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TRANSFER PROPERTIES OF MIXTURES OF RAREFIED NEUTRAL GASES. HYDROGEN–ARGON SYSTEM

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A method is proposed for generalization of the transport properties of mixtures of rarefied gases on the basis of their particle-interaction potentials and relations of the molecular-kinetic theory. A simultaneous processing of data on the viscosity of a binary Ar–H_2 *mixture and its components as well as data on the concentration-diffusion and thermal-diffusion coefficients of this mixture has been carried out by the weight method. The parameters of three functions of the Lennard-Jones (m–6) potential of interaction between the Ar atoms and* H_2 *molecules in the indicated mixture were determined. Tables of reference data on the transport properties of the Ar–*H_2 *mixture at a temperature of 200–2000 K and a concentration x(Ar) varying from 0 to 1 were calculated.*

Keywords: *gases, gas mixtures, transport properties, hydrogen, argon, viscosity, diffusion coefficient, thermal-diffusion coefficient, interaction potential, reference data.*

Introduction. Mixtures of gases kept under relatively low pressure play an important role in nature (cosmos, earth's atmosphere and biosphere) as well as in man-made apparatus and processes. Many processes of heat and mass transfer proceed in gaseous media representing, as a rule, mixtures of gases.

From the standpoint of molecular physics, the thermodynamic properties of the indicated gaseous mixtures can be represented in the first approximation as properties of mixtures of ideal gases, and the transport properties of such systems (their viscosity, heat conductivity, and concentration and thermal-diffusion coefficients) can be considered within the framework of the model of pair collisions of atoms and molecules of their components. The theory of transport properties of monatomic gases, based on solution of the Boltzmann equation, is a rigorous theory [1, 2]. To calculate the properties of substances by relations of this theory, it is necessary to know their pair potentials of interatomic interactions. Currently, the "true" interaction potentials of the classical representatives of monoatomic gases — inert gases in the ground electron state — have been determined on the basis of simultaneous analysis of experimental data on their molecular beams, molecular spectra, second virial coefficients, and transport properties and results of the corresponding quantum-mechanical calculations. It has been possible to represent these potentials with a high degree of accuracy by the parametric functions $U(R, \mathbf{a})$ with a number of parameters a_i = 8–10 [3]. On the basis of the indicated potentials and relations of the molecular-kinetic theory, tables of reference data on the viscosity and heat conductivity of a group of rarefied gases (He, ..., Xe) at temperatures lower than 5000 K were calculated [4]. Knowledge of the "true" interaction potentials of rarefied gases makes it possible to develop, with the use of the mathematical apparatus of the molecular-kinetic theory, basic standards for calculating the transport properties of these gases (see, e.g., [5]).

In the case of pair interactions between atoms and molecules and interactions between molecules, two additional factors should be taken into account: 1) the dependence of the trajectories of collisions of particles on the angles φ, i.e., the nonspherisity of the interaction potentials $U(R, \varphi)$; 2) the thermal excitation of molecules and the energy exchange between the translational and internal (rotational and vibrational) degrees of freedom of the colliding

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particles. In the case where particles with nonspherical potentials interact, the collision cross sections are averaged over the collision angles; this averaging is reflected in one form or another in the molecular-kinetic theory and, in part, leads to the concept of the energy-dependent effective collision cross section $\langle Q^l(E)\rangle$ and the effective potential $\langle U(R)\rangle$ [2, 6]. In the calculation of the transport properties of gases, the averaging of collision cross sections is a more rigorous procedure. The energy exchange between the translational and internal degrees of freedom of the colliding particles in a gaseous mixture influences, first of all, the heat conductivity and the volume viscosity of this mixture and, partially, its thermal diffusion. Because of this, the transport properties of gases should be calculated with account for the additional characteristics of inelastic collisions, such as the temperature-dependent rotational $Z_{\mathrm{rot}}(T)$ and vibrational $Z_{\mathrm{vib}}(T)$ collision numbers [7, 8]. Another approach involves the averaging of the trajectories of collisions of particles interacting under the action of nonspherical potentials with account for the inelastic processes [9]. At present the relaxation characteristics of only a small group of two- and three-atom molecular gases of the type of N_2, CO, CO_2 [10, 11] have been adequately investigated experimentally and theoretically, and these characteristics for other atom–molecule pairs and pairs of different-kind molecules are less understood. This circumstance makes the verification of the corresponding calculation methods and the estimation of the reliability of the calculation data on the transport properties of mixtures of gases including molecular components difficult. Thus, to calculate the transport properties of rarefied gaseous mixtures with components i and j, it is necessary to know the potentials of homogeneous U_{ii}, U_{jj} and heterogeneous (crossed) U_{ij} interactions. The heterogeneous interactions prevail in a mixture including more than three components. For example, in a mixture with five components the number of crossed interactions is equal to ten. The potentials U_{ii} and U_{jj} are determined as a rule on the basis of experimental and calculation data for individual substances.

Role of Data on the Diffusion D_{12} and Thermal-Diffusion α_T Coefficients of Gaseous Mixtures in the Generalization of Their Properties. In the calculations of the thermophysical properties of gaseous mixtures the heterogeneous-interaction potentials or their parameters are usually determined with the use of nonrigorous rules of combining the potentials of individual substances: $U_{ij} = f(U_{ii}, U_{jj})$, or their parameters: $a_{ij} = f(a_{ii}, a_{jj})$ [12]. The molecular-kinetic theory expressions for the second virial coefficient, the viscosity, and the heat conductivity of a mixture of gases include integrals of both the potentials of homogeneous and heterogeneous interactions. On the basis of these data, the potentials U_{ij} of mixtures are determined in the process of generalization of their properties or by the known potentials U_{ii} and U_{jj} [13], or as a result of the simultaneous processing of data on the properties of these mixtures and their constituent gases, or by varying the parameters of all the potentials [14]. In these procedures of verification of the thermophysical properties of gaseous mixtures on the basis of their interaction potentials, of special importance are data on the interdiffusion and thermal-diffusion coefficients of these mixtures. The point is that the theoretical dependences of the molecular-kinetic theory in the first approximation for the interdiffusion coefficient $[D_{12}]_1$ and the thermal-diffusion coefficient $\alpha_T(L)$ of a Lorentzian gaseous mixture, in which the content of the light component $x_2 \to 0$, include integrals of only the potentials of crossed interactions. For example, the expression for the interdiffusion coefficient of such a mixture at a pressure $p = 1$ atm has the form

$$[D_{12}]_1 = 2.628\cdot10^{-3}\,[T^3\,(2\mu)^{-1}]^{1/2}/\langle Q_{12}^{(11)}\rangle\,, \tag{1}$$

where $\langle Q_{12}^{(11)}\rangle = \langle d^2\Omega^{(11)*}(T^*, a_3\,...)\rangle_{12}$. At $i = j$, expression (1) is used for calculating the self-diffusion coefficient. It should be noted that, in the second approximation of the molecular-kinetic theory, the coefficients $[D_{12}]_1$ and $[D_{12}]_2^{\mathrm{qL}}$ are equal for the quasi-Lorentzian mixture in which the concentration of the heavy component $x_1 \to 0$. The dependence of $[D_{12}]_2$ on the composition of a mixture on the isotherm usually does not exceed 2%. All the listed factors determine the special role of experimental data on the diffusion coefficient of a binary mixture in the determination of its heterogeneous-interaction potential. The formula for the crossed viscosity $[\eta_{12}]_1$ of a binary mixture of gases, entering, as a part, into the expression for its viscosity, is similar in form to expression (1). The denominator of the expression for $[\eta_{12}]_1$ includes another collision integral $\langle Q_{12}^{(22)}\rangle$, and the use of one and the same potential $U_{12}(R)$ provides internal consistency of the integrals $\langle Q_{12}^{(11)}\rangle$ and $\langle Q_{12}^{(22)}\rangle$ and, in the final analysis, consistency of the reference data on the viscosity, diffusion, and translational heat conductivity.

The concentration processes occur usually under nonisothermic conditions with thermal diffusion, which should be taken into account when real processes are simulated. The dependence of the thermal-diffusion coefficient of a binary mixture $\alpha_T(T, x) = (D^T/D_{12})/[x(1-x)]$ on its concentration x is stronger by an order of magnitude than that of the diffusion coefficient. Within the framework of the model of elastic collisions, the temperature dependence α_T $(T, x = \text{const})$ is determined by the multiplier $(6C^* - 5)$, where $C^* = \Omega_{12}^{(12)*}/\Omega_{12}^{(11)*}$. Thus, the thermal-diffusion coefficient, as well as the diffusion coefficient, is a source of direct information on the potential of heterogeneous interactions. In the same approximation of the molecular-kinetic theory, the thermal-diffusion coefficient is directly related to the derivative of the interdiffusion coefficient with respect to the temperature by the relation

$$\alpha_T(L) = (6C^* - 5)/2 = 2 - (d\ln[D_{12}]_1^{\mathrm{qL}}/d\ln T)_P\,. \tag{2}$$

In a sense, the role of the thermal-diffusion coefficient in relation to the interdiffusion coefficient is similar to the role of the heat capacity in relation to the change in the enthalpy on the isobar. The relation between the interdiffusion and thermal-diffusion coefficients of a gaseous mixture should be taken into account in the analysis of experimental data on its transfer properties. The process of thermal diffusion is associated not only with mass transfer, but also with energy transfer. Therefore, in the generalization of experimental data on the thermal-diffusion coefficient of a gaseous mixture with molecular components, one should take into account the possible contribution of the inelastic collisions into the computational relations for this coefficient [15]. Unfortunately, at present the available information on the relaxation characteristics of the processes of energy exchange between the translational and internal degrees of freedom in the collisions of different-kind molecules is limited, which reguires additional investigations in this direction.

In the USSR, in the second half of the past century the interdiffusion and thermal-diffusion coefficients of gas mixtures have been much investigated, in particular, in experiments carried out at the Leningrad Polytechnical Institute, the Kazakh State University (Alma-Ata), the Ural Polytechnical University (Sverdlovsk), the Kazan' Chemical-Technological Institute, and the A. V. Luikov Heat and Mass Transfer Institute (Minsk). Results of these investigations, including methodical recommendations, descriptions of equipment, and experimental data, are presented in the Institute collected articles, journal articles, and GSSSR collections (GSSSD — State service of Standard and Reference Data) [16]. The results obtained formed the basis for tens of Candidate dissertations and several Doctor's dissertations [17–23] as well as for a number of monographs [24–27]. The investigations on the interdiffusion and thermal-diffusion coefficients of gaseous mixtures, carried out in different countries, were mainly concerned with the temperature range 90–1000 K; in this case, the error in determination of these coefficients was of the order of 2–5%, and only in some works performed at room temperatures was the error in estimation of the indicated coefficients decreased to several fractions of a percent [28]. The systems investigated comprised inert and atmospheric gases, their mixtures, and mixtures of these gases with lower hydrocarbons and polar gases — water vapor, ammonia, lower alcohols, and freons. If the range of reduced temperatures T^*, investigated in the above-indicated works, is represented on the scale of the well depth ε_{12} of the potential $U_{12}(R)$ and the value of $\varepsilon_{12} \approx 100$ K characteristic of the indicated constituent gases is taken, the temperature range of investigation of the interdiffusion and thermal-diffusion coefficients will be equal to $T^* \sim 1$–10. For mixtures with He and H_2 the upper boundary of the temperature range of investigation of their properties reaches $T^* = 50$–100. In this situation the generalization of results of experimental investigations on the properties of such mixtures on the basis of their interaction potentials should be performed with special emphasis on the characteristics of the repulsive branch of the heterogeneous-interaction potentials of the indicated mixtures.

Experimental works on the interdiffusion coefficient carried out up to 1969 are listed in the known review [29]. In this review, the semiempirical dependences $D_{12}(T)$ are presented for 74 mixtures of technologically important gases. Collections of experimental data on the interdiffusion coefficients of gaseous mixtures are presented in the review of Andrussov [30], in the handbook of Vargaftik [31, 32], and in monograph [33]. The new volume of the Landolt–Bornstein reference book [34] includes tables of experimental data on the interdiffusion coefficient of gaseous systems obtained after 1970. Unfortunately, information on the investigations of the interdiffusion coefficient of gases, carried out in the CIS countries in this period, are absent in this publication. In a number of reference books, information on the interdiffusion coefficient of gases on the isobar are represented in the form of the power law $D/D_0 = (T/T_0)^n$ where, as a rule, $T_0 = 298$ K and the exponent $n = \text{const}$. A collection of approximately 150 pairs of inorganic substances used in gas-transport metallurgy processes and processes of electronic engineering [35] can be found in the indicated series of works. Monograph [36] also includes reference data on the interdiffusion coefficient of atoms of alkali metals and metals of the zinc group in inert and atmospheric gases. It should be noted that the use of the above-indicated power law with a constant exponent n for the interdiffusion coefficient is reasonable only for a narrow temperature range, especially in the region of $T^* = 1$–10. An imposing collection of reference data on the interdiffusion coefficient of ~1360 carbon compounds (up to $C_{28}H_{46}O_4$) is presented in handbook [37]. However, in this publication there are no references to information sources for concrete systems.

The number of experimental works on the thermal-diffusion coefficient is smaller as compared to that for the interdiffusion coefficient. Their bibliography is presented in monographs [25–27, 38] and in review [39]. A large collection of experimental data on the thermal-diffusion separation $q = x_1(T_2) - x_1(T_1)$ in binary mixtures, obtained up to 1970, is presented in handbooks [31, 32]. Monograph [27] includes analogous tables of data obtained at the Heat and Mass Transfer Institute of the National Academy of Sciences of Belarus for mixtures of inert gases, mixtures of helium with NH_3, N_2O, and H_2, mixtures of argon and nitrogen with lower alcohols, and so on. Tables of data on the recommended values of the interdiffusion coefficient and the thermal-diffusion separation for a number of gas systems are stored in the GSSSD funds [40].

Interaction Potentials. Experimental data on the thermophysical properties of substances are analyzed and generalized on the basis of their interaction potentials and molecular-kinetic theory relations with the use of additional explicit or implicit information on the properties of these substances. In this case, the following apriori information can be used:

a) results of quantum-mechanical calculations of particle interaction potentials;

b) results of physical experiments on molecular beams and the spectroscopy of interatomic and intermolecular interactions;

c) relations between the structural characteristics of atoms and molecules and the parameters of their potentials [41–43], and estimates of the parameters of the heterogeneous interaction potentials with the use of combination rules for the parameters of the homogeneous interaction potentials;

d) results of generalization of the properties of different gases and gaseous mixtures on the basis of the similarity theory that is usually used in molecular physics in terms of the method of corresponding states [44–46].

Not going into details, we will point to two circumstances: 1) in the last few years each of the indicated directions of investigations has been developed rapidly due to the improvement of the experimental technique and the development of computer facilities; 2) the experience gained in investigations on the interaction potentials and the thermophysical properties of substances points to the fact that the two-parameter theory of corresponding states going back to the Van der Waals equation does not satisfy modern requirements on the accuracy of experiments. For example, at present, additional parameters — Pitzer factor [46] or the Filippov factor [45] — are used in the generalization of the thermodynamic properties of nonpolar gases. Analogously, in the generalization of data on the second virial coefficient and the transport properties of gaseous mixtures with the use of the corresponding states of the standard gas argon the authors of [47] used, in addition to the geometric parameter d (collision diameter) and the energy parameter ε (potential-well depth), two or three characteristics concerning the special features of the interaction potentials at small and large distances [48]. Nevertheless, the group of such substances considered in [47, 48] does not include helium and molecular hydrogen. In our opinion, this is explained not only by the quantum properties of these substances appearing at low temperatures but also by the fact that the pair interaction potentials of the indicated gases and their mixtures have more smooth repulsive branches in the energy range corresponding to the temperature range 100–2000 K being considered. Recently the transport properties of gaseous hydrogen have been generalized by the Mason–Kestin scheme; in this case, argon was not used as a basis and new generalization dependences for reduced collision integrals were proposed [49].

As noted above, data on the thermal-diffusion coefficient of a gaseous mixture carry important information on its heterogeneous-interaction potential. Therefore, it would appear reasonable to perform correlation of the properties of gaseous mixtures and determination of their interaction potentials with the use of results of generalization of the indicated data by the similarity methods for mixtures of inert [50, 51] and nonpolar [52] gases and mixtures with polar components [53]. The experiments on the thermal-diffusion coefficient of gaseous mixtures are limited, with rare exception [54], by the temperature range 600–800 K, which corresponds, for mixtures of inert and atmospheric gases, excepting helium and hydrogen, to the reduced temperatures $T^* = 7$–8. At the same time, the temperature range of investigation of the thermal-diffusion coefficient of mixtures with helium and hydrogen on the scale of reduced temperatures reaches $T^* = 10$–18, and the corresponding generalized data are of great importance for analysis of the properties of such substances and determination of their heterogeneous-interaction potentials by data of a different, in particular, for hydrogen-containing gaseous mixtures.

An important stage of the analysis and generalization of the transport properties of gaseous mixtures is the choice of models of particle-interaction potentials. In this connection, two directions of calculations — individual calculations and mass ones — should be differentiated.

On the one hand, at present, detailed trajectory calculations of the collision cross sections and the integrals of elastic and inelastic collisions are carried out for individual pairs of monatomic and diatomic gases, such as the inert gas N_2 and the inert gas H_2, on the basis of the nonspherical interaction potentials, and then the correspondence between the physical information (beams, spectra) and the thermophysical one (second virial coefficient, viscosity, interdiffusion coefficient, etc.) is analyzed [9]. The perspectives and problems of this approach is an object of modern investigations [55].

On the other hand, the initial micro- and macroinformation on the interaction potentials and properties of a large majority of gases is absent, and for many users of interest is only the final product — tables of thermophysical properties, independently of the accuracy of their determination.

Mass calculations give estimates of the parameters ε and d of the two-parameter Lennard-Jones (12–6) potentials (L-J (12–6)) or (exp–6). The procedures of obtaining of these parameters involve correlations of the thermodynamic properties of substances (their critical parameters, density, surface tension, etc.) and the parameters of their potentials [35, 56] with the use of the properties of crystals [57], the method of increments for complex molecules [58], and so on. In these procedures the parameters of the heterogeneous–interaction potentials are determined by a combination of estimates obtained for individual substances. The parameters of the L-J (12–6) potential of a large group of ~250 individual substances are used in a number of computational complexes, e.g., on the transport properties of the combustion products of propellants [56, 59, 60].

It should be noted that all characteristics of the L-J (12–6) potential — the complex $4\varepsilon d^6$ as an analog of the dispersion-energy coefficient C_6, the slope of the potential at the point of minimum, and the slope of the high-energy portion of the repulsive branch — are overstated as a rule. At the same time, this potential for pure gases and their mixtures allows one to satisfactorily (within the limits of several percents) estimate a number of thermophysical parameters of these substances, e.g., their second virial coefficient, viscosity, and interdiffusion coefficient, in a limited temperature range, especially if each property of a substance is determined by its own set of parameters [1]. However, as an analysis has shown, it is precisely the transport properties of mixtures of gases at temperatures higher than the room temperatures, to which the values of $T^* > 1$ correspond, for which the repulsive branch of the L-J (12–6) potential is too sharp as applied to mixtures with He and H_2 and is too smooth for mixtures with heavy globular molecules [61, 62]. At present, improved models and procedures of calculation of combustion processes and associated calculations of heat and mass transfer with estimation of the influence coefficients depending on many factors [63] are actively developed. In this situation the requirements for the quality of the reference data on the transport properties of gases are increased. Clearly investigations on the interaction potentials of substances and the possibilities of their application to the description of the properties of these substances can be realistically placed into the category of fundamental works.

The properties of gases and gas mixtures can be described with the use of a large number of small-parameter functions $U(R)$ alternative to the L-J (12–6) potential [2]. In the present work we use the generalized L-J (m–6) potential with a varied exponent $m \sim$ var of the repulsive branch

$$\frac{U(R)}{\varepsilon} = \frac{6}{m-6} X^{-m} - \frac{m}{m-6} X^{-6} , \tag{3}$$

where $X = R/R_e$, $U(X = 1)/\varepsilon = -1$. The collision diameter d is determined from the condition $U(d) = 0$ and is equal to $R_e(m/6)^{1/(m-6)}$.

The L-J (m–6) potential:

1) degenerates into the traditional L-J (12–6) potential at $m = 12$, which allows one to use the experience gained in work with this potential;

2) allows one to vary the slope of the repulsive branch over wide limits from the limiting smooth curve at $m = 6$ [64] to a sharp one at $m \rightarrow \infty$ [1];

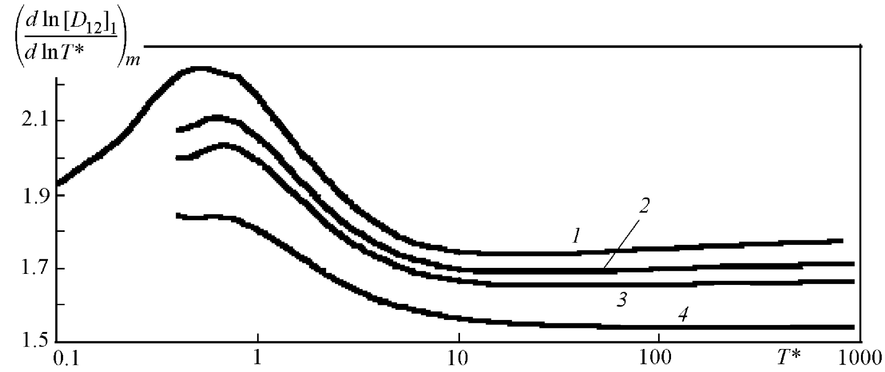

Fig. 1. Isolines of the logarithmic derivatives $(d \ln [D_{12}]_1/d \ln T^*)_m$ at different values of the exponent m: $m = 6$ (1), 9 (2), 12 (3), and 50 (4).

3) for this potential, collision integrals were calculated with a high degree of accuracy throughout the range of change in $m = 6$–∞ [64, 65], and they were approximated in the temperature range $T^* = 0.4$–200 and in the range of change in $m = 8$–∞ with the use of simple algebraic expressions [66];

4) in a medium with polar gases, in addition to the spherically symmetric potential, a potential dependent on the mutual orientation of particles is formed due to the interaction of the two dipoles μ_1 and μ_2. As applied to the L-J (m–6) potential, one can pass in a certain approximation to the averaged potential with overdetermined parameters $d(\mu_1, \mu_2, m)$ and $\varepsilon(\mu_1, \mu_2, m)$ [67]. The parameters of a mixture of polar and nonpolar gases can be analogously transformed with account for the induced dipole-dipole interaction;

5) a statistical processing of data with the use of the two-parameter functions like the L-J (12–6) or the L-J (m–6) potential with a fixed exponent m gives underestimated errors in the calculated values of parameters. The use of three-parameter potentials with a varied exponent m usually allows one to obtain more realistic estimates of the corresponding errors;

6) as the experience shows, the use of the one-type L-J (m–6) functions for the homogeneous and heterogeneous interactions allows one to fit, in a wide temperature range, the experimental data on the transport properties of individual gases to those of the corresponding gaseous mixtures within typical errors of their determination (Ar, N_2, H_2, H_2O) [68, 69].

One-type potential functions are also convenient for formation of the error matrix of parameters in the process of approximation of the corresponding experimental data; this matrix is used then for estimation of the errors in the calculated reference data on the transport properties of substances.

For illustration of the possibilities of the L-J (m–6) potential, Fig. 1 shows isolines of the derivative of the diffusion coefficient $(d \ln [D_{12}]_1/d \ln T)_m$ depending on the reduced temperature T^* for $m = 6$, 9, 12, and 50. Recall that $[D_{12}]_1 = [D_{12}]_2^{qL}$ [1]. To this derivative corresponds the exponent n in the power law of the diffusion coefficient $D(T)$ that is frequently used for generalization of experimental data. As an analysis has shown, in the limit of high temperatures, the indicated derivative tends asymptotically to the value $1.5 + 1/m$. At the left of $T^* = 1$ the isolines for m = const have maxima that reach the value 2.24 at $m = 6$. Behind the maximum on the isolines, the derivative decreases sharply with increase in the temperature to $T^* \sim 10$. It is evident that in the generalization of experimental data on $D(T)$ in the indicated temperature region with the use of a power law the exponent n = const cannot be used, and the use of the linear temperature dependence of the type of $n = n_0 - b(T - T_0)$ [30] should be controlled.

For the thermal-diffusion coefficient, as well as for the diffusion coefficient, the isolines $\alpha_T(T)$ for the L-J (m–6) function separate markedly at m = const. Below are data on the thermal-diffusion coefficient at $T^* = 10$ and characteristic values of m for the Lorentzian mixture of two gases at $x_2 \to 0$, calculated with the use of relation (2):

m	6	9	12	18
$\alpha_T(L)$	0.253	0.300	0.332	0.372

For typical mixtures with helium, the temperature $T^* = 10$ corresponds to the room temperatures at which a large number of experimental data were obtained. Thus, comparison of the experimental and calculated values of the thermal-diffusion coefficients of gaseous mixtures can give, in a number of cases, estimates of the exponent m for the potentials of heterogeneous interactions before a detailed statistical processing of the corresponding experimental data is performed.

Method of Simultaneous Processing of Data. The apparatus of the molecular-kinetic theory of gases and gaseous mixtures allows one to perform a simultaneous processing of experimental data on the thermophysical properties of substances on the basis of common interaction potentials. For coordination of the properties of substances and search for their interaction potentials with optimum parameters, the least square method (LSM) is usually used in the form

$$S = \sum_i \sum_j W_{kl} \left[Y_{kl}(T, x) - y_{kl}(T, x, \mathbf{a})\right]^2 \Rightarrow \min , \tag{4}$$

where $W_{kl} = (\delta_{kl} Y_{kl})^{-2}$ are the weights of experimental points that are calculated with the use of the estimated relative errors of data. The form of expression (4) and each of its terms is written with some assumptions.

The LSM variant in the form of (4) is written for a system of independent random quantities. In this case, only the diagonal terms are retained in the complete weight matrix of the initial LSM, and the matrix representation of the LSM degenerates into the scalar form (4). It is assumed that the errors of the temperature and composition of a substance determined at Y_{kl} and y_{kl} are exact, and the corresponding errors in their assignment are involved in the estimates of δ_{kl}.

The molecular-kinetic theory formulas for the second virial coefficient, viscosity, and heat conductivity of a gaseous mixture include both integrals of homogeneous interactions ii, jj and integrals of heterogeneous interactions ij. This makes it possible to coordinate the experimental data for individual substances and their mixtures. As noted above, the collision integrals necessary for calculation of the transport properties of substances are determined for the L-J (m–6) potentials with the use of approximation expressions [66]. For a binary mixture of gases, the three potentials U_{11}, U_{22}, and U_{12} with nine parameters d_{11}, d_{22}, d_{12}; ε_{11}, ε_{22}, ε_{12}; m_{11}, m_{22}, m_{12} are considered, and functional (4) is minimized by these parameters. For mixtures with polar molecules, the dipole moments μ_1 and μ_2 are additionally used as varied parameters. The models of the heat conductivity and the thermal-diffusion coefficient of the molecular components of mixtures include expressions for the collision numbers of their rotational relaxation with their own parameters Z_{ii}^0 and Z_{jj}^0.

The expressions for calculation of the transport properties of rarefied gases were obtained on the basis of the Boltzmann equation in the form of expansions of different orders in terms of Sonin polynomials. In the present work the following dependences satisfying the accuracy of the experimental data were used. The interdiffusion coefficient was calculated in the second approximation of the molecular-kinetic theory $[D_{12}]_2 = [D_{12}]_1 f_D(T, x)$, where $[D_{12}]_1$ is determined by relation (1) and the multiplier $f_D(T, x)$ depends weakly on the composition of a mixture [1, 2].

The viscosity of a binary mixture of gases $\eta_{\mathrm{mix}}(T)$ is determined by the Storvick–Mason scheme [70] in such a way that the structure of the general expression corresponds to the first approximation of the molecular-kinetic theory for gaseous mixtures; however, the partial coefficients $[\eta_{11}(T)]_2$, $[\eta_{22}(T)]_2$, and $[\eta_{12}(T)]_2$ involved in it are determined in the second approximation. The viscosity of individual substances is calculated automatically in the second approximation.

The heat conductivity of a gaseous mixture is calculated by the dependence [1]

$$\lambda_{\mathrm{mix}}(T, x) = \lambda_{\mathrm{mix}}^{\mathrm{mon}}(T, x) + \sum_n \left[\lambda_i(T) - \lambda_i^{\mathrm{mon}}(T)\right]\left[1 + \sum_{i \neq j} (x_j D_{ii})/(x_i D_{ij})\right]^{-1} . \tag{5}$$

With allowance made for the real accuracy of experimental data on the heat conductivity of gases, the partial values of the heat conductivity λ_i and λ_i^{mon}, involved in $\lambda_{\mathrm{mix}}^{\mathrm{mon}}$, are calculated in the first approximation of the molecular-kinetic theory.

The thermal-diffusion coefficient $\alpha_T(T, x)$ of a binary mixture of gases was calculated in the nonvanishing first approximation of the theory in the initial Chapman–Cowling formulation [1].

Functional (4) can also include, as the initial data, apriori values of the parameters a_i of the potentials of homogeneous and heterogeneous interactions with their own estimates of errors.

TABLE 1. Initial Data on the Viscosity of Argon, Hydrogen, and Ar–H_2 Mixture

Authors	T, K	Number of points	$x(H_2)$	Notes
Trautz, Ludewigs [72]	293–523	24	0.13–0.94	Capillary method for variable flow rate, δ = 2–3%
Trautz, Binkele [73]	293–523	16	0.30–0.63	Capillary method for variable flow rate, δ = 2%
Lierde van [74]	288, 290	5	0.14–0.64	Disk method, Knudsen regime, δ = 10%
Heath [75]	291	9	0–1	Capillary method, points on the graph [75]
Guevara, McInteer, Wageman [76]	1102–2128	23	1	Capillary method for constant flow rate, δ = 1.5–2%
Clifford, Kestin, Wakeham [77]	298, 308	4	0.35, 0.71	Disk method, δ = 0.4%
GSSSD R 233–87 [78]	200–1500	14	1	Tables of recommended reference data, δ = 1–1.5%
GSSSD 138–89 [4]	200–2500	14	0	Tables of standard reference data, δ = 0.75–0.5–2%
May, Berg, Moldover, [79]	298–394	25	0, 1	Capillary method, δ = 0.2%

The weights W_{kl} applied to functional (4) merit special consideration. It is precisely these weights that make the minimized functional dimensionless, which makes it possible to perform a simultaneous processing of experimental data having different dimensions. In accordance with the sense of the least square method, the errors of the experimental points in (4) should correspond to the spread of the random variables relative to the average ones. However, real experimental data always include systematic errors. The proficiency of experimenters is substantially determined by their ability to estimate the systematic errors of an experiment and then to propose broadened fiducial estimates of measurement results. A theorist developing reference data agrees with estimates of an experimenter or propose its own quantities that, in the form of the multipliers $W_{kl} = (\delta_{kl} Y_{kl})^{-2}$ for each point of a data array, appear in the expression for functional (4). Thus, the LSM functional in the form of (4) is inconsistent at least twice with the premises of the least square method, first, when all the points are considered as independent random quantities, and, second, when systematic components are introduced into the estimates of errors. At the same time, all the details of formation of functional (4) are strictly algorithmic, which provides the reproduction of the procedure of obtaining reference data.

The variance-covariance matrix (matrix of errors) of the parameters $\mathbf{D}(\mathbf{a})$ generated by the least square method allows one to obtain, using the rule of extension of errors, the dispersions of the calculated data and their average square errors $s(\mathbf{y})$. In turn, for going from the quantities $s(\mathbf{y})$ to the total (reliable) errors of the reference data $\Delta_P(\mathbf{y})$ at a confidence coefficient P, the following expression is used:

$$\Delta_P(\mathbf{y}) = t_P s(\mathbf{y}) . \qquad (6)$$

As an analysis [71] has shown, to obtain the upper estimates of the confidence coefficients on condition that the content of the statistical procedures is retained, it is appropriate to assign the errors to the 90% probability, and, for the quantile, to take the value of t_P = 5.

Ar–H_2 Mixture. The above-described procedure of generalization of experimental data on the basis of relations of the molecular-kinetic theory of gases and the interaction potentials of gaseous mixtures was used for obtaining reference data for N_2–O_2, N_2–CO_2, N_2–Ar [68], N_2–H_2 [69], and H_2–H_2O mixtures. Below are results of analysis and generalization of data on the transport properties of an Ar–H_2 mixture. This work is a continuation of the program on obtaining reference data on the properties of hydrogen-containing mixtures. The coordination procedure did not include consideration of data on the second virial coefficient of the indicated mixture, which made it possible to disregard the difference between the effective potentials of the equilibrium and nonequilibrium (transport) properties of its components and data on the heat conductivity of the mixture because of the absence of information on the crossed rotational collision numbers $Z_{\text{Ar–H}_2}(T)$ of its components.

Table 1 presents information on the works devoted to investigating the viscosity of gases, the results of which were used for coordination of data obtained for the Ar–H_2 mixture and determination of its interaction potentials.

Reliable experimental and reference data on the viscosity of gaseous argon and hydrogen were obtained for a wide range of temperatures. The coordination procedure included the consideration of the standard reference data for argon [4] in the temperature range 200–2500 K, the recommended reference data for hydrogen [78] in the temperature

TABLE 2. Initial Data on the Interdiffusion Coefficient

Authors	T, K	Number of points	Notes
Waldman [80]	293	1	Two-flask method, $\delta = 5\%$
Strehlow [81]	288, 354, 418	3	Loschmidt method, $\delta = 10\%$
Paul, Srivastava [82]	303–418	2	Two-flask method, $\delta = 5\%$
Westenberg, Frazier [83]	295–1069	6	Point-source method, $x(H_2) \to 0$, $\delta = 4–5\%$
Ivakin, Suetin [84]	295–473	7	Loschmidt method, $\delta = 3\%$
Mason, Weissman, Wendt [85]	293	4	Two-flask method, $\delta = 5\%$
Suetin, Loiko, Kalinin, Gerasimov [86]	118–296	5	Two-flask method, $\delta = 3\%$
Suetin, Kalinin, Loiko [87]	476–695	4	Two-flask method, $\delta = 4\%$
Kosov, Kurlapov, Solonitsyn [88]	296–809	14	Two-flask method + stationary-flow method, $\delta = 5\%$
Loiko, Kalinin, Suetin [89]	77.2	1	Two-flask apparatus, $\delta = 5\%$
Harris, Bell, Dunlop [90]	300	10	Loschmidt method, semiconductor katharometer of composition, $x(H_2) = 0.04–0.95$, $\delta = 0.7\%$
Harris, Bell [91]	300	7	Loschmidt method, semiconductor katharometer of composition, $x(H_2) = 0.04–0.95$, $\delta = 0.7\%$
Wahby et al [92]	237–423		Two-flask method, variant of relative measurements, $D(299.6\text{ K}) = 0.820\text{ cm}^2/\text{sec}$, $x(H_2) = 0.95$, $\delta = 0.6\%$
Trengove, Dunlop [93]	300	2	Two-flask method, $x(H_2) = 0.01, 0.99$; $\delta = 0.5\%$
Wahby, Los [94]	237–419	8	Two-flask apparatus, variant of relative measurements, $D(299.6\text{ K}) = 0.820\text{ cm}^2/\text{sec}$, $x(H_2) = 0.95$, $\delta = 0.6\%$
Dunlop, Robjohns, Bignell [28]	200–340	18	Two-flask method, semiconductor katharometer of composition, $x(H_2) = 0.9$, $\delta = 0.5\%$

range 250–1500 K, and the high-temperature experimental data [76]. The new precise-measurement data on the viscosity of the indicated gases [79] were taken into account. To these data correspond collision integrals $\langle \Omega_{11}^{(22)*} \rangle$ and $\langle \Omega_{12}^{(22)*} \rangle$ in the calculation expressions, and this information forms the basis for determination of the homogeneous-interaction potentials.

The viscosity of the Ar–H_2 mixture was determined only in five experimental works in a narrow temperature range 288–523 K [72–75, 77]. The errors of these data presented in Table 1 were considered by us with account for the experience of the previous generalizations [68, 69]. In [74], experimental data on the viscosity of the indicated mixture were obtained for the Knudsen regime by the oscillating-disk method, and the results extrapolated to the atmospheric pressure with a large error were included into the data-processing procedure.

The calculated temperature dependence of the viscosity of the Ar–H_2 mixture includes the crossed-interaction integral $\langle \Omega_{12}^{(22)*} \rangle$. However, as follows from Table 1, the data array obtained for the Ar–H_2 mixture is limited by the narrow temperature range, which increases the importance of the data on the interdiffusion coefficient for determination of the heterogeneous-interaction potential $U_{\text{Ar–H}_2}$. Information on measurements of the interdiffusion coefficient of the Ar–H_2 mixture are presented in Table 2, in the note of which the errors of the experimental data and data on the hydrogen concentration $x(H_2)$ in the mixture are given. If the value of $x(H_2)$ is absent, it is assumed that it is equal to 0.5.

The works listed in Table 2 can be divided into two groups. One group is concerned with the measurements of the interdiffusion coefficient in the temperature range 200–400 K with an error <1% [28, 90–94]. These works clearly demonstrate the concentration dependence of the interdiffusion coefficient. Another group includes a series of works carried out in the wide temperature range 200–1000 K with an error 3–5% [84, 86–88, 95]. The data obtained in the indicated works form an imposing basis for determination of the integral $\langle \Omega_{11}^{(22)*} \rangle$ and the interaction potential U_{12}. In the determination of this potential, of importance are also the low-temperature data of [86, 89]. As an analysis has shown, the collision integral $\langle \Omega_{12}^{(22)*} \rangle$ of the Ar–H_2 mixture at $T > 100$ K can be calculated without consideration for the quantum correction against the background of the errors of the experimental data on the interdiffusion coefficient. In [96], results of control measurements of the interdiffusion coefficient of mixtures of hydrogen with inert gases

TABLE 3. Initial Data on the Thermal-Diffusion Coefficient

Authors	T_{av}, K	$x(H_2)$	Number of points	Notes
Ibbs [97]	325, 375	0.06–0.85	6	Two-flask method, δ = 10%
Ibbs, Grew, Hirsk [98]	200–250	0.47, 0.56	12	Two-flask method, δ = 10%
Waldman [80]	293	0.05–0.95	10	Diffusion thermal effect, δ = 10%
Mason, Waissman, Wendt [85]	239, 355		3	Two-flask method, δ = 10%
Bogatyrev, Makletsova [20, 54]	377, 464	0.07–0.95	30	Two-flask method, δ = 10%
Trengove, Dunlop [93]	300	0.2, 0.5, 0.8	3	Two-flask method, δ = 3%
Dunlop, Robjohns, Bignell [28]	255, 350	0.19–0.79	8	Two-flask method, δ = 2%
Shashkov, Zolotukhina [our data]	150–450	0.5	9	Similarity method, δ = 5%

in the temperature range 240–320 K are presented. In these measurements, the Loschmidt method and the method of two flasks were used. The discrepancy between these data and the disagreement between them and the corresponding measurement results [28, 94] did not exceed 0.25%, which is evidence of the high level of the indicated works.

As noted above, data on the thermal-diffusion separation in a gaseous mixture, recalculated for the thermal-diffusion coefficient, give valuable information on the temperature behavior of the interdiffusion coefficient and allow one to discriminate the interaction potentials, in particular, their branches. Table 3 lists the works used for coordination of the data being considered. Following the common practice, the experimental data obtained for the temperature range T_1, T_2 were assigned to the mean-logarithmic temperature [38]. In the further processing, for the experimental data of [54, 80, 85, 97, 98] the error of ~10% was taken with account for the discrepancy between the results of different works. During a number of years, measurements of the thermal-diffusion coefficient of many binary mixtures, in particular of Ar–H_2, were conducted at the University of Adelaide (Australia) [28, 93]. For these data, the error of 2–3% was taken with account for careful calibrations of the equipment being used and the degree of coordination of different series of measurements.

In our work, a special generalization of experimental data on the thermal-diffusion coefficient of mixtures of hydrogen with inert gases and nitrogen was carried out with the use of the similarity-theory method by the algorithm of [52, 53]. The results of this generalization for the Ar–H_2 mixture of equimolar concentration at a temperature of 150–450 K were also included in the procedure of simultaneous data processing with an error of 5%:

T, K	150	160	200	250	300	350	400	450
α_T	0.181	0.192	0.232	0.271	0.300	0.317	0.333	0.345

Finally, as a one further type of information, the following apriori estimates of the parameters of the L-J (m–6) potentials of the interacting pairs, obtained from analysis of literature data with the use of rules of combining parameters, are included, clearly with their own errors, in the minimized functional (4):

1) for the potential of argon $U_{11} : d_{11} = 3.35 \pm 0.01$ A, $\varepsilon_{11} = 141.5 \pm 7$ K, $m_{11} = 11.5 \pm 1$;

2) for the potential of hydrogen $U_{22} : d_{22} = 3.1 \pm 0.15$ A, $\varepsilon_{22} = 25.7 \pm 2.5$ K, $m_{22} = 8.0 \pm 0.75$;

3) for the potential of Ar–H_2 $U_{12} : d_{12} = 3.27 \pm 0.25$ A, $\varepsilon_{12} = 56.7 \pm 6$ K, $m_{12} = \pm 0.7$.

Results and Discussion. The simultaneous processing of data (347 points) was carried out with minimization of functional (4) by nine parameters of three L-J (m–6) potentials. The sum of the weighted squares of deviations S = 119. The optimum parameters and their mean-square errors are as follows:

1) for the potential of argon $U_{11} : d_{11} = 3.377 \pm 0.018$ A, $\varepsilon_{11} = 133.48 \pm 4.3$ K, $m_{11} = 11.0 \pm 0.55$;

2) for the potential of hydrogen $U_{22} : d_{22} = 3.140 \pm 0.026$ A, $\varepsilon_{22} = 25.67 \pm 2.4$ K, $m_{22} = 8.0 \pm 0.15$;

3) for the potential of Ar–H_2 $U_{12} : d_{12} = 3.230 \pm 0.021$ A, $\varepsilon_{12} = 61.23 \pm 2.2$ K, $m_{12} = 9.0 \pm 0.15$.

For convenience of users, the near-optimum round-off values of the parameter m were used and then the increments Δm were limited prior to the first iteration in the minimization of functional (4).

Practically all the data processed on the viscosity and the interdiffusion and thermal-diffusion coefficients deviated from the calculation surface within the limits of the predetermined errors. It should be noted that for a number of parameters the estimated errors were less than $\delta < 1\%$.

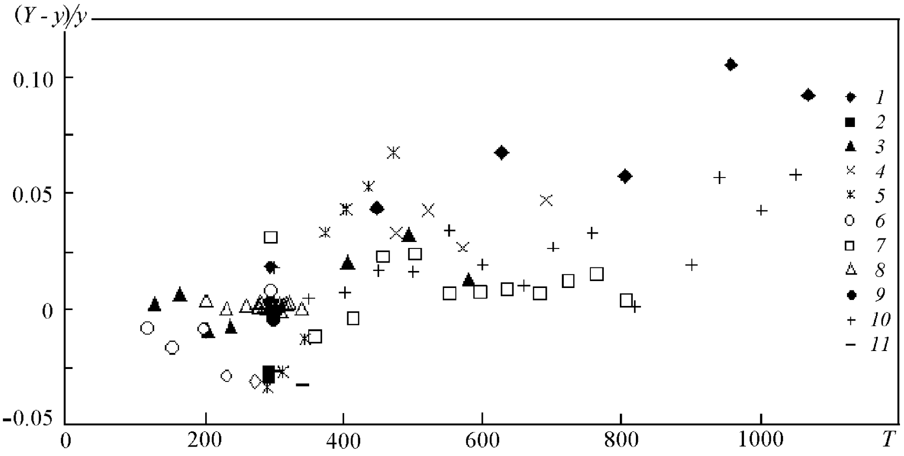

Fig. 2. Graph of derivatives of the experimental data on the interdiffusion coefficient from the calculated ones: 1) [83]; 2) [85]; 3, 4) [86]; 5, 6) [87]; 7) [88]; 8) [28]; 9) [90]; 10) [95]; 11) [82].

The precision data on the viscosity of pure Ar and H_2 at a temperature of 200–400 K [79] deviated from the corresponding calculated dependence by less than 0.15%. The reference data for argon [4] at T = 1000–2000 K were systematically larger by 1–1.5% than the calculated ones. The calculated curve for hydrogen at $T > 1000$ K lies between the experimental [76] and reference [78] data. Thus, it may be suggested that the use of the L-J (m–6) potential with a varied exponent m for description of the viscosity of the indicated gases in the temperature range 200–2000 K is fully warranted.

Figure 2 shows the deviations of experimental data on the interdiffusion coefficient from the calculated ones. It is seen that, at $T > 400$ K, the experimental points lie systematically higher (within the estimated errors). This situation can be explained in the following way. In the temperature range 200–350 K, the deviations of the precision experimental data [28, 90–94] from the calculated curve fall within the range 0.2–0.3% and are random in character. In accordance with the meaning of the least square method, the contribution of the values of $Y(T, x)$ determined with an error of 0.3–0.5% to the minimized functional (4) is larger by 100 times than that of the analogous data with an error of 3–5%, and the potential determined, if it satisfies the physics of the effect, will reflect, first of all, the precision data.

On the other hand, the experimental data [88, 95] obtained on the equipment realizing the method of a stationary flow with a slot or capillaries had systematic deviations. These works do not give information on calibration experiments carried out with well-studied substances, in particular, inert gases, for which the "true" interaction potentials are known and the interdiffusion coefficients were determined with a high degree of accuracy [3]. As noted above, the data on the viscosity of pure argon and hydrogen at a temperature varying within a wide range can be generalized with a high accuracy with the use of the three-parameter L–J potential, and the prognosis possibility of this potential as applied to the data on the interdiffusion coefficient is beyond doubt. It is only desirable to widen the temperature range in which the viscosity of the mixture is measured and to refine the experimental data on its interdiffusion coefficient at $T > 500$ K.

As for the thermal-diffusion coefficient, we did not reveal systematic deviations of the experimental data from the calculated ones in the first approximation.

The reference data on the viscosity and the interdiffusion and thermal-diffusion coefficients of hydrogen and argon calculated by their interaction potentials and the molecular-kinetic theory relations are presented in Tables 4–6. These tables also include estimates of the enlarged errors of the reference data, calculated by relation (6). The matrix of errors of the indicated parameters is found in the www.thermophysics.ru portal.

The properties of individual substances presented in Tables 4–6 at $x = 0.1$: the viscosity of hydrogen and argon (Table 4), the self-diffusion coefficients $[D_{11}]_2$ and $[D_{22}]_2$ (Table 5), and the values of the isotopic thermal-diffusion coefficient α_T^0 (Table 6), were determined at x = 0, 1. The values of errors are given as a rule with two sig-

TABLE 4. Viscosity η and Estimates of the Error δ

T, K	η, μPa·sec δ, %	Molar fraction *x*(Ar)						
		0	0.2	0.4	0.5	0.6	0.8	1
1	2	3	4	5	6	7	8	9
200	η	6.74	12.03	14.19	14.78	15.18	15.64	15.84
	δ	1.3	0.7	0.6	0.6	0.65	0.7	0.75
250	η	7.88	14.34	17.11	17.88	18.43	19.08	19.40
	δ	0.75	0.65	0.55	0.5	0.5	0.5	0.44
300	η	8.95	16.47	19.79	20.74	21.42	22.26	22.70
	δ	0.5	0.65	0.55	0.5	0.4	0.35	0.35
350	η	9.95	18.46	22.28	23.40	24.20	25.22	25.76
	δ	0.4	0.65	0.55	0.5	0.4	0.35	0.4
400	η	10.91	20.33	24.63	25.89	26.81	27.99	28.63
	δ	0.45	0.65	0.55	0.5	0.45	0.4	0.5
450	η	11.83	22.12	26.85	28.26	29.29	30.61	31.34
	δ	0.5	0.6	0.55	0.5	0.5	0.5	0.55
500	η	12.72	23.83	28.98	30.52	31.64	33.10	33.92
	δ	0.6	0.6	0.55	0.55	0.55	0.55	0.65
550	η	13.58	25.47	31.02	32.68	33.90	35.49	36.39
	δ	0.65	0.6	0.6	0.55	0.6	0.6	0.7
600	η	14.42	27.06	32.98	34.76	36.07	37.78	38.75
	δ	0.7	0.6	0.6	0.6	0.6	0.7	0.8
650	η	15.24	28.61	34.88	36.77	38.16	39.99	41.03
	δ	0.8	0.55	0.6	0.6	0.65	0.75	0.85
700	η	16.04	30.11	36.73	38.72	40.19	42.12	43.23
	δ	0.85	0.6	0.6	0.65	0.7	0.8	0.95
750	η	16.82	31.57	38.52	40.62	42.16	44.20	45.37
	δ	0.9	0.55	0.6	0.7	0.75	0.9	1.0
800	η	17.59	33.00	40.27	42.46	44.08	46.22	47.44
	δ	1.0	0.55	0.65	0.7	0.8	0.95	1.1
850	η	18.34	34.40	41.97	44.26	45.96	48.18	49.47
	δ	1.0	0.55	0.7	0.75	0.85	1.00	1.2
900	η	19.09	35.77	43.64	46.02	47.79	50.10	51.44
	δ	1.1	0.55	0.7	0.8	0.9	1.1	1.25
950	η	19.82	37.12	45.28	47.75	49.58	51.98	53.37
	δ	1.15	0.6	0.75	0.85	0.95	1.15	1.3
1000	η	20.54	38.44	46.89	49.44	51.33	53.82	55.26
	δ	1.2	0.6	0.8	0.9	1.0	1.2	1.4
1050	η	21.25	39.74	48.46	51.10	53.05	55.62	57.11
	δ	1.3	0.6	0.8	0.95	1.1	1.3	1.5
1100	η	21.95	41.02	50.01	52.73	54.74	57.39	58.92
	δ	1.35	0.6	0.85	1.0	1.1	1.4	1.6
1150	η	22.64	42.28	51.54	54.34	56.40	59.13	60.70
	δ	1.4	0.65	0.9	1.05	1.2	1.45	1.65
1200	η	23.32	43.52	53.04	55.91	58.04	60.84	62.46
	δ	1.5	0.8	1.0	1.1	1.25	1.5	1.75
1250	η	24.00	44.74	54.52	57.47	59.65	62.52	64.18
	δ	1.5	0.7	1.0	1.15	1.3	1.6	1.85
1300	η	24.66	45.96	55.98	59.00	61.24	64.18	65.88
	δ	1.55	0.75	1.05	1.2	1.4	1.65	1.9
1350	η	25.32	47.15	57.42	60.52	62.81	65.82	67.56
	δ	1.6	0.8	1.1	1.25	1.45	1.75	2.0
1400	η	25.98	48.33	58.84	62.01	64.35	67.43	69.21
	δ	1.65	0.8	1.1	1.3	1.5	1.8	2.1
1450	η	26.63	49.50	60.24	63.48	65.88	69.02	70.83
	δ	1.7	0.85	1.2	1.4	1.55	1.9	2.15
1500	η	27.27	50.65	61.63	64.94	67.38	70.59	72.44
	δ	1.75	0.9	1.2	1.4	1.6	1.95	2.2
1550	η	27.90	51.80	63.01	66.38	68.87	72.14	74.03
	δ	1.8	0.9	1.25	1.5	1.65	2.0	2.3
1600	η	28.54	52.93	64.36	67.80	70.34	73.68	75.60
	δ	1.85	1.0	1.3	1.5	1.7	2.1	2.4
1650	η	29.16	54.05	65.71	69.21	71.80	75.19	77.15
	δ	1.9	1.1	1.35	1.6	1.8	2.15	2.45
1700	η	29.78	55.16	67.04	70.61	73.24	76.70	78.68
	δ	1.95	1.0	1.4	1.6	1.8	2.2	2.5
1750	η	30.40	56.26	68.35	71.99	74.67	78.18	80.20
	δ	2.0	1.05	1.45	1.7	1.9	2.3	2.6
1800	η	31.01	57.35	69.66	73.36	76.08	79.65	81.70
	δ	2.0	1.1	1.5	1.7	1.95	2.3	2.7
1850	η	31.61	58.43	70.95	74.71	77.48	81.11	83.19
	δ	2.1	1.1	1.5	1.75	2.0	2.4	2.75
1900	η	32.21	59.50	72.23	76.05	78.87	82.55	84.66
	δ	2.1	1.15	1.55	1.8	2.05	2.45	2.8
1950	η	32.81	60.57	73.50	77.38	80.24	83.98	86.12
	δ	2.15	1.2	1.6	1.85	2.1	2.5	2.9
2000	η	33.40	61.62	74.76	78.70	81.60	85.39	87.56
	δ	2.2	1.2	1.65	1.9	2.15	2.6	2.95

TABLE 5. Diffusion Coefficients $[D]_2$ at $p = 1$ atm and Estimates of the Error δ

T, K	$[D]_2$, cm^2/sec; δ, %	Molar fraction x(Ar)						
		0	0.2	0.4	0.5	0.6	0.8	1
1	2	3	4	5	6	7	8	9
200	$[D]_2$	0.75	0.4	0.41	0.41	0.41	0.41	—
	δ	2.0	0.65	0.65	0.65	0.65	0.65	—
250	$[D]_2$	1.10	0.60	0.60	0.61	0.61	0.61	0.13
	δ	1.4	0.55	0.55	0.55	0.55	0.55	1.1
300	$[D]_2$	1.50	0.83	0.83	0.83	0.84	0.84	0.18
	δ	1.1	0.50	0.50	0.50	0.50	0.50	1.2
350	$[D]_2$	1.95	1.08	1.09	1.09	1.09	1.10	0.24
	δ	1.00	0.5	0.5	0.5	0.5	0.5	1.35
400	$[D]_2$	2.45	1.37	1.37	1.38	1.38	1.38	0.31
	δ	1.0	0.55	0.55	0.55	0.55	0.55	1.5
450	$[D]_2$	2.99	1.67	1.68	1.69	1.69	1.70	0.38
	δ	1.1	0.6	0.6	0.6	0.6	0.6	1.65
500	$[D]_2$	3.58	2.00	2.02	2.02	2.03	2.03	0.46
	δ	1.2	0.7	0.7	0.7	0.7	0.7	1.8
550	$[D]_2$	4.21	2.36	2.37	2.38	2.38	2.39	0.54
	δ	1.3	0.8	0.8	0.8	0.8	0.8	2.0
600	$[D]_2$	4.88	2.73	2.75	2.76	2.77	2.78	0.63
	δ	1.35	0.9	0.9	0.9	0.9	0.9	2.1
650	$[D]_2$	5.59	3.13	3.15	3.16	3.17	3.18	0.73
	δ	1.45	1.0	1.0	1.0	1.0	1.0	2.3
700	$[D]_2$	6.34	3.55	3.58	3.59	3.60	3.61	0.83
	δ	1.5	1.05	1.05	1.05	1.05	1.05	2.4
750	$[D]_2$	7.13	3.99	4.02	4.03	4.04	4.06	0.93
	δ	1.6	1.15	1.15	1.15	1.15	1.15	2.6
800	$[D]_2$	7.96	4.46	4.49	4.50	4.51	4.53	1.04
	δ	1.7	1.25	1.25	1.25	1.25	1.25	2.7
850	$[D]_2$	8.83	4.94	4.97	4.99	5.00	5.02	1.15
	δ	1.75	1.35	1.35	1.35	1.35	1.35	2.85
900	$[D]_2$	9.74	5.44	5.48	5.50	5.51	5.53	1.27
	δ	1.8	1.45	1.45	1.45	1.45	1.45	3.0
950	$[D]_2$	10.68	5.96	6.01	6.02	6.04	6.06	1.39
	δ	1.9	1.55	1.55	1.55	1.55	1.55	3.1
1000	$[D]_2$	11.65	6.50	6.55	6.57	6.58	6.61	1.52
	δ	2.0	1.6	1.6	1.6	1.6	1.6	3.2
1050	$[D]_2$	12.66	7.06	7.11	7.13	7.15	7.18	1.65
	δ	2.0	1.7	1.7	1.7	1.7	1.7	3.35
1100	$[D]_2$	13.71	7.64	7.70	7.72	7.74	7.77	1.78
	δ	2.1	1.8	1.8	1.8	1.8	1.8	3.5
1150	$[D]_2$	14.79	8.24	8.30	8.32	8.34	8.37	1.92
	δ	2.15	1.9	1.9	1.9	1.9	1.9	3.6
1200	$[D]_2$	15.91	8.85	8.92	8.94	8.96	9.00	2.06
	δ	2.2	1.95	1.95	1.95	1.95	1.95	3.7
1250	$[D]_2$	17.06	9.48	9.55	9.58	9.60	9.64	2.21
	δ	2.25	2.05	2.05	2.05	2.05	2.05	3.8
1300	$[D]_2$	18.24	10.13	10.21	10.24	10.26	10.30	2.36
	δ	2.3	2.15	2.15	2.15	2.15	2.15	3.9
1350	$[D]_2$	19.45	10.80	10.88	10.91	10.94	10.98	2.51
	δ	2.4	2.2	2.2	2.2	2.2	2.2	4.0
1400	$[D]_2$	20.70	11.48	11.57	11.60	11.63	11.68	2.67
	δ	2.4	2.3	2.3	2.3	2.3	2.3	4.1
1450	$[D]_2$	21.98	12.19	12.28	12.31	12.34	12.39	2.83
	δ	2.5	2.4	2.4	2.4	2.4	2.4	4.2
1500	$[D]_2$	23.29	12.91	13.00	13.04	13.07	13.12	3.00
	δ	2.5	2.45	2.45	2.45	2.45	2.45	4.3
1550	$[D]_2$	24.64	13.64	13.74	13.78	13.82	13.87	3.17
	δ	2.6	2.5	2.5	2.5	2.5	2.5	4.4
1600	$[D]_2$	26.01	14.39	14.50	14.54	14.58	14.64	3.34
	δ	2.6	2.6	2.6	2.6	2.6	2.6	4.5
1650	$[D]_2$	27.42	15.16	15.27	15.32	15.36	15.42	3.52
	δ	2.7	2.65	2.65	2.65	2.65	2.65	4.55
1700	$[D]_2$	28.86	15.95	16.06	16.11	16.15	16.22	3.70
	δ	2.7	2.7	2.7	2.7	2.7	2.7	4.65
1750	$[D]_2$	30.33	16.75	16.87	16.92	16.96	17.03	3.88
	δ	2.75	2.8	2.8	2.8	2.8	2.8	4.75
1800	$[D]_2$	31.83	17.56	17.69	17.74	17.79	17.86	4.07
	δ	2.8	2.85	2.85	2.85	2.85	2.85	4.8
1850	$[D]_2$	33.36	18.40	18.53	18.59	18.63	18.71	4.26
	δ	2.85	2.9	2.9	2.9	2.9	2.9	4.9
1900	$[D]_2$	34.92	19.25	19.39	19.44	19.49	19.57	4.46
	δ	2.9	2.95	2.95	2.95	2.95	2.95	5.0
1950	$[D]_2$	36.51	20.11	20.26	20.32	20.37	20.45	4.65
	δ	2.9	3.0	3.0	3.0	3.0	3.0	5.1
2000	$[D]_2$	38.13	20.99	21.14	21.20	21.26	21.34	4.86
	δ	2.95	3.1	3.1	3.1	3.1	3.01	5.15

TABLE 6. Thermal-Diffusion Coefficient α_T and Estimates of the Error δ

T, K	α_T; δ, %	Molar fraction x(Ar)						
		0	0.2	0.4	0.5	0.6	0.8	1
1	2	3	4	5	6	7	8	9
200	α_T	0.44	0.32	0.26	0.24	0.23	0.20	0.13
	δ	12.5	6.00	6.1	6.1	6.15	6.2	24.0
250	α_T	0.46	0.36	0.30	0.28	0.26	0.22	0.21
	δ	8.9	3.7	3.75	3.75	3.75	3.8	12.0
300	α_T	0.47	0.39	0.32	0.30	0.28	0.24	0.28
	δ	6.9	3.0	2.9	2.9	2.85	2.8	9.4
350	α_T	0.47	0.40	0.34	0.31	0.29	0.26	0.33
	δ	5.8	3.0	2.85	2.8	2.75	2.65	8.2
400	α_T	0.47	0.42	0.35	0.32	0.30	0.26	0.36
	δ	5.2	3.3	3.1	3.0	3.0	2.85	7.7
450	α_T	0.47	0.43	0.36	0.33	0.31	0.27	0.39
	δ	4.9	3.8	3.5	3.4	3.3	3.2	7.7
500	α_T	0.47	0.43	0.36	0.34	0.31	0.28	0.41
	δ	4.7	4.2	3.9	3.7	3.6	3.5	7.8
550	α_T	0.47	0.44	0.37	0.34	0.32	0.28	0.43
	δ	4.7	4.5	4.2	4.1	3.9	3.8	8.0
600	α_T	0.47	0.44	0.37	0.34	0.32	0.28	0.45
	δ	4.7	4.9	4.5	4.35	4.2	4.0	8.2
650	α_T	0.47	0.45	0.37	0.35	0.32	0.28	0.46
	δ	4.75	5.2	4.8	4.6	4.5	4.25	8.4
700	α_T	0.47	0.45	0.38	0.35	0.32	0.29	0.47
	δ	4.8	5.5	5.05	4.9	4.7	4.45	8.6
750	α_T	0.47	0.45	0.38	0.35	0.33	0.29	0.48
	δ	4.9	5.8	5.3	5.1	4.9	4.65	8.8
800	α_T	0.47	0.45	0.38	0.35	0.33	0.29	0.49
	δ	4.95	6.00	5.5	5.3	5.1	4.9	9.0
850	α_T	0.46	0.45	0.38	0.35	0.33	0.29	0.49
	δ	5.0	6.25	5.7	5.5	5.3	5.0	9.2
900	α_T	0.46	0.45	0.38	0.35	0.33	0.29	0.50
	δ	5.1	6.5	5.9	5.65	5.5	5.15	9.3
950	α_T	0.46	0.46	0.38	0.35	0.33	0.29	0.50
	δ	5.1	6.7	6.1	5.8	5.6	5.3	9.5
1000	α_T	0.46	0.46	0.38	0.35	0.33	0.29	0.51
	δ	5.2	6.9	6.2	6.0	5.8	5.4	9.6
1050	α_T	0.46	0.46	0.38	0.36	0.33	0.29	0.51
	δ	5.3	7.1	6.4	6.1	5.9	5.5	9.74
1100	α_T	0.46	0.46	0.38	0.36	0.33	0.29	0.51
	δ	5.3	7.2	6.5	6.3	6.0	5.7	9.9
1150	α_T	0.46	0.46	0.38	0.36	0.33	0.29	0.51
	δ	5.4	7.4	6.7	6.4	6.2	5.8	10.0
1200	α_T	0.46	0.46	0.38	0.36	0.33	0.29	0.52
	δ	5.4	7.6	6.8	6.5	6.3	5.9	10.1
1250	α_T	0.46	0.46	0.38	0.36	0.33	0.29	0.52
	δ	5.50	7.71	6.94	6.64	6.38	5.97	10.22
1300	α_T	0.46	0.46	0.38	0.36	0.33	0.29	0.52
	δ	5.55	7.85	7.06	6.75	6.49	6.06	10.33
1350	α_T	0.46	0.46	0.39	0.36	0.33	0.29	0.52
	δ	5.60	8.00	7.18	6.86	6.59	6.15	10.43
1400	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.52
	δ	5.65	8.13	7.29	6.97	6.69	6.24	10.53
1450	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.70	8.26	7.40	7.07	6.78	6.32	10.62
1500	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.75	8.39	7.51	7.17	6.87	6.40	10.71
1550	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.80	8.51	7.61	7.26	6.96	6.47	10.79
1600	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.84	8.63	7.71	7.35	7.04	6.55	10.87
1650	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.88	8.75	7.80	7.44	7.12	6.62	10.95
1700	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.93	8.86	7.89	7.52	7.20	6.69	11.03
1750	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	5.97	8.97	7.98	7.60	7.28	6.75	11.10
1800	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	6.01	9.08	8.07	7.68	7.35	6.81	11.17
1850	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	6.05	9.18	8.15	7.76	7.42	6.88	11.24
1900	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	6.08	9.28	8.24	7.83	7.49	6.94	11.30
1950	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	6.12	9.38	8.31	7.91	7.56	6.99	11.36
2000	α_T	0.45	0.46	0.39	0.36	0.33	0.29	0.53
	δ	6.16	9.47	8.39	7.98	7.62	7.05	11.43

TABLE 7. Comparison of Experimental [28] and Calculated Values of the Interdiffusion Coefficient (p = 1 atm, $x(H_2)$ = 0.99)

Diffusion coefficients	T, K						
	200	230	260	280	300	310	340
D_{12} [28]	.4033	.5155	.6408	.7312	.8240	.8696	1.0221
D_{12} [99]	.4110	5266	.6538	.7435	.8380	.8873	1.0412
D_{12} [our data]	.4016	.515	.640	.729	.822	.870	1.022

nificant digits or with a step of 0.05%. The pattern of change in the errors of the reference data presented in the indicated tables is an independent object for analysis of the properties of the gaseous mixture.

In the works of Iranian authors [99, 100] devoted to generalization of the transport properties of a mixture of the rarefied gases Ar and H_2, the centrally-symmetric interaction potentials were determined within the framework of the broadened principle of corresponding states [47, 48]. The collision integrals $\Omega^{(11)*}$ and $\Omega^{(22)*}$ for argon were used as the basis of comparison. In [99], tables of calculated values of the viscosity and the interdiffusion and thermal-diffusion coefficients at 200–3273 K and the heat conductivity at a temperature of up to 1273 K are presented. In [100], the parameters of the Morse–Spline–Van der Waals function of the $U_{\text{Ar–H}_2}$ potential and tables of the transport properties of an Ar–H_2 mixture determined at x = 0.5 are presented. The multistage iteration procedure of determination of interaction potentials of substances, realized in [99, 100], was oriented first at all, to the use of experimental data on their viscosity. However, the question of how and with which weights data on the interdiffusion coefficient should be considered remains open. The authors of these works do not give estimates of the errors in the data calculated by them.

For analysis of the quality of the results of [99], in Table 7 experimental data on the interdiffusion coefficient of an Ar–H_2 mixture (δ = 0.2–0.3%) [28] are compared with the corresponding calculation data. It is seen that the calculated values [99] are systematically larger than the experimental ones, and their deviations exceed the errors of the experimental data by several times. In the tables of [100], the values of the thermal-diffusion coefficient determined at x = 0.5 are presented; they first increase with increase in the temperature, pass through the maximum α_T = 0.458 in the region of 625 K, and then decrease sharply to 0.202 at 1273 K. Such behavior of the thermal-diffusion coefficient is in contradiction with the commonly accepted view of the temperature dependence of the thermal-diffusion coefficient of mixtures of neutral gases as well as with the analogous data presented in [99]. Special calculations carried out within the framework of our project with the use of the $U_{\text{Ar–H}_2}$ potential [100] has shown that the data obtained agree with the corresponding tabulated data within 1–2% for the viscosity and within 7–9% for the interdiffusion coefficient and did not provide support for the existence of a maximum of α_T at temperatures lower than 1273 K.

For a mixture of rarefied gases Ar and H_2, tables of recommended GSSSD reference data on the interdiffusion coefficient at T = 294 K were developed for all concentrations of the components [101]. The errors of these data were estimated at 5%. The following values of the interdiffusion coefficient were obtained: D_{12} = 0.845 cm^2/sec for the mixture at $x(H_2) \to 0$ and D_{12} = 0.674 cm^2/sec for the mixture at $x(H_2) \to 1$. This sharp change in the interdiffusion coefficient on the isotherm is in conflict with the known facts, and the interdiffusion-coefficient value calculated for the quasi-Lorentzian mixture is lower by approximately 25% than its experimental values [28, 91, 92] and the data presented in Table 5.

In a series of works the nonspherical interaction potential $U_{\text{Ar–H}_2}(R, \varphi)$ was determined on the basis of spectroscopic data and data on the elastic and inelastic scattering of molecular beams [102, 103]. In this works the following values of the parameters of the spherically symmetric portion of the potential function were obtained: $d_{12} \sim$ 3.2 A and $\varepsilon_{12}/k \sim$ 73 K. Calculation of the collision integrals and the transport properties of gases in which the interaction of particles is determined by the nonspherical potentials represents an independent problem. In [102], two values of the interdiffusion coefficient, 0.809 and 0.813 cm^2/sec, were calculated at T = 300 K and normal pressure; these values were lower by approximately 1% than the corresponding experimental data [28, 90, 91] measured with an error of ~0.2–0.3% (see also Table 5). The simultaneous quantitative analysis of physical and thermophysical data is a direction of our further investigations.

CONCLUSIONS

1. The molecular-kinetic theory of rarefied gaseous mixtures and their particle interaction can be used to advantage as a basis for generalization of the transport properties of these mixtures in the temperature range investigated in the present work.

2. If the normative reference data on the viscosity of argon and hydrogen are available, the procedure of verification of the properties of their mixture becomes much simpler and the data on its property become much more reliabile.

3. The available data on the viscosity of the Ar–H_2 mixture were obtained with a limited accuracy for a definite temperature range. In this case, it is precisely the procedure of simultaneous processing of data on the interdiffusion and thermal-diffusion coefficients of a gaseous mixture that allows one to reliably determine the crossed-interaction potential of this mixture and then to calculate the coordinated set of its properties.

4. It has been established that the three-parameter L-J (m–6) potentials can be used to advantage for solving the problem considered. However, because of the limited number of parameters of this potential function, the estimation of its completeness from the standpoint of probability representations of the mathematical statistics remains an open question. In the future, the use of a larger number of interaction potentials having, in part, more flexible repulsive branches for analysis and improvement of the properties of rarefied gaseous mixtures seems promising.

5. The generalization of experimental data on the thermal-diffusion coefficient carried out within the framework of the theoretical model of elastic collisions of atoms and molecules did not reveal marked variances between these data in the process of their approximation. However, it should be noted that the relaxation characteristics of the collisions of atoms with molecules and of molecules with each other are as yet imperfectly understood. This limits the analysis and discrimination of theoretical models for calculating the heat conductivity and the thermal-diffusion coefficient of gaseous mixtures with account for inelastic collisions of atoms and molecules in them. Investigations in this direction should be continued.

6. The information obtained on the Ar–H_2 system and the experience gained in work with this system are planned to use for generalization of the properties of the Kr–H_2 and Xe–H_2 mixtures.

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NOTATION

a, vector of the potential-function parameters; a_3, a_4, additional parameters of a potential; b, coefficient of a polynomial; C^*, ratio between the collision integrals; D_{12}, interdiffusion (binary diffusion) coefficient, cm^2/sec; D^T, thermal-diffusion coefficient of a binary mixture, m^2/(sec·K); $\mathbf{D}(\mathbf{a})$, errors matrix of parameters; d_{ij}, collision diameter of the potential function, A; E, kinetic collision energy of particles; f_D, correction for the interdiffusion coefficient determined in the first approximation of the molecular-kinetic theory; k, Boltzmann constant; L, Lorentzian mixture at a concentration of the light component $x_2 \to 0$; m, exponent of the repulsive branch of the Lennard-Jones potential; n, exponent of the temperature dependence of the interdiffusion coefficient; p, pressure, atm; P, probability; $Q^{(l,s)}$, collision integral, A^2, l = 1, 2, s = 1, 2; $Q^{(l,s)*}$, reduced collision integral; q, separation factor; qL, quasi-Lorentzian mixture at a concentration of the heavy component $x_1 \to 0$; R, distance between the centers of mass in the interaction potential; R_e, equilibrium distance in the potential function; s, estimate of the mean-square error; S, sum of weighted squares of deviations; $s(x)$, mean-square error; t_P, quantile of the composite function of the error distribution; T, temperature, K; $T^* = T/\varepsilon_{12}$, reduced temperature; U, particle interaction potential; x, mole fraction of a mixture component; y_{kl}, calculated point l in the data array k; Y_{kl}, experimental point l in the data array k; W_{kl}, weights of experimental points; Z, rotational collision number; α_T, thermal-diffusion coefficient; δ, relative error, %; $\Delta_P(\mathbf{y})$, total error at a probability coefficient P; ε (or ε/k, parameter of the potential function of the intermolecular interaction, J (or K); η, viscosity, μPa·sec; λ, heat conductivity, W/(m·K); $\lambda_{\text{mix}}^{\text{mon}}$, heat conductivity of a mixture of components interacting under the action of real potentials but having no internal degrees of freedom, W/(m·K); λ_i, heat conductivity of the ith component calculated for molecules with account for the energy exchange between the translational and internal degrees of freedom, W/(m·K); φ, angle function of the nonspherical potential; μ, reduced molecular mass of particles; μ_i, dipole moment of a molecule; $\Omega^{(l,s)*}$, reduced collision integral. Subscripts: *i*, *j*, mixture components; *k*, *l*, point *l* in the data array *k*; mix, mixture; rot, rotational; vib, vibrational; mon, with no account for the internal degrees of freedom; av, average; *, reduced; e, equilibrium; 0, initial value.

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