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TRANSFER COEFFICIENT OF MULTICOMPONENT AIR WITH SUBLIMATION
 PRODUCTS OF GRAPHITE

V. V. Ryabov

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An economical method of calculating the viscosity, thermal conductivity, binary diffusion coefficient, and thermodiffusion coefficient is proposed, for a mixture of dissociated air and subliming graphite.

1. In investigating the heat transfer in a high-temperature viscous shock layer close to a graphite surface which is undergoing breakdown, the transfer coefficients (viscosity, heat conduction, diffusion) of multicomponent air with an admixture of the sublimation products of graphite must be determined.

Calculations of the characteristic parameters of molecular transfer both by kinetic theory [1] and by approximate methods [2, 3] are based on data regarding the elastic interaction potentials between the mixture components. Experimental investigation of the transfer coefficients to date has been restricted to the range $T \leq 2000$ K [4, 5], and, as noted in [6], rigorous quantum-mechanical calculation of elastic-interaction processes is only possible for atoms and molecules with a simple electron structure [7]. The basic sources of information on the interaction of molecules at high temperatures are experimental results on the scattering of fast molecular beams. Numerous data on the molecule-molecule and atom-molecule interaction were obtained in [8-11], for example. The first investigations [11] were based on the assumption of an inverse power dependence of the interaction potential $V = K/R^s$ on the distance between the centers of mass of the molecules. At the same time, analysis of the experimental conditions in [9] shows that s is not constant. Evidently, the most preferable form of repulsive potential is the exponential approximation $V = A \exp(-\beta R)$. This approximation has been successfully used to describe the interaction of identical atoms (especially of inert gases [9]). The additive-potential method, proposed in [11] for approximate calculation of $V(R)$, gives good accuracy in calculating the parameters of the atom-molecule and molecule-molecule interaction. According to this method, the interaction potential of the molecules AB and CD is written as follows [9]

$$V(R) = V(r_{AC}) + V(r_{BC}) + V(r_{AD}) + V(r_{BD}), \quad (1)$$

where r is the interatomic distance.

The potential in Eq. (1), averaged over equiprobable orientations, determines the effective spherically symmetric potential corresponding to the point force center. Here, it is possible to choose parameters of the exponential function approximating cumbersome analytical expressions for the mean potential. This procedure was described in detail in [12,

13]. The combinatorial rule [9] is usually employed to make up the currently unknown data on the interaction potential

$$V_{ij} = \sqrt{V_{ii}V_{jj}}. \quad (2)$$

Using the values of the collision integrals $\Omega_{ij}^{(\ell,s)}$ [1, 14], the transfer coefficients of the multicomponent gas mixture may be calculated by the well-known method [1, 3, 15]. According to [14], the expression for the $\Omega^{(\ell,s)}$ integrals calculated for the exponential repulsive potential $V(R) = A \exp(-\beta R)$ takes the form

$$\Omega^{(\ell,s)} = 4 \left(\frac{\pi kT}{2} \right)^{1/2} I^{(\ell,s)}(\gamma) \gamma^2 / \beta^2, \quad \gamma = \ln \left(\frac{A}{kT} \right). \quad (3)$$

Here k is the Boltzmann constant; the function $I^{(\ell,s)}(\gamma)$ for various ℓ, s and values $3.5 \leq \gamma \leq 28.5$ was tabulated in [14]. In the present work, these dependences are approximated by cubic splines of defect 1.

Using Eq. (3), it is simple to calculate the viscosity values η_i of the individual components and the binary-diffusion coefficient \mathcal{D}_{ij} [1, 14]

$$\eta_i = \frac{1,3346 \cdot 10^{-6} (M_i T)^{1/2}}{\gamma^2 / \beta^2 I^{(2,2)}(\gamma)}, \quad \text{N} \cdot \text{sec} / \text{m}^2, \quad (4)$$

$$\frac{\rho \mathcal{D}_{ij}}{M} = \frac{0,40038 \cdot 10^{-6} \beta^2}{\gamma^2 I^{(1,1)}(\gamma)} \sqrt{\frac{2T_i(M_i + M_j)}{M_i M_j}}, \quad \text{kmole} / \text{m} \cdot \text{sec}. \quad (5)$$

Note also that the viscosity and self-diffusion coefficient are simply related [1]

$$\frac{\rho \mathcal{D}_{ii}}{\eta_i} = \frac{6}{5} A^*, \quad A^* = \frac{\Omega^{(2,2)*}}{\Omega^{(1,1)*}}, \quad (6)$$

$$\Omega^{(\ell,s)*} = \frac{\Omega^{(\ell,s)}}{\Omega_{\text{TB.cfp}}^{(\ell,s)}} = \frac{8\gamma^2 / \beta^2 I^{(\ell,s)}(\gamma)}{(s+1)! \left[1 - \frac{1}{2} \frac{1 + (-1)^\ell}{1+l} \right] \sigma^2}, \quad (7)$$

where σ is the intermolecular-potential parameter of solid spheres [1].

2. Use of the additive-potential method [1] permits approximate calculation of the dependences $V(R)$ by the above method for pairs of molecules C, O, N, O₂, N₂, NO, CN, CO, CO₂, C₂, C₃, C₄, and C₅ forming the gas mixture of dissociated air and breakdown products of graphite in the range $0.6 \leq R \leq 5.5 \text{ \AA}$. The results obtained are approximated by an exponential function, using the least-square method. The basis functions adopted are the atom-atom interaction potentials, the parameters of which are taken from [6, 13]. Data on the molecular structure are taken from [16].

The values of the parameters A, β of the exponential repulsive potential obtained in the present work are shown in Table 1. These values are compared with the data of [9, 13]. The agreement of the results is basically good (an asterisk marks correlating values in Table 1). Most of the information on the interaction potentials (especially with the participation of C, O atoms) is complete, thanks to the additive-potential method used here.

For dissociated air with the ablation products of graphite at temperatures $T = 2000-10,000 \text{ K}$, the values of the reduced $\Omega_{ij}^{(\ell,s)*}$ integrals are calculated, as well as the viscosity and binary-diffusion coefficient, according to Eqs. (3)-(7). The best agreement of the present results on $\Omega_{ij}^{(\ell,s)*}$ is with [6, 9, 13]. Here the values of the collision integrals calculated using the potentials with the parameters in Table 1 are intermediate between the results of other works; see the bibliography in [6]. These differences in the given temperature range may reach 50%.

The viscosity values η_i obtained by the given method for the individual components (Table 2) are compared with the data for O₂, N₂, CO₂ from [4]. With a relative error of around 10%, the results are in good correlation. Comparison of the present data with those of [17] for η_i, \mathcal{D}_{ij} reveals only qualitative agreement. Note that, in [17], the Lennard-Jones potential was used; this potential is of strictly limited applicability, as noted in [15], for example. As an example, values of $\rho \mathcal{D}_{ij} / M$ at $T = 4000 \text{ K}$ are shown in Table 3.

TABLE 1. Parameter Values of Exponential Protential

Interacting molecules		A, eV	$\beta, \text{\AA}^{-1}$	Interacting molecules		A, eV	$\beta, \text{\AA}^{-1}$
C	C	0,3480E+03	0,3370E+01	N ₂	N ₂ *	0,4157E+04	0,2573E+01
C	O	0,7005E+03	0,3755E+01	N ₂	NO	0,5238E+02	0,1761E+01
C	N	0,1730E+03	0,3025E+01				
C	O ₂	0,1997E+04	0,3659E+01	N ₂	CN	0,6217E+03	0,2724E+01
C	N ₂	0,3985E+03	0,2956E+01	N ₂	CO	0,2217E+02	0,1432E+01
C	NO	0,9476E+03	0,3330E+01	N ₂	CO ₂	0,4738E+04	0,3118E+01
C	CN	0,5874E+03	0,3109E+01	N ₂	C ₂	0,9748E+03	0,2890E+01
C	CO	0,1368E+04	0,3496E+01	N ₂	C ₃	0,1945E+04	0,2760E+01
C	CO ₂	0,4843E+04	0,3500E+01	N ₂	C ₄	0,2983E+03	0,1855E+01
C	C ₂	0,9069E+03	0,3276E+01	N ₂	C ₅	0,9161E+03	0,1946E+01
C	C ₃	0,2076E+04	0,3117E+01	NO	NO*	0,2678E+04	0,3303E+01
C	C ₄	0,3055E+03	0,2060E+01	NO	CN	0,1568E+04	0,3084E+01
C	C ₅	0,1603E+04	0,2264E+01	NO	CO*	0,4075E+04	0,3473E+01
O	O*	0,1410E+04	0,4140E+01	NO	CO ₂	0,1613E+05	0,3510E+01
O	N	0,3482E+03	0,3410E+01	NO	C ₃	0,2871E+02	0,1515E+01
O	O ₂ *	0,4530E+04	0,4039E+01	NO	C ₃	0,7526E+02	0,1649E+01
O	N ₂ *	0,8604E+03	0,3331E+01	NO	C ₄	0,1947E+04	0,2380E+01
O	NO*	0,2142E+04	0,3717E+01	NO	C ₅	0,5863E+03	0,1781E+01
O	CN	0,1302E+04	0,3488E+01	CN	CN	0,9506E+03	0,2879E+01
O	CO	0,3095E+04	0,3882E+01	CN	CO	0,2371E+04	0,3253E+01
O	CO ₂	0,1310E+05	0,3879E+01	CN	CO ₂	0,7868E+04	0,3268E+01
O	C ₂	0,2032E+04	0,3654E+01	CN	C ₂	0,1523E+04	0,3047E+01
O	C ₃	0,5815E+04	0,3498E+01	CN	C ₃	0,3329E+04	0,2921E+01
O	C ₄	0,1580E+05	0,3310E+01	CN	C ₄	0,8944E+03	0,2162E+01
O	C ₅	0,2517E+04	0,2351E+01	CN	C ₅	0,2374E+04	0,2167E+01
N	N*	0,8600E+02	0,2680E+01	CO	CO*	0,6280E+04	0,3646E+01
N	O ₂ *	0,9057E+03	0,3320E+01	CO	CO ₂ *	0,2494E+05	0,3654E+01
N	N ₂ *	0,1849E+03	0,2614E+01	CO	C ₂	0,3836E+04	0,3418E+01
N	NO*	0,4286E+03	0,2983E+01	CO	C ₃	0,1009E+05	0,3285E+01
N	CN	0,2715E+03	0,2771E+01	CO	C ₄	0,1771E+04	0,2345E+01
N	CO	0,6157E+03	0,3149E+01	CO	C ₅	0,1125E+05	0,2531E+01
N	CO ₂	0,1855E+04	0,3156E+01	CO ₂	CO ₂	0,1049E+06	0,3666E+01
N	C ₂	0,4116E+03	0,2937E+01	CO ₂	C ₂	0,1414E+05	0,3440E+01
N	C ₃	0,9713E+02	0,1978E+01	CO ₂	C ₃	0,3537E+05	0,3297E+01
N	C ₄	0,4308E+03	0,2210E+01	CO ₂	C ₄	0,1423E+05	0,2666E+01
N	C ₅	0,2466E+03	0,1785E+01	CO ₂	C ₅	0,1079E+05	0,2303E+01
O ₂	O ₂	0,1485E+05	0,3964E+01	C ₂	C ₂	0,2431E+04	0,3209E+01
O ₂	N ₂	0,2316E+04	0,3267E+01	C ₂	C ₃	0,5807E+04	0,3084E+01
O ₂	NO*	0,6373E+04	0,3644E+01	C ₂	C ₄	0,1183E+05	0,2904E+01
O ₂	CN	0,3600E+04	0,3416E+01	C ₂	C ₅	0,1529E+04	0,2021E+01
O ₂	CO	0,9683E+04	0,3808E+01	C ₃	C ₃	0,1036E+04	0,2218E+01
O ₂	CO ₂	0,4268E+05	0,3822E+01	C ₃	C ₄	0,3750E+04	0,2316E+01
O ₂	C ₂	0,6001E+04	0,3587E+01	C ₃	C ₅	0,2704E+04	0,1998E+01
O ₂	C ₃	0,1623E+05	0,3433E+01	C ₄	C ₄	0,1870E+04	0,1927E+01
O ₂	C ₄	0,1011E+04	0,2169E+01	C ₄	C ₅	0,4640E+04	0,1939E+01
O ₂	C ₅	0,1312E+05	0,2550E+01	C ₅	C ₅	0,7891E+02	0,9718E+00

TABLE 2. Viscosity Values of Components $\eta_i \cdot 10^4, \text{N}\cdot\text{sec}/\text{m}^2$

T, K	C	O	N	O ₂	N ₂	NO	CN	CO	CO ₂	C ₂	C ₃	C ₄	C ₅
2000	0,770	0,957	0,791	0,781	0,655	0,727	0,645	0,723	0,572	0,627	0,442	0,338	0,222
3000	1,319	1,587	1,424	1,252	1,116	1,193	1,079	1,172	0,899	1,030	0,738	0,557	0,400
6000	1,825	2,149	2,039	1,656	1,539	1,603	1,468	1,561	1,175	1,385	1,003	0,752	0,575
8000	2,309	2,672	2,654	2,024	1,942	1,982	1,833	1,918	1,422	1,714	1,251	0,932	0,750
10000	2,780	3,171	3,275	2,367	2,333	2,340	2,183	2,253	1,651	2,025	1,489	1,104	0,928

3. The use of accurate expressions for the viscosity, thermal conductivity, and multi-component-diffusion coefficient of a mixture of complex gas composition on the basis of the kinetic theory of gases [1, 15] may be inapplicable because of the great consumption of machine time for the calculation. Considerably simpler expressions may be obtained using the bifurcational approximation for the binary-diffusion coefficients [2, 3, 18]. According to [2, 3, 18], the binary-diffusion coefficient may be approximately written in the form

$$\mathcal{D}_{ij} = \frac{D}{F_i F_j}, \quad (8)$$

where $D = D(p, T)$ is the parameter of the given multicomponent mixture, and $F_i(T)$ is the parameter of the i -th component. The possibility of using the correlation relation in Eq. (8) was analyzed in [2, 3] for various mixtures of complex composition. The results of the present work also indicate high accuracy of the correlation in Eq. (8) for an air mixture containing the sublimation products of graphite.

TABLE 3. Accurate (I) and Approximate (II) Values of $\rho \mathcal{D}_{ij}/2M \cdot 10^5$ (kmole/m·sec) for a Mixture with $T = 4000$ K

Interacting molecules		I	II	Interacting molecules		I	II
C	C	0,8165	0,8438	N ₂	N ₂	0,2953	0,3008
C	O	0,7733	0,7740	N ₂	NO	0,2850	0,2931
C	N	0,7953	0,8134	N ₂	CN	0,3001	0,3043
C	O ₂	0,4958	0,4839	N ₂	CO	0,2900	0,2991
C	N ₂	0,5095	0,5038	N ₂	CO ₂	0,2105	0,2092
C	NO	0,5009	0,4909	N ₂	C ₂	0,3050	0,3061
C	CN	0,5089	0,5098	N ₂	C ₃	0,2113	0,2123
C	CO	0,5068	0,5010	N ₂	C ₄	0,1512	0,1546
C	CO ₂	0,3556	0,3505	N ₂	C ₅	0,1157	0,1168
C	C ₂	0,5077	0,5126	NO	NO	0,2874	0,2856
C	C ₃	0,3509	0,3556	NO	CN	0,2960	0,2965
C	C ₄	0,2511	0,2589	NO	CO	0,2936	0,2915
C	C ₅	0,1872	0,1956	NO	CO ₂	0,2027	0,2039
O	O	0,7230	0,7099	NO	C ₂	0,2907	0,2982
O	N	0,7495	0,7461	NO	C ₃	0,2022	0,2069
O	O ₂	0,4520	0,4438	NO	C ₄	0,1451	0,1506
O	N ₂	0,4721	0,4621	NO	C ₅	0,1071	0,1138
O	NO	0,4602	0,4502	CN	CN	0,3038	0,3079
O	CN	0,4706	0,4675	CN	CO	0,3025	0,3027
O	CO	0,4658	0,4596	CN	CO ₂	0,2122	0,2117
O	CO ₂	0,3181	0,3215	CN	C ₂	0,3074	0,3097
O	C ₂	0,4695	0,4702	CN	C ₃	0,2129	0,2148
O	C ₃	0,3150	0,3262	CN	C ₄	0,1529	0,1564
O	C ₄	0,2207	0,2374	CN	C ₅	0,1157	0,1182
O	C ₅	0,1610	0,1794	CO	CO	0,2993	0,2975
N	N	0,7745	0,7842	CO	CO ₂	0,2064	0,2081
N	O ₂	0,4729	0,4665	CO	C ₂	0,3051	0,3044
N	N ₂	0,4831	0,4857	CO	C ₃	0,2075	0,2112
N	NO	0,4764	0,4732	CO	C ₄	0,1473	0,1537
N	CN	0,4845	0,4914	CO	C ₅	0,1094	0,1162
N	CO	0,4833	0,4830	CO ₂	CO ₂	0,1423	0,1456
N	CO ₂	0,3405	0,3379	CO ₂	C ₂	0,2141	0,2129
N	C ₂	0,4856	0,4942	CO ₂	C ₃	0,1464	0,1477
N	C ₃	0,3355	0,3428	CO ₂	C ₄	0,1056	0,1075
N	C ₄	0,2450	0,2496	CO ₂	C ₅	0,0792	0,0813
N	C ₅	0,1857	0,1886	C ₂	C ₂	0,3101	0,3114
O ₂	O ₂	0,2775	0,2775	C ₂	C ₃	0,2147	0,2161
O ₂	N ₂	0,2916	0,2889	C ₂	C ₄	0,1557	0,1573
O ₂	NO	0,2835	0,2815	C ₂	C ₅	0,1153	0,1189
O ₂	CN	0,2935	0,2923	C ₃	C ₃	0,1498	0,1499
O ₂	CO	0,2888	0,2873	C ₃	C ₄	0,1116	0,1091
O ₂	CO ₂	0,1967	0,2010	C ₃	C ₅	0,0856	0,0825
O ₂	C ₂	0,2956	0,2940	C ₄	C ₄	0,0843	0,0794
O ₂	C ₃	0,1987	0,2039	C ₄	C ₅	0,0655	0,0600
O ₂	C ₄	0,1396	0,1485	C ₅	C ₅	0,0509	0,0454
O ₂	C ₅	0,1030	0,1122				

TABLE 4. Parameters for Approximate Calculation of Binary-Diffusion Coefficients

Molecules	C	O	N	O ₂	N ₂	NO	CN	CO	CO ₂	C ₂	C ₃	C ₄	C ₅
F_i	0,573	0,625	0,595	1,0	0,961	0,986	0,950	0,966	1,381	0,944	1,361	1,869	2,473

The values of F_i for the given mixture are found by the least-squares method for the values of the binary-diffusion coefficients calculated from Eq. (5). The values of $D(p, T)$ for O_2 were taken as the normalizing quantities. Table 4 gives the values of F_i for the system O-N-C with an exponential repulsive potential. The parameters F_i depend weakly on the temperature. Analysis shows that, when $2000 \leq T \leq 10,000$ K, for the given gas mixture, F_i are constant, to an accuracy of 3.5%.

The results of comparing the approximate values obtained using Eq. (8) with the accurate values in Eq. (5) are shown in Table 3 for $T = 4000$ K. The mean relative error is no more than 3%, and the maximum error of the complex $\rho \mathcal{D}_{ij}/M$ is 11.5%, which corresponds to the interaction O-C₅, C₅-C₅, etc.; these interactions are rarely realized in the given conditions. Note also that the given model leads to a mean absolute error of the value of $\rho \mathcal{D}_{ij}/M$ more than an order of magnitude less than for the model of equal diffusion coefficients, which confirms the conclusions of [2, 3]. Thus, the use of the approximate correlational dependence in Eq. (8) is entirely valid.

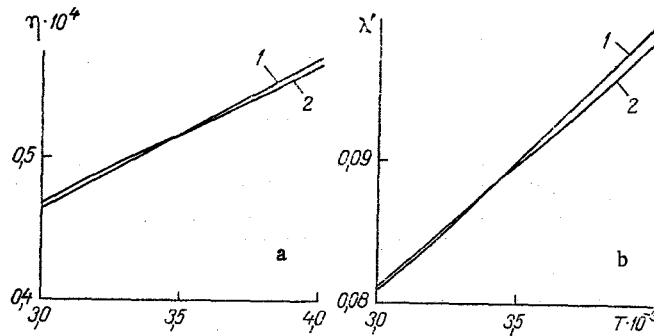


Fig. 1. Dependence of the viscosity η (a) and "frozen" thermal conductivity (b) on the temperature: 1) $p = 10^4$ Pa; 2) 10^6 . η , N·sec/m²; λ , W/m·K; T , K.

4. Following [3], the expression for the diffusional flux j_i is written in the form

$$j_i + D_i^T \nabla T = - \frac{\rho D}{\mu_1 M} [\mu_2 \nabla z_i + (z_i - \alpha_i) \nabla \mu_2 + \mu_p \nabla \ln p],$$

$$z_i = \frac{M_i x_i}{\mu_2 F_i}, \quad x_i = \frac{M}{M_i} \alpha_i, \quad \mu_1 = \sum_{j=1}^v \frac{\alpha_j F_j}{M_j} M, \quad (9)$$

$$\mu_2 = \sum_{j=1}^v \frac{M_j x_j}{F_j}, \quad \mu_p = \mu_2 \left[z_i \left(1 - \frac{M_i}{M} \right) - \alpha_i \left(1 - \sum_{j=1}^v \frac{M_j z_j}{M} \right) \right].$$

Here D_i^T is the thermodiffusion coefficient, for which the following approximate relation is assumed to be valid below [3]

$$D_i^T = c_t \frac{\rho D}{\mu_1 M} \mu_2 (z_i - \alpha_i). \quad (10)$$

After simple algebraic manipulations, Eq. (9) is brought to the form

$$j_i = - \frac{\eta}{Sc_i} \left[\nabla \alpha_i + \alpha_i \sum_{j=1}^{v-1} (R_{iv} - R_{ij}) \nabla \alpha_j + \hat{\mu}_p \nabla \ln p + \hat{c}_{ti} \nabla \ln T \right]. \quad (11)$$

In deriving Eq. (11), it is taken into account that

$$\frac{\rho D}{\mu_1 F_i} = \frac{\eta}{Sc_i}; \quad \hat{\mu}_p = \frac{F_i}{M} \mu_p; \quad \hat{c}_{ti} = c_t \left(1 - \frac{F_i \mu_2}{M} \right) \alpha_i, \quad R_{ij} = \frac{F_i}{F_j} + \left(1 - \frac{F_i \mu_2}{M} \right) \frac{M}{M_j}.$$

Equation (11) expresses the diffusional flux of the i -th component in terms of the mass-concentration gradients using the effective Schmidt numbers Sc_i and the modified thermodiffusional relations \hat{c}_{ti} . Using Eq. (11) permits significant reduction in the time of the gas-dynamic calculations and the use of simple iterative algorithms.

5. The use of Eq. (8) in combination with approximate methods of calculating the transfer coefficients (for example, the methods of Uilki and Saksen [15]) permits significant simplification of the calculation of the viscosity η and the "frozen" thermal conductivity λ' of the multicomponent gas mixture. According to [3], the following formulas are used for η and λ'

$$\eta = \frac{\rho D \mu_2}{\mu_1 b M}, \quad b = 1,473; \quad (12)$$

$$\lambda' = \lambda'_0 + \lambda_{in}, \quad (13)$$

$$\lambda'_0 = \frac{15}{4} R_g \frac{\rho D}{\mu_1 M b} \sum_{j=1}^v \frac{x_j F_j}{1,065 - \frac{x_j F_j}{\mu_1} 0,065}, \quad (14)$$

$$\lambda_{in} = \frac{\rho D}{\mu_1 M} R_g \sum_{k=1}^v \frac{x_k}{F_k} c_{in,k}. \quad (15)$$

In calculating the thermal conductivity of multiatomic gases λ' , the contribution of the internal degrees of freedom of the molecules λ_{in} is taken into account by introducing the Eucken correction in Eq. (15) [1, 3].

The results of calculating the viscosity and thermal conductivity of equilibrium-dissociated air with the sublimation products of graphite (the elements O, N, C are taken in the proportion 10.625:48.571:12.5 mole/kg) using Eqs. (12)-(15) for temperatures $T = 3000$ - 4000 K are shown in Fig. 1 for pressures $p = 10^4$ and 10^6 Pa, respectively. The equilibrium concentrations of the 13 mixture components are determined by the method of [19]. According to the estimates in [3, 15], the difference in the results of accurate [1] and approximate approaches, over the whole temperature range, is no more than 5% for the viscosity and 10% for the "frozen" thermal conductivity. The given results indicate the possibility of using the approximate method to obtain the transfer coefficients of mixtures of complex composition to an accuracy acceptable for practical purposes.

NOTATION

ρ , density; p , pressure; T , temperature; M , molecular weight; x_i , molar concentration of i -th component; α_i , mass concentration of i -th component; V , interaction potential; \mathcal{D}_{ij} , binary-diffusion coefficient; D_i^T , thermodiffusion coefficient; η , viscosity; λ' , "frozen" thermal conductivity; R , distance between centers of mass of molecules; $\Omega_{ij}^{(l,s)}$, collision integrals; R_g , universal gas constant.

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