

Comparative study of homogeneous nucleation rate models for wet steam condensing flows

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Abstract—To accurately describe the homogeneous nucleation process in a wet-steam dual-phase flow and to improve the reliability of computations of the condensing steam flow, classical homogeneous nucleation theories were derived and summarized based on the molecular agglomerate thermodynamic free energies. To obtain more accurate homogeneous nucleation rates, various modified homogeneous nucleation rate models were described. Experimental data were used in a comparative study of these models to select a nucleation rate model to provide calculated values for engineering projects. This study indicates that the main difference between different nucleation theories lies in the difference in the influence of temperature on the nucleation rate.

Keywords: Two-phase Flow, Homogeneous Nucleation Rate, Wet Steam, Condensing Flows, Thermodynamics, Nucleation Temperature

INTRODUCTION

Issues, such as the degradation of flow efficiency and reduced economic benefits of mechanical systems, occur when a turbine flow path is under long-term water corrosion; the blades working under long-term wet-steam working conditions may also break [1]. Baumann noted in 1910 that a 1% increase in wetness at a low-pressure stage of a steam turbine induces a 1% reduction in its efficiency [2]. If the pure steam is not affected by foreign particles from the external environment and is away from solid surfaces, the steam will not condense even if it expands beyond the saturation threshold, but it will instead continue to expand to form super-saturated steam. Large amounts of tiny water droplets will form that accompany the continued steam expansion, which will restore the steam to a balanced state; the duration of nucleation depends on the degree of super-saturation. The size distribution of the water droplets is the main influential factor for the characteristics of a wet-steam flow, and studying the variation pattern of the nucleation rate is the key to understanding the wet-steam properties.

According to classical thermodynamics theory, the condensation nuclei satisfy the Boltzmann distribution, and based on this, the critical free energy and critical radius can be calculated. Using this method, Volmer and Weber derived a nucleation equation to calculate the nucleation rate in 1926 [3]; thereafter, Becker and Döring modified and perfected the equation to form the classical nucleation theory [4]. The nucleation rate model proposed by Lothe et al. resulted in a calculated value that is 17 orders of magnitude higher than that based on the classical nucleation theory [5]. Feder analyzed these two nucleation rate theories and noted that the

results that follow Lothe's model are not consistent with the experimental data; however, due to the limitations of the experimental conditions, they could only present semi-qualitative results. Later, Courtney proposed a non-steady state nucleation model, which was based on fewer assumptions, and it improved the accuracy of the calculation of the nucleation rate [6]. Even after over a century of development, a completely reliable nucleation model is still not available; engineering projects still use the classical Becker-Döring theory.

To obtain a more accurate homogeneous nucleation rate, we addressed the free energy during the formation of molecular aggregation from a thermodynamics aspect and derived a classical homogeneous nucleation model. Additionally, we comparatively investigated various homogeneous nucleation rate models combined with experimental data. The suitable application ranges of different models were obtained, which provide a reference for wet-steam condensate flow simulations.

DYNAMIC EQUATIONS FOR NUCLEATION

The molecular aggregation and separation process in supersaturated steam is similar to that in overheated steam; the difference is that the molecular aggregation size in supersaturated steam exceeds the critical radius, and the aggregation grows gradually due to the Gibbs free energy. Under this condition, the distribution equation for steady-state molecular aggregation can no longer accurately describe the growth and decay of the molecular aggregation. A molecular aggregation, g , can collide and combine with another molecular aggregation of any size, or it can split into smaller aggregations. However, because the number of poly-molecular aggregations in low-pressure steam is far less than the number of individual molecules, collisions between poly-molecular aggregations rarely occur [7,8]. Thus, the growth and decay of the molecular aggrega-

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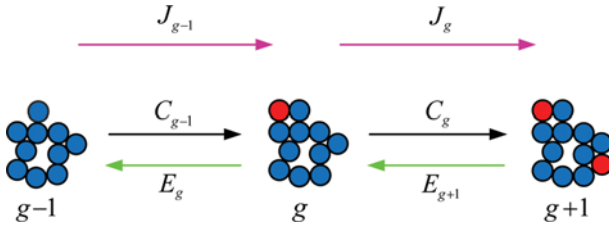


Fig. 1. The dynamic process of nucleation.

tion is assumed to occur through the capturing and losing of single molecules. Fig. 1 shows the dynamic process of the growth and decay of the molecular aggregation.

The net growth rate at which the molecular aggregation per unit volume of steam (g) changes to molecular aggregation ($g+1$) is defined as the nucleation rate, which is expressed as J_g .

$$J_g = C_g f_g - E_{g+1} f_{g+1} \quad (1)$$

where, C_g is the condensation speed of the molecular aggregation g , f_g is the amount of molecular aggregation g per unit volume of steam, and E_g is the evaporation speed of the molecular aggregation ($g+1$).

When solving for the nucleation rate using Eq. (1), the expressions of C_g and E_g must be known. C_g is related to the surface area of the molecular aggregation, molecule collision rate, and condensation coefficient. When approximating the molecular aggregation g as a sphere, the following equation is true:

$$C_g = q_c a_0 g^{2/3} \frac{p}{\sqrt{2\pi m k T_g}} \quad (2)$$

In this equation, q_c is the condensation coefficient, a_0 is the single molecule surface area (m^2), p is steam pressure (Pa), m is the mass of a single molecule (kg), k is the Boltzmann constant, and T is the gas phase temperature (K).

The aggregation evaporation speed satisfies the following equation:

$$E_g = q_e a_0 g^{2/3} \frac{p_s(T_r)}{\sqrt{2\pi m k T_r}} \exp\left(-\frac{2\sigma}{\rho_l R T_r}\right) \quad (3)$$

Here, q_e is the evaporation coefficient, T_r is the temperature of the molecular aggregation, σ is the surface tension of the molecular aggregation (N/m), ρ_l is the liquid-phase density (kg/m^3), and R is the gas constant of water steam.

Because the molecular aggregation is in an equilibrium state, the equation, $C_g n_g = E_{g+1} n_{g+1}$, is satisfied, where n_g and n_{g+1} are the number of molecular aggregations g and ($g+1$) per unit volume of steam, respectively. Therefore, the following relation can be obtained from Eq. (1):

$$\frac{J_g}{C_g n_g} = \frac{C_g f_g}{C_g n_g} - \frac{E_{g+1} f_{g+1}}{E_{g+1} n_{g+1}} = \frac{f_g}{n_g} - \frac{f_{g+1}}{n_{g+1}} \approx -\frac{\partial}{\partial g} \left(\frac{f_g}{n_g} \right) \quad (4)$$

The rate of change for the number of molecular aggregations with respect to time is:

$$\frac{\partial f_g}{\partial t} = J_{g-1} - J_g = -\frac{J_g - J_{g-1}}{\Delta g} \approx \frac{\partial J_g}{\partial g}, \Delta g = 1 \quad (5)$$

By substituting Eq. (4) into Eq. (5) to cancel out J_g , the governing differential equation for homogenous nucleation is obtained:

$$\frac{\partial f_g}{\partial t} = \frac{\partial}{\partial g} \left[C_g n_g \frac{\partial}{\partial g} \left(\frac{f_g}{n_g} \right) \right] \quad (6)$$

The literature [9] indicates that when the nucleation rate can reach a steady state in less than $1 \mu s$, the transient state can be ignored. Thus, in the calculations for a condensing flow, the nucleation rate is generally viewed as instantaneously reaching a steady state. The molecular aggregation radius in the steady state can be obtained from Boltzmann's law:

$$n_g \approx n_1 \exp\left(-\frac{\Delta G}{k T_g}\right) \quad (7)$$

Here, n_g and n_1 are the total number of molecular aggregations g and single molecules per unit volume of steam, respectively, and ΔG is the Gibbs free energy.

Eq. (8) is obtained by integrating Eq. (4) on both sides:

$$J_{CL} \int_{g=1}^{\infty} \frac{dg}{C_g n_g} = \int_{g=1}^{\infty} -d\left(\frac{f_g}{n_g}\right) \quad (8)$$

where J_{CL} is the classical steady-state nucleation rate ($/kg \cdot s$).

The right hand side of Eq. (8) has two extreme conditions:

$$\begin{cases} g \rightarrow 1, f_g/n_g = 1 \\ g \rightarrow \infty, f_g/n_g = 0, J_{CL} = \left[\int_{g=1}^{\infty} \frac{dg}{C_g n_g} \right]^{-1} \end{cases} \quad (9)$$

Eq. (10) is obtained by substituting Eq. (7) into Eq. (8):

$$J_{CL} = \left[\int_{g=1}^{\infty} \frac{1}{C_g n_1 \exp[-\Delta G/k T_g]} dg \right]^{-1} \quad (10)$$

In Eq. (10), C_g is proportional to $g^{2/3}$. Compared with the exponential term, the change in C_g is very slow, whereas the exponential term decreases rapidly as g increases. When the denominator reaches the minimum value, the exponential term will increase as g increases. The maximum value of the integrand corresponds to the minimum value of the denominator, and it has a very sharp peak [10]. Therefore, C_g can be approximated by C_{g^*} . Eq. (11) is obtained by expanding ΔG in a Taylor expansion with respect to g :

$$\Delta G = \Delta G^* + (g - g^*) \left(\frac{\partial \Delta G}{\partial g} \right)_{g=g^*} + \frac{(g - g^*)^2}{2} \left(\frac{\partial^2 \Delta G}{\partial g^2} \right)_{g=g^*} + \dots \quad (11)$$

By keeping up to the second order term, because $(\partial \Delta G / \partial g)_{g=g^*} = 0$, Eq. (10) can be simplified as:

$$J_{CL} = C_{g^*} n_1 Z \exp\left(-\frac{\Delta G^*}{k T_g}\right) \quad (12)$$

where Z is the Zeldovich factor, and it is expressed as [11,12]:

$$Z = \left\{ \int_{g=1}^{\infty} \exp\left[\frac{(g - g^*)}{2 k T_g} \left(\frac{\partial^2 \Delta G}{\partial g^2} \right)_{g=g^*} \right] dg \right\}^{-1} \approx \left[\frac{-1}{2 \pi k T_g} \left(\frac{\partial^2 \Delta G}{\partial g^2} \right)_{g=g^*} \right]^{1/2} = \frac{1}{3} \left(\frac{\theta}{\pi} \right)^{1/2} (g^*)^{-2/3} \quad (13)$$

MODIFICATIONS OF THE NUCLEATION MODEL

1. Classical Model

The classical nucleation model assumes that the temperatures of all water droplets are the same as the gas-phase temperature, and it does not consider the condensation latent heat produced by the phase transformation. For wet air flow, the isothermal assumption of the classical nucleation theory is reasonable. The classical nucleation rate per unit mass is:

$$J_{CL} = q_c \frac{\rho_g^2}{\rho_l} \left(\frac{2\sigma}{\pi m^3} \right)^{1/2} \exp\left(-\frac{4\pi r^* \sigma}{3kT_g}\right) \quad (14)$$

2. Non-isothermal Modified Model

The classical nucleation theory takes for granted that the temperatures in both the gas and liquid phases are the same, $T_r = T_g$, and the latent heat released by the condensation of gas-phase molecules is not considered. If a large amount of stagnant gases exists in the space, such as a diffusion cloud chamber or wet air flow in a nozzle, the isothermal assumption is reasonable. However, the temperature difference between the two phases of the wet-steam flow cannot be ignored. As known from Eq. (3), the temperature for molecular aggregation has a great influence on the evaporation speed; thus, the non-isothermal effect has an influence on the nucleation rate.

Although the non-isothermal effect is very important, relevant research on the non-isothermal nucleation process seldom takes into account the energy transfer. Kantrowitz et al. derived a modified relation equation on the basis of isothermal nucleation theory [13]. Afterward, Feder verified this model and performed more detailed analysis [14]. Wyslouzil and Seinfeld studied this problem indepth and concluded that the governing equation for the condensation speed is related to the energy transfer in small-scale aggregations [15]. However, current studies still have difficulty precisely describing a non-isothermal nucleation process.

The Kantrowitz non-isothermal nucleation model places a multiplier on the classical nucleation model, and the expression is:

$$J_{NCL} = \psi J_{CL} = \frac{J_{CL}}{1 + \eta} \quad (15)$$

The expression for η is:

$$\eta = q_c \frac{\rho_g R}{\alpha \sqrt{2\pi RT_g}} \frac{h_{fg}}{RT_g} \left(\frac{h_{fg}}{RT_g} - \frac{1}{2} \right) \quad (16)$$

where α is the heat-exchange coefficient for the surface of the molecular aggregations with radius r^* (W/(m²·K)), and h_{fg} is the latent heat of vaporization (kJ/kg).

3. Courtney Model

The influence of the partial pressure during molecular aggregation is not considered in the classical nucleation rate model. One study [16] modified the model with the aggregation free energy:

$$\Delta G = a_0 \sigma g^{2/3} - kT_g(g-1)\ln S \quad (17)$$

Using Eq. (17), the nucleation model can be modified as:

$$J_{CN} = q_c \frac{\rho_g \rho_{gs}}{\rho_l} \left(\frac{2\sigma}{\pi m^3} \right)^{1/2} \exp\left(-\frac{4\pi r^* \sigma}{3kT_g}\right) \quad (18)$$

In this equation, ρ_{gs} is the saturation density that corresponds to the steam temperature T_g . The quantity of ΔG in the classical nucleation theory does not satisfy the law of mass action; however, Eq. (17) after the modification does satisfy the mass consistency, and it satisfies the requirement of Kashchiev's first nucleation theorem [17]. Currently, the modification by Courtney is accepted and verified by physicists [18].

4. An Internally Consistent Modified Model

A careful examination of Eq. (17) reveals that when g equals 1, the free energy during molecular aggregation is $\Delta G = a_0 \sigma$ and not $\Delta G = 0$. To restore the ΔG consistency, Girschik and Chu proposed an internally consistent modification method [19]:

$$\frac{\Delta G}{kT_g} = \frac{a_0 \sigma}{kT_g} (g^{2/3} - 1) - (g-1)\ln S = \theta (g^{2/3} - 1) - (g-1)\ln S \quad (19)$$

After modification via this method, the nucleation model has an additional modification factor of e^θ , and the modified nucleation model becomes:

$$J_{ICCT} = q_c \frac{\rho_g \rho_{gs}}{\rho_l} \left(\frac{2\sigma}{\pi m^3} \right)^{1/2} \exp\left(\theta - \frac{4}{27} \frac{\theta^3}{(\ln S)^2}\right) \quad (20)$$

When T_g equals 300 K, the value of the modification factor e^θ is approximately 10^3 . Thus, the modification accuracy of the internally consistent classical theory is questionable. The argument among scholars is the following: Is it really necessary to make the distribution of every single molecule approach the actual situation? Because the molecular aggregation can only be considered to be a droplet when its size reaches a certain scale, modifying ΔG to satisfy the consistency conditions for extreme cases may be meaningless [11]. However, Katz believes that e^θ is a simple modification of the impact of the droplet radius on the surface tension [20].

5. A Modified Model for the Empirical Function

Although the calculation results from the classical nucleation model are quite consistent with the experimental data, an offset of a certain degree still exists. Wölk and Strey analyzed the low-temperature experimental data, and they constructed an empirical function to modify the classical nucleation model; the specific form is [21]:

$$J_{WS} = J_{CL} \exp\left(-27.56 + \frac{6.5 \times 10^3}{T_g}\right) \quad (21)$$

Because the nucleation rate in this model is derived from water vapor nucleation experimental data at $T_g = 220$ – 260 K, the calculation results fit the data very well in this range, but the rationale under other conditions has yet to be verified.

RESULTS AND DISCUSSION

Currently, a generally applicable nucleation rate model does not exist, and many scholars accept this fact. Wilemski conducted an extensive indepth study of the existing nucleation models [22] and concluded that a completely reliable nucleation model for computing the nucleation rate that is applicable to all working conditions does not exist. We studied the calculation accuracy of different models by comparing the theoretical calculations and experimental results. The parameters that describe the thermal physical prop-

erty of water were acquired from the literature [23], and the experimental data for the nucleation rate were acquired from references [23]. The condensation coefficient q_c was 1, and the steam temperature was between 220 K and 260 K. The calculated results were compared with pulse chamber test data, and the results are shown in Fig. 2.

As shown in Fig. 2, the calculated value for J_{CN} is 0.1 times that for J_{CL} , and the calculated value for J_{ICCT} is 10^3 – 10^4 times that for J_{CL} . The internally consistent classical theory has quite a large deviation, and its authenticity requires further study. Meanwhile, the

calculated value for the nucleation rate by J_{NCL} is 1/200–1/100 of J_{CL} , which is in agreement with the results in the literature [12]. The calculated average values for the Wölk-Strey nucleation rate, J_{NCL} , under all work conditions are close to the experimental values. When T_g equals 260 K and 250 K, the calculated values for J_{WS} are similar to those for J_{CN} , and both are 1/10th of the calculated values for J_{CL} , as shown in Figs. 2(a) and (b). When T_g equals 240 K, the calculated values for J_{WS} are completely consistent with the experimental data, as shown in Fig. 2(c). When T_g equals 230 K, the calculated values for J_{WS} and J_{CL} are on the same order of mag-

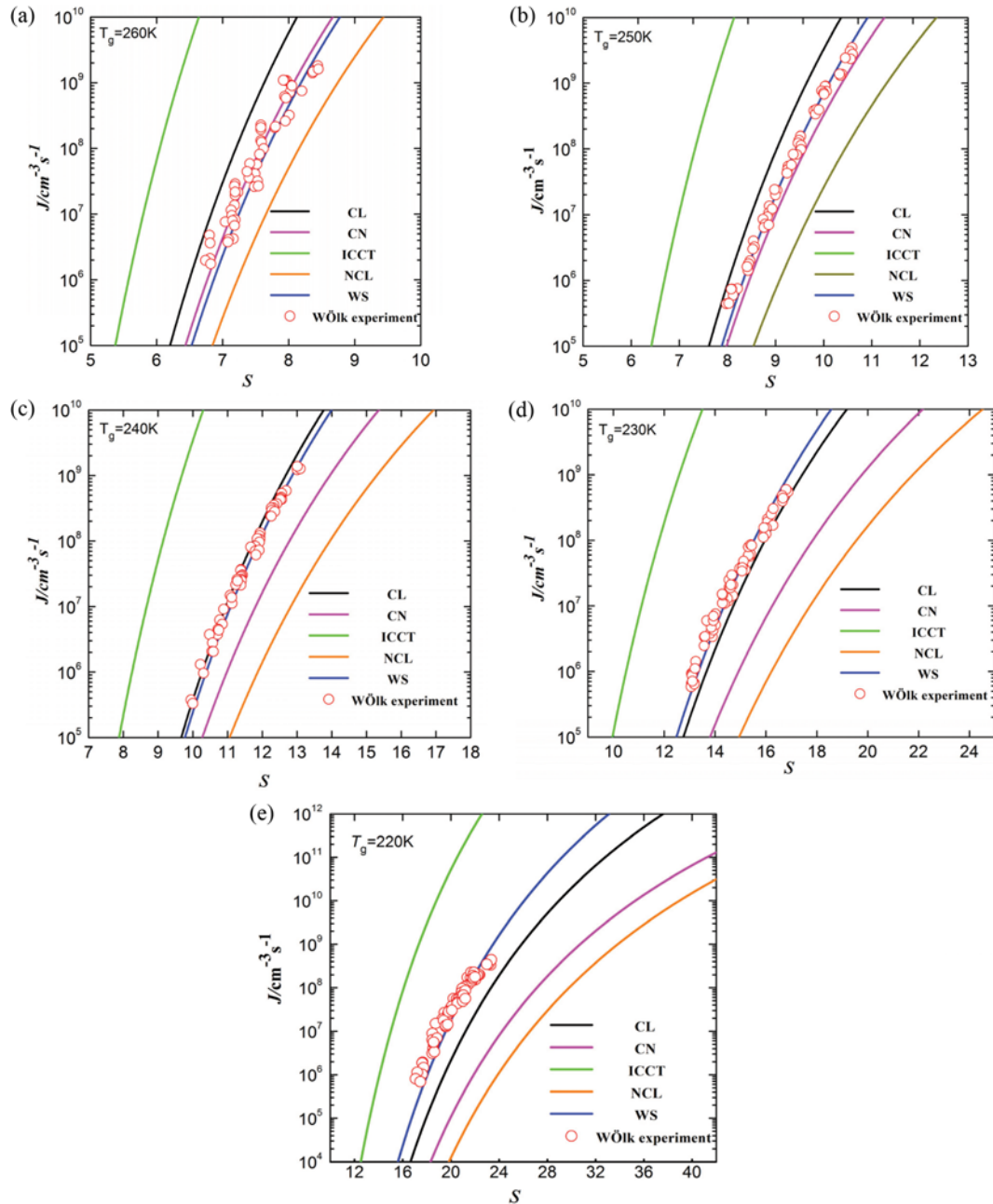


Fig. 2. Comparison of the calculated results of the nucleation rates using various water-steam nucleation models. The nucleation rate given by Eq. (14) with J_{CL} is the black curve. The nucleation rate given by Eq. (15) with J_{NCL} is the brown curve. The nucleation rate given by Eq. (18) with J_{CN} is the pink curve. The nucleation rate given by Eq. (20) with J_{ICCT} is the green curve. The nucleation rate given by Eq. (21) with J_{WS} is the blue curve. Wölk experimental data is from reference [23].

nitude (see Fig. 2(d)). When T_g equals 220 K, the difference between the calculated values for J_{WS} and J_{CL} becomes larger, and the difference increases as supersaturation S increases. Meanwhile, the calculated values for J_{WS} are approximately ten-times those for J_{CL} . This is the opposite from when T_g equals 260 K and 250 K, and the calculated values for J_{WS} show the trend of approaching, as shown in Fig. 2(e).

According to the calculated results, the calculated values for J_{CL} completely coincide with the experimental data when $T_g=240$ K. The reason for such highly accurate predictive values from the classical nucleation theory under these working conditions is worth pondering. Under these working conditions, further studies are required to determine which part of the free energy during the molecular aggregation is canceled and in what manner it is can-

celed. As T_g increases, the calculated values for J_{CL} begin to deviate from the experimental values, and the deviation becomes ever greater. When T_g is higher than 240 K, as the supersaturation S increases, the deviation of J_{CL} from the data becomes ever greater. Generally, the calculated values for J_{CL} are greater than the experimental data. When T_g is lower than 240 K, the calculated values for J_{CL} also deviate from the data, but they are lower than the experimental values.

When the steam temperature is fixed, the slopes of the J - S curves of different nucleation models are very similar. This indicates that the main influential factor on different models is the steam temperature, and this influence is barely reflected in the J - S curves. Therefore, the S - T_g curves (Fig. 3) were plotted to study the impact of the temperature on the nucleation rate.

Fig. 3 shows the relation between the supersaturation S and temperature T_g when the nucleation rates are $J=10^0 \text{ cm}^{-3} \text{ s}^{-1}$ and $J=10^4 \text{ cm}^{-3} \text{ s}^{-1}$. The data in Fig. 3(a) came from the literature [24], and those in Fig. 3(b) came from references [25] and [26]. The study indicates that when T_g is lower than 240 K, the supersaturation values calculated by the Courtney nucleation model are in good agreement with the experimental data. However, when T_g is higher than 240 K, the calculated values slightly deviate from the experimental values. Under this condition, the calculated values for J_{NCL} from the non-isothermal nucleation model and for J_{WS} from the Wölk-Strey nucleation model better match the experimental values. Thus, when T_g is higher than 240 K, the J_{WS} from the Wölk-Strey nucleation model achieves good results for both nucleation prediction and supersaturation prediction. In addition to the internally consistent nucleation model, J_{ICCT} the calculated values from all other models deviate only slightly from the experimental values. However, because the J - S curve is very steep, a very small supersaturation variation can lead to a larger deviation in the nucleation rates.

In the wet-steam flow in the final stage of a steam turbine, the temperature during the nucleation process is 320 K. As suggested by the above analysis, the selection of the Wölk-Strey nucleation model for calculating J_{WS} and the selection of the non-isothermal nucleation model for calculating J_{NCL} are both more reasonable.

CONCLUSIONS

For molecular aggregations that contain a liquid core, the treatment in this paper is generally applicable. Nucleation dynamics were utilized to derive a classical homogeneous nucleation theory. Various forms of modifications for the classical Becker-Döring theory were described to obtain a homogeneous nucleation rate model with higher accuracy. A comparison of different homogeneous nucleation models in combination with experimental data was conducted, and the following conclusions were made:

1. In the temperature range of 220-260 K, the J_{CN} calculated values are approximately 1/10 of the classical nucleation rate, J_{ICCT} is 10^3 - 10^4 times J_{CL} , and J_{NCL} is 1/2000-1/100 of J_{CL} .
2. In the temperature range of 220-260 K, J_{WS} is precisely consistent with experimental values. When T_g equals 260 K and 250 K, J_{WS} is approximately 1/10 of J_{CL} , and when T_g equals 240 K, the calculated values for J_{WS} are exactly the same as J_{CL} . When T_g equals 230 K, the calculated values for J_{WS} are greater than the calculated

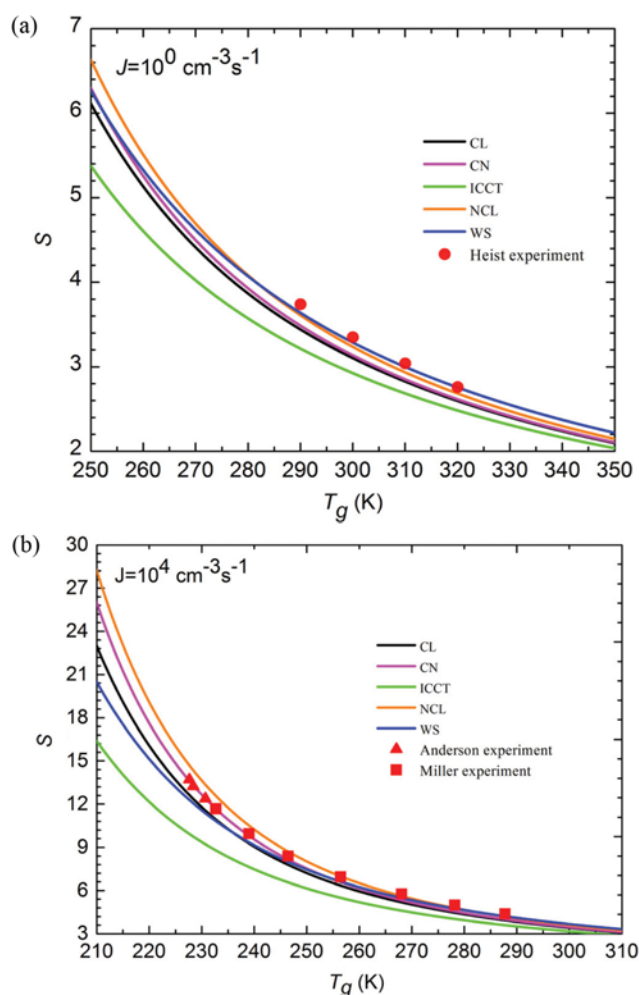


Fig. 3. Comparison of the calculated results of supersaturation for a fixed nucleation rate. The nucleation rate given by Eq. (14) with J_{CL} is the black curve. The nucleation rate given by Eq. (15) with J_{NCL} is the brown curve. The nucleation rate given by Eq. (18) with J_{CN} is the pink curve. The nucleation rate given by Eq. (20) with J_{ICCT} is the green curve. The nucleation rate given by Eq. (21) with J_{WS} is the blue curve. Heist experimental data is from reference [24]. Anderson experimental data is from reference [25]. Miller experimental data is from reference [26].

values for J_{CL} , but they are still on the same order of magnitude. When T_g equals 220 K, the calculated values for J_{WS} are approximately 10 times those for J_{CL} .

3. When T_g is higher than 240 K, J_{CL} deviates from the experimental values, and the deviation increases as the temperature increases. When T_g is lower than 240 K, as the temperature decreases, the calculated values for J_{CL} become smaller than the experimental ones; however, when T_g equals 240 K, the calculated values for J_{CL} are precisely consistent with the experimental values.

4. The steam temperature has a large influence on the nucleation rate. When T_g is lower than 240 K, the supersaturation values calculated by the Courtney nucleation model are more consistent with the experimental values. However, when T_g is higher than 240 K, the supersaturation values calculated using the Wölk-Strey model and the non-isothermal nucleation model better match the experimental values.

5. For wet-steam flow in the final stage of a steam turbine, the selection of the Wölk-Strey nucleation model and the non-isothermal nucleation model for performing the calculations is more reasonable. Under medium temperature conditions, the Wölk-Strey nucleation model has higher accuracy, and under low temperature conditions, the calculated values for J_{ICCT} are closer to the experimental values.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this paper.

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