

## SOLUTION OF CERTAIN PROBLEMS IN THE THERMAL DIFFUSION OF BINARY GASES

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*Two variants of the experimental method of unique determination of the thermal-diffusion ratio ( $K_t$ ) of binary gases are described. A formula expressing the temperature and concentration dependence of  $K_t$  and obtained by a thermodynamic method is proposed.*

More than 80 years have passed since Chapman and Enskog discovered (1911–1917) the thermal-diffusion effect in gas mixtures. Nonetheless, some problems of the thermal diffusion of binary mixtures remain to be solved, in particular:

- (a) unique determination of the thermal-diffusion constant of binary gases by an experimental method;
- (b) description of the temperature and concentration dependence of the thermal-diffusion constant ( $\alpha_t$ ) by a thermodynamic method.

Below we present the basic results of our work, which are satisfactory solutions of the problems mentioned above.

Traditional experimental methods of determination of  $K_t$  (or  $\alpha_t$ ) rely mainly on the following theoretical prerequisites [1]:

$$\bar{K}_t = \frac{x_{10} - x_{1h}}{\ln(T_0/T_h)}; \quad (1)$$

$$\bar{\alpha}_t = \frac{\ln q}{\ln(T_0/T_h)}, \quad q = \frac{x_{10}(1 - x_{1h})}{x_{1h}(1 - x_{10})}. \quad (2)$$

In (1) and (2),  $\bar{K}_t = K_t(\bar{x}_1, \bar{T})$  and  $\bar{\alpha}_t = \alpha_t(\bar{x}_1, \bar{T})$ , where  $\bar{x}_1$  and  $\bar{T}$  are certain undetermined values of  $x_1$  and  $T$  from the intervals

$$T_h \leq T \leq T_0 \quad \text{and} \quad x_{1h} \leq x_1 \leq x_{10}, \quad (3)$$

therefore, within the framework of the theoretical prerequisites (1) and (2) it is, in principle, impossible to uniquely determine  $\bar{K}_t$  or  $\bar{\alpha}_t$ . Since  $K_t$  and  $\alpha_t$  are interrelated by the relation

$$K_t = x_1(1 - x_1)\alpha_t, \quad (4)$$

in what follows we will use formula (1) for the sake of simplicity.

We propose two variants of the experimental method of unique determination of  $K_t$ .

*First Variant.* As the theoretical basis one employs the differential equation

$$K_t = T \frac{dx_1}{dT} \quad (5)$$

with the boundary conditions

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$$T = T_0, \quad x_1 = x_{10} \quad \text{for } z = 0, \quad T = T_h, \quad x_1 = x_{1h} \quad \text{for } z = h. \quad (6)$$

Based on the designed experiments on thermal-diffusion separation in stationary thermal diffusion, one constructs the interpolation formula (from the data of experimental measurements)

$$\Delta x_1 = f(\Delta T) \quad \text{or} \quad x_1 = x_{1h} + f(T - T_h) \quad (7)$$

with a relative error no higher than the relative experimental error  $\Delta x_1$ . Having determined  $dx_1/dT$  from (7) and having substituted it into (5), we find

$$K_t = Tf'(T - T_h). \quad (8)$$

Equations (7) and (8) uniquely determine  $K_t$ .

*Second Variant.* It involves analytical determination of the gradients  $dx_1/dz$  and  $dT/dz$  by modeling mathematically the combined diffusive heat and mass transfer in the case of stationary thermal diffusion in a two-cylinder thermal-diffusion apparatus on the basis of the linear phenomenological equations of thermal dynamics. As a result, in a medium-pulse frame of reference we obtain the formula [2]

$$K_t = \frac{\mu p}{R} \frac{\lambda T}{\rho D} \left( x_1 \sqrt{\mu_1} + x_2 \sqrt{\mu_2} \right) \frac{\int_{x_{10}}^{x_{1h}} \frac{dx_1}{(x_1 \sqrt{\mu_1} + x_2 \sqrt{\mu_2}) \varphi(x_1)}}{\int_{T_h}^{T_0} \frac{Tf(T)}{D} dT}, \quad (9)$$

which, however, determines  $K_t$  not for any pair of values  $\bar{x}_1$  and  $\bar{T}$  from the intervals (3) but only for the pairs that satisfy the integral relation

$$\int_{x_{10}}^{\bar{x}_1} \frac{dx_1}{(x_1 \sqrt{\mu_1} + x_2 \sqrt{\mu_2}) \varphi(x_1)} = \frac{\int_{x_{10}}^{x_{1h}} \frac{dx_1}{(x_1 \sqrt{\mu_1} + x_2 \sqrt{\mu_2}) \varphi(x_1)}}{\int_{T_0}^{T_h} \frac{Tf(T)}{D} dT} \int_{T_0}^{\bar{T}} \frac{Tf(T)}{D} dT. \quad (10)$$

It must be noted that the degree of adequacy of formula (9) is attributed to the adequacy of the linear thermal-dynamics equations.

Following the first variant and employing the thermal-diffusion separations of an He–Ar mixture measured in experiments [1, 3], we have constructed the interpolation formula

$$\alpha_t = \frac{x_{1h}(1 - x_{1h})}{x_1(1 - x_1)} \left\{ H + \frac{\varphi}{[(T - 293) + \varphi]^2} \left[ \frac{1.0234}{1.8281 - x_{1h}} - 0.2425 \right] \right\} T, \quad (11)$$

where

$$\varphi = 608.33x_{1h}^3 - 1150.17x_{1h}^2 + 643.07x_{1h} + 394.18;$$

$$H = 10^{-4} (-50.07x_{1h}^5 + 122.88x_{1h}^4 - 116.14x_{1h}^3 + 46.58x_{1h}^2 - 6.8x_{1h} + 2.72);$$

$$x_1 = x_{1h} + x_{1h}(1 - x_{1h}) \left\{ H + \frac{1}{T - 293 + \varphi} \left[ \frac{1.0234}{1.8281 - x_{1h}} - 0.2425 \right] \right\} (T - 293). \quad (12)$$

Formulas (11) and (12) determine  $\alpha_t$  with an average relative error of  $\pm 2\%$  in the intervals

$$100 \leq T \leq 1000 \text{ K} \quad \text{and} \quad 0.1 \leq x_1 \leq 0.9 \quad (13)$$

when the maximum is smaller than 5%. Thus, the error of formulas (11) and (12) lies within the experimental error [1, 3].

An advantage of formulas (11) and (12) is that in using them one can calculate the separation in the process of stationary thermal diffusion for any values of  $T_0$ ,  $T_h$ , and  $x_{1h}$  for He–Ar.

With the use of formulas (11) and (12) we have developed a draft table of recommended reference data which contains about nine thousand values of  $\alpha_t$  corresponding to the intervals

$$100 \leq T \leq 1000 \text{ K} \quad \text{and} \quad 0.01 \leq x_1 \leq 0.98 \quad (14)$$

with the steps

$$\Delta x = 0.01 \quad \text{and} \quad \Delta T = 10 \text{ K}.$$

The draft table is currently undergoing an examination by experts at the State Standard of Turkmenistan for recommended reference data.

Thus, the first variant of the proposed experimental method of unique determination of  $K_t$  is quite satisfactory and opens up the way for the development of standard reference data on  $\alpha_t$  for binary gases. Furthermore, by mathematical modeling of the combined diffusive heat and mass transfer in binary gas mixtures under isothermal and iso-concentration conditions, we have obtained the formula

$$K_t = \frac{\bar{\mu} \left( \bar{x}_1 \sqrt{\mu_1} + \bar{x}_2 \sqrt{\mu_2} \right)^2 x_1 x_2 \bar{\lambda} T}{\mu_1 \sqrt{\mu_2} [H(T) - C(\bar{x}_1)] \left( x_1 \sqrt{\mu_1} + x_2 \sqrt{\mu_2} \right) \bar{x}_1 \bar{x}_2 \bar{\rho} D}, \quad (15)$$

which expresses the temperature and concentration dependences of  $K_t$  accurate to the undetermined constant  $C(\bar{x}_1)$ . Logically,  $K_t$  is invariant relative to  $\bar{x}_1$ . Formula (15) is correct within the framework of the validity of the linear phenomenological equations of nonequilibrium thermal dynamics. Thus, within the framework of linear thermal dynamics the problem of the temperature and concentration dependences of  $K_t$  (or  $\alpha_t$ ) is solved by the thermodynamic method.

The values of  $K_t$  calculated from formulas (9) and (15) are adequate, in practice. Formulas (11) and (15) with account for (4), conversely, disagree not only quantitatively but also qualitatively. This leads us to state that the linear phenomenological equations of thermal dynamics do not correspond to the situation of heat and mass transfer in the binary gas mixture that is formed in the two-cylinder thermal-diffusion apparatus in the process of thermal diffusion. Therefore, the second variant of the experimental method of unique determination of  $K_t$  cannot be recommended as a reliable method.

The formulas of kinetic theory also fail to satisfactorily describe the tabulated data on  $\alpha_t$  for the He–Ar mixture.

In closing, we note that the present paper contains a preliminary discussion of the scientific results obtained. We are planning a number of publications in the future which present the subject matter of these investigations in greater detail.

## NOTATION

$K_t$ , thermal-diffusion relation;  $\alpha_t$ , thermal-diffusion constant;  $T_0$  and  $T_h$ , temperatures of the hot and cold cylinders of the thermal-diffusion apparatus respectively, K;  $x_{10}$  and  $x_{1h}$ , mole fraction of the light gas in the hot and cold cylinders respectively;  $z$ , longitudinal coordinate along the connecting tube of the two-cylinder apparatus, m;  $h$ , length of the connecting tube, m;  $\Delta x_1 = x_1 - x_{1h}$ ;  $\Delta T = T - T_h$ ;  $p$ , total pressure;  $R$ , universal gas constant, J/(mole·K);  $\mu_1$ ,  $\mu_2$ , and  $\mu$ , mole masses of the light, heavy, and binary gases, kg/mole;  $\lambda = \varphi(x_1)f(T)$ , thermal conductivity,

$J/(m^2 \cdot K \cdot sec)$ ;  $\varphi(x_1)$ , empirical function of  $x_1$ ;  $f(T)$ , empirical function of  $T$ ;  $\rho$ , density of the mixture;  $kg/m^3$ ;  $T$ , temperature of the mixture, K;  $D$ , interdiffusion coefficient,  $m^2/sec$ ;  $H(T) = H_1(T) - H_2(T)$ , difference of the specific mass enthalpies of the light and heavy gases, J/kg;  $x_2$ , mole fraction of the heavy gas;  $\bar{\mu} = \mu(\bar{x}_1)$ ;  $\bar{\rho} = \rho(\bar{x}_1, T)$ ;  $\bar{\lambda} = \lambda(\bar{x}_1, T)$ ;  $C(\bar{x}_1)$ , undetermined constant;  $\bar{x}_1$ , certain fixed value of  $x_1$ . Subscript: t, thermal-diffusion.

## REFERENCES

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