Cavitation in a metallic liquid: Homogeneous nucleation and growth of nanovoids

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Large-scale molecular dynamics (MD) simulations are performed to investigate homogeneous nucleation and growth of nanovoids during cavitation in liquid Cu. We characterize in detail the atomistic cavitation processes by following the temporal evolution of cavities or voids, analyze the nucleation behavior with the mean first-passage time (MFPT) and survival probability (SP) methods, and discuss the results against classical nucleation theory (CNT), the Tolman equation for surface energy, independent calculation of surface tension via integrating the stress profiles, the Johnson-Mehl-Avrami (JMA) growth law, and the power law for nucleus size distributions. Cavitation in this representative metallic liquid is a high energy barrier Poisson processes, and the steady-state nucleation rates obtained from statistical runs with the MFPT and SP methods are in agreement. The MFPT method also yields the critical nucleus size and the Zeldovich factor. Fitting with the Tolman’s equation to the MD simulations yields the surface energy of a planar interface (∼0.9 J m⁻²) and the Tolman length (0.4–0.5 Å), and those values are in accord with those from integrating the stress profiles of a planar interface. Independent CNT predictions of the nucleation rate (10³⁻³ to 10⁻³ s⁻¹ m⁻³) and critical size (3–4 Å in radius) are in agreement with the MFPT and SP results. The JMA law can reasonably describe the nucleation and growth process. The size distribution of subcritical nuclei appears to follow a power law with an exponent decreasing with increasing tension owing to coupled nucleation and growth, and that of the supercritical nuclei becomes flattened during further stress relaxation due to void coalescence. © 2014 AIP Publishing LLC.

I. INTRODUCTION  

Cavitation or the formation of bubbles in metastable liquids is of both scientific and applied interests.1–6 In order to understand cavitation in real liquids with preexisting bubbles or impurities, it would be helpful to understand homogeneous nucleation of cavities first. Liquids can be regarded as an extreme case of plastically deformed solids, and cavitation in liquids is analogous to void nucleation in solids with much more complicated phenomena and likely mechanisms. Therefore, studying liquid cavitation can also help us gain insights into damage in solids. Molecular dynamics (MD) simulations are ideal for revealing the microscopic cavitation process and underlying mechanisms since nucleation is largely initiated at sub-nm and sub-ns scales.

Cavitation is a special case of phase changes: liquid-vapor/vacuum transitions. In first-order phase transitions, spatial and temporal fluctuations in physical properties are required to overcome the inherent energy barrier to form critical nuclei of the daughter phase.7 As a result, it is extremely difficult to directly observe critical nucleation and to determine related parameters including critical nucleus size and nucleation rate,8–10 because we define a critical nucleus at the local maximum where the probability of the forward transition equals to that of the backward transition. There are different methods to assess these parameters from atomistic simulations, and two important ones are the mean first-passage time (MFPT) method and the survival probability (SP) method for Poisson processes.11,12 Despite their success in certain processes including condensation, it would be useful to apply and compare these methods in cavitation.

Cavitation is a fluctuation-driven process and can be described with classical nucleation theory (CNT).7,13–18 Molecular simulations and statistical theories have been attempted within the context of CNT.19–27 However, direct application of CNT to cavitation is difficult due to the lack of reliable knowledge on such parameters as surface tension and prefactor. For instance, assuming a constant value of surface tension (equal to that for a planar surface) may underestimate nucleation rates with CNT.19,20,28 A recent work considered the Tolman length effect and found agreement in nucleation rate between CNT and MD simulations of cavitation in a binary metallic liquid, but with inevitable assumptions on some parameters including critical size.29

To characterize nucleation parameters including critical size, the Tolman length and nucleation rate, and to connect simulations to transition state and nucleation theories with minimum assumptions on certain parameters, we choose a representative metallic liquid (Cu), and perform MD simulations of cavitation under isotropic tension. We describe in
detail the atomistic cavitation processes by following the temporal evolution of cavities or voids, analyze the nucleation behavior with the MFPT and SP methods, and discuss the results against CNT, the Tolman equation for surface energy, independent calculation of surface tension via integrating the stress profiles, the Johnson-Mehl-Avrami (JMA) growth law,30–32 and the power law for nucleus size distributions.10

Our work shows agreement between the MFPT method, SP method, and CNT, and consistency in surface energy is found for different methods. Section II addresses the methodology of MD simulations, void analysis, and the MFPT and SP methods. We present the results and discussion in Sec. III and main conclusions in Sec. IV.

II. METHODOLOGY

We use the embedded atom method (EAM) potential33 to describe the atomic interactions in Cu. This potential has been fitted to reproduce physical properties such as stacking fault energy and elastic moduli,33 and widely used in a large number of simulations, including melting curve and equations of state.10,34 Our MD simulations employ the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS).35 We apply the constant temperature-pressure (NPT) ensemble for constructing the initial liquid configurations, and the constant volume-temperature ensemble (NVT) for cavitation simulations. All simulations are conducted under three-dimensional (3D) periodic boundary conditions.

The time step for integrating the equation of motion is 1 fs. Temperature is controlled with a Hoover thermostat. The initial liquid Cu configurations are obtained by melting single crystal Cu at zero pressure, and then equilibrated at 1400 K for cavitation simulations. The liquid configurations of a cubic shape are subjected to an incremental, isotropic, tensile strain of $6 \times 10^{-4}$ every 1000 steps. Two system sizes are attempted of 864 000 atoms and 2 916 000 atoms. Discussions refer to the smaller system size unless noted otherwise.

There are different methods in quantifying nucleation of a daughter phase (nanovoids) from a parent phase (Cu liquid), and here we explore the survival probability method12 for Poisson processes, and the mean first-passage time method.10,11 Nucleation rate can be obtained via the SP method, and nucleation rate, critical nucleus size, and the Zeldovich factor, with the MFPT method. Both methods require numerous independent runs for sufficient statistics. For each statistic run, we change the random number seed for assigning initial velocities in NVT simulations.

In the presence of a high cavitation energy barrier, the appearance of a supercritical cluster or nucleus becomes a random event, whose probability is described by a Poisson distribution

$$P_k(t) = \frac{1}{k!} \left( \frac{t}{\tau_n} \right)^k \exp \left( -\frac{t}{\tau_n} \right).$$  \hspace{1cm} (1)

Here, $P_k(t)$ denotes the probability for $k$ clusters of size larger than $n$ to appear at time $t$, and $\tau_n^{-1}$ is the average appearance rate of these clusters. Instead of calculating $P_k(t)$ literally, it is equivalent but easier to characterize the random nucleation events with the probability for a system not to have nucleated any clusters of size $n$ after a time $t$, i.e., the survival probability ($P_s$) of the metastable parent phase (liquid). For a fully random process, it is given by

$$P_s(t) = P_{k=0}(t) = \exp \left( -\frac{t}{\tau_n} \right).$$  \hspace{1cm} (2)

Monitoring the evolution of $P_s$ can thus provide a straightforward way to evaluate the nucleation rate. In our simulations, $P_s$ is obtained as the ratio of the number of runs without any nuclei beyond size $n$ to the total number of runs. $\tau_n$ is essentially $\tau_J$ in the MFPT method described below.

The steady-state nucleation or barrier-crossing rate $J$, the critical nucleus size $n^*$, and the Zeldovich factor $Z$, can be evaluated with the MFPT method.11,36,37 Here, we follow the general MFPT methodology37 which has been presented and applied to different physical processes by Wedekind and co-workers,11,38,39 Bartell and Wu,40 and Zheng et al.10 For most activated processes, such as nucleation of cavities/voids (daughter phase) from a liquid (parent phase), the dynamics can be described by the Fockker-Planck equation41

$$\frac{\partial P(x, t)}{\partial t} = \frac{\partial}{\partial x} \left[ D_0 e^{-\frac{U(x)}{k_B T}} \frac{\partial}{\partial x} \left( P(x, t) e^{\frac{U(x)}{k_B T}} \right) \right].$$  \hspace{1cm} (3)

Here, $x$ is the reaction coordinate, $D_0$ represents the generalized diffusion coefficient, $k_B$ is the Boltzmann constant, and $U(x)$ denotes the excess Gibbs free energy. $P(x, t)$ is the probability for the system sitting at a specific $x$ for a given time $t$. We define the mean-first passage time, $\tau(x_0; a, b)$, as the average time consumed for the system initially at state $x_0$ to leave the domain $[a, b]$ for the first time. For reflecting boundary point $a$ and absorbing boundary $b$, the MFPT is expressed as follows:37

$$\tau(x_0; a, b) = \int_{x_0}^{b} \frac{1}{D_0} e^{\frac{U(x)}{k_B T}} dy \int_{a}^{x} e^{-\frac{U(y)}{k_B T}} dy.$$  \hspace{1cm} (4)

With fixed $x_0$, $\tau(x_0; a, b)$ simplifies to $\tau(b)$. For an energy barrier of sufficient height, $\tau(b)$ adopts a sigmoidal shape, and a transition state appears at the inflection point $x^*$. Since the MFPTs are significantly longer than the characteristic diffusion time $1/D_0$, we approximate

$$\frac{\partial^2 \tau(b)}{\partial b^2} \bigg|_{b=x^*} \approx 0.$$  \hspace{1cm} (5)

We then have

$$\tau(b) = \frac{\tau_J}{2} \left[ 1 + \text{erf}((b - x^*) c) \right].$$  \hspace{1cm} (6)

$\tau_J$ is proportional to the inverse of the steady-state transition rate ($J^{-1}$), and $c$ denotes the local curvature at the top of the barrier

$$c = \sqrt{\frac{|U''(x^*)|}{2k_B T}},$$  \hspace{1cm} (7)

and the Zeldovich factor $Z = c/\sqrt{\pi}$.

In the case of cavitation, $b$ corresponds to the size of the largest void nucleus ($n_{\text{max}}$), $x^* = n^*$, and $J = (\tau_J V)^{-1}$ ($V$ $\equiv$ volume). We obtain $\tau(b) = \tau(n_{\text{max}})$ via averaging among $N$ independent runs. We calculate $n_{\text{max}}(t)$ for each run with
III. RESULTS AND DISCUSSION

Our main purpose is to characterize homogeneous void nucleation and growth process in a representative metallic liquid at atomistic scales. In this method, we first divide the simulation supercell into numerous regularly spaced grids in three dimensions. For a given grid point, if there is a Cu atom within a distance less than \( d_{\text{cut}} \) from it, this grid point is excluded from the list of void voxels; otherwise, it is counted as a void voxel. \( d_{\text{cut}} \) and the grid width are about 0.2 nm in our simulations, and a void voxel is comparable to a vacancy in size. Two void voxels belong to the same cluster (void nucleus) if they are within the nearest-neighbor distance of each other. The number of void voxels contained in a cluster is void size, \( n \).

A void analysis method is used to characterize void (cavity) nucleation and growth at atomistic scales. In this method, we first divide the simulation supercell into numerous regularly spaced grids in three dimensions. For a given grid point, if there is a Cu atom within a distance less than \( d_{\text{cut}} \) from it, this grid point is excluded from the list of void voxels; otherwise, it is counted as a void voxel. \( d_{\text{cut}} \) and the grid width are about 0.2 nm in our simulations, and a void voxel is comparable to a vacancy in size. Two void voxels belong to the same cluster (void nucleus) if they are within the nearest-neighbor distance of each other. The number of void voxels contained in a cluster is void size, \( n \). The trajectories are recorded every 1 ps. The \( n_{\text{max}}(t) \) curves of all these runs are similar to each other, except that the rapid increase in \( n_{\text{max}} \) occurs at different instants, i.e., with different waiting periods (Fig. 3). Note that \( n = 1 \) corresponds to the average size of a liquid Cu atom, or a single vacancy.

The MFPT \( (\tau) \) is obtained by averaging over 100 runs (Fig. 4). \( T(n_{\text{max}}) \) shows a sigmoidal shape but without a well-defined plateau. As pointed previously, this indicates that the growth rate of a critical nucleus is limited and its growth occurs at similar time scales as its nucleation, i.e., nucleation is coupled with growth. In such cases, Eq. (6) is still valid.

![Figure 1. Snapshots of void nucleation and growth during isotropic tension. Only atoms with coordination number (CN) \( \leq 5 \) are plotted, and nanovoids are enclosed within these atoms. Red, gray, and green refer to atoms with CN = 5, 4, and 3, respectively.](image)

![Figure 2. Pressure evolution during isotropic tension and cavitation at 1400 K.](image)
though. Fitting to $\tau(n_{\text{max}})$ with Eq. (6) yields $n^* = 5.74$ which corresponds to a radius of $r = 2.7$ Å, $\tau_J = 61.86$ ps, and $c = 0.42$. It follows that $J = 1/(\tau_J V) \approx 1.25 \times 10^{33}$ s$^{-1}$ m$^{-3}$, and $Z = 0.24$.

In the survival probability or SP method, the probabilities are independent of which cluster size $n$ is chosen as a threshold if the energy barrier is sufficiently high. So we regard a system as “nucleated” whenever the largest cluster in it exceeds an arbitrarily chosen size, $n = 10$. The survival probability curve $P_s(t)$ is obtained from the same 100 independent runs as in the MFPT method, and fitting to $P_s(t)$ with Eq. (2), yields $\tau_J = 62.38$ ps (Fig. 5), which is in accord with the MFPT result $\tau_J = 61.86$ ps. This agreement indicates that both methods are accurate for evaluating the rate of homogeneous nucleation [$J = 1/(\tau_J V)$].

It would be interesting to compare the critical nucleation results obtained with the MFPT and SP methods against independent, CNT predictions. A required parameter in CNT is the surface energy ($\gamma$). For a sufficiently large void in a steady state where the void is in equilibrium with the surrounding liquid, $\gamma$ can be calculated from the Young-Laplace equation

$$\gamma = \frac{\Delta P r}{2},$$

(9)

where $\Delta P$ is the pressure difference across the cavity surface, and $\Delta P = |P|$ since the internal pressure within a void is essentially zero for liquid Cu; $r$ is the radius of the cavity at a steady state. We therefore construct voids of different radii within the liquid Cu at 1400 K, equilibrate the system, and then measure the steady-state cavity size and stresses to calculate the corresponding $\gamma$. The results of $\gamma$ for different cavity sizes are shown in Fig. 6 (circles). Fitting with the Tolman's equation,

$$\gamma(r) = \frac{\gamma_0}{1 + \frac{\delta}{r}},$$

(10)

yields the Tolman length $\delta = 0.53$ Å, and the surface energy for a planar surface $\gamma_0 = 0.911$ J/m$^2$.

Given the importance of surface energy, we calculate independently $\gamma_0$ using the method by Nijmeijer et al. A 3D system with coexisting liquid and vacuum is constructed at 1400 K. The free surfaces of the liquid Cu slab are parallel to the $xy$-plane, and their normals are along the $z$-axis. Planar surface tension can be obtained with

$$\gamma_0 = \frac{1}{2} \int_0^{L_z} [\sigma_n(z) - \sigma_t(z)] dz.$$  

(11)

Here, $L_z$ denotes the box length of the $z$-axis, $\sigma$ is stress, and subscripts $n$ and $t$ refer to normal and tangential, respectively.
After equilibration, the density and stress profiles along the \( z \)-axis are measured [Fig. 7(a)]. If we divide the liquid slab along the \( z \)-axis into \( N_b \) bins, Eq. (11) can be rewritten as

\[
\gamma_0 = \frac{1}{2} \sum_{k=1}^{N_b} \gamma(k),
\]

with

\[
\gamma(k) = \frac{L_z}{N_b} \left\{ \sigma_{zz}(k) - \frac{1}{2} [\sigma_{xx}(k) + \sigma_{yy}(k)] \right\}.
\]

We thus obtain \( \gamma_0 \approx 0.903 \text{ J/m}^2 \), in excellent agreement with 0.911 J/m\(^2\) from fitting with the Tolman’s equation. This lends support to the methodology of obtaining \( \gamma_0 \) and \( \delta \) by fitting the Tolman’s equation to simulations of voids with different sizes.

In classical nucleation theory, the driving force for nucleation of a spherical void is

\[
\Delta G = 4 \pi r^2 \gamma - \frac{4}{3} \pi r^3 |P|,
\]

where the first (surface) term is the free energy gain due to surface tension of a cavity and the second (bulk) term is the reduction while creating the cavity. In CNT, one often assumes a step-function-like interface (\( \delta = 0 \) or \( \delta \ll r \)). However, considering the Tolman length is necessary. The radius of the critical nucleus \( r^* \) satisfies

\[
\left( \frac{1 + \frac{3 \delta}{r^*}}{1 + \frac{2 \delta}{r^*}} \right)^2 r^* = 1,
\]

where \( r^*_0 = 2 \gamma_0 / |P| \) for \( \delta = 0 \). Using values obtained above, i.e., \( \delta = 0.53 \text{ Å} \) and \( \gamma_0 = 0.911 \text{ J/m}^2 \), we have \( r^* = 3.44 \text{ Å} \). For the critical radius, the surface energy is 0.7 J/m\(^2\) according to Eq. (10).

\( \Delta G \) peaks at the critical value \( \Delta G^* \) with the critical radius \( r^* \). From Eq. (14), we have \( \Delta G^* = 1.71 \text{ eV} \) for \( P = -4.52 \text{ GPa} \) and \( T = 1400 \text{ K} \). Given \( \Delta G^* \), CNT allows us to estimate the nucleation rate at the critical size with

\[
J = J_0 \exp \left( -\frac{\Delta G^*}{k_B T} \right),
\]

where \( J_0 \) is a prefactor depending on several parameters. In Bogach and Utkin’s work, \( J_0 \) is expressed as

\[
J_0 = \frac{N \gamma}{V \eta} \sqrt{\frac{\gamma}{k_B T}}.
\]

Here, \( N \) denotes the total number of Cu atoms within a volume \( V \), and \( \eta \) is the viscosity. \( \gamma \) is 3.74 mPa s for liquid Cu at \( T = 1400 \text{ K} \). We therefore obtain \( J_0 \approx 7 \times 10^{46} \text{ s}^{-1} \text{ m}^{-3} \), and \( J = 4 \times 10^{34} \text{ s}^{-1} \text{ m}^{-3} \) from Eq. (16). The nucleation rate obtained from CNT here is comparable to those from the MFPT and SP methods; the CNT result is slightly higher by approximately one order of magnitude, but such a discrepancy is reasonable as discussed below.

Given the functional form of nucleation rate in CNT [Eq. (16)], a slight variation in \( \Delta G^* \) may be amplified and propagated into \( J \). \( \Delta G^* \) is also sensitive to surface energy \( \gamma \). Thus, a small error in \( \gamma \) may cause a drastic difference in the calculated nucleation rate. For example, assuming \( \gamma_0 = 0.903 \text{ J/m}^2 \) as obtained from the stress profiles [Eq. (11)], we obtain \( \delta = 0.42 \text{ Å} \) and \( \gamma = 0.73 \text{ J/m}^2 \) near the transition point (153 ps). The nucleation rate can then be calculated from CNT as \( J \approx 4 \times 10^{33} \text{ s}^{-1} \text{ m}^{-3} \) from Eq. (16), which is close to the MFPT and SP results (1.25 \times 10^{33} \text{ s}^{-1} \text{ m}^{-3} ).

The critical nucleus \( r^* \) is also an important parameter to be elaborated here. \( r^* = 2.7 \text{ Å} \) obtained from the MFPT method is smaller than that from CNT (3.44 Å). However, \( r^* = 2.7 \text{ Å} \) from MFPT only accounts for strict void space without considering the diffuse interface or transition zone between the void and the liquid. In order to find a characteristic length of the transition zone, we analyze the liquid-cavity interface profiles for the planar interface (Fig. 7). The density \( \rho \) profile can be described by a sigmoidal function,

\[
\rho(z) = \rho_{\min} + \frac{\rho_{\max} - \rho_{\min}}{2} \left[ 1 - \text{tanh} \left( \frac{z - z_0}{2w} \right) \right],
\]

where \( z_0 \) is the center of the interface, \( z \) is the distance, and \( w \) denotes a characteristic interface length scale. Fitting to the simulation results with Eq. (18) yields \( w = 0.65 \text{ Å} \) for the planar interface at 1400 K. If we simply add the characteristic length scale \( w \) to \( r^* = 2.7 \text{ Å} \) from MFPT, \( r^* \) becomes 3.35 Å, in excellent agreement with the CNT value (3.44 Å). If we take the 10%-90% width of the interface (4.394w = 2.86 Å) as the interface thickness, then the corrected \( r^* \) is \( \sim 4 \text{ Å} \), with half the interface thickness (1.4 Å) considered. The critical sizes as obtained from CNT and MFPT are in a reasonable agreement. We also fit the differential stress profile \( \sigma_{n-t} \) with the Gaussian function,

\[
\sigma_{n-t}(z) = A \exp \left[ -\frac{(z - z_0)^2}{2 \mu^2} \right],
\]

where \( A \), \( z_0 \), and \( \mu \) are fitting parameters. \( \mu \) is the standard deviation representing the width of the Gaussian, and \( \mu = 2w \approx 1.35 \text{ Å} \) (Fig. 7). The differential stress peaks when density nearly reaches its maximum value. Thus, either \( \mu \) from the stress profile or \( w \) from the density profile can be
used to characterize the interface thicknesses. Interestingly, \( w \) (0.65 Å) is close to the Tolman length \( \delta \), and may be used to estimate the latter.

The phenomenological JMA law is often utilized to describe the growth dynamics of a daughter phase (nanovoids) from its parent phase (liquid Cu). In our work, the JMA law relates the growth time to void volume fraction \( f_V \),

\[
f_V(t) = f_m[1 - \exp(-k t^n)],
\]

where \( f_m \) is the maximum volume fraction of cavity prescribed by the stress and temperature conditions, \( k \) is a prefactor, and \( n \) is the characteristic growth exponent. For 3D linear growth of noncontact and fixed-number spherical nuclei, we have \( n = 3 \), while \( n = 4 \) and \( n > 4 \) for fixed-number spherical nuclei, a constant, and an increasing nucleation rate, respectively. Fitting to the late stages of the simulations with the JMA law yields \( n \sim 7 \) (Fig. 8).

To better understand the mechanism of critical nucleation, we examine the size distribution of subcritical nuclei as well as its evolution at different stages of cavitation. We thus perform MD simulations of cavitation in a much larger system (2 916 000 atoms), and plot the size distribution of all the nuclei including isolated individual void voxels in logarithm-logarithm scales (Fig. 9). The distributions of small nuclei follows a power law at different times of cavitation, i.e.,

\[
F(n) = \mathcal{F}_0 n^{-\alpha},
\]

where \( \mathcal{F} \) denotes the multiplicity or frequency of void nuclei of size \( n \). The exponent \( \alpha \) is in the range of 3–6, greater than 2.2 predicted from 3D percolation model, likely because of homogeneous, independent nucleation events in the early stages. \( \alpha \) decreases with increasing time and tension. This implies that the driving force or tension affects the \( \alpha \) exponent directly. When the tension is relatively low, small nuclei form via fluctuations, leading to larger \( \alpha \); increasing tension assists the growth of supercritical nuclei and their coalescence, resulting in smaller \( \alpha \). The deviation from the power law and flattening for supercritical nuclei at later stages could be caused by finite size effect in MD simulations, or due to coalescence-dominated growth with reduced nucleation under stress relaxation.

**IV. CONCLUSION**

MD simulations of homogeneous nucleation and growth of nanovoids during cavitation are conducted on liquid Cu. Cavitation in this metallic liquid is a high energy barrier Poisson process, and the steady-state nucleation rates obtained with the MFPT and SP methods are in agreement. In addition, the MFPT method yields the critical nucleus size and the Zeldovich factor. Via fitting the Tolman’s equation to the MD simulations, we obtain the surface energy of a planar interface (~0.9 J m\(^{-2}\)) and the Tolman length (0.4–0.5 Å); those values agree with those from integrating the stress profiles of a planar interface. Independent CNT predictions of the nucleation rate \( (10^{33–34} \text{s}^{-1} \text{m}^{-3}) \) and critical size \( (3–4 \text{Å} \text{in radius}) \) are in accord with those from the MFPT and SP methods. The JMA law can reasonably describe the nucleation and growth process. The size distribution of subcritical and small nuclei appears to follow a power law with an exponent decreasing with increasing tension due to coupled nucleation and growth, and that of the supercritical nuclei becomes flattened during further stress relaxation owing to void coalescence.

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