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VAPOR PRESSURE, DENSITY, VISCOSITY AND REFRACTIVE INDEX OF DIMETHYL SULFOXIDE + 1,4-DIMETHYLBENZENE SYSTEM

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This work reports the experimental results of the isothermal vapor-liquid equilibrium data between 303.15 K and 333.15 K, densities, viscosities and refractive indices from 298.15 to 323.15 K of the dimethyl sulfoxide + 1,4-dimethylbenzene system over the entire range of mixture composition. The obtained PTX data were correlated and estimated by the Wilson, NRTL and UNIFAC models. The excess Gibbs energy and activity coefficients were calculated and compared with the others excess properties. Excess molar volumes, viscosity deviations and deviations in refractivity were calculated from experimental data; all the computed quantities were fitted to the Redlich-Kister equation. The resulting excess functions were interpreted in terms of the interactions between the molecules in the binary system.

Keywords: VLE data, mixture properties, correlative and predictive methods, molecular interactions

Introduction

Continuing our work on the thermodynamic of the nonelectrolite systems, the present paper is a part of a study of the binary systems containing dimethyl sulfoxide (DMSO) and aromatic hydrocarbons, interesting as mixed solvents [1]. Dimethyl sulfoxide is a versatile nonaqueous, aprotic, highly polar self-associated solvent (having a dipole moment μ =3.90 D at 298.15 K) used extensively in kinetic studies, electrochemistry and as a solvent for polymers. Binary mixtures of DMSO with aromatic solvents are interesting in studies of polymer miscibility, polymer phase diagrams, and preferential interactions in mixed solvents [2].

This work reports isothermal experimental vapor liquid equilibrium (VLE) data at temperatures in the range from 303.15 K to 333.15 K and experimental data of density, viscosity and refractive index in the temperature range from 298.15 to 323.15 K for the dimethyl sulfoxide + 1,4-dimethylbenzene system.

The VLE data were correlated by means of Wilson [3] and NRTL [4] models; this work has also served to test the predictive capacity of the group contribution methods: original UNIFAC [5] and UNIFAC-Dortmund [6,7].

Excess molar volumes, viscosity deviations and deviations in refractivity were calculated from experimental data; all the computed quantities were fitted to the Redlich-Kister equation.

After our knowledge, VLE data for this binary system are not numerous [8]; there is only one source in the literature [9]. No literature data on densities, viscosities and refractive indices are available for this system, except on densities at 303.15 K [10].

Experimental

Materials

The used substances were purified by distillation. Dimethyl sulfoxide was distilled under vacuum at 0.8-0.9 kPa and 338.65 K. The analytical-reagent-grade 1,4-dimethylbenzene from Fluka (p.a.) was used without further purification. The purity was checked by refractive index, density and gas chromatography. It was better than 99.5 mass per cent. The experimental values of density, refractive index and viscosity of the pure components are presented in *Table 1* and compared with literature values.

Table 1. Experimental and literature values for density (ρ), refractive index (n_D) and viscosity (η) of the pure components

T, K	10^{-3} ρ, kg·m^{-3}		n_D		η, mPa·s	
	lit.	exptl	lit.	exptl	lit.	exptl
dimethyl sulfoxide						
293.15	1.10050[13], 1.10053[14]	1.10073				
298.15	1.0962[11], 1.0954[12]	1.09574	1.4770[11]	1.4768		2.0388
303.15	1.09050[15]	1.09074	1.4752[14]	1.4733	1.830[14]	1.8405
313.15	1.08046[16]	1.08075	1.4700[14]	1.4694	1.534[14]	1.5373
1,4-dimethylbenzene						
293.15	0.8610[17]	0.86144				
298.15	0.8565[19], 0.85670[20]	0.85712	1.49320[10], 1.49286[19], 1.4930[21]	1.4930	0.603[18]	0.6281

Vapor pressure

The total vapor pressure of the binary mixture was measured at temperatures between 303.15 K and 333.15 K by a static method, with a glass isoteniscope of the Smith and Menzies type, with some modifications [22], which allows the measurements of VLE at subatmospheric pressures at temperatures up to 423.15 K. Vapor pressures are measured by means of a Hg manometer and a cathetometer, readings accurate to within 0.026 kPa. The temperature was controlled by a U-10 type thermostat and measured with an Hg thermometer with an estimate accuracy of 0.05 K. The substances were degassed prior to the measurements by freezing-thawing processes under vacuum. The vapor phase volume of the apparatus being relatively small, calculation showed that the error in composition due to vaporization is within this limit.

Density, Viscosity and Refractive Index

Densities, ρ, of the pure solvents and the mixtures were measured with an Anton Paar DMA 4500 densimeter with a precision of ±0.00005 g cm^{-3}, between 298.15 and 323.15 K. The DMA cell was calibrated with ultra pure water at atmospheric pressure. The sample size was 0.7 cm^3, and the sample thermostat was controlled to ±0.01 K.

Kinematic viscosities, ν, of the pure components and their mixtures were determined at 298.15, 303.15, 313.15 and 323.15 K using an Ubbelohde capillary viscosimeter for which the uncertainty of the flow time measurement was ±0.1 s; the corresponding uncertainty in the kinematic viscosity was ± 0.001 10^{-6} m^2 s^{-1}. At least four times flow measurements were performed for each composition and temperature, and the results were averaged. The viscosimeter was calibrated using double distilled water; it was placed in a thermostat maintained constant within ± 0.05 K.

Refractive indices values for the D-line, n_D, were measured with a thermostated Abbe refractometer with a precision of ±0.0001. All measurements were done in a thermostat maintained at ± 0.05 K. The binary mixtures were prepared by mass; the precision of the mole fraction is estimated to be better than ± 0.0002.

Results and Discussion

The measurements of the total pressure were carried out at various compositions on the large temperature range. For each binary mixture the dependencies between experimental pressure and temperature were established. The functions P = *f*(T) obtained by polynomial regression were used to calculate the VLE data at 303.15 K, 313.15 K, 323.15 K and 333.15 K. The smoothed data resulted from experimental measurements are reported in *Table 2*.

Table 2 Vapor-liquid equilibrium data for the binary dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system from 303.15 to 333.15 K. Smoothed values of the pressure by polynomial regression

T, K	P, kPa			
X_1	303.15	313.15	323.15	333.15
0.0000	1.55	2.65	4.34	6.86
0.1235	1.67	2.50	4.33	7.21
0.2098	1.50	2.41	4.37	7.20
0.3029	1.19	2.19	4.00	6.93
0.4059	1.36	2.10	3.86	6.64
0.6348	1.09	1.91	3.60	6.17
0.8022	1.09	1.71	3.05	5.11
0.9078	0.74	1.40	2.35	3.59
0.9554	0.64	0.85	1.47	2.48
1.0000	0.11	0.22	0.41	0.73

The temperature dependence of the pure component vapor pressures, P_i^o, was calculated using the Antoine equation:

$$\ln P_i^o\,(kPa) = A_i - \frac{B_i}{T(K) - C_i} \qquad (1)$$

where the Antoine constants A_i, B_i and C_i are reported in *Table 3*.

Table 3 Result of correlation and prediction with Wilson, NRTL and UNIFAC models for dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system

Compound	A_i	B_i	C_i
dimethyl sulfoxide[23]	15.86168	4717.396	41.59
1,4-dimethylbenzene[24]	14.08127	3346.646	57.84

The various models are used for correlation and prediction of binary VLE data: the equations based on local composition concept (Wilson and NRTL) and the group contribution methods UNIFAC.

The results of correlation with Wilson and NRTL models are presented in *Table 4*, which includes the model parameters values and the statistical parameters: average percentage deviation in pressure, ΔP, and standard deviation, σ.

Prediction of VLE for the binary system at 303.15, 313.15, 323.15 and 333.15 K has been carried out by the original UNIFAC method and its modification, UNIFAC-Dortmund. The group interaction parameters were those published by Reid [25], Hansen [26] and Gmehling [6,7]. The results of prediction are also presented in *Table 4.*

The Wilson and NRTL give a similar and good representation of experimental data (except at 303.15 K temperature); σ and ΔP characterise the data as satisfactory.

It can be observed that the description of this system by the original UNIFAC method shows good agreement with the experimental data, like Wilson and NRTL models; the modified UNIFAC gives less satisfactory results, especially at high temperature.

The experimental and calculated (with Wilson model) total pressures vs. liquid and vapor compositions for 303.15 K, 313.15 K, 323.15 K and 333.15 K are presented in *Figure 1*. In the same phase diagram the curves represent calculated data and the points, experimental data. The activity coefficients were calculated with Wilson equation for binary systems. *Figure 2* presents the dependence of the activity coefficients of the composition at 313.15 K, the limiting values at infinite dilution of these coefficients being given. We have also computed and represented the variation of the excess Gibbs energy of the system, which shows positive deviations from the ideal. The comparison of maximum obtained value of G^E with literature, at 313.15 K, is satisfactory [9].

Table 4 Experimental values of density, kinematic viscosity and refractive index of dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system from 298.15 to 323.15 K

Model	Parameters, J mol^{-1}	Statistical parameters	
		σ [a], mmHg	ΔP [b], %
303.15 K			
Wilson	$\Delta\lambda_{12}$=4143.8± 770 $\Delta\lambda_{21}$=2412.5±1004	0.86	7.14
NRTL (α=0.3)	Δg_{12}=4067.7±1113 Δg_{21}=1442.6± 891	0.91	7.54
UNIFAC	Original UNIFAC UNIFAC-Dortmund		6.25 9.92
313.15 K			
Wilson	$\Delta\lambda_{12}$=3764.3±217 $\Delta\lambda_{21}$= 346.4±326	0.41	2.64
NRTL (α=0.3)	Δg_{12}=4444.2±309 Δg_{21}= 969.4±222	0.37	2.10
UNIFAC	Original UNIFAC UNIFAC-Dortmund		7.99 4.54
323.15 K			
Wilson	$\Delta\lambda_{12}$=6261.8±431 $\Delta\lambda_{21}$=1163.9±284	0.86	2.27
NRTL (α=0.3)	Δg_{12}=2620.0±435 Δg_{21}=3451.0±535	1.15	2.93
UNIFAC	Original UNIFAC UNIFAC-Dortmund		3.79 10.20
333.15 K			
Wilson	$\Delta\lambda_{12}$=9770.9±728 $\Delta\lambda_{21}$= 455.2±172	1.07	1.91
NRTL (α=0.3)	Δg_{12}=1649.8±343 Δg_{21}=5777.3±636	2.00	3.85
UNIFAC	Original UNIFAC UNIFAC-Dortmund		3.48 14.20

[a] Standard deviation $\sigma = \{\Sigma_i^N (P_{calc}(i) - P_{exp}(i))^2/(N-M)\}^{0.5}$ (N: number of data points and M: number of estimated parameters).

[b] Average percentage deviation in pressure $\Delta P = (100/N)\cdot\Sigma_i^N |P_{calc}(i) - P_{exp}(i)|/ P_{exp}(i)$

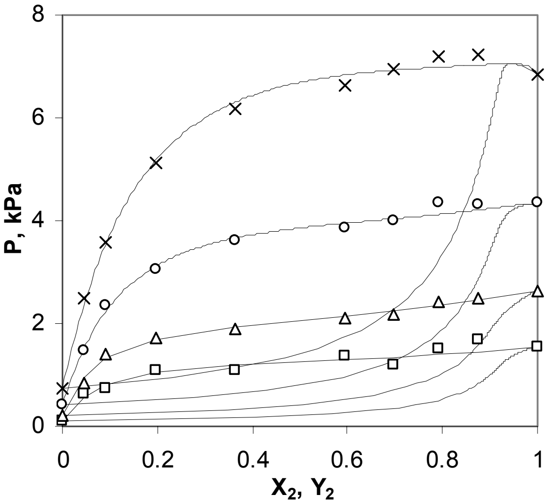

Fig. 1 Experimental vapor pressures of dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system at 303.15 K (□); 313.15 K (Δ); 323.15 K (o); 333.15 K (×); correlation with Wilson model (—)

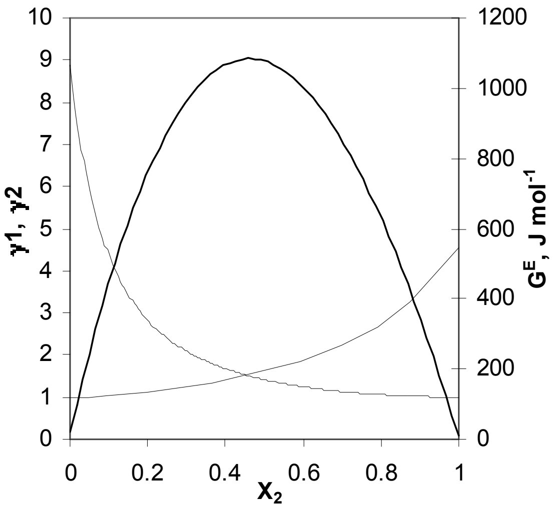

Fig. 2 Activity coefficients and excess Gibbs energy for the dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system calculated with Wilson model at 313.15 K

Densities, viscosities and refractive indices were measured in the temperature range from 298.15 to 323.15 K and the measured values were listed in *Table 5*. The values of excess molar volume, V^E, viscosity deviation, $\Delta\nu$, and deviation in molar refractivity, ΔR, were calculated from the experimental data according to the following equations:

$$V^E = V_m - \sum_{i=1}^{2} V_i X_i \tag{2}$$

$$\Delta\nu = \nu_m - \sum_{i=1}^{2} \nu_i X_i \tag{3}$$

$$\Delta R = R_m - \sum_{i=1}^{2} R_i \varphi_i \tag{4}$$

where X_i and φ_i represent the mole fraction and volume fraction, respectively; V_m, ν_m, R_m, the properties of mixtures, V_i, ν_i, R_i, the properties of the pure components. The values of R were calculated from the Lorentz-Lorenz equation [27].

The experimental values of V^E, $\Delta\nu$, and ΔR have been fitted to the Redlich-Kister [28] type polynomials:

$$Y = X_1 X_2 \sum_{k=0}^{n} A_k \left(2X_1 - 1\right)^k \tag{5}$$

where Y represent the excess molar volume, viscosity deviation and deviation in molar refractivity, and A_k represents the parameters. A nonlinear least-squares method was used to estimate the adjustable parameters A_k. The values of A_k and standard deviation σ are given in *Table 6*.

Table 5 Parameters of Eq. 5 and standard deviations of excess functions of dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system

X_1	$10^{-3}\cdot\rho$, $kg\cdot m^{-3}$	$10^{6}\cdot\nu$, $m^2\cdot s^{-1}$	n_D	X_1	$10^{-3}\cdot\rho$, $kg\cdot m^{-3}$	$10^{6}\cdot\nu$, $m^2\cdot s^{-1}$	n_D
298.15 K							
0.0000	0.85712	0.855	1.4930	0.5969	0.96896	1.236	1.4858
0.1056	0.87326	0.902	1.4927	0.7009	0.99631	1.337	1.4836
0.1975	0.88813	0.950	1.4920	0.8022	1.02625	1.447	1.4813
0.2988	0.90577	1.010	1.4908	0.8978	1.05785	1.561	1.4791
0.4033	0.92574	1.079	1.4893	1.0000	1.09574	1.698	1.4768
0.5126	0.94902	1.163	1.4875				
303.15 K							
0.0000	0.85277	0.692	1.4907	0.5969	0.96423	1.110	1.4835
0.1056	0.86880	0.745	1.4901	0.7009	0.99151	1.227	1.4813
0.1975	0.88363	0.797	1.4893	0.8022	1.02138	1.360	1.4790
0.2988	0.90121	0.861	1.4882	0.8978	1.05292	1.506	1.4765
0.4033	0.92112	0.936	1.4868	1.0000	1.09074	1.687	1.4733
0.5126	0.94434	1.028	1.4851				
313.15 K							
0.0000	0.84405	0.746	1.4854	0.5969	0.95490	1.012	1.4792
0.1056	0.85999	0.782	1.4847	0.7009	0.98205	1.079	1.4772
0.1975	0.87472	0.816	1.4840	0.8022	1.01179	1.152	1.4749
0.2988	0.89220	0.858	1.4831	0.8978	1.04318	1.227	1.4723
0.4033	0.91200	0.906	1.4820	1.0000	1.08075	1.316	1.4694
0.5126	0.93510	0.963	1.4805				
323.15 K							
0.0000	0.83526	0.578		0.5969	0.94543	0.857	
0.1056	0.85108	0.612		0.7009	0.97244	0.934	
0.1975	0.86571	0.647		0.8022	1.00203	1.020	
0.2988	0.88308	0.690		0.8978	1.03326	1.113	
0.4033	0.90276	0.740		1.0000	1.07075	1.227	
0.5126	0.92574	0.802					

Table 6

Function	A_0	A_1	A_2	A_3	σ [a]
298.15 K					
$10^6\cdot V^E$, $m^3\cdot mol^{-1}$	-0.7875	0.0235	-0.7956	0.1458	0.0007
$10^6\cdot\Delta\nu$, $m^2\cdot s^{-1}$	-0.4946	-0.0705			0.0004
$10^6\cdot\Delta R$, $m^3\cdot mol^{-1}$	-8.6332	2.0491	-0.5638	0.1739	0.0022
303.15 K					
$10^6\cdot V^E$, $m^3\cdot mol^{-1}$	-0.7718	0.0122	-0.7718	0.1051	0.0011
$10^6\cdot\Delta\nu$, $m^2\cdot s^{-1}$	-0.4112	-0.1051	0.0130		0.0002
$10^6\cdot\Delta R$, $m^3\cdot mol^{-1}$	-8.6048	2.2879	-0.7394		0.0058
313.15 K					
$10^6\cdot V^E$, $m^3\cdot mol^{-1}$	-0.7908	-0.0116	-0.8715		0.0009
$10^6\cdot\Delta\nu$, $m^2\cdot s^{-1}$	-0.3002	-0.0443			0.0002
$10^6\cdot\Delta R$, $m^3\cdot mol^{-1}$	-8.5246	2.1822	-0.8037	0.5005	0.0041
323.15 K					
$10^6\cdot V^E$, $m^3\cdot mol^{-1}$	-0.7815	-0.0221	-0.9102		0.0026
$10^6\cdot\Delta\nu$, $m^2\cdot s^{-1}$	-0.2340	-0.0220	0.0017		0.0001
$10^6\cdot\Delta R$, $m^3\cdot mol^{-1}$					

[a] Standard deviation $\sigma = \{\Sigma_i^N (Y_{calc}(i) - Y_{exp}(i))^2/(N-M)\}^{0.5}$ (N: number of data points and M: number of estimated parameters).

The dependence of excess molar volume V^E on the mole fraction of DMSO is presented in *Figure 3*. The negative values of V^E suggest the presence of specific interactions between the mixing components. V^E shows negligible temperature dependence.

Plots of excess viscosity versus the mole fraction of DMSO are shown in *Figure 4*. The $\Delta\nu$ are negative over the whole composition range; a rise of temperature makes an increase of Δν. The viscosity of a mixture depends on the molecular interactions between components: generally, mixtures with strong interactions between different molecules show positive viscosity deviations, while for mixtures without specific strong interactions, viscosity deviations are negative. In our mixtures the breaking of dipole-dipole interactions of DMSO contributes to the reduction of the viscosity during the mixing process. On the other hand, self-association of DMSO and "π electron interactions" in DMSO + 1,4-dimethylbenzene system [29] increase the viscosity, but the effect is not as important as the breaking of self-interactions. Consequently, the excess viscosity of all our mixtures is moderately negative; similarly, V^E values remain negative. The presence of intramolecular interaction is reflected in positive deviations from Raoult's law, therefore, positive values for G^E.

The excess molar refractivity is shown in *Figure 5.* The ΔR values are negative for the whole composition range for all mixtures. The values are independent of temperature, as predicted by the theory, the molar refractivity depending only on the wavelength of light used for measurement.

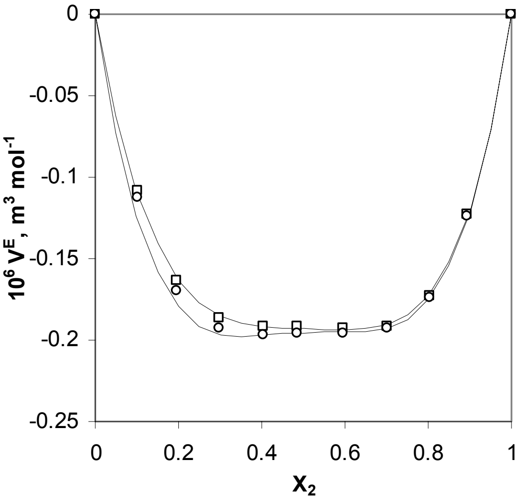

Fig. 3 Excess molar volumes for dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system at 298.15 (◊); 303.15 K (□); 313.15 K (Δ); 323.15 K (o); correlation with Redlich-Kister equation (—)

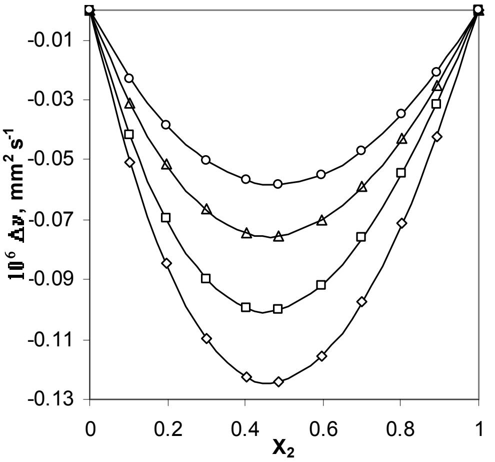

Fig. 4 Viscosity deviations for dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system for the temperatures given in Figure 3; correlation with Redlich-Kister equation (—)

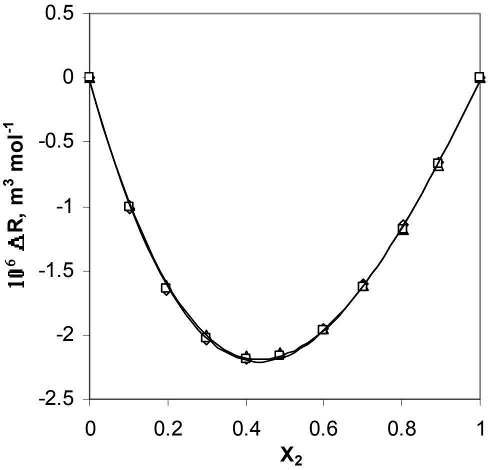

Fig. 5 Deviations in molar refractivity for dimethyl sulfoxide (1) + 1,4-dimethylbenzene (2) system for the temperatures given in Figure 3; correlation with Redlich-Kister equation (—)

Conclusions

The experimental data concerning the isothermal vapor-liquid equilibria in the binary 1,4-dimethylbenzene + DMSO system have been presented. Eight various mixtures containing 1,4-dimethylbenzene + DMSO have been analyzed. A good agreement between experimental and calculated values of the pressure has been observed for Wilson and NRTL correlative methods and for original UNIFAC- predictive method.

Experimental densities, kinematic viscosities and refractive indices data for the binary system DMSO + 1,4-dimethylbenzene have been measured at atmospheric pressure between 298.15 K and 323.15 K. Excess functions have been calculated and fitted to the Redlich-Kister equation. The behaviour of this mixture has been discussed in terms of inter and intramolecular interactions.

SYMBOLS

A_i, B_i, C_i	Antoine's equation parameters
A_k	Redlich-Kister's parameters
G^E	excess Gibbs energy
g_{ij}	parameters of NRTL equation
M	number of model parameters
N	number of experimental points
n_D	refractive index
ΔP	average percentage deviation in pressure
$P_{\exp}$	experimental total pressure
P_{calc}	computed total pressure
P_i^o	vapor pressure of *i*-th pure component
ΔR	deviation in molar refractivity
R_m	molar refractivity of mixture
R_i	molar refractivity of the *i*-th pure component
T	absolute temperature
V^E	excess molar volume
V_m	molar volume of mixture
V_i	molar volume of the *i*-th pure component
X_i	liquid-phase mole fractions
γ_k	activity coefficients
λ_{ij}	parameters of Wilson equation
μ	dipole moment
η, ν	dynamic and cinematic viscosity, respectively
$\Delta\nu$	viscosity deviation
ν_m	kinematic viscosity of mixture
ν_i	kinematic viscosity of the *i*-th pure component
ρ	density
σ	standard deviation
φ_i	volume fraction

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