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STABILITY OF AIR NUCLEUS IN LIQUID WATER AND CAVITATION INCEPTION ON MARINE ENGINEERING

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ABSTRACT

The micro air nucleus widely distributed in the ocean is a necessary condition for the cavitation of hydraulic machinery in seawater. In order to study the stability of air nucleus in seawater and cavitation inception, the computational domain of water molecules with air nucleus was studied using the method of molecular dynamics simulation, and the transient characteristics of air nucleus in liquid water were obtained. The key factors influencing nuclei stability were analyzed. The results showed that air nucleus with a certain mass could maintain the dynamic equilibrium in liquid water. The internal density of air nuclei had a critical value that allowed the nuclei to stably exist in water. The air nuclei mass was the decisive factor in its equilibrium volume in water, and the two were positively correlated. The internal density of air nuclei was negatively correlated with the nuclei radius when the nuclei was stable in water. Liquid surface tension was an important factor affecting the stability of the air nuclei. The larger the initial radius of nuclei, the smaller the water pressure, and the more likely the cavitation occurs

Keywords: Air nuclei, Water, Molecular dynamics, Stability, Surface tension

INTRODUCTION

In the field of marine engineering, cavitation is widespread in the operation of hydraulic machinery such as propellers and underwater vehicles, which affects the safety and stability of equipment operation. However, for a long time, many scholars have focused on the macroscopic level of cavitation research, but less on the microscopic mechanism of cavitation. The cavitation phenomenon is a microscopic to macroscopic transient development process. Therefore, in-depth study of the cavitation microscopic mechanism is a necessary means to improve the understanding of the nature of cavitation. In the ocean, the micro air nucleus are widely distributed, and the micro air nuclei as a typical cavitation nucleus is a necessary condition for the cavitation of hydraulic machinery in seawater. Micro nuclei research has profound implications for surface science, fluid dynamics, biological sciences, and some applications [1-2]. In recent decades, the nuclei in the liquid can exist stably has been the focus of academic debate [3-6]. As early as 1950, Epstein and Plesset demonstrated that free nucleus cannot be stably present in stationary fluids. They either float under buoyancy and escape from free surface or dissolve gradually under surface tension [7]. However, at present the theory and experiment of cavitation are based on the existence of air nuclei, which is called "nuclei paradox"[8]. Therefore, people have envisioned a variety of cavitation kernel models to avoid this contradiction [9-10], but all kinds of models have their own flaws and limitations. In order to study the microscopic mechanism of gas nuclei growth, collapse and stability in liquid, people tried to observe the gas nuclei in water by experiments and microscopy [11-12]. However, nanoscale micro gas nucleus are difficult to observe directly through experiments. More recently, molecular dynamics simulation has become a powerful tool for studying microscopic nuclei. The researchers studied the gas nuclei stability in the liquid from the aspects of the density and volume of the gas nuclei and the surface tension of the liquid.

The calculation results of Zhang L et al. [14] showed that the nano nuclei with high internal density had a longer lifetime in molecular dynamics simulation.

Yoshida-Honmachi et al. [13] studied the growth and collapse of cavity in water by using molecular dynamics method under NVT ensemble. It was found that the stability of the nano cavity in the water has a certain relationship with the size of the cavity. Shin-ichi Tsuda et al. [17] studied the nucleation of L-J fluid under NVE ensemble and found that there was a critical radius of nuclei growth. The nuclei larger than the critical radius spontaneously grew, whereas the nuclei smaller than the critical radius tended to disappear. Takenori Yamamoto et al. [15] calculated that the critical radius of the helium nano-nucleus in water is 1 nm.

Yoshida-honmachi et al. [13] used the Y-L equation to estimate and found that the surface tension of water has no obvious correlation with the radius of the cavity; Takenori Yamamoto et al. [15] used NAMD software to simulate and calculate the surface tension of liquid helium relative to the gas nuclei radius is a convex function; Yan Hong [16] used molecular dynamics to study the surface tension of the argon. The results show that the surface tension of the argon decreases with the increase of the nucleus radius; Dupont V et al. [18] found the relationship between surface tension and the nuclei radii of curvature has a decisive influence on the stability of the nano nucleus.

It can be seen that although the researchers believe that the surface tension of the liquid, the volume and density of the gas nuclei are factors affecting the stability of the nuclei in the liquid, there is no final conclusion in this research field, and even some research results are mutually contradictory.

In the field of microscopic cavitation,Yijin Mao et al. [19] used molecular dynamics simulation with TIP3P water molecule model to show that the hydrodynamic model based on the Rayleigh-Plesset equation was ineffective in predicting the growth and annihilation of nano nucleus in liquid water. The conclusion of previous studies based on molecular dynamics has not yet established a reliable connection between micro-phenomena and macroscopic experiments. This paper attempts to explore the microscopic phenomena of coexistence of nano air nuclei and liquid water, and further reveals its regular characteristics connected with the macro phenomenon.

SIMULATION METHOD

The model used in this paper was to take a cubic region as the computational domain in an infinite space and apply periodic boundary conditions around it. Considering the calculation time and accuracy and other factors, the length of the model in the x, y, z direction was taken as 5.3nm. The air nuclei was set in the center of the calculation domain, and the initial radii of the air nuclei were set as $R_0 = 0.8$ nm, 1.0 nm and 1.2 nm, respectively, and the density of water molecules outside the air nuclei was 0.997 g/cm^3. The model section was shown in Figure 1. Air is a mixture of substances, of which about 78% of nitrogen, oxygen accounts for about 21%. In this simulation, the material used was 79% nitrogen and 21% oxygen molecules, ignoring other trace components. The velocity initialization satisfied the Maxwell-Boltzmann distribution.

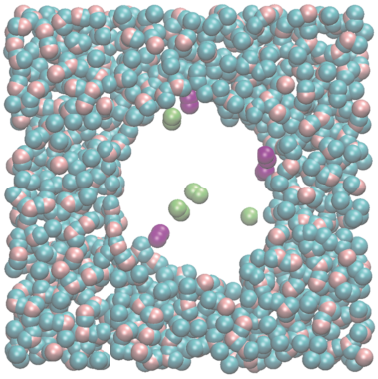


Fig.1. System initial state

In this paper, the potential energy of molecules was taken into account Coulomb force, intermolecular force and so on. The potential energy function between molecules a and b can be seen as the sum of the potential energy of Lennard-Jones and Coulomb force. The specific expression is as follows [20],

$$U_{ab} = \sum_{i}^{a}\sum_{j}^{b}\frac{k_c q_{a_i} q_{b_j}}{r_{a_i b_j}} + \sum_{i}^{a}\sum_{j}^{b} 4\varepsilon_{a_i b_j}\left[\left(\frac{\sigma_{a_i b_j}}{r_{a_i b_j}}\right)^{12} - \left(\frac{\sigma_{a_i b_j}}{r_{a_i b_j}}\right)^{6}\right] \tag{1}$$

Where *a* and *b* denote two different molecules, subscripts i and j denote two different atoms, kc is the electrostatic constant, and the potential well depth ε and the characteristic length σ are potential energy parameters. The calculation software adopted the open source software Lammps, and the water molecule adopted the TIP4P model. Newton's equations of motion were solved numerically using the leap frog algorithm. The long range force calculation used particle-particle-particle-mesh (PPPM) algorithm [21], The PPPM solver maps the water molecule's hydrogen and oxygen atoms charge in a 3D mesh then uses 3D fast Fourier transforms to solve the Poisson equation on each mesh. Finally, it interpolates from the grids to calculate the electric field and corresponding long-range Coulomb force on each charged atom [22-24].

The main parameters of the simulation were as follows: mass of nitrogen atom of 14.007 g/mol, mass of oxygen atom of

15.999 g/mol, mass of hydrogen atom of 1.008 g/mol and time step of 0.1 fs. The charge amount of an oxygen atom in the water molecules was -1.1128*e*, the charge amount of a hydrogen atom was 0.5564*e*. The cut off radius of L-J potential function was 0.85nm. The length parameter σ and the energy parameter ε of the L-J potential function were differentiated according to the atoms that acted. $\sigma_{O1\text{-}O1}$ =0.31589nm,$\varepsilon_{O1\text{-}O1}$ =0.1852 Kcal/mol when the oxygen atoms in the water molecules interact with each other; $\sigma_{N\text{-}N}$ =0.3501nm,$\varepsilon_{N\text{-}N}$ =0.1670 Kcal/mol when the nitrogen atoms interact with each other; $\sigma_{O2\text{-}O2}$ =0.2860nm, $\varepsilon_{O2\text{-}O2}$ =0.2280 Kcal/mol when the oxygen atoms in the oxygen molecules interact with each other [25]. Since the Van der Waals radius of the hydrogen atoms is much smaller than the van der Waals radius of the oxygen atoms, the Van der Waals interaction range of H-H and H-O is obscured by the range of O-O interaction, so we ignore Van der Waals interaction between H-H. The interaction parameters ε_{ij} and σ_{ij} for different atoms was given by the Lorentz-Berthelot formula [17]:

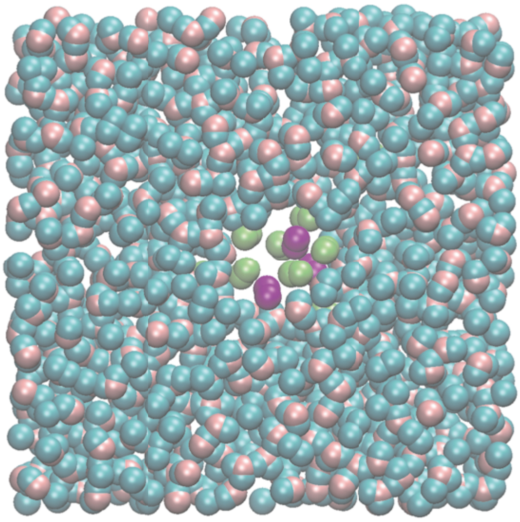
$$\sigma_{ij} = \frac{1}{2}\left(\sigma_i + \sigma_j\right) \tag{2}$$

$$\varepsilon_{ij} = \sqrt{\varepsilon_i \cdot \varepsilon_j} \tag{3}$$

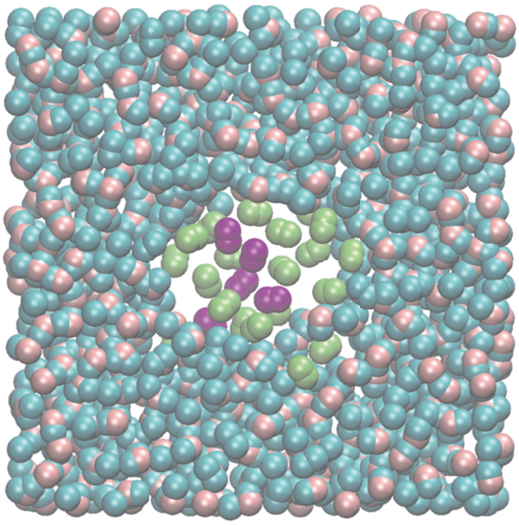
AIR NUCLEUS CHANGE IN VOLUME IN WATER

The purpose of this molecular dynamics calculation was to simulate the process of nanoscale air nuclei that was not dissolved in liquid water in nature. In the past, molecular dynamics studies used NVT ensemble (NVE) to study the gas nuclei in liquids [26]. However, the author believes that the volume-invariant system cannot reflect the real process of gas nuclei change, because the volume change rate of liquid is very small. When the total volume of liquid and gas is defined, the volume of gas also tends to remain stable, which is inconsistent with the actual situation. In the simulation process of this paper, the Nose/Hoover hot bath method was adopted to keep the temperature of the system constant [27]. At the same time, the liquid water was controlled under pressure, and the pressure was set to 1atm. In this process, the total system volume and the air nuclei volume are free to change.

a) Air nuclei contraction



b) Air nuclei stability existence



c) Air nuclei expansion

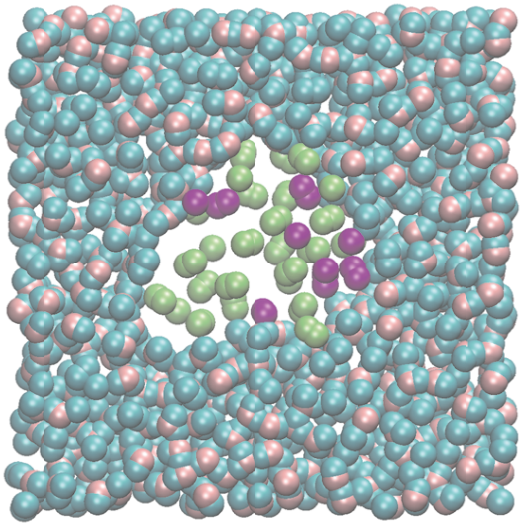
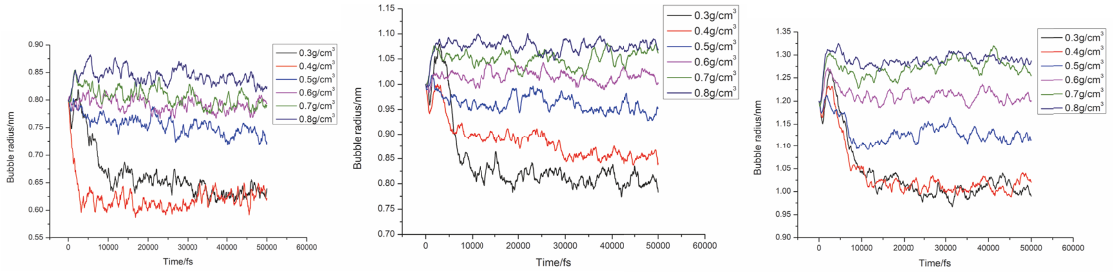
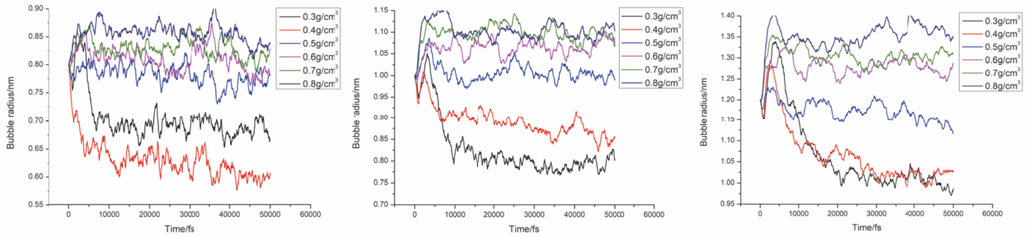


Fig. 2. Three states of air nucleus in liquid water



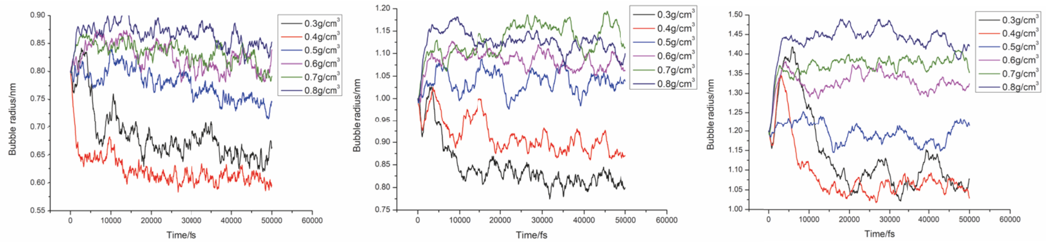
a) initial radius of 0.8 nm *b) initial radius of 1.0 nm* *c) initial radius of 1.2 nm*

T=298K



d) initial radius of 0.8 nm *e) initial radius of 1.0 nm* *f) initial radius of 1.2 nm*

T=348K



g) initial radius of 0.8 nm *h) initial radius of 1.0 nm* *i) initial radius of 1.2 nm*

T=388K

Fig.3. Variations of air nucleus with different initial radii

Voronoi tessellation method was used in this paper to estimate the size of air nuclei, that is, define a computation that calculates the volume of the Voronoi cell around each atom. Any point in an atom's Voronoi cell is closer to that atom than any other. The number of faces of the Voronoi cell is equal to the number of nearest neighbors of the central atom. In this way, the sum of volume of the Voronoi cell around nitrogen atoms and oxygen atoms in the air molecules was added to obtain a value V, and we know the radius of the nucleus R0 at the initial moment, so that the radius of the nuclei at any moment can be deduced:

$$R = R_0 * \left(\frac{V}{V_0}\right)^{1/3} \tag{4}$$

V_0 is the initial volume of all air molecules.

Observe the change process of the air nucleus, we can find that there were three states of the air nucleus in the liquid water: contraction, stable existence and expansion, as shown in Figure 2. It has been observed that the nano air nucleus were poorly soluble in water and the molecules remained clustered even when the air nuclei contracted to a very small volume.

Comparing the changes of the air nucleus at different temperatures, it was found that the time for the air nucleus to reach equilibrium was almost constant with the increase of temperature. Under the same initial conditions, the trends in the volume of air nuclei in the systems with different temperatures were consistent, in higher temperature systems, the equilibrium volume of the air nucleus was slightly larger, especially in the air nuclei with higher initial density. This is related to the thermal motion of the molecules and the surface tension of the liquid. This will be discussed further below.

The changes of nuclei volume with different initial radii in Fig. 3 are analyzed. The air nuclei volume varied dramatically during the first 15000fs of the calculation, and then gradually moved toward the dynamic equilibrium. That is, the air nuclei volume fluctuated within a certain range, but the average value remained stable. This dynamic balance is determined by the state characteristics of the gas: the calculation system temperature was constant, when the volume of air nuclei with a certain mass was greater than the equilibrium state, the air nuclei pressure would be reduced, the pressure of liquid water in the outside world, The air nuclei are forced to shrink and re-tend to their equilibrium values; On the contrary, when the nuclei volume was less than the equilibrium state, the pressure in the air nuclei would increase, and the air nuclei would then expand to the equilibrium value.

The volume change of the air nuclei was not only related to the initial volume of the air nuclei, but also to the internal density of the air nuclei. The air nucleus of three initial sizes showed a shrinking tendency when the initial density of the air nucleus was less than 0.4g/cm3. The air nuclei with the initial radius of 0.8nm shrank more rapidly and the air nuclei with the initial radius of 1.0nm shrank more slowly. For the air nuclei with the initial radius of 0.8nm, when the internal initial density was about 0.5-0.6g/cm3, the nuclei slowly shrank and then balanced, and when the initial density was about 0.7g/cm3, the air nuclei could stably exist in liquid water, When the initial density was greater than 0.7g/cm3, the air nuclei began to show an expansion trend; For the air nuclei with initial radii of 1.0 nm and 1.2 nm, the nuclei slowly shrank when the initial internal density was 0.5g/cm3, when the internal initial density was 0.6g/cm3, the air nuclei could stably exist in liquid water, and when the internal initial density was more than 0.6g/cm3, the nuclei expanded. We can infer that there was a critical value for the internal density of the air nuclei. When the density was smaller than the critical value, the air nuclei shrank gradually. When the density was equal to the critical value, the air nuclei could stably exist in the liquid water. When the density was larger than the critical value, the air nuclei showed an expansion trend. The critical value was negatively correlated with the air nuclei volume, and the larger the air nuclei volume, the smaller the critical value.

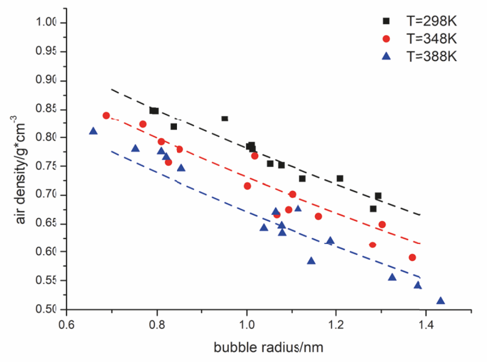


Fig. 4. Relationship Between Air Nuclei Radius and Internal Density

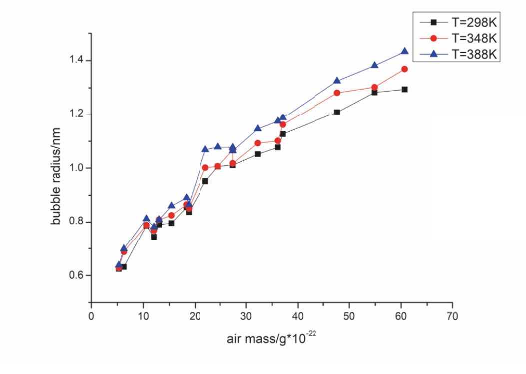


Fig.5. Relationship between Air nuclei Mass and Nuclei Radius

Figure 4 shows the relationship between nuclei density and nuclei radius when the system was in a state of dynamic equilibrium. The general trend is analyzed. It can be concluded that the density of air nuclei was negatively correlated with the nuclei radius at equilibrium, verifying the conjecture described above. At higher temperatures, the density in the same volume of air nuclei is less. We used the function fitting map data points, and according to the obtained function speculated that the air nuclei with a radius of 3.3nm can exist stably in 298K water when its internal density is 0.00129g/cm3.

We further investigate the relationship between air nuclei mass and nuclei radius when the system was in a state of dynamic equilibrium, as shown in Figure 5. It shows that the balance volume of the air nuclei was essentially determined by the gas mass contained in the nuclei. The gas mass in the nuclei was positively correlated with the nuclei radius at equilibrium, and the same mass of air nuclei is larger in higher temperature water

INFLUENCE OF WATER SURFACE TENSION ON AIR NUCLEUS

A balanced interface with stable nuclei inside and liquid outside can be used Young-Laplace (Y-L) equation:

$$p_g = p_1 + \frac{2\tau}{R} \tag{5}$$

Where p_g for the noncondensable gas pressure inside nuclei; p_l for the liquid pressure outside the nuclei; R for the nuclei radius (This article neglects the tiny differences between the nuclei equimolar surface and the surface of tension [12]); τ for the surface tension of the liquid. Studies have shown that Y-L equation (5) still holds when applied to nano nucleus [13].

In this paper, the air pressure pg and liquid water pressure p1 were calculated under the micro-canonical ensemble (NPT) by molecular dynamics to calculate the surface tension of the liquid water through the Y-L equation (5).

The stress tensor for a single atom i is given by:

$$S_{ab} = -\left[\begin{array}{l} mv_a v_b + \frac{1}{2}\sum_{n=1}^{N_p}\left(r_{1a}F_{1b} + r_{2a}F_{2b}\right) + \frac{1}{2}\sum_{n=1}^{N_b}\left(r_{1a}F_{1b} + r_{2a}F_{2b}\right) + \\ \frac{1}{3}\sum_{n=1}^{N_a}\left(r_{1a}F_{1b} + r_{2a}F_{2b} + r_{3a}F_{3b}\right) + Kspace\left(r_{ia}, F_{ib}\right) + \sum_{n=1}^{N_f} r_{ia}F_{ib} \end{array}\right] \tag{6}$$

Where a and b take the values x, y, z to generate six components of a symmetric tensor. The first term is a kinetic energy contribution for atom i. The second term is a pairwise energy contribution where n loops over the N_p neighbors of atom i, r_1 and r_2 are the positions of the 2 atoms in the pairwise interaction, and F_1 and F_2 are the forces on the 2 atoms resulting from the pairwise interaction. The third term is a bond contribution of similar form for the N_b bonds which atom i is part of. The fourth term is the KSpace contribution from long-range Coulombic interactions. Finally, there is a term for the N_f fixs that apply internal constraint forces to atom i.

Pressure calculation formula is:

$$p = \frac{-\left(S_{xx} + S_{yy} + S_{zz}\right)}{3V} \tag{7}$$

Where V is the sum of volume of the Voronoi cell around same type molecules.

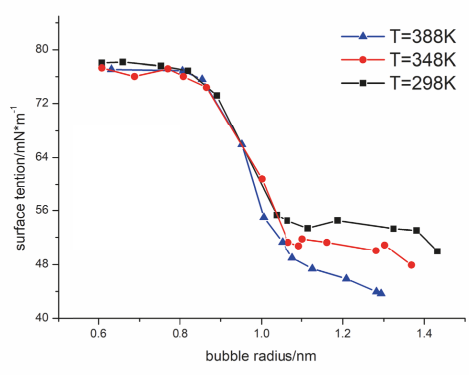


Fig.6. Relationship between surface tension of liquid water and air nuclei radius

After 30000 fs, the air nuclei volume, the liquid water pressure, and the pressure inside the air nuclei had stabilized. Therefore, the surface tension of the liquid water was calculated by averaging the air nuclei radius, the liquid water pressure, and the pressure inside the air nuclei after 30000 fs. This paper calculated and analyzed the surface tension of liquid water in 13 different size air nuclei systems. It can be seen from Figure 6 that as the air nuclei radius increased, the calculated liquid water surface tension generally decreased, which is consistent with the trend of the calculated simulation in [16],but not consistent with the trend of surface tension in [13,15]. The surface tension of liquid water in a 0.6 nm radius air nuclei system was about 77 mN/m. The surface tension of liquid water drastically decreased in systems with air nuclei radius of 0.8-1.1 nm. The surface tension of liquid water in system with 1.1 nm radius air nuclei was only 47 mN/m. The surface tension of liquid water slowly decreased in systems with air nuclei radius greater than 1.1 nm. The point data fitting function showed that when the air nuclei radius tended to infinity, the surface tension of liquid water at about 43mN/m, which was smaller than the experimental results 72mN/m, and it was close to results calculated in [13] , but different from the calculated data in [15].

This tendency of the surface tension of liquid water to decrease as the air nuclei radius increases can explain the negative correlation between the above-mentioned density critical value of the air nuclei in liquid water and the volume of the cavitated nuclei. According to the balance equation, when the pressure outside the nuclei of the air remains the same, the smaller the surface tension of the liquid water, the smaller the pressure needed to maintain balance within the air nuclei. According to the gas equation of state, when the temperature is constant, Increase, then the density decreases, the pressure value is also reduced, so that the air just reached the kinetic equilibrium nuclear.

The surface tension of liquids at different temperatures has a small difference when the nuclei radius is less than 1.0 nm, and the difference is significant when the nuclei radius is greater than 1.0 nm. Overall the higher the temperature, the smaller the surface tension of the liquid. This also explains the reason for the larger volume of air nuclei in the higher temperature system above.

This tendency of the surface tension of water to decrease as the air nuclei radius increased can explain the negative correlation between the above-mentioned density critical value of the air nuclei and the volume of the nuclei. According to the balance equation, when the pressure outside the nuclei remains unchanged, the smaller the surface tension of the liquid water, the smaller the pressure needed to maintain balance within the air nuclei. According to the gas state equation, when the temperature is constant and volume increase, then the density decreases, the pressure value is also reduced, so that the air nuclei just reaches the kinetic equilibrium.

SYSTEM STATE CHANGE DURING CAVITATION

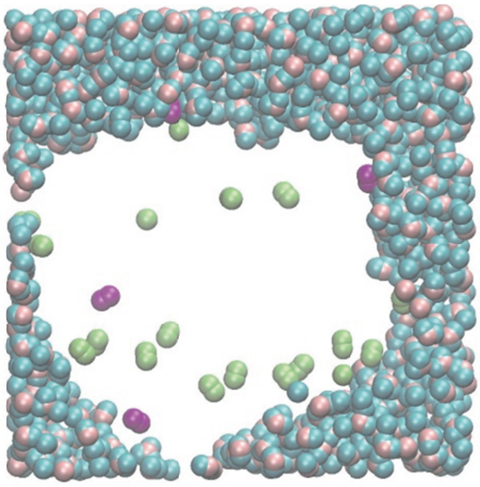
Taking the above equilibrium process as a relaxation process, we further studied the cavitation process of liquid water containing air nucleus. We chose systems in which the air nuclei was stable in water, and controlled the pressure of liquid water to promote cavitation. We observed the volume change of air nucleus during cavitation development, and monitored the potential energy change of the whole system. In this process, the system temperature remained at 298K, the system was unrestricted and could be freely changed. A visual image of the cavitation development system is shown in Figure 7. Once cavitation occurred, the air nuclei volume

Tab. 1. Cavitation Calculation Results

Initial nuclei radius (nm)	Initial PotEng (kcal/mol)	Pressure (atm)	Cavitation	End PotEng (kcal/mol)	Cavitation Timestep (fs)
0.8	-43443.054	-500	No	-42910.93	/
0.8	-43443.054	-700	Yes	-44752.241	61000
0.8	-43443.054	-1000	Yes	-45408.247	34000
1.0	-42655.704	-400	No	-42388.551	/
1.0	-42655.704	-500	Yes	-43334.101	168000
1.2	-41864.537	-300	No	-41853.478	/
1.2	-41864.537	-400	Yes	-42944.457	101500
1.2	-41864.537	-500	Yes	-42227.299	60500

increased rapidly and the entire process was irreversible until the air nuclei collapsed.

a) Initial development of cavitation



b) Full development of cavitation

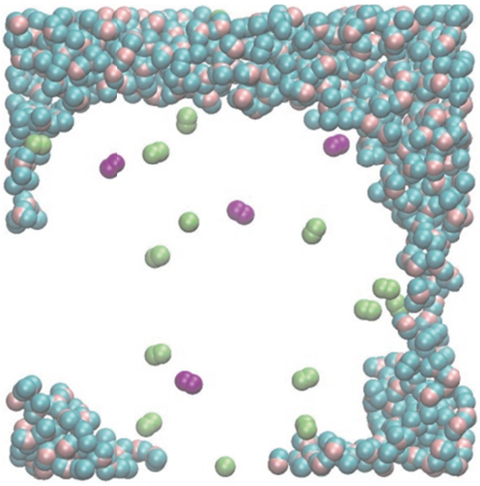


Fig.7. System state change during cavitation

After calculating the gas-liquid equilibrium above, we selected three systems in which air nucleus could exist stably and performed 8 sets of calculations. The results are shown in the table 1.

We found that the liquid negative pressure value and initial nuclei radius are the key factors affecting the occurrence of cavitation. The cavitation of system with initial nuclei radius of 0.8 nm occurs when the water pressure reaches -700 atm. The cavitation of system with initial nuclei radius of 1.0 nm occurs when the water pressure reaches -500 atm. The cavitation of system with initial nuclei radius of 1.2 nm occurs when the water pressure reaches -400 atm. The critical time for cavitation in an air nuclei system with initial radius of 1.2 nm is 101500 fs when the water pressure reaches -400 atm, and the critical time for cavitation when the water pressure reaches -500 atm is 60500 fs; The critical time for cavitation in an air nuclei system with initial radius of 1.0 nm is 168000 fs when the water pressure reaches -500 atm; It can be seen that the system can remain stable under a small negative pressure, and cavitation occurs when the negative pressure reaches a critical value, and the larger the negative pressure, the shorter the critical time for cavitation of the system. A certain size of air nucleus corresponds to a certain cavitation critical negative pressure, and the smaller the air nucleus, the larger the cavitation critical negative pressure. Under the same negative pressure, the cavitation of the smaller air nuclei system takes a longer time. Therefore, it can be seen that the larger the initial radius of the air nuclei, the larger the liquid negative pressure, and the more likely the cavitation occurs.

The above-mentioned conclusion that the surface tension of the liquid water decreases as the nuclei radius increases can also be considered as the cause of the cavitation critical negative pressure becoming smaller. The cavitation characteristics described above can also be considered from the perspective of system potential energy. It can be seen from the table that the potential energy of the system is reduced if cavitation occurs; the initial potential energy of the system with smaller air nucleus is smaller, and the system is more stable. Cavitation has free energy barriers, and a small negative pressure does not allow the system to cross the cavitation free energy barrier, so cavitation cannot occur.

CONCLUSION

Through the molecular dynamics simulation method, the stability of air nucleus in liquid water was studied and the key factors that affect its stability were analyzed. The following conclusions are drawn:

1. Air nucleus with a certain mass could maintain the dynamic equilibrium in liquid water. In the external liquid pressure, the air nuclei pressure and liquid surface tension together, the air nucleus in the water gradually reached the dynamic equilibrium. There were three states of the air nucleus in the liquid water: contraction, stable existence and expansion. There was a critical value for the internal density of the air nuclei. When the density was smaller than the critical value, the air nuclei shrank gradually. When the density was equal to the critical value, the air nuclei could stably exist in the liquid water. When the density was larger than the critical value, the air nuclei showed an expansion trend. The critical value was negatively correlated with the air nuclei volume, and the larger the air nuclei volume, the smaller the critical value. We speculated that the air nuclei with a radius of 3.3nm can exist stably in water when its internal density is 0.00129g/cm3.
2. The balance volume of the air nuclei was essentially determined by the gas mass contained in the nuclei. The gas mass in the nuclei was positively correlated with the nuclei radius at equilibrium.
3. Liquid surface tension is an important factor affecting the stability of the air nuclei. With the increase of air nuclei radius, the interfacial tension of liquid water tends to decrease. The higher the temperature, the smaller the surface tension of the liquid.

4. The larger the initial radius of the air nuclei, the smaller the water pressure, and the more likely the cavitation occurs.

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