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A molecular dynamics simulation of droplet evaporation

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Abstract

A molecular dynamics (MD) simulation method is developed to study the evaporation of submicron droplets in a gaseous surrounding. A new methodology is proposed to specify initial conditions for the droplet and the ambient fluid, and to identify droplet shape during the vaporization process. The vaporization of xenon droplets in nitrogen ambient under subcritical and supercritical conditions is examined. Both spherical and non-spherical droplets are considered. The MD simulations are shown to be independent of the droplet and system sizes considered, although the observed vaporization behavior exhibits some scatter, as expected. The MD results are used to examine the effects of ambient and droplet properties on the vaporization characteristics of submicron droplets. For subcritical conditions, it is shown that a spherical droplet maintains its sphericity, while an initially non-spherical droplet attains the spherical shape very early in its lifetime, i.e., within 10% of the lifetime. For both spherical and non-spherical droplets, the subcritical vaporization, which is characterized by the migration of xenon particles that constitute the droplet to the ambient, exhibits characteristics that are analogous to those reported for "continuum-size" droplets. The vaporization process consists of an initial liquid-heating stage during which the vaporization rate is relatively low, followed by nearly constant liquid-temperature evaporation at a "pseudo wet-bulb temperature". The rate of vaporization increases as the ambient temperature and/or the initial droplet temperature are increased. For the supercritical case, the droplet does not return to the spherical configuration, i.e., its sphericity deteriorates sharply, and its temperature increases continuously during the "vaporization" process.

Keywords: Molecular dynamics simulations; Submicron droplet; Subcritical and supercritical evaporation

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1. Introduction

The physics of droplet vaporization has attracted much interest during the past decades due to the crucial role played by sprays and atomizers in numerous propulsion, industrial, and environmental systems. Amongst them, spray combustion perhaps represents the most important application, considering the range of devices that rely on liquid–fuel combustion as their main source of energy. Consequently, many researchers in this area have developed important fundamental theories [1,2] on the evaporation of fuel droplets, taken as a single entity or included in a spray. The droplet sizes studied in these investigations have ranged from several microns to a few millimeters. Modeling the vaporization of such droplets has relied on the equations developed in continuum fluid mechanics. Although the sizes involved were small, they remained large enough to neglect the detailed molecular structure of the droplet. In addition, the macroscopic sizes have allowed extensive experimentation on their evaporation behavior, thus facilitating validation of the continuum-based models.

Nonetheless, not much is known about the evaporation of droplets when their initial sizes are in the submicron range. For this size range the continuum assumption becomes questionable: the molecular structure of the droplet is no longer negligible, since the

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droplet size is only a few orders of magnitude greater than the size of its constituent molecules. From a mathematical perspective, many variables, such as density, temperature, etc., which are assumed to be continuous functions of space and time lose this characteristic. The proper description for the vaporization of these smaller droplets, thus, requires a new methodology.

Molecular dynamics delivers this novel approach. A molecular dynamics (MD) study of any physical phenomena attempts to simulate the real behavior of nature by identifying each molecule and following their motion in time through the basic laws of classical mechanics. The system behavior and temporal evolution of its thermodynamic and transport properties are, therefore, nothing more than the cumulative result of individual molecular motion.

The simulation of macroscopic droplet evaporation using molecular dynamics is not yet possible due to the prohibitively large computational requirements both in terms of CPU time and memory. However, a system involving a submicron droplet is small enough to successfully apply MD. Studying these smaller droplets through MD will yield two primary benefits: (1) such an investigation will expand the knowledge of droplet evaporation to sizes that are yet to be tested; (2) through molecular analysis of the vaporization process, a better understanding will be acquired of the molecular physics of evaporating droplets of all sizes. Models developed in the studies of larger droplets generally rely on certain assumptions (for example: assuming the droplet to remain spherical throughout the process; neglecting second order effects such as the Soret and Dufour effects; assuming thermodynamic equilibrium at the droplet surface). A molecular model of a vaporizing droplet has no need for these assumptions. Rather, if these were correct, MD would simply confirm them.

Another motivation for developing MD approaches to study droplet phenomena stems from the consideration that many important droplet processes involve scales that cannot be resolved using continuum-based methods. For instance, the phase change and interfacial transport processes associated with transcritical and supercritical droplet vaporization involve subcontinuum scales, and, therefore, require phenomenological models [1–4] for their representation in a continuum approach. While such models may be appropriate to simulate droplet vaporization under subcritical conditions, they become questionable under transcritical/supercritical conditions. The MD simulations can be used to examine the applicability of these models under various conditions. The droplet vaporization under supercritical conditions also involves significant shape deformations [5] due to vanishing surface tension. Since these deformations have length scales ranging from the droplet size to the subcontinuum scale, continuum-based models [6–8] may not be able to represent them accurately. Moreover, atomization under supercritical conditions would result in non-spherical droplets with a dynamically deforming interface. MD methods may be ideally suited to simulate the vaporization and deformation behavior of these droplets.

Motivated by these considerations, the present study employs a MD approach to investigate the vaporization of submicron droplets. The major objectives of this investigation are: (i) examine the effect of system size on the simulated vaporization behavior, (ii) characterize differences and similarities between the vaporization of subcontinuum and continuum-size droplets, (iii) characterize the effects of ambient and droplet properties on the vaporization of submicron droplets, and (iv) examine the tendency of a droplet to remain spherical under subcritical and supercritical conditions.

Previous investigations using MD methods for the simulation of droplet vaporization have been relatively few. Long et al. [9] used a MD methodology to simulate the vaporization of an argon droplet in its own vapor under subcritical conditions. Kaltz et al. [10] extended this investigation to oxygen droplets evaporating in supercritical hydrogen and helium environments. The oxygen droplet was shown to exhibit a heating period followed by a constant-temperature evaporation for subcritical and moderately supercritical pressures. For highly supercritical pressures, however, the droplet temperature was shown to never reach a wet-bulb state, increasing throughout the droplet lifetime. Walther and Koumoutsakos [11] combined certain aspects of previous studies for the MD simulations of an argon droplet in its own vapor. An "adaptive tree data structure" was proposed to determine the list of neighbors surrounding each particle, in order to avoid the computation of interatomic separation distance of one atom relative to all the others.

The above investigations represent an excellent start towards the development of MD methods for predicting droplet vaporization under various conditions. However, considerable future efforts will be needed to make MD methods a reliable simulation tool, and to apply them to situations, including supercritical and deforming interfaces, that are not tractable using continuum approaches. The future efforts should also focus on simulating microscopic (of the order of microns) rather than nano-droplets being investigated here, as well as developing realistic yet tractable potential models for more complex molecules to represent practical fuels. The present study represents one such effort. It focuses on the molecular physics of submicron droplet evaporation for a new binary system: a xenon droplet evaporating in nitrogen ambient. An alternative technique to identify the droplet shape, as well as a different methodology for specifying the desired initial conditions for droplet and ambient are presented. In addition to examining the effects of various parameters on the vaporization of submicron droplets, the vaporization and shape histories of initially non-spherical droplets under subcritical and supercritical conditions are investigated.

2. Model formulation

The first step in a MD simulation is to choose the model that best represents the interaction between elementary particles. From a classical mechanics standpoint, it is commonly accepted that atoms and molecules exert intermolecular forces, which are described through a potential energy function. The present study employs the Lennard-Jones 12-6 potential [12], whose analytical form is

$$U_{ij}(r_{ij}) = 4\varepsilon_{ij} \cdot \left[\left(\frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^{6} \right] \tag{1}$$

In the above expression r_{ij} represents the intermolecular separation distance between particles i and j. Furthermore, ε_{ij} and σ_{ij} are the minimum energy and the zero energy separation distance relative to the pair. Data for ε and σ of a wide range of atoms and small molecules are widely available [13,14]. For cross interactions between different species, it is common to use mixing rules such as the Lorentz-Berthelot [15], which are given by

$$\varepsilon_{ij} = \sqrt{\varepsilon_i \varepsilon_j} \tag{2}$$

$$\sigma_{ij} = \tfrac{1}{2} \cdot (\sigma_i + \sigma_j) \tag{3}$$

The Lennard-Jones potential model used here is known to be adequate for many non-quantum monatomic fluids (Ar, Xe, Ne, etc.), as well as small diatomic and polyatomic compounds (N_2, O_2, CH_4, etc.) [13–16]. In the present study, we have chosen to examine the evaporation characteristics of Xe droplet in a N_2 ambient. We chose the Xe/N_2 system because this system allows droplet behavior to be studied over a wide range of conditions, including subcritical, transcritical, and supercritical pressures. For this system, the following LJ parameters were used: $\varepsilon_{\mathrm{Xe}} = 221 k_{\mathrm{B}}$, $\sigma_{\mathrm{Xe}} = 0.410$ nm and $\varepsilon_{\mathrm{N}_2} = 95.9 k_{\mathrm{B}}$, $\sigma_{\mathrm{N}_2} = 0.371$ nm, where k_{B} is the Boltzmann's constant. The cut-off radius was chosen to be $3\sigma_{\mathrm{Xe}}$. The critical temperatures of N_2 and Xe are 126.2 and 289.2 K respectively while the critical pressures are 33.9 and 58.4 bar respectively. This allows a rather wide temperature region for a stable liquid droplet, with no substantial condensation of the ambient gas into the droplet.

In a MD simulation, specifying the total number of particles N and the initial average number density δ for the simulated system yields the overall system volume: $V = N/\delta$, which in the present investigation is a cube. Initially all N particles are placed at the lattice sites of a faced-centered cube (FCC) in the simulation system. The liquid xenon droplet of an initial diameter D is placed at the center of the cube, surrounded by the gaseous species (nitrogen). A specified number of nitrogen molecules are then removed from their FCC nodes in the ambient (outside the droplet region) in order to establish the desired initial ambient density. For example, in a typical system studied, $N = 5662$, while the number of molecules constituting the droplet and the ambient are 3559 and 2103, respectively. It should be further noted that the volume occupied by the ambient is considerably larger than that occupied by the droplet in our simulations (cf. Fig. 1).

The numerical scheme adopted to solve the Newtonian equations of motion for every particle is based on the Gear's fifth order predictor–corrector algorithm [12]. The nature of the Lennard-Jones interaction is such that the acceleration of any one molecule, at a given instant in time, is only a function of the separation distances, r_{ij}. Therefore, setting the initial positions corresponds to setting the initial accelerations. Gear's algorithm also requires the assignment of initial velocities of all the system particles. These are assigned randomly as a Gaussian distribution based on the desired initial temperature, using the equations of statistical mechanics [12].

The conditions for the numerical algorithm to initiate the computation of particle trajectories are now specified. As the particles start to move, the xenon crystal melts, losing memory of its initial orderly structure and acquiring the structure of the liquid state. As the simulation evolves, some xenon atoms evaporate and mix with nitrogen molecules and the vapor content of the ambient increases. Due to the small sizes involved (the initial droplet diameter is taken to be 6.88 nm), distinguishing the condensed phase from the gaseous phase becomes an important challenge. The identification of the droplet is based on dynamically monitoring the profiles of liquid particle number density in the three system coordinates. Assuming the droplet to be inscribed in a regular ellipsoid, the length of its x-axis is calculated using the following procedure. Imagine slicing the system cube into forty equal slices in the x direction, with each slice housing a certain number of particles. The length of the x-axis of the droplet is then calculated according to the number of continuous slices with xenon particles greater than (or equal to) a given threshold value based on the liquid density. Additional details are provided in Ref. [17].

The time interval required by the xenon FCC crystal to become a liquid droplet will be referred to as the initialization period. During this period the crystal is left to melt and no datum is collected. Also, during this period, the velocities of both xenon and nitrogen particles are first scaled to achieve the desired initial droplet

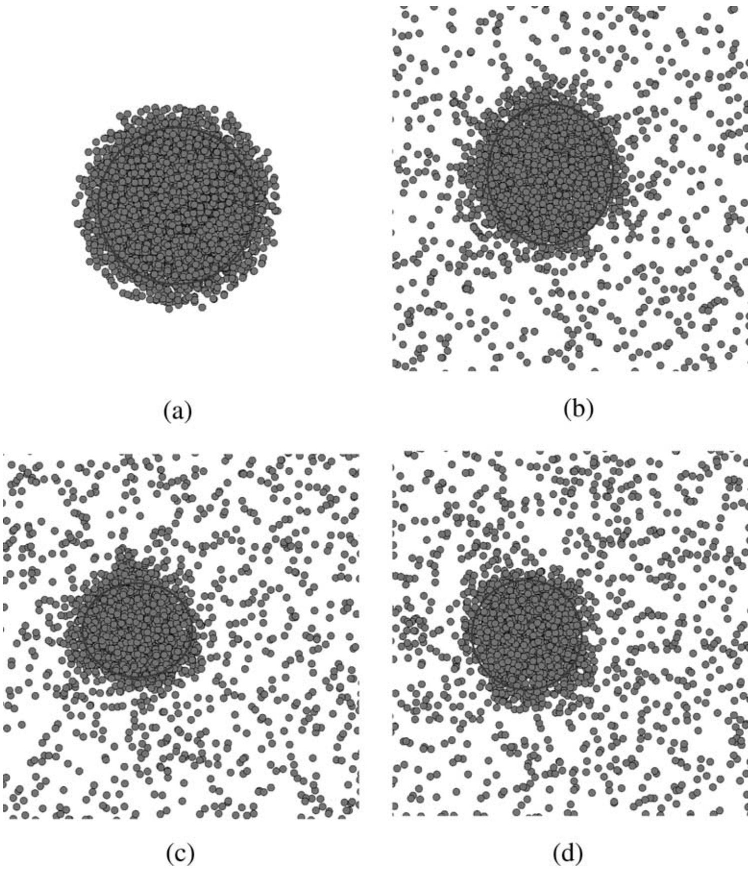

Fig. 1. Snapshots of the xenon droplet during the vaporization process at (a) $t = 0$, (b) 510 ps, (c) 926 ps and (d) 1030 ps. Only xenon molecules are shown.

and ambient temperatures, respectively, while maintaining a motionless center of mass. Details are provided in Ref. [17]. Once the velocity scaling is stopped, which typically involves 100 time steps, the system is left to adjust further for a brief time period (typically 1000 time steps). During this period, the ambient and droplet particle velocities will generally adapt to the system configuration. The end of the initialization period corresponds to the start of the production part of the simulation and, therefore, represents the origin of time.

3. Results

3.1. Droplet evaporation

In order to assess the capability of the MD model to distinguish droplet from ambient during the vaporization process, snapshots of the simulated system were recorded at different instances in time. Fig. 1 presents these snapshots or instantaneous "pictures" for a xenon droplet, with an initial diameter of 6.88 nm and initial density of 2600 kg/m^3 (2000 xenon atoms), evaporating in a subcritical nitrogen environment. The droplet configuration is shown at different instances in its lifetime, with its liquid–vapor interface identified using the procedure outlined earlier. A "crown" of vapor atoms appears to be consistently surrounding the droplet. In addition, the vaporizing droplet is shown to lose, to some extent, its spherical shape at random instances in its lifetime. An important observation here is that the algorithm developed to distinguish droplet from ambient does an excellent job, and, to a large extent, the droplet maintains its spherical shape.

3.2. Effect of initial droplet and system sizes

To assess the sensitivity of the MD simulations to the initial droplet and system size used, results were obtained for three systems of different sizes. The droplet diameters for the three systems were 3.44, 5.14, and 6.88 nm, and the corresponding number of xenon particles were 205, 783, and 2000, respectively. For all three cases, the initial ambient (nitrogen) temperature and density were 650 K and 8.5 kg/m^3 respectively. The volumes of the primary system domain were 2.6×10^3 nm^3 8.8×10^3 nm^3 and 20.8×10^3 nm^3 respectively.

Fig. 2 presents the evaporation behavior for the three cases. The number of droplet particles, normalized by the initial value, is plotted versus non-dimensional time, τ, which is defined as the actual time elapsed, t, divided by the time required for half the droplet to evaporate, $t_{1/2}$: $\tau = t/t_{12}$. Half the droplet is considered evaporated when half the initial droplet particles have migrated to the vapor phase. The simulations are essentially independent of the system size, although fluctuations are larger for the smaller system.

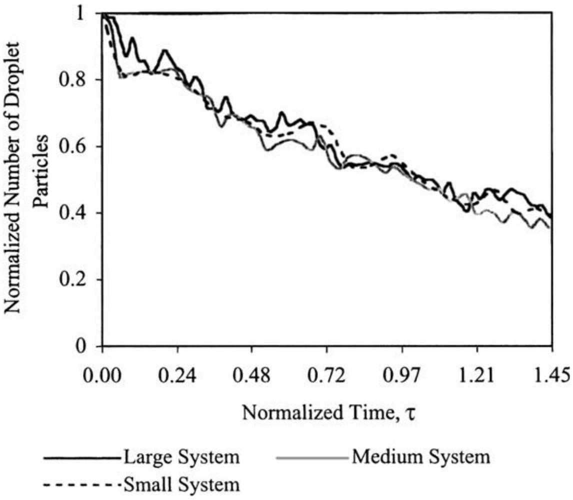

Fig. 2. Total number of droplet particles, normalized by their initial value, plotted versus the normalized time for the three systems of different initial sizes.

The *droplet sphericity* is a good measure of the tendency of a droplet to maintain a spherical shape during the vaporization process. The droplet sphericity, S, is defined as the ratio of the smallest half axis length to the largest:

$$S = \frac{\min\{a, b, c\}}{\max\{a, b, c\}} \tag{4}$$

Here a, b, c represent the three droplet axial lengths. Fig. 3 shows the temporal variation of S during the vaporization process for the three system sizes. As the system size increases, the sphericity appears to maintain a value consistently closer to unity, implying that under subcritical conditions, a submicron droplet maintains the spherical shape during its lifetime.

3.3. Vaporization history

To examine the fundamental physics of submicron droplet evaporation, the behavior of a xenon droplet was analyzed for different ambient conditions. The initial ambient density was 8.5 kg/m^3 and the initial pressures for the three cases were 26, 16 and 9 bar, corresponding to the initial ambient temperatures of 1030, 650 and 330 K respectively (after the initialization period). The initial droplet temperature was fixed at 165 K. It should be noted that the system pressure changes during the simulation due to two effects. The first is due to the decrease in ambient temperature, caused by the heat transfer from ambient to droplet and by the addition of cooler xenon vapor molecules into the ambient (Fig. 5b). This decreases the system pressure. On the

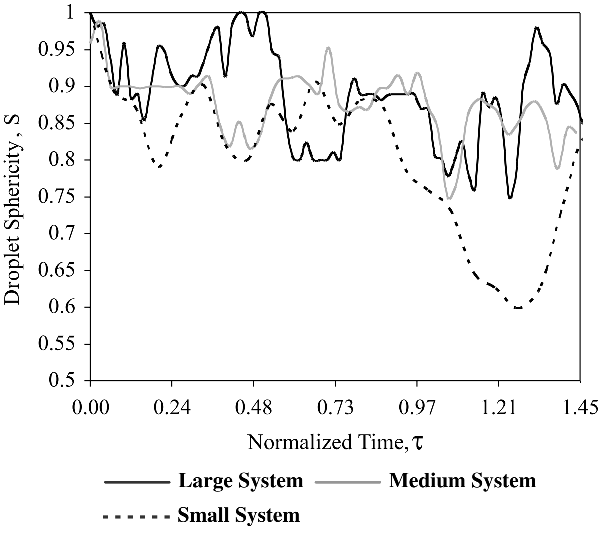

Fig. 3. Droplet sphericity plotted versus the normalized time for the three systems discussed in the context of Fig. 2.

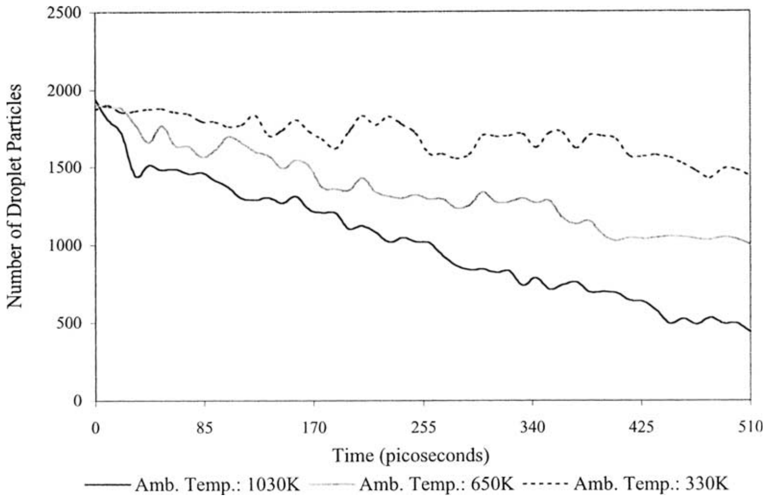

Fig. 4. Temporal evolution of the number of droplet particles for different ambient temperatures.

other hand, the system pressure increases as the ambient density increases due to the addition of xenon vapor molecules. Since the effect of ambient temperature generally dominates, the system pressure decreases during the simulation. For example, for the case with the ambient temperature of 330 K, the system pressure decreases from 9 to 7 bar during the simulation time of 500 ps.

Fig. 4 presents the vaporization history in terms of the temporal variation of the number of droplet particles for three different ambient temperatures. The corresponding droplet and ambient temperature histories are shown in Fig. 5. As expected, the evaporation process is characterized by the loss of liquid xenon particles from the droplet, and their migration to the ambient. The vaporization rate increases as the ambient temperature is increased; for example, increasing the ambient temperature from 330 to 1030 K increases the vaporization rate by a factor of more than three. The enhanced vaporization rate is directly attributable to the higher droplet temperature corresponding to the higher ambient temperature. The droplet temperature plots, presented in Fig. 5a, exhibit two distinct stages of droplet vaporization: (1) a liquid-heating stage characterized by relatively slow vaporization, and (2) a constant-liquid-temperature evaporation at the "pseudo wet-bulb temperature". It is interesting to note that a similar behavior is observed for larger or "continuum-size" droplets evaporating under subcritical conditions [1,2]. In addition, Fig. 5a indicates that both the liquid heat-up time and the pseudo wet-bulb temperature increase as the ambient temperature is increased. This behavior is also analogous to that observed in continuum-based studies dealing with larger droplets. It should also be noted that for the lowest ambient temperature case, the droplet temperature decreases with time. This is attributed to the fact that due to the relatively small temperature difference between the droplet and the ambient gas, the heat supplied from the ambient gas is less than that required for vaporization. A similar behavior has been observed in studies dealing with continuum-size droplets [18].

In Fig. 5b, we present the temporal variation of ambient temperature during the evaporation process. The ambient temperature in the context of MD simulations represents an average temperature of all the nitrogen and xenon particles that are in the ambient. The ambient temperature decreases with time due to the incoming "cooler" xenon vapor particles, and due to the transfer of energy from ambient to droplet. The behavior is similar to that observed for macro-size droplets. For the latter case, the gas film near the droplet experiences a similar decrease in temperature, and the gas-film thickness is of the order of the droplet diameter. For the MD simulations, the ambient size is of the order of the submicron droplet diameter, and its temperature decreases due to heat transfer to the evaporating droplet, and due to the addition of xenon vapor particles into the ambient.

3.4. Evaporation of an initially non-spherical droplet

One of the fundamental assumptions in the study of macroscopic droplet evaporation has been that the droplet remains spherical during its lifetime. For subcritical vaporization, especially under non-convective or mildly convective conditions, the assumption is justified since the Weber number is relatively small due to the finite surface tension. However, under transcritical and supercritical conditions, as surface tension goes to zero, the assumption of spherical shape becomes highly

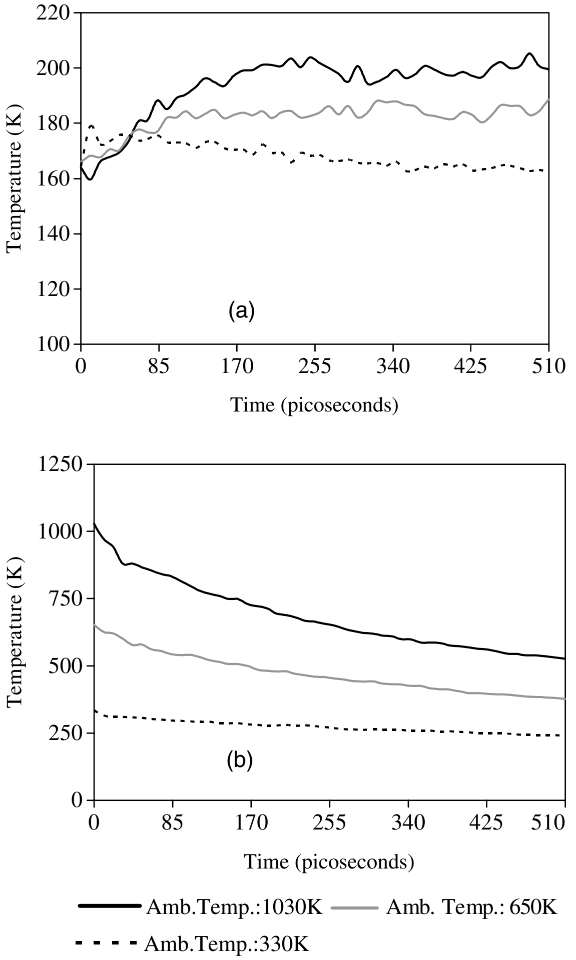

Fig. 5. Temporal evolution of (a) droplet temperature, and (b) ambient temperature for the three cases discussed in the context of Fig. 4.

questionable. In order to address this issue, MD simulations are used to study the vaporization of initially non-spherical droplets under both subcritical and supercritical conditions. An initially elliptic xenon droplet with an initial density of 2250 kg/m^3 and comprising of 1800 atoms is considered. For the subcritical case, the initial liquid temperature is 165 K, while the ambient temperature and pressure are 650 K and 16 bar respectively. For the supercritical case, the initial liquid temperature is 266 K, and the ambient temperature and pressure are 1320 K and 78 bar respectively.

Fig. 6 presents the temporal variation of sphericity (S) and temperature for an initially elliptic droplet for both the subcritical and supercritical conditions. For both cases, the initial value of S is 0.65, defined by the following half-axis values: $a \cong 4.50$ nm, $b \cong 2.90$ nm and $c \cong 3.25$ nm. For the subcritical case, the initially elliptical droplet attains the spherical shape rather early in its lifetime, and its vaporization follows the typical subcritical behavior, which has been described in the preceding section. The droplet temperature increases to the quasi wet-bulb temperature value, as indicated in Fig. 6b, with the subsequent vaporization occurring at this temperature. The rapid attainment of the spherical shape for an initially elliptical droplet under subcritical conditions is also shown by the snapshots of xenon atoms in Fig. 7. In contrast, for the supercritical case, the droplet never returns to the spherical, i.e., the minimum energy configuration. Rather, sphericity of the droplet deteriorates rapidly and noticeably, and its temperature continues to increase throughout the "vaporization" process (cf. Fig. 6). Similar variation in droplet temperature under supercritical conditions has been observed in the study of macroscopic droplets [6,19], as well as that of nano-size droplets [10].

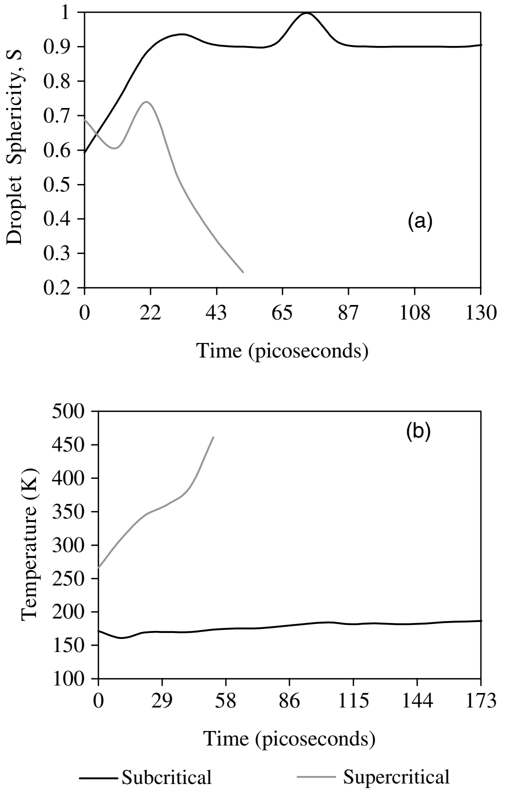

Fig. 6. Temporal variation of (a) sphericity and (b) temperature of an initially elliptical droplet evaporating under subcritical and supercritical conditions.

As discussed in the context of Fig. 6, a subcritical microscopic droplet, with a "disrupted" initial shape, has a strong tendency to recover the spherical configuration. The driving force that leads the droplet back to its traditional shape is determined primarily by the liquid temperature. To evaluate the effect of liquid temperature

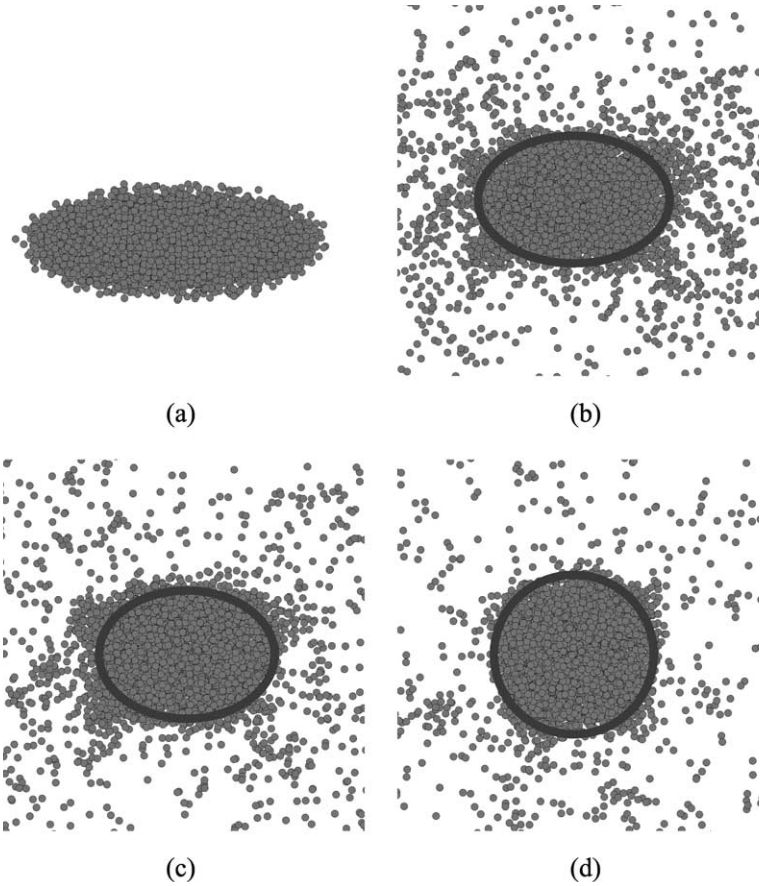
(a) (b) (c) (d)

Fig. 7. Snapshots of xenon atoms for an initially non-spherical droplet at (a) $t = 0$ ps, (b) 10 ps, (c) 16 ps and (d) 36 ps for the subcritical case. Only xenon particles are shown.

on the rate of change of droplet sphericity, the behavior of the subcritical droplet was studied under different initial conditions.

The influence of initial droplet temperature on the temporal evolution of droplet shape is presented in Fig. 8. The three droplets become nearly spherical within few picoseconds. In addition, it is observed that the tendency of a non-spherical droplet to attain a spherical shape is reduced as the initial droplet temperature is increased. For example, at $t \geqslant 50$ pico-seconds (ps), the droplet with higher initial temperature ($T = 195$ K) is less spherical than those with lower initial temperatures ($T = 165$ and 130 K). Surface tension is a measure of the overall attractive force acting on the surface, exerted by the inner bulk molecules, that causes the droplet surface to attain the spherical shape. Therefore, in an equilibrium quiescent environment, a droplet with a higher surface tension will have a greater tendency to remain spherical. Since surface tension generally decreases with increasing liquid temperature, the results in Fig. 8 are

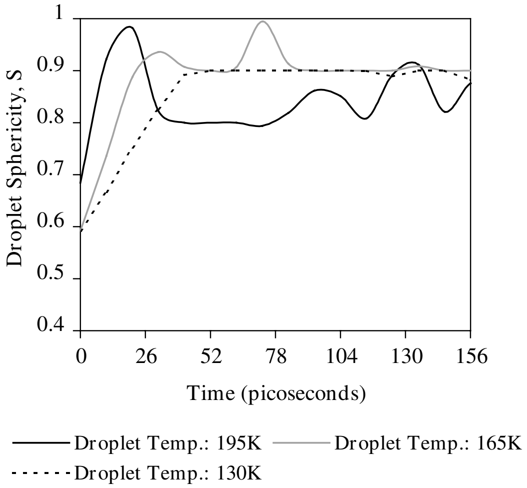

Fig. 8. Temporal variation of the sphericity of three initially elliptical droplets with different initial temperatures.

consistent with this behavior. However, for $t < 25$ ps, the results in Fig. 8 may appear to be somewhat in contrast with this behavior, but they simply represent fluctuations in the droplet shape, and the amplitude of fluctuations generally increases at higher liquid temperature as observed in our simulations.

4. Conclusions

Molecular dynamics (MD) simulations have been used to study the vaporization behavior of submicron droplets. A new methodology has been developed to specify initial conditions for the droplet and ambient, and to identify droplet shape during the vaporization process. The vaporization characteristics of a xenon droplet in nitrogen ambient have been investigated. Results have focused on (1) the sensitivity of MD simulations to the system and droplet sizes, (2) the vaporization behavior of spherical and non-spherical droplets, (3) the effects of ambient and droplet properties on the vaporization process, and (4) the tendency of a droplet to retain its spherical shape under both subcritical and supercritical conditions.

The MD simulations are shown to be essentially independent of the primary system size, although the predicted vaporization behavior shows less scatter when a larger system is employed. The predicted droplet sphericity is also shown to be higher, i.e., closer to unity with the larger system.

The evaporation process is characterized by the migration of xenon particles from the droplet to the ambient. The rate of migration or vaporization increases as the ambient temperature is increased, which is directly attributable to the higher droplet temperature corresponding to the higher ambient temperature. Analogous to the vaporization of macro-size droplets, the vaporization process associated with submicron droplets consists of a liquid-heating stage during which the vaporization rate is relatively low, followed by a nearly constant liquid-temperature evaporation at the "pseudo wet-bulb temperature".

MD simulations are also used to examine the tendency of a submicron droplet to retain its spherical shape under subcritical and supercritical conditions. The vaporization of an initially elliptic droplet is analyzed. For the subcritical case, an initially elliptical droplet attains the spherical shape rather early in its lifetime, and its vaporization follows the typical subcritical behavior. The tendency to return to spherical shape is initially (at $t \leqslant 25$ ps) higher for a droplet with higher initial temperature. This can be attributed to the highly non-equilibrium process initially and the enhanced loss of the higher energy "vertex" molecules, even though the surface tension is reduced at higher liquid temperature. At later times ($t \geqslant 50$ ps), however, the droplet with higher initial temperature becomes less spherical. For the supercritical case, the droplet never returns to the spherical configuration. Rather, sphericity of the droplet deteriorates rapidly and noticeably, and its temperature increases continuously during the "vaporization" process.

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