

Volumetric behaviour and vapour-liquid equilibrium of dimethyl disulfide + *n*-alkanol binary mixtures

Najla Ben Mahdoui^{a,b}, Manuela Artal^b, Monia Hichri^a, Carlos Lafuente^{b,*}

^a Université de Tunis EL Manar, Faculté des Sciences, Département de Chimie, Laboratoire des Matériaux, Cristalochimie et Thermodynamique Appliquée, LR15ES01, 2092 Tunis, Tunisia.

^b Departamento de Química Física, Facultad de Ciencias, Universidad de Zaragoza, 50009 Zaragoza, Spain.

* Corresponding author, e-mail: celadi@unizar.es

Abstract

In this paper, we report densities at two temperatures (303.15 and 323.15) K and at atmospheric pressure ($p = 0.1$ MPa) of the binary mixtures containing dimethyl disulfide and a linear alkanol (methanol, ethanol, 1-propanol or 1-butanol). The isothermal vapour-liquid equilibrium for these systems was also determined at the same two temperatures while the pressure range for vapour-liquid equilibrium measurement depends on both the mixture and temperature considered. The vapour-liquid equilibrium data were found thermodynamically consistent. From experimental data excess volumes and excess Gibbs functions were obtained and correlated with composition using the Redlich-Kister polynomial expansion and the Wilson equation, respectively. The values calculated for both excess properties were positive. The excess volumes, unlike the excess Gibbs functions, increase with the length of the n -alkanol chain.

Keywords: n -Alkanols; Dimethyl disulfide; Isothermal vapour-liquid equilibrium; Volumetric behaviour.

1. Introduction

In the evolution of the oil industry [1-6], the fractional distillation has been one of the most important processes. This physical separation technique, based on the different volatility of the compounds into the mixture, has since been the key step applied to crude oil refining. During the refining, the petroleum industry products are generally subjected to hydrotreating steps. These steps involve treating petroleum feeds in the presence of a hydrotreatment catalyst to remove certain heteroatoms in the petroleum fractions. Of all the hetero-elements in crude oil, the sulfur has the most important effects on refining processes [7-10]. Hydrotreatment catalysts are used to convert organosulfur components to hydrogen sulfide gas (H_2S), a process known as hydrodesulfurization (HDS). However, these catalysts are active in hydrotreatment operations only in metal sulfides form. Therefore, the sulfurization of these catalysts, before being used, is an important step to obtain the maximum of their performances in HDS. Several practical experiments prove that the sulphurization procedure has an important effect on stability and activity of an hydrotreatment catalyst [11,12]. Hallie et al. [13] reported on these hydrogen sulphurization procedures which are performed directly in hydrotreating reactors. These different techniques of catalyst sulfidation were compared and the results show that sulphurization with a liquid feed supplemented with a spiked feedstock having the property of decomposing at low temperature is the best sulphurization technique [14]. Dimethyl disulfide (DMDS) is the most frequently used compound as sulfurizing agent due to its lower cost and higher sulfur content (68%) [15]. Worldwide, there are many hydrotreating units, which produce transportation fuels containing between 500-3000 parts per million (ppm) sulfur. Nowadays, the stringent, reduced-sulfur specifications between 10 and 15 ppm, are considered as a difficult challenge [16]. In view of this increasingly stringent environmental requirements for

sulfur content in fuels, there are much research effort has gone into improving refining procedures. For this reason, it will be necessary to increase the physicochemical properties databases of sulfur compounds used in the refining of crude oil. Among these properties, we were concentrating on the volumetric and vapour-liquid equilibrium behaviour.

In the present study, we report experimental results (densities and vapour pressures) for the binary systems: dimethyl disulfide + linear alcohol (methanol, ethanol, 1-propanol or 1-butanol). Densities were measured at two temperatures (303.15 and 323.15 K) and at atmospheric pressure ($p = 0.1$ MPa). Vapour pressures (isothermal vapour-liquid equilibrium) were determined at the same temperatures and at variable pressure range. From the experimental data we have obtained the corresponding excess molar volumes and excess Gibbs functions. These excess properties have been correlated using the Redlich-Kister expansion [17] for excess molar volume and the Wilson equation [18] for excess Gibbs function. A literature survey shows that some works related with this study have been performed. Zudkevitch et al. [19] measured the isothermal VLE of dimethyl disulfide + methanol mixture at the temperatures 310.95 and 336.35 K. Uusi-Kyyny et al. [20] determined the isothermal VLE of the dimethyl disulfide + ethanol system at $T = 343.15$ K. Recently, we have measured the isobaric vapour liquid equilibrium of these systems at $p = 40.000$ and 101.325 kPa [21].

2. Experimental section

2.1. Chemicals used

Chemical specifications are given in Table 1. Due to the high mass purity of these chemicals no further treatment of purification was employed. The water content of the

Table 1 Chemical description

Compound	CAS No.	Source	Mass fraction purity ^a	Analysis method	Water content / ppm
Dimethyl disulfide	624-92-0	Aldrich	0.995	GC	364
Methanol	67-56-1	Sigma-Aldrich	0.998	GC	148
Ethanol	64-17-5	Acros	0.998	GC	133
1-Propanol	71-23-8	Sigma-Aldrich	0.998	GC	195
1-Butanol	71-36-3	Sigma-Aldrich	0.999	GC	187

^aAs stated by the supplier

pure compounds was measured in our laboratory using the Karl-Fischer method with an automatic titrator (Crison KF 1S-2B).

2.2. Apparatus and procedure

The densities of the pure liquids and binary mixtures have been measured using an Anton Paar DMA 5000 densimeter. The cell temperature is controlled by an integrated Peltier thermostat at ± 0.005 K. The apparatus was calibrated using ultra-pure water supplied by the GmbH SH calibration service, and dry air. The uncertainty of density measurements is $0.05 \text{ kg}\cdot\text{m}^{-3}$. Mixtures were prepared by mass using a Sartorius Semimicro balance CP225-D. The uncertainty of the mass determination was 0.01 mg. In order to avoid variations in composition by evaporation, the samples were prepared immediately prior to performing measurements and were kept in airtight stopper glass vials. Taking into account the uncertainty in the water content of pure substances and mass determination, the uncertainty in the mole fraction was estimated to be 0.0005.

The vapour pressures of the pure compounds and the phase equilibria of the binary mixtures was performed using a dynamic ebulliometer (Fischer Labodest), previously described [21,22], equipped with a Cottrell pump. It is an ebulliometer with recirculation of both phases, after leaving the separation chamber the vapour phase is condensed and returned to the liquid phase. The equilibrium pressure was measured by

means of a Digiquartz215A-102 pressure transducer with an uncertainty of 0.05 kPa. An Automatic Systems Laboratories (model F25) thermometer with a PT100 probe was employed to determine the equilibrium temperature with an uncertainty of 0.02 K. Once the equilibrium was established, samples of both the liquid and condensed vapour phases were taken. The corresponding compositions were determined by densitometry, previously the calibration curves density-composition were obtained. The estimated uncertainty in mole fraction is 0.002.

3. Results and discussion

3.1. Pure compounds

In this section, we report data of densities, ρ , at $p = 0.1$ MPa and vapour pressures, p° , for the pure liquids: dimethyl disulfide, methanol, ethanol, 1-propanol and 1-butanol. The working temperatures were 303.15 and 323.15 K.

Table 2 Comparison between experimental and literature values of the pure compounds properties at the working temperatures. Liquid density, ρ , at pressure $p = 0.1$ MPa, and vapour pressure, p° .^a

Compound	$\rho / \text{kg}\cdot\text{m}^{-3}$				p° / kPa			
	$T = 303.15 \text{ K}$		$T = 323.15 \text{ K}$		$T = 303.15 \text{ K}$		$T = 323.15 \text{ K}$	
	Exp. ^b	Lit.	Exp.	Lit.	Exp.	Lit.	Exp.	Lit.
Dimethyl disulfide	1051.136	1051.38 [23]	1028.496		4.925	4.916 [24] ^c 4.924 [25]	12.490	12.39 [24] ^c 12.44 [25]
Methanol	781.808	781.627 [26]	762.615	762.581 [26]	21.888	21.914 [26]	55.735	55.684 [26]
Ethanol	780.778	781.221 [26]	763.601	763.613 [26]	10.465	10.555 [26]	29.545	29.660 [26]
1-Propanol	795.538	795.58 [27]	779.082	779.11 [27]	3.800	3.8526 [28]	12.185	12.10 [29]
1-Butanol	801.918	801.94 [27]	786.607	786.30 [27]	1.225	1.200 [30]	4.505	4.548 [29]

^aFor density measurements, standard uncertainties are $u(T) = 0.005 \text{ K}$, $u(p) = 1 \text{ kPa}$, and the combined expanded uncertainty is $U_c(\rho) = 0.05 \text{ kg}\cdot\text{m}^{-3}$ with 0.95 level of confidence ($k = 2$). For VLE measurements, standard uncertainties are $u(T) = 0.02 \text{ K}$ and $u(p) = 0.05 \text{ kPa}$. ^b These densities have been already published in reference [21]. ^c Correlated vapour pressures values.

The experimental values are given in Table 2 and they are compared with the literature [23-30]. Due to the high number of references for methanol and ethanol, we have chosen for them recommended data from REFPROP (version 9.0) [25]. In general terms, a good agreement is observed. The average deviation in density is 0.02 %, the biggest deviation (0.06 % or 0.44 kg·m⁻³) is shown by the density values of ethanol at $T = 303.15$ K. On the other hand, the average deviation in vapour pressure is 0.66 %, the biggest deviation (2 % or 0.025 kPa) is observed for the vapour pressure value of 1-butanol at $T = 303.15$ K; it must be taken into account that this value is near to the lowest vapour pressure measurable in our equipment. These deviations between experimental and literature values are close to the corresponding experimental uncertainties.

3.2. Binary mixtures

3.2.1. Volumetric properties

The densities, ρ , for the binary mixtures containing dimethyl disulfide and methanol, ethanol, 1-propanol, or 1-butanol have been measured at $T = 303.15$ and 323.15 K and at $p = 0.1$ MPa. These values, which are collected in Table S1 of the supplementary material, were used to calculate the corresponding excess molar volumes, V^E , from the following equation:

$$V^E = \sum_{i=1}^2 x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right) \quad (1)$$

where x_i , M_i , and ρ_i are respectively, the mole fraction, the molar mass, and the density of component i , the densities of the components are in the same physical state as the mixture [31], and ρ is the density of the mixture. The calculated excess molar volumes are also collected in Table S1.

The excess molar volumes have been correlated with composition using the Redlich-Kister expansion [17]:

$$V^E = x_1 x_2 \sum_{i=0}^n A_i (x_1 - x_2)^i \quad (2)$$

being x_i the mole fraction of component i , and A_i the adjustable parameters, which have been determined using the least square method. These values and the corresponding standard deviations are shown in Table 3.

The excess molar volumes, V^E , as function of the dimethyl disulfide mole fraction at temperatures 303.15 and 323.15 K are plotted in Figs. 1 and 2, respectively. These V^E plots are not symmetrical, they are slightly shifted to the rich region in dimethyl disulfide. The V^E values increase in the sequence methanol < ethanol < 1-propanol < 1-butanol, that is, the longer the aliphatic chain of 1-alkanol the greater excess molar volume. The V^E maximum values are also shifted to dimethyl disulfide following the same sequence. Moreover, for all the mixtures the excess molar volume

Table 3 Parameters of the Redlich-Kister equation, A_i , and standard deviations, $\sigma(V^E)$.

T / K	$A_0 \times 10^6 / \text{m}^3 \cdot \text{mol}^{-3}$	$A_1 \times 10^6 / \text{m}^3 \cdot \text{mol}^{-3}$	$A_2 \times 10^6 / \text{m}^3 \cdot \text{mol}^{-3}$	$A_3 \times 10^6 / \text{m}^3 \cdot \text{mol}^{-3}$	$\sigma(V^E) \times 10^6 / \text{m}^3 \cdot \text{mol}^{-3}$
Dimethyl disulfide (1) + methanol (2)					
303.15	0.2719	0.0691	-0.0610	-0.0239	0.0008
323.15	0.3955	0.0705	-0.0398	0.0401	0.0011
Dimethyl disulfide (1) + ethanol (2)					
303.15	0.3927	0.1143	-0.0720	0.0675	0.0010
323.15	0.6642	0.2825	-0.1157		0.0017
Dimethyl disulfide (1) + 1-propanol (2)					
303.15	0.5542	0.1895	-0.0488	0.1396	0.0013
323.15	0.9157	0.2869	0.0275	0.3469	0.0019
Dimethyl disulfide (1) + 1-butanol (2)					
303.15	0.6605	0.2511	0.0921	0.1947	0.0015
323.15	1.0967	0.3735	0.3062	0.5422	0.0025

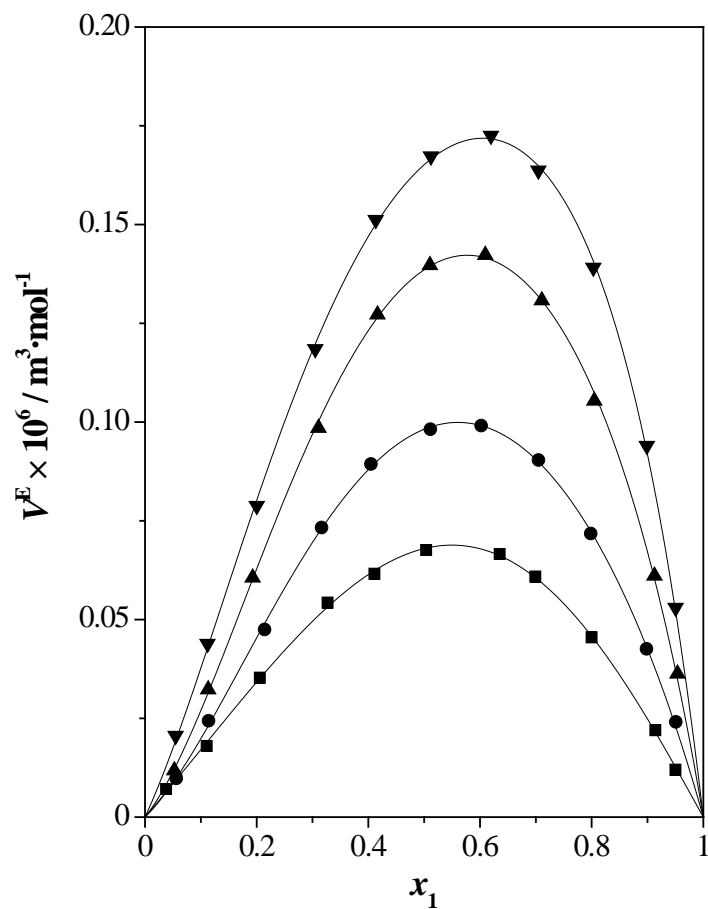


Figure 1. Excess molar volumes, V^E , as a function of the mole fraction, x_1 , at $p = 0.1$ MPa and at $T = 303.15$ K for the binary mixtures dimethyl disulfide (1) + alkanol (2): (■) methanol; (●) ethanol; (▲) 1-propanol; (▼) 1-butanol.

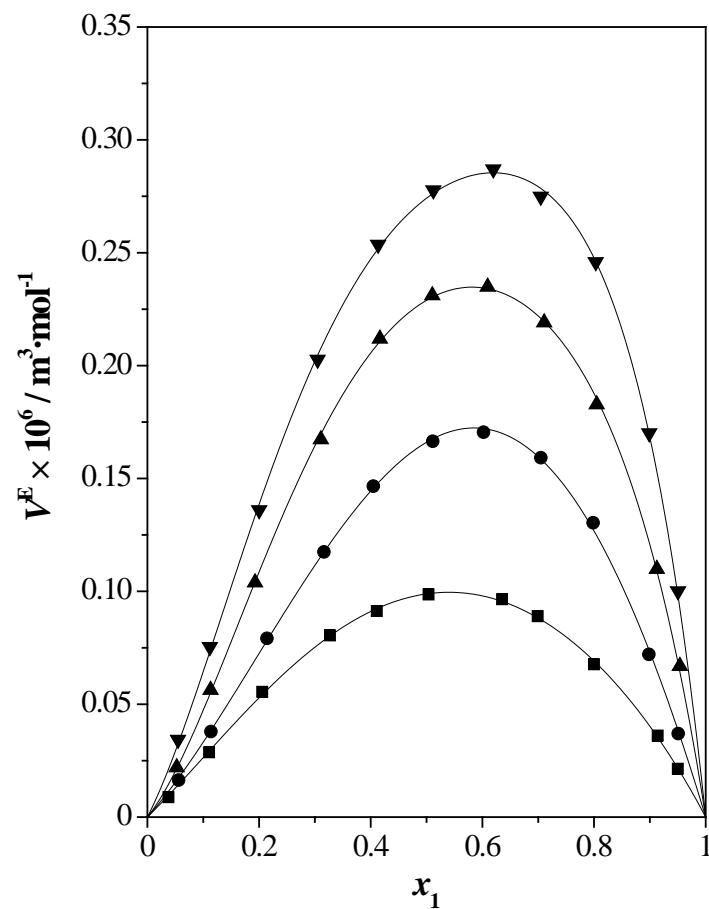


Figure 2. Excess molar volumes, V^E , as a function of the mole fraction, x_1 , at $p = 0.1$ MPa and at $T = 323.15$ K for the binary mixtures dimethyl disulfide (1) + alkanol (2): (■) methanol; (●) ethanol; (▲) 1-propanol; (▼) 1-butanol.

increases with temperature, this increase is similar for the mixtures containing ethanol, 1-propanol and 1-butanol and slightly higher than for the mixture with methanol.

3.2.2. Vapour-liquid equilibrium

The isothermal VLE for the binary systems dimethyl disulfide with methanol, ethanol, 1-propanol, or 1-butanol have been determined at the temperatures 303.15 and 323.15 K. Experimental data (T , p , x_i , y_i) are listed in Table S2 of the supplementary material. These values were correlated with the Wilson model [18] based on the following basic equations:

$$G^E = RT (x_1 \ln \gamma_1 + x_2 \ln \gamma_2) \quad (3)$$

$$\ln \gamma_i = -\ln \left(\sum_j x_j \Lambda_{ij} \right) + 1 - \sum_k \frac{x_k \Lambda_{ki}}{\sum_j x_j \Lambda_{kj}} \quad (4)$$

$$\Lambda_{ij} = \frac{V_j^\circ}{V_i^\circ} \exp \left(-\frac{\lambda_{ij} - \lambda_{ii}}{RT} \right) \quad (5)$$

in the above equations: G^E is the excess Gibbs function; R is the universal gas constant; T is the equilibrium temperature, x_i , γ_i and V_i° are, respectively, the liquid mole fraction, the activity coefficient and the liquid molar volume of component i . Λ_{ij} are the dimensionless model parameters, and $(\lambda_{ij} - \lambda_{ii})$ denotes the adjustable Wilson parameters expressed in $\text{J}\cdot\text{mol}^{-1}$. These latter parameters have been obtained by minimizing the following objective function [34] in terms of experimental and calculated pressures:

$$F = \sum_{i=1}^n \left(\frac{p^{exp} - p^{cal}}{p^{exp}} \right)_i^2 \quad (6)$$

Taking into account the non-ideality of the vapour phase and the variation of the Gibbs energies of the pure compounds with pressure, the calculated pressure, p^{cal} , have been determined as follows [33,34]:

$$p^{cal} = \sum_{i=1}^2 x_i \gamma_i p_i^{\circ} \exp \left[\frac{(V_i^{\circ} - B_{ii})(p - p_i^{\circ}) - (1 - y_i)^2 p \delta_{ij}}{RT} \right] \quad (7)$$

$$\delta_{ij} = 2B_{ij} - B_{ii} - B_{jj} \quad (8)$$

where B_{ii} and B_{ij} are the second virial coefficient of component i and the cross second virial coefficient, respectively. These values were estimated using the Tsonopoulos method [35-37]. The other symbols have been previously described.

The correlation results, Wilson parameters and deviations for pressure, Δp , and vapour phase mole fraction, Δy , are shown in Table 4. The biggest deviations, $\Delta p = 0.038$ kPa and $\Delta y = 0.0060$, indicate that the correlation for these systems is adequate and that the newly measured VLE data are reliable. According to the test proposed by Van Ness et al. [38] described by Fredenslund et al. [39], the experimental data are consistent if $\Delta y < 0.01$ and as can be sighted in Table 4, all the regarded systems satisfy this condition.

The pressure-composition diagrams, p - x_1 - y_1 , both experimental and calculated from the Wilson equation are plotted in Figs. 3-6 along with previous isothermal VLE results [19, 20]. All the systems, at the two working temperatures, present an azeotrope whose coordinates (p_{az} , $x_{1,az}$) are listed in Table 5; the azeotropic coordinates have obtained taking into account that the pressure-composition diagram must present an extremum and the composition of both phases must be equal. As the temperature

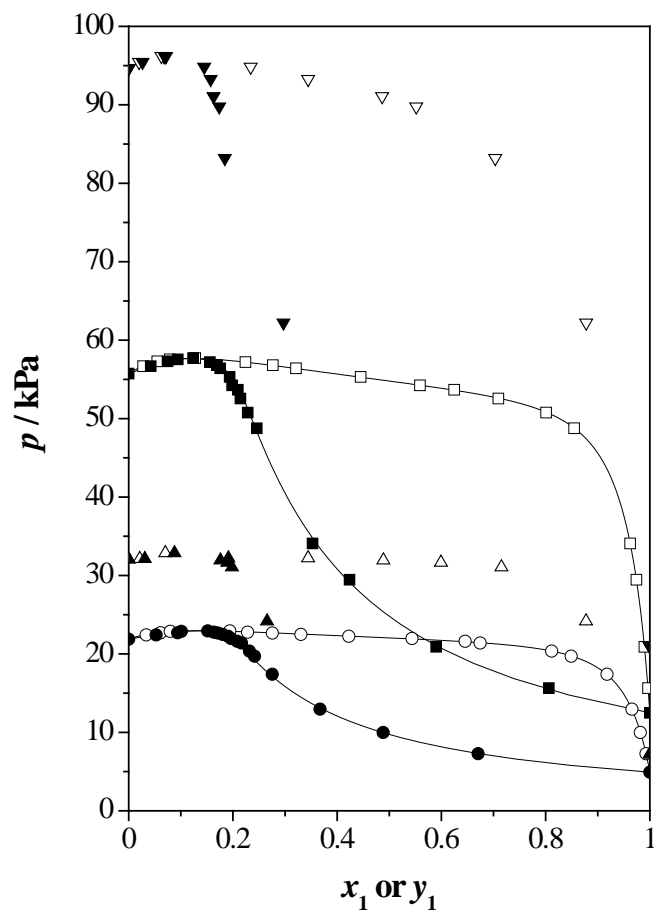


Figure 3. p - x_1 - y_1 diagrams for the binary mixture dimethyl disulfide (1) + methanol (2): (○, ●) experimental data at $T = 303.15$ K; (□, ■) experimental data at $T = 323.15$ K; (—) Wilson correlation; (△, ▲) experimental data at $T = 335.95$ K from reference [19]; (▽, ▼) experimental data at $T = 310.95$ K from reference [19].

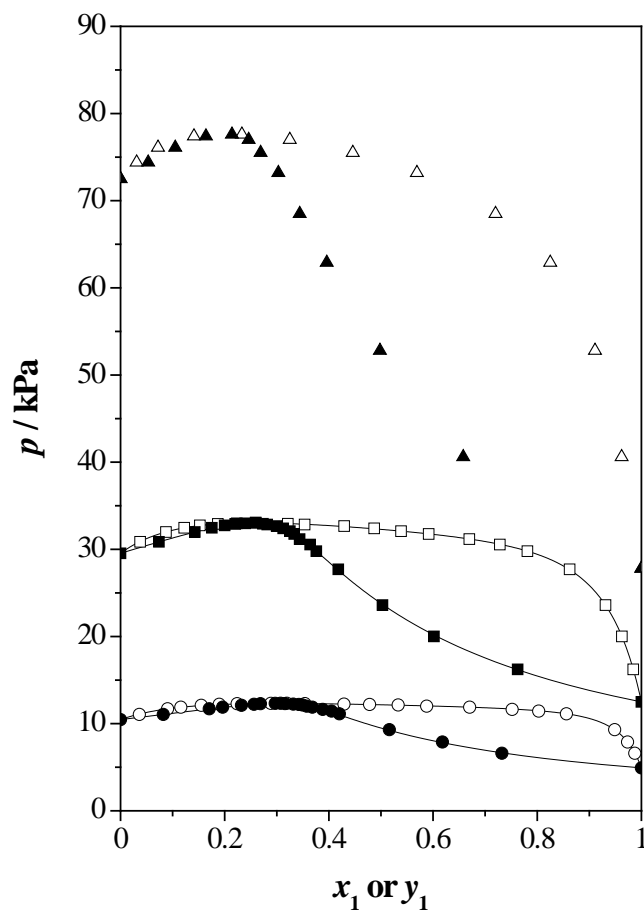


Figure 4. p - x_1 - y_1 diagrams for the binary mixture dimethyl disulfide (1) + ethanol (2): (○, ●) experimental data at $T = 303.15$ K; (□, ■) experimental data at $T = 323.15$ K; (—) Wilson correlation; (△, ▲) experimental data at $T = 343.15$ K from reference [20].

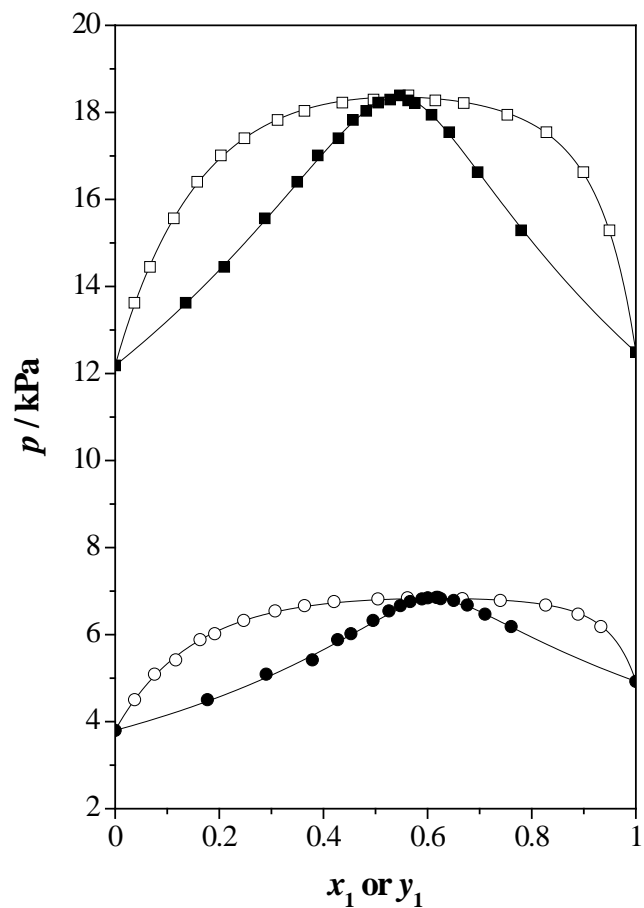


Figure 5. p - x_1 - y_1 diagrams for the binary mixture dimethyl disulfide (1) + 1-propanol (2): (○, ●) experimental data at $T = 303.15$ K, (□, ■) experimental data at $T = 323.15$ K; (—) Wilson correlation

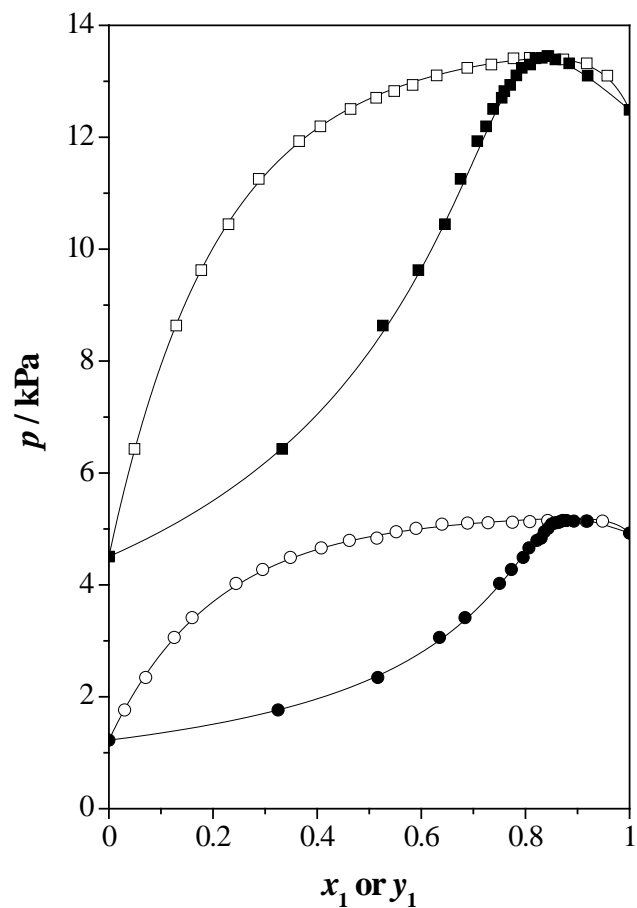


Figure 6. p - x_1 - y_1 diagrams for the binary mixture dimethyl disulfide (1) + 1-butanol (2): (○, ●) experimental data at $T = 303.15$ K, (□, ■) experimental data at $T = 323.15$ K; (—) Wilson correlation.

increases, the azeotrope moves to lower dimethyl disulfide compositions. For a given temperature, the same effect is observed decreasing the carbons number of the n -alkanol. A direct comparison between our experimental results and previous VLE and azeotropic data is not possible because the temperatures don't match. However, as it can be seen in Figs. 3 and 4, and in Table 5 our VLE and azeotropic results are consistent with previous information reported for these systems.

Table 4 Parameters of the Wilson equation, $\lambda_{ij}-\lambda_{ii}$, average deviation in pressure, Δp , and average deviation in vapour phase composition, Δy .

T / K	$\lambda_{12}-\lambda_{11} / \text{J}\cdot\text{mol}^{-1}$	$\lambda_{21}-\lambda_{22} / \text{J}\cdot\text{mol}^{-1}$	$\Delta p / \text{kPa}$	Δy
Dimethyl disulfide (1) + methanol (2)				
303.15	1443.76	7200.31	0.038	0.0037
323.15	1036.97	7927.86	0.031	0.0041
Dimethyl disulfide (1) + ethanol (2)				
303.15	1392.33	6475.81	0.020	0.0043
323.15	1412.56	5625.98	0.023	0.0026
Dimethyl disulfide (1) + 1-propanol (2)				
303.15	1576.84	5033.78	0.021	0.0060
323.15	1400.69	4559.38	0.029	0.0029
Dimethyl disulfide (1) + 1-butanol (2)				
303.15	1572.70	4880.46	0.025	0.0033
323.15	1611.80	4046.10	0.026	0.0044

From the correlation of VLE data, we have calculated the excess Gibbs functions, G^E , for the investigated systems and the values are given in Table S2. The plots of these excess functions versus liquid composition are presented in Figs. 7-8. All mixtures show positive G^E values over the whole composition. The excess Gibbs functions show the maximum values close to the equimolecular composition. At a given temperature, the excess Gibbs function decreases with the increase in the carbon chain length of linear alcohols. Except for the system containing methanol, the temperature

effect on G^E is small, decreasing slightly with temperature. On the other hand, for dimethyl disulfide + methanol system, the G^E maximum value increases with increasing temperature from $1308.5 \text{ J mol}^{-1}$ at $T = 303.15 \text{ K}$ to $1353.8 \text{ J mol}^{-1}$ at $T = 323.15 \text{ K}$.

Thermodynamically, all the studied systems show positive deviations from ideal behaviour over the entire composition range, although for excess molar volume these deviations are not too high. Excess properties could be explained on one hand considering energetic factors, the predominance of the weakening interactions (dipole-dipole interactions in dimethyl disulfide and hydrogen bonding in the alcohols) among molecules in the pure liquids during the mixing process over the heteroassociation between unlike molecules [40] leads to positive values. On the other hand, the structural factors also play an important role, especially in excess molar volume, it can be taken into account the worst interstitial accommodation with increasing chain length of the alkanol that leads to higher V^E values.

Table 5 Azeotropic pressure, p_{az} , and composition, $x_{1,az}$.^a

System	T / K	p_{az} / kPa	$x_{1,az}$
Dimethyl disulfide (1) + methanol (2)	303.15	22.9	0.135
	310.95 [19]	33.0	0.1253
	323.15	57.7	0.121
Dimethyl disulfide (1) + ethanol (2)	336.35 [19]	96.2	0.0882
	303.15	12.3	0.307
	323.15	33.0	0.255
Dimethyl disulfide (1) + 1-propanol (2)	343.15 [20]	78.1	0.199
	303.15	6.8	0.617
	323.15	18.3	0.544
Dimethyl disulfide (1) + 1-butanol (2)	303.15	5.2	0.888
	323.15	13.4	0.836

^a Standard uncertainties are $u(T) = 0.02 \text{ K}$, $u(p) = 0.05 \text{ kPa}$, $u(x_i) = 0.002$.

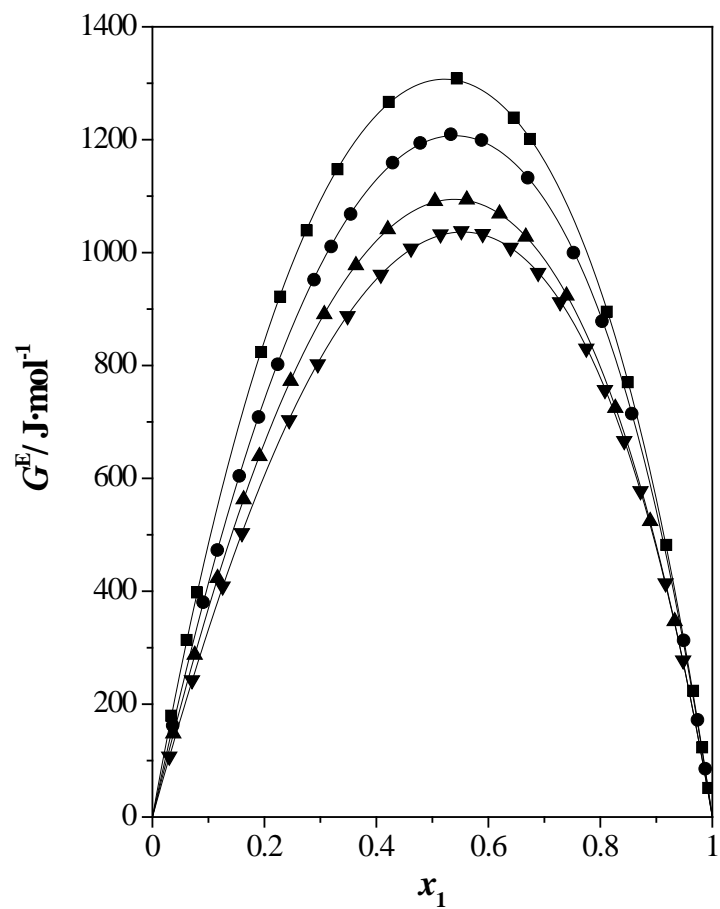


Figure 7. Calculated excess Gibbs function, G^E , as a function of the mole fraction, x_1 , at $T = 303.15$ K for the binary mixtures dimethyl disulfide (1) + alkanol (2): (■) methanol; (●) ethanol; (▲) 1-propanol; (▼) 1-butanol.

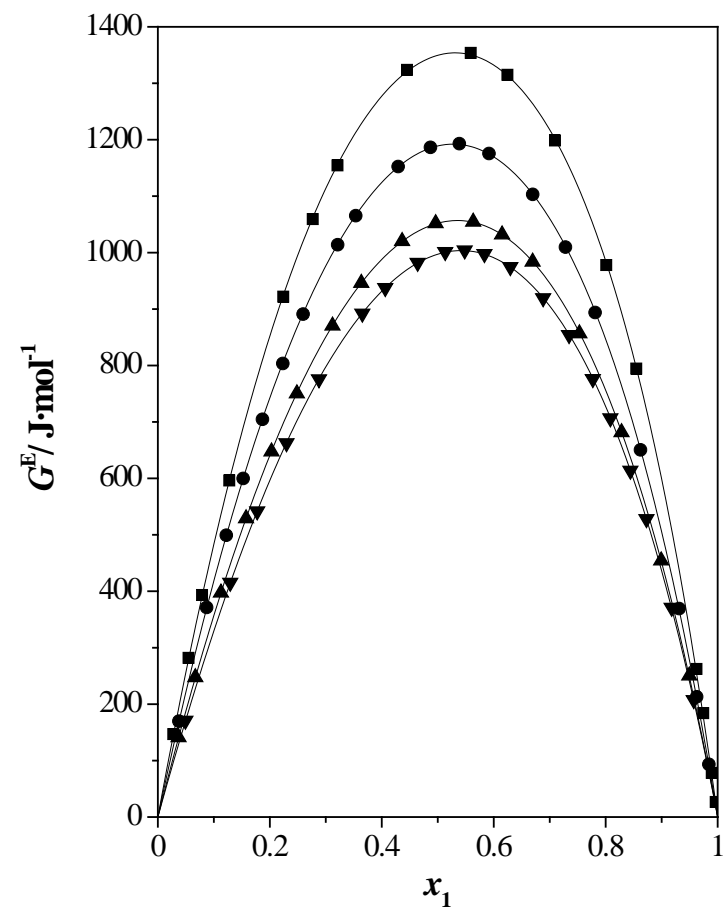


Figure 8. Calculated excess Gibbs function, G^E , as a function of the mole fraction, x_1 , at $T = 323.15$ K for the binary mixtures dimethyl disulfide (1) + alkanol (2): (■) methanol; (●) ethanol; (▲) 1-propanol; (▼) 1-butanol.

4. Conclusions

This work reported experimental density data for binary mixtures containing dimethyl disulfide with C1-C4 *n*-alkanols at $p = 0.1$ MPa, and at $T = (303.15$ and $323.15)$ K. For these systems and temperatures, the isothermal vapour liquid equilibrium was determined and azeotropes were observed. From density values, the excess molar volumes were calculated and fitted by a Redlich-Kister polynomial. The phase equilibrium was correlated using the Wilson equation and the excess Gibbs energies were obtained. Positive deviations from ideality were obtained both the volumetric and the phase behavior. The results showed that the declining of the interactions in the pure compounds is more important than the attraction between different molecules in these mixtures.

Acknowledgments

This research was supported by Gobierno de Aragón (Grant E31_17R), Fondo de Desarrollo Regional “Construyendo Europa desde Aragón” and the Ministry of superior education and scientific research of Tunisia. The authors would like to thank their financial assistance.

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SUPPLEMENTARY MATERIAL

Volumetric behaviour and vapour-liquid equilibrium of dimethyl disulfide + *n*-alkanols binary mixtures

Najla Ben Mahdoui ^{a,b}, Manuela Artal ^b, Carlos Lafuente ^{b,*}, Monia Hichri ^a

^a Université de Tunis EL Manar, Faculté des Sciences, Département de Chimie, Laboratoire des Matériaux, Cristalochimie et Thermodynamique Appliquée, LR15ES01, 2092 Tunis, Tunisia.

^b Departamento de Química Física, Facultad de Ciencias, Universidad de Zaragoza, 50009 Zaragoza, Spain.

* Corresponding author, e-mail: celadi@unizar.es

Table S1

Experimental densities, ρ and calculated excess molar volumes, V^E , for the dimethyl disulfide(1) + 1-alkanol (2) systems at $p = 0.1$ MPa and at the working temperatures.^a

x_1	$\rho / \text{kg}\cdot\text{m}^{-3}$	$V^E \times 10^6 / \text{m}^3\cdot\text{mol}^{-1}$	x_1	$\rho / \text{kg}\cdot\text{m}^{-3}$	$V^E \times 10^6 / \text{m}^3\cdot\text{mol}^{-1}$
Dimethyl disulfide (1) + methanol (2) at $T = 303.15$ K					
0.0000	781.808		0.6353	994.210	0.0666
0.0378	802.980	0.0071	0.6992	1006.041	0.0609
0.1106	839.060	0.0180	0.7999	1022.904	0.0455
0.2057	878.572	0.0353	0.9141	1039.770	0.0220
0.3271	919.701	0.0543	0.9501	1044.675	0.0120
0.4110	943.552	0.0616	1.0000	1051.136	
0.5036	966.452	0.0676			
Dimethyl disulfide (1) + methanol (2) at $T = 323.15$ K					
0.0000	762.615		0.6353	971.797	0.0965
0.0378	783.431	0.0088	0.6992	983.531	0.0890
0.1106	818.823	0.0287	0.7999	1000.296	0.0677
0.2057	857.640	0.0555	0.9141	1017.087	0.0361
0.3271	898.164	0.0805	0.9501	1021.995	0.0214
0.4110	921.685	0.0913	1.0000	1028.496	
0.5036	944.315	0.0986			
Dimethyl disulfide (1) + ethanol (2) at $T = 303.15$ K					
0.0000	780.778		0.6021	967.928	0.0991
0.0560	802.994	0.0098	0.7048	991.584	0.0904
0.1139	824.617	0.0244	0.7984	1011.726	0.0718
0.2142	859.315	0.0475	0.8982	1031.854	0.0426
0.3162	891.369	0.0733	0.9508	1041.944	0.0241
0.4047	916.973	0.0894	1.0000	1051.136	
0.5111	945.419	0.0982			
Dimethyl disulfide (1) + ethanol (2) at $T = 323.15$ K					
0.0000	763.601		0.6021	946.119	0.1705
0.0560	785.279	0.0164	0.7048	969.342	0.1592
0.1139	806.374	0.0379	0.7984	989.208	0.1304
0.2142	840.142	0.0792	0.8982	1009.271	0.0721
0.3162	871.401	0.1175	0.9508	1019.346	0.0370

Table S1.Continuation

x_1	$\rho / \text{kg}\cdot\text{m}^{-3}$	$V^E \times 10^6 / \text{m}^3 \cdot \text{mol}^{-1}$	x_1	$\rho / \text{kg}\cdot\text{m}^{-3}$	$V^E \times 10^6 / \text{m}^3 \cdot \text{mol}^{-1}$
0.4047	896.338	0.1466	1.0000	1028.496	
0.5111	924.087	0.1665			
Dimethyl disulfide (1) + 1-propanol (2) at $T = 303.15$ K					
0.0000	795.538		0.6094	959.857	0.1423
0.0525	811.178	0.0118	0.7108	984.358	0.1308
0.1132	828.807	0.0323	0.8048	1006.529	0.1054
0.1928	851.313	0.0606	0.9125	1031.308	0.0611
0.3107	883.505	0.0985	0.9538	1040.685	0.0363
0.4164	911.286	0.1272	1.0000	1051.136	
0.5103	935.259	0.1397			
Dimethyl disulfide (1) + 1-propanol (2) at $T = 323.15$ K					
0.0000	779.082		0.6094	938.494	0.2349
0.0525	794.254	0.0220	0.7108	962.431	0.2191
0.1132	811.330	0.0563	0.8048	984.163	0.1828
0.1928	833.106	0.1039	0.9125	1008.632	0.1099
0.3107	864.262	0.1673	0.9538	1017.971	0.0669
0.4164	891.202	0.2119	1.0000	1028.496	
0.5103	914.517	0.2311			
Dimethyl disulfide (1) + 1-butanol (2) at $T = 303.15$ K					
0.0000	801.918		0.6197	952.724	0.1725
0.0549	815.025	0.0205	0.7046	974.140	0.1637
0.1120	828.678	0.0439	0.8031	999.294	0.1391
0.2002	849.860	0.0788	0.8988	1024.135	0.0940
0.3049	875.149	0.1185	0.9505	1037.819	0.0530
0.4133	901.564	0.1511	1.0000	1051.136	
0.5123	925.964	0.1673			
Dimethyl disulfide (1) + 1-butanol (2) at $T = 323.15$ K					
0.0000			0.6197	931.991	0.2870
0.0549	799.243	0.0343	0.7046	952.767	0.2749
0.1120	812.370	0.0754	0.8031	977.163	0.2459
0.2002	832.733	0.1360	0.8988	1001.530	0.1701
0.3049	857.051	0.2027	0.9505	1015.087	0.1001

Table S1.Continuation

x_1	$\rho / \text{kg}\cdot\text{m}^{-3}$	$V^E \times 10^6 / \text{m}^3\cdot\text{mol}^{-1}$	x_1	$\rho / \text{kg}\cdot\text{m}^{-3}$	$V^E \times 10^6 / \text{m}^3\cdot\text{mol}^{-1}$
0.4133	882.514	0.2536	1.0000	1028.496	
0.5123	906.099	0.2777			

^a Standard uncertainties are $u(T) = 0.005 \text{ K}$, $u(p) = 1 \text{ kPa}$, and $u(x_1) = 0.0005$, and the combined expanded uncertainty is $U_c(\rho) = 0.05 \text{ kg}\cdot\text{m}^{-3}$ with 0.95 level of confidence ($k = 2$).

Table S2

Isothermal VLE for dimethyl disulfide(1) + 1-alkanol (2) systems at the working temperatures. Experimental data: equilibrium pressure, p , liquid-phase, x_1 , and vapor-phase, y_1 , mole fractions. Calculated properties: activity coefficients, γ_i and excess Gibbs function, G^E .^a

p / kPa	x_1	y_1	γ_1	γ_2	$G^E / \text{J}\cdot\text{mol}^{-1}$
Dimethyl disulfide (1) + methanol (2) at $T = 303.15 \text{ K}$					
21.880	0.0000	0.0000			
22.430	0.0336	0.0526	7.532	1.004	179.6
22.700	0.0611	0.0940	6.471	1.011	313.7
22.885	0.0796	0.1011	5.892	1.018	397.9
22.930	0.1941	0.1516	3.694	1.095	823.8
22.775	0.2281	0.1634	3.305	1.128	921.9
22.660	0.2756	0.1713	2.873	1.183	1039.3
22.490	0.3305	0.1803	2.489	1.259	1147.7
22.250	0.4223	0.1907	2.025	1.425	1267.0
21.970	0.5437	0.1964	1.619	1.757	1308.5
21.610	0.6455	0.2082	1.387	2.206	1239.2
21.395	0.6743	0.2166	1.333	2.381	1201.3
20.360	0.8116	0.2317	1.134	3.836	895.3
19.715	0.8490	0.2414	1.093	4.588	770.4
17.410	0.9177	0.2754	1.034	7.049	482.1
12.955	0.9657	0.3672	1.007	10.850	223.6
9.990	0.9818	0.4882	1.002	13.061	123.3
7.285	0.9926	0.6705	1.000	15.036	51.5
4.925	1.0000	1.0000			
Dimethyl disulfide (1) + methanol (2) at $T = 323.15 \text{ K}$					
55.735	0.0000	0.0000			
56.680	0.0276	0.0424	6.763	1.002	146.9
57.300	0.0549	0.0745	5.946	1.008	282.0
57.550	0.0790	0.0943	5.353	1.015	393.3
57.710	0.1276	0.1236	4.423	1.038	596.3
57.205	0.2241	0.1558	3.231	1.109	921.9
56.810	0.2767	0.1695	2.798	1.164	1059.4
56.410	0.3210	0.1755	2.507	1.219	1154.5
55.310	0.4452	0.1937	1.928	1.435	1323.7

Table S2. (Continuation)

p / kPa	x_1	y_1	γ_1	γ_2	$G^E / \text{J}\cdot\text{mol}^{-1}$
54.240	0.5589	0.1985	1.583	1.751	1353.8
53.675	0.6245	0.2096	1.434	2.020	1315.0
52.575	0.7094	0.2144	1.280	2.540	1198.6
50.765	0.8011	0.2285	1.151	3.536	977.9
48.770	0.8547	0.2458	1.091	4.579	794.5
34.080	0.9622	0.3528	1.009	10.471	262.4
29.455	0.9743	0.4238	1.005	12.072	184.0
20.910	0.9895	0.5900	1.001	14.808	78.3
15.645	0.9965	0.8063	1.000	16.460	26.6
12.490	1.0000	1.0000			
Dimethyl disulfide (1) + ethanol (2) at $T = 303.15 \text{ K}$					
10.465	0.0000	0.0000			
11.075	0.0362	0.0820	5.510	1.003	162.2
11.705	0.0904	0.1703	4.524	1.016	380.5
11.890	0.1158	0.1961	4.158	1.026	473.1
12.105	0.1550	0.2322	3.684	1.046	604.4
12.220	0.1895	0.2563	3.337	1.067	708.7
12.290	0.2238	0.2686	3.044	1.093	802.2
12.330	0.2886	0.2969	2.598	1.154	952.0
12.340	0.3193	0.3073	2.425	1.190	1010.9
12.300	0.3540	0.3157	2.253	1.235	1068.4
12.240	0.4290	0.3317	1.947	1.357	1159.4
12.205	0.4782	0.3424	1.784	1.458	1194.3
12.145	0.5330	0.3493	1.631	1.599	1209.7
12.005	0.5878	0.3573	1.501	1.778	1199.4
11.885	0.6703	0.3683	1.339	2.158	1132.7
11.630	0.7519	0.3882	1.212	2.761	999.9
11.435	0.8029	0.4050	1.147	3.357	878.4
11.130	0.8560	0.4202	1.089	4.327	714.7
9.310	0.9487	0.5161	1.016	8.445	313.0
7.895	0.9735	0.6180	1.005	11.059	172.2
6.600	0.9873	0.7322	1.001	13.231	85.6
4.925	1.0000	1.0000			

Table S2.(Continuation)

p / kPa	x_1	y_1	γ_1	γ_2	G^E / J·mol ⁻¹
Dimethyl disulfide (1) + ethanol (2) at $T = 323.15$ K					
29.545	0.0000	0.0000			
30.855	0.0376	0.0738	4.999	1.003	169.8
31.980	0.0872	0.1426	4.197	1.014	371.3
32.475	0.1223	0.1753	3.751	1.028	499.3
32.745	0.1524	0.2000	3.429	1.043	599.8
32.915	0.1869	0.2201	3.113	1.063	704.7
32.975	0.2232	0.2394	2.832	1.090	803.6
33.055	0.2595	0.2595	2.592	1.121	891.1
32.930	0.3212	0.2734	2.259	1.186	1014.0
32.845	0.3535	0.2816	2.114	1.227	1065.5
32.670	0.4293	0.2998	1.832	1.345	1152.5
32.400	0.4870	0.3123	1.662	1.460	1186.5
32.085	0.5386	0.3252	1.533	1.589	1193.0
31.755	0.5916	0.3335	1.421	1.754	1175.6
31.175	0.6696	0.3438	1.285	2.084	1103.2
30.550	0.7282	0.3641	1.202	2.436	1009.8
29.785	0.7811	0.3760	1.138	2.878	893.9
27.705	0.8622	0.4181	1.062	3.974	650.5
23.605	0.9311	0.5029	1.018	5.769	369.4
20.005	0.9626	0.6018	1.006	7.172	213.1
16.225	0.9843	0.7625	1.001	8.540	93.4
12.490	1.0000	1.0000			
Dimethyl disulfide (1) + 1-propanol (2) at $T = 303.15$ K					
3.800	0.0000	0.0000			
4.505	0.0373	0.1771	4.539	1.002	147.8
5.090	0.0755	0.2899	4.039	1.009	287.3
5.420	0.1163	0.3787	3.598	1.022	423.3
5.885	0.1629	0.4273	3.183	1.042	562.7
6.020	0.1912	0.4528	2.968	1.058	639.3
6.325	0.2469	0.4953	2.611	1.097	772.6
6.545	0.3070	0.5257	2.300	1.151	890.7
6.665	0.3635	0.5477	2.062	1.217	977.5
6.760	0.4203	0.5665	1.863	1.299	1041.2

Table S2.(Continuation)

p / kPa	x_1	y_1	γ_1	γ_2	$G^E / \text{J}\cdot\text{mol}^{-1}$
6.820	0.5046	0.5891	1.626	1.461	1091.1
6.845	0.5614	0.6003	1.496	1.607	1093.8
6.855	0.6200	0.6169	1.381	1.802	1069.0
6.470	0.8890	0.7102	1.047	4.509	524.2
6.185	0.9328	0.7605	1.019	5.931	346.9
4.925	1.0000	1.0000			
Dimethyl disulfide (1) + 1-propanol (2) at $T = 323.15 \text{ K}$					
12.185	0.0000	0.0000			
13.625	0.0370	0.1355	3.931	1.002	141.2
14.450	0.0669	0.2095	3.625	1.006	247.6
15.565	0.1127	0.2873	3.227	1.018	397.4
16.405	0.1576	0.3497	2.901	1.035	529.1
17.010	0.2031	0.3891	2.623	1.058	647.5
17.405	0.2483	0.4288	2.388	1.088	750.4
17.825	0.3120	0.4567	2.112	1.141	870.4
18.035	0.3635	0.4822	1.928	1.195	946.1
18.225	0.4363	0.5051	1.711	1.294	1020.2
18.295	0.4963	0.5281	1.565	1.400	1051.8
18.390	0.5633	0.5469	1.427	1.552	1054.5
18.275	0.6150	0.5627	1.338	1.704	1031.9
18.215	0.6695	0.5756	1.256	1.909	983.7
17.945	0.7531	0.6077	1.152	2.360	856.8
17.545	0.8283	0.6416	1.081	3.014	681.6
16.625	0.8992	0.6962	1.031	4.065	454.3
15.290	0.9493	0.7797	1.009	5.332	250.5
12.490	1.0000	1.0000			
Dimethyl disulfide (1) + 1-butanol (2) at $T = 303.15 \text{ K}$					
1.225	0.0000	0.0000			
1.765	0.0301	0.3250	3.959	1.001	107.3
2.345	0.0707	0.5165	3.588	1.006	242.6
3.060	0.1255	0.6350	3.167	1.020	408.6
3.415	0.1599	0.6840	2.941	1.033	503.1
4.025	0.2443	0.7503	2.483	1.078	703.2
4.275	0.2954	0.7730	2.259	1.116	802.3

Table S2.(Continuation)

p / kPa	x_1	y_1	γ_1	γ_2	$G^E / \text{J}\cdot\text{mol}^{-1}$
4.490	0.3486	0.7957	2.059	1.167	887.7
4.660	0.4079	0.8067	1.869	1.238	961.0
4.795	0.4619	0.8221	1.720	1.319	1007.2
4.835	0.5146	0.8302	1.593	1.419	1032.5
4.950	0.5517	0.8363	1.513	1.505	1038.2
5.010	0.5899	0.8435	1.439	1.610	1033.1
5.085	0.6397	0.8500	1.351	1.779	1008.9
5.105	0.6889	0.8550	1.275	1.997	964.2
5.110	0.7282	0.8591	1.220	2.223	912.3
5.120	0.7750	0.8644	1.162	2.578	830.1
5.130	0.8083	0.8665	1.125	2.917	756.8
5.150	0.8428	0.8722	1.090	3.381	666.3
5.150	0.8718	0.8791	1.065	3.904	577.6
5.140	0.9166	0.8930	1.031	5.118	414.3
5.140	0.9480	0.9188	1.014	6.499	277.7
4.925	1.0000	1.0000			
Dimethyl disulfide (1) + 1-butanol (2) at $T = 323.15 \text{ K}$					
4.505	0.0000	0.0000			
6.430	0.0492	0.3329	3.439	1.003	170.9
8.635	0.1294	0.5262	2.880	1.021	415.3
9.625	0.1775	0.5944	2.611	1.039	541.9
10.445	0.2298	0.6456	2.362	1.066	662.6
11.255	0.2879	0.6755	2.128	1.105	775.8
11.930	0.3653	0.7076	1.871	1.177	892.2
12.195	0.4063	0.7243	1.756	1.224	937.6
12.505	0.4641	0.7378	1.614	1.307	982.1
12.705	0.5134	0.7544	1.508	1.394	1001.4
12.825	0.5481	0.7595	1.442	1.467	1004.2
12.935	0.5834	0.7707	1.380	1.554	997.8
13.105	0.6296	0.7826	1.307	1.690	974.6
13.240	0.6883	0.7930	1.226	1.912	919.5
13.300	0.7346	0.8088	1.170	2.144	854.3
13.410	0.7771	0.8192	1.126	2.418	776.2
13.420	0.8084	0.8315	1.097	2.673	706.7

Table S2.(Continuation)

p / kPa	x_1	y_1	γ_1	γ_2	$G^E / \text{J}\cdot\text{mol}^{-1}$
13.450	0.8441	0.8422	1.068	3.040	613.9
13.390	0.8728	0.8575	1.047	3.416	528.1
13.320	0.9178	0.8840	1.022	4.228	371.1
13.100	0.9574	0.9195	1.006	5.306	207.4
12.490	1.0000	1.0000			

^a Standard uncertainties are $u(T) = 0.02 \text{ K}$, $u(p) = 0.05 \text{ kPa}$, $u(x_1) = 0.002$, and $u(y_1) = 0.002$.