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# Heterogeneous condensation on insoluble spherical particles: Modeling and parametric study

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## HIGHLIGHTS

- A full model for the whole heterogeneous condensation processes is developed.
- Two new criteria for condensation ability of a system are proposed.
- The relations of the condensation ability with initial conditions are evaluated.
- A stronger condensation ability can be achieved at the medium initial conditions.

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## ABSTRACT

A full heterogeneous condensation model is developed to describe the water vapor condensation on an insoluble spherical particle. The new model divides the whole process of the heterogeneous condensation into three stages: nucleation, transition and growth, and considers the conservation of mass and energy of system. The model is incorporated with the population balance equation and results in a numerical method, and validated by comparison with the experimental data. In addition, two new criteria are proposed to evaluate the ability of heterogeneous condensation. The effects of particle properties and initial conditions on the whole process of heterogeneous condensation are then evaluated. It is found that the relations of the condensation ability with the wetting degree, the particle size or the initial saturation are all different from the monotonous tendency as found in the classical heterogeneous condensation theory. The parametric study suggests that a stronger ability of heterogeneous condensation can be achieved with a medium value of the wetting degree, the particle size, the initial saturation or the initial temperature.



## 1. Introduction

The condensation from vapor to droplet is usually divided into three steps (Wagner, 1982): nucleation, growth and coagulation. For a single droplet, only the former two steps should be considered. The condensation can be activated by two ways, namely heterogeneous condensation and homogeneous condensation. The heterogeneous condensation which takes place on particles is much common in daily life, because the energy barrier is lower than that for homogeneous condensation which occurs in the interior of a uniform substance (Kozisek et al., 2004). Heterogeneous condensation plays an important role in many fields such as atmospheric physics (Mason, 1971; Maattanen et al., 2007), crystal study (Liu et al., 1997; Chow et al., 2002) and gas cleaning technology (Heidenreich and Ebert, 1995; Yan et al., 2011). Especially in the separation of submicron particle from gas, the heterogeneous condensation as a preconditioning technique can improve the particle removal efficiency substantially (Schaber, 1995; Johannessen et al., 1997; Heidenreich et al., 2000; Ehrig et al., 2002).

Study on heterogeneous condensation has a very long history. Volmer (1929) examined the heterogeneous nucleation on an insoluble flat surface in detail and proposed the classical heterogeneous nucleation theory. Then Twomey (1959) confirmed Volmer's theory through experiments. Fletcher (1958) established a theory by extending Volmer's theory to spherical particles. In the study of droplet growth, Wagner (1982) developed a first-order theory of droplet growth and reviewed the results of various experimental investigations. Gyarmathy (1982) combined the growth models in the continuum limit and in the free molecular limit into a new growth model, which is valid for the arbitrary droplet radius.

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In spite of the continuous advance and the wide application for the study of heterogeneous condensation, the whole process of heterogeneous condensation on insoluble spherical particles has not been systematically considered yet. One difficult is how to bridge nucleation to growth by including the transition process. The transition process in heterogeneous condensation is very complex for simulation, because the spherical cap shape makes it difficult to calculate the growth rate of embryos, and the co-existence of the former embryos growth and the new embryos appearance complicates the process further. In the literature, there are some models to deal with the whole process of heterogeneous condensation approximately. Liu et al. (1997) proposed a two-dimensional kinetic process to simulate the whole heterogeneous condensation of crystals. Heiler (1999) constructed a three-dimensional model of heterogeneous condensation on particles, in which the formation and growth of embryos are assumed to occur on a flat surface to simplify the calculation. However, this model still has some limitations especially when embryo's radius is not much smaller than particle's. The transition stage was actually considered by Tammaro et al. (2012), but the number of embryos formed and the velocity of their growth were very large in their experimental conditions so that the transition stage was neglected. As a result, it was assumed by Tammaro et al. (2012) that heterogeneous condensation on particles will be activated once the condition reaches the critical saturation, and will continue the same way as the growth of a homogenous liquid droplet whose size is equal to the particle's. As already claimed by Tammaro et al. (2012), this model may fail to describe the first instants of embryos growth when nucleation rate or the wetting degree of particle is small.

In this paper, we will develop a full model to include the whole process of heterogeneous condensation on insoluble spherical particles in a closed and adiabatic system. By dividing the whole process of the heterogeneous condensation into three stages: nucleation, transition and growth, the new model bridges the gap between the nucleation and the growth in the heterogeneous condensation. Instead of the classical criteria, two new criteria for condensation ability are proposed based on the new model. We shall evaluate the effects of particle properties and initial conditions on the whole process of heterogeneous condensation by comparing new results with the classical model predictions. In Section 2, the new model is described in detail. The numerical method of our model is introduced in Section 3. Validation of the new model, and two new criteria for condensation ability are presented in Section 4. In Section 5, the effects of particle properties and initial conditions on condensation are then evaluated by our model. A brief summary of our model and results is given in Section 6.

## 2. Physical model

We firstly present the assumptions in our physical model. Then the three stages of the heterogeneous condensation are introduced in details. At last, the classical theories of heterogenous nucleation and droplet growth, which are used in our model, are described briefly.

### 2.1. Assumptions

The process of heterogeneous condensation on insoluble spherical particles is analyzed based on the following assumptions:

(1) The mixture gas is thermally and calorically perfect, which is reasonable for the considered condition with temperature and pressure, and the volume occupied by the liquid phase is neglected (a good approximation for a small wetness).

(2) The particles are uniformly distributed (every particle is static, the interaction between particles and the effect of gravity are not considered). As a result, it is reasonable to consider a single particle in a finite volume $V_c$ filled with the mixture gas.

(3) The distance between particles is large enough compared with the particle size. Typical concentrations of atmospheric particles are in the range of $10^4\,\mathrm{cm}^{-3}$ in rural and $10^6\,\mathrm{cm}^{-3}$ in urban regions (Schaber, 1995). Therefore, the distance between particles i.e. 100 μm is much larger than the particle size in our consideration which is less than μm-scale. As a result, the finite volume $V_c$ containing a single particle does not exchange mass nor energy with its surroundings, i.e. the conservation of mass and energy in this volume can be applicable, which may lead to the depletion of vapor and the increase of temperature during condensation.

(4) Assuming that the heterogeneous particle is smooth, the nuclei are formed uniformly on the particle surface.

(5) The contact angle $\theta$ between the embryo and the particle is constant when the embryo grows.

Based on the assumptions (2) and (3), it is reasonable to analyze the condensation process on a single particle in the closed volume $V_c$ with the conservation of mass and energy.

### 2.2. Three stages of heterogeneous condensation

In our model, the whole process of heterogeneous condensation on insoluble spherical particles is divided into three stages as sketched in Fig. 1. The first stage is heterogeneous nucleation, which is the beginning of the condensation process. In this stage, a number of embryos with critical radius $r^*$ appear on the particle surface (Note that the superscript '*' denotes critical parameters hereafter.). Based on the nucleation theory of Fletcher (1958), the increasing rate of embryo's number is the nucleation rate. The second stage is the transition from nucleation to growth. In this stage, the former embryos continue to grow and new critical embryos are formed simultaneously. We assume that all embryos can be born anywhere which is around the nucleating particle and there is no intergrowth between embryos. The last stage is the growth of the droplet. When the total surface area of the particle is covered by embryos, it can be assumed that a thin liquid film is formed uniformly on the particle surface. In this stage, the condensation can be considered as the growth of a homogenous liquid droplet which contains the solid particle and the liquid film (Tammaro et al., 2012).

The produce of embryos (liquid) will lead to the depletion of vapor and the release of latent heat. As a result, the saturation and the temperature of ambient condition will change during the condensation process. When the saturation reduces to unit, the condensation will be terminated. Based on the division of condensation process in our model, if the total surface area of the particle can be covered by embryos in the process of condensation, all three stages occur. Otherwise, the process of condensation only contains the stages of nucleation and transition. Therefore, a


**Fig. 1.** Three stages of the whole process of heterogeneous condensation on an insoluble spherical particle.