



# The Stefan outflow in a multicomponent vapor–gas atmosphere around a droplet and its role for cloud expansion



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## ABSTRACT

A new comprehensive analysis of Stefan's flow caused by a free growing droplet in the vapor–gas atmosphere with several condensing components is presented. This analysis, based on the nonstationary heat and material balance and diffusion transport equations, shows the appearance of the Stefan inflow in the vicinity of the growing droplet and the Stefan outflow at large distances from the droplet as a consequence of nonisothermal condensation. For an ensemble of droplets in the atmospheric cloud, this outflow provides an increase of the total volume of the cloud, which can be treated as cloud thermal expansion and leads to the rise of the cloud as a whole due to increasing its buoyancy. We have formulated the self-similar solutions of the nonstationary diffusion and heat conduction equations for a growing multicomponent droplet and have derived analytical expressions for the nonstationary velocity profile of Stefan's flow and the expansion volume of the vapor–gas mixture around the growing droplet. To illustrate the approach, we computed these quantities in the case of droplet of stationary composition in air with several specific vapors ( $\text{C}_2\text{H}_5\text{OH}/\text{H}_2\text{O}$ ;  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ ;  $\text{H}_2\text{O}$ ).

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## 1. Introduction

Growth and evaporation of small droplets are key phenomena in the physics of aerosols and clouds. The regularities of phase changes and latent heat release have a global influence on the Earth atmosphere and climate (Heintzenberg & Charlson, 2009; Kessler, 1969; Pruppacher & Klett, 2010). Cloud microphysical schemes are a central part of every model of the atmosphere. In numerical weather simulation, they are important for quantitative precipitation forecasts.

Our goal in this paper is to give a detailed microphysical analysis of one of the mechanisms of thermal cloud expansion in the Earth atmosphere. This mechanism is associated with the Stefan flow caused by nonisothermal growth of small droplets in the cloud. The Stefan flow of the vapor–gas mixture around a droplet compensates the diffusive flux of noncondensable gas molecules from the impenetrable surface of the growing droplet. This flow under ordinary atmospheric conditions is usually considered to be a small effect (Clement, 2008; Fuchs, 1959; Kuchma & Shchekin, 2011, 2012). However there are some atmospheric situations where the Stefan flow may be important. The particle scavenging efficiency of vapor-grown ice crystals falling from mixed clouds proves to be very high due to the Stefan flow which pushes airborne particles away from the surface of the supercooled droplets evaporating in the vicinity of an ice crystal (Vittori, 1984). The influence of the Stefan

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flow on the heat balance of a char particle should be taken into account in oxy-fuel combustion (temperature  $\approx 1400$  K), because the presence of Stefan's flow greatly weakens the heat transfer by conduction and accelerates the heat loss from the particle, which makes the particle temperature lower (Zhou, Yu, & Ou, 2013). Another effect, as follows from Kuchma, Shchekin, Martyukova, and Lezova (2016), owes to nonstationary heating a vapor–gas mixture around a growing droplet (due to releasing the condensation heat) which produces the Stefan outflow at large distances from the droplet. For an ensemble of droplets in the cloud, such effect results in increase of the total volume of the cloud that can be treated in physics of clouds as cloud thermal expansion followed by the rise of the cloud as a whole due to buoyancy. In the case of several vapors, also the vertical structure of a cloud with droplets of different size and composition can change. Thus we have an example where a small effect on a level of a single droplet becomes an origin for strong mass and heat transport on the macroscopic scale of a cloud.

We will formulate below the theory of the Stefan flow in the case of multi-component nonisothermal and nonstationary growth of a free droplet with non-ideal composition. As a first step, we will derive in Section 2 the general expressions for the velocity of the Stefan flow based on the material and enthalpy balance equations at nonstationary diffusion. In Section 3, we will consider the self-similar solutions of the nonstationary diffusion and heat conduction equations for a growing multicomponent droplet and obtain analytical expressions for the whole nonstationary velocity profile of Stefan's flow and the expansion volume of the vapor–gas mixture around the growing droplet. Finally in Section 4, we will illustrate the approach by computing velocity profile of Stefan's flow and the expansion volume of the vapor–gas mixture around the growing droplet for droplets of stationary composition in air with several specific vapors (binary mixture of ethanol–water  $C_2H_5OH/H_2O$  vapors; binary mixture of sulfuric acid–water  $H_2SO_4/H_2O$  vapors, and pure water vapor  $H_2O$ ).

## 2. General regularities for a multicomponent Stefan flow

### 2.1. Definitions and boundary conditions

Consider a small free spherical droplet suspended in the ideal mixture of several condensable vapors and a non-condensable carrier gas. The condensable components are assumed to be miscible in the droplet at any ratios, and the solution in the droplet may be non-ideal. The droplet is tiny enough to provide that the specific times of establishing internal thermal and chemical equilibrium in the droplet are small in comparison with the specific time of changing the droplet radius in time. As a consequence, the droplet is internally uniform in composition and temperature. The droplet can grow or evaporate depending on the ratio between the actual concentrations of vapors far away from the droplet and the concentrations of the vapors at saturation with the solution in the droplet. We will take into account releasing or absorbing the latent heat at condensation or evaporation. As a result, the temperatures of the droplet and surrounding vapor–gas mixture may differ. The regimes of vapor and heat transfer to or from the droplet are diffusive. The total pressure in the vapor–gas mixture is considered to be constant. Gravity is neglected.

We set the origin of the coordinate system in the center of the spherical droplet with radius  $R(t)$  changing in time  $t$ . In view of the spherical symmetry, the local vapor concentration  $n_i(r, t)$  of the condensable component  $i$  ( $i = 1, 2, \dots, k$ ), i.e., the local number of molecules of  $i$ th component per unit volume, and the local temperature  $T(r, t)$  in the vapor–gas mixture at any time moment  $t$  depend only on distance  $r$  from the center of the droplet to the observation point. The corresponding concentration of the non-condensable carrier gas will be denoted by  $n_g(r, t)$ . Thereby the total local volume concentration of molecules in the vapor–gas mixture is determined as

$$n(r, t) = n_g(r, t) + \sum_{i=1}^k n_i(r, t), \quad (2.1)$$

The boundary conditions for the vapor concentrations  $n_i(r, t)$  and temperature  $T(r, t)$  can be written in the form

$$n_i(r, t) \xrightarrow{r \rightarrow \infty} n_{i0}, \quad n_i(r, t) \Big|_{r=R(t)} = n_{i\infty}(\{x\}, T_d), \quad i = 1, 2, \dots, k, \quad (2.2)$$

$$n(r, t) \xrightarrow{r \rightarrow \infty} n_0 = n_{g0} + \sum_{i=1}^k n_{i0}, \quad (2.3)$$

$$T(r, t) \xrightarrow{r \rightarrow \infty} T_0, \quad T(r, t) \Big|_{r=R(t)} = T_d, \quad (2.4)$$

where  $n_{i0}$  and  $n_{g0}$  are the bulk concentration of  $i$ th component and carrier gas, respectively,  $T_0$  is the temperature far away from droplet,  $n_{i\infty}(\{x\}, T_d)$  is the equilibrium concentration of the  $i$ th component over a planar surface of the liquid solution at droplet temperature  $T_d$  (we neglect the effect of the curvature of droplet surface on the  $i$ th vapor equilibrium pressure and concentration) and composition  $\{x\} \equiv \{x_1, x_2, \dots, x_k\}$ . The molar concentration  $x_i$  of condensible components in the droplet (the total number of droplet components coincides with the number of the vapor components in the vapor–gas mixture) is defined by

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