1 and \( b(z) \) is less than 3/2 for all \( z > 1 \). In a confined system such as the computational domain in MD, liquid field is narrow and the liquid inertia is clearly weaker than that in original RP. This effect is mainly reflected to Eq. (1). On the other hand, R.H.S. of Eq. (1) is almost the same as that in the original RP except \( p_{\text{ave}} \) (In the original RP, \( p_{\text{ave}} \) in Eq. (1) becomes pressure given at a place sufficiently far from the bubble).

In the numerical solution of RP or CRP, \( p(R) \) was given as the saturation vapour pressure in the case of one-component fluid while it was estimated based on the assumption that the gas inside the bubble shows a polytropic change as an ideal gas in the case of two-component fluid. Equation (1) was numerically solved by 4th order Runge-Kutta method.

3. Results and Discussion

Figure 1(a) shows the bubble radius change of the collapsing bubble in the one-component case. As is shown, fairly good correspondences are obtained between MD, RP, and CRP. If we judge a bubble
“collapsed” when its radius becomes around 1nm, difference of the collapsing time between MD, RP or CRP is estimated to be less than 15%. Here, we can figure that both RP and CRP quite well reproduce the bubble collapse in this case. Next, we compared the solutions of the two equations (RP and CRP) and MD result in liquid oxygen with helium gas. Figure 1(b) shows each radius change in the case of the two-component case, and as is shown, bubble radius in MD shows “rebound”, and this phenomenon is also reproduced in RP and CRP qualitatively. And quantitatively, CRP reproduces the MD result better than RP. The first rebound time in CRP becomes shorter and closer to that in MD compared with RP, and it is originated from the appropriate evaluation of the inertial term in CRP (Weaker inertia reflected to CRP in Eq. (1) makes the rebound time shorter). This result indicates that bubble dynamics equation on the basis of fluid dynamics in the framework of continuum mechanics can be applied even to a nano-scale bubble if boundary condition is appropriately taken into account.

4. Conclusion
A validation of the application of bubble dynamics equation to a nano-scale bubble was conducted employing Molecular Dynamics (MD) simulation. In this study, not only oxygen as pure fluid but also oxygen including helium as non-condensable gas were simulated for the validation. As a result, the bubble radius changes observed in the MD simulation were reproduced fairly well by Rayleigh-Plesset (RP) equation, and by Confined Rayleigh-Plesset (CRP) equation, which was originally derived in this paper in order to compensate the essential difference of boundary condition between RP and MD simulation. The present results indicate that bubble dynamics equation on the basis of fluid dynamics as continuum mechanics can be applied even to a nano-scale bubble if we appropriately take the boundary condition into account.

References
[1] Okumura, H. and Ito, N. 2003 Nonequilibrium molecular dynamics simulations of a bubble Nonequilibrium molecular dynamics simulations of a bubble, PHYSICAL REVIEW E 67, 045301(R)
[2] Holyst, R., Litniewski, M., and Garstecki P. 2010 Large-scale molecular dynamics verification of the Rayleigh-Plesset approximation for collapse of nanobubbles PHYSICAL REVIEW E 82, 066309
[3] Holyst, R., Litniewski, M., and Garstecki P. 2012 Collapse of a nanoscopic void triggered by a spherically symmetric traveling sound wave PHYSICAL REVIEW E 85, 056303
[4] Allen, M. P. and Tildesley D. J. 1987 Computer Simulation of Liquids. Clarendon Press
[5] Maruyama S. and Kimura T. 2000 A Molecular Dynamics Simulation of a Bubble Nucleation on Solid Surface Int. J. Heat & Technology 18-supplement1, 69-74