EQUATIONS FOR DETERMINING THE THERMODIFFUSION RATIO OF A BINARY GAS MIXTURE

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Differential equations describing the temperature and concentration dependence of the thermodiffusion ratio for binary gas mixtures and saturated vapor–gas mixtures are derived on the basis of linear phenomenological equations for the total molecular flux of mass and heat.

The thermodiffusion phenomenon has found wide application in connection with obtaining substances with a high degree of purity. It accompanies processes of heat and mass transfer in the presence of a temperature gradient and can make an appreciable contribution to them. Therefore, neglect of the thermodiffusion effect prevents one from estimating the degree of reliability of a calculated or measured quantity [1].

Calculated values of the thermodiffusion ratio obtained on the basis of the known theoretical methods differ by one to two orders of magnitude from measured values of it in the low-pressure region (up to 1 bar) [2]. A qualitative similarity of the results is not even satisfied in certain cases [1]. The known experimental methods of determining the coefficient of the thermodiffusion ratio of a binary gas mixture are not applicable for a vapor–air mixture.

The absence of reliable methods of calculating the thermodiffusion ratio of a gas mixture, and the imperfection of the experimental method of determining the thermodiffusion ratio of a binary gas mixture in application to a vapor–gas mixture with a nonuniform gas component, limit the possibilities of theoretical research on combined heat and mass transfer in evaporation and condensation processes with allowance for the thermodiffusion effect and diffusional heat conduction.

In this paper we propose a method enabling one to obtain differential equations for determining the thermodiffusion ratio of a binary gas mixture and a vapor–gas mixture.

According to [3], for a binary mixture of ideal gases at a constant total pressure the system of phenomenological equations for the mass- and heat-flux densities, in the case of the absence of convection, has the form

$$\mathbf{J}_1 = -R\frac{\mu_1\gamma_{11}}{x_1x_2}\nabla x_1 - \gamma_{10}\frac{\mu_1}{T^2}\nabla T, \tag{1}$$

$$\mathbf{q} = -\frac{\gamma_{00}}{T^2}\nabla T - R\frac{\gamma_{10}}{x_1x_2}\nabla x_1 + \mathbf{J}_1\left(\frac{\partial \mathrm{H}}{\partial c_1}\right). \tag{2}$$

Expressing ∇x_1 through $\nabla c_1 = \nabla(\rho_1/\rho)$ in these equations, we find

$$\mathbf{J}_1 = -R\frac{\gamma_{11}}{x_1x_2}\frac{\mu_1^2\mu_2}{[\mu_1+(\mu_2-\mu_1)c_1]^2}\nabla c_1 - \gamma_{10}\frac{\mu_1}{T^2}\nabla T, \tag{3}$$

$$\mathbf{q} = -\frac{\gamma_{00}}{T_2}\nabla T - R\frac{\gamma_{10}}{x_1x_2}\frac{\mu_1\mu_2}{[\mu_1+(\mu_2-\mu_1)c_1]^2}\nabla c_1 + \mathbf{J}_1\left(\frac{\partial \mathrm{H}}{\partial c_1}\right). \tag{4}$$

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Equations (3) and (4) can be considered as the thermodynamic generalization of Fick's and Fourier's laws, where the roles of the proper coefficients of diffusion and thermal conductivity are played by the quantities

$$D = \frac{R}{\rho}\,\frac{\gamma_{11}}{x_1 x_2}\,\frac{\mu_1^2\mu_2}{[\mu_1 + (\mu_2 - \mu_1)c_1]^2}, \tag{5}$$

$$\lambda = \gamma_{00}/T^2. \tag{6}$$

In addition, they contain cross coefficients of transfer, which are expressed by the dimensionless relation

$$K_t = \frac{\mu_1\gamma_{10}}{\rho D T}. \tag{7}$$

With allowance for (5)-(7), Eqs. (3) and (4) take the form

$$\mathbf{J}_1 = -\rho D\left(\nabla c_1 + \frac{K_t}{T}\nabla T\right), \tag{8}$$

$$\mathbf{q} = -\lambda\nabla T - \frac{\rho R}{\mu_1}\,\frac{DT}{x_1 x_2}K_t\,\frac{\mu_1\mu_2}{[\mu_1 + (\mu_2 - \mu_1)c_1]^2}\nabla c_1 + \mathbf{J}_1\left(\frac{\partial \mathrm{H}}{\partial c_1}\right). \tag{9}$$

Expressing x_1 and x_2 through c_1 and substituting (9), we have

$$\mathbf{q} = -\lambda\nabla T - \frac{\rho R}{\mu_1}\,\frac{DTK_t}{c_1(1 - c_1)}\nabla c_1 + \mathbf{J}_1\frac{\partial \mathrm{H}}{\partial c_1}. \tag{10}$$

For steady diffusion, the concentration and temperature fields in a binary mixture will be described by the system of equations

$$\operatorname{div}\left[-\rho D\left(\nabla c_1 + \frac{K_t}{T}\nabla T\right)\right] = 0, \tag{11}$$

$$\operatorname{div}\left[-\lambda\nabla T - \frac{\rho R}{\mu_1}\,\frac{DTK_t}{c_1(1 - c_1)}\nabla c_1 + \mathbf{J}_1\frac{\partial \mathrm{H}}{\partial c_1}\right] = 0, \tag{12}$$

$$\rho = \frac{\mu_1\mu_2}{(\mu_2 - \mu_1)c_1 + \mu_1}\,\frac{p}{RT} \tag{13}$$

or, for the case of one-dimensional diffusion,

$$\frac{d}{dx}\left[-\rho D\left(\frac{dc_1}{dx} + \frac{K_t}{T}\,\frac{dT}{dx}\right)\right] = 0, \tag{14}$$

$$\frac{d}{dx}\left[-\lambda\frac{dT}{dx} - \frac{\rho R}{\mu_1}\,\frac{DTK_t}{c_1(1 - c_1)}\,\frac{dc_1}{dx} + \mathbf{J}_1\frac{\partial \mathrm{H}}{\partial c_1}\right] = 0, \tag{15}$$

$$\rho = \frac{\mu_1\mu_2}{(\mu_2 - \mu_1)c_1 + \mu_1}\,\frac{p}{RT}. \tag{16}$$

Assuming that D, K_t, and λ are variables, i.e., $D = D(T)$, $K_t = K_t(c_1, T)$, and $\lambda = \lambda(c_1, T)$, we find

$$\begin{aligned}
&\frac{d^2c_1}{dx_2} + \frac{K_t}{T}\frac{d^2T}{dx^2} + \frac{1}{\rho}\frac{d\rho}{\partial c_1}\left(\frac{dc_1}{dx}\right)^2 + \left(\frac{K_t}{\rho T}\frac{\partial\rho}{\partial T} + \right.\\
&\left. + \frac{K_t}{DT}\frac{dD}{dT} + \frac{1}{T}\frac{\partial K_t}{\partial T} - \frac{K_t}{T^2}\right)\left(\frac{dT}{dx}\right)^2 + \left(\frac{1}{\rho}\frac{d\rho}{dT} + \right.\\
&\left. + \frac{1}{D}\frac{dD}{dT} + \frac{K_t}{\rho T}\frac{\partial\rho}{dc_1} + \frac{1}{T}\frac{\partial K_t}{\partial c_1}\right)\frac{dc_1}{dx}\frac{dT}{dx} = 0.
\end{aligned} \tag{17}$$

$$\begin{aligned}
&\frac{d^2c_1}{dx^2} + \frac{\lambda\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{d^2T}{dx^2} + \left[\frac{1}{\rho}\frac{\partial\rho}{\partial c_1} + \frac{1}{K_t}\frac{\partial K_t}{\partial c_1} + \right.\\
&\left. + \frac{2c_1-1}{c_1(1-c_1)} + \frac{\mu_1 c_1(1-c_1)}{RTK_t}\frac{\partial^2\mathrm{H}}{\partial c_1^2}\right]\left(\frac{dc_1}{dx}\right)^2 + \left[\frac{\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{\partial\lambda}{\partial T} + \right.\\
&\left. + \frac{\mu_1 c_1(1-c_1)}{RT^2}\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\right]\left(\frac{dT}{dx}\right)^2 + \left[\frac{\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{\partial\lambda}{\partial c_1} + \right.\\
&+ \frac{1}{\rho}\frac{\partial\rho}{\partial T} + \frac{1}{D}\frac{dD}{dT} + \frac{1}{T} + \frac{1}{K_t}\frac{\partial K_t}{\partial T} + \frac{\mu_1 c_1(1-c_1)}{RT^2}\frac{\partial^2\mathrm{H}}{\partial c_1^2} + \\
&\left. + \frac{\mu_1 c_1(1-c_1)}{RTK_t}\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\right]\frac{dc_1}{dx}\frac{dT}{dx} = 0.
\end{aligned} \tag{18}$$

The system of equations (17) and (18) is a mathematical model of the temperature and concentration fields formed in a binary mixture during combined heat and mass transfer in the case of a mixture of variable density, concentration, and temperature (p = const).

From the system of equations (17)-(18) we find $K_t(c_1, T)$ from the known functions $\lambda(c_1, T)$, $D(T)$, and $H(c_1, T)$ under certain assumptions about the parameters determining the state of a binary mixture.

1. In a binary mixture in which one-dimensional, steady, molecular heat and mass transfer occur, let p = const and ρ = const over the entire volume, which corresponds to the conditions of the Chapman–Enskog scheme – the mixture is in a state of mechanical equilibrium: $K_t = K_t(T, c_1)$ is the true value of the thermodiffusion ratio, while the system of equations (17)-(18) is written in the form

$$\begin{aligned}
&\frac{d^2c_1}{dx^2} + \frac{K_t}{T}\frac{d^2T}{dx^2} + \left(\frac{K_t}{DT}\frac{dD}{dT} + \frac{1}{T}\frac{\partial K_t}{\partial T} - \frac{K_t}{T^2}\right)\left(\frac{dT}{dx}\right)^2 + \\
&+ \frac{1}{D}\frac{dD}{dT}\frac{dc_1}{dx}\frac{dT}{dx} = 0,
\end{aligned} \tag{19}$$

$$\begin{aligned}
&\frac{d^2c_1}{dx^2} + \frac{\lambda\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{d^2T}{dx^2} + \left[\frac{1}{K_t}\frac{\partial K_t}{\partial c_1} + \frac{2c_1-1}{c_1(1-c_1)} + \right.\\
&\left. + \frac{\mu_1 c_1(1-c_1)}{RTK_t}\frac{\partial^2\mathrm{H}}{\partial c_1^2}\right]\left(\frac{dc_1}{dx}\right)^2 + \left[\frac{\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{\partial\lambda}{\partial T} + \right.\\
&\left. + \frac{\mu_1 c_1(1-c_1)}{RT^2}\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\right]\left(\frac{dT}{dx}\right)^2 + \left[\frac{\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{\partial\lambda}{\partial c_1} + \right.\\
&+ \frac{1}{D}\frac{dD}{dT} + \frac{1}{T} + \frac{1}{K_t}\frac{\partial K_t}{\partial T} + \frac{\mu_1 c_1(1-c_1)}{RT^2}\frac{\partial^2\mathrm{H}}{\partial c_1} + \\
&\left. + \frac{\mu_1 c_1(1-c_1)}{RTK_t}\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\right]\frac{dc_1}{dx}\frac{dT}{dx} = 0.
\end{aligned} \tag{20}$$

With allowance for the relations

$$\frac{dc_1}{dx}=\frac{dc_1}{dT}\,\frac{dT}{dx}\,;\quad \frac{d^2c_1}{dx^2}=\frac{d^2c_1}{dT^2}\left(\frac{dT}{dx}\right)^2+\frac{dc_1}{dT}\,\frac{d^2T}{dx^2}$$

Eqs. (19) and (20) take the form

$$\frac{d^2T}{dx^2}+\left[\frac{1}{D}\,\frac{dD}{dT}+\frac{\dfrac{d^2c_1}{dT^2}+\dfrac{1}{T}\,\dfrac{dK_t}{dT}-\dfrac{K_t}{T^2}}{\dfrac{dc_1}{dT}+\dfrac{K_t}{T}}\right]\left(\frac{dT}{dx}\right)^2=0, \tag{21}$$

$$\frac{d^2T}{dx^2}+\frac{f_1(T)+\varphi_1(T)}{\dfrac{dc_1}{dT}+\dfrac{\lambda\mu_1c_1(1-c_1)}{\rho RDTK_t}}\left(\frac{dT}{dx}\right)^2=0, \tag{22}$$

where

$$f_1(T)=\left[\frac{2c_1-1}{c_1(1-c_1)}\,\frac{dc_1}{dT}+\frac{1}{D}\,\frac{dD}{dT}+\frac{1}{T}+\frac{1}{K_t}\,\frac{dK_t}{dT}+\right.$$
$$\left.+\frac{\mu_1c_1(1-c_1)}{RTK_t}\,\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\right]\frac{dc_1}{dT}\,;$$
$$\varphi_1(T)=\frac{d^2c_1}{dT^2}+\frac{\mu_1c_1(1-c_1)}{\rho RDTK_t}\,\frac{d\lambda}{dT}+\frac{\mu_1c_1(1-c_1)}{RT^2}\,\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\,.$$

Equating the corresponding coefficients of Eqs. (21) and (22) to each other, after certain transformations we obtain the following differential equation for determing the thermodiffusion ratio:

$$\frac{dK_t}{dT}=\frac{A_1-a}{C_1-C_2'}K_t^2+\frac{(B_1-b)+(A_1C_2'-aC_1)}{C_1-C_2'}K_t+\frac{B_1C_2'-bC_1}{C_1-C_2'}\,, \tag{23}$$

where

$$a=\frac{1}{D}\,\frac{dD}{dT}-\frac{1}{T}\,;\quad b=\frac{T}{D}\,\frac{dD}{dT}\,\frac{dc_1}{dT}\,;\quad C_2'=T\frac{dc_1}{dT}\,,$$
$$B_1=\frac{\mu_1c_1(1-c_1)}{RT}\left[\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}+\frac{1}{\rho D}\,\frac{d\lambda}{dT}\right]\Big/\frac{dc_1}{dT}\,;$$
$$C_1=\frac{\lambda\mu_1c_1(1-c_1)}{\rho RDT}\Big/\frac{dc_1}{dT}\,,$$
$$A_1=\left[\frac{2c_1-1}{c_1(1-c_1)}\,\frac{dc_1}{dT}+\frac{1}{D}\,\frac{dD}{dT}+\frac{1}{T}\right]+\left[\frac{d^2c_1}{dT^2}+\right.$$
$$\left.+\frac{\mu_1c_1(1-c_1)}{RT^2}\,\frac{\partial^2\mathrm{H}}{\partial c_1\partial T}\right]\Big/\frac{dc_1}{dT}\,.$$

Equation (23) is the Riccati equation, a general solution of which cannot yet be obtained. Therefore, it can be solved by numerical methods on a computer. For ideal gas mixtures its solution has physical meaning in the temperature interval

$$\frac{\mu_1p}{\rho R}\leqslant T\leqslant\frac{\mu_2p}{\rho R} \tag{24}$$

for each fixed value of the density of the binary mixture, since it is assumed that ρ = const over the entire volume of the mixture. For a certain boundary condition, Eq. (23) uniquely determines the temperature dependence and, through it, the concentration dependence of the thermodiffusion ratio of binary gas mixtures in the approximation of the assumptions made.

The determination of the correct boundary condition for Eq. (23) on the basis of theoretical considerations presents serious difficulty. Two different boundary conditions for this equation can be set up. They follow from the condition that one of the components of the binary mixture disappears at the limits of the interval (24), and hence the thermodiffusion ratio vanishes. But these boundary conditions have the drawback that they do not correspond to internal points of the interval (24). In connection with this, difficulties of a mathematical character, connected with questions of the stability and convergence of the solution obtained, arise in the numerical solution of Eq. (23).

For convenience in the further analysis of Eq. (23), we rewrite it in the form

$$\frac{dK_t}{dT} = AK_t^2 + BK_t + C, \tag{25}$$

where

$$A = \frac{A_1 - a}{C_1 - C_2'}; \quad B = \frac{(B_1 - b) + (A_1 C_2' - aC_1)}{C_1 - C_2'};$$

$$C = \frac{B_1 C_2' - bC_1}{C_1 - C_2'}.$$

An analysis of the structure of Eq. (25), under the assumption that K_t is a smooth continuous function of the temperature, gives reason to assume that in the most general case K_t has a local maximum and minimum in the interval (24). It is determined from the condition

$$dK_t/dT = 0,$$

i.e.,

$$K_t = \frac{-B \pm \sqrt{B^2 - 4AC}}{2A}. \tag{26}$$

Hence, within the interval there may be one value of the temperature at which the thermodiffusion ratio is reduced to zero. In the transition through this temperature, the thermodiffusion ratio changes sign. This temperature is called the inversion temperature [4].

Those binary mixtures which do not have inversion temperatures must, in accordance with Eq. (26), have either a local maximum or a local minimum within the interval (24), which corresponds to the temperature determined from the equation

$$B^2 - 4AC = 0. \tag{27}$$

Then the value of the extremum is determined from the formula

$$K_t = -\frac{B}{2A}. \tag{28}$$

When the condition (27) is satisfied for a given binary mixture, as the boundary condition for Eq. (25) one can take

$$K_t = -\frac{B}{2A}; \quad T = T_0,$$

where T_0 is a root of Eq. (27).

Although a particular solution of Eq. (25) is obtained in the form of the temperature dependence of K_t, it does not express the temperature dependence of K_t referred to some definite concentration. In it the concentration of the mixture varies simultaneously with variation of the temperature, in accordance with Eq. (16). But by varying the density of the mixture, one can, in principle, obtain a set of solutions of Eq. (25) enabling one to determine only the temperature dependence or only the concentration dependence of K_t referred to some definite concentration or temperature.

We note that the results of a numerical solution of Eq. (25) by the Runge–Kutta method as applied to an N_2–CO_2 binary gas mixture at a total pressure of $1.013 \cdot 10^5$ Pa and a density $\rho = 1.1372$ kg/m³ in the temperature range of $300 \leq T \leq 471.42$°K yield "reasonable" values for K_t.

2. Now let us consider the case of combined heat and mass transfer in a saturated binary vapor–gas mixture. In such a mixture the pressure of the saturated vapor is a unique function of the temperature. At a constant pressure, the density and concentration of the mixture are functions only of the temperature. With allowance for this, the system of equations (17)-(18) is written in the form

$$\frac{d^2T}{dx^2} + \left[\frac{1}{\rho}\frac{d\rho}{dT} + \frac{1}{D}\frac{dD}{dT} + \frac{\dfrac{d^2c_1}{dT^2} + \dfrac{1}{T}\dfrac{dK_t}{dT} - \dfrac{K_t}{T^2}}{\dfrac{dc_1}{dT} + \dfrac{K_t}{T}}\right]\left(\frac{dT}{dx}\right)^2 = 0, \tag{29}$$

$$\frac{d^2T}{dx^2} + \frac{f_2(T) + \varphi_2(T)}{\dfrac{dc_1}{dT} + \dfrac{\lambda\mu_1 c_1(1-c_1)}{\rho RDTK_t}}\left(\frac{dT}{dx}\right)^2 = 0, \tag{30}$$

where

$$\varphi_2(T) = \frac{d^2c_1}{dT^2} + \frac{\mu_1 c_1(1-c_1)}{\rho RDTK_t}\frac{d\lambda}{dT} + \frac{\mu_1 c_1(1-c_1)}{RT^2}\frac{\partial^2 \mathrm{H}}{\partial c_1 \partial T};$$

$$f_2(T) = \left[\frac{2c_1 - 1}{c_1(1-c_1)}\frac{dc_1}{dT} + \frac{1}{\rho}\frac{d\rho}{dT} + \frac{1}{D}\frac{dD}{dT} + \frac{1}{T} + \right.$$
$$\left. + \frac{1}{K_t}\frac{dK_t}{dT} + \frac{\mu_1 c_1(1-c_1)}{RTK_t}\frac{\partial^2 \mathrm{H}}{\partial c_1 \partial T}\right]\frac{dc_1}{dT}.$$

Equating the coefficients of (29) and (30), we obtain a differential equation for determining the thermodiffusion ratio for a saturated vapor–gas mixture,

$$\frac{dK_t}{dT} = \frac{A_1' - a'}{C_1' - C_2'}K_t^2 + \frac{(B_1' - b') + (A_1'C_2' - a'C_1')}{C_1' - C_2'}K_t + \frac{B_1'C_2' - b'C_1'}{C_1' - C_2'}, \tag{31}$$

where

$$a' = \frac{1}{\rho}\frac{d\rho}{dT} + \frac{1}{D}\frac{dD}{dT} - \frac{1}{T}; \quad C_2' = T\frac{dc_1}{dT};$$

$$b' = \left(\frac{T}{\rho}\frac{d\rho}{dT} + \frac{T}{D}\frac{dD}{dT}\right)\frac{dc_1}{dT};$$

$$A_1' = \left[\frac{2c_1 - 1}{c_1(1-c_1)}\frac{dc_1}{dT} + \frac{1}{\rho}\frac{d\rho}{dT} + \frac{1}{D}\frac{dD}{dT} - \frac{1}{T}\right] +$$
$$+ \left[\frac{d^2c_1}{dT^2} + \frac{\mu_1 c_1(1-c_1)}{RT^2}\frac{\partial^2 \mathrm{H}}{\partial c_1 \partial T}\right] \Big/ \frac{dc_1}{dT};$$

$$B_1' = \frac{\mu_1 c_1(1-c_1)}{RT}\left[\frac{\partial^2 \mathrm{H}}{\partial c_1 \partial T} + \frac{1}{\rho D}\frac{d\lambda}{dT}\right] \Big/ \frac{dc_1}{dT};$$

$$C_1' = \frac{\lambda\mu_1 c_1(1-c_1)}{\rho RDT} \Big/ \frac{dc_1}{dT}.$$

Equation (31) as applied to a saturated vapor—air mixture was made concrete in [5], where the results of a numerical solution in the temperature range of $273 \leqslant T \leqslant 373°K$ are presented.

Further research on the determination of the thermodiffusion ratios of various binary gas and vapor—gas mixtures is required for the fullest determination of the possibilities and the reliability of the above analytic method of determining the thermodiffusion ratios of binary gas mixtures and saturated vapor—gas mixtures on the basis of the differential equations derived.

We note that the idea of the above method can be used, in principle, to determine the thermodiffusion ratios in liquids and solids.

NOTATION

γ_{00}, γ_{10}, γ_{11}, kinetic coefficients; x_1, x_2, molar fractions of the components of the mixture; ∇, gradient symbol; J_1, vector of flux density of a substance; μ_1, μ_2, molecular masses of the components of the mixture; R, universal gas constant; T, local temperature of the mixture; H_1, H_2, specific enthalpies of the components of the mixture; ρ, density of the mixture; ρ_1, density of the light component of the mixture; c_i, mass concentration of the light component of the mixture; D, diffusion coefficient; λ, coefficient of thermal conductivity of the mixture; K_t, thermodiffusion ratio of the mixture; p, pressure of the mixture; n, power of the temperature dependence of the diffusion coefficient of the binary mixture under consideration; p_0, pressure of the mixture under standard conditions; D_0, diffusion coefficient under standard conditions; α_t, thermodiffusion constant of the mixture; q, vector of heat-flux density; H, specific thermodynamic function of the mixture.

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