Evaporation and Growth of Multicomponent Droplets in Random Dense Clusters

T. Elperin
elperin@menix.bgu.ac.il

B. Krasovitov
The Pearlstone Center for Aeronautical Engineering Studies,
Department of Mechanical Engineering,
Ben-Gurion University of the Negev,
P.O. Box 653,
Beer-Sheva 84105,
Israel

Interaction between evaporating (growing) droplets in binary arrays and large random clusters of droplets of different sizes is analyzed in a quasi-steady approximation using the modified method of expansion into irreducible multipoles. Evaporation and condensation of binary arrays and clusters of droplets (i) composed of volatile components and (ii) composed of a volatile and a nonvolatile component was studied. The analytical and numerical results of the investigation are presented in terms of heat and mass correction factors. Solution of the transient problem is obtained, and the evaporation rate is determined. It is shown that for droplets of ideal solution with different compositions, Nusselt and Sherwood numbers depend on the concentration of components inside each droplet. When dense cloud contains both pure droplets and droplets containing soluble nuclei, interactions between temperature and concentration fields causes growth of some droplets in a cluster and evaporation of the other droplets. It is shown that inside an evaporating large cluster of multicomponent droplets composed of volatile components, recondensation occurs whereby evaporating droplets act as mass sources and heat sinks and growing droplets act as mass sink and heat sources. The results of this study are of relevance in the analysis of dynamics of droplet size distribution in clouds, artificial modification of clouds and precipitation, in-cloud pollutants scavenging, and in the analysis of evaporation and combustion of multicomponent (blended) liquid fuels.

1 Introduction

Mass and energy exchange between a dispersed phase and an ambient gas is encountered in numerous atmospheric and industrial processes. The applications include condensation of hydrocarbons and water vapor in stack gases, spray drying, and spray combustion, etc. Condensational droplet growth on nuclei, consisting of hygroscopic substances, is very important in such fields as separation of submicron particles from gases and scavenging of the pollutants (Heidenreich and Ebert, 1995). Single drop evaporation (combustion) models yield the parameters and evaporation (combustion) characteristics of the individual droplets. However, clusters of droplets with high concentration of droplets are encountered in various engineering and environmental applications (see e.g., Bellan and Harstad, 1995). In this case, the droplets behave quite differently from the isolated droplets. The importance of droplet interaction effects has not gone unrecognized in the literature. It is known that interactive problems can be treated by five methods: (i) bispherical coordinates—suitable for two-drop arrays (Carstens et al., 1970; Umemura et al., 1981); (ii) method of images—suitable for arrays less than 20 drops (Labowsky, 1978, 1980a, b); (iii) point sources method—suitable for dilute arrays of 1000 drops (Deutch et al., 1976; Ray and Davis, 1980; Annamalai et al., 1993); (iv) continuum methods—suitable for dilute sprays (Chiu and Liu, 1977; Annamalai et al., 1988; Tsai and Sterling, 1991); (v) method of expansion into irreducible multipoles—suitable for large dense random clusters of droplets (particles) (Elperin and Krasovitov 1994a, 1994b).

State of the art in the above methods was reviewed by Annamalai and Ryan, 1992; Annamalai et al., 1993, 1994; Elperin and Krasovitov 1994a, 1994b. The theoretical methods (1)–(4) deal primarily with droplet evaporation (combustion) in various limiting cases, such as: regular structure of cluster, finite array, dilute clusters, monosized clusters, etc. The method of expansion into irreducible multipoles which was developed in our previous works (Elperin and Krasovitov, 1994a, 1994b) is applicable to more realistic problems and is particularly suitable for dense random clusters of droplets (particles). The droplets that are used in various engineering applications are mainly composed of two or more components. However, in spite of the importance of this field there are only a few works (Labowsky, 1980a; Annamalai et al., 1993) that address evaporation of clusters of multicomponent droplets.

The effects of interactions on the evaporation (combustion) rate are estimated using the correction factor. For an array containing N multicomponent droplets, the correction factor of ith species for the jth droplet is determined as (Annamalai et al., 1993)

$$\eta_{ij} = \frac{(\dot{m}_i)_j}{(\dot{m}_{iso,i})_j} \tag{1}$$

where the isolated droplet has the same composition as the jth droplet in the array.

It is well known that interactive effects could both retard and enhance the rate of evaporation of the interacting droplets. It was shown previously (see e.g., Labowsky, 1980a; Annamalai et al., 1993) that the latter occurs under some restrictive conditions such as buoyant convective effects and when droplets of dissimilar composition are considered. In a case where interactive effects retard the rate of evaporation, the correction factor is less then 1 ($\eta < 1$, "negative" interactions) and in case when the interactive effects enhance the rate of evaporation the correction factor is greater then 1 ($\eta > 1$, "positive" interactions).

However, the above mentioned theoretical results were obtained for constant droplet diameters, compositions, and center-to-center distances. In reality, the compositions and sizes of droplets change with time. The latter implies that the correction factor η depends on time and, therefore, could give rise to the

Contributed by the Heat Transfer Division for publication in the JOURNAL OF HEAT TRANSFER. Manuscript received by the Heat Transfer Division February 26, 1996; revision received January 17, 1997; Keywords: Condensation, Evaporation, Sprays/Droplets. Associate Technical Editor: S. Ramadhyani.

mass and heat sources and sinks inside the cluster. Xiong et al. (1984) conducted evaporation experiments on a two-drop array of pure single component droplets (high volatile heptane drop and low volatile hexadecane drop at an ambient temperature 24°C) and showed that the hexadecane drop acts as a mass sink, and a heat source for the heptane drop. Thus, the diameter of the hexadecane droplet increases as a result of condensation of vapors from the heptane droplet. In this study it is shown that heat and mass sinks and sources within the cluster of multicomponent droplets containing volatile and a nonvolatile components can also occur in a case of condensation on hygroscopic nuclei inside the cluster.

The first theoretical investigation of the transient problem where the sizes of particles inside a random cluster of char/carbon particles change with time was performed by Elperin and Krasovitov (1994b). In the latter work, the results obtained using expansion into irreducible multipoles were validated by comparison with the available experimental data. The purpose of the present study is to develop the theory of evaporation and condensation of dense random clusters of droplets with dissimilar compositions. In the next section some important evaporation characteristics of random clusters of droplets are discussed.

2 Model and Assumptions

2.1 Evaporation of a Single Multicomponent Droplet.

(A) Evaporation of Droplets With a Volatile and Nonvolatile Components. Consider a spherical droplet with high thermal conductivity containing volatile and nonvolatile components immersed in a stagnant binary gas mixture at temperature T_∞, with the concentration of volatile species $C_{1,\infty}$. The influence of convection on the evaporation is neglected. Assume also that the characteristic times of heat and diffusive relaxation are small compared to total time of evaporation so that the steady-state evaporation model is valid.

Under the above assumptions, the system of mass and energy conservation equations is shown in Table 1. Table 2 provides the corresponding boundary conditions that include the boundary conditions (7) – (8) allowing for the occurrence of the temperature and concentration jumps at the surface of a droplet and for the occurrence of the cross effects (see Yalamov et al., 1990). These boundary conditions are supplemented with the condition of continuity of energy flux at the droplet surface (9) and conditions at infinity (10).

Solution of the above system of equations for large-size droplets (without temperature and concentration jumps), in case of a diffusion regime of evaporation, yields the following expressions for the heat and mass fluxes at the droplet surface (Elperin and Krasovitov, 1995):

$$q_T = 4\pi R \int_{T_\infty}^{T_s} k_e dT,$$

$$q_1 = 4\pi R \frac{(C_{1,s}(T_s) - C_{1,\infty})}{\int_{T_\infty}^{T_s} \frac{k_e}{nD} dT} \int_{T_\infty}^{T_s} k_e dT \qquad (11)$$

where q_T and q_1 are integral heat and mass fluxes at the droplet surface, correspondingly, T_∞ is the temperature of the ambient gas and temperature at the droplet surface T_s can be found from the following transcendental algebraic equation:

$$\int_{T_\infty}^{T_s} k_e dT_e \left(1 + m_1 L \frac{(C_{1,s}(T_s) - C_{1,\infty})}{\int_{T_\infty}^{T_s} (k_e/nD) dT_e}\right) + R\gamma\gamma_0(T_s^4 - T_\infty^4) = \frac{q_w}{4\pi R} \qquad (12)$$

where γ and γ_0 are emissivity and Stefan-Boltzmann's constant, correspondingly, q_w is the total power of internal heat sources and $C_{1,s}(T_s)$ is the relative concentration at the droplet surface that is related to the drop temperature through phase equilibrium relations. In case of ideal solutions, the value of the relative concentration $C_{1,s}(T_s)$ is determined by the Raoult's law:

$$C_{1,s}(T_s) = X_{1,w} C_{1,s}^{(0)}(T_s) \qquad (13)$$

and the Clapeyron-Clausius equation:

$$C_{1,s}^{(0)}(T_s) = C_{1,s}(T_0) \exp\left(\frac{L\mu_\nu}{R_g}\left(\frac{1}{T_0} - \frac{1}{T_s}\right)\right) \qquad (14)$$

where $C_{1,s}^{(0)}(T_s)$ is the relative concentration of saturated vapor at the surface of a pure liquid, μ_ν is the molar mass of volatile species and R_g is the universal gas constant. For non-ideal solu-

Nomenclature

$C_k = n_k/n$, dimensionless concentration of k-th specie
D = coefficient of binary diffusion, $m^2 \cdot s^{-1}$
$\mathbf{j}_i$ = mass flux density, $m^{-2} \cdot s^{-1}$
$\mathbf{j}_T$ = heat flux density, $W \cdot m^{-2}$
h = enthalpy, $J \cdot kg^{-1}$
k = coefficient of thermal conductivity, $W \cdot (m \cdot K)^{-1}$
$K_T = D_T(T, C_1)/D$ thermal diffusion ratio
$K_T^{(T)}$, $K_n^{(n)}$ = coefficients of temperature and concentration jumps, m
$K_T^{(n)}$, $K_n^{(T)}$ = cross coefficients, m
L_{ij} = center-to-center distance between droplets, m
L = latent heat of evaporation, $J \cdot kg^{-1}$
m_k = mass of the molecules of k-th species, kg
M' = mass of soluble nucleus, kg
M_d = mass of a droplet, kg
M_j = mass of j-th volatile species, kg
n = molecular number density, m^{-3}
Nu = Nusselt number
q_k = integral mass flux of the molecules of k-th specie, s^{-1}
q_T = integral heat flux, W
q_w = total power of internal sources, W
R_i = radius of i-th droplet, m
R_g = universal gas constant, $J \cdot (mol \cdot K)^{-1}$
r = radius coordinate, m
Sh = Sherwood number
T = temperature, K
X_k = molar fraction of k-th species

Greek

γ = Stefan-Boltzmann constant, $W \cdot m^{-2} \cdot K^{-4}$
γ_0 = integral emissivity
ζ = heat correction factor
η = mass correction factor
μ' = molar mass of soluble hygroscopic nucleus, $kg \cdot mol^{-1}$
μ_v = molar mass of a solvent vapor, $kg \cdot mol^{-1}$
ρ = density, $kg \cdot m^{-3}$
σ = volume fraction
$\tilde{\sigma}$ = surface tension, $N \cdot m^{-1}$

Subscript

i, j = number of a droplet, number of species (Tables 1–3)
β = number of species
s = value at a droplet surface
1, 2 = volatile specie and ambient gas
∞ = value at infinity

Superscript

(e) = value outside a droplet
i = number of a droplet
(s) = value at the surface
(w) = value within a droplet

Table 1 Governing equations

$$\operatorname{div} \Gamma_\phi = 0 \tag{2}$$

Equation	Γ_ϕ: General equations	Γ_ϕ: Linearized equations
Mass conservation	$\mathbf{j}_i$	∇C_i
Energy conservation	$\mathbf{j}_T$	∇T_e

Droplets of a volatile and nonvolatile component

$$\mathbf{j}_i = n_i \mathbf{v} - \frac{m_j n^2}{\rho^{(e)}} D(\nabla C_i + (-1)^{i+1} K_T \nabla \ln T_e) \tag{3}$$

$$\mathbf{j}_T = \sum_i h_i m_i \mathbf{j}_i - k_e \nabla T_e \tag{4}$$

$$(i, j = 1, 2,\ i \neq j)$$

$$C_\nu = n_\nu / \sum_i n_i, \quad C_2 + C_1 = 1$$

Multicomponent droplets of volatile components

$$\mathbf{j}_i = \frac{\sum_k C_k}{\sum_k \frac{C_k}{D_{ik}}} n \nabla C_i + \frac{\sum_k \frac{\mathbf{j}_k}{D_{ik}}}{\sum_k \frac{C_k}{D_{ik}}} C_i \tag{5}$$

$$\mathbf{j}_T = \sum_i h_i m_i \mathbf{j}_i - k_e \nabla T_e \tag{6}$$

$$i, k = \overline{1, \Lambda + 1}, \quad i \neq k, \quad \sum_{i=1}^{\Lambda+1} C_i = 1$$

tions the value of the relative concentration $C_{1,s}(T_s)$ can be found from the following formula (see Mason, 1971, pp. 24–30):

$$C_{1,s}(T_s) = C_{1,s}^{(0)}(T_s)\left(1 + \frac{2\mu_\nu \tilde{\sigma}}{\rho R_g T_s} \frac{1}{R} - \frac{3i\mu_\nu}{4p\rho\mu'} \frac{M'}{R^3}\right) \tag{15}$$

where $\tilde{\sigma}$ is the surface tension, ρ is the density of solvent, μ_ν and μ' are the molar masses of vapor of the solvent and of soluble hygroscopic nucleus, i is van't Hoff's factor which depends on the chemical nature and the degree of dissociation (i.e., on the concentration) of the solute and M' is the mass of a soluble hygroscopic nucleus.

Integrals in expressions (11) and transcendental Eq. (12) can be calculated using dependencies of thermal conductivity and diffusivity on temperature and concentrations of species (see e.g., Lefebvre, 1989, Chapter VIII).

In the case of evaporation of a droplet immersed into a stagnant binary gas mixture with small temperature differences, in the neighborhood of the droplet, the system of Eqs. (2) can be linearized (see Table 1). In the latter system of equations it is taken into account that in a binary gas mixture, the sum of the relative concentrations is equal to unity (i.e., $C_1 + C_2 = 1$).

Finally, the rate of droplet evaporation can be found from the following differential equation:

$$\frac{dM_d}{dt} = -m_1 q_1 \tag{16}$$

where M_d is the mass of a droplet and t is time.

(B) Droplets of Arbitrary Composition. Consider a single multicomponent droplet composed of Λ volatile components immersed into a stagnant mixture of $\Lambda + 1$ components with temperature T_∞. It is assumed that Λ constituent species of the ambient gas mixture are volatile components of the droplet and $\Lambda + 1$th species is an ambient gas. The influence of natural convection on evaporation is neglected. Since the characteristic times of thermal and diffusion relaxation are small, we can consider a steady-state evaporation. Under the above assumptions, the evaporation of a multicomponent droplet can be described by a system of conservation Eqs. (2). However, the densities of energy and mass fluxes in a multicomponent gas mixture are given by Eqs. (5)–(6) (see Table 1).

Using the effective values of velocity and diffusivity (Frank-Kamenetskii, 1969, Chapter IV):

$$\mathbf{v}_i = \frac{1}{n} \frac{\sum_k \frac{\mathbf{j}_k}{D_{ik}}}{\sum_k \frac{C_k}{D_{ik}}}, \quad \bar{D}_i = \frac{\sum_k C_k}{\sum_k \frac{C_k}{D_{ik}}} \tag{17}$$

in the Eq. (6) the mass conservation equation can be written as follows:

$$\operatorname{div}(n_i \mathbf{v}_i + \bar{D}_i n \nabla C_i) = 0 \tag{18}$$

where $\bar{D}_i$ is effective diffusivity that can be calculated using Wilke's rule.

When the temperature differences in the neighborhood of a droplet are small, the Stefan flux can be neglected and transport coefficients are constant. In this case, Eq. (18) can be linearized; one is presented in Table 1.

Finally, the rate of droplet evaporation can be found from the following differential equation:

$$\frac{dM_d}{dt} = -\sum_j m_j q_j. \tag{19}$$

2.2 Modified Method of Expansion into Irreducible Multipoles. A detailed description of the modified method of expansion into irreducible multipoles and its application to the evaporation and combustion of dense random clusters of droplets and particles can be found in Elperin and Krasovitov 1994a, 1994b. For the sake of completeness, a brief discussion of this method is presented below.

Consider a system containing N droplets whose centers are located at $\mathbf{r} = \mathbf{r}_i$ and which have radii R_i, where $i = \overline{1, N}$. The location of each droplet is fixed in a system of coordinate X, Y, Z the origin of which can be located in the center of each droplet. The location of jth droplet can be determined in the system of coordinate X_i, Y_i, Z_i which is connected with the center of each droplet by the vector $\mathbf{L}_{ij}$. Similarly, the location of ith droplet can be determined in the coordinate system X_j, Y_j, Z_j by the vector $\mathbf{L}_{ji} = -\mathbf{L}_{ij}$.

Let U denote concentration or temperature fields which are

Table 2 Boundary conditions and conditions at infinity

The existence of temperature and concentration jumps at the surface of a droplet:

$$(T_e - T_w)|_{r=R} = K_T^{(T)} \left.\frac{\partial T_e}{\partial r}\right|_{r=R} + K_T^{(n)} T_\infty \left.\frac{\partial C_1}{\partial r}\right|_{r=R} \tag{7}$$

$$(C_1 - C_1^{(s)}(T_w))|_{r=R} = K_n^{(n)} \left.\frac{\partial C_1}{\partial r}\right|_{r=R} + \frac{K_n^{(T)}}{T_\infty} \left.\frac{\partial T_e}{\partial r}\right|_{r=R} \tag{8}$$

The continuity of energy flux at the droplet surface:

$$\frac{W_T}{4\pi R^2} = (\sum_i (L_i - h_i) m_i j_{r,i} + j_{r,T} + j_{r,R})|_{r=R} \tag{9}$$

Conditions at infinity:

$$T_e|_{r\to\infty} = T_\infty, \quad C_1|_{r\to\infty} = C_{1\infty} \tag{10}$$

governed by Eq. (2) (see Table 1) where flux Γ_ϕ is determined by the following equation:

$$\Gamma_\phi = \nabla U. \tag{20}$$

Notably, most practical applications involve high temperature differences during evaporation (combustion), and the Stefan's flux must be taken into account. Moreover, the energy and mass conservation equations are nonlinear on the account of dependence of heat and mass transfer coefficients on temperature (see Table 1). However, methods were developed which allow conversion of the Stefan flow (SF) problem into a non-Stefan flow (NSF) problem (Annamalai, 1993) and conversion of the nonlinear problem into the Laplace equation (Elperin and Krasovitov 1994a, 1994b). Therefore, the general problem can reduced to the solution of the Dirichlet, Neumann, or mixed boundary value problem for the Laplace equation in the domain exterior to N arbitrary located droplets.

The boundary condition at the surface of ith droplet and the condition at infinity read:

$$U|_{S_i} = U_i^{(s)} = \text{const}, \quad U|_{|\mathbf{r}-\mathbf{r}_i|\to\infty} \to 0. \tag{21}$$

In case when Eq. (2) is linearized, the solution U in the vicinity of the ith droplet can be represented as a superposition:

$$U = U_i + \delta U_i, \quad \delta U_i = \sum_{\substack{j=1 \\ j\neq i}}^{N} U_j \tag{22}$$

where U_i is the field generated by the ith droplet and δU_i is the perturbation which is caused by the presence of $N-1$ other droplets. Note that in order to simplify notations, hereafter, we omit index i near the solution U in the vicinity of the ith droplet. Note also that the field function in the neighborhood of the ith droplet has singularities at the centers of other $N-1$ droplets.

The field function (e.g., temperature or concentration) can be represented as a tensor expansion suggested by Schmitz and Felderhof (1982):

$$U_i = \sum_{n=0}^{\infty} A^i_{\nu_1\ldots\nu_n} x_i^{-(2n+1)} \overrightarrow{x^i_{\nu_1}\ldots x^i_{\nu_n}}$$

$$\delta U_i = \sum_{n=0}^{\infty} B^i_{\nu_1\ldots\nu_n} \overrightarrow{x^i_{\nu_1}\ldots x^i_{\nu_n}} \tag{23}$$

where the tensor $\overrightarrow{x^i_{\nu_1}\ldots x^i_{\nu_n}}$, defined as follows:

$$\overrightarrow{x^i_{\nu_1}\ldots x^i_{\nu_n}} = \frac{(-1)^n}{(2n-1)!!}\, x_i^{2n+1} \nabla^i_{\nu_1}\ldots\nabla^i_{\nu_n}\left(\frac{1}{x_i}\right) \tag{24}$$

is a symmetrical tensor with $2n+1$ independent components; $\nabla^i_{\nu_k} = \partial_{x^i_{\nu_k}}$, $x^i_{\nu_k} = (\tilde{x}_{\nu_k} - \tilde{x}^i_{\nu_k})/R_i$, $\tilde{x}_{\nu_k}$ and $\tilde{x}^i_{\nu_k}$ ($\nu_k = \overline{1,3}$, $k = \overline{1,n}$) are current Cartesian coordinates and the Cartesian coordinate of the center of the ith sphere, respectively, and $x_i = \sqrt{\sum_{\nu_k} (x^i_{\nu_k})^2}$ (for details see, e.g., Hess and Köhler, 1980). It is shown below that the latter representation of the solution allows to satisfy the boundary conditions at the surface of each droplet. Since U_i and δU_i are harmonic functions, the coefficients $A^i_{\nu_1\ldots\nu_n}$ and $B^i_{\nu_1\ldots\nu_n}$ are the constant symmetrical tensors that are traceless in any pair of their indices. These tensors must be determined from the boundary conditions.

Using the boundary condition (21) we can express the coefficients $A^i_{\nu_1\ldots\nu_n}$ via the coefficients $B^i_{\nu_1\ldots\nu_n}$:

$$A^i_0 = U_i^{(s)} - B^i_0, \quad \text{for} \quad n = 0$$

$$A^i_{\nu_1\ldots\nu_n} = -B^i_{\nu_1\ldots\nu_n}, \quad \text{for} \quad \forall n \geq 1. \tag{25}$$

Substituting (25) into expansion (23) and using (22) we find that

$$U = U_i^{(s)}(1/x_i) + \sum_{n=0}^{\infty} B^i_{\nu_1\ldots\nu_n}(1 - x_i^{-(2n+1)})\overrightarrow{x^i_{\nu_1}\ldots x^i_{\nu_n}}. \tag{26}$$

According to (22), the formula for field perturbation in the neighborhood of the ith droplet reads as follows:

$$\delta U_i = \sum_{\substack{j=1 \\ j\neq i}}^{N} \left[U_j^{(s)}(1/x_j) - \sum_{n=0}^{\infty} B^j_{\nu_1\ldots\nu_n} x_j^{-(2n+1)} \overrightarrow{x^j_{\nu_1}\ldots x^j_{\nu_n}}\right]. \tag{27}$$

Suppose that the origins of the systems of Cartesian coordinates O_k ($k = 1, \ldots, N$) are located at the centers of each droplet. Then, if the axes of these systems are parallel (i.e., $O_1X_1 \| O_2X_2 \| \ldots \| O_NX_N$; $O_1Y_1 \| O_2Y_2 \| \ldots \| O_NY_N$; $O_1Z_1 \| O_2Z_2 \| \ldots \| O_NZ_N$), the Taylor's series expansion of $x_j^{-(2n+1)}\overrightarrow{x^j_{\nu_1}\ldots x^j_{\nu_n}}$ can be written as follows:

$$x_j^{-(2n+1)}\overrightarrow{x^j_{\nu_1}\ldots x^j_{\nu_n}} = \sum_{k=0}^{\infty} \omega_{kn}\epsilon_j^{n+1}\epsilon_i^k \Omega_{\nu_1\ldots\nu_n\mu_1\ldots\mu_k}(\mathbf{L}_{ij})\overrightarrow{x^i_{\mu_1}\ldots x^i_{\mu_k}} \tag{28}$$

where

$$\epsilon_i = \frac{R_i}{|\mathbf{L}_{ij}|}, \ \epsilon_j = \frac{R_j}{|\mathbf{L}_{ij}|},$$

$$\omega_{kn} = \frac{(-1)^{k+n}[2(n+k)-1]!!}{k!(2n-1)!!}, \quad \nabla^{(i,j)}_{\nu_k} = \partial_{L^{(i,j)}_{\nu_k}},$$

$$\Omega_{\nu_1\ldots\nu_n\mu_1\ldots\mu_k}(\mathbf{L}_{ij}) = \frac{(-1)^{k+n}L_{ij}^{k+n+1}}{[2(n+k)-1]!!} \times \left[\nabla^{(i,j)}_{\nu_1}\ldots\nabla^{(i,j)}_{\nu_n}\nabla^{(i,j)}_{\mu_1}\ldots\nabla^{(i,j)}_{\mu_k}\left(\frac{1}{L_{ij}}\right)\right]$$

and $L^{(i,j)}_{\nu_k}$, Cartesian coordinates of the vector $\mathbf{L}_{ij}$.

Substituting expansion (28) into the expression (27), and using expression (23), we can obtain the infinite system of linear algebraic equations for the determination of the coefficients $B^i_{\nu_1\ldots\nu_n}$:

$$B^i_{\nu_1\ldots\nu_n} = B^{i(0)}_{\nu_1\ldots\nu_n} - \sum_{\substack{j=1 \\ j\neq i}}^{N}\sum_{k=0}^{\infty} B^j_{\mu_1\ldots\mu_k}\omega_{nk}\epsilon_j^{k+1}\epsilon_i^n \Omega_{\nu_1\ldots\nu_n\mu_1\ldots\mu_k}(\mathbf{L}_{ij}) \tag{29}$$

where

$$B^{i(0)}_{\nu_1\ldots\nu_n} = \sum_{\substack{j=1 \\ j\neq i}}^{N} U_j^{(s)}\omega_{n0}\epsilon_j\epsilon_i^n \Omega_{\nu_1\ldots\nu_n}(\mathbf{L}_{ij}).$$

The system of Eqs. (29) must be solved by a truncation procedure. Our past experience with the use of expansion into irreducible multipoles showed that convergence characteristics of the system are good, and only a few equations are required to obtain a high accuracy.

2.3 Evaporation of Dense Random Clusters of Multicomponent Droplets.

(A) Interactive Evaporation and Growth of Droplets With Volatile and Nonvolatile Component. Consider a cluster of N spherical droplets composed of volatile and nonvolatile components. It is assumed that there are no internal heat sources inside the droplets and the temperature differences in the neighborhood of each droplet are small. Assume that the characteristical times

of heat and diffusive relaxation are small and we can consider a steady-state evaporation.

Under the above assumptions, the system of mass and energy conservation equations reads:

$$\operatorname{div} \tilde{\Gamma}_\phi = 0 \tag{30}$$

where the flux $\tilde{\Gamma}_\phi$ is determined by equations presented in Table 3. Boundary conditions for conservation Eqs. (30) read:

$$\tilde{C}^{(e)}|_{S_i} = \tilde{C}_i^{(s)}, \quad \tilde{T}^{(e)}|_{S_i} = \tilde{T}_i^{(s)} \tag{31}$$

$$Lm_1 n D^{(e)} (\nabla \tilde{C}^{(e)} \mathbf{n}_r)|_{S_i} + k^{(e)} (\nabla \tilde{T}^{(e)} \mathbf{n}_r)|_{S_i} = k^{(w)} (\nabla \tilde{T}_i^{(w)} \mathbf{n}_r)|_{S_i}. \tag{32}$$

Conditions at infinity read:

$$\tilde{T}^{(e)}|_{|\mathbf{r}-\mathbf{r}_i|\to\infty} \to 0, \quad \tilde{C}^{(e)}|_{|\mathbf{r}-\mathbf{r}_i|\to\infty} \to 0. \tag{33}$$

Note that in case of small temperature differences in the neighborhood of each droplet we can expand the nondimensional vapor concentration at the gas-liquid interface in Taylor series and keep only the linear term:

$$\tilde{C}^{(e)}|_{S_i} = \tilde{C}_i^{(s)} = C_s^i(T_\infty) + \left.\frac{\partial C_s^i}{\partial \tilde{T}_s^i}\right|_{\tilde{T}_s^i=0} \tilde{T}_s^i + \dots$$

where $C_s^i = C_{1,s}^i - C_{1,\infty}$ and $\tilde{T}_s^i = T_{i,s} - T_\infty$. Concentrations $C_{1,s}^i$ can be determined by the Raoult's law (for ideal solutions) and by the formula (15) (for nonideal solutions).

Using the approach discussed in Section 2.2, the temperature and concentration distributions in the neighborhood of the ith droplet can be obtained as follows:

$$\tilde{C}^{(e)} = C^{i(0)} + \sum_{n=0}^{\infty} (B^i_{\nu_1\dots\nu_n}(1 + x_i^{-(2n+1)}\lambda^i_{bn}) + Q^i_{\nu_1\dots\nu_n} x_i^{-(2n+1)}\lambda^i_{qn}) \overline{x^i_{\nu_1} \dots x^i_{\nu_n}} \tag{34}$$

$$\tilde{T}^{(e)} = T_e^{i(0)} + \sum_{n=0}^{\infty} (Q^i_{\nu_1\dots\nu_n}(1 + x_i^{-(2n+1)}\varphi^i_{qn}) + B^i_{\nu_1\dots\nu_n} x_i^{-(2n+1)}\varphi^i_{bn}) \overline{x^i_{\nu_1} \dots x^i_{\nu_n}} \tag{35}$$

where $C^{i(0)} = x_i^{-1}\lambda_n^{i(0)}\delta_{0n}$ and $T_e^{i(0)} = x_i^{-1}\varphi_n^{i(0)}\delta_{0n}$ are nonperturbed fields of concentration and temperature generated by ith droplet.

The temperature distribution inside the ith droplet is determined by the following formula:

$$\tilde{T}_i^{(w)} = \varphi_n^{i(0)}\delta_{0n} + \sum_{n=0}^{\infty} (\varphi^i_{bn} B^i_{\nu_1\dots\nu_n} + (1 + \varphi^i_{qn}) Q^i_{\nu_1\dots\nu_n}) \overline{x^i_{\nu_1} \dots x^i_{\nu_n}}. \tag{36}$$

In (34)–(36)

$$\lambda_n^{i(0)} = \frac{(C^i_{1,s}(T_\infty) - C_{1,\infty})((n+1)k^{(e)} + nk_i^{(w)})}{(n+1)(k^{(e)} + \gamma^i D^{(e)} n^{(e)} Lm_1) + nk_i^{(w)}},$$

$$\lambda^i_{bn} = -\frac{\lambda_n^{i(0)}}{(C^i_{1,s}(T_\infty) - C_{1,\infty})},$$

$$\lambda^i_{qn} = \frac{(2n+1)k^{(e)}\gamma^i}{(n+1)(k^{(e)} + \gamma^i D^{(e)} n^{(e)} Lm_1) + nk_i^{(w)}},$$

$$\gamma^i = C^i_{1,s}(T_\infty)\frac{L\mu_1}{R_g T_\infty^2},$$

$$\varphi_n^{i(0)} = -\frac{(n+1)(C^i_{1,s}(T_\infty) - C_{1,\infty})Lm_1 n^{(e)} D^{(e)}}{(n+1)(k^{(e)} + \gamma^i D^{(e)} n^{(e)} Lm_1) + nk_i^{(w)}},$$

$$\varphi^i_{bn} = -\frac{\varphi_n^{i(0)}}{(C^i_{1,s}(T_\infty) - C_{1,\infty})},$$

$$\varphi^i_{qn} = \frac{-n(k_i^{(w)} - k^{(e)}) - (n+1)Lm_1 n^{(e)} D^{(e)}\gamma^i}{(n+1)(k^{(e)} + \gamma^i D^{(e)} n^{(e)} Lm_1) + nk_i^{(w)}}.$$

As can be seen from above formulas the coefficient of heat conductivity inside the droplet does not enter into the temperature and concentration distributions for $n = 0$. The analysis of these expressions and estimations of the coefficients for $n > 0$ show that for a cluster of droplets with high thermal conductivity the solution of the problem can be carried out in the region outside the droplets only.

Using expansion (28) the unknown coefficients $B^i_{\nu_1\dots\nu_n}$ and $Q^i_{\nu_1\dots\nu_n}$ can be found from the following system of equations:

$$B^i_{\nu_1\dots\nu_n} = B^{i(0)}_{\nu_1\dots\nu_n} + \sum_{\substack{j=1\\ j\neq i}}^{N}\sum_{k=0}^{\infty} (\lambda^j_{bk} B^j_{\mu_1\dots\mu_k} + \lambda^j_{qk} Q^j_{\mu_1\dots\mu_k}) \times \omega_{nk}\epsilon_j^{k+1}\epsilon_i^n \Omega_{\nu_1\dots\nu_n\mu_1\dots\mu_k}(\mathbf{L}_{ij}) \tag{37}$$

$$Q^i_{\nu_1\dots\nu_n} = Q^{i(0)}_{\nu_1\dots\nu_n} + \sum_{\substack{j=1\\ j\neq i}}^{N}\sum_{k=0}^{\infty} (\varphi^j_{bk} B^j_{\mu_1\dots\mu_k} + \varphi^j_{qk} Q^j_{\mu_1\dots\mu_k}) \times \omega_{nk}\epsilon_j^{k+1}\epsilon_i^n \Omega_{\nu_1\dots\nu_n\mu_1\dots\mu_k}(\mathbf{L}_{ij}) \tag{38}$$

where

$$B^{i(0)}_{\nu_1\dots\nu_n} = \sum_{\substack{j=1\\ j\neq i}}^{N} \lambda_0^{j(0)}\omega_{k0}\epsilon_j\epsilon_i^k \Omega_{\nu_1\dots\nu_n}(\mathbf{L}_{ij}) \text{ and}$$

$$Q^{i(0)}_{\nu_1\dots\nu_n} = \sum_{\substack{j=1\\ j\neq i}}^{N} \varphi_0^{j(0)}\omega_{k0}\epsilon_j\epsilon_i^k \Omega_{\nu_1\dots\nu_n}(\mathbf{L}_{ij}).$$

The solution of the above system of coupled linear algebraic equations can be found by Jacobi-Zeidel iterations with successive underrelaxation and can be calculated with any accuracy of the nth power of the small parameter ϵ. All the calculations in this study were performed with the accuracy $O(\epsilon^5)$.

Table 3 Formulation of parameters for the problem of condensation (evaporation) of a cluster of droplets containing a volatile and non volatile component

Conservation equation	$\tilde{\Gamma}_\phi$	Definitions of parameters
Mass (outside droplets)	$\nabla\tilde{C}^{(e)}$	$\tilde{C}^{(e)} = C_1^{(e)} - C_{1,\infty}$, $C_{1,\infty} = \lim_{\lvert\mathbf{r}-\mathbf{r}_i\rvert\to\infty} C_1^{(e)}$, $C_k^{(e)} = n_k^{(e)}/\sum_k n_k^{(e)}$
Energy (outside droplets)	$\nabla\tilde{T}^{(e)}$	$\tilde{T}^{(e)} = T^{(e)} - T_\infty$, $T_\infty = \lim_{\lvert\mathbf{r}-\mathbf{r}_i\rvert\to\infty} T^{(e)}$
Energy (inside droplets)	$\nabla\tilde{T}_i^{(w)}$	$\tilde{T}_i^{(w)} = T_i^{(w)} - T_\infty$, $i = \overline{1, N}$

As was suggested by Labowsky (1980a) for evaporating clusters it is more convenient to represent the obtained results in terms of correction factor. In the notations of the present solution, the heat and mass correction factors could be determined as follows:

$$\zeta_i = -\frac{1}{4\pi R_i \varphi_0^i}\int (\nabla \tilde{T}^{(e)}\mathbf{n}_r)|_{S_i} R_i^2 \sin\vartheta_i\, d\vartheta_i\, d\varphi_i$$

$$\eta_i = -\frac{1}{4\pi R_i \lambda_0^i}\int (\nabla \tilde{C}^{(e)}\mathbf{n}_r)|_{S_i} R_i^2 \sin\vartheta_i\, d\vartheta_i\, d\varphi_i \quad (39)$$

where $\varphi_0^i = \varphi_n^{i(0)}$ $(n = 0)$ and $\lambda_0^i = \lambda_n^{i(0)}$ $(n = 0)$.
Keeping only the first term in expansions (34), (35) we obtain

$$\zeta_i = 1 + \frac{\varphi_{b0}^i}{\varphi_0^i} B_0^i + \frac{\varphi_{q0}^i}{\varphi_0^i} Q_0^i, \quad \eta_i = 1 + \frac{\lambda_{b0}^i}{\lambda_0^i} B_0^i + \frac{\lambda_{q0}^i}{\lambda_0^i} Q_0^i. \quad (40)$$

The expressions for Sherwood and Nusselt numbers read:

$$\mathrm{Sh}_i = 2\,\frac{\lambda_0^i + \lambda_{b0}^i B_0^i + \lambda_{q0}^i Q_0^i}{\lambda_0^i + (1 + \lambda_{b0}^i) B_0^i + \lambda_{q0}^i Q_0^i},$$

$$\mathrm{Nu}_i = 2\,\frac{\varphi_0^i + \varphi_{b0}^i B_0^i + \varphi_{q0}^i Q_0^i}{\varphi_0^i + \varphi_{b0}^i B_0^i + (1 + \varphi_{q0}^i) Q_0^i}. \quad (41)$$

Finally, the rate of evaporation can be found from the Eq. (16) in which the value of mass flux q_1 takes the form

$$q_1^i = \int n D^{(e)} (\nabla \tilde{C}^{(e)}\mathbf{n}_r)|_{S_i} R_i^2 \sin\vartheta_i\, d\vartheta_i\, d\varphi_i. \quad (42)$$

(B) Interactive Evaporation and Growth of Clusters of Multicomponent Droplets With Arbitrary Volatile Components. Similar to the previous section, we consider a system containing N spherical multicomponent droplets immersed into a stagnant mixture of $\Lambda + 1$ components with temperature T_∞. It is assumed that Λ constituent species of the ambient gas mixture are volatile components of the droplet and $\Lambda + 1$th species is an ambient gas. The analysis of evaporation (condensation) of clusters of multicomponent droplets with arbitrary volatile components and small temperature differences in the neighborhood of the droplets will be performed in case of droplets composed of ideal mixtures. Ideal mixtures approximation can be used when mixtures are formed by substances with similar molecular properties (e.g., hexane-heptane, benzene-chlorobenzene, etc.). Since the characteristic times of heat and diffusion relaxation are small, we can consider the steady-state evaporation (condensation). Moreover as it was shown in the previous section when the coefficient of conductivity of droplets is much higher then the coefficient of heat conductivity of the ambient gas, the solution of the problem can be carried out in the domain outside the droplets only. Thus, under the above assumptions the linearized system of mass and energy conservation equations reads:

$$\nabla^2 \tilde{T}^{(e)} = 0, \quad \nabla^2 \tilde{C}_\beta^{(e)} = 0. \quad (43)$$

The boundary conditions for the above system of conservation equations (43) can be written as follows:

$$\tilde{T}^{(e)}|_{S_i} = \tilde{T}_i^{(s)}, \quad \tilde{C}_\beta^{(e)}|_{S_i} = \tilde{C}_{\beta,i}^{(s)} \quad (44)$$

$$k^{(e)}(\nabla \tilde{T}^{(e)}\mathbf{n}_r)|_{S_i} + \sum_{\beta=1}^{\Lambda} L_\beta m_\beta n^{(e)} \bar{D}_\beta (\nabla \tilde{C}_\beta^{(e)}\mathbf{n}_r)|_{S_i} = 0 \quad (45)$$

where the values $\tilde{T}^{(e)}$ and $\tilde{C}_\beta^{(e)}$ are defined similarly to the parameters shown in Table 3, subscript β denotes the number of species, and $\bar{D}_\beta$ is the effective diffusion coefficient.

Conditions at infinity read as

$$\tilde{C}_\beta^{(e)}|_{|\mathbf{r}-\mathbf{r}_i|} \to 0, \quad \tilde{T}^{(e)}|_{|\mathbf{r}-\mathbf{r}_i|} \to 0. \quad (46)$$

Using the approach described in the above sections, the following expressions for the temperature and concentration distributions in the neighborhood of the ith droplet were obtained:

$$\tilde{T}^{(e)} = T^{i(0)} + \sum_{n=0}^{\infty} \Big(x_i^{-(2n+1)} \sum_{\beta=1}^{\Lambda} \alpha_{\beta,gn}^i G_{\beta,\nu_1\ldots\nu_n}^i + (1 + \alpha_{hn}^i x_i^{-(2n+1)}) H_{\nu_1\ldots\nu_n}^i \Big) \overline{x_{\nu_1}^i \ldots x_{\nu_n}^i} \quad (47)$$

$$\tilde{C}_\beta^{(e)} = C_\beta^{i(0)} + \sum_{n=0}^{\infty} \Bigg(x_i^{-(2n+1)} \psi_{\beta,hn}^i H_{\nu_1\ldots\nu_n}^i + \Bigg(1 + \gamma_\beta^i \sum_{l=1}^{\Lambda} \Big[\alpha_{l,gn}^i - \frac{1}{\gamma_\beta^i}\delta_{l\beta} \Big] x_i^{-(2n+1)} \Bigg) G_{\beta,\nu_1\ldots\nu_n}^i \Bigg) \overline{x_{\nu_1}^i \ldots x_{\nu_n}^i} \quad (48)$$

where

$$T_e^{(0)} = \alpha_0^i x_i^{-1}, \quad C_\beta^{i(0)} = \psi_\beta^{i(0)} x_i^{-1},$$

$$\psi_\beta^{i(0)} = C_{\beta,i}^{(s)}(T_\infty)\Big(1 + \frac{L_\beta \mu_\beta}{R_g T_\infty^2}\alpha_0^i \Big)$$

$$\alpha_{hn}^i = \frac{k^{(e)} n - (n+1)\sum_{\beta=1}^{\Lambda} L_\beta m_\beta n^{(e)} \bar{D}_\beta \gamma_\beta^i}{(n+1)(k^{(e)} + \sum_{\beta=1}^{\Lambda} L_\beta m_\beta n^{(e)} \bar{D}_\beta \gamma_\beta^i)},$$

$$\alpha_{\beta,gn}^i = \frac{(2n+1) L_\beta m_\beta n^{(e)} \bar{D}_\beta}{(n+1)(k^{(e)} + \sum_{\beta=1}^{\Lambda} L_\beta m_\beta n^{(e)} \bar{D}_\beta \gamma_\beta^i)},$$

$$\alpha_0^i = \frac{-\sum_{\beta=1}^{\Lambda} L_\beta m_\beta n^{(e)} \bar{D}_\beta C_{\beta,i}^{(s)}(T_\infty)}{k^{(e)} + \sum_{\beta=1}^{\Lambda} L_\beta m_\beta n^{(e)} \bar{D}_\beta \gamma_\beta^i},$$

$$\gamma_\beta^i = C_{\beta,i}^{(s)}(T_\infty)\frac{L_\beta \mu_\beta}{R_g T_\infty^2}, \quad \psi_{\beta,hn}^i = \gamma_\beta^i (1 + \alpha_{hn}^i),$$

and $\delta_{l\beta}$ is Kronecker's symbol.

Using formula (28), the unknown coefficients $G_{\beta,\nu_1\ldots\nu_n}^i$ and $H_{\nu_1\ldots\nu_n}^i$ can be found from the following system of linear algebraic equations:

$$H_{\nu_1\ldots\nu_n}^i = H_{\nu_1\ldots\nu_n}^{i(0)} + \sum_{\substack{j=1\\ j\neq i}}^{N} \sum_{k=0}^{\infty} \Big(\alpha_{hk}^j H_{\nu_1\ldots\nu_k}^j + \sum_{\beta=1}^{\Lambda} \alpha_{\beta,gk}^j G_{\beta,\nu_1\ldots\nu_k}^j \Big) \Theta_{nk}^{(i,j)} \quad (49)$$

$$G_{\beta,\nu_1\ldots\nu_n}^i = G_{\beta,\nu_1\ldots\nu_n}^{i(0)} + \sum_{\substack{j=1\\ j\neq i}}^{N} \sum_{k=0}^{\infty} \Bigg(\psi_{\beta,hk}^j H_{\nu_1\ldots\nu_k}^j + \gamma_\beta^j \sum_{l=1}^{\Lambda} \Big[\alpha_{\beta,gl}^j - \frac{1}{\gamma_\beta^j}\delta_{l\beta} \Big] G_{\beta,\nu_1\ldots\nu_k}^j \Bigg) \Theta_{nk}^{(i,j)} \quad (50)$$

where

$$\Theta_{nk}^{(i,j)} = \omega_{nk} \epsilon_j^{k+1} \epsilon_i^n \Omega_{\nu_1\ldots\nu_n\mu_1\ldots\mu_k}(\mathbf{L}_{ij}).$$

Similar to previous analysis, the solution of the above system of coupled linear algebraic equations can be found by Jacobi-Zeidel iterations with successive underrelaxation and can be calculated with any accuracy of the nth power of the small parameter ϵ.

In notations of the above solution, the heat and mass correction factors could be determined as follows:

$$\zeta_i = 1 + \frac{1}{\alpha_0^i} \sum_{\beta=1}^{\Lambda} \alpha_{\beta,g0}^i G_{\beta,0}^i + \frac{(1+\alpha_{h0}^i)}{\alpha_0^i} H_0^i$$

$$\eta_{\beta,i} = 1 + \frac{\gamma_\beta^{i(0)}}{\psi_\beta^{i(0)}} \sum_{l=1}^{\Lambda} \left[\alpha_{l,gn}^i - \frac{1}{\gamma_\beta^{i(0)}} \delta_{l\beta} \right] G_{\beta,0}^i + \frac{\psi_{\beta,hn}^i}{\psi_\beta^{i(0)}} H_0^i. \quad (51)$$

For simplicity, we assume that the total droplet volume equals to the sum of the mixing species volumes. The error of the latter approximation is about 2.5 percent. Then, using this approximation, the rate of evaporation can be found from the following system of equations:

$$\frac{dR_i}{dt} = -\sum_{\beta=1}^{\Lambda} \frac{m_\beta}{4\pi R_i^2 \rho_\beta^{(0)}} q_\beta^i(R_i, M_\beta^i)$$

$$\frac{dM_\beta^i}{dt} = -m_\beta q_\beta^i(R_i, M_\beta^i) \quad (52)$$

with the following initial conditions: $R_i = R_{i0}$, $M_\beta^i = M_{\beta,0}^i$ at $t = 0$.

In (52), $\rho_\beta^{(0)}$ is the density of pure βth species, $q_\beta^i(R_i, M_\beta^i)$ is the integral mass flux of βth species, and M_β^i is the mass of the βth species within the ith droplet.

3 Results and Discussion

The solutions derived in the previous sections have been applied to the analysis of interactions between multicomponent droplets and the effect of these interactions on evaporation (condensation) rates for variety configurations of evaporating (growing) clusters. The results were obtained for binary arrays and large clusters of droplets (i) composed of volatile components and (ii) composed of a volatile and a soluble nonvolatile component.

The results of calculation of Nusselt and Sherwood numbers for binary array of droplets consisting of a volatile and a soluble nonvolatile component are shown in Figs. 1 and 2 where two droplets of the same size but with different mole fraction of a volatile species (droplet 1, volatile species: 80 percent; droplet 2, volatile species: 20 percent) are considered. In the calculations it was assumed that the mixture of a volatile component and a soluble nonvolatile component constitutes the ideal solution; octane, heptane, and hexane were considered volatile components. As can be seen from these plots, Nusselt and Sherwood numbers strongly depend on the kind of volatile species and on the distance between droplets. When the relative distance tends to infinity (i.e., $L/R \to \infty$) Sherwood and Nusselt numbers asymptotically approach the value 2, appropriate for a single droplet. In a case when the relative distance decreases, Nusselt and Sherwood numbers of predominantly volatile droplet (droplet 1, with 80 percent volatile species) increase and Nusselt and Sherwood numbers of a droplet with a lower content of volatile component (droplet 2, with 20 percent volatile species) decrease. Therefore, at small distances between droplets, the evaporation rate of a droplet with a higher content of a volatile component increases and a droplet with lower content of a volatile component grows, and its surface temperature increases. Notably, Nusselt and Sherwood numbers increase beyond 2 when octane is the volatile species and the L/R ratio varies in the range from 2 to 3.

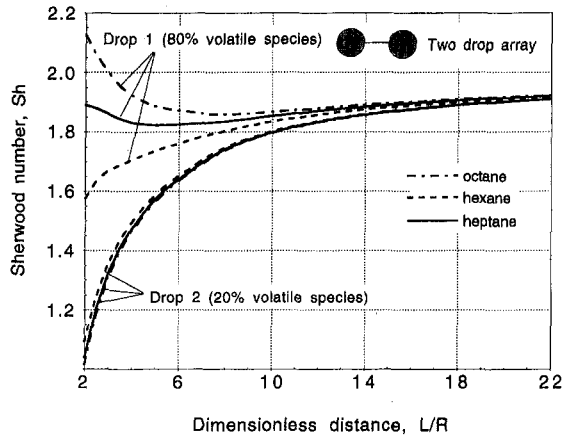

Fig. 2 Sherwood numbers for monosized binary array of droplets with volatile and nonvolatile species (drop 1–80 percent volatile species; drop 2–20 percent volatile species)

Interactive growth of droplets on hygroscopic nuclei is encountered in various environmental applications. In particular, small aerosol soluble particles act as nuclei for droplet condensation in atmospheric clouds. In this case, the mixture of a volatile component and a soluble nonvolatile component constitutes the non-ideal solution. In this connection, the evaporation of a binary array, comprised of a pure droplet and a droplet consisting of volatile and a nonvolatile soluble nucleus, was considered. The results of calculating the evaporation rate of a pure water droplet located in the neighborhood of a droplet consisting of water and a hygroscopic nucleus of NaCl are presented in Fig. 3. The calculations were performed for droplets with the radius 3 μm and for the ambient temperature T_∞ = 278 K, pressure p = 1 atm, mass of soluble nucleus M' =

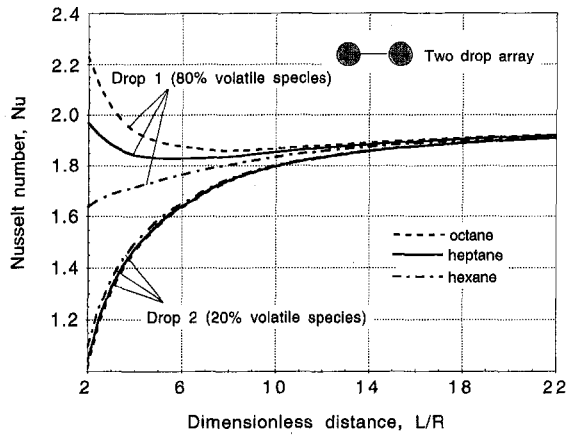

Fig. 1 Nusselt numbers for monosized binary array of droplets with volatile and nonvolatile species (drop 1–80 percent volatile species; drop 2–20 percent volatile species)

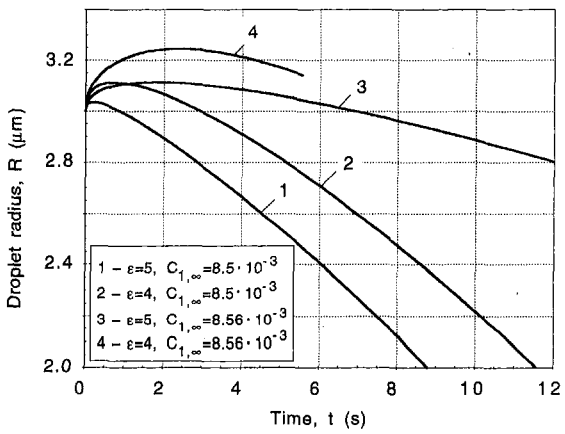

Fig. 3 Evaporation rate of pure water droplet located in the neighborhood of droplet consisting of water and hygroscopic nucleus of NaCl

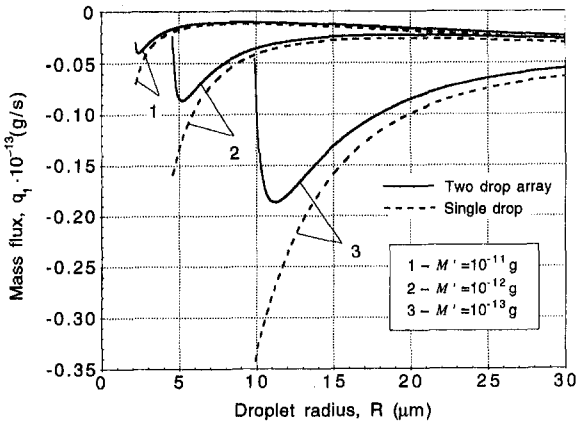

Fig. 4 Mass flux versus radius. Comparison the results obtained for a growing two monosized drop array and single drop contain hygroscopic nuclei NaCl (M'—the mass of hygroscopic nucleus).

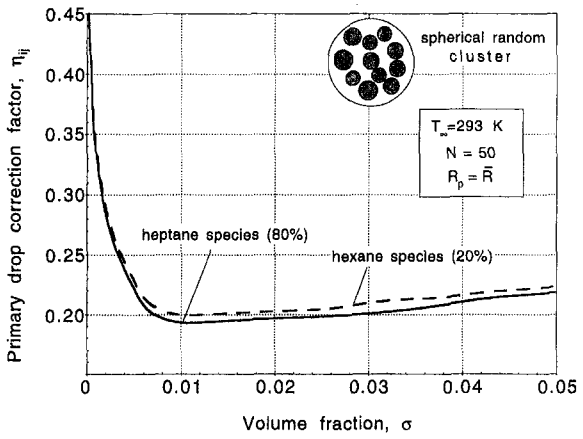

Fig. 6 Average mass correction factor of primary droplet versus volume fraction (radius of the primary droplet is equal to the average radius)

$10^{-10}g$ and various ambient concentrations of volatile species, and center-to-center distances (in this plot ϵ is the L/R ratio). As can be seen from this plot for the considered values of the center-to-center distances and ambient concentrations of volatile species, the pure water droplet grows first and only then the droplet begins to evaporate. In this instance, the droplet containing soluble nucleus (droplet 1) acts as a heat source and a mass sink that causes an increase of vapor concentration in the neighborhood of the droplet. Thus, the pure water droplet (droplet 2) located in the vicinity of droplet 1 grows. When the radii of the pure water droplet and salty droplet increase, the concentration of the solution within the droplet containing a soluble nucleus (droplet 1) decreases. The latter retards the condensation rate at the pure droplet and decreases vapor concentration in the vicinity of the droplets. Thus, inside the dense clusters and clouds containing pure droplets, droplets containing soluble nuclei heat and mass sources and sinks can occur. Therefore, when a dense cloud contains both pure water droplets and droplets containing soluble nuclei, interaction between temperature and concentration fields causes the growth of some droplets in a cluster and evaporation of other droplets. In the case of a finite symmetrical array of compositionally dissimilar interacting droplets this phenomena was originally predicted by Labowsky (1980a) who used the method of images. Later, the theoretical model of recondensation in multicomponent droplet arrays was developed using the point sources method by Annamalai (1993).

The dependence of mass flux on the radius for the array of two identical droplets containing hygroscopic nuclei of NaCl is shown in Fig. 4. In this plot, the solid lines denote the interacting droplets and the dashed lines denote the single droplets. The calculations were performed for the ambient temperature T_∞ = 278 K, ambient pressure p = 1 atm, dimensionless concentration of volatile species $C_{1,\infty}$ = 0.009 and various values of the nuclei's mass. As it can be seen from this plot, the dependence of the mass flux on droplet radius has the minimum in a case of interactive evaporation of droplets. The negative values of mass flux mean that condensation of droplets containing soluble nuclei occurs. Thus, in case of a monosized cluster of droplets the radius of hygroscopic nuclei, which enables the growth of the droplets in the cluster, can be calculated. In clusters with droplet size distribution, the size distribution of hygroscopic nuclei, which enables growth of droplets in a dense cluster, can be determined using the developed theory.

The results of calculation of the average mass correction factor for the primary droplet, $\bar{\eta}_p$, located inside a spherical cluster with log-normal distribution of droplet radii and uniform spatial distribution of droplet are shown in Fig. 5. Notably, the effect of droplet size distribution width on correction factor is small (see Elperin and Krasovitov, 1994b). The calculations were performed for a primary droplet (droplet located in the center of the cloud) with the radii equal to the average radius (solid lines) and 0.5 of average radius (dashed lines). In all calculations presented hereafter, droplet radii and center-to-center distances between droplets are normalized by the average radius. These results are obtained for N = 50 and 100 droplets (N is the number of droplets in the cluster) with octane as

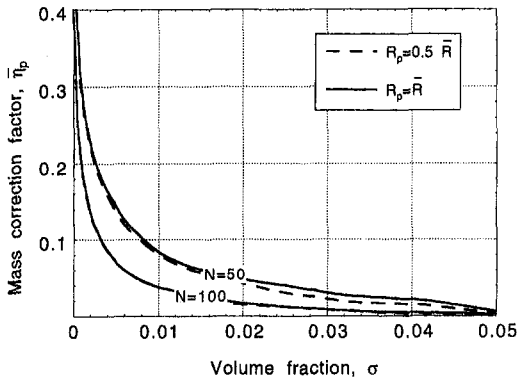

Fig. 5 Variation of average mass correction factor of primary droplet with volatile and nonvolatile component versus volume fraction (N—the number of droplets within the cluster)

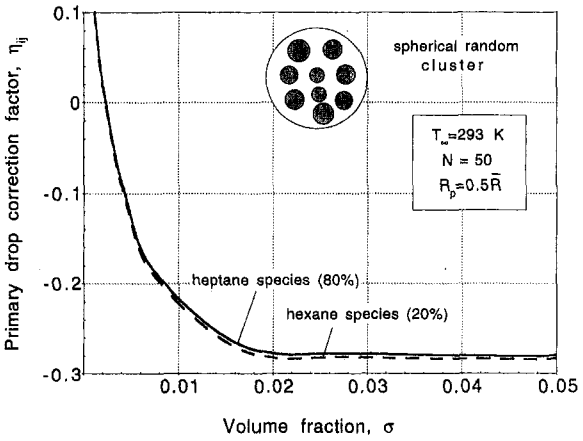

Fig. 7 Average mass correction factor of primary droplet versus volume fraction (radius of the primary droplet is equal to 0.5 of the average radius)

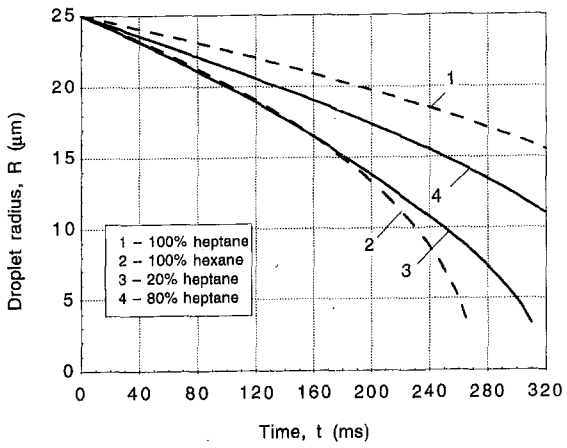

Fig. 8 Droplet radius versus time. Comparison of the results obtained for binary arrays of pure droplets (1—array of heptane droplets; 2—array of hexane droplets) with the results obtained for two drop array of heptane-hexane droplets (4—droplet 1: 80 percent heptane and 20 percent hexane, 3—droplet 2: 20 percent heptane and 80 percent hexane).

a volatile component (octane: 80 percent; soluble nonvolatile component: 20 percent) when the droplet volume fraction is up to five percent inclusive. Note that when droplet volume fraction equals five percent, the average center-to-center dimensionless droplet spacing L_{ij}/R_p (where R_p is the radius of the primary droplet) between neighboring droplets is of order 5, i.e., such clusters are fairly dense. The value of the correction factor for the primary droplet, $\overline{\eta}_p$, was calculated as an average from the calculations of the correction factor $\overline{\eta}_p$ for 100 various randomly sampled configurations of droplets in the cluster.

The results of calculating the average mass correction factor for the primary droplet located inside a spherical random cluster with log-normal distribution of droplet radii and uniform spatial distribution of droplet are shown in Figs. 6 and 7. The calculations were performed for a primary droplet (droplet located in the center of the cloud) with the radii equal to the average radius, Fig. 6, and 0.5 of average radius, Fig. 7. Similar to the previous in the calculations, droplet radii and center-to-center distances between the droplets are normalized by the radius of the primary (trial) droplet. These results are obtained for N = 50 droplets (N is the number of droplets in the cluster) composed of the binary mixture of 80 percent heptane and 20 percent hexane when the droplet volume fraction is up to five percent inclusive. As it can be seen from Fig. 6, the dependence of mass correction factors of heptane and hexane species on volume fraction has a minimum. Remarkably, the increase of the correction factor for more dense clusters occurs due to the phenomena of recondensation whereby evaporating droplets act as mass sources and heat sinks and growing droplets act as mass sink and heat sources. The calculations presented in Fig. 6 were performed for the primary droplet with the radius equal to the average radius of droplets in the cloud. The results of calculations for the primary droplet with 0.5 of the average radius are shown in Fig. 7. As it can be seen from this plot, the primary drop correction factor reverses sign in case of more dense clusters. The latter implies condensation of the primary droplet.

The solution presented above was applied to the analysis of interactions between multicomponent droplets composed of a binary mixture of volatile components. The dependencies of radius on time for an array of two multicomponent droplets is presented in Fig. 8. Droplet 1 has a composition of 80 percent heptane and 20 percent hexane and droplet 2 has a composition of 20 percent heptane and 80 percent hexane. The dashed lines, 1 and 2 in Fig. 8, are plotted for arrays of two heptane and hexane droplets of equal radii correspondingly. The calculations were performed for ambient temperature T_∞ = 283 K, ambient pressure p = 1 atm, and dimensionless concentration of volatile species $C_{\text{Hept},\infty} = 0$ and $C_{\text{Hex},\infty} = 0$.

4 Conclusions

The quasi-steady evaporation of multicomponent drop arrays and random clusters of multicomponent droplets has been studied using the modified method of expansion into irreducible multipoles developed by Elperin and Krasovitov 1994a, 1994b. The evaporation and growth of binary arrays and large random clusters of droplets (i) composed of volatile components and (ii) composed of a volatile and a nonvolatile component were considered. The analytical and numerical results of the investigation are presented in terms of heat and mass correction factors. Transient evaporation and condensation of multicomponent droplet clusters is analyzed, and the evaporation rate is determined. It is shown that for droplets of ideal solution with different compositions, Nusselt and Sherwood numbers depend on the concentration of components inside each droplet. When dense cloud contains both pure droplets and droplets containing soluble nuclei, interactions between temperature and concentration fields causes growth of some droplets in a cluster and evaporation of the other droplets. Remarkably, at a certain stage the growth of the droplets slows down and they begin to evaporate. The developed theory allows to determine the size distribution of hygroscopic nuclei which enables growth of droplets in a dense cloud. It is shown also that inside an evaporating large cluster of multicomponent droplets composed of volatile components, the phenomena of recondensation occurs whereby evaporating droplets act as mass sources and heat sinks and growing droplets act as mass sink and heat sources. The results of this study are of relevance in the analysis of dynamics of droplet size distribution in clouds, artificial modification of clouds and precipitation, in-cloud pollutants scavenging, and in-cloud analysis of evaporation and combustion of multicomponent (blended) fuels.

Acknowledgment

This work was partially supported by the German-Israeli Foundation for Scientific Research (Grant No. I-008-303.06/95).

References

Annamalai, K., Ramalingam, S., Dahdah, T., and Chi, D., 1988, "Group Combustion of a Cylindrical Cloud of Char/Carbon Particles," *Trans. ASME,* Vol. 110, pp. 190–200.

Annamalai, K., and Ryan, W., 1992, "Interactive Processes in Gasification and Combustion. Part I: Liquid Drop Arrays and Clouds," *Prog. Energy Combust. Sci.*, Vol. 18, pp. 221–295.

Annamalai, K., Ryan W., and Chandra, S., 1993, "Evaporation of Multicomponent Drop Arrays," ASME JOURNAL OF HEAT TRANSFER, Vol. 115, pp. 707–716.

Annamalai, K., Ryan, W., and Dhanapalan, S., 1994, "Interactive Processes in Gasification and Combustion—Part III: Coal/Char Particle Arrays, Streams and Clouds," *Prog. Energy Combust. Sci.*, Vol. 20, pp. 487–618.

Bellan, J., and Harstad, K., 1995, "Unsteady Injection of Sequences of Drop Clusters in Vortices Depicting Portions of a Spray," *Atomization and Sprays,* Vol. 5, pp. 17–44.

Carstens, J. C., Williams, A., and J. T. Zung, 1970, "Theory of Droplet Growth in Clouds: II Diffusional Interaction Between Two Growing Droplets," *J. Atmosph. Sci.*, Vol. 27(8), pp. 798–807.

Chiu, H. H., and Liu, T. M., 1977, "Group Combustion of Liquid Droplets," *Comb. Sci. Tech.*, Vol. 17, pp. 127–142.

Deutch, J. M., Felderhof, B. U., and Saxton, M. J., 1976, "Competitive Effects in Diffusion Controlled Reactions," *J. Chem. Phys.*, Vol. 11, pp. 4559–4563.

Eperin, T., and Krasovitov, B., 1994a, "Analysis of Evaporation and Combustion of Random Clusters of Droplets by a Modified Method of Expansion Into Irreducible Multipoles," *Atomization and Sprays,* Vol. 4(1), pp. 79–97.

Elperin, T., and Krasovitov, B., 1994b, "Combustion of Cylindrical and Spherical Random Clusters of Char/Carbon Particles," *Comb. Sci. Tech.*, Vol. 102(1–6), pp. 165–180.

Elperin, T., and Krasovitov, B., 1995, "Radiation, Thermal Diffusion and Kinetic Effects in Evaporation and Combustion of Large and Moderate Size Fuel Droplets," *Int. J. Heat and Mass Transfer,* Vol. 38(3), pp. 409–418.

Frank-Kamenetskii, D. A., 1969, *Diffusion and Heat Transfer in Chemical Kinetics,* Plenum Press, New York.

Heidenreich, S., and Ebert, F., 1995, "Condensatinal Droplet Growth as a Preconditioning Technique for the Separation of Submicron Particles From Gases," *Chem. Eng. Process.,* Vol. 34, pp. 235–244.

Hess, S., und Köhler, W., 1980, *Formeln zur Tensor—Rechnung,* Palm & Enke, Erlangen.

Labowsky, M., and Rosner, D. E., 1978, " 'Group' Combustion of Droplets in the Fuel Clouds: I. Quasi-Steady Predictions," *Adv. Chem. Ser.,* Vol. 166, pp. 63–79.

Labowsky, M., 1980a, "Transfer Rate Calculations for Compositionally Dissimilar Interacting Particles," *Chem. Eng. Sci.,* Vol. 35, pp. 1041–1048.

Labowsky, M., 1980b, "Calculation of the Burning Rates of Interacting Fuel Droplets," *Combust. Sci. Technol.,* Vol. 22, pp. 217–226.

Lefebvre, A. H., 1989, *Atomization and Sprays,* Hemisphere Publishing Co., New York.

Mason, B. J., 1971, *The Physics of Clouds,* 2nd ed., Clarendon Press, Oxford.

Ray, A. K., and Davis, E. J., 1980, "Heat and Mass Transfer With Multiple Particle Interactions: Part I. Droplet Evaporation," *Chem. Eng. Commun.,* Vol. 6, pp. 61–79.

Schmitz, R., and Felderhof, B. U., 1982, "Creeping Flow About a Sphere," *Physica,* Vol. 92A, pp. 423–437.

Tsai, J. T., and Sterling, A. M., 1991, "The Combustion of a Linear Droplet Array in a Convective Coaxial Potential Flow," *Combust. Flame,* Vol. 86, pp. 189–202.

Umemura, A., Ogawa S., and Oshima, N., 1981, "Analysis of Interaction Between Two Burning Fuel Droplets With Different Sizes," *Comb. and Flame,* Vol. 43, pp. 111–119.

Xiong, T. Y., Law, C. K., and Miyasaka, K., 1984, "Interactive Vapoarization and Combustion of Binarydroplet Systems," *20th Symposium (International) on Combustion,* pp. 1781–1787.

Yalamov, Yu. I., Schuckin, E. R., and Alekhin, E. I., 1990, "Calculation of Temperature and Concentration of Discontinuities in a Multicomponent Gas Mixture," *High Temp.,* Vol. 28(2), pp. 184–190.