Correlations

Viscosity, Second *pVT* Virial Coefficient, and Diffusion of Binary Mixtures of Small Alkanes CH_4, C_2H_6, C_3H_8, *n*-C_4H_{10}, *i*-C_4H_{10}, *n*-C_5H_{12}, *i*-C_5H_{12}, and $C(CH_3)_4$ Predicted by Means of an Isotropic Temperature-Dependent Potential

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The isotropic (*n*-6) Lennard-Jones temperature-dependent potential (LJTDP) together with the Hohm−Zarkova−Damyanova mixing rules is used to predict second interaction *pVT* virial coefficients $B_{AB}(T)$, interaction viscosities $\eta_{AB}(T)$, and diffusion coefficients $D_{AB}(T)$ for all 28 binary mixtures of the alkanes CH_4, C_2H_6, C_3H_8, *n*-C_4H_{10}, *i*-C_4H_{10}, *n*-C_5H_{12}, *i*-C_5H_{12}, and $C(CH_3)_4$ at low densities. Where possible, the obtained equilibrium and transport data are compared to existing measurements. In general, good agreement is found between experiment and theory. On the basis of these findings, fitting formulas and tables are presented which allow for a fast and reliable estimation of the aforementioned properties in the temperature range between (180 and 1200) K.

Introduction

It is nowadays impossible to overestimate the importance of multicomponent fluid mixtures containing alkanes since a dominant portion of methane and other lower alkanes can be found in any natural gas. It is, therefore, not astonishing that different journals like the *Journal of Natural Gas Chemistry* or *Natural Gas & Electricity* are specialized on technological, physical, and chemical aspects of natural gas behavior. Not only due to the broad range of different compositions of natural gas, reliable simulation techniques for predicting the thermophysical properties of these multicomponent mixtures are of the utmost importance.[1–6] In this context, it becomes clear that experimental and theoretical laboratory studies on the thermophysical behavior of pure alkanes and their well-defined mixtures provide indispensable basic knowledge. Since the costs of computer resources continuously decrease, there are many attempts to obtain, e.g., diffusion coefficients, liquid−vapor equilibrium curves, or second cross virial coefficients of binary, ternary, or even quaternary alkane mixtures by different simulation and estimation techniques.[1,7–10] Most of these theoretical methods are based on a detailed description of the intermolecular interactions which are responsible for thermophysical properties like the second *pVT* virial coefficient B, viscosity η, or the diffusion coefficient D. To handle the vast number of different binary mixtures containing alkanes, mixing rules of the intermolecular interaction potential parameters are often used which relate the unlike interaction AB between two particles A and B to the like interactions AA and BB.[11] Although it is now accepted that the very simple Lorentz−Berthelot mixing rules do not give the correct potential parameters of unlike interactions, they are still widely used because in some cases they nevertheless allow for a reliable prediction of some thermophysical properties of binary mixtures.[12–19] Despite their partial success, in many cases extensions of the Lorentz−Berthelot mixing rules or completely different combination rules give better results for binary alkane mixtures.[20–25] In our recent work on the thermophysical properties of low-density binary alkane mixtures, we have developed a flexible extension of the physically reasonable Tang−Toennies mixing rules.[25,26] This new scheme is called Hohm−Zarkova−Damyanova (HZD) mixing rules. We have shown that in general these more sophisticated HZD mixing rules work better compared to the old Lorentz−Berthelot combination scheme as long as the thermophysical properties B, η, and D of binary mixtures are considered.[25] The present paper is a considerable extension of our preliminary study on the binary mixtures of small alkanes.[25] Here, a complete study of second interaction *pVT* virial coefficients $B_{AB}(T)$, interaction viscosities $\eta_{AB}(T)$, and binary diffusion coefficients $D_{AB}(T)$ for all 28 binary mixtures of the alkanes CH_4, C_2H_6, C_3H_8, *n*-C_4H_{10}, *i*-C_4H_{10}, *n*-C_5H_{12}, *i*-C_5H_{12}, and $C(CH_3)_4$ is presented. These properties are obtained via the HZD mixing rules applied to the intermolecular interaction potential parameters of the pure alkanes which are tabulated by Zarkova et al.[27] It should be mentioned that contrary to most of the other studies our potential model is able to account simultaneously for transport and equilibrium thermophysical properties.

Theoretical Section

Calculation of B_{AB}, η_{AB}, and D_{AB}. To obtain the properties B_{AB}, η_{AB}, and D_{AB} of the binary mixtures, we rely on our model of the Lennard-Jones temperature-dependent potential (LJTDP). The LJTDP is explained in detail in Hohm and Zarkova.[28] Briefly, the intermolecular interaction energy $U_{AB}(R, T)$ between two particles A and B is described via

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$$U_{AB}(R,T) = \frac{\varepsilon_{AB}^{(eff)}(T)}{n_{AB}-6}\left[6\left(\frac{R_{mAB}^{(eff)}(T)}{R}\right)^{n_{AB}} - n_{AB}\left(\frac{R_{mAB}^{(eff)}(T)}{R}\right)^{6}\right] \quad (1)$$

where $R_{mAB}^{(eff)}(T)$ is an effective equilibrium distance; $\varepsilon_{AB}^{(eff)}(T)$ is the potential well-depth at temperature T; and n_{AB} is the repulsive parameter and R the center-of-mass distance. For like interactions, we have A = B. In our model, the temperature dependence of $R_{mAA}^{(eff)}(T)$ is due to vibrational excitation of the intramolecular modes of the molecule. A statistical mechanical analysis of this excitation leads to the formulation $R_{mAA}^{(eff)}(T) = R_{mAA}^{(eff)}(T = 0\text{ K}) + \delta_0 \cdot f(T)$, where $R_{mAA}^{(eff)}(T = 0\text{ K})$ and δ_0 (the first vibrationally excited level enlargement) are two of the four temperature-independent parameters of the LJTDP. The function $f(T)$ can be calculated for any T from the known vibrational frequencies of the molecule.[28] The other two temperature-independent parameters of the LJTDP are n_{AA} and $\varepsilon_{AA}^{(eff)}(T = 0$ K). Once $R_{mAA}^{(eff)}(T)$, $\varepsilon_{AA}^{(eff)}(T)$, and n_{AA} are known for the pure substances, the parameters $R_{mAB}^{(eff)}(T)$, $\varepsilon_{AB}^{(eff)}(T)$, and n_{AB} describing the interaction between unlike molecules can be deduced via mixing rules. In this work, we use our recently developed Hohm−Zarkova−Damyanova (HZD) mixing rules.[25] For the unlike interaction parameters we have

$$n_{AB} = \frac{n_{AA}+n_{BB}}{2} \quad (2)$$

$$\left(R_{mAB}^{(eff)}(T)\right)^{n_{AB}-6} = \frac{\left\{0.5\left[\left(\varepsilon_{AA}^{(eff)}(T)\right)^{\phi}\left(R_{mAA}^{(eff)}(T)\right)^{\psi} + \left(\varepsilon_{BB}^{(eff)}(T)\right)^{\phi}\left(R_{mBB}^{(eff)}(T)\right)^{\psi}\right]\right\}^{1/\phi}}{\left(\varepsilon_{AA}^{(eff)}(T)\varepsilon_{BB}^{(eff)}(T)\right)^{1/2}\left(R_{mAA}^{(eff)}(T)R_{mBB}^{(eff)}(T)\right)^{3}} \quad (3)$$

with

$$\phi = \frac{1}{1+n_{AB}}, \quad \psi = \frac{n_{AB}}{1+n_{AB}} \quad (4)$$

and

$$\varepsilon_{AB}^{(eff)}(T) = \left(\varepsilon_{AA}^{(eff)}(T)\varepsilon_{BB}^{(eff)}(T)\right)^{1/2} \cdot \frac{\left(R_{mAA}^{(eff)}(T)R_{mBB}^{(eff)}(T)\right)^{3}}{\left(R_{mAB}^{(eff)}(T)\right)^{6}} \frac{2\alpha_A\alpha_B\left(C_6^{AA}C_6^{BB}\right)^{1/2}}{C_6^{AA}\alpha_B^2 + C_6^{BB}\alpha_A^2} \quad (5)$$

where α_A and α_B are the dipole-polarizabilities and C_6^{AA} and C_6^{BB} are the dispersion-interaction energy constants of molecules A and B, respectively. These quantities can be obtained from direct measurements and experimentally determined dipole−oscillator strength distributions. For the alkanes under study, precise values or at least very reliable estimates of α and C_6 are available.[25]

The measurable quantities of the binary mixtures are the second mixture pVT virial coefficient $B_{mix}(T)$, the viscosity of the mixture $\eta_{mix}(T)$, and the binary diffusion coefficient $D_{AB}(T)$. They are related to the corresponding properties describing the like (AA and BB) and unlike (AB) interactions via[11]

$$B_{mix}(T) = B_{AA}(T)x_A^2 + 2B_{AB}(T)x_Ax_B + B_{BB}(T)x_B^2 \quad (6)$$

where for our spherically symmetric potential energy model we have for both, A = B and A ≠ B

$$B_{AB}(T) = -2\pi N_A\int_0^{\infty}\left(\exp\left(-\frac{U_{AB}(R,T)}{k_BT}\right)-1\right)R^2dR \quad (7)$$

k_B and N_A being Boltzmann's and Avogadro's constants, respectively. Furthermore, we use

$$\eta_{mix} = \frac{1+Z_\eta}{X_\eta + Y_\eta} \quad (8)$$

with

$$X_\eta = \frac{x_A^2}{\eta_{AA}} + \frac{2x_Ax_B}{\eta_{AB}} + \frac{x_B^2}{\eta_{BB}} \quad (9)$$

$$Y_\eta = \frac{3}{5}A_{AB}^{*}\left\{\frac{x_A^2}{\eta_{AA}}\frac{M_A}{M_B} + \frac{2x_Ax_B}{\eta_{AB}}\left(\frac{(M_A+M_B)^2}{4M_AM_B}\right)\left(\frac{\eta_{AB}^2}{\eta_{AA}\eta_{BB}}\right) + \frac{x_B^2}{\eta_{BB}}\frac{M_B}{M_A}\right\} \quad (10)$$

$$Z_\eta = \frac{3}{5}A_{AB}^{*}\left\{x_A^2\frac{M_A}{M_B} + 2x_Ax_B\left[\frac{(M_A+M_B)^2}{4M_AM_B}\left(\frac{\eta_{AB}}{\eta_{AA}} + \frac{\eta_{AB}}{\eta_{BB}}\right) - 1\right] + x_B^2\frac{M_B}{M_A}\right\} \quad (11)$$

In eqs 6 to 11, x_A and x_B are the mole fractions of the components A and B with molar masses M_A and M_B. η_{AA} and η_{BB} are the respective viscosities of the pure components A and B. $A_{AB}^{*} = \Omega_{AB}^{(2,2)*}/\Omega_{AB}^{(1,1)*}$ is the ratio of the reduced collision integrals $\Omega_{AB}^{(1,1)*}$ and $\Omega_{AB}^{(2,2)*}$. The latter ones are necessary for the description of the viscosity η_{AB} and diffusion coefficients D_{AB}

$$\eta_{AB} = \frac{5}{16\pi N_A\sigma_{AB}^2(T)\Omega_{AB}^{(2,2)*}(T^*)}\sqrt{2\pi k_BN_AT\frac{M_AM_B}{M_A+M_B}} \quad (12)$$

$$D_{AB} = \frac{3}{5}\frac{k_BN_AT}{p}\frac{M_A+M_B}{M_AM_B}A_{AB}^{*}\eta_{AB} \quad (13)$$

wnere $T^* = k_BT/\varepsilon_{AB}^{(eff)}(T)$ is the reduced temperature and $\sigma_{AB}(T) = R_{mAB}^{(eff)}(T)(6/n_{AB})^{1/(n_{AB}-6)}$.

Results and Discussion

Description of Tables with $R_{mAB}^{(eff)}(T)$, $\varepsilon_{AB}^{(eff)}(T)$, $B_{AB}(T)$, $\eta_{AB}(T)$, and $D_{AB}(T)$. The potential parameters $R_{mAB}^{(eff)}(0) \equiv R_{mAB}^{(eff)}(T = 0\text{ K})$, $\varepsilon_{AB}^{(eff)}(0) \equiv \varepsilon_{AB}^{(eff)}(T = 0\text{ K})$, and n_{AB} for the pure alkanes (A = B) and their binary mixtures (A ≠ B) are presented in Tables 1, 2, and 3, respectively. The calculated potential parameters $R_{mAB}^{(eff)}(T)$ and $\varepsilon_{AB}^{(eff)}(T)$ as well as the recommended thermophysical properties $B_{AB}(T)$, $\eta_{AB}(T)$, and $D_{AB}(T)$ are given for all 28 binary mixtures in the temperature range between (180 and 1200) K as Supporting Information. For a fast evaluation and compact representation, these calculated properties $R_{mAB}^{(eff)}(T)$, $\varepsilon_{AB}^{(eff)}(T)$, $B_{AB}(T)$, $\eta_{AB}(T)$, and $D_{AB}(T)$ are presented in the form of fitting formulas. $R_{mAB}^{(eff)}(T)$ is fitted to the dimensionless expression

$$\left[R_{mAB}^{(eff)}(T) - R_{mAB}^{(eff)}(0)\right]/(10^{-10}\text{ m}) = A_1\exp(-A_2/(T/\text{K})) + A_3\exp(-A_4/(T/\text{K})) \quad (14)$$

In the case of the dimensionless properties $P(T) \equiv [\varepsilon_{AB}^{(eff)}(T)/k_B]/\text{K}$, $\eta_{AB}(T)/(\mu\text{Pa}\cdot\text{s})$, and $10^6D_{AB}(T)/(\text{m}^2\cdot\text{s}^{-1})$, we use the fitting formula

$$P(T) = \sum_{i=1}^{5}A_i(T/\text{K})^{i-1} \quad (15)$$

whereas the most suitable function for the dimensionless second interaction virial coefficient was found to be

Table 1. Potential Parameters at $T = 0$ K for Pure Alkanes and Their Mixtures: Equilibrium Distance $10^{10}R_{\mathrm{mAB}}^{(\mathrm{eff})}$ ($T = 0$ K)/m

	CH_4	C_2H_6	C_3H_8	n-C_4H_{10}	i-C_4H_{10}	n-C_5H_{12}	i-C_5H_{12}	$C(CH_3)_4$
CH_4	3.868	4.158	4.399	4.576	4.616	4.682	4.703	4.783
	C_2H_6	4.447	4.689	4.866	4.905	4.971	4.992	5.072
		C_3H_8	4.930	5.107	5.147	5.213	5.234	5.314
			n-C_4H_{10}	5.284	5.324	5.390	5.411	5.491
				i-C_4H_{10}	5.363	5.429	5.450	5.530
					n-C_5H_{12}	5.495	5.516	5.596
						i-C_5H_{12}	5.537	5.617
							$C(CH_3)_4$	5.697

Table 2. Potential Parameters at $T = 0$ K for Pure Alkanes and Their Mixtures: Potential Well Depth ($\varepsilon_{\mathrm{AB}}^{(\mathrm{eff})}(T = 0\ \mathrm{K})/k_{\mathrm{B}}$)/K

	CH_4	C_2H_6	C_3H_8	n-C_4H_{10}	i-C_4H_{10}	n-C_5H_{12}	i-C_5H_{12}	$C(CH_3)_4$
CH_4	220.78	283.56	324.99	362.69	349.86	411.00	388.29	369.69
	C_2H_6	364.18	417.39	465.81	449.34	527.86	498.70	474.80
		C_3H_8	478.38	533.87	515.00	605.00	571.56	544.18
			n-C_4H_{10}	595.80	574.74	675.17	637.87	607.30
				i-C_4H_{10}	554.42	651.30	615.32	585.83
					n-C_5H_{12}	765.12	722.84	688.20
						i-C_5H_{12}	682.90	650.18
							$C(CH_3)_4$	619.02

Table 3. Potential Parameters for Pure Alkanes and Their Mixtures: Repulsive Parameter n_{AB}

	CH_4	C_2H_6	C_3H_8	n-C_4H_{10}	i-C_4H_{10}	n-C_5H_{12}	i-C_5H_{12}	$C(CH_3)_4$
CH_4	21.63	21.96	22.88	21.37	22.18	26.06	22.24	29.46
	C_2H_6	22.28	23.20	21.69	22.50	26.38	22.56	29.78
		C_3H_8	24.12	22.61	23.42	27.30	23.48	30.70
			n-C_4H_{10}	21.10	21.91	25.79	21.97	29.19
				i-C_4H_{10}	22.72	26.60	22.78	30.00
					n-C_5H_{12}	30.48	26.66	33.88
						i-C_5H_{12}	22.84	30.06
							$C(CH_3)_4$	37.28

$$B_{\mathrm{AB}}(T)/(\mathrm{cm}^3\cdot\mathrm{mol}^{-1}) = \sum_{i=1}^{4} A_i(T/\mathrm{K} - A_5)^{1-i} \qquad (16)$$

The fitting parameters for all 28 binary mixtures are given in Tables 4 and 5.

Comparison with Available Experimental Data of B_{AB}, B_{mix}, η_{AB}, η_{mix}, and D_{AB}. In Table 6, reference is given to $N = 697$ available experimental data points of B_{AB}, B_{mix}, η_{AB}, η_{mix}, and D_{AB} for 20 binary mixtures measured by different methods between 1931 and 2001. The experimental data of the second *pVT* virial coefficient B_{AB} and B_{mix} are taken from the compilation of Dymond et al.[29] The viscosities η_{mix} and η_{AB} were measured by Trautz and Sorg,[42] Abe et al.,[43,65] Bicher and Katz,[51] Giddings et al.,[52] Kestin and Yata,[55] and Küchenmeister et al.[63] The tabulated diffusion coefficients in Table 6 are either obtained by direct measurements (Trautz and Müller,[44] Gotoh et al.,[47] Gover,[46] Arora et al.[48]) or recalculated from measured viscosities via eq 13. This method of recalculation is widely used to obtain diffusion coefficients. It compensates the scarcity of experimentally measured D_{AB} values. As discussed by Marrero and Mason,[66] the most favorable cases for obtaining D_{AB} from measured η_{AB} are those where the two components have equal masses or where the heavier component is a trace gas. The accuracy is between 1 % and 13 % and is comparable to that of the most precisely measured diffusion data. Usually such recalculated data are considered as "experimental" ones.[66]

It is obvious that some mixtures like those of CH_4 with C_2H_6 and C_3H_8 have been explored more intensively than others (e.g., i-C_4H_{10} with C_2H_6, n-C_4H_{10}, and $C(CH_3)_4$, or n-C_4H_{10} with n-C_5H_{12} and $C(CH_3)_4$). There are no measured thermophysical properties at all for eight binary mixtures: i-C_4H_{10} with n-C_5H_{12}; i-C_5H_{12} with C_2H_6, C_3H_8, n-C_4H_{10}, i-C_4H_{10}, n-C_5H_{12}, $C(CH_3)_4$; n-C_5H_{12} with $C(CH_3)_4$. Our findings are now compared to the existing measurements. For those properties and mixtures which have not been considered in our preliminary paper on the binary alkane mixtures,[25] deviation plots are given in Figures 1 to 5. The remaining deviation plots are fully presented in the Supporting Information. We will first concentrate on the directly measurable properties B_{mix} and η_{mix}, which are calculated according to eqs 6 and 12. In Figures 1 and 2 we present deviation plots B_{mix} for the mixtures of CH_4 with C_2H_6, C_3H_8, and $C(CH_3)_4$. In most cases, we observe that the deviations between experimental and calculated second virial coefficients $B_{\mathrm{mix}}^{\mathrm{exptl}} - B_{\mathrm{mix}}^{\mathrm{calcd}}$ lie inside the experimentally determined error bounds of the measurements. The same holds for η_{mix}, where our calculated values generally deviate by no more than $\pm$ 1.5 % from the measured ones.

In many experimental papers, it is common practice to report on the unlike interaction properties B_{AB} and η_{AB}. In contrast to B_{mix} and η_{mix}, they are independent of the composition of the binary mixture. However, they are not directly accessible by experiment and rely on the known properties B_{AA} and B_{BB} or η_{AA} and η_{BB}, respectively, of the pure components A and B. Therefore, their uncertainty inevitably is higher than that for B_{mix} and η_{mix}, and their values depend on the actually chosen set of B_{AA}, B_{BB}, η_{AA}, and η_{BB}, respectively. In the case of B_{AB}, we note that the deviations $B_{\mathrm{AB}}^{\mathrm{exptl}} - B_{\mathrm{AB}}^{\mathrm{calcd}}$ are nearly always negative and that $B_{\mathrm{AB}}^{\mathrm{calcd}}$ in some cases lies outside of the error bar of the experimentally obtained $B_{\mathrm{AB}}^{\mathrm{exptl}}$. This is especially the case for the mixtures CH_4–n-C_5H_{12} and CH_4–$C(CH_3)_4$. However, as already mentioned[67] and discussed in detail by Zarkova et al.,[27] the works of Strein et al.[59] and Bellm et al.[60] dealing with binary mixtures of CH_4 and $C(CH_3)_4$ contain systematic errors. Therefore, they cannot serve as a test case for our results. Except for the mixture between CH_4 and n-C_4H_{10}, the agreement between the experimentally determined η_{AB} and our calculations is within $\pm$ 1.5 %. In the case of CH_4–n-C_4H_{10}, however, the difference is up to -5 % (see Figure 3).

Table 4. Fit Parameters According to Equations 14 to 16 for Binary Alkanes Mixtures. I. Mixtures of Type $C_mH_{2m+2}-C_nH_{2n+2}$, m =1,2 and $m < n < 6$

mixture	property P	A_1	A_2	A_3	A_4	A_5	predicted accuracy of P $\Delta P = P_{exptl} - P_{calcd}$
$CH_4-C_2H_6$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.060311	604.723	0.441260	2143.783	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	276.198	$4.82846 \cdot 10^{-3}$	$-5.80560 \cdot 10^{-5}$	$2.18536 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	71.387	$4.07304 \cdot 10^{4}$	$-7.39468 \cdot 10^{5}$	$3.86892 \cdot 10^{8}$	-	-3 cm^3·mol^{-1} to -6 cm^3·mol^{-1}
	η_{AB}/μPa·s	-0.34240	$3.90434 \cdot 10^{-2}$	$-1.63679 \cdot 10^{5}$	$1.79564 \cdot 10^{-9}$	$1.03034 \cdot 10^{-12}$	-1 % to 1 %
	10^6D/m^2·s^{-1}	-0.41262	$2.78253 \cdot 10^{-3}$	$1.88155 \cdot 10^{-4}$	$-8.42940 \cdot 10^{-8}$	$2.00051 \cdot 10^{-11}$	5 %
$CH_4-C_3H_8$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.093770	450.562	0.552478	1945.876	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	303.799	$-2.04972 \cdot 10^{-2}$	$-5.81723 \cdot 10^{-5}$	$2.55141 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/ cm^3·mol^{-1}	93.237	$5.54799 \cdot 10^{4}$	$-2.38303 \cdot 10^{5}$	$6.60747 \cdot 10^{8}$	-	-8 cm^3·mol^{-1}
	η_{AB}/μPa·s	-0.18587	$3.41906 \cdot 10^{-2}$	$-1.08023 \cdot 10^{-5}$	$-2.61233 \cdot 10^{-9}$	$2.35801 \cdot 10^{-12}$	-2 % to 0 %
	10^6D /m^2·s^{-1}	-0.12684	$1.77622 \cdot 10^{-4}$	$1.58289 \cdot 10^{-4}$	$-7.36686 \cdot 10^{-8}$	$1.75540 \cdot 10^{-11}$	5 %
$CH_4-n\text{-}C_4H_{10}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.165569	338.175	0.897233	1749.189	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	322.797	$-6.94531 \cdot 10^{-2}$	$-6.42659 \cdot 10^{-5}$	$3.66784 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	122.556	$7.53145 \cdot 10^{4}$	$1.75452 \cdot 10^{6}$	$1.07872 \cdot 10^{9}$	-	-20 cm^3·mol^{-1}
	η_{AB}/μPa·s	-0.23697	$3.15769 \cdot 10^{-2}$	$-7.53059 \cdot 10^{-6}$	$-6.36256 \cdot 10^{-9}$	$3.71874 \cdot 10^{-12}$	-5 % to -1 %
	10^6D/m^2·s^{-1}	0.18279	$-3.36682 \cdot 10^{-3}$	$1.50501 \cdot 10^{-4}$	$-7.79116 \cdot 10^{-8}$	$1.94180 \cdot 10^{-11}$	-4 % to 4 %
$CH_4-i\text{-}C_4H_{10}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.134286	281.359	0.699503	1637.582	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	307.158	$-6.01840 \cdot 10^{-2}$	$-4.85833 \cdot 10^{-5}$	$2.87887 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	118.451	$6.89254 \cdot 10^{4}$	$6.07152 \cdot 10^{5}$	$8.68834 \cdot 10^{8}$	-	-20 cm^3·mol^{-1}
	η_{AB}/μPa·s	-0.29450	$3.25369 \cdot 10^{-2}$	$-1.11804 \cdot 10^{-5}$	$-2.49429 \cdot 10^{-9}$	$2.44378 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	-0.04789	$-9.24208 \cdot 10^{-4}$	$1.42803 \cdot 10^{-4}$	$-7.22957 \cdot 10^{-8}$	$1.81845 \cdot 10^{-11}$	0 % to 5 %
$CH_4-n\text{-}C_5H_{12}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.159716	331.454	0.798395	1838.710	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	357.543	$-7.81659 \cdot 10^{-2}$	$-4.04479 \cdot 10^{-5}$	$2.38127 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	134.473	$8.48517 \cdot 10^{4}$	$2.34468 \cdot 10^{6}$	$1.46061 \cdot 10^{9}$	-	-25 cm^3·mol^{-1} to -50 cm^3·mol^{-1}
	η_{AB}/μPa·s	0.12934	$2.73508 \cdot 10^{-2}$	$-3.18691 \cdot 10^{-6}$	$-8.39940 \cdot 10^{-9}$	$4.02357 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	0.17597	$-2.24520 \cdot 10^{-3}$	$1.29680 \cdot 10^{-4}$	$-6.37117 \cdot 10^{-8}$	$1.51599 \cdot 10^{-11}$	
$CH_4-i\text{-}C_5H_{12}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.182377	372.845	0.817007	1969.156	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	335.408	$-8.61813 \cdot 10^{-2}$	$-1.59342 \cdot 10^{-5}$	$1.22434 \cdot 10^{-2}$	-	
	-$B_{AB}(T)$/cm^3·mol^{-1}	128.610	$8.02048 \cdot 10^{4}$	$1.20067 \cdot 10^{6}$	$1.22221 \cdot 10^{9}$	-	-20 cm^3·mol^{-1} to 20 cm^3·mol^{-1}
	η_{AB}/μPa·s	-0.12881	$2.93876 \cdot 10^{-2}$	$-6.17583 \cdot 10^{-6}$	$-6.13524 \cdot 10^{-9}$	$3.39733 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	0.09152	$-2.08647 \cdot 10^{-3}$	$1.32503 \cdot 10^{-4}$	$-6.47368 \cdot 10^{-8}$	$1.55218 \cdot 10^{-11}$	
$CH_4-C(CH_3)_4$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.091762	443.697	0.377857	1960.703	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	318.029	$-3.32654 \cdot 10^{-2}$	$-2.13209 \cdot 10^{-5}$	$1.06617 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	120.619	$6.67405 \cdot 10^{4}$	$-1.18322 \cdot 10^{6}$	$8.01571 \cdot 10^{8}$	-	-80 cm^3·mol^{-1} to -20 cm^3·mol^{-1}
	η_{AB}/μPa·s	$4.80953 \cdot 10^{-2}$	$2.90504 \cdot 10^{-2}$	$-9.62727 \cdot 10^{-6}$	$-1.38988 \cdot 10^{-9}$	$1.68007 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	-0.21912	$2.09241 \cdot 10^{-3}$	$1.16433 \cdot 10^{-4}$	$-5.28725 \cdot 10^{-8}$	$1.26669 \cdot 10^{-11}$	-2 % to 2 %
$C_2H_6-C_3H_8$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.148290	493.130	0.988949	2021.637	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	415.477	$-2.58561 \cdot 10^{-2}$	$-1.38058 \cdot 10^{-4}$	$6.07506 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	135.473	$1.02341 \cdot 10^{5}$	$2.62243 \cdot 10^{6}$	$1.83472 \cdot 10^{9}$	9.56163	-6 cm^3·mol^{-1}
	η_{AB}/μPa·s	0.64430	$2.55691 \cdot 10^{-2}$	$9.52547 \cdot 10^{-6}$	$-1.95450 \cdot 10^{-8}$	$7.19334 \cdot 10^{-12}$	-1 % to 1 %
	10^6D/m^2·s^{-1}	0.59431	$-5.79521 \cdot 10^{-3}$	$1.14291 \cdot 10^{-4}$	$-5.16403 \cdot 10^{-8}$	$1.10555 \cdot 10^{-11}$	5 %
$C_2H_6-n\text{-}C_4H_{10}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.211386	372.235	1.321383	1840.941	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	454.593	$-9.60761 \cdot 10^{-2}$	$-1.39046 \cdot 10^{-4}$	$7.44028 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	171.415	$1.29749 \cdot 10^{5}$	$4.03662 \cdot 10^{6}$	$2.22205 \cdot 10^{9}$	18.11643	-20 cm^3·mol^{-1}
	η_{AB}/μPa·s	0.78055	$2.20107 \cdot 10^{-2}$	$1.58986 \cdot 10^{-5}$	$-2.57624 \cdot 10^{-8}$	$9.21859 \cdot 10^{-12}$	-2 % to 0 %
	10^6D/m^2·s^{-1}	0.77783	$-7.66858 \cdot 10^{-3}$	$1.05139 \cdot 10^{-4}$	$-5.03673 \cdot 10^{-8}$	$1.08163 \cdot 10^{-11}$	3 %
$C_2H_6-i\text{-}C_4H_{10}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.176903	323.041	1.116534	1765.057	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	433.325	$-8.25149 \cdot 10^{-2}$	$-1.18460 \cdot 10^{-4}$	$6.29765 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	165.568	$1.21102 \cdot 10^{5}$	$2.46970 \cdot 10^{6}$	$1.82468 \cdot 10^{9}$	18.26227	
	η_{AB}/μPa·s	0.63865	$2.34790 \cdot 10^{-2}$	$1.16949 \cdot 10^{-5}$	$-2.18879 \cdot 10^{-8}$	$8.06672 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	0.64917	$-6.45270 \cdot 10^{-3}$	$1.02202 \cdot 10^{-4}$	$-4.91889 \cdot 10^{-8}$	$1.09858 \cdot 10^{-11}$	-1 % to 6 %
$C_2H_6-n\text{-}C_5H_{12}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.203492	364.726	1.227504	1909.053	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	505.824	-0.10672	$-1.16879 \cdot 10^{-4}$	$6.09736 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	186.065	$1.43838 \cdot 10^{5}$	$4.02154 \cdot 10^{6}$	$2.51228 \cdot 10^{9}$	25.99098	-50 cm^3·mol^{-1}
	η_{AB}/μPa·s	1.03927	$1.99225 \cdot 10^{-2}$	$1.45667 \cdot 10^{-5}$	$-2.19126 \cdot 10^{-8}$	$7.48975 \cdot 10^{-12}$	
	10^6D /m^2·s^{-1}	0.64380	$-5.62162 \cdot 10^{-3}$	$8.82900 \cdot 10^{-5}$	$-3.94321 \cdot 10^{-8}$	$7.95049 \cdot 10^{-12}$	
$C_2H_6-i\text{-}C_5H_{12}$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.228300	401.436	1.251932	2007.494	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	478.720	-0.11767	$-8.14788 \cdot 10^{-5}$	$4.42540 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	180.254	$1.41482 \cdot 10^{5}$	$3.83178 \cdot 10^{6}$	$2.56554 \cdot 10^{9}$	20.58290	
	η_{AB}/μPa·s	0.86093	$2.07456 \cdot 10^{-2}$	$1.51020 \cdot 10^{-5}$	$-2.32490 \cdot 10^{-8}$	$8.10206 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	0.64534	$-6.11346 \cdot 10^{-3}$	$9.12080 \cdot 10^{-5}$	$-4.07758 \cdot 10^{-8}$	$8.30483 \cdot 10^{-12}$	
$C_2H_6-C(CH_3)_4$	$10^{10}R^{(eff)}_{mAB}(T)$/m	0.145573	487.313	0.814464	2041.884	-	
	$(\varepsilon^{(eff)}_{AB}(T)/k_B)$/K	450.455	$-4.22542 \cdot 10^{-2}$	$-9.48295 \cdot 10^{-5}$	$4.19641 \cdot 10^{-8}$	-	
	$-B_{AB}(T)$/cm^3·mol^{-1}	170.028	$1.24619 \cdot 10^{5}$	$1.243119 \cdot 10^{6}$	$1.97243 \cdot 10^{9}$	18.43639	
	η_{AB}/μPa·s	0.79169	$2.26770 \cdot 10^{-2}$	$6.82366 \cdot 10^{-6}$	$-1.46848 \cdot 10^{-8}$	$5.32409 \cdot 10^{-12}$	
	10^6D/m^2·s^{-1}	0.35965	$-2.91837 \cdot 10^{-3}$	$8.15930 \cdot 10^{-5}$	$-3.53375 \cdot 10^{-8}$	$7.47623 \cdot 10^{-12}$	0 % to 4 %

In Figures 4 and 5, two deviation plots for D_{AB} are presented for various mixtures of i-C_4H_{10} and $C(CH_3)_4$ with some of the other lower alkanes. As for the viscosities η_{AB}, we do not give any error bars to the experimental results. The stated accuracy of 1 % seems to be too small in many cases. Arora et al.[48] have measured diffusion coefficients D_{AB} in the temperature range between (275 and 323) K and gave their results in the form of fitting polynomials. All the other experimental data are given as a set of single data points. The diffusion coefficients measured by Arora et al.[48] for mixtures of CH_4 with C_2H_6, C_3H_8, and n-C_4H_{10} are well within ± 5 % of our calculations. Trautz and Müller[44] have directly measured binary diffusion coefficients D_{AB} for $CH_4-C_2H_6$, $CH_4-C_3H_8$, and $C_2H_6-C_3H_8$. Their data in general show the largest deviation from our

Table 5. Fit Parameters According to Equations 14 to 16 for Binary Alkanes Mixtures. II. Mixtures of Type $C_mH_{2m+2}-C_nH_{2n+2}$, $2 < m < 6$ and $m \leq n < 6$

mixture	property	A_1	A_2	A_3	A_4	A_5	predicted accuracy of P: $\Delta P = P_{\mathrm{exptl}} - P_{\mathrm{calcd}}$
$C_3H_8-n-C_4H_{10}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.251867	370.030	1.436982	1818.205	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	540.913	−0.14967	$-1.39033 \cdot 10^{-4}$	$8.23275 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	209.322	$1.66048 \cdot 10^{4}$	$2.86037 \cdot 10^{6}$	$2.60659 \cdot 10^{9}$	34.76575	−20 cm$^3\cdot$mol^{-1} to 10 cm$^3\cdot$mol^{-1}
	η_{AB}/μPa$\cdot$s	1.24216	$1.77053 \cdot 10^{-2}$	$2.21359 \cdot 10^{-5}$	$-2.90245 \cdot 10^{-8}$	$9.68923 \cdot 10^{-12}$	−1 % to 2 %
	10^6D/m$^2\cdot$s^{-1}	0.77914	$-7.02604 \cdot 10^{-3}$	$8.09473 \cdot 10^{-5}$	$-3.65192 \cdot 10^{-8}$	$7.20718 \cdot 10^{-12}$	2 %
$C_3H_8-i-C_4H_{10}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.217045	328.951	1.23484	1747.681	-	
	$(\varepsilon_{\mathrm{AB}}{}^{(\mathrm{eff})}(T)/k_B)$/K	516.764	−0.13207	$-1.19037 \cdot 10^{-4}$	$7.04585 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	202.812	$1.57012 \cdot 10^{5}$	$1.46761 \cdot 10^{6}$	$2.20477 \cdot 10^{9}$	33.93285	
	η_{AB}/μPa$\cdot$s	1.09383	$1.90812 \cdot 10^{-2}$	$1.85159 \cdot 10^{-5}$	$-2.59218 \cdot 10^{-8}$	$8.83681 \cdot 10^{-12}$	0 % to 1 %
	10^6D/m$^2\cdot$s^{-1}	0.67810	$-6.14821 \cdot 10^{-3}$	$7.90561 \cdot 10^{-5}$	$-3.60180 \cdot 10^{-8}$	$7.46295 \cdot 10^{-12}$	
$C_3H_8-n-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.244360	364.101	1.340241	1876.196	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	606.230	−0.16565	$-1.16577 \cdot 10^{-4}$	$6.89921 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	226.092	$1.84866 \cdot 10^{5}$	$2.278908 \cdot 10^{6}$	$3.01512 \cdot 10^{9}$	43.62552	−20 cm$^3\cdot$mol^{-1} to 0 cm$^3\cdot$mol^{-1}
	η_{AB}/μPa$\cdot$s	1.36556	$1.72174 \cdot 10^{-2}$	$1.74339 \cdot 10^{-5}$	$-2.20025 \cdot 10^{-8}$	$6.93424 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.57515	$-4.57045 \cdot 10^{-3}$	$6.55291 \cdot 10^{-5}$	$-2.59983 \cdot 10^{-8}$	$4.33487 \cdot 10^{-12}$	
$C_3H_8-i-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.269241	394.852	1.359527	1960.988	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	575.704	−0.17555	$-7.85080 \cdot 10^{-5}$	$5.05818 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	219.680	$1.82413 \cdot 10^{5}$	$2.23802 \cdot 10^{6}$	$3.03241 \cdot 10^{9}$	38.76052	
	η_{AB}/μPa$\cdot$s	1.28987	$1.71478 \cdot 10^{-2}$	$1.99663 \cdot 10^{-5}$	$-2.51236 \cdot 10^{-8}$	$8.09860 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.62680	$-5.35488 \cdot 10^{-3}$	$6.86305 \cdot 10^{-5}$	$-2.79357 \cdot 10^{-8}$	$4.93695 \cdot 10^{-12}$	−4 % to 8 %
$C_3H_8-C(CH_3)_4$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.183226	447.609	0.923166	1956.170	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	541.749	$-8.62576 \cdot 10^{-2}$	$-1.00298 \cdot 10^{-4}$	$5.02072 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	208.128	$1.64598 \cdot 10^{5}$	$1.11710 \cdot 10^{4}$	$2.43900 \cdot 10^{9}$	35.49813	
	η_{AB}/μPa$\cdot$s	1.11291	$1.97535 \cdot 10^{-2}$	$1.09628 \cdot 10^{-5}$	$-1.64984 \cdot 10^{-8}$	$5.45122 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.37353	$-2.77726 \cdot 10^{-3}$	$6.16194 \cdot 10^{-5}$	$-2.42669 \cdot 10^{-8}$	$4.45381 \cdot 10^{-12}$	0 % to 6 %
$n-C_4H_{10}-i-C_4H_{10}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.294024	310.904	1.585166	1703.787	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	583.921	−0.22477	$-1.06912 \cdot 10^{-4}$	$8.11982 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	248.839	$1.96264 \cdot 10^{5}$	$1.56997 \cdot 10^{6}$	$2.73362 \cdot 10^{9}$	44.45207	3 cm$^3\cdot$mol^{-1}
	η_{AB}/μPa$\cdot$s	1.38175	$1.53608 \cdot 10^{-2}$	$2.57124 \cdot 10^{-5}$	$-3.19082 \cdot 10^{-8}$	$1.05270 \cdot 10^{-11}$	−1 % to 1.5 %
	10^6D/m$^2\cdot$s^{-1}	0.75926	$-6.90534 \cdot 10^{-3}$	$6.99766 \cdot 10^{-5}$	$-3.21407 \cdot 10^{-8}$	$6.37922 \cdot 10^{-12}$	80 %
$n-C_4H_{10}-n-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.320039	335.980	1.679610	1797.760	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	687.859	−0.27373	$-1.03845 \cdot 10^{-4}$	$8.22331 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	277.043	$2.28759 \cdot 10^{5}$	$2.09999 \cdot 10^{6}$	$3.76262 \cdot 10^{9}$	54.91403	−40 cm$^3\cdot$mol^{-1} to −0 cm$^3\cdot$mol^{-1}
	η_{AB}/μPa$\cdot$s	1.59481	$1.45523 \cdot 10^{-2}$	$2.19296 \cdot 10^{-5}$	$-2.51451 \cdot 10^{-8}$	$7.61914 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.61311	$-4.92527 \cdot 10^{-3}$	$5.64069 \cdot 10^{-5}$	$-2.20815 \cdot 10^{-8}$	$3.30942 \cdot 10^{-12}$	
$n-C_4H_{10}-i-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.342976	357.376	1.690568	1853.496	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	653.802	−0.27831	$-6.59390 \cdot 10^{-5}$	$6.28011 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	268.546	$2.26160 \cdot 10^{5}$	$1.73610 \cdot 10^{6}$	$3.68094 \cdot 10^{9}$	50.76393	
	η_{AB}/μPa$\cdot$s	1.57267	$1.38708 \cdot 10^{-2}$	$2.60155 \cdot 10^{-5}$	$-2.96638 \cdot 10^{-8}$	$9.22298 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.66887	$-5.71271 \cdot 10^{-3}$	$5.91196 \cdot 10^{-5}$	$-2.34763 \cdot 10^{-8}$	$3.67858 \cdot 10^{-12}$	
$n-C_4H_{10}-C(CH_3)_4$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.249612	367.930	1.257716	1803.773	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	616.256	−0.17276	$-1.07592 \cdot 10^{-4}$	$6.82071 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	259.492	$2.08924 \cdot 10^{5}$	$6.84221 \cdot 10^{5}$	$3.18275 \cdot 10^{9}$	46.27762	
	η_{AB}/μPa$\cdot$s	1.32787	$1.71716 \cdot 10^{-2}$	$1.55163 \cdot 10^{-5}$	$-1.99427 \cdot 10^{-8}$	$6.28760 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.42631	$-3.33418 \cdot 10^{-3}$	$5.31666 \cdot 10^{-5}$	$-2.08888 \cdot 10^{-8}$	$3.50783 \cdot 10^{-12}$	15 %
$i-C_4H_{10}-n-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.289441	308.106	1.477818	1743.132	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	659.222	−0.24813	$-8.69952 \cdot 10^{-5}$	$7.04144 \cdot 10^{-8}$	-	
	-$B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	269.340	$2.19311 \cdot 10^{5}$	$7.70084 \cdot 10^{5}$	$3.30292 \cdot 10^{9}$	53.17794	
	η_{AB}/μPa$\cdot$s	1.48470	$1.54351 \cdot 10^{-2}$	$1.98680 \cdot 10^{-5}$	$-2.36831 \cdot 10^{-8}$	$7.33474 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.54747	$-4.40415 \cdot 10^{-3}$	$5.53739 \cdot 10^{-5}$	$-2.19303 \cdot 10^{-8}$	$3.52195 \cdot 10^{-12}$	
$i-C_4H_{10}-i-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.310289	329.400	1.484490	1798.144	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	626.700	−0.25359	$-5.095467 \cdot 10^{-2}$	$5.20987 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	261.241	$2.16438 \cdot 10^{5}$	$4.91330 \cdot 10^{5}$	$3.23621 \cdot 10^{9}$	49.04756	
	η_{AB}/μPa$\cdot$s	1.44389	$1.50035 \cdot 10^{-2}$	$2.32692 \cdot 10^{-5}$	$-2.75737 \cdot 10^{-8}$	$8.73859 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.62332	$-5.34925 \cdot 10^{-3}$	$5.86330 \cdot 10^{-5}$	$-2.41021 \cdot 10^{-8}$	$4.20325 \cdot 10^{-12}$	
$i-C_4H_{10}-C(CH_3)_4$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.214810	326.577	1.058231	1722.128	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	591.276	−0.15547	$-8.60230 \cdot 10^{-5}$	$5.63390 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	250.693	$1.98034 \cdot 10^{5}$	$-1.14411 \cdot 10^{6}$	$2.65244 \cdot 10^{9}$	45.70077	
	η_{AB}/μPa$\cdot$s	1.23062	$1.79468 \cdot 10^{-2}$	$1.36082 \cdot 10^{-5}$	$-1.84922 \cdot 10^{-8}$	$5.97038 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.36545	$-2.83760 \cdot 10^{-3}$	$5.20963 \cdot 10^{-5}$	$-2.05956 \cdot 10^{-8}$	$3.66454 \cdot 10^{-12}$	0 % to 6 %
$n-C_5H_{12}-i-C_5H_{12}$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.334716	352.822	1.594136	1906.243	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	740.929	−0.30929	$-3.76183 \cdot 10^{-5}$	$4.77696 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	290.579	$2.53884 \cdot 10^{5}$	$9.62763 \cdot 10^{5}$	$4.54644 \cdot 10^{9}$	60.26773	
	η_{AB}/μPa$\cdot$s	1.56300	$1.51099 \cdot 10^{-2}$	$1.78191 \cdot 10^{-5}$	$-1.952059 \cdot 10^{-8}$	$5.50512 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.42881	$-3.11967 \cdot 10^{-3}$	$4.53935 \cdot 10^{-5}$	$-1.43209 \cdot 10^{-8}$	$1.24297 \cdot 10^{-12}$	
$n-C_5H_{12}-C(CH_3)_4$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.242333	362.270	1.161240	1869.398	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	700.157	−0.19589	$-7.71905 \cdot 10^{-5}$	$5.18909 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	275.503	$2.35672 \cdot 10^{5}$	$-1.39337 \cdot 10^{6}$	$3.84120 \cdot 10^{9}$	56.58860	
	η_{AB}/μPa$\cdot$s	1.31571	$1.77557 \cdot 10^{-2}$	$1.00096 \cdot 10^{-5}$	$-1.26905 \cdot 10^{-8}$	$3.59608 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.27178	$-1.66062 \cdot 10^{-3}$	$4.19176 \cdot 10^{-5}$	$-1.33039 \cdot 10^{-8}$	$1.50474 \cdot 10^{-12}$	
$i-C_5H_{12}-C(CH_3)_4$	$10^{10}R^{(\mathrm{eff})}_{\mathrm{mAB}}(T)$/m	0.266823	392.880	1.1795768	1965.961	-	
	$(\varepsilon^{(\mathrm{eff})}_{\mathrm{AB}}(T)/k_B)$/K	666.093	−0.20632	$-3.85382 \cdot 10^{-5}$	$3.32327 \cdot 10^{-8}$	-	
	$-B_{\mathrm{AB}}(T)$/cm$^3\cdot$mol^{-1}	270.149	$2.31530 \cdot 10^{5}$	$-8.25448 \cdot 10^{5}$	$3.80168 \cdot 10^{9}$	51.81179	
	η_{AB}/μPa$\cdot$s	1.33391	$1.72826 \cdot 10^{-2}$	$1.28190 \cdot 10^{-5}$	$-1.58236 \cdot 10^{-8}$	$4.69431 \cdot 10^{-12}$	
	10^6D/m$^2\cdot$s^{-1}	0.29633	$-2.01732 \cdot 10^{-3}$	$4.34193 \cdot 10^{-5}$	$-1.41113 \cdot 10^{-8}$	$1.68355 \cdot 10^{-12}$	

Table 6. Set of Available Experimental Data for Thermophysical Properties of Mixtures[a]

mixture	reference	property	N	ΔT (K)
$CH_4-C_2H_6$	Michels and Nederbragt[30] (1939)	B_{AB}	3	273 to 323
		B_{mix}	12	273 to 323
	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Hoover et al.[32] (1968)	B_{AB}	3	215 to 273
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Wormald et al.[34] (1979)	B_{AB}	8	241 to 303
	Katayama et al.[35] (1980)	B_{AB}	1	298
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
	Estrada-Alexanders and Trusler[37] (1994)	B_{AB}	8	200 to 375
	Trusler[38] (1994)	B_{AB}	9	200 to 375
	McElroy and Fang[39] (1994)	B_{AB}	5	303 to 343
		B_{mix}	13	303 to 343
	Blanke and Weiss[40] (1995)	B_{AB}	7	273 to 333
	Hou et al.[41] (1996)	B_{AB}	2	300 to 320
		B_{mix}	6	300 to 320
	Trautz and Sorg[42] (1931)	η_{mix}	20	293 to 523
	Abe et al.[43] (1978)	η_{AB}	5	298 to 468
		η_{mix}	15	298 to 468
		D_{AB}	5	298 to 468
	Trautz and Müller[44] (1935)	D_{AB}	5	273 to 523
	Weissman[45] (1964)	D_{AB}	4	293 to 523
	Gover[46] (1967)	D_{AB}	1	298
	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 438
	Arora et al.[48] (1980)	D_{AB}	[b]	275 to 323
$CH_4-C_3H_8$	Trusler et al.[49] (1996)	B_{AB}	7	225 to 375
	Wormald et al.[34] (1979)	B_{AB}	10	243 to 302
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Barker and Linton[50] (1963)	B_{mix}	5	378 to 511
	Trautz and Sorg[42] (1931)	η_{mix}	12	293 to 523
	Bicher and Katz[51] (1943)	η_{mix}	20	298 to 498
	Giddings et al.[52] (1966)	η_{mix}	8	311 to 411
	Abe et al.[43] (1978)	η_{mix}	15	298 to 468
		η_{AB}	5	298 to 468
		D_{AB}	5	298 to 468
	Weissman[45] (1964)	D_{AB}	4	293 to 523
	Trautz and Müller[44] (1935)	D_{AB}	5	273 to 523
	Gotoh et al.[47] (1974)	D_{AB}	4	298 to 438
	Arora et al.[48] (1980)	D_{AB}	[b]	275 to 323
$CH_4-n\text{-}C_4H_{10}$	Wormald et al.[34] (1979)	B_{AB}	10	277 to 394
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Beattie and Stockmayer[53] (1942)	B_{AB}	7	423 to 573
	Pompe and Spurling[54] (1976)	B_{AB}	10	348 to 573
	Kestin and Yata[55] (1968)	η_{mix}	8	293 to 303
		D_{AB}	2	293 to 303
	Abe et al.[43] (1978)	η_{mix}	15	298 to 468
		η_{AB}	5	298 to 468
		D_{AB}	5	298 to 468
	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 436
	Arora et al.[48] (1980)	D_{AB}	[b]	275 to 323
$CH_4-i\text{-}C_4H_{10}$	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Gotoh et al.[47].(1974)	D_{AB}	3	298 to 438
$CH_4-n\text{-}C_5H_{12}$	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Pecsok and Windsor[56] (1968)	B_{AB}	2	298 to 323
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Massoudi and King[57] (1973)	B_{AB}	1	298
	Wormald et al.[34] (1979)	B_{AB}	9	319 to 404
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
$CH_4-i\text{-}C_5H_{12}$	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Pecsok and Windsor[56] (1968)	B_{AB}	2	298 to 323
$CH_4-C(CH_3)_4$	Hamann et al.[58] (1955)	B_{AB}	8	303 to 403
		B_{mix}	32	303 to 403
	Strein et al.[59] (1971)	B_{AB}	11	296 to 493
	Bellm et al.[60] (1974)	B_{AB}	10	300 to 550
	Baughman et al.[61] (1975)	B_{AB}	7	200 to 258
	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 436
$C_2H_6-C_3H_8$	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Fontalba et al.[62] (1988)	B_{AB}	18	274 to 356
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
	Abe et al.[43] (1978)	η_{mix}	17	298 to 468
		η_{AB}	5	298 to 468
		D_{AB}	5	298 to 468

Table 6. Continued

mixture	reference	property	N	ΔT (K)
	Trautz and Sorg[42] (1931)	η_{mix}	12	313 to 373
	Trautz and Müller[44] (1935)	D_{AB}	5	273 to 523
	Weissman[45] (1964)	D_{AB}	4	293 to 523
	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 438
	Gover[48] (1967)	D_{AB}	1	298
$C_2H_6-n\text{-}C_4H_{10}$	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Wormald et al.[34] (1979)	B_{AB}	3	305 to 363
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
	Abe et al.[43] (1978)	η_{mix}	15	298 to 468
		η_{AB}	5	298 to 468
		D_{AB}	5	298 to 468
	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 437
$C_2H_6-i\text{-}C_4H_{10}$	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 437
$C_2H_6-n\text{-}C_5H_{12}$	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Pecsok and Windsor[56] (1968)	B_{AB}	1	298
	Massoudi and King[57] (1973)	B_{AB}	1	298
	Jaeschke et al.[36] (1988)	B_{AB}	3	273 to 313
$C_2H_6-C(CH_3)_4$	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 437
$C_3H_8-n\text{-}C_4H_{10}$	Mason and Eakin[31] (1961)	B_{AB}	1	289
	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
	Jaeschke et al.[36] (1988)	B_{AB}	4	273 to 333
	Abe et al.[43] (1978)	η_{mix}	19	298 to 468
		D_{AB}	5	298 to 468
	Gotoh et al.[47] (1974)	D_{AB}	2	298 to 437
$C_3H_8-i\text{-}C_4H_{10}$	Küchenmeister et al.[63] (2001)	η_{mix}	14	298 to 626
		η_{AB}	14	298 to 626
		D_{AB}	17	298 to 626
	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 437
$C_3H_8-n\text{-}C_5H_{12}$	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
$C_3H_8-C(CH_3)_4$	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 438
$n\text{-}C_4H_{10}-i\text{-}C_4H_{10}$	Connolly[64] (1962)	B_{AB}	7	344 to 444
	Abe et al.[65] (1979)	η_{mix}	20	298 to 373
		η_{AB}	5	298 to 373
		D_{AB}	5	298 to 373
$n\text{-}C_4H_{10}-n\text{-}C_5H_{12}$	Dantzler et al.[33] (1968)	B_{AB}	4	298 to 373
$n\text{-}C_4H_{10}-C(CH_3)_4$	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 438
$i\text{-}C_4H_{10}-C(CH_3)_4$	Gotoh et al.[47] (1974)	D_{AB}	3	298 to 438

[a] N is the number of experimental data points measured in the temperature range ΔT. [b] The authors have measured binary diffusion coefficients over the temperature range (275 to 323) K at constant mole fraction and then fitted them to polynomials.

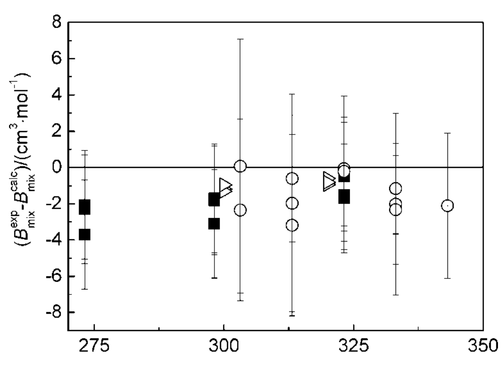

Figure 1. Deviations $B_{mix}^{exptl} - B_{mix}^{calcd}$ between experimental and calculated second pVT mixture virial coefficients of $CH_4-C_2H_6$ mixtures of different compositions: ○, McElroy and Fang;[39] ■, Michels and Nederbragt;[30] open triangle pointing right, Hou et al.[41]

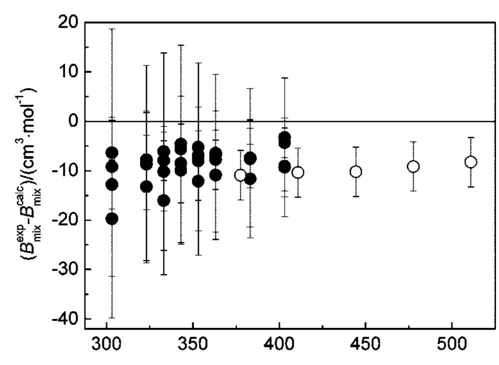

Figure 2. Deviations $B_{mix}^{exptl} - B_{mix}^{calcd}$ between experimental and calculated second pVT mixture virial coefficients: ●, Hamann et al.,[58] $CH_4-C(CH_3)_4$ mixtures of different compositions; ○, Barker and Linton,[50] equimolar $CH_4-C_3H_8$ mixture.

calculations of up to 30 %. We expect this deviation to be larger than their experimental error bars, which, however, are not given in their publication.[44] Gotoh et al.[47] have examined 14 mixtures relevant to us and presented the most comprehensive experimental study on the binary diffusion coefficients of small alkanes. They state a probable accuracy of 1 %. Their data coincide with our results to within −3 % − +10 %, except for $n\text{-}C_4H_{10}-C(CH_3)_4$ where a difference of up to 20 % is observed. The same high accuracy of 1 % was also stated by Gover.[46] His results of D_{AB} for mixtures of C_2H_6 with CH_4 and C_3H_8 do not deviate by more than 2 % from our calculations. In view of the experimentally obtained diffusion coefficients D_{AB}, our calculations are well within a range of −5 % and +10 % of the majority of the data. The agreement becomes much better if

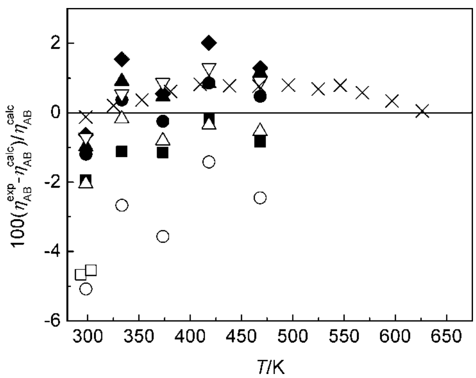

Figure 3. Relative deviations $100\cdot(\eta_{AB}^{exptl} - \eta_{AB}^{calcd})/\eta_{AB}^{calcd}$ between experimental and calculated interaction viscosities: ●, $CH_4-C_2H_6$; ■, $CH_4-C_3H_8$; ○, $CH_4-n\text{-}C_4H_{10}$; ▲, $C_2H_6-C_3H_8$; △, $C_3H_8-n\text{-}C_4H_{10}$; ◆, $i\text{-}C_4H_{10}-n\text{-}C_4H_{10}$ (all Abe et al.[43]); ▽, $C_2H_6-n\text{-}C_4H_{10}$ (Abe et al.[65]); □, $CH_4-n\text{-}C_4H_{10}$ (Kestin and Yata[55]); ×, $C_3H_8-i\text{-}C_4H_{10}$ (Küchenmeister et al.[63]).

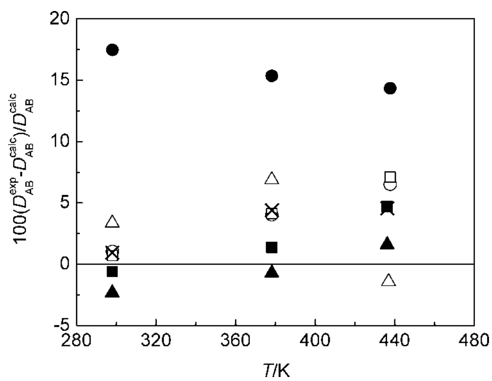

Figure 4. Relative deviations $100\cdot(D_{AB}^{exptl} - D_{AB}^{calcd})/D_{AB}^{calcd}$ between experimental[47] and calculated binary diffusion coefficients: ×, $CH_4-i\text{-}C_4H_{10}$; ▲, $CH_4-C(CH_3)_4$; △, $C_2H_6-i\text{-}C_4H_{10}$; ■, $C_2H_6-C(CH_3)_4$; □, $C_3H_8-C(CH_3)_4$; ●, $n\text{-}C_4H_{10}-C(CH_3)_4$; ○, $i\text{-}C_4H_{10}-C(CH_3)_4$.

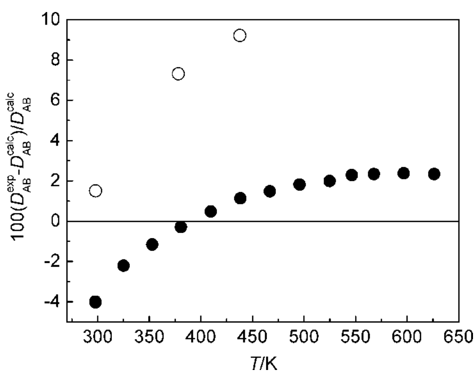

Figure 5. Relative deviations $100\cdot(D_{AB}^{exptl} - D_{AB}^{calcd})/D_{AB}^{calcd}$ between experimental and calculated binary diffusion coefficients of $C_3H_8-i\text{-}C_4H_{10}$ mixtures: ●, Küchenmeister et al.;[63] ○, Gotoh et al.[47]

we compare our results with diffusion coefficients D_{AB} recalculated from measured interaction viscosities η_{AB} according to eq 13. Abe et al.[43] have explored six mixtures and recalculated D_{AB} from their measured η_{AB}. Their results are in very good agreement with our calculations. The deviations range from ± 0.5 % for $CH_4-C_2H_6$ and $C_2H_6-n\text{-}C_4H_{10}$ up to a maximum value of 4 % for $C_3H_8-n\text{-}C_4H_{10}$. In a later work on $n\text{-}C_4H_{10}-i\text{-}C_4H_{10}$, the results of Abe et al.[65] deviate by 90 % from our calculations, although the underlying η_{AB} (Figure 3) and η_{mix} coincide with our calculated viscosities to within ± 1.5 %. We assume an erroneous calculation of D_{AB} by Abe et al.[65] Weissman[45] has used η_{mix} measured by Trautz and Sorg[42] to obtain D_{AB} for $CH_4-C_2H_6$, $CH_4-C_3H_8$, and $C_2H_6-C_3H_8$. For the latter two mixtures, his results deviate by no more than 5 % from our findings, where for the first mixture deviations between 3 % and 8 % are observed. A recent experimental work of Vogel and coworkers[63] presents 14 viscosities η_{mix} of the equimolar binary mixture $C_3H_8-i\text{-}C_4H_{10}$ measured in the range between (298 and 627) K with an unsurpassed uncertainty not higher than 0.3 %. Using these data, the authors obtained the interaction viscosity coefficients η_{AB} and the binary diffusion coefficients D_{AB} with only slightly higher uncertainty. As can be seen in Figure 5, their diffusion coefficients do agree fairly well with our calculations, and the underlying viscosities do not deviate by more than 1 % from our data presented in this work.

For those mixtures where comparison of our calculated thermophysical data to experiments is possible, we try to extract a range of confidence of our fitting formulas. Their expected accuracy is given in the last column of Tables 4 and 5. However, it is very hard to predict reliable error bounds for the missing entries. Based on our knowledge with the LJTDP, a conservative estimate of the accuracies of B_{AB}, η_{AB}, and D_{AB} is (20 to 30) $cm^3\cdot mol^{-1}$, ± 2 %, and ± 7.5 %, respectively. It seems that especially B_{mix} could be obtained with a slightly better accuracy of (10 to 20) $cm^3\cdot mol^{-1}$.

Conclusions

The isotropic (n-6) Lennard-Jones temperature-dependent potential (LJTDP) is used to calculate second interaction virial coefficients B_{AB} and interaction viscosities η_{AB}, second mixture virial coefficients B_{mix} and mixture viscosities η_{mix}, and binary diffusion coefficients D_{AB} of all 28 binary mixtures of the alkanes CH_4, C_2H_6, C_3H_8, $n\text{-}C_4H_{10}$, $i\text{-}C_4H_{10}$, $n\text{-}C_5H_{12}$, $i\text{-}C_5H_{12}$, and $C(CH_3)_4$ in the temperature range between (180 and 1200) K. The potential parameters $\varepsilon_{AB}^{(eff)}(T)$, $R_{mAB}^{(eff)}(T)$, and n_{AB} of the unlike interaction between two alkanes A and B are obtained from the corresponding parameters $\varepsilon_{AA}^{(eff)}(T)$, $R_{mAA}^{(eff)}(T)$, and n_{AA} of the pure alkanes via the Hohm−Zarkova−Damyanova (HZD) mixing rules. Our approach is checked against the limited number of experimentally obtained thermophysical properties of the binary mixtures $CH_4-C_2H_6$ (B_{AB}, B_{mix}, η_{mix}, η_{AB}, D_{AB}), $CH_4-C_3H_8$ (B_{AB}, B_{mix}, η_{mix}, η_{AB}, D_{AB}), $CH_4-n\text{-}C_4H_{10}$ (B_{AB}, η_{mix}, η_{AB}, D_{AB}), $CH_4-i\text{-}C_4H_{10}$ (B_{AB}, D_{AB}), $CH_4-n\text{-}C_5H_{12}$ (B_{AB}), $CH_4-i\text{-}C_5H_{12}$ (B_{AB}), $CH_4-C(CH_3)_4$ (B_{AB}, B_{mix}, D_{AB}), $C_2H_6-C_3H_8$ (B_{AB}, η_{mix}, η_{AB}, D_{AB}), $C_2H_6-n\text{-}C_4H_{10}$ (B_{AB}, η_{mix}, η_{AB}, D_{AB}), $C_2H_6-i\text{-}C_4H_{10}$ (D_{AB}), $C_2H_6-n\text{-}C_5H_{12}$ (B_{AB}), $C_2H_6-C(CH_3)_4$ (D_{AB}), $C_3H_8-n\text{-}C_4H_{10}$ (B_{AB}, η_{mix}, D_{AB}), $C_3H_8-i\text{-}C_4H_{10}$ (η_{mix}, η_{AB}, D_{AB}), $C_3H_8-n\text{-}C_5H_{12}$ (B_{AB}), $C_3H_8-C(CH_3)_4$, (D_{AB}), $n\text{-}C_4H_{10}-i\text{-}C_4H_{10}$ (B_{AB}, η_{mix}, η_{AB}, D_{AB}), $n\text{-}C_4H_{10}-n\text{-}C_5H_{12}$ (B_{AB}), $n\text{-}C_4H_{10}-C(CH_3)_4$ (D_{AB}), $i\text{-}C_4H_{10}-C(CH_3)_4$ (D_{AB})). In general, we observe a good agreement between our calculations and the directly measurable properties B_{mix} and η_{mix}. In the case of B_{AB} and η_{AB}, the agreement becomes slightly worse but still acceptable. For the binary diffusion coefficients D_{AB}, our calculations mostly lie outside the error bars of the directly measured properties. Very good agreement, however, is observed if the comparison is made to diffusion coefficients D_{AB} recalculated from measured interaction viscosities η_{AB}.

To conclude, we must state that there is a strong need for further experimental studies on the thermophysical properties of binary mixtures. Such studies can also help to check and improve our approach of the application of the LJTDP to mixtures.

Having now some confidence in our approach, the tabulated interaction properties of the alkanes under study and their binary mixtures will allow for a better prediction of thermophysical properties of binary and multicomponent vapor mixtures for the needs of the gas and oil industry.

Supporting Information Available:

Tables of recommended thermophysical properties of all 28 mixtures considered as well as deviation plots between experimental and calculated thermophysical properties are given. This material is available free of charge via the Internet at http://pubs.acs.org.

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