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THE DYNAMICS OF CONDENSATION NUCLEI IN A THERMAL DIFFUSION CHAMBER†

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The evolution of a spherical liquid drop formed by homogeneous nucleation inside a thermal diffusion chamber is investigated. The equation of motion of the drop in non-uniform temperature fields and the composition of the gaseous mixture are obtained, taking into account the phase transition at the drop-gas interface as well as gravitation. © 1998 Elsevier Science Ltd. All rights reserved.

Investigations of homogeneous nucleation, carried out in a thermal diffusion chamber [1, 2], required a more detailed study of the motion of the aerosol particles acted upon by temperature gradients and affected by the composition of the gaseous mixture (thermophoresis and diffusiophoresis), taking into account phase transitions at the interface. By the action of the forces of thermodiffusiophoresis a particle inside the chamber initially moves vertically upwards, and then, on reaching a critical size, begins to fall. The value of the coordinate at which inversion of the particle velocity occurs is determined experimentally. The purpose of the present paper is to find the trajectory of the particle motion which is accompanied by condensation growth of the particle.

1. FORMULATION OF THE PROBLEM

For a specified geometry of the thermal diffusion chamber, the temperature of the hot and cold walls and the type of working liquid, the temperature distribution, the distribution of the concentration of the working-liquid vapour and the vapour supersaturation are calculated. The region $z = z_0$, where supersaturation is a maximum, is the region where nucleation of a drop is most probable. Relations are also known which enable one to calculate the mean radius of the nucleation centre R_0—the initial size of the growing drop. If the latter is fairly small, the perturbation of the temperature and concentration fields will also be small, and they can be neglected. Otherwise we can calculate the perturbation of the temperature field and the vapour concentration field of the working material of a single drop, i.e. we can obtain the values of the temperature and concentration of the gaseous mixture in the region of the drop, assuming the number density of the drops themselves to be fairly small [3, 4]. It must be borne in mind here that one must take into account the gas–liquid phase transition at the interface in the boundary conditions.

In both of these cases it is easy to obtain the force with which the non-uniform gas acts on a particle situated in it [3, 4], and also investigate the kinetics of particle growth in this case. In this paper we consider the case of fairly small Knudsen numbers (large drop dimensions).

The first part of the programme (obtaining the temperature and concentration fields inside the thermal diffusion chamber) was published previously in [1, 2]. We will give a description of the method of calculating the fields perturbed by the presence of a growing (or evaporating) drop, and of the forces acting on it from the gas side. By then solving the equation of motion of a drop of variable mass in a field made up of gravity and thermodiffusiophoresis forces, we can determine its trajectory of motion. The final aim is to obtain an equation for calculating the coordinate z at which the particle velocity vanishes. The value of the vertical coordinate which the particle reaches before it begins to fall under gravity, is determined in a direct experiment [1, 2].

2. THE TEMPERATURE AND CONCENTRATION FIELDS IN THE NEIGHBOURHOOD OF THE DROP

The distribution of the temperature T_i inside the drop, of the radius R, the temperature T and the concentration C outside it can be written in the form [3]

$$T_i = T_0 + (\mathbf{A}\cdot\mathbf{r}),\quad T = T_\infty + \delta T\frac{R}{r} + (\operatorname{grad} T\cdot\mathbf{r}) + ([\mathbf{A} - \operatorname{grad} T]\cdot\mathbf{r})\left(\frac{R}{r}\right)^3 \tag{2.1}$$

$$C = C_\infty + \delta C\frac{R}{r} + (\operatorname{grad} C\cdot\mathbf{r}) + \left(\left[\frac{\partial C_s}{\partial T}\mathbf{A} - \operatorname{grad} C\right]\cdot\mathbf{r}\right)\left(\frac{R}{r}\right)^3$$

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The constants of integration **A**, δT, δC can be found from the boundary conditions on the drop surface

$$T=T_i,\quad C=C_s(T),\quad \varkappa\frac{\partial T}{\partial r}+N\left(\frac{\partial C}{\partial r}+\frac{k_T}{T}\frac{\partial T}{\partial r}\right)=\varkappa_i\frac{\partial T_i}{\partial r},\quad N=\frac{LD\rho_s}{C_s(1-C_s)} \tag{2.2}$$

Here gradt T and grad C are the temperature and concentration gradients of the vapour of the working material far from the drop, T_∞, C_∞ are the values of the temperature and concentration far from the drop in a horizontal plane passing through the drop centre, T_0 is the temperature at the drop centre, $\varkappa$ is the thermal conductivity of the gas, $\varkappa_i$ is the thermal conductivity of the condensed phase, L is the specific latent heat of the phase transition, ρ_s is the density, C_s is the concentration of saturated vapour of the working substance, D is the mutual diffusion coefficient of the components of the mixture and k_T is the thermal-diffusion ratio.

In order not to complicate the description with unimportant details, we have neglected the Dufour effect here.

When writing the solution of (2.1) it is assumed that the non-uniformity of the drop temperature $\delta T_i = T_i(R) - T_0$ is fairly small. The concentration of saturated vapour (the subscript s) in the region of the interface can be represented in the form $C_s(T) = C_s(T_0) + (\partial C_s/\partial T)\delta T$. Consequently $\partial C = C_s(T_0) - C_\infty$. The effect of the curvature of the drop surface on the saturated vapour density close to it is ignored.

The third equation of (2.2) is equivalent to the following two relations

$$-\left(\varkappa+N\frac{k_T}{T}\right)(3\,\mathrm{grad}\,T-2\mathbf{A})+\varkappa_i\mathbf{A}-N\left(3\,\mathrm{grad}\,C-2\mathbf{A}\frac{\partial C}{\partial T}\right)=0 \tag{2.3}$$

$$\left(\varkappa+N\frac{k_T}{T}\right)\delta T+N\delta C=0 \tag{2.4}$$

We have from (2.3)

$$\mathbf{A}=\frac{3}{2}\left(1+\frac{1}{2}\frac{\varkappa_i}{\varkappa}Q\right)^{-1}\mathrm{grad}\,T,\quad Q=\left[1+\frac{N}{\varkappa T}\left(k_T+T\frac{\partial C_s}{\partial T}\right)\right]^{-1}$$

while for $\delta T = T_0 - T_\infty$ we obtain

$$\partial T=-N\left(\varkappa+N\frac{k_T}{T}\right)^{-1}[C_s(T_0)-C_\infty]$$

3. THE THERMODIFFUSIOPHORESIS FORCE

We will now calculate the force acting on the drop from the side of the non-uniform gaseous mixture—the thermodiffusiophoresis force. We have for the components of the gas velocity on the drop surface in the Stokes approximation (η is the dynamic viscosity and p is the gas pressure)

$$v_r=\frac{h}{R^2}+v_c[1-2a+2b]\cos\theta,\quad v_\theta=-v_c[1-a-b]\sin\theta$$

where a and b are constants. The force with which the gas acts on the drop has the form $F = 8\pi\eta R a v_c$, where v_c is the velocity of the centre of mass of the gas with respect to the drop.

We will write the boundary conditions for the radial and tangential components of the gas velocity on the drop surface in the form

$$v_r=v^{(0)}+v_D\cos\theta=-\frac{N}{\rho L}\left(\frac{\partial C}{\partial r}+k_T\frac{\partial\ln T}{\partial r}\right) \tag{3.1}$$

$$v_\theta=v_{\mathrm{sl}}\sin\theta=k_{\mathrm{ts}}\frac{\eta}{\rho T}\frac{1}{R}\frac{\partial T}{\partial\theta}+k_{\mathrm{ds}}D\frac{1}{R}\frac{\partial C}{\partial\theta} \tag{3.2}$$

Here v_D is the diffusion velocity, v_{sl} is the slip velocity, k_{ts} is the thermal slip coefficient and k_{ds} is the diffusion slip coefficient.

Determining the constants of integration we obtain

$$F=6\pi\eta R\left(v_c-\frac{1}{3}v_D+\frac{2}{3}v_{\mathrm{sl}}\right) \tag{3.3}$$

4. THE CHANGE IN THE DROP SIZE

The spherically symmetrical component of the expression for the radial velocity of the gas $v^{(0)}$ defines the mass flux on the particle

$$\frac{\partial m}{\partial t}=-\iint_{\Sigma}\rho v^{(0)}d\Sigma=-\frac{N}{L}\left(\delta C+\frac{k_T}{T}\delta T\right)4\pi R$$

Bearing Eq. (2.4) in mind, we arrive at the well-known relation $\partial m/\partial t = 4\pi RI(t)/L$, $I(t) = \varkappa\delta T$.

On the other hand, taking into account the expression for the mass of a spherical drop of density r_i, we have $\partial m/\partial t = 4\pi R^2\rho_i dR/dt$, i.e. $dR^2/dt = 2I(t)/(\rho_i L)$. After integration and changing to the variable z ($z = v(z)t$, where $v(z)$ is the drop velocity in a system connected with the chamber walls), we obtain

$$R^2(z)=R_0^2+\frac{2}{\rho_i L}\int_{z_0}^{z}I(z')\frac{dz'}{v(z')}$$

5. THE EQUATION OF MOTION OF THE DROP

We will now consider the motion of a drop in a thermal diffusion chamber taking the variable mass into account. Using expression (3.3) we can write the equation of motion in the form

$$v\frac{1}{m}\frac{dm}{dt}+\frac{dv}{dt}=\frac{2\pi\eta R}{m}(3v_c-v_D+2v_{sl})-g$$

But

$$\frac{1}{m}\frac{dm}{dt}=\frac{1}{\frac{4}{3}\pi R^3\rho_i}4\pi R\frac{\varkappa\delta T}{L}=\frac{3I}{\rho_i R^2 L},\quad v_c=-(v-v_{St}),\quad v_{St}=-\frac{N}{\rho L}\left(\operatorname{grad}C+\frac{k_T}{T}\operatorname{grad}T\right)$$

(v_{st} is the velocity of Stefan flow).

Hence, we have the following for the equation of motion of the drop

$$\frac{dv}{dt}+\frac{3v(t)}{\rho_i R^2(t)}\left(\frac{I}{L}+\frac{3}{2}\eta\right)-\frac{3\eta}{2\rho_i R^2(t)}(2v_{sl}+3v_{St}-v_D)+g=0$$

Changing to the variable z, we obtain the equation

$$v(z)\left(\frac{I(z)}{\eta(z)L}+\frac{3}{2}\right)+\frac{\rho_i R^2(z)}{3\eta(z)}\left(v(z)\frac{dv}{dz}+g\right)=v_{sl}(z)+\frac{3}{2}v_{St}-\frac{1}{2}v_D(z)$$

the solution of which enables us to obtain the coordinate of the inversion of the drop velocity, compare it with the measured value and hence judge the adequacy of the model employed.

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