

Molecular dynamics of rupture phenomena in a liquid thread

Satoyuki Kawano*

Institute of Fluid Science, Tohoku University, 2-1-1 Katahira, Aobaku, Sendai 980-8577, Japan

(Received 23 December 1997; revised manuscript received 4 May 1998)

The interfacial motions of a liquid thread with nanometer-order diameter are numerically analyzed by the use of molecular dynamics simulations of up to 10 278 Lennard-Jones molecules in three dimensions. The rupture phenomena in a liquid thread and the formation process of ultrafine liquid particles are successfully simulated for various conditions. The numerical results of the interfacial phenomena in the liquid thread, which include the unstable wave motion and the rupture time, are quantitatively compared with the theoretical results based on the classical linear instability theories. Consequently, it is found that the numerical results of wavelength are in reasonable agreement with those obtained using the inviscid linear instability theory. The results obtained here, which are concerned with the rupture of ultrafine liquid thread, will provide fundamental information on the liquid atomization phenomena from a microscopic viewpoint. [S1063-651X(98)08309-3]

PACS number(s): 47.20.Ma, 47.20.Dr, 47.11.+j, 02.70.Ns

I. INTRODUCTION

Liquid atomization and spray systems have received considerable attention because of their wide range of applications, e.g., for power, propulsion, heat/mass exchange, and material processing industries. Although the accumulation of fundamental information regarding liquid atomization is strongly required, the phenomena are extremely complex and are not fully investigated from the viewpoint of fluid dynamical analogy. Difficulties in the investigation arise due to the rupture phenomena in the continuous liquid [1]. There exist a large number of theoretical works on the analogy of atomization phenomena based on the instability theory such as the well-known Rayleigh instability [2] or on the numerical simulation technique by the use of a MAC method [3]. These works have been effective for the rough estimation of the macroscopic flow mechanism of atomization and the liquid particle size produced. However, their treatments are limited to the macroscale phenomena such as the interfacial wave motion in the continuous liquid. It should be noted that the rupture phenomena in the continuous liquid, e.g., the formation process of liquid particles by the torn-off liquid thread, are mainly governed by the molecular force and cannot be treated by the continuum dynamics such as Navier-Stokes equations. The lack of microscopic fundamental knowledge of the rupture phenomena is a major hindrance in the further advancement of the research of not only liquid atomization but also fluid interfacial motions. Although the molecular dynamics simulations technique should be highly effective, there exist only a few works on fluid interfacial motion with regard to the liquid atomization phenomena [4,5] because the molecular dynamics simulations require extremely large computations. From the viewpoint of the physical interests and the engineering applications, theoretical analysis using a simplified model of the rupture phenomena based on molecular dynamics is essential and remains to

be performed as the latest frontier of fluid mechanics research.

The present research focuses on the unstable motion of a vapor-liquid interface in a circular liquid thread, which is the most basic and important phenomenon in liquid atomization. The behavior of the interface and the rupture phenomena in the liquid thread with nm-order diameter are numerically studied by the use of molecular dynamics simulations of 607 to 10 278 Lennard-Jones molecules in three dimensions. Consequently, the rupture of the liquid thread and the formation process of ultrafine liquid particles are successfully simulated. Furthermore, the present numerical results of the interfacial phenomena, which consist of the unstable wave motion on the liquid thread interface and the rupture time of the thread, are compared with the analytical results based on the well-known instability theories. The validity of applying molecular dynamics simulations to the liquid interfacial phenomena is also discussed in detail. The present research will provide further information on the rupture phenomena in a liquid thread and useful suggestions for future research on the fluid interfacial motions such as the liquid atomization.

II. PROCEDURE FOR MOLECULAR DYNAMICS SIMULATIONS

The basic equations and the computational procedure are based on typical molecular dynamics simulations [6]. The Lennard-Jones 12-6 potential ϕ is used as

$$\phi(r) = 4\varepsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right], \quad (1)$$

where r denotes the distance between two molecules, and where the parameters ε and σ denote the representative scales of energy and length, respectively. The molecular dynamics simulations were performed using a number of molecules placed initially in a cubical box termed a fundamental cell with periodic boundary conditions in all three dimensions. To prevent an $O(M^2)$ computation due to interactions between all pairs of molecules, the potential was cut off at a distance $r = 2.5\sigma$, where M denotes the molecular number. A

*FAX: +81-22-217-5311.

Electronic address: kawano@ifs.tohoku.ac.jp

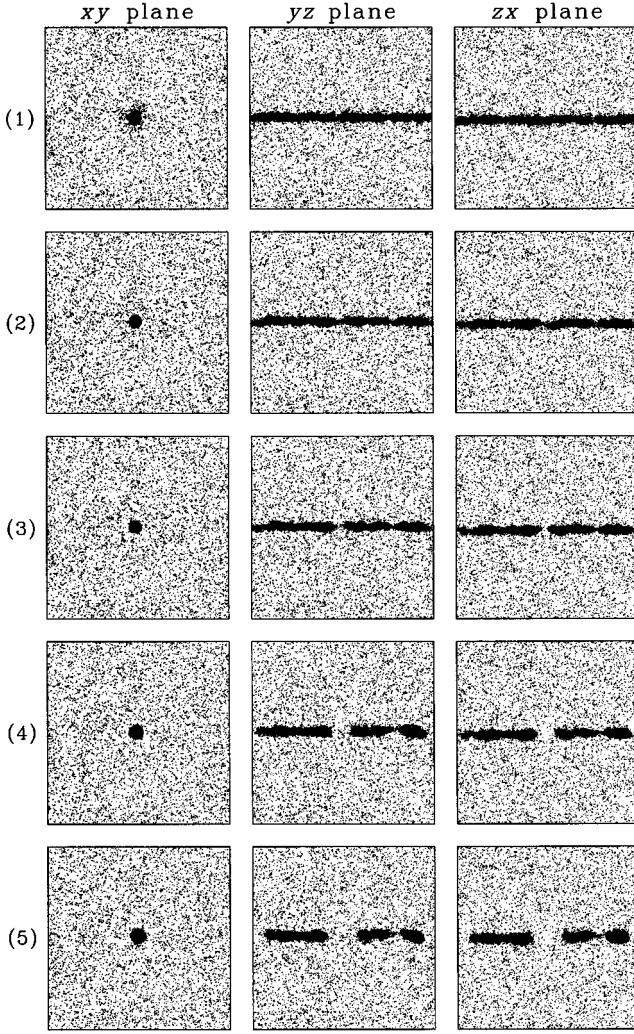


FIG. 1. Unstable motion of the vapor-liquid interface in the liquid thread at $L^*=120$, $R^*=3$, $T^*=0.75$, $\rho_L^*=0.819$, and $\rho_V^*=0.0031$: (1) $t^*=60$; (2) $t^*=120$; (3) $t^*=180$; (4) $t^*=240$; (5) $t^*=300$.

minimum image convention technique and a standard neighbor list to keep track of which molecules were actually interacting at a given time interval 0.1τ were used, where $\tau = \sqrt{m\sigma^2/48\epsilon}$ denotes the representative time scale and the mass m of the molecule is taken as the representative mass scale. The computational time was close to $O(M)$. Under the simulation techniques described above, the motion of the molecules was obtained by the use of a leapfrog algorithm with a time step of 0.001τ . As the initial conditions, the molecules in a vapor phase were distributed in a fundamental cell and the molecules in a liquid phase were distributed within the cylindrical region, which corresponded to the liquid thread by the use of random numbers. The cylindrical region was set in the center of the cell. The initial velocities of molecules were decided by the use of normal random numbers, so that the velocity of each molecule had a Maxwell distribution. The achievement of the equilibrium state was confirmed by obtaining the radial distribution function and by observing the temperature of the system. The agreements of the thermal properties such as critical point of the Lennard-Jones fluid between the results based on the present

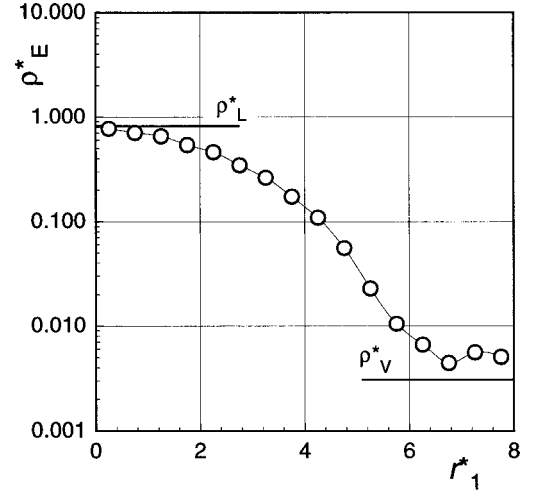


FIG. 2. Dimensionless local density ρ_E^* distribution in the fundamental cell at $L^*=120$, $R^*=3$, $T^*=0.75$, $\rho_L^*=0.819$, and $\rho_V^*=0.0031$.

simulation code developed here and previous ones [7] were confirmed, which are omitted here.

III. RESULTS AND DISCUSSION

A. Unstable motion of vapor-liquid interface

The interfacial motions of the liquid thread are discussed in the case of the temperature $T^*=0.75$, liquid density $\rho_L^*=0.819$ and vapor density $\rho_V^*=0.0031$, where superscript * denotes the dimensionless value. These dimensionless values correspond to $T=70.0$ K, $\rho_L=1223$ kg/m³, and $\rho_V=4.63$ kg/m³ in argon. Here, in the case of argon, the parameters $\sigma=3.54\times 10^{-10}$ m, $\epsilon/k_B=93.3$ K, and $m=6.64\times 10^{-26}$ kg are used [8], where $k_B=1.38\times 10^{-23}$ J/K denotes Boltzmann constant. The computational conditions of T^* , ρ_L^* , and ρ_V^* were chosen considering the phase diagram of the Lennard-Jones fluids. That is, T^* was set at a value higher than the triple point temperature [7]. The values ρ_L^* and ρ_V^* were set to be the liquid density and vapor density, respectively, in the coexistence properties for the Lennard-Jones fluids [9]. In the present research, the number of molecules, cell volume, and temperature of the system were set to be constant.

Figure 1 shows the unstable motion of the vapor-liquid interface in the liquid thread at the fundamental cell length $L^*=120$ and initial liquid thread radius $R^*=3$, where t^* denotes the time. Cartesian coordinates (x,y,z) are used. In Fig. 1, the dimensionless values correspond to $L=42.5$ nm, $R=1.06$ nm, and $60\tau=22.0$ ps in argon. The dot in Fig. 1 indicates the center of the molecule. The viewing point is moved so that the center of mass of all the molecules in the fundamental cell coincides at the center of the cell. It was confirmed, by obtaining the radial distribution function, that the thread maintained a liquid state and the ambience had a vapor state, and that they had already reached the equilibrium state before $t^*=10$. From Fig. 1, it is found that the unstable motion of the liquid thread interface is already present at $t^*=60$, the rupture of the liquid thread occurs from $t^*=120$ to 180, and finally, the molecules begin to form ultrafine liquid particles by the contraction motion of

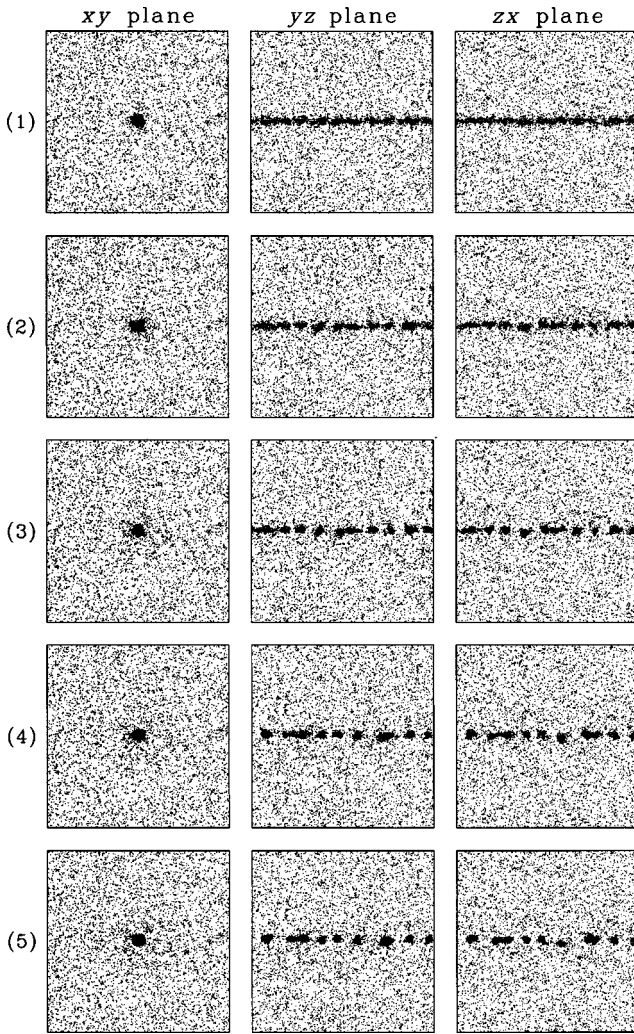


FIG. 3. Unstable motion of the vapor-liquid interface in the liquid thread at $L^*=120$, $R^*=2$, $T^*=0.75$, $\rho_L^*=0.819$, and $\rho_V^*=0.0031$: (1) $t^*=30$; (2) $t^*=60$; (3) $t^*=90$; (4) $t^*=120$; (5) $t^*=150$.

molecules in the axial direction of the liquid thread after $t^*=180$. An explicit perturbation on the vapor-liquid interface in the liquid thread is not imposed. Note that multiple liquid particles are formed in the fundamental cell. In previous simulations of the rupture of liquid thread [5], only one liquid particle was formed in the cell. In the molecular dynamics simulations using periodic boundary conditions, the wavelength in the vapor-liquid interface in the liquid thread, which corresponds to the particles size and the particle number in the fundamental cell, must be independent of the cell length. To investigate the dynamical analogy in the microscale atomization phenomena, a relatively large fundamental cell length in comparison with the wavelength in the vapor-liquid interface in the liquid thread is needed. If the contraction velocity of the liquid thread in the axial direction, immediately after the rupture, is relatively large and the fundamental cell length is relatively small, there is a high possibility of the formation of only one liquid particle in the cell by the simulation technique with the periodic boundary conditions. The dimensional velocity V of contraction motion in the axial direction of the liquid thread is 48.3 m/s at $t^*=180$ and 145 m/s at $t^*=300$ in argon. The velocity V

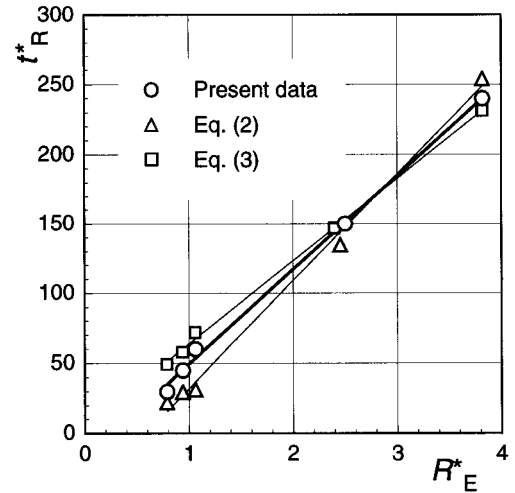


FIG. 4. Comparison of the dimensionless rupture time t_R^* of the liquid thread between the numerical results and the linear instability theories.

increases with time. It can be said, from Fig. 1, that the atomization process in the liquid thread is successfully simulated.

Figure 2 shows the dimensionless local density ρ_E^* distribution in the fundamental cell at the equilibrium state, which is averaged in the axial direction of the liquid thread at $10 \leq t^* \leq 90$. The computational conditions are the same as those in Fig. 1. Here, r_1^* in Fig. 2 denotes the dimensionless radial distance from the central axis of the liquid thread at the initial state. As shown in Fig. 2, ρ_E^* becomes large and approaches the constant value as r_1^* becomes small, which corresponds to the liquid density ρ_L^* , and ρ_E^* becomes small and approaches the vapor density ρ_V^* as r_1^* becomes large. Furthermore, the so-called interfacial layer is well simulated in the region of about $2 < r_1^* < 6$, which is the transient state between vapor and liquid from the microscopic viewpoint. The interfacial layer is an important future research subject in the microscale fluid interfacial phenomena because the macroscopic fluid dynamical theory cannot describe the detailed structure of the layer. Because ρ_E^* is rapidly varied by r_1^* and due to the existence of the interfacial layer, the exact definition of the vapor-liquid interface is difficult. In the present research, liquid thread radius R_E^* at the equilibrium state is defined by the point $\rho_E^* = (\rho_L^* + \rho_V^*)/2$. It is estimated that $R_E^* = 2.45$ in Fig. 2.

The case of thinner liquid thread is discussed here. Figure 3 shows the unstable motion of the vapor-liquid interface in the liquid thread at $L^*=120$ and $R^*=2$. From Fig. 3, it is seen that the rupture time decreases and the more uniform sized particles are produced in comparison with Fig. 1. The velocity V is 96.5 m/s at $t^*=90$ in argon. Eight to nine particles are produced owing to the small wavelength. Coalescence of the particles can be seen at $t^*=90-120$. The very thin liquid thread does not evanesce and the atomization process in the liquid thread is successfully simulated. From the definition of R_E^* described above, it is estimated that $R_E^* = 0.939$.

B. Comparison with linear instability theories

The numerical results of wavelength in the vapor-liquid interface and the rupture time of the liquid thread are com-

TABLE I. All the conditions of the present simulation, the numerical results of number N of the ultrafine liquid particles produced in the fundamental cell and the numerical results of dimensionless wave number k' ($=kR_E$) in the liquid thread interface.

L^*	R^*	M	ρ_L^*	ρ_V^*	R_E^*	N	k'
40	2	607	0.819	0.0031	0.788	3–4	0.433
80	2	2407	0.819	0.0031	1.06	6	0.500
120	2	6587	0.819	0.0031	0.939	8–9	0.418
120	3	8124	0.819	0.0031	2.45	3	0.386
120	4	10278	0.819	0.0031	3.82	2	0.400

pared with the results of macroscopic linear instability theories. In the linear instability theory [2,10], the assumption $\eta = A \exp(\omega t - ikz)$ is introduced, where η denotes the displacement of unstable wave on the liquid thread interface, A the small amplitude, ω the growth rate, and k the wave number. In particular, ω in inviscid fluid can be expressed as

$$\omega = \left[\frac{\gamma}{R_E^3 \rho_L} \frac{I_1(k')}{I_0(k')} k' (1 - k'^2) \right]^{1/2}, \quad (2)$$

where γ denotes the tension of the liquid thread interface, I_n the n th-order modified Bessel function of the first kind and k' the dimensionless wave number, which is made dimensionless by the use of R_E . On the other hand, ω in fluid in the limit of high viscosity can be expressed as

$$\omega = \frac{\gamma}{6R_E \mu_L} (1 - k'^2), \quad (3)$$

where μ_L denotes the liquid viscosity. The dimensionless wave number can be written as $k' = 2\pi R_E^* N / L^*$ in the present research, where N denotes the number of ultrafine liquid particles produced in the fundamental cell. Figure 4 shows the rupture time t_R^* of the liquid thread under various conditions. Note that the numerical values include the time to the equilibrium state for the molecules initially placed in the fundamental cell to form a fluid, which is an $O(1)$ small time. The equation $t_R^* = C \omega^{-1}$ is used, where A/R_E is set to be constant. Although γ and μ_L can be obtained from the microscopic viewpoint at the equilibrium state, they are treated as constant values. All the conditions of the present simulation, the numerical results of N and the numerical results of k' are shown in Table I. From Fig. 4, the results obtained by the instability theories agree qualitatively with the numerical values, where the constant C is decided by the least squares method. Koplík and Banavar [5] concluded that the behavior of unstable motion of the liquid thread was well described by Eq. (3). In the present results, it is found that the behavior of unstable motion of the liquid thread is well described both by Eqs. (2) and (3). However, from the engineering viewpoint in the liquid atomization phenomena, there is a disadvantage in using Eq. (3), that is, the prediction of wave number, which is closely connected to the particles size produced, is impossible. The inviscid theory in Eq. (2) is suitable for the macroscopic atomization research and pro-

vides the useful suggestion that the most unstable wave with $k' = 0.697$, at which ω takes the largest value, has been thought to appear in the actual phenomena. This value of k' is independent of constant C . From Table I, it is found that the numerical results of k' are almost constant and not very far from the value of 0.697 in spite of the different approach in the interfacial dynamics. Precisely speaking, all the numerical values are smaller than the value of 0.697.

IV. CONCLUSIONS

The rupture phenomena in a liquid thread with nm-order diameter are studied by the use of three-dimensional molecular dynamics simulations in the Lennard-Jones potential. The main features of the present research are as follows.

(1) The atomization phenomena in the liquid thread, which consist of the unstable wave motion on the vapor-liquid interface, the interface rupture, the axial contraction motion of the torn-off liquid thread and the formation of liquid particles, are successfully simulated for various conditions. The local density distribution and the structure of the interfacial layer in the liquid thread are investigated quantitatively from the microscopic viewpoint.

(2) Comparisons of the rupture time between the present results and the macro-scale linear instability theories are made in detail. It is found that the numerical results of rupture time agree qualitatively both with the highly viscid theory and the inviscid theory.

(3) The numerical results of wave number are in good quantitative agreement with the well-known inviscid linear instability theory because the dimensionless wave number has an almost constant value of 0.386–0.500, where the inviscid linear instability theory results in the value of 0.697. This result provides the validity of applying the molecular dynamics simulations to the fluid interfacial phenomena such as the liquid atomization.

ACKNOWLEDGMENTS

The author wishes to thank Professor Emeritus Hiroyuki Hashimoto, Tohoku University, and Professor Taku Ohara, Institute of Fluid Science, Tohoku University, for valuable discussions.

- [1] S. Kawano, H. Hashimoto, H. Togari, A. Ihara, T. Suzuki, and T. Harada, *Atomization Sprays* **7**, 359 (1997).
- [2] D. B. Bogoy, *Annu. Rev. Fluid Mech.* **11**, 207 (1979).
- [3] F. H. Harlow and J. E. Welch, *Phys. Fluids* **8**, 2182 (1965).
- [4] S. M. Thompson, K. E. Gubbins, J. P. R. B. Walton, R. A. R. Chantry, and J. S. Rowlinson, *J. Chem. Phys.* **81**, 530 (1984).
- [5] J. Koplik and J. R. Banavar, *Phys. Fluids A* **5**, 521 (1993).
- [6] M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids* (Oxford University Press, Oxford, 1987).
- [7] B. Smit, *J. Chem. Phys.* **96**, 8639 (1992).
- [8] R. C. Reid, J. M. Prausnitz, and B. E. Poling, *The Properties of Gases and Liquids* (McGraw-Hill, New York, 1988).
- [9] A. Z. Panagiotopoulos, N. Quirke, M. Stapleton, and D. J. Tildesley, *Mol. Phys.* **63**, 527 (1988).
- [10] S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Stability* (Oxford University Press, Oxford, 1968).