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Densities, Viscosities and Refractive Indices for Binary and Ternary Mixtures of Dimethylacetamide + Dimethylbenzylamine + Dichloromethane

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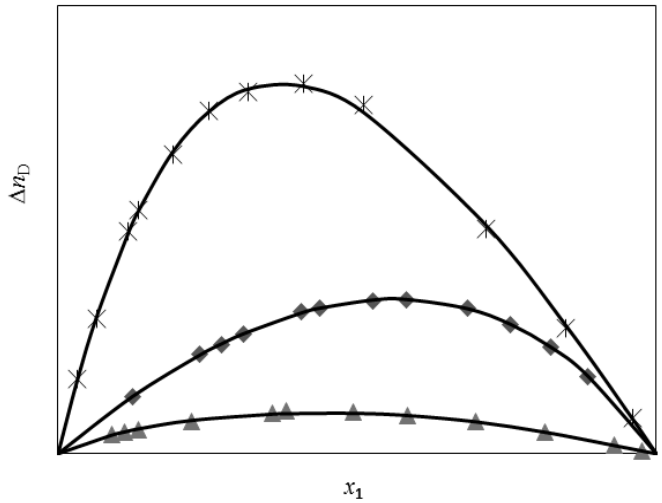
ABSTRACT

Density (ρ), viscosity (η), and refractive index (n_D) for ternary systems include dimethylacetamide, dimethylbenzylamine, and dichloromethane, as well as the related binaries such as dimethylacetamide + dimethylbenzylamine, dimethylacetamide + dichloromethane, and dimethylbenzylamine + dichloromethane, were obtained at ambient pressure. Excess volumes, V_m^E, viscosity deviations $\Delta\eta$, and refractive index deviations Δn_D of the mixtures were obtained using the Redlich-Kister and the Cibulka relations for binary and ternary systems, respectively. Adopted values for coefficient and standard deviations were reported. The experimental data for ternary system were also fitted to Tsao-Smith, Radjkovic, and Kohler models. The intermolecular interactions were analyzed based on the experimental findings.

GRAPHICAL ABSTRACT



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Introduction

This research aims to test theories of the liquid state, understand intermolecular interactions, and predict solvent behavior. However, physico-chemical properties such as excess functions depend upon various factors between like and unlike molecules. Here, we focus on measuring density, viscosity, and refractive index for selected systems [1-3]. The binary and ternary mixtures have been analyzed using Redlich-Kister and Cibulka relations, respectively [4,5]. Additionally, the experimental results for the ternary system fit the Tsao-Smith, Radjkovic, and Kohler models. These findings aim to contribute to the knowledge of thermodynamic properties in mixtures, which can ultimately aid in engineering design and operations.

Experimental

Chemicals

Dimethylcetamide (>99.5%), dimethyl-benzylamine (>99%) and dichloromethane (>99.5%) were purchased from Merck. The purity was confirmed through GC, and their density and refractive index were compared to the literature data presented in **Table 1** [6,7]. The chemicals were stored in opaque containers and were purified through fractional distillation before use.

Apparatus and Procedure

Density was taken using an Anton Parr DMA 4500 densimeter in static mode, with an estimated uncertainty of ±1 × 10^{-5} g.cm^{-3}. Refractive indices were measured using an Abbe refractometer with an uncertainty of ±4 × 10^{-5}. Dynamic viscosities at 298.15 K were measured using an Ubbelohde viscometer, with an uncertainty of ±10^{-3} mPa.s, and viscosity was expressed as [8]:

$$\eta = \rho\left(kt - \frac{c}{t}\right) \quad (1)$$

where k and c are viscometer parameters, and t is the time for flow.

Table 1. Comparison of Measured Densities,Viscosities, and Refractive Indices of the pure Components with Literature Values at 298.15 K

	ρ (g.cm^{-3})		η (mPa.s)		n_D	
Component	**Exp**	**Lit^a**	**Exp**	**Lit^a**	**Exp**	**Lit^a**
dimethylacetamide	0.93636	0.9366	0.945	0.943 ^{b}	1.4356	1.4357
dimethylbenzylamine	0.89503	0.8940	1.800	1.796	1.4985	1.4991
dichloromethane	1.31669	1.31610	0.423	0.400	1.4212	1.4220

^{a} Ref [6] ^{b} Ref [7]

Standard uncertainties u are: u (T) = 0.01 K, u (P) = 0.05 MPa, and expanded uncertainties U are: U (ρ) = 3 ∗10^{-4} g·cm^{-3}, U (η) = 0.02 (mPa.s) and U (n_D) = 3 ∗10^{-5} with 0.95 level of confidence

Results and discussion

The V_m^E , calculated as follows:

$$V_m^E = \sum_{i=1}^{N} x_i M_i \left(\rho_{mix}^{-1} - \rho_i^{-1}\right) \quad (2)$$

where ρ and M_i are density and molar mass, respectively. The resulting values of V_m^E for dimethylacetamide and dimethylbenzylamine demonstrated negative values throughout the

range of mole fractions, while for dimethylacetamide + dichloromethane and dimethylbenzylamine + dichloromethane showed a sign inversion in V_m^E over part of the concentration range (x≅0.65). **Figure 1** displays V_m^E and density and V_m^E values have been presented (**Table 2**). The occurrence of favorable bipolar-bipolar intermolecular potential between unlike molecules due to their polarity is suggested to explain this observation. Additionally, the presence of alkyl groups with a positive inductive effect causes further intensification of this polarity since the negative charge congregates on the electronegative atoms. Consequently, there is a significant reduction in volume in this system. For the dimethyl acetamide + dichloromethane and dimethylbenzylamine + dichloromethane systems, a sign change from positive to negative in the zones (x≅0.65) occurred. This is because at higher concentrations of amide and amine, the molecules form cages, and dichloromethane molecules occupy the spaces between these molecules, leading to a contraction in volume. However, at high concentrations of dichloromethane, undesirable interactions between dissimilar molecules lead to a positive deviation from the ideal state.

The viscosity deviations, $\Delta\eta$ was calculated using equation (3) [9]:

$$\Delta\eta = \eta - \sum_{i=1}^{N} x_i \eta_i \quad (3)$$

where η and η_i are the viscosity of the mixture and pure components, respectively. Table 2 lists η and $\Delta\eta$ in binary systems and graphically showed in **Figure 2**. The results indicate that all systems exhibit negative $\Delta\eta$ values, with parabolic curves and the highest value observed within the x 0.5 zone. Negative $\Delta\eta$ values suggest that factors such as molecular size, shape, and spatial formation are also effective in addition to force and specific interactions among molecules. For instance, in the dimethylacetamide + dimethylbenzylamine system, the alkyl group and benzenoid loop's difference in shape and size leads to spatial inhibition, rendering the molecules unable to approach each other closely. As a result, the solution has a lower viscosity than each pure ingredient, resulting in negative viscosity deviation.

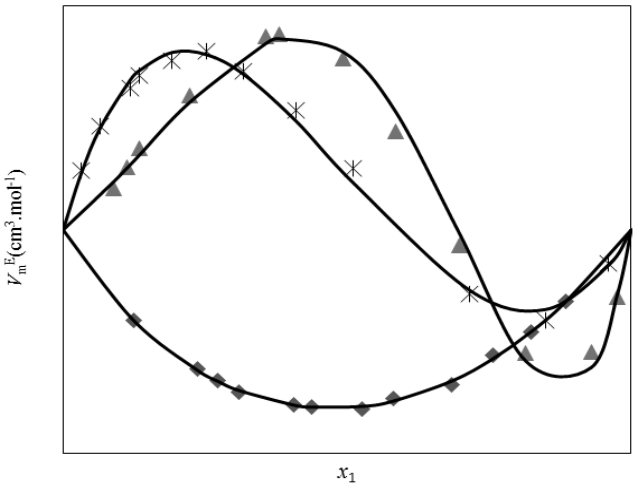

Figure 1. Variation of excess molar volume V_m^E with mole fraction x_1 for the binary systems at T = 298.15K:◆ dimethylacetamide + dimethylbenzylamine; ▲ dimethyl acetamide + dichloromethane ; ∗ dimethylbenzylamine + dichloromethane. Solid curves were calculated from the Redlich–Kister equation

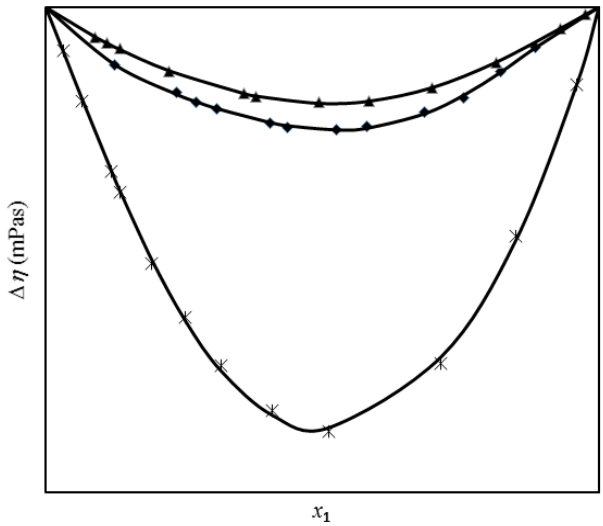

Figure 2. Variation of deviation in the viscosity $\Delta\eta$ with mole fraction x_1 for the binary systems at T = 298.15K: ◆ dimethylacetamide + dimethylbenzylamine; ▲ -dimethylacetamide + dichloromethane ; ✳ dimethylbenzylamine + dichloromethane. Solid curves were calculated from the Redlich–Kister equation

Table 2. Experimental Densities ρ, Viscosities η ,Refractive Indices n_D , Excess Molar Volumes V_m^E, Deviation in the Viscosity $\Delta\eta$, and Deviation in the Refractive Index Δn_D for the Binary Systems at 298.15 K

x	ρ (g.cm^{-3})	η (mPa.s)	n_D	V_m^E (cm^{3}mol^{-1})	$\Delta\eta$ (mPa.s)	Δn_D
(x) *dimethylacetamide* + (1 – x) *dimethylbenzylamine*						
0.0000	0.89503	1.795	1.4991			
0.1238	0.89910	1.664	1.4938	0.122-	0.021-	0.0026
0.2371	0.90290	1.558	1.4885	-0.187	-0.032	0.0044
0.2726	0.90413	1.524	1.4867	-0.202	-0.035	0.0049
0.3099	0.90546	1.490	1.4848	0.217-	0.037-	0.0054
0.4059	0.90894	1.404	1.4797	-0.234	-0.043	0.0063
0.4373	0.91013	1.376	1.4779	-0.238	-0.044	0.0065
0.5260	0.91363	1.300	1.4727	-0.240	-0.045	0.0069
0.5816	0.91586	1.255	1.4692	0.227-	-0.044	0.0070
0.6847	0.92039	1.173	1.4622	-0.207	-0.039	0.0065
0.7563	0.92361	1.117	1.4569	-0.168	-0.033	0.0058
0.8231	0.92690	1.071	1.4517	-0.137	-0.023	0.0048
0.8858	0.93010	1.026	1.4464	-0.096	-0.015	0.0035
1.0000	0.93636	0.945	1.4357			

Table 2. Continued…

x	ρ (g.cm^{-3})	η (mPa.s)	n_D	V_m^E (cm^{3}mol^{-1})	$\Delta\eta$ (mPa.s)	Δn_D
(*x*) *dimethylacetamide* + (1 – *x*) *dichloromethane*						
0.0000	1.31669	0.423	1.4220			
0.0890	1.26865	0.458	1.4241	0.056	0.011-	0.0009
0.1117	1.25675	0.468	1.4245	0.083	0.013-	0.0010
0.1338	1.24540	0.478	1.4249	0.109	0.015-	0.0011
0.2232	1.20220	0.516	1.4265	0.180	0.024-	0.0014
0.3578	1.14324	0.578	1.4287	0.260	0.032-	0.0018
0.3806	1.13403	0.588	1.4291	0.262	0.033-	0.0019
0.4938	1.09118	0.646	1.4306	0.230	0.035-	0.0018
0.5849	1.06000	0.694	1.4317	0.137	0.034-	0.0017
0.6986	1.02405	0.758	1.4330	-0.017	0.030-	0.0014
0.8141	0.99020	0.828	1.4341	-0.165	0.021-	0.0009
0.8601	0.97686	0.857	1.4345	-0.180	0.015-	0.0007
0.9298	0.95698	0.901	1.4351	-0.161	0.008-	0.0004
0.9766	0.94350	0.930	1.4355	-0.090	-0.003	0.0001
1.0000	0.93636	0.945	1.4357			
(*x*) *dimethylbenzylamine* + (1 – *x*) *dichloromethan*						
0.0000	1.31669	0.423	1.4220			
0.0319	1.28466	0.451	1.4274	0.093	-0.016	0.0029
0.0655	1.25450	0.478	1.4326	0.153	0.035-	0.0055
0.1178	1.21295	0.523	1.4405	0.202	0.061-	0.0094
0.1348	1.20048	0.539	1.4428	0.219	0.069-	0.0104
0.1922	1.16237	0.591	1.4499	0.239	0.095-	0.0131
0.2526	1.12710	0.653	1.4564	0.251	0.115-	0.0149
0.3170	1.09440	0.724	1.4624	0.223	0.133-	0.0159
0.4096	1.05388	0.833	1.4697	0.169	0.150-	0.0161
0.5110	1.01649	0.964	1.4764	-0.090	0.157-	0.0150
0.6235	0.98178	1.130	1.4830	-0.039	-0.146	0.0129
0.7152	0.95692	1.269	1.4874	-0.082	0.132-	0.0103
0.7811	0.94095	1.376	1.4903	-0.120	0.115-	0.0081
0.9226	0.91007	1.633	1.4960	-0.074	-0.052	0.0029
1.0000	0.89503	1.795	1.4991			

Equation (4) defines the deviation in refractive index, Δn_D [10]:

$$\Delta n_D = n_D - \sum_i^n x_i\, n_{D,i} \quad (4)$$

where n_D and $n_{D,i}$ are the refractive index of mixtures and pure components, respectively. Across all composition ranges studied Δn_D was positive. **Figure 3** displays Δn_D *vs.* mole fractions, and results are given in **Table 2**. In the case of the dimethylacetamide + dimethylbenzylamine system, the positive deviation in refractive index can be attributed to stronger bipolar-bipolar interactions between molecules, given the strong polarity of both molecules. These interactions result in increased electron density in the mixture, causing a decrease in the speed of light passing through and increasing the refractive index.

The increase in light speed in solutions with lower viscosity results in the positive deviation observed in the refractive index.

The V_m^E, $\Delta\eta$, and Δn_D calculated using the Redlich-Kister polynomial function to correlate data.

$$\Delta Q_{ij} = x_i x_j \sum_{k=0}^{m} A_k (x_j - x_i)^k \quad (5)$$

where ΔQ_{ij} shows to V_m^E, $\Delta\eta$ or Δn_D A_k is an parameter and σ as standard deviation calculated as:

$$\sigma = \left[\sum_{i=1}^{n} \frac{(\Delta Q_i^{exp} - \Delta Q_i^{cal})^2}{(n-p)}\right]^{\frac{1}{2}} \quad (6)$$

where ΔQ_i^{exp} and ΔQ_i^{cal} refer to the experimental and calculated value. n and p are the numbers of experimental points and several adjustable coefficients. **Table 3** presents the values of the parameters A_k together with the standard deviation σ.

Densities, viscosities, refractive indices, excess molar volumes, deviations in viscosity, and refractive index for ternary mixtures of system dimethylacetamide + dimethylbenzylamine + dichloromethane are presented in **Table 4**. The V_m^E, $\Delta\eta$, and Δn_D for the ternary system were correlated using the Cibulka equation (7):

$$\Delta Q_{123} = \Delta Q_{bin} + x_1 x_2 x_3 \Delta_{123} \quad (7)$$

and

$$\Delta Q_{bin} = \sum_{i=1}^{3} \sum_{j>i}^{3} \Delta Q_{ij} \quad (8)$$

where ΔQ_{123} refers to V_m^E, $\Delta\eta$ and Δn_D for ternary mixtures. ΔQ_{ij} in equation (8) is the binary contribution of each *i-j* pair to the V_m^E, $\Delta\eta$ and Δn_D given by Equation 5 with the parameters shown in Table 3. The ternary contribution term Δ_{123} was correlated as:

$$\Delta_{123} = B_0 + B_1 x_1 + B_2 x_2 \quad (9)$$

The ternary parameters B_0, B_1 and B_2 were extracted, and their values with standard deviations were given in Table 3.

The experimental results for V_m^E, of ternary system fitted to models such as Radjkovic [11], TSAO–Smith [12], and Kohler [13].

The Radjkovic model is defined as:

$$V_{123}^E = V_{12}^E + V_{13}^E + V_{23}^E \quad (10)$$

$$V_{123}^E = x_1 x_2 \left(\sum_{n=0}^{k} A_{12}(n)(x_1 - x_2)^n\right) + x_1 x_3 \left(\sum_{n=0}^{k} A_{13}(n)(x_3 - x_1)^n\right) + x_2 x_3 \left(\sum_{n=0}^{k} A_{23}(n)(x_2 - x_3)^n\right) \quad (11)$$

A_{12}, A_{13} and A_{23} values are the adjustable parameters in binary systems.

The Tsao–Smith model is defined as:

$$V_{123}^E = \frac{V_{12}^E x_2}{(1-x_1)} + \frac{V_{13}^E x_3}{(1-x_1)} + V_{23}^E (1 - x_1) \quad (12)$$

The Kohler model is given by:

$$V_{123}^E = V_{12}^E (x_1 + x_2)^2 + V_{13}^E (x_1 + x_3)^2 + V_{23}^E (x_2 + x_3)^2 \quad (13)$$

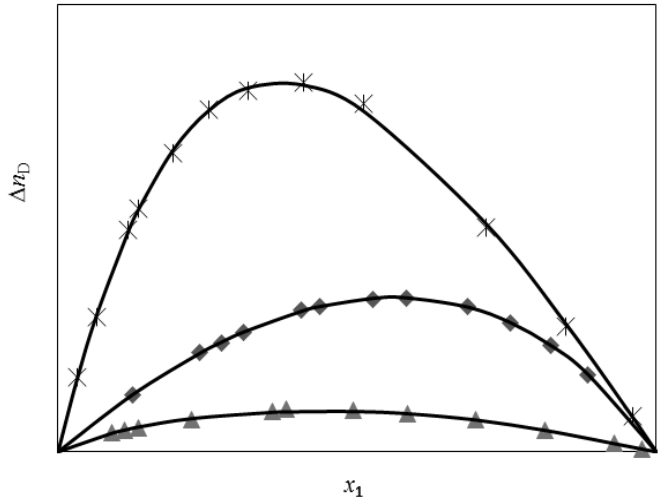

Figure 3. Variation of deviation in the refractive index Δn_D with mole fraction x_1 for the binary systems at T = 298.15K: ◆ dimethylacetamide + dimethylbenzylamine; ▲ dimethylacetamide + dichloromethane ; ∗ dimethylbenzylamine + dichloromethane. Solid curves were calculated from the Redlich–Kister equation

Table 3. Binary Coefficients of the Redlich–Kister Equation at 298.15K and Ternary Coefficients of the Cibulka Equation for V_m^E, $\Delta\eta$, and Δn_D at 298.15 K

ΔQ_{ij}	A_0	A_1	A_2	A_3	σ
		(x) dimethylacetamide + (1 – x) dimethylbenzylamine			
V_m^E (cm^3·mol^{-1})	-0.953	0.083	-0.1375	0.0576	3.1×10^{-3}
$\Delta\eta$ (mPa·s)	-0.1812	-0.0218	0.014	0.0905	6.3×10^{-4}
Δn_D	0.0274	0.0067	0.0023	0.0004	3×10^{-5}
		(x) dimethylacetamide + (1 – x) dichloromethane			
V_m^E (cm^3·mol^{-1})	0.955	-1.055	-2.627	-1.376	9.1×10^{-2}
$\Delta\eta$ (mPa·s)	-0.142	-0.008	0.021	0.025	2.3×10^{-4}
Δn_D	0.007	-0.001	0.001	-0.002	9.7×10^{-5}
		(x) dimethylbenzylamine + (1 – x) dichloromethan			
V_m^E (cm^3·mol^{-1})	0.26	-1.675	0.576	-0.552	8.1×10^{-2}
$\Delta\eta$ (mPa·s)	-0.625	0.003	-0.024	-0.139	6.1×10^{-3}
Δn_D	0.062	-0.031	0.006	0.001	9.1×10^{-4}
ΔQ_{123}	B_0	B_1	B_2		σ
V_m^E (cm^3·mol^{-1})	-1.81855	6.40231	0.460098		2×10^{-2}
$\Delta\eta$ (mPa·s)	0.39927	-0.84597	-0.51009		4×10^{-3}
Δn_D	0.024273	0.027428	0.028409		3.8×10^{-4}

Table 4. Experimental Densities ρ, Viscosities η ,Refractive Indices n_D , Excess Molar Volumes V_m^E, Deviation in the Viscosity $\Delta\eta$, and Deviation in the Refractive Index Δn_D for the Ternary System of Dimethylacetamide, Dimethylbenzylamine and Dichloromethane at 298.15 K

x_1	x_2	ρ (g.cm^{-3})	η (mPa.s)	n_D	V_m^E (cm^3.mol^{-1})	$\Delta\eta$ (mPa.s)	Δn_D
0.0505	0.0401	1.25193	0.480	1.4297	0.123	-0.024	0.0039
0.0512	0.0810	1.21815	0.513	1.4363	0.181	-0.047	0.0074
0.0530	0.2091	1.13308	0.636	1.4526	0.223	-0.101	0.0138
0.0538	0.2537	1.10870	0.684	1.4569	0.216	-0.114	0.0146
0.0554	0.3930	1.04590	0.846	1.4687	0.112	-0.144	0.0156
0.0558	0.4419	1.02730	0.907	1.4724	0.072	-0.150	0.0155
0.0577	0.5954	0.97760	1.124	1.4814	-0.059	-0.143	0.0127
0.0593	0.6485	0.96255	1.204	1.4840	-0.087	-0.136	0.0112
0.0614	0.8176	0.92170	1.496	1.4919	-0.140	-0.077	0.0060
0.0816	0.8546	0.91044	1.579	1.4931	-0.130	-0.055	0.0041
0.0614	0.8757	0.90948	1.604	1.4942	-0.115	-0.048	0.0038
0.1011	0.0401	1.22760	0.497	1.4306	0.146	-0.034	0.0041
0.1055	0.2090	1.11485	0.669	1.4527	0.210	-0.095	0.0131
0.1105	0.3933	1.03112	0.884	1.4689	0.080	-0.135	0.0151
0.1160	0.5948	0.96540	1.169	1.4810	-0.083	-0.128	0.0116
0.2025	0.0402	1.18161	0.548	1.4319	0.198	-0.036	0.0040
0.2063	0.1226	1.12695	0.621	1.4428	0.229	-0.078	0.0085
0.2108	0.2091	1.07980	0.717	1.4526	0.198	-0.102	0.0116
0.2154	0.2992	1.03898	0.827	1.4609	0.127	- 0.117	0.0129
0.2234	0.3855	1.00490	0.943	1.4676	0.042	-0.124	0.0128
0.2260	0.4910	0.97103	1.088	1.4747	-0.058	-0.125	0.0117
0.2311	0.5954	0.94185	1.259	1.4808	-0.143	-0.099	0.0097
0.2372	0.7035	0.91548	1.449	1.4861	-0.193	-0.059	0.0066
0.3032	0.0401	1.13975	0.592	1.4338	0.243	-0.044	0.0045
0.3165	0.2092	1.04701	0.781	1.4530	0.181	-0.094	0.0105
0.3316	0.3926	0.97651	1.025	1.4681	-0.021	-0.108	0.0113
0.3472	0.5951	0.91960	1.354	1.4805	-0.205	-0.063	0.0077
0.4045	0.0402	1.10088	0.641	1.4345	0.262	-0.049	0.0039
0.4128	0.1233	1.05573	0.733	1.4449	0.219	-0.074	0.0077
0.4221	0.2089	1.01680	0.841	1.4538	0.132	-0.087	0.0099
0.4314	0.2987	0.98253	0.968	1.4612	-0.015	-0.089	0.0103
0.4414	0.3933	0.95167	1.105	1.4681	-0.106	-0.086	0.0039
0.4513	0.4918	0.92400	1.268	1.4745	-0.209	-0.063	0.0084
0.5052	0.0402	1.06583	0.708	1.4359	0.218	-0.034	0.0039

Table 4. Continued…

x_1	x_2	ρ (g.cm^{-3})	η (mPa.s)	n_D	V_m^E (cm^3.mol^{-1})	$\Delta\eta$ (mPa.s)	Δn_D
0.5272	0.2092	0.98891	0.908	1.4540	-0.030	-0.076	0.0086
0.5518	0.3931	0.92834	1.190	1.4682	-0.198	-0.059	0.0083
0.6060	0.0402	1.03378	0.750	1.4369	0.114	-0.044	0.0035
0.6321	0.2092	0.96300	0.982	1.4542	-0.099	-0.057	0.0074
0.6475	0.2984	0.93305	1.111	1.4617	-0.189	-0.058	0.0078
0.7073	0.0402	1.00415	0.810	1.4379	-0.036	-0.037	0.0031
0.7376	0.2093	0.93792	1.051	1.4546	-0.177	-0.043	0.0064
0.8583	0.0404	0.96244	0.911	1.4388	-0.187	-0.015	0.0019
0.8677	0.0809	0.94577	0.964	1.4433	-0.159	-0.020	0.0032
0.9089	0.0403	0.94850	0.938	1.4395	-0.153	-0.014	0.0019

Conclusion

Density, viscosity, and refractive index measurements were experimentally conducted for systems of dimethylacetamide, dimethylbenzylamine, and dichloromethane. Results showed that some solutions exhibited positive values for V_m^E, while others showed negative values. The strong bipolar-bipolar interactions between the molecules of dimethylacetamide + dimethylbenzylamine system resulted in a negative deviation, whereas undesirable interactions between dissimilar molecules in the dimethylacetamide + dichloromethane and dimethylbenzylamined + dichloromethane systems led to a positive deviation. The behavior of the solution was the result of these mutual effects. Negative values for viscosity deviation $\Delta\eta$ indicated that factors such as molecular size, the shape of the ingredients, and spatial formation of molecules, in addition to the existence of forces and specific interactions among the mixture's ingredients, were also important. Shape and size of the ingredients of the alkyl group and benzenoid loop caused spatial inhibition, preventing the molecules from becoming closer to each other. As a result, the solution had a lower viscosity than each of the pure ingredients, leading to a negative viscosity deviation. Furthermore, the deviation values for the refractive index in all mole fractions were positive, with negative $\Delta\eta$ values confirming this observation.

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Disclosure statement

The authors declare that they have no conflict of interest.

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