



# Condensation rate of light vapour molecules in the transition regime

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## ABSTRACT

In order to overcome uncertainties of semi-empirical models based on the flux matching theory and to resolve discrepancies between more accurate interpolation formulas, obtained from various solutions of the Boltzmann equation, a new approach, similar to that recently proposed by the authors to consideration of the heavy vapour molecules condensation, is applied to light vapours. In this limit the transport of light vapour molecules to the central particle can be strictly considered as specific random walks, characterized by the exponential distribution of the elementary displacement distances for each walker migrating with a fixed speed and changing direction randomly, whereas the condensation rate is related to the mean volume swept per unit time by the migrating effective particle of radius equal to the radius of the central particle. Results of numerical calculations of the condensation rate at different values of the Knudsen number are approximated by the interpolation expression with a relatively high accuracy in comparison with the traditional expressions.

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## 1. Introduction

The steady state flow of vapour molecules to a spherical particle in a sample of volume  $V \rightarrow \infty$ , when the particle radius  $R$  is sufficiently large compared to the mean free path  $\lambda_v$  of the diffusing (with the diffusivity  $D$ ) vapour molecules (or  $Kn = \lambda_v/R \ll 1$ , where  $Kn$  is the Knudsen number), is given by Maxwell's solution of the continuum transport equation:

$$F_c = 4\pi DR(n_v - n_s), \quad (1)$$

where  $n_v = N_v/V$  is the mean concentration of vapour molecules and  $n_s$  is their saturation concentration (i.e. at vapour–solid equilibrium). This expression can be applied to calculation of the spherical particle growth kinetics

$$\frac{1}{\Omega_p} \frac{dV_p}{dt} = F_c, \quad (2a)$$

where  $V_p$  is the particle volume,  $\Omega_p$  is the volume of a condensed vapour molecule in the particle, or

$$\frac{dR}{dt} = \frac{D\Omega_p}{R}(n_v - n_s), \quad (2b)$$

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under the steady state growth condition,  $R^{-1} dR/dt \ll \tau_{ss}^{-1}$ , where  $\tau_{ss} \approx R^2/\pi D$  is the characteristic time for attainment of the steady state solution of the vapour diffusion problem with the fixed value of  $R$  (i.e. with the immobile boundary), which can be deduced from analysis of the general, non-stationary solution of the diffusion problem,  $F_c = 4\pi D_v R(n_v - n_s)(1 + R/\sqrt{\pi D t})$  (cf., e.g. Fuchs, 1934). This provides an insignificant restriction on the applicability of the steady state approximation,  $(n_v - n_s)\Omega_p \ll \pi$ , and thus Eqs. (2a) and (2b) can be used with a good accuracy.

At the other extreme,  $Kn \gg 1$ , the expression for the vapour flow based on the kinetic theory of gases can be used

$$F_{fm} = \pi R^2 \bar{u}_v (n_v - n_s) \alpha = \frac{R \bar{u}_v \alpha}{4D} F_c, \quad (3)$$

where  $\bar{u}_v = (8kT/\pi m_v)^{1/2}$  is the mean thermal speed of vapour molecules,  $\alpha$  is the molecular accommodation coefficient. Both expressions are no longer valid in the transition regime, when the mean free path of the diffusive vapour molecules becomes comparable with the particle radius,  $Kn \approx 1$ .

Early investigations of Knudsen aerosol condensation used the flux matching theory of Fuchs (1934), that is, by considering the non-continuum effects to be limited to a region  $R \leq r \leq \Delta$  beyond the droplet surface and assuming that for  $r \geq \Delta$  continuum theory applies. The absorbing sphere radius  $\Delta$ , then, is of the order of the mean free path  $\lambda_v$  and within this inner region the simple kinetic theory of gases is assumed to apply. Fuchs, by matching the fluxes for the two domains at  $r = \Delta$ , obtained the flux ratio as follows:

$$\frac{F}{F_c} = \frac{1 + \Delta/R}{1 + \Delta/R + \alpha R \bar{u}_v / 4D_v}. \quad (4)$$

The value  $\Delta$  was not specified in the original model and must be adjusted empirically or estimated by independent theory. Several choices for  $\Delta$  have been proposed; the simplest, due to Fuchs, is  $\Delta = 0$ . Dahneke (1983) suggested  $\Delta = \lambda_v$ , and using  $\lambda_v = 2D_v/\bar{u}_v$  in definition of the Knudsen number (designated here as  $Kn_{Da}$ ), obtained

$$\frac{F}{F_c} = \frac{1 + Kn_{Da}}{1 + 2Kn_{Da}(1 + Kn_{Da})\alpha^{-1}}, \quad (5)$$

but numerous other possibilities exist, as reviewed by Davis (1983).

Besides, the basic equations used in the flux matching theory cannot be strictly justified and thus the obtained expressions for the condensation flux can be used only for qualitative consideration. Indeed, the diffusion equation for the vapour molecules concentration, which is applied in the flux matching theory outside the absorbing sphere, is valid under the general condition that the length scale  $L$  of the concentration variation is large in comparison with the mean free path,  $L \gg \lambda_v$ . Taking into account that the diffusion concentration profile substantially varies outside the absorbing sphere (of radius  $\Delta \approx R + \lambda_v$ ) on the scale of  $L \approx \Delta$ , the condition of the diffusion equation validity in the vicinity of the absorbing sphere takes the form,  $\Delta \gg \lambda_v$ , which results in  $R \gg \lambda_v$ . This condition significantly restricts applicability of the flux matching theory to small Knudsen numbers,  $Kn = \lambda_v/R \ll 1$ , i.e. only small corrections to the flux (Eq. (1)), in the near-continuum regime can be properly evaluated in the flux matching approach.

The concentration distributions of the diffusing species and background gas in the transition regime are governed rigorously by the Boltzmann equation. However, there does not exist a general solution to the Boltzmann equation valid over the full range of Knudsen numbers for arbitrary masses of the diffusing species,  $m_v$ , and the background gas,  $m_g$ ; nonetheless, the problem can be studied more rigorously in the two limiting cases of heavy and light vapour molecules, when the partial vapour pressure is much less than the gas pressure and thus the vapour–vapour collisions can be neglected.

The heavy vapour molecules with large  $z = m_v/m_g \gg 1$  can be considered as Brownian particles and thus the condensation rate can be calculated using the collision kernel of the Smoluchowski coagulation equation, as recently suggested by Gopalakrishnan and Hogan (2011) and Veshchunov and Azarov (2012).

The problem can be simplified also in the opposite case of small mass ratio,  $z = m_v/m_g \ll 1$ . Different approaches to consideration of this case was thoroughly analysed in the review paper of Davis (1983). The problem assumes considerable similarity to the problems encountered in the neutron transport studies, under assumptions that the concentration and velocity distribution of the gas molecules are only slightly perturbed by the evaporation process and that once a vapour molecule encounters the surface of a particle its probability of sticking is unity,  $\alpha = 1$ . Namely, it was noticed that the problem of a flux, to a black sphere, of neutrons, diffused isotropically by heavy atoms, is completely equivalent to the examined problem of droplet growth for this case,  $z = m_v/m_g \rightarrow 0$ . The former problem has received considerable attention from the workers in neutron transport theory and some more accurate solutions have been obtained. In this approach, Fuchs and Sutugin (1971) fitted Sahni's (1966) theoretical solution to the Boltzmann equation by means of the expression

$$\frac{F}{F_c} = \frac{1 + Kn_{FS}}{1 + 1.7104Kn_{FS} + (4/3)Kn_{FS}^2}, \quad (6)$$

which correctly represents asymptotic solutions for large and small Knudsen numbers; however, becomes approximate in the transition regime, where series expansion of the Bessel function (with a finite argument) in the integral equation analysed by Sahni (1966) cannot be truncated after the first terms. Besides, some additional deviation of values calculated according to this interpolation formula (Eq. (6)), from numerical results of Sahni attained 2–6%. The mean free path included in the definition of the

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