NON-STEADY-STATE THERMAL DIFFUSION AND THERMAL CONDUCTIVITY IN GAS MIXTURES

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Expressions are obtained which characterize effective thermal conductivity coefficients of gas mixtures, as measured by various experimental methods.

Recently, non-steady-state methods have begun to be employed to measure the thernal conductivity of gas mixtures. Examples are the shock tube method [1-3] and the non-steady-state heated filament method [4-7]. Comparison of experimental data on thermal conductivity of gas mixtures shows that experimental data obtained by steady-state methods for one and the same mixtures under identical conditions do not agree with data obtained by non-steady-state methods. Thus, it is of definite interest in the steady and nonsteady states with reference to the conditions occurring in experimental determination of thermal conductivity.

The present study will be limited to consideration of some results of an experimental determination of thermal conductivity of helium—argon mixtures, measured by the steady-state heated filament method, the non-steady-state heated filament method, and the non-steady-state shock tube method.

We will first consider the physical aspects on which these methods of thermal conductivity measurements are based.

The state of the gas mixture is characterized by three thermodynamic quantities: concentration and any two of three other quantities: temperature, pressure, and density. Accordingly, we can dinstinguish two cases of gas mixture states.

1. The specific volume of the system is constant, while concentration, temperature, and pressure vary. These conditions correspond to measurement of the thermal conductivity coefficient by the non-steady-state heated filament method.

2. The specific volume, concentration, and temperature of the system vary, while pressure remains constant. This corresponds to measurement by the steady-state heated filament or non-steady-state shock tube methods.

We note that [8] evaluated the question of describing the heat-transfer process in a gas the state of which is defined by density and temperature.

We will describe the processes of mass and energy transfer in gas mixtures for the two cases described above. The system of equations for mass and energy fluxes has the form

$$\begin{gathered}\vec{J}_1 = L_{11}(\vec{X}_1 - \vec{X}_2) + L_{1u}\vec{X}_u,\\ \vec{J}_u = L_{u1}(\vec{X}_1 - \vec{X}_2) + L_{uu}\vec{X}_u.\end{gathered} \tag{1}$$

Equations for the thermodynamic forces, obtained with the relationship $Tds = du + pd\bar{v} - \sum_{k=1}^{2} \bar{\mu}_k dc_k,$ in the first case (ρ = const) have the form

$$\begin{gathered}\vec{X}_u = -\nabla T/T^2,\\ \vec{X}_1 - \vec{X}_2 = -\frac{1}{T}\left(\frac{\partial \bar{v}}{\partial c_1}\nabla p - \frac{\partial h}{\partial c_1}\nabla \ln T + c_2^{-1}\frac{\partial \bar{\mu}_1}{\partial c_1}\nabla c_1\right),\end{gathered} \tag{2}$$

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and in the second (p = const)

$$\vec{X}_u = -\frac{5}{3}\,\frac{\nabla T}{T^2},$$
$$\vec{X}_1 - \vec{X}_2 = -\frac{1}{T}\left[\left(p\,\frac{\partial \bar{v}}{\partial c_1} - \frac{\partial h}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1\right]. \tag{3}$$

System (1) with thermodynamic forces written in the form of Eq. (2) has the form (here we use the condition $\nabla \ln p = \nabla \ln T$, $L_{1u} = U_1^* L_{11}$)

$$\vec{J}_1 = -\frac{L_{11}}{T}\left[\left(U_1^* - \frac{\partial h}{\partial c_1} + p\,\frac{\partial \bar{v}}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1 = -\frac{L_{11}}{T}\left[\left(\theta_1^* + p\,\frac{\partial \bar{v}}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1\right],\right.$$
$$\vec{J}_u = -\frac{L_{11}}{T}\,U_1^*\left[\left(p\,\frac{\partial \bar{v}}{\partial c_1} - \frac{\partial h}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1\right] - L_{uu}\,\frac{\nabla T}{T^2}. \tag{4}$$

With thermodynamic forces in the form of Eq. (3), Eq. (1) has the form

$$\vec{J}_1 = -\frac{L_{11}}{T}\left[\left(\frac{5}{3}\,U_1^* - \frac{\partial h}{\partial c_1} + p\,\frac{\partial \bar{v}}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1\right] = -\frac{L_{11}}{T}\left[\left(\theta_1^* + p\,\frac{\partial \bar{v}}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1\right],$$
$$\vec{J}_u = -\frac{L_{11}}{T}\,U_1^*\left[\left(p\,\frac{\partial \bar{v}}{\partial c_1} - \frac{\partial h}{\partial c_1}\right)\frac{\nabla T}{T} + c_2^{-1}\,\frac{\partial \bar{\mu}_1}{\partial c_1}\,\nabla c_1\right] - \frac{5}{3}\,L_{uu}\,\frac{\nabla T}{T^2}. \tag{5}$$

We will introduce the notation

$$\frac{L_{11}}{T}\,c_2^{-1}\left(\frac{\partial \bar{\mu}_1}{\partial c_1}\right)_{p,T} = \rho D_{12}^{\text{п}}, \quad \frac{L_{11}}{T}\left(\theta_1^* + p\,\frac{\partial \bar{v}}{\partial c_1}\right) = \rho D_{1\text{ef}}^{T}\,c_1(1-c_1). \tag{6}$$

Here $D_{12}^{\text{п}}$ is the mutual diffusion coefficient stemming from pressure difference (in case of Eq. (4)), or density difference (in case of Eq. (5)). With consideration of Eq. (6), Eq. (4) takes on the form

$$\vec{J}_1 = -\rho D_{12}^{\text{п}}\left(\nabla c_1 + k_{T_{\text{ef}}}\,\frac{\nabla T}{T}\right),$$
$$\vec{J}_u = -\left[\lambda_{\text{tr}} + \frac{pD_{12}}{T}\,\alpha_T^{U_1^*}\left(\frac{p\partial \bar{v}/\partial c_1 - \partial h/\partial c_1}{c_2^{-1}\,\partial \bar{\mu}_1/\partial c_1} - k_{T_{\text{ef}}}\right)\right]\nabla T = -\lambda_{\text{ef}}^{\text{п}}\,\nabla T. \tag{7}$$

$$k_{T_{\text{ef}}} = \frac{D_{1\text{ef}}^{T}\,c_1(1-c_1)}{D_{12}}; \quad \lambda_{\text{tr}} = \frac{L_{uu}}{T^2}, \quad \alpha_T^{U_1^*} = \frac{U_1^*}{c_1\partial \bar{\mu}_1/\partial c_1};$$

$$D_{12} = D_{12}^{\text{п}}\,\frac{m_1 m_2}{(m_1 c_2 + m_2 c_1)^2}.$$

If the state is stationary ($\nabla p = 0$), then the thermodiffusion ratio is defined as

$$k_{T_{\text{tr}}} = \theta_1^*/c_2^{-1}\,\partial \bar{\mu}_1/\partial c_1.$$

If the state is non-steady ($\nabla p \neq 0$), then the effective value of the thermodiffusion ratio is

$$k_{T_{\text{ef}}} = \frac{\theta_1^* + p\partial \bar{v}/\partial c_1}{c_2^{-1}\,\partial \bar{\mu}_1/\partial c_1}.$$

The expression for the effective thermal conductivity coefficient, measured by the non-steady-state heated filament method, is characterized by the relationship

$$\lambda_{\text{ef}}^{\text{п}} = \lambda_{\text{tr}} + \frac{pD_{12}}{T}\,\alpha_T^{U_1^*}\,\tilde{k}_T, \tag{8}$$

where

$$\tilde{k}_T = k_{T_{\text{ef}}} + \frac{p\partial \bar{v}/\partial c_1 - \partial h/\partial c_1}{c_2^{-1}\,\partial \bar{\mu}_1/\partial c_1}.$$

We note that the sign of $k_{t\,\mathrm{ef}}$ is positive, since the thermodiffusion ratio of the mixture the thermal conductivity of which is measured by the non-steady-state heated filament method is always negative.

With consideration of Eq. (6), Eq. (5) takes on the form

$$\vec{J}_1 = -\rho D''_{12}\left(\nabla c_1 + k_{\mathrm{T_{ef}}}\frac{\nabla T}{T}\right),$$

$$\vec{J}_u = -\left[\frac{5}{3}\lambda_{\mathrm{tr}} + \frac{pD_{12}}{T}\alpha_{\mathrm{T}}^{U_1^*}\left(\frac{p\partial\bar{v}/\partial c_1 - \partial h/\partial c_1}{c_2^{-1}\partial\bar{\mu}_1/\partial c_1} - k_{\mathrm{T_{ef}}}\right)\right]\nabla T = -\lambda_{\mathrm{ef}}\nabla T. \tag{9}$$

Since the thermodiffusion ratio of a mixture the thermal conductivity of which is measured, for example, by the steady-state heated filament (non-steady-state shock tube) method, is always positive, while the effective value of the thermodiffusion ratio of a mixture, the specific volume of which varies, is defined by the expression

$$k_{\mathrm{T_{ef}}} = \frac{\frac{5}{3}U_1^* - \partial h/\partial c_1 + p\partial\bar{v}/\partial c_1}{c_2^{-1}\partial\bar{\mu}_1/\partial c_1} = \frac{\theta_1^* + p\partial\bar{v}/\partial c_1}{c_2^{-1}\partial\bar{\mu}_1/\partial c_1},$$

then the expression for the thermal conductivity coefficient of the mixture in the steady state has the form

$$\lambda_{\mathrm{ef}\,\infty} = \frac{5}{3}\left(\lambda_{\mathrm{tr}} - \frac{pD_{12}}{T}\alpha_{\mathrm{T}}^{U_1^*}k_{\mathrm{T}}^{U_1^*}\right),$$

while the thermal conductivity coefficient at the initial moment of time

$$\lambda_0 = \frac{5}{3}\lambda_{\mathrm{tr}} + \frac{pD_{12}}{T}\alpha_{\mathrm{T}}^{U_1^*}\overset{\circ}{k}_{\mathrm{T}}; \quad \overset{\circ}{k}_{\mathrm{T}} = \frac{p\partial\bar{v}/\partial c_1 - \partial h/\partial c_1}{c_2^{-1}\partial\bar{\mu}_1/\partial c_1},$$

whence

$$\lambda_0 - \lambda_{\mathrm{ef}\,\infty} = \frac{pD_{12}}{T}k_{\mathrm{T}}^{U_1^*}\alpha_{\mathrm{T_{ef}}}.$$

If the mixture as a whole is immobile, then $\lambda_0 = \lambda_{\mathrm{tr}}$ and

$$\lambda_{\mathrm{ef}\,\infty} = \lambda_{\mathrm{tr}} - \frac{pD_{12}}{T}k_{\mathrm{T}}^{U_1^*}\alpha_{\mathrm{T_{ef}}}. \tag{10}$$

We will demonstrate that the effective thermal conductivity coefficient of a helium–argon mixture as measured by the non-steady-state heated filament method is not equal to the effective thermal conductivity coefficient as measured by the steady-state heated filament method, making use of Eqs. (8) and (10) to show that $\lambda_{\mathrm{ef}}^{\mathrm{n}} \neq \lambda_{\mathrm{ef}\infty}$. In fact,

$$\lambda_{\mathrm{ef}}^{\mathrm{n}} - \lambda_{\mathrm{ef}\,\infty} = \frac{pD_{12}}{T}\alpha_{\mathrm{T}}^{U_1^*}(k_{\mathrm{T_{ef}}} + \tilde{k}_{\mathrm{T}}) > 0, \tag{11}$$

since for a helium–argon mixture $k_{\mathrm{T_{ef}}} + \tilde{k}_{\mathrm{T}} > 0$, while $\alpha_{\mathrm{T}}^{U_1^*}$ is always greater than zero.

We will illustrate the validity of Eq. (11) by comparing experimental data on thermal conductivity presented in [5] ($\lambda_{\mathrm{ef}}^{\mathrm{n}}$, nonstationary heated filament method) and [9] ($\lambda_{\mathrm{ef}\infty}$, stationary heated filament method) (Table 1).

As is evident from the table, the divergence between the results can exceed 7%, which cannot be connected to experimental error in the data, since the uncertainty in [5] was no greater than 0.2%, while that of [9] was ±1%. However, it should be noted here that the data of both [5] and [9] were presented graphically, which can introduce a certain error into the values presented in Table 1.

We will demonstrate the validity of Eqs. (8) and (10) and explain the cause of the poor agreement between experimental and theoretical data on the thermal conductivity of inert gas mixtures. By theoretical data we mean data obtained with the above approximations of formal molecular-kinetic theory, based on the Chapman–Enskog method, which can be identified with true values of the thermal conductivity coefficient.

TABLE 1. Comparison of Effective Thermal Conductivity Coefficients of Helium–Argon Mixture at T = 300°K, as Measured by Non-Steady State (λ^n) and Steady-State (λ_∞) Methods ($\lambda \cdot 10^3$ W/m·deg K)

x_2	λ^n	λ_∞	$\frac{\lambda^n - \lambda_\infty}{\lambda^n} \cdot 100\%$
0	154,6	151,0	2,1
0,23	95,4	87,9	7,9
0,35	75,8	70,0	7,7
0,64	42,04	39,85	5,2
0,81	29,3	27,9	4,8
1,0	17,8	17,7	0,56

According to Eq. (8), the experimental data on thermal conductivity measured by the non-steady-state heated filament method will be higher than the theoretical values. This conclusion agrees with the comparison of theoretical and experimental data on thermal conductivity of helium–argon and neon–argon mixtures presented in [5].

According to Eq. (10), experimental values of thermal conductivity measured by the steady-state heated filament method will be lower than the theoretical values. This is confirmed by a comparison of experimental and theoretical results on thermal conductivity of mixtures of heavy inert gases at low temperatures (T < 300°K), and is a result of nonisothermal mutual diffusion caused by density differences. At high temperatures this effect is insignificant and $\lambda_{ef_\infty} \approx \lambda_{tr}$.

On the basis of the above, the question arises of which thermal conductivity data are close to the true values — those obtained by the non-steady-state heated filament method (λ^n_{ef}), or those of the steady-state heated filament method ($\lambda_{ef\infty}$). In the author's opinion, in the temperature range T > 300°K, the steady-state method is preferable, while at T < 300°K the non-steady-state method is to be preferred.

However, no matter what method is used to measure effective mixture thermal conductivity, it is necessary to correct the experimental data for the corresponding effects, and to use only true values in comparing experimental data with each other and theoretical values.

We will demonstrate that the effective thermal conductivity measured by the steady-state heated filament method is not equal to the effective coefficient measured by the non-steady-state shock tube method.*

We will first note that below, by $\vec{J}_u$ we mean the reduced energy flux, i.e.,

$$\vec{J}_u = \vec{\tilde{J}}_u - \frac{\partial h}{\partial c_1} \vec{J}_1,$$

where $\vec{\tilde{J}}_u$ is the total energy flux. The concept of reduced energy flux is used to describe the process of energy transfer in mixtures when the mixture as a whole is immobile (thermal conductivity is measured by the steady-state heated filament method). If the mixture as a whole moves (non-steady-state heated filament method), then to describe the process of energy transfer we use the concept of total energy flux. In this case

$$\vec{\tilde{J}}_u = \vec{J}_u + \frac{\partial h}{\partial c_1} \vec{J}_1 = \left(U_1^* + \frac{\partial h}{\partial c_1}\right) \vec{J}_1 - \lambda_{ef\,\infty} \nabla T = -\lambda_{ef}^n \nabla T, \qquad (12)$$

whence $\lambda_{ef}^n - \lambda_{ef\,\infty} = -\frac{5}{3} \frac{pD_{12}}{T} \tilde{\alpha}_T k_T^{U_1^*}$; $\lambda_{ef\,\infty}$ is the effective thermal conductivity coefficient measured by the non-steady-state heated filament method;

$$\tilde{\alpha}_T = \frac{U_1^* + \partial h/\partial c_1}{c_1 \partial\bar{\mu}_1/\partial c_1} > 0, \quad k_T^{v_1^*} > 0.$$

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*The divergence between thermal conductivity data for gases obtained by steady-state heated filament and non-steady-state shock tube methods was first noted by N. B. Vargaftik.

According to Eq. (12), $\lambda^n_{ef} < \lambda_{ef\infty}$, which is confirmed by comparison of the results of [10, 11] on thermal conductivity of helium-argon mixture thermal conductivity (measurements at temperatures up to 1500°K were performed by the steady-state heated filament method [10], and in the 1500-6000°K range by the shock tube method [11]). A comparison of the results of [10, 11] is presented in [12].

In conclusion, we will discuss the causes of poor agreement between theoretical and experimental data on the thermodiffusion constant of binary gas mixtures. By theoretical data, we understand, as before, the above approximations of formal molecular-kinetic theory. The greatest quantity of experimental data on thermodiffusion constants of gas mixtures has been obtained by the two-reservoir method [13, 14]. In this method the thermodiffusion constant is measured in the following manner: the experimental chamber is evacuated, placed in a steady-state thermal regime, and filled with mixture to the required pressure, i.e., the temperature, concentration, and density change, while the pressure remains constant. According to Eq. (9), the thermodiffusion constant

$$\alpha_{T_{ef}} = \frac{\theta_1^* + p\partial\bar{v}/\partial c_1}{c_1\partial\bar{\mu}_1/\partial c_1} = \alpha_{T_{tr}} + \alpha_T^\rho,$$

where $\alpha_{T_{tr}} = \theta_I^*/c_1\partial\bar{\mu}_1/\partial c_1$; $\alpha_T^\rho = p\partial\bar{v}/\partial c_1/c_1\partial\bar{\mu}_1/\partial c_1$. Here $\alpha_{T_{tr}}$ characterizes the true value of the thermodiffusion constant; α_T^ρ is the effect of change in mixture specific volume on nonisothermal mass transfer.

Thus, in our opinion, the cause of poor agreement between theoretical and experimental data on thermodiffusion constants is the fact that aside from the true value, the experimental data contain an additional contribution produced by the effect of change in specific volume on mass transfer.

NOTATION

λ^n_{ef}, $\lambda_{ef\infty}$, effective thermal conductivities as measured by non-steady-state and steady-state methods; λ_0, thermal conductivity of uniformly mixed mixture; p, pressure; ρ, density; T, temperature, x_i, c_i, molar and mass fractions of component i; $\vec{J}_i$, mass flux of component i; k_T, thermodiffusion ratio; α_T, thermodiffusion constant; D_{12}, D^T_1, mutual diffusion and thermodiffusion coefficients; m, mass; s, u, $\bar{v}$, h, specific entropy, energy, volume, and entropy; θ^*, heat of transfer; U^*, energy of transfer; $\vec{X}_u$, $\vec{X}_i$, thermodynamic forces; $\bar{\mu}$, chemical potential.

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AN EXPERIMENTAL STUDY OF THE FILTERING CAPACITY OF A THERMAL DIFFUSION CHAMBER CONTAINING A CONTROL ELEMENT

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Experimental results are presented on the deposition of a finely divided aerosol in a planar thermal-diffusion flow chamber containing a control element [1].

Diffusiophoresis has been used [2-7] to remove aerosols from gases (directional motion of the aerosol particles in a concentration gradient). The particle velocity is directly proportional to the concentration gradient in the diffusing component [8, 9], so thermal-diffusion chambers (TD chambers) are used to produce high stationary gradients [1, 3, 6, 7]. A major disadvantage of such a filtering device is that there is a positive supersaturation in the working space [10], which leads to vapor flow to the aerosol particles (by heterogeneous condensation), which tends to reduce the concentration gradient and thus reduces the filtration performance. This is very important if the particle concentration is more than 10^{10}-10^{11} m^{-3}, but it can be eliminated in a TD chamber with a control element, whose operation has been described in detail in [11]. The control element takes the form of a heated grid permeable to the vapor molecules placed between continuously wetted surfaces differing in temperature, one of which produces the vapor and the other absorbs it. If the temperature of the grid is equal to the temperature of the heater (evaporator), the state of the vapor—gas mixture will be that of negative supersaturation in the region of the control element, which is called the working zone, but with a vapor concentration gradient determined as in an ordinary TD chamber by the temperatures of the cooler and heater and by the distance between them.

A quantitative measure of the filtration performance is provided by the breakthrough coefficient K_b, which is equal to the ratio of the aerosol concentrations at the outlet and inlet. The performance is commonly measured by a method based on nephelometry of a monodisperse aerosol before and after the filter [12]. If the aerosol is finely divided, some enlargement of the particles is produced before nephelometry in the supersaturated vapor of a high-boiling substance in a device of KUST type [13]. This method cannot be used to measure the breakthrough coefficient for a TD chamber because of the enlargement of the particles by condensation at the outlet from the TD chamber due to cooling of the air flow with its high vapor content in the pipelines of the nephelometer and KUST.

During preliminary studies, it was shown that the aerosol may be deposited completely in the TD chamber.

Zero value of K_b (the absence of particles at the exit from the TD chamber is readily detected) is attained at a certain value of the volume flow rate Q* through the TD working zone, which is subsequently called the critical value. The value of Q* will be dependent on the working and geometrical parameters of the TD chamber, and also on the physical characteristics of the aerosol and vapor—gas medium.

We have measured Q* ($K_b = 0$) for various conditions in the TD chamber and various parameters for a finely divided sodium chloride aerosol (Knudsen number $Kn \gg 1$) with the

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