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# Molecular clustering physical model of steam condensation and the experimental study on the initial droplet size distribution

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ABSTRACT

A molecular clustering physical model is proposed to describe the state of steam molecules in bulk steam phase before condensing on the cooled surface. Based on the general feature of nucleation processes, it is presumed that the steam molecules become clusters with the Lognormal size distribution in bulk steam phase firstly before condensing onto the cooled surface. In order to prove this physical model, the condensation of moist air on the surfaces with different wettability is observed using a high speed camera and microscope. The size distribution of the pre-coalescing droplets with radius of several microns is obtained, and for the primary droplets, about 3–10 nm, the size distribution is generated based on the experimental images reported in literature. It is concluded that the size distribution of the droplets from initial to the pre-coalescing size both satisfy the Lognormal distribution function, this proves the present physical model to be reasonable and consistence with the cluster physical theory. In addition, the observation that the initial condensation mode of moist air on the surfaces (either hydrophobic or hydrophilic) is always dropwise confirms the physical model further.

## 1. Introduction

It is now accepted by most investigators that dropwise condensation is a nucleation process [1–5]. It was considered that there were two simultaneously reverse processes, condensing and evaporating, on the nucleus surfaces. Only when the condensing rate was larger than the latter, the droplet could grow up; the droplet size at this moment was called the minimal size, which could be calculated by Kelvin theory [6–8]. Subsequently, the droplet growing process was divided into two stages [9], mainly through direct condensation on it self's surface and mainly through the coalescence between droplets, by the critical size.

Most of the previous investigations on droplet size distribution are based on steady condensation process. Le Fevre and Rose [10] assumed a form for the distribution function which had the correct behavior for the limiting cases of very large and very small droplets. And later, Rose and Glicksman [9] proposed a universal form of time-averaged distribution function for large droplets which grew primarily by coalescence with smaller droplets, by introducing a simplified model of sequence of events occurring during the time interval between successive sweepings by departing droplets. Graham and Griffith [11] investigated the distribution of larger size droplets and obtained their distribution function, however, for the droplets less than 10 microns, the distributions had to be inferred from the heat transfer measurements because of the limitation of the experimental instruments. In order to deal with the effect of numerous coalescences between droplets on the droplet size distribution, Gose et al. [12], Tanasawa and Tachibana [13] and Glicksman and Hunt [14] attempted to simulate the entire process of dropwise condensation, namely growth, coalescence and re-nucleation of droplets, by using a computer. Wu and Maa [15] deduced the droplet size distribution of the droplets less than the critical radius and growing up mainly through the direct condensation of steam based on the population balance theory, and for the larger droplets, the droplet size distribution function proposed by Le Fevre and Rose [10] was still used. Subsequently, Mousa [6], Vemuri and Kim [7] developed the small droplet size distribution based on the work of Wu and Maa [15]. Wu et al. [8] put forward a random fractal model to simulate the size and spatial distribution from the primary to the departing droplets in dropwise condensation. Lan et al. [16] rebuilt the spatial conformation of droplet distribution into the temporal conformation based on the random fractal model and reflected the dynamic characteristics of dropwise condensation and developed the dropwise condensation heat transfer model with the liquid–solid surface free energy difference effect. In addition, Le Fevre [8] guessed that the droplet size

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distributions might be decided simply geometrically, independent of the heat transfer process.

For the limitation of experimental conditions, the behaviors of the micro-droplets with size of several microns or smaller are not studied adequately, so the physical views before and right after steam nucleation are still unclear. Since the fact accepted by most investigators that dropwise condensation is a nucleation process, this process must have the general feature of nucleation processes. In fact, the formation of a new phase in a bulk phase always occurs by either homogeneous or heterogeneous nucleation [17], i.e. all of the phase changes are induced by nucleation which is triggered by molecular clustering. From this point of view, the molecular clustering should be the key issue of investigating the initial steam condensation.

Information regarding the cluster dynamics in phase change heat transfer provides a basis to better understand the microscale transport processes, which has been introduced into the boiling, especially the microscale boiling heat transfer. Peng et al. [18] investigated the role of perturbations on the dynamics of clusters, discussed the dynamics of clusters formed with internal or external perturbations, and proposed a pressure fluctuation model to physically interpret the occurrence of fictitious boiling. Based on the molecule self-assemble principle, Wang et al. [19] proposed the critical aggregation concentration concept of active molecules to describe the physical situation before nucleus form. Tian et al. [20] studied the aggregation of active molecules inside metastable bulk phase using thermodynamics theory, deduced expression of critical aggregation concentration, determined energy distribution of active molecules inside bulk phase at superheat and supercooling limit state, and used the molecule aggregation theory to describe gas–liquid phase transition process.

However, for the phase change heat transfer process of condensation, the understanding on the microscale transport process is still deficient.

Based on the general feature of nucleation processes, the present paper proposes a physical model to describe the state of steam molecules in bulk steam phase before condensing on cooled surface by introducing the molecular clustering concept. Subsequently, the physical model is proved to be reasonable by the experimental investigations on the size distributions of two groups of droplets: primary droplets with several nanometers in radius and pre-coalescing droplets with several microns in radius, according to the cluster physics theory.

## 2. Physical model of steam condensation introduced in molecular clustering concept

As a typical phase change process, condensation must be induced by nucleation (usually heterogeneous nucleation for the presence of the cooled surface) which is triggered by molecular clustering. During condensation process the conditions for molecular clustering are sufficient: In the vicinity of cooled walls or condensates, the existence of clusters depends on the condition of energy transfer between molecules of the surrounding gas, molecules approaching the cooled wall have higher temperatures (energies) than departing molecules that have transferred their energies to the wall, the energy exchanges between these oncoming and departing molecules of different temperatures provide a sufficient condition for molecular clustering (as shown in Fig. 1), the cluster size formed increases closer to the wall, and the thickness of cluster zone depends on the thermal condition of the molecular system and the processes of their energy transfer [21,22].

The classic dropwise condensation theory considered that the primary droplets nucleated from random nucleation sites which were probably pits and grooves in the surface, with the continuous addition of steam molecules condensed. The droplets grew up to a critical radius and the most of heat transferred by the surface was through those droplets smaller than the critical radius; after then, the coalescences between them became the main approach to grow up further, the contribution of this part of droplets to the overall heat transfer mainly was the surface area cleaning and sweeping by their coalescing and departing [1,4]. The distributions and movement characteristics of the droplets with large size were investigated sufficiently, while the understanding of the pre-nucleation situation was still insufficient. The initial condensation process has been given a special attention and been described using the concept of molecular clustering in the present paper, as shown in Fig. 2.

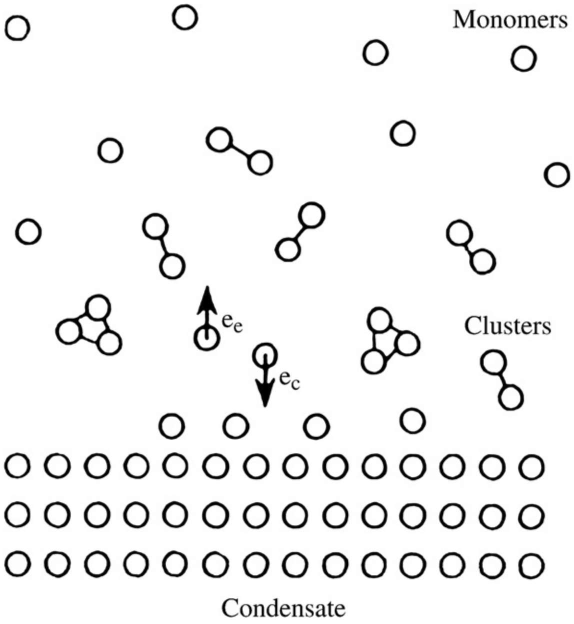

**Fig. 1.** Cluster formation in the vicinity of condensate [22]: evaporating molecules have lower energy ($e_e$) than condensing molecules ($e_c$); $e_e < e_c$ makes favorable conditions for cluster formation.

For the existence of surface subcooling, the steam molecules become clusters before condensing on the cooled surface, and a certain cluster size distribution forms in the bulk steam phase close to the surface (as in Appendix). After then, the clusters contact the cooled surface and deposit on it randomly, acting as the nucleate embryo. As their energies will not dissipate instantly, some of the deposited clusters are able to migrate on the surface in a limited

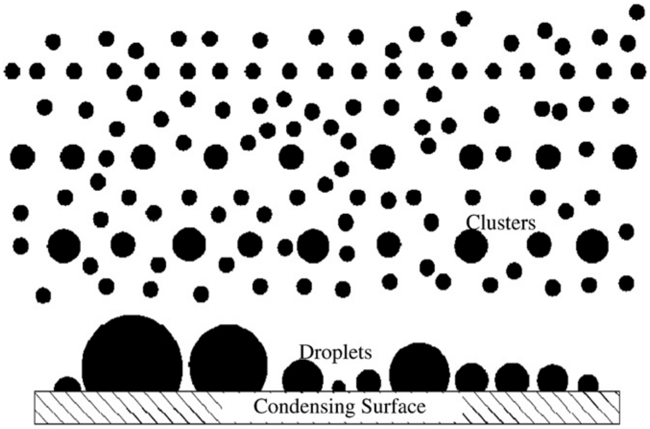

**Fig. 2.** New concept of vapor condensation.

area and incorporate or be incorporated by the other clusters on their ways, accompanied by the condensation of other clusters from vapor phase on their surfaces. At the same time, the evaporation on the surface is on-going as well. After energies have been dissipated, the clusters stay on the locations with high free energies [23], such as pits and grooves in the surface, finally. If the seated, post-incorporated clusters were not smaller than the minimal size defined in the classic dropwise condensation theory, they become the primary droplets and grow up through the condensation of the other clusters on their surfaces. It means that the primary droplets are not formed on the nucleation sites suddenly, they are composed of several water molecular clusters, and that the condensation heat transfer elements are not only the steam molecular monomers, but also the molecular clusters, they condense on the surface in the way of group by group, just like "quantum".

Bréchignac et al. [24] studied the nucleation and growth on a cluster scale for preformed antimony clusters soft landed on an amorphous carbon substrate, the incident cluster size was used as a parameter to monitor independently the average island size and island density, they mentioned that the distribution of the "islands" formed on the cooled surface by deposited clusters was identical to that of the incident clusters. Furthermore, the cluster physics states that the size distribution will display a Lognormal function feature if the molecules become clusters, equation (1) [23]:

$$n(r) = \frac{1}{\sqrt{2\pi}\sigma r}\exp\left(-\frac{\left(\ln\frac{r}{\bar{r}}\right)^2}{2\sigma^2}\right) \tag{1}$$

Where $n(r)$ is cluster size distribution function, $r$ the radius of cluster, $\bar{r}$ the mean radius of clusters, and $\sigma$ standard deviation.

To prove the clustering model for steam condensation, the presupposition is whether the steam molecules become clusters before condensing on the cooled surface. According to the investigating results of Wang [23] and Bréchignac et al. [24], if the size distributions of the primary droplets and the droplets growing up through direct condensation without coalescence on the condensing surface satisfy the Lognormal distribution function, the same size distribution should exist in the bulk gas phase and steam molecules should become clusters before condensing, then the present physical model can be proved. In addition, the conclusion can be drawn that the initial condensation mode should be "dropwise" both for dropwise and filmwise condensation, based on the present model. For this aim, we analyzed the size distributions of the droplets with size of several nanometers based on the experimental images of Liu

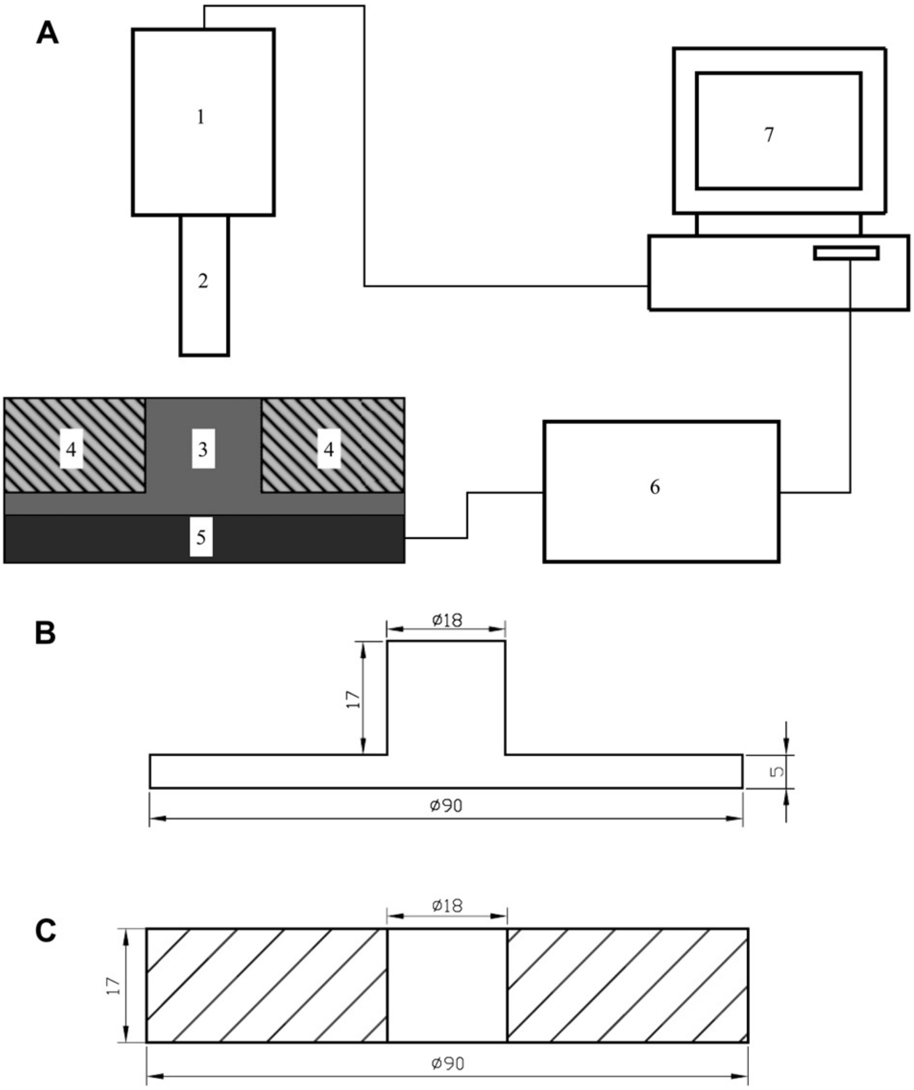

**Fig. 3.** Experimental apparatus and part dimensions. A – Layout of experimental apparatus; 1 – High speed camera, 2 – Microscope Lens, 3 – Test section (copper block), 4 – PTFE, 5 – Temperature-controllable unit, 6 – Controller, 7 – Computer. B – Dimensions of condensing block (mm); C – Dimensions of PTFE insulation (mm).

et al. [5] and several microns based on the present experimental observation respectively, and compared the condensation processes of moist air on the surfaces with different wettability.

## 3. Experimental investigations

### 3.1. Experimental apparatus

As shown in Fig. 3-A, the present experimental apparatus consists of image acquisition unit, condensing unit and temperature controlling unit. Experimental images are acquired by a high speed camera (PHOTRON, FASTCAM APX-RS) mounted with a set of microscope lenses (HIROX, CX10C), its highest shooting speed is 3000 fps at $1024 \times 1024$ pixels, maximum 250,000 fps with lowering the resolutions. With various objective lenses, the magnification can achieve 7000×. The images stored in the camera need to be transferred to computer through FireWire cable after every set of experiments. The condensing unit is made up of a machined copper block and PTFE insulation, the radius of condensing surface is 9 mm, as shown in Fig. 3-B and C. The temperature controlling unit includes a constant temperature platform and a temperature adjusting instrument, based on the Peltier effect, the surface temperature of the platform can vary in the range of $-20$–150 °C. The copper block fitted with PTFE insulation is placed on the platform with silica gel in the gap, for reducing the contact thermal resistance.

### 3.2. Experimental procedure

For the condensation of saturated steam, the initial stage is ultra instantaneous, in order to obtain the universal droplet distribution images, the visual field observed by microscope and the magnification should be as large as possible, the shooting rate and the field brightness at the largest resolution will not be sufficient to acquire the clear initial condensation images, so it is hardly to get the transient experimental images, even using the high speed camera.

Leach et al. [25] studied the dropwise condensation of water vapor from a naturally cooling, hot water reservoir onto a hydrophobic polymer film and a silanized glass slide by direct observation and simulations. Under these conditions, droplet growth was considerably slower than in most steady-state experiments, allowing for some phenomena (e.g., coalescence-induced nucleation, vapor depletion around individual drops) that either did not occur under steady-state conditions or occurred too rapidly for convenient characterization, just like “stretches the time scale” of experimental measurements. According to their method, we designed the microscale visualization experiments of the moist air condensation on the horizontal surfaces with different wettability:

(1) The condensing surface was polished using nylon cloth and polishing paste with grain size of 0.5 μm firstly, and then rinsed by acetone and deionized water repeatedly, until the treated surface was hydrophilic (surface A); for another condensing surface, after the same polishing and cleaning operations as surface A, it was promoted by being dipped in a 2.5 mmol/l solution of 1-octadecanethiol dissolved in ethanol and then heated to be dry at about 200 °C (Surface B). The mean contact angles on surface A and B were 73.9° and 110.8° (measured by DataPhysics OCAH200 Contact Angle Instrument, as shown in Fig. 4), respectively.

(2) Placing the condensing block fitted with PTFE insulation on the temperature-controllable platform, focusing on the condensing surface, and then adjusting the platform temperature slowly until the tiny dews appeared on the surface, at the same time, the whole process was recorded by the high speed camera.

(3) Transferring the images stored in camera to a computer and analyzing them using image analysis package.

In the present experiments, the initial moist air condensation processes were observed directly using optical method, the shooting rate was set to 125 fps, and the minimal droplet size observed was about 1 μm. Since the droplets smaller than 1 μm were hardly to be distinguished clearly using optical method, for the size distribution of the smaller droplets, the experimental images obtained by Liu et al. [5] were analyzed. They applied electron probe microanalyzer (EPMA) to analyze the variation of the chemical composition of the surfaces of monocrystalline silicon substrate sputtered a novel plating magnesium film before and after condensation of steam, which were used to deduce the initial condensation state, and obtained the initial condensation images with the initial condensation nucleus with size of 3–10 nm, as shown in Fig. 5.

## 4. Results and discussions

### 4.1. Size distribution of the droplets before coalescing (with size of several microns)

Adjusting the surface temperature, the dew point condensation images with droplet size of several microns on the surface B with contact angle of 110.8° were got, as shown in Fig. 6, with the

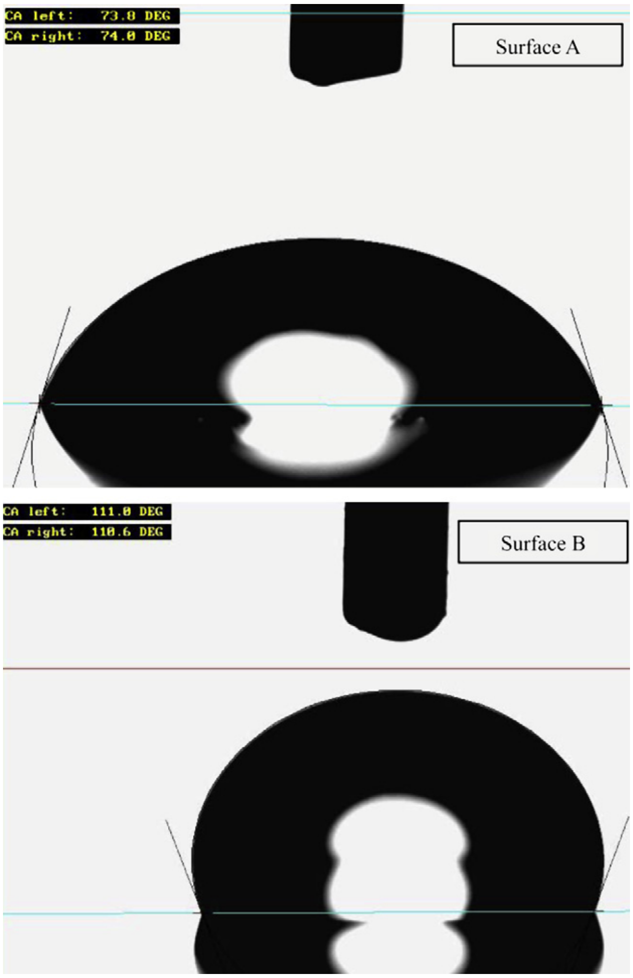

**Fig. 4.** Contact angles on surfaces A and B.

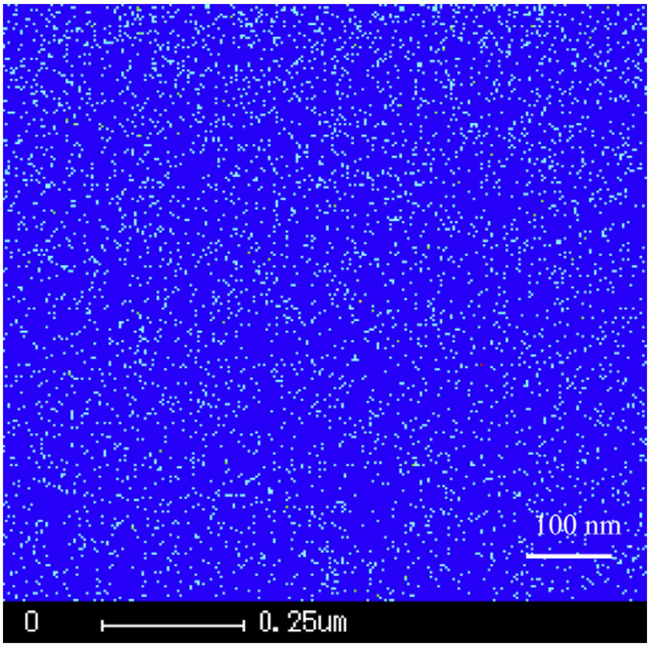

**Fig. 5.** Initial condensation image of experiments of Liu et al. [5] (8# surface: Average subcooling 4.6 K; Condensation time 20 s).

atmospheric pressure of 98.2 kPa, environmental temperature of 25.9 °C, and humidity of 75–80%, surface subcooling of about 5.5 K, and condensation time of about 20 s. It is obvious that the droplet number densities along the grooves are larger than that on the other surface area; this is agreed with the reference [26]. Although the clusters contact the cooled surface and deposit on it randomly, but their remained energies allow them to migrate on the surface in a limited area and coalesce with the clusters on their ways, then stay on the locations with higher free energies [23], such as pits and grooves. In this case, the droplet number densities in the vicinities of pits and grooves in the surface should be higher relatively. As shown in Fig. 9, the dew point condensing processes on surfaces A and B are identical before obvious coalescence occurs between droplets, so only the droplet size distributions on surface B were determined with an image analysis package, ImagePro Plus (Cold Spring, USA). The software first identifies the dark droplet zones and then calculates the area and radius of each droplet in the image based on the calibrated size scale by imaging the Olympus grid with known size, the minimum droplet radius can be resolved clearly is about 1 μm. Subsequently, the distribution of the droplets at the moment of about 20s after nucleating is generated by binning these measurements into interval of 0.5 micron, as shown in Fig. 7. The scattering points are the present experimental size distribution, and the solid line is the Lognormal distribution function. The result indicates that, the size distribution of the investigated droplets with size of several microns agrees well with the Lognormal distribution function, the total droplet number density is about $6.1 \times 10^9$ droplet/m$^2$ on the surface and the surface area fraction covered by droplets is about 20%. Due to the low growth rate of the dew droplets, these droplets are growing up by direct condensation, and no coalescence is observed. Therefore, it can be concluded that the steam molecules became clusters in the bulk vapor phase according to the reference [23].

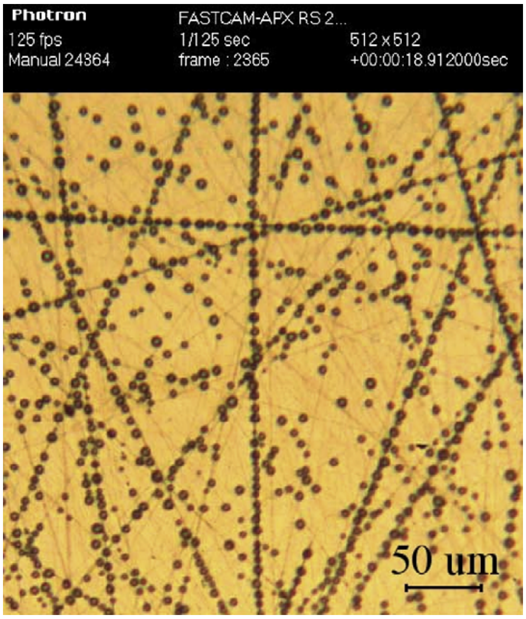

**Fig. 6.** Initial condensation image for surface B (Atmospheric pressure: 98.2 kPa; Environmental temperature: 25.9 °C; Humidity: 75–80%; Subcooling: about 5.5 K; Condensation time: about 20 s).

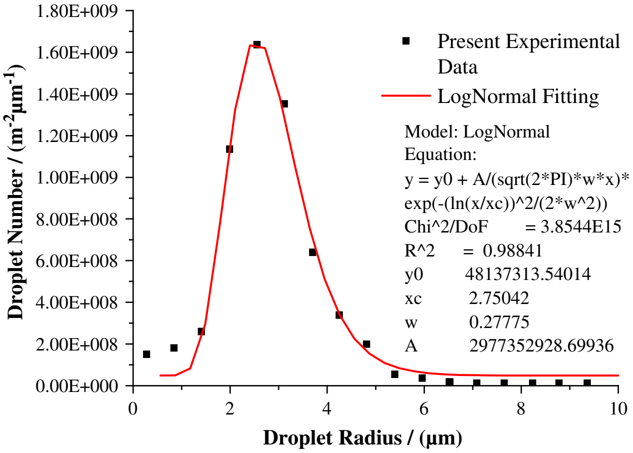

**Fig. 7.** Droplet size distribution for surface B.

## 4.2. Size distribution of the initial droplets (with size of several nanometers)

According to the thermodynamic theory, the primary condensation droplet has a size of about 10 nm [6–8], while the

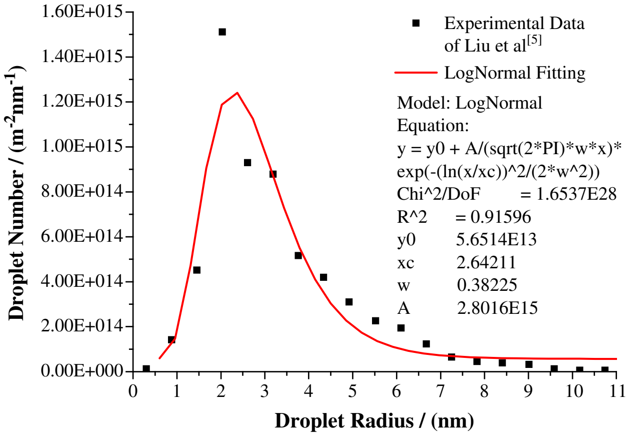

**Fig. 8.** Droplet size distribution from the experimental image in literature.

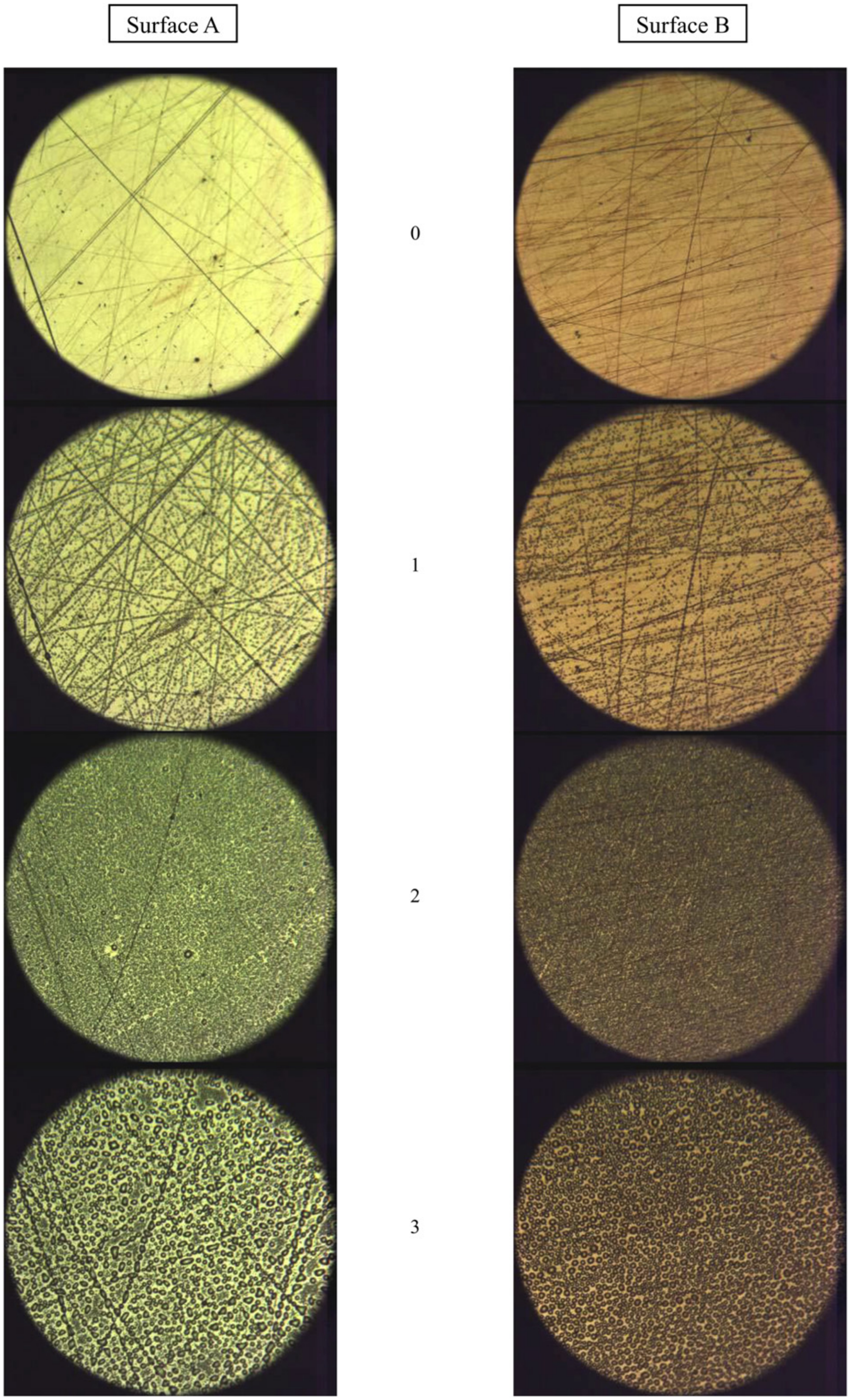

**Fig. 9.** Condensation processes on surfaces with different wettability (Atmospheric Pressure: 98.2 kPa; Environmental Temperature: 25.9 °C; Humidity: 75–80%; Subcooling: about 5.5 K; Interval about 10 s).

present experimental instrument cannot acquire the images of these droplets currently. Fortunately, Liu et al. [5] reported the experimental image of the primary droplets, as shown in Fig. 5. Image analysis method is used to obtain the size distribution (the interval is 0.5 nm), as shown in Fig. 8. The scattering points are the size distribution derived from the experimental image, and the solid line is the Lognormal distribution function. The result indicates that, the size distribution of the droplets with size of several nanometers satisfies the Lognormal distribution function too, the total droplet number density is $5.9 \times 10^{15}\ \mathrm{m}^{-2}$ and the surface covering fraction is about 2%. So, according to cluster physics theory, the conclusion again confirms the molecular clustering concept of the present physical model.

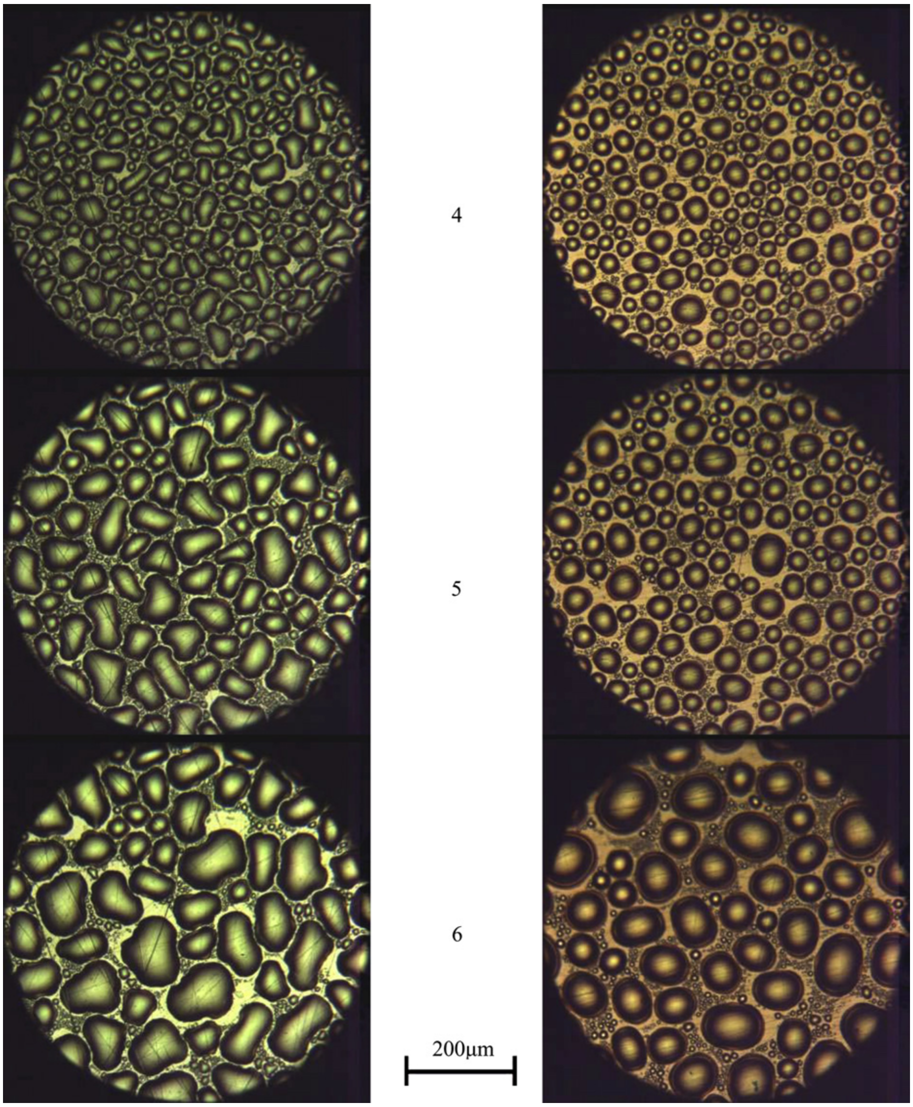

**Fig. 9.** (*continued*).

### 4.3. Comparison of the initial condensation mode on the surfaces with different wettability

Based on the present physical model, no matter how the surface wettability is, the initial mode of condensation on it should be "dropwise", i.e. the separated nuclei should be generated on the surface firstly. So the moist air condensation experiments are conducted on surfaces with different wettability resulting in different condensation modes, filmwise and dropwise. The comparison of the two condensation processes with each other is shown in Fig. 9. The images show that the initial stages of the two condensation modes are identical: nucleating firstly, then growing up through direct condensation and coalescing at critical size (Fig. 9-2). After the critical size, the discrepancy between the two surfaces occurs, the contact lines of droplets on hydrophobic surface B come back to be circular immediately after coalescing with others, for the smaller liquid–solid interaction. On the contrary, the contact lines of droplets on hydrophilic surface A are pinned and remain the shapes right after coalescing. The phenomena indicate that both filmwise and dropwise condensations have the same origin: nucleation triggered by molecular clustering, and the same pre-coalescence growing processes. The steady-state condensation mode is decided by the liquid–solid interface effect, i.e. filmwise condensation will form if the liquid–solid interaction is larger than the liquid surface tension, and the dropwise mode will be on the contrary [27]. In nature, the liquid–solid interface effect decides the condensation mode through affecting the behavior of contact line of condensate [28].

## 5. Concluding remarks

To describe the state of steam molecules in bulk steam phase before condensing onto the cooled surface, a physical model is proposed by introducing clustering concept, based on the general feature of nucleation processes. It is suggested that the steam molecules become clusters prior to the condensation on the cooled surface, the Lognormal size distribution forms resultantly. The condensation heat transfer elements are not only the steam molecular monomers, but also the molecular clusters, and they condense on the surface in the way of group by group. Based on the theory of cluster physics, the size distribution of clusters which have just deposited on substance can indicate the distribution of the clusters which are in the bulk steam phase, i.e. Lognormal distribution, if the primary droplets show a Lognormal size distribution too, the model proposed in the present paper could be proved. So the size distributions of droplets with several nanometers and several microns are investigated corresponding to the primary and pre-coalescing droplets, respectively. The following conclusions can be drawn:

(1) Based on the experimental images of the moist air condensation, the droplets with radius of several microns which are prior to coalescing have a Lognormal distribution feature, the total number density is about $6.1 \times 10^9\ \mathrm{m}^{-2}$ and array more closely in the vicinities of pits and grooves in the surface;

(2) Based on the initial condensation images of steam on magnesium film obtain by EPMA reported in literature, the primary droplets with radius of 3–10 nm have a Lognormal distribution feature also, and the total number density is about $5.9 \times 10^{15}\ \mathrm{m}^{-2}$;

(3) The comparison of moist air condensations on surfaces with different wettability show that the initial stages of the two condensation modes are identical, from nucleating to growing up through direct condensation and then to coalescing, their discrepancy just occurs after coalescing between droplets. The contact lines of droplets on hydrophobic surface B can come back to be circular immediately, while those on hydrophilic surface A are pinned and remain the shapes right after the coalescing. It can be concluded that the liquid–solid interface effect decides the condensation mode through affecting the behavior of contact line of condensate.

## 6. Outlook remarks

For the difficulty to directly observe the behaviors of steam molecules and clusters experimentally, the further investigation should be focused on the molecular dynamic simulation for the processes of steam molecules clustering and from clusters to the primary droplets on the cooled surface.

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## Appendix. The clustering evolution processes of molecules

The clustering evolution processes of molecules are similar with the reaction kinetics with corresponding reaction rates [18,22]. This kinetic approach to clustering is also obtained with adequate assumptions of the kinetic rate coefficients, $k_{mn}$:

$$A_m + A_n \xrightarrow{k^+_{mn}} A_{mn}$$

$$A_{m+n} \xrightarrow{k^-_{mn}} A_m + A_n$$

where $A_i$ means the cluster composed of $i$ molecules, $i$ is from 1 to infinity, $mn$ means a loose cluster construction, $m+n$ means a compact cluster construction. The reaction rate or formation rate of cluster $[A_{m+n}]^\circ$ is given by:

$$[A_{m+n}]^\circ = k^+_{mn}[A_m][A_n] - k^-_{mn}[A_m][A_n]$$

where $[A]$ is the number density of cluster $A$. The kinetic rate coefficient $k$ can be given by

$$k^+_{mn} = \iint P_{mn}\sigma_{mn}|\upsilon_m - \upsilon_n| f(\upsilon_m) f(\upsilon_n)\,\mathrm{d}\upsilon_m \mathrm{d}\upsilon_n$$

$$\sigma_{mn} = \pi(r_m + r_n)^2$$

where $\upsilon_m$ is the translational velocity of cluster $m$, $f(\upsilon_m)$ the velocity distribution of cluster $m$, $\sigma_{mn}$ is the collision cross-section, $P_{mn}$ is the formation probability of $A_{mn}$ at the collision of $A_m$ and $A_n$, $P_{mn}$ can be predicted analytically with the molecular dynamics simulation or estimated empirically with experimental data, and $r_m$ and $r_n$ are the radius of cluster $m$ and $n$, respectively. The reverse reaction rate coefficient $k^-_{mn}$ can be obtained by the equilibrium condition of the reaction in the same manner as the forward chemical reaction.

So the total formation rate of cluster $i$ is then given by:

$$\begin{aligned}[A_i]^\circ = {} & \sum_{m=1}^{i-1} k^+_{mn}[A_m][A_n]_{(n=i-m)} - \sum_{m=1}^{i_{\max}-i} k^+_{mi}[A_m][A_i] \\ & - \sum_{m=1}^{i-1} k^-_{mn}[A_i]_{(n=i-m)} + \sum_{m=i+1}^{i_{\max}} k^-_{mn}[A_m]_{(n=m-i)}\end{aligned}$$

On the right side of the above equation, the four terms are the formation rate from association of smaller clusters, the growth rate, the dissociation rate, and the formation rate from the dissociation of larger clusters of cluster $A_i$, respectively.

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