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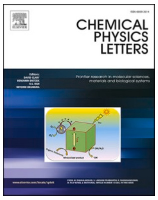

Numerical investigation of effects of particle properties on nucleation process in a sulfuric acid–water vapor mixture: Homogeneous versus heterogeneous

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A B S T R A C T

The pollution caused by sulfuric acid mist emissions has attracted attention due to demand of low emissions. In this work, molecular dynamics simulation was used to investigate effects of particle properties on nucleation process in a sulfuric acid–water vapor mixture. The effects of particle properties, including particle-vapor interaction and particle shape was analyzed in detail. The results show that with the increase of the particle-vapor interaction, heterogeneous nucleation gradually occupies an advantage in the competition with homogeneous nucleation. For non-spherical particles, the vapor molecules are more prone to heterogeneous nucleation on the flat surface.

1. Introduction

The rapid development of society and economy has brought about huge energy consumption, which is caused by fossil fuels, resulting in a large number of pollutant emissions [1]. As the thermal power and other industries generally achieve ultra-low emissions of particulate matter, sulfur dioxide and other conventional pollutants, the pollution caused by sulfuric acid mist emissions has attracted attention.

Existing studies [2–7] have shown that the analysis and prediction of physical migration of sulfuric acid during flue gas cooling are based on two kinds of nucleation mechanisms: homogeneous and heterogeneous. The nucleation rate of supersaturated vapor on an ideal surface can be calculated using a classical heterogeneous nucleation model [8]. However, in the actual nucleation process, nucleation occurs in the active parts of solid surface. The surface properties have an important influence on the nucleation of vapor, while the shape and size have an important influence on the growth stage [9]. Lee et al [10]. studied the nucleation characteristics of organic vapor on soot particles, they found that there was a huge deviation between the nucleation rate calculated by using Fletcher's heterogeneous nucleation theory and the experimental data. The fly ash in flue gas has a wide particle size range, covering from tens of nanometers to hundreds of microns, and fly ash has different shapes, including approximate spherical shape and irregular shape. The surface properties of the fly ash are also inconsistent due to different fly ash compositions [11], which will directly affect the heterogeneous nucleation characteristics of sulfuric acid. Due to the coexistence of sulfuric acid and water in the flue gas, it is not suitable to use the conventional unary heterogeneous nucleation theory to analyze the binary heterogeneous nucleation occuring on the surface of fly ash. Homogeneous nucleation also occurs when supersaturation ratio is relatively high. Noppel [12], Vehkamaki et al. [13] used binary homogeneous nucleation theory to predict the formation process of acid droplets in the atmospheric environment, but parameters such as equilibrium constant and saturated vapor pressure required for model calculation were determined by trial calculation and backward through comparison with experimental data. Due to the lack of basic data, the binary homogeneous nucleation theory cannot be directly used to analyze the homogeneous nucleation process of gaseous sulfuric acid in flue gas. It is necessary to study the binary homogeneous nucleation characteristics of sulfuric acid and water under flue gas conditions, explore and establish the corresponding binary homogeneous condensation model on this basis.

With the rapid development of computers and computing power, molecular dynamics simulation (MD) has been used to study nucleation process. Compared with the traditional experimental and theoretical methods, MD can observe the nucleation process from the molecular scale, dynamically track different molecules in the nucleation process, obtain the required information through statistics. Some scholars have

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studied the nucleation process of sulfuric acid water with MD. Doyle [14] first carried out quantitative analysis of homogeneous nucleation process of sulfuric acid and water, and found that adding a small amount of sulfuric acid in water can effectively improve the nucleation rate of water vapor. Subsequent studies [15–17] found that the nucleation rate observed in experiments was several orders of magnitude lower than the nucleation rate predicted by traditional theories, and the stabilization effect on vapor by the hydrate formation is thought to be responsible for this reduction in nucleation rate [18]. After further analysis of hydrate structure, scholars found that hydrate coagulation rate plays a leading role in the effect of enhanced nucleation, because hydrate is a multi-shell structure composed of sulfuric acid inner shell and water diffusion shell [19]. Although scholars have gained a certain understanding of the homogeneous nucleation process of sulfuric acid and water, in the actual flue gas emission process, the supersaturation ratio is always not high enough and homogeneous condensation is difficult to occur, thus the nucleation process of sulfuric acid in flue gas is often dominated by heterogeneous nucleation. Compared with the discrete micro-droplets of sulfuric acid formed by homogeneous nucleation, the sulfuric acid droplets of heterogeneous nucleation on fly ash surface are easier to be desulfurized by conventional desulfurization method. Therefore, increasing the proportion of sulfuric acid molecules in heterogeneous nucleation is beneficial to improve desulfurization efficiency and reduce sulfuric acid emissions. However, as far as the authors know, there is no specific study on the heterogeneous nucleation process of sulfuric acid–water vapor mixture, and it is not clear how to determine the proportion of heterogeneous nucleated sulfuric acid molecules in space. It is necessary to study the mechanism of heterogeneous nucleation of sulfuric acid–water vapor mixture.

A large number of scholars have explored the influence mechanism of parameters in heterogeneous nucleation process. Xu et al. [20] studied the heterogeneous condensation process of water vapor on smooth surfaces and found that condensation is more likely to occur on hydrophilic surfaces. By changing the wettability of particle surface, Rose [21] and Schmidt et al. [22] proposed two different heterogeneous nucleation modes: droplet-like nucleation and film-like nucleation, these studies are essentially studies on the influence of solid–liquid interaction. Suh and Yasuoka [23] analyzed the heterogeneous nucleation process of argon vapor under three-dimensional spherical particles of different sizes, they found that the seed size has a reciprocal effect on the growth rate, but showed to be insignificant on the homogeneous nucleation characteristics. Subsequently, Suh and Yasuoka also studied the heterogeneous nucleation process of cubic [24] and cuboid particles [25], and found that supersaturation ratio and particle shape have an important influence on the nuclei growth process. The above studies indicate that particle properties (including particle surface wettability, particle size and particle shape) have an important influence on the heterogeneous nucleation and nuclei growth process. However, the vapor molecules in the above studies are mainly simple fluids, the heterogeneous nucleation of sulfuric acid–water vapor mixture is more complicated. For example, the heterogeneous nucleation process starts from the adsorption and wetting of vapor molecules on the particle surface. Since the surface tension of sulfuric acid and water is different, the wettability of sulfuric acid and water on solid surface also varies, thus the adsorption state of vapor molecules is closely related to the composition of the vapor mixture. Therefore, the influence mechanism of parameters in the heterogeneous nucleation process of vapor mixture is different from that of pure working fluids. The spatial distribution of sulfuric acid in flue gas determines the desulfurization efficiency of subsequent desulfurization methods. In order to predict the spatial distribution of sulfuric acid in the nucleation process, it is necessary to recognize the key parameters of the heterogeneous nucleation process of sulfuric acid on the fly ash surface and its influence mechanism on the nucleation process.

In this work, MD simulation was used to investigate effects of particle properties on nucleation process in a sulfuric acid–water vapor mixture. The influence mechanism of sulfuric acid mole fraction on nucleation and nuclei growth process was revealed. The effects of particle properties, including particle-vapor interaction and particle shape on the heterogeneous nucleation and nuclei growth process was analyzed in detail.

2. Simulation details

In this work, all the simulations were performed by LAMMPS (Large-scale Atomic/Molecular Masssively Parallel Simulator) package [26]. To simulate the nucleation process in a sulfuric acid–water vapor mixture, a simulation system of 300*300*300 Å^3 is created as shown in Fig. 1. An ideal spherical carbon particle with the FCC crystal structure for simplification is placed in the center of the box, the carbon particle diameter $d = 7$ nm, 10,000 vapor molecules are arranged in the space around the carbon particle, corresponding to the vapor phase density of around 13.2 kg/m^3. Periodic boundary conditions are adopted in all dimensions. In order to study the difference in dynamic characteristics of nucleation process for sulfuric acid–water vapor mixture with different components, three different cases ($x_{\text{SA}} = 0.001$, 0.052, 0.09) were simulated, the corresponding molecular numbers of sulfuric acid are 10, 520, 900 respectively.

Before nucleation, the system ran 2 ns in the Nose–Hoover NVT (constant molecular number, volume and temperature) ensemble with a time step 1 fs to obtain the equilibrium sulfuric acid–water mixture. The temperature is set to 500 K. In the process of vapor phase equilibrium, to avoid premature nucleation on particle surface, the interaction between particle and vapor molecule was set to 0. In this work, the nucleation process is also realized by NVT, when the vapor mixture reaches equilibrium, the temperature of the system is reduced to target temperature with a decrease rate of 1000 K/ps, then the temperature of the system will keep constant for 4 ns with a time step 1 fs, the supersaturated vapor molecules will collide and nucleate to realize nucleation and condensation process.

In this study, OPLS-AA (optimized potentials for liquid simulations-all atoms) model [27] was applied to sulfuric acid–water vapor molecules, and the SPC/E (Extended Single Point Charge) model was used for water molecules [28]. The potential energy is expressed as follows:

$$U_{\text{Conf}} = \sum_{\text{bonds}} k_d (d - d_0)^2 + \sum_{\text{angles}} k_\varphi (\varphi - \varphi_0)^2 + \sum_{\text{dihedral}} k_\chi [1 + cos(n\chi - \delta)] + \sum_i \sum_{j>i} \{4\varepsilon_{ij} [(\frac{\sigma_{ij}}{d_{ij}})^{12} - (\frac{\sigma_{ij}}{d_{ij}})^{6}] + \frac{1}{4\pi\varepsilon_0} \frac{q_i q_j}{d_{ij}}\} \tag{1}$$

Where U_{Conf} is the pair potential energy; σ_{ij} and ε_{ij} are the length and energy parameters between atoms i and j, and d_{ij} is the distance between atoms; d, φ, χ are bond length, angle and dihedral. The rest nomenclatures are potential parameters. The cutoff distance is 10 Å. The long-range effect due to electrostatic potential is resolved by a particle–particle particle-mesh approach with an accuracy of 1.0×10^{-4}. All the potential parameters are listed in supplementary information. The interactions between solid carbon atoms was modeled by Lennard-Jones (LJ) potential, the interaction parameters refer to the existing work [29]. All LJ parameters for unlike atoms are obtained from the Lorentz–Berthelot combining rule [30]. The temperature dependent equilibrium properties of sulfuric acid–water mixture with different compositions are calculated and the reliability of the potential model is verified. The details of the calculation are attached in the supplementary materials. In this work, the interaction between particles and the vapor molecules is the driving force of heterogeneous nucleation, when the interaction between particles and vapor molecules is weak, it takes too long for the heterogeneous nucleation to occur, and the heterogeneous nucleation process cannot be effectively observed during the simulation period. Conversely, when the interaction force between particles and the vapor molecules is strong, vapor molecules will quickly adsorb and connect on the particle surface to form liquid films covering the particle surface, which is actually surface condensation rather than nucleation.

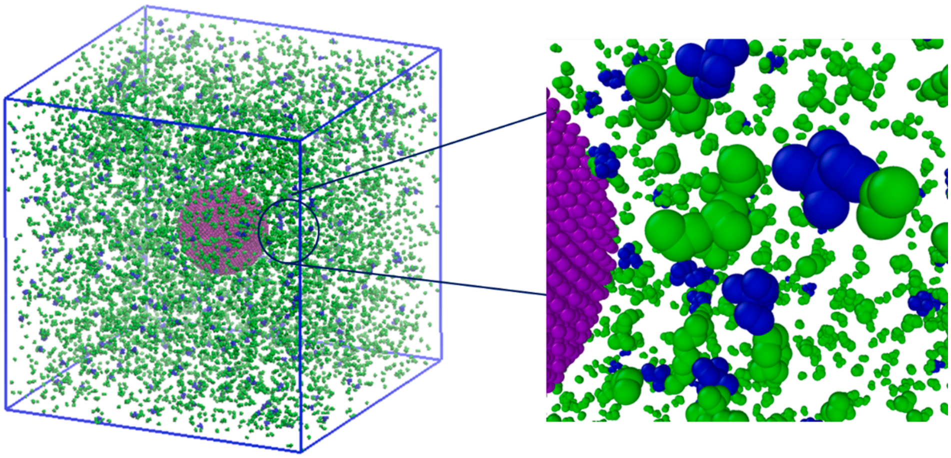


Fig. 1. Schematic diagram of initial system ($d = 7$ nm is shown in the sketch): green are water molecules, blue are sulfuric acid molecules, purple are carbon atoms. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Therefore, the interaction force between carbon particles and gas molecules in this study is modified by multiplying the coefficient 2.0 in consideration of the above effects.

The definition of clusters is according to the Stillinger criterion [31], when the distance between two molecules is less than a certain value, the two molecules are considered to belong to the same cluster, the value corresponds to the first trough of the liquid radial distribution function, which is around 3.6 Å in this work.

3. Results and discussions

3.1. Cluster analysis

The supersaturation ratio selected in this work is 9.6 corresponding to the target temperature of 400 K. The details of how to select appropriate supersaturation ratio is attached in the supplementary materials. The Yasuoka-Matsumoto method [32] was adopted in this work, which is always used to calculate the critical size and stable nucleation rate in molecular dynamics. The basic principle of this method is to calculate the slope of cluster number curve when the number of molecules is greater than a certain value. When the number of cluster molecules reaches the critical nucleation number, the slope of cluster number curve tends to be stable. Therefore, the nucleation rate can be obtained by calculating the slope of cluster number curve. This method is usually used for homogeneous nucleation, but existing studies have shown that it is also suitable for heterogeneous condensation when the particle size is large enough [33]. Since the homogeneous nucleation rate is defined as the number of nucleation nuclei generated per unit volume in unit time, and the heterogeneous nucleation rate is defined as the number of nucleation nuclei generated per unit area in unit time, both of which can be calculated from the slope of the cluster change curve larger than the critical nucleation number. By changing the determination value of cluster molecular number, the critical number of nucleated molecules for heterogeneous nucleation and homogeneous nucleation in this work is acquired of about 10 and 25, respectively.

3.2. Compositional effects on heterogeneous nucleation

Heterogeneous nucleation can be divided into three stages. In the first stage, supersaturated gas molecules adsorb on the surface of particles, which is called the "start-up" stage according to previous study [33], at this stage, the number of molecules adsorbed on the surface of particles increases slowly. The second stage is "nucleation" stage, in which the number of clusters on the particle surface increases linearly. The third stage is the "nuclei growth" stage, in which small nuclei coalesce to form larger nuclei and the clusters grow rapidly.

In order to quantitatively analyze the difference of nucleation characteristics between systems with different compositions, it is necessary to define the classification criteria of each stage and compare the effects of compositions by stages. By changing the mole fraction of sulfuric acid in the vapor mixture, the number of clusters above the threshold size as a function of time for sulfuric acid–water mixtures with different compositions is statistically obtained, as shown in Fig. 2. In this work, the time point corresponding to the cluster number reaching 1 for the first time is selected as the start time of nucleation stage. It can be seen from the figure that with the increase of sulfuric acid mole fraction, the start time of nucleation stage is significantly advanced, and the rate of heterogeneous nucleation is also greatly increased.

In order to distinguish the nucleation stage from the nuclei growth stage, the average size of the adsorbed clusters on the particle surface was calculated as shown in Fig. 3, It can be seen that the average cluster size keeps relatively constant in the early stage, and then gradually grows, and the cluster growth rate increases gradually, which is because the larger the cluster surface area is, the more vapor molecules absorbed on the surface of the nuclei. Meanwhile, with the increase of the mole fraction of sulfuric acid, the cluster growth rate becomes faster, but the promoting effect of the mole fraction of sulfuric acid on the nuclei growth rate gradually weakens. In this work, the time point at which the average cluster size reaches 10 is defined as the end of the nucleation

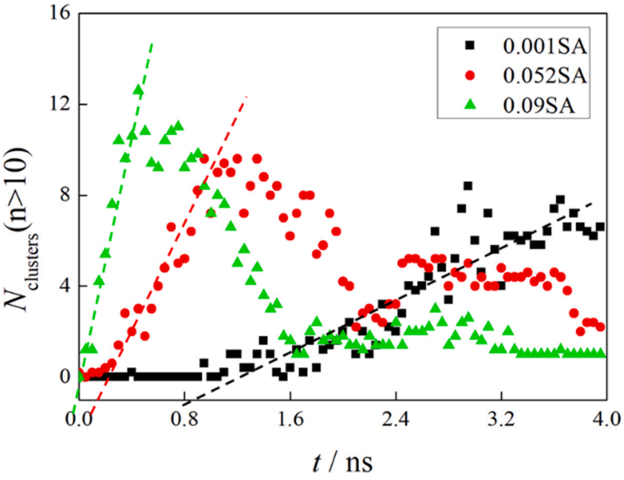


Fig. 2. Number of clusters above the threshold size as a function of time for sulfuric acid–water mixtures with different compositions. The nucleation rate is the slope of the linear fit (dashed line).

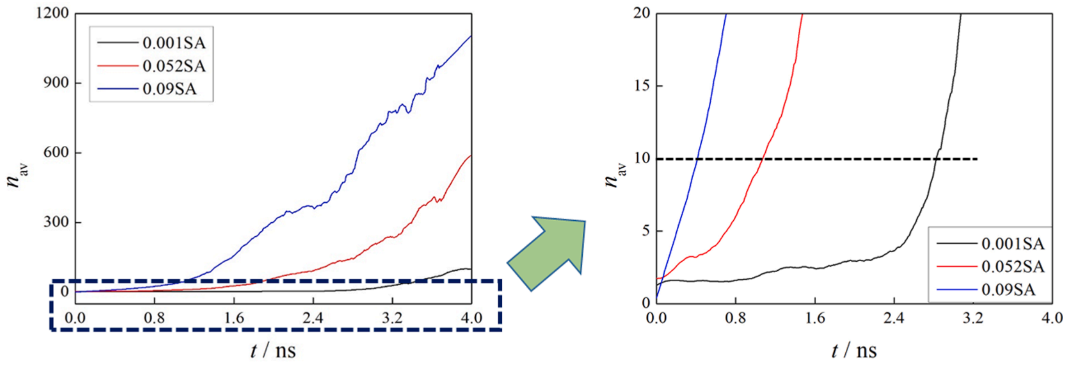


Fig. 3. The variation of the average cluster size with time for sulfuric acid–water mixtures with different compositions.

stage, then the duration of the nucleation stage for sulfuric acid–water mixtures with different compositions can be obtained in combination with the start time of nucleation stage mentioned above.

Parameters for heterogeneous nucleation of sulfuric acid–water mixtures with different compositions are summarized as shown in Table 1, t1 and t2 are the start time and the end time of nucleation stage, respectively. The results show that sulfuric acid molecules are more likely to be adsorbed on particle surface, and more sulfuric acid molecules can significantly promote the heterogeneous nucleation and nuclei growth. Besides, with the increase of sulfuric acid mole fraction, the nucleation rate enhancement effect is more significant, while the nuclei growth rate enhancement effect is gradually weakened.

Compared with the influence of compositions on homogeneous nucleation in previous study [19], the increase of sulfuric acid mole fraction in heterogeneous nucleation has a more significant promoting effect on nucleation rate. In order to reveal the influence mechanism of compositions on nucleation characteristics, the composition of adsorbed molecules in different systems was analyzed, and the molar fraction of sulfuric acid in adsorbed molecules on particle surface changes with time was calculated as shown in Fig. 4. In combination with Fig. 2, it can be seen that at nucleation stage, the sulfuric acid mole fraction increases rapidly and reaches the peak value, then decreases gradually at nuclei growth stage. The mole fraction of sulfuric acid on the surface of particles is much higher than the average sulfuric acid mole fraction of the system, indicating that the preferential adsorption of sulfuric acid molecules on particles magnifies the influence of sulfuric acid mole fraction on nucleation process, leading to the result that the increase of sulfuric acid mole fraction enhances the heterogeneous nucleation rate much more than that of homogeneous nucleation.

3.3. The effects of particle-vapor interaction

3.3.1. Particle-vapor interaction

As mentioned above, the interaction between particles and vapor is the driving force of molecular adsorption and heterogeneous nucleation on particle surface, which has a significant influence on the nucleation process. In fact, the interaction between particle and vapor determines the solid–liquid contact angle, which is the surface wetting characteristics of particles at macroscale. In order to compare and analyze the difference of heterogeneous nucleation under different particle-vapor interactions, take the case of $x_{SA} = 0.052$ as the object, the particle-vapor interaction was adjusted by adding energy factor α, and the energy factors were set as 0.5, 1.0, 2.0 and 5.0, respectively. The larger the energy factor is, the greater the particle-vapor interaction is, and the more lyophilic the particle surface is. Snapshots of the vapor molecules and clusters on the surface for different surface wettability at $t = 4$ ns are attached in the supplementary materials.

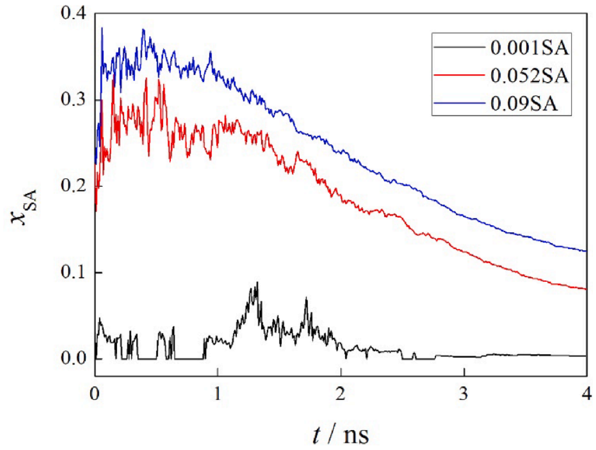


Fig. 4. Molar fraction of sulfuric acid in adsorbed molecules on particle surface changes with time.

Table 1
Parameters for heterogeneous nucleation of sulfuric acid–water mixtures with different compositions.

x_{SA}	0.001	0.052	0.09
t_1 / ns	1.17	0.25	0.03
t_2 / ns	2.83	1.08	0.42
Nucleation rate, J / (10^{25}m^{-2}s^{-1})	1.7	6.7	19.4
Average growth rate in “nuclei growth stage”, J_a / ns^{-1}	75.2	197.6	308.1

In order to quantitatively compare the difference of heterogeneous nucleation for different particle wettability, the analysis method is the same as that for the influence of compositional effects. The number of clusters above the threshold size as a function of time and the variation of the average cluster size with time for different particle wettability are calculated, as shown in Fig. 5. It can be seen from the figure that the heterogeneous nucleation rate increases significantly with the enhancement of the lyophilic characteristics of particle surface. Due to the enhancement of lyophilic property, the area covered by nucleated clusters on the particle surface is increased, and the number of adsorbed vapor molecules is positively correlated with the coverage area. Therefore, the nuclei growth rate is sensitive to the enhancement of lyophilic properties on the particle surface under the combined action of the enhancement of particle-vapor interaction and the increase of adsorption area. However, when the surface of particles is completely covered ($\alpha = 5.0$, $t = 1.5$ ns), the adsorption area remains nearly unchanged, and the nuclei growth rate slows down and remains relatively stable.

In the nucleation process of sulfuric acid–water binary system, sulfuric acid molecules can be divided into three forms: exist in the surrounding vapor phase in the form of monomer molecules, adsorbed to

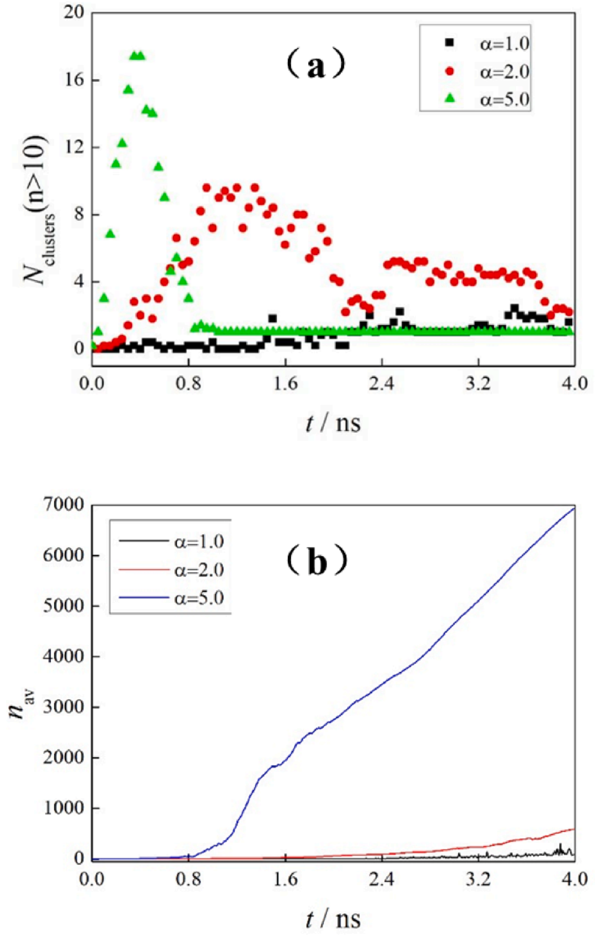
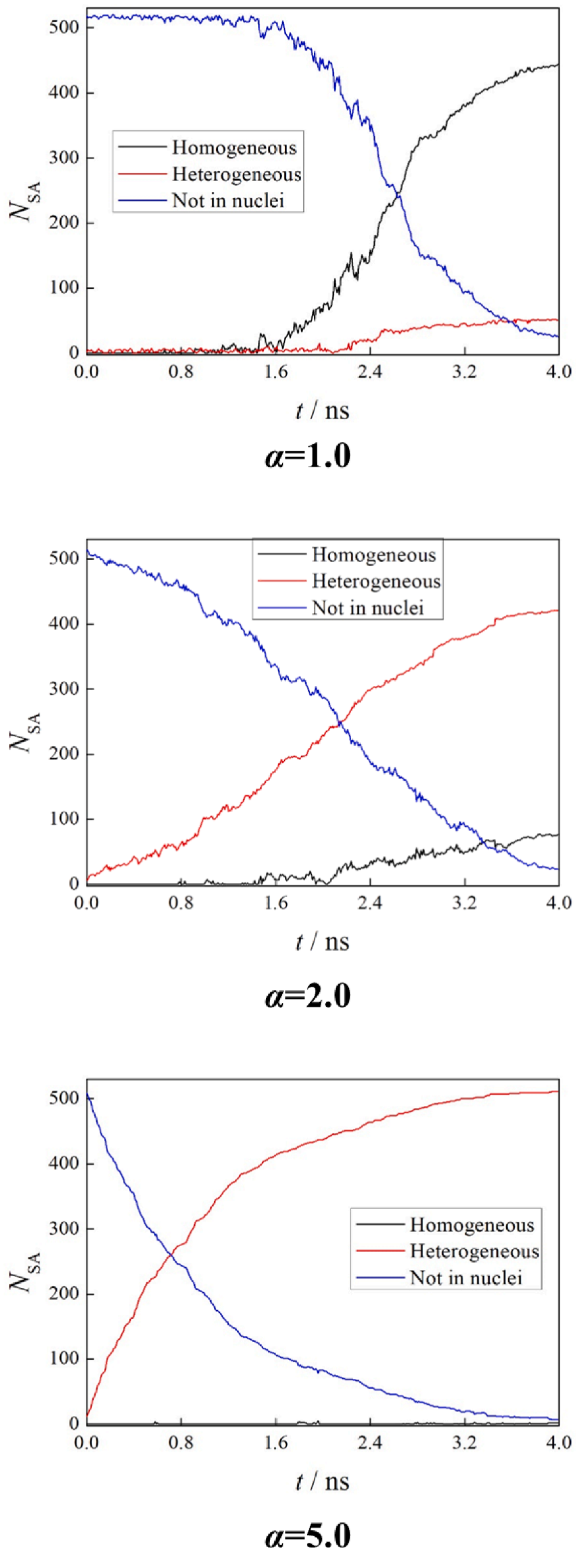


Fig. 5. (a) Number of clusters above the threshold size as a function of time for different particle wettability, (b) The variation of the average cluster size with time for different particle wettability.

the particle surface for heterogeneous nucleation and homogeneous nucleation in the vapor phase. Dynamic distribution of sulfuric acid molecules for different particle wettability are calculated, as shown in Fig. 6, in the figure, N_{SA} represents the number of sulfuric acid molecules. It can be seen from the figure that with the enhancement of lyophilic characteristics, heterogeneous nucleation gradually occupies an advantage in the competition with homogeneous nucleation. When the particle surface is completely wetted, heterogeneous nucleation can completely inhibit the occurrence of homogeneous nucleation. Due to the higher intensity of heterogeneous nucleation, the decrease rate of monomer molecules will gradually increase with the increase of lyophilicity of particles.

3.3.2. *Particle sizes and shapes*

The influence mechanism of particle size is also studied. Since the difference between the nucleation rate values for different nanoparticle sizes are very small within a limited range of particle sizes, this part is attached in supplementary materials for reference. In the above simulations on heterogeneous nucleation, the particles were assumed to be ideal spheres, but the actual fly ash particles in the flue gas have irregular shapes, and the influence of particle morphology on nucleation process could not be simply ignored. To analyze the influence of particle shape, spherical, cylindrical and cubic particles were designed respectively. The schematic diagram of different particle morphology is attached in the supplementary material. The size selection criterion is to ensure the same surface area of particles in the three forms. For spherical particles, the probability of nucleation sites is the same everywhere on the particle surface, but for other forms, the probability of different sites becoming the sites of heterogeneous nucleation is no longer consistent.



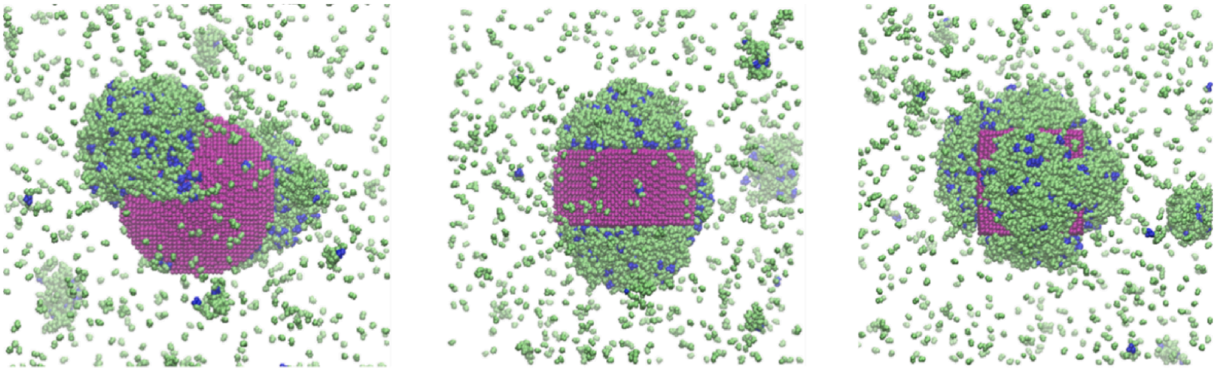
α=1.0

α=2.0

α=5.0

Fig. 6. Dynamic distribution of sulfuric acid molecules for different particle wettability.

The heterogeneous nucleation process of particles with different shape was calculated, and Fig. 7 shows the snapshots of the vapor molecules and clusters on the surface for different particle shape at $t = 4$ ns, it can be seen from the figure that for cylindrical and cubic particles,



sphere cylinder cube

Fig. 7. Snapshots of the vapor molecules and clusters on the surface for different particle shape at $t = 4$ ns.

the vapor molecules are more prone to heterogeneous nucleation on the flat surface.

In order to quantitatively compare the difference of heterogeneous nucleation for different particle shape, The number of clusters above the threshold size as a function of time and the variation of the average cluster size with time for different particle shape are calculated, as shown in Fig. 8. It can be seen from the figure that the closer the particle shape is to the spherical shape, the lower the heterogeneous nucleation rate on the particle surface, which is consistent with the higher condensation rate on the rough surface at the macro scale [34]. The difference of nucleation rate among different particle shapes is small, indicating that nucleation rate is less sensitive to particle shape. In the process of nuclei growth, the smaller the sphericity, the faster the nuclei growth rate. The reason for the difference is that the surface area of the adsorbed clusters is also different due to the different particle shapes. In combination with Fig. 7, it can be seen that the surface area of adsorbed clusters is positively correlated with the number of continuous surfaces on the particle. The more continuous surfaces there are (1 for spherical particle, 3 for cylindrical particle and 6 for cubic particle), the larger the surface area of adsorbed clusters, the faster the nuclei growth rate.

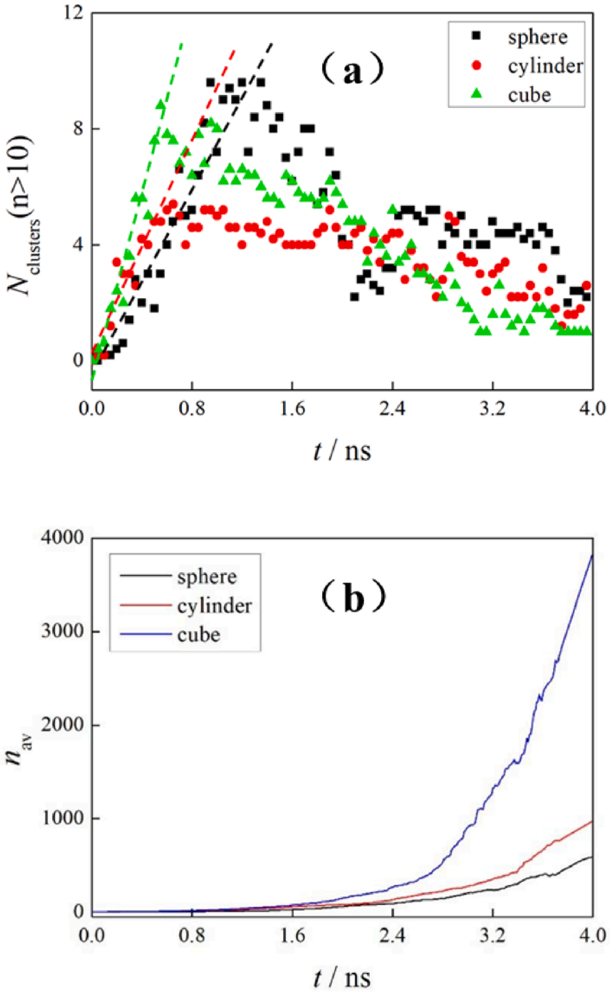


Fig. 8. (a) Number of clusters above the threshold size as a function of time for different particle shape, (b) The variation of the average cluster size with time for different particle shape.

4. Conclusions

In this work, MD simulation was used to investigate effects of particle properties on nucleation process in a sulfuric acid–water vapor mixture. The influence mechanism of sulfuric acid mole fraction on nucleation growth process and spatial distribution of different components was revealed. The effects of particle properties, including particle-vapor interaction and particle shape on the heterogeneous nucleation and nuclei growth process were analyzed in detail. The main conclusions are as following:

(1) More sulfuric acid molecules can significantly promote the heterogeneous nucleation and nuclei growth. Besides, with the increase of sulfuric acid mole fraction, the nucleation rate enhancement effect is more significant, while the nuclei growth rate enhancement effect is gradually weakened. In the heterogeneous nucleation process, the increase of sulfuric acid mole fraction enhances the heterogeneous nucleation rate much more than that of homogeneous nucleation, because the preferential adsorption of sulfuric acid molecules by particles magnifies the effect of sulfuric acid mole fraction on nucleation process.
(2) With the increase of the particle-vapor interaction, more vapor molecules adsorbed and nucleated on the particle surface, heterogeneous nucleation gradually occupies an advantage in the competition with homogeneous nucleation. When the particle surface is completely wetted, heterogeneous nucleation can completely inhibit the occurrence of homogeneous nucleation. Due to the higher intensity of heterogeneous nucleation, the total nucleation rate still increases with the increase of lyophilicity of particles.
(3) For non-spherical particles, the vapor molecules are more prone to heterogeneous nucleation on the flat surface. In the nuclei growth process, the smaller the sphericity, the faster the nuclei growth rate, which means for the simulations in this work, the surface area of adsorbed clusters is positively correlated with the

number of continuous surfaces on the particle. The more continuous surfaces there are (1 for spherical particle, 3 for cylindrical particle and 6 for cubic particle), the larger the surface area of adsorbed clusters, the faster the nuclei growth rate.

CRediT authorship contribution statement

Xinghui Wu: Conceptualization, Methodology, Formal analysis, Writing – original draft. **Zuozhou Tang:** Investigation. **Shuhang Lyu:** Writing – review & editing. **Qiang Song:** Funding acquisition, Validation, Supervision. **Yuanyuan Duan:** Validation, Supervision. **Zhen Yang:** Validation, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cplett.2022.139582.

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