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VISCOSITY STUDIES ON SOLUTE–SOLVENT INTERACTIONS IN DIETHYL MALONATE–BRANCHED ALCOHOL SYSTEMS

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ABSTRACT

Diethyl malonate binary mixtures with 3-methyl-1-butanol, 2-methyl-1-propanol, 2-propanol, and 2-butanol were evaluated at intervals of 5 K throughout the ester's full composition range from 303.15–318.15 K during a dynamic viscosity and density analysis. In order to summarise the experimental results, viscosity deviations (Δη) and excess Gibbs free energy of activation for viscous flow (ΔG*E) were calculated. The results were correlated with the Redlich–Kister polynomial model to determine fitting parameters and standard deviations. Additionally, several semi-empirical models have been proposed for predicting theoretical viscosities, including Grunberg-Nissan, Heric-Brewer, Katti-Chaudhri, Hind et al., McAllister, Kendall–Monroe, and Frenkel. The Hind et al. and McAllister four-body approaches were the closest to experimental values. In these binary systems, the observed variations are further analysed in relation to intermolecular interactions

Keywords: Dynamic Viscosity, Density, Diethyl Malonate, Molecular Collaboration.

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INTRODUCTION

Molecular thermodynamics offers valuable insights into the nature of molecular interactions between molecules, which are essential to the analysis and design of mechanical systems that transfer heat or mass.[1] In this context, a binary system comprising diethyl malonate (DEM) and branched alcohols[2] is investigated due to its significance in various industrial applications. Diethyl malonate, an ester derived from malonic acid, is a clear, colourless liquid that possesses a mild fruity odour often compared to that of freshly crushed apples. It is extensively utilised in the fragrance sector and functions as a crucial step in the production of barbiturates, vitamins B_1 and B_6, and diverse flavouring compounds.[3] Alcohols, owing to their versatility, are extensively used not only as solvents but also as functional additives across diverse sectors, including pharmaceuticals, cosmetics, perfumes, flavour formulations, paint strippers, dyes, disinfectants, and veterinary medicine.[4] The current examination explores the viscometric performance of binary systems composed of diethyl malonate and branched alcohols through the measurement of viscosity, viscosity deviation, and excess Gibbs free energy of activation for viscous flow. The experimental data were interpreted using several established semi-empirical viscosity relations, including the models of Grunberg-Nissan, Katti-Chaudhari, Heric-Brewer, Hind *et al.* McAllister, Kendall-Monroe, and Frenkel models approach derived from Eyring's theory.[3-8] The predictive capability of these models was evaluated by comparing the calculated values with the experimentally observed results.[9-10] While the physicochemical properties of ester-containing liquids have been widely reported, systematic studies on mixtures of diethyl malonate with branched alcohols are scarce. In this context, the present work contributes novel and detailed viscometric data for diethyl malonate mixed with four different branched alcohols, allowing a comparative assessment of their behaviour over a range of temperatures. Viscosity

and density measurements were experimentally determined for binary mixtures of diethyl malonate with 3-methyl-1-butanol, 2-methyl-1-propanol, 2-propanol, and 2-butanol over a temperature range of 303.15–318.15 K at four fixed intervals. Using the experimentally obtained density and viscosity values, viscosity deviations (Δη) and the excess Gibbs free energy of activation for viscous flow (ΔG^{*E}) were evaluated. Measurements were performed over the full range of mixture compositions to gain insight into the nature and magnitude of intermolecular interactions among the components. The analysis enables a detailed understanding of molecular associations within the systems and elucidates the role of alcohol molecular structure and temperature in governing the observed viscometric behaviour.

EXPERIMENTAL

Material and Methods

Diethyl malonate and the selected branched alcohols were obtained from Merck and purified following established procedures reported in the literature.[11,12] Before use, all pure liquids were stored over activated molecular sieves to remove residual moisture. Binary mixtures were again prepared by mass using an electronic balance (Shimadzu AY120) with an accuracy of $\pm 1.0 \times 10^{-7}$ kg and stored in airtight containers to avoid contamination. The uncertainty associated with mole fraction determination was within $\pm 1.0 \times 10^{-4}$. Each mixture was thoroughly homogenised, and all experimental measurements were completed within 24 h of sample preparation. The expanded uncertainty in molar quantities was estimated to be below 0.004% at a confidence level of 95% (k = 2). Density (ρ) measurements of the pure components and their corresponding mixtures were performed using a double-arm pycnometer with a nominal volume of 10^{-5} m^3.[12,13] The reported density values represent the average of three independent measurements, with an estimated uncertainty of approximately 2×10^{-4}. The reproducibility of mole fraction measurements was maintained within ±0.0002. Density and viscosity were measured at precisely maintained temperatures with the aid of a microprocessor-controlled circulating water bath, which provided thermal regulation to within ±0.01 K. Throughout the experimental procedure, special precautions were implemented to prevent sample evaporation and ensure the reliability of the measured data.[14,15] The experimentally determined density and viscosity values at 303.15 K were further validated through comparison with available literature data, as presented in Table-1.

Table-1: Comparative Analysis of the Pure Liquids' Density (ρ) and Viscosity (η) at 303.15 K using Data from Published Literature

Compound	ρ (10^{-3} kg m^{-3})		η (m Pa s)	
	Expt.	Lit.	Expt.	Lit.
Diethyl malonate	1.0433	1.0443[15]	1.730	1.732[20]
3-methyl 1-butanol	0.8078	0.8075[16]	2.920	2.927[21]
2-methyl 1-propanol	0.7933	0.7936[17]	2.855	2.845[2]
2-propanol	0.7764	0.7766[18]	1.801	1.80[18]
2-butanol	0.7984	0.7984[19]	2.585	2.597[22]

Background Theory

Viscosity deviations (Δη) of binary liquid mixtures were computed using Equation (1), where x_i and η_i are the mole fraction and viscosity of pure component I, respectively, and η is the mixture's viscosity.[15,16]

$$\Delta\eta = \eta_{12} - x_1\eta_1 - x_2\eta_2 \qquad (1)$$

The viscosity behaviour of the mixtures was analysed using widely accepted relations, namely the Grunberg-Nissan, Katti-Chaudhri, Heric-Brewer, Hind *et al*., McAllister, Kendall–Monroe, and Frenkel models, all rooted in Eyring's absolute reaction rate theory.[15]

$$\ln\eta = x_1 \ln\eta_1 + x_2 \ln\eta_2 + x_1 x_2 G_{12} \qquad (2)$$

These models incorporate interaction or association parameters to describe non-ideal molecular interactions in binary systems.[3–10] The McAllister four-body association model was employed to correlate kinematic viscosity data by accounting for interactions among like and unlike molecules.[7] The excess Gibbs free energy of activation for viscous flow (ΔG^{*E}) was evaluated from viscosity and molar volume

data and correlated using the Redlich–Kister polynomial equation, with the quality of fit assessed via standard deviation analysis.[13,14]

RESULTS AND DISCUSSION

In Table-2, the dynamic viscosities, viscosity deviations, and Gibbs free energies of activation of four binary liquid mixtures were determined experimentally over the temperature range of 303.15–318.15 K, namely diethyl malonate in combination with 3-methyl-1-butanol, 2-methyl-1-propanol, 2-propanol, and 2-butanol. The composition dependence of the deviation and excess functions was correlated using the Redlich–Kister polynomial relationship[15], with explicit consideration of temperature effects. The optimized Redlich–Kister coefficients (A_i^{-1}) for each binary system, along with the corresponding standard deviation (σ) evaluated as the root mean square deviation, are presented in Table-3. These fitted parameters and excess functions were subsequently utilised to elucidate the nature and strength of intermolecular interactions between diethyl malonate and the respective alcohols in the investigated mixtures.

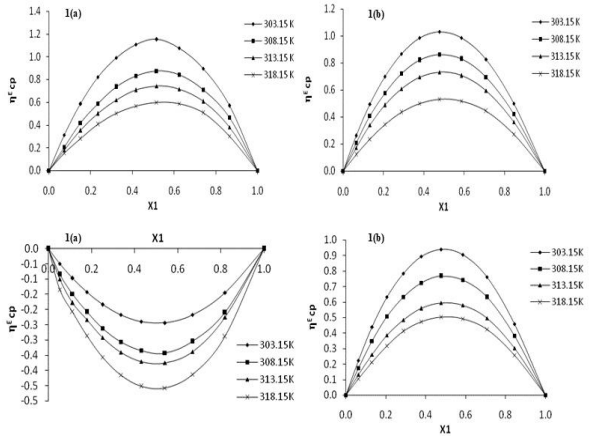

Fig.-1: Variation of Excess Viscosity (Δη / mPa·s) as a Function of the Mole Fraction of Diethyl Malonate (x_1) for the Binary Mixtures: Diethyl Malonate + system(a): 3-methyl-1-butanol, System(b):2-methyl-1-propanol, System(c): 2-propanol and System (d): 2-butanol (2)

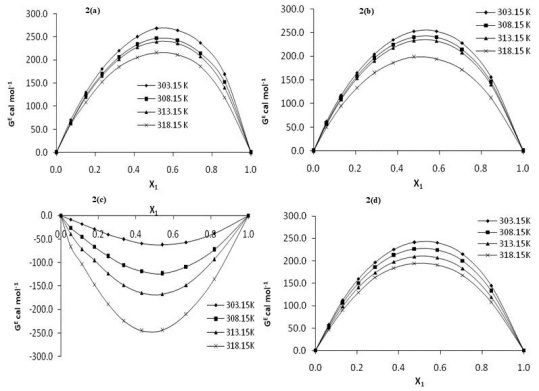

Fig.-2: Dependence of the Excess Gibbs Free Energy of Activation for Viscous Flow, ΔG^{*E} (cal mol^{-1}), on the Mole Fraction of Diethyl Malonate (x_1) for the Binary Systems: Diethyl Malonate + System (a):3-methyl-1-butanol, System (b):2-methyl-1-Propanol, System(c): 2-propanol and System(d): 2-butanol (2).

Viscosity Deviation

In binary mixes of 3-methyl-1-butanol, 2-methyl-1-propanol, 2-propanol, and 2-butanol with diethyl malonate at T = (303.15 to 318.15) K with a rise of 5K, Fig.-1 illustrates the change in viscosity, Δη, with the mole fraction of diethyl malonate (x1). Regardless of intermolecular pressures, the size and shape of the segments determine the value and extent of Δη. The deviancy in viscosity is positive for all binaries and temperatures, as shown in Fig.-1(a), 1(b), and 1(d). However, in Fig.-1(c), the viscosity deviation is negative for the mixture diethyl malonate + 2-proponal at all temperatures. The outright estimation of viscosity deviation of system (a), (b), (c) & (d) expands directly and spans to a most extreme incentive at x_1~ 0.5163 for (system (a)), x_1~ 0.4775 for (system (b)), x_1~ 0.5408 for (system (c)), x_1~ 0.5855 for (system (d)) after which step by step diminishes until to an unadulterated condition of diethyl malonate.

The Magnitude of the Viscosity Deviation (Δη) and its Sign rely upon the aggregate impact of components, for example, atomic size, shape and intermolecular forces.[23] The positive estimations of deviation in viscosity show the presence of a particular connection, for example, the improvement of hydrogen holding of the sort O-H……O. The negative estimations of deviation in viscosity in the systems of diethyl malonate with 2- propanol suggest that shared loss of explicit communications in like atoms exceeds the particular associations among not at all like particles.[24,25] Negative deviation in viscosity may likewise happen where dispersion forces are predominant, especially, for the systems having different molecular sizes.[25, 26] Specific concoction collaborations, including hydrogen bond improvement and

dipole-dipole associations among various portions of fluid mixtures, make the structure more conservative and result in positive viscosity deviations. The sign and magnitude of Δη furthermore shift with structural qualities of fluid sections emerging from geometrical fitting of one portion into the structure of another fragment due to distinctions in molecular size and shape of segments. According to Kauzmann and Eyring, a mixture's viscosity strongly depends on its entropy, which is related to the fluid's structure.[27] Vogel and Weiss explained that whereas mixtures with positive deviations from Raoult's law and no explicit relationships exhibit negative viscosity deviations, mixtures with firm connections between different particles and negative deviations from Raoult's law exhibit positive viscosity deviations.[28] The variety in Δη with the mole fraction of diethyl malonate is introduced in Table-2. The deviation in viscosity variety invigorates a qualitative assessment of the strength of the intermolecular connections. The deviancy in viscosity, Δη, from the linear reliance on mole fraction reveals that it is positive for the three binary mixtures investigated and negative for diethyl malonate with the 2-propanol system. It is moreover evident from the outcomes that Δη assessments of the mixtures diminish as the test temperatures rise, thereby suggesting an expansion in the fluidity of the mixtures. It has been represented for those positive deviations in Δη that occur in the mixtures where specific associations are generally responsible for the connection among the fragment molecules. Similar behaviour in Δη was moreover suggested.[29]

Table-2: For Binary Mixes of Diethyl Malonate with Branched Alcohols at Different Temperatures, the Active Viscosity, η (m Pa·s), Deviancy in Viscosity, Δη (m Pa·s), and Excess Gibbs Free Energy of Activation of Viscous Flow, ΔG*E (cal mol-1) were Measured

	303.15^{0}K			308.15^{0}K			313.15^{0}K			318.15^{0}K		
X_1	H_1	$\Delta\eta_1$	ΔG^{*E}	η_1	X_1	H_1	$\Delta\eta_1$	ΔG^{*E}	η_1	X_1	H_1	$\Delta\eta_1$
diethyl malonate + 3-methyl 1-butanol												
0.0000	2.920	0.0000	0.0000	2.055	0.0000	0.0000	1.775	0.0000	0.0000	1.529	0.0000	0.0000
0.0733	3.145	0.3120	70.2600	2.230	0.2080	63.3200	1.930	0.1780	62.9600	1.673	0.1520	61.6300
0.1510	3.325	0.5850	129.9200	2.405	0.4180	122.7400	2.079	0.3520	119.8900	1.791	0.2780	108.9100
0.2337	3.462	0.8200	181.1700	2.538	0.5890	168.8900	2.200	0.5000	165.6000	1.909	0.4050	152.9600
0.3217	3.526	0.9890	220.0700	2.642	0.7330	206.7100	2.292	0.6200	201.6500	1.999	0.5040	185.5000
0.4157	3.531	1.1060	250.0100	2.698	0.8310	233.2000	2.349	0.7070	227.6300	2.053	0.5680	205.5000
0.5163	3.459	1.1530	268.2200	2.696	0.8750	246.9100	2.353	0.7430	239.6000	2.075	0.6000	215.2200
0.6241	3.254	1.0770	264.0000	2.613	0.8410	242.8100	2.293	0.7170	235.6400	2.051	0.5880	211.6000
0.7400	2.936	0.8970	237.1600	2.430	0.7100	214.8100	2.146	0.6070	207.7300	1.956	0.5050	186.1100
0.8649	2.460	0.5690	169.0700	2.130	0.4670	152.7000	1.879	0.3800	140.2800	1.741	0.3030	118.4300
1.0000	1.730	0.0000	0.0000	1.602	0.0000	0.0000	1.456	0.0000	0.0000	1.424	0.0000	0.0000
diethyl malonate + 2-methyl 1-propanol												
0.0000	2.855	0.0000	0.0000	2.435	0.0000	0.0000	2.111	0.0000	0.0000	1.623	0.0000	0.0000
0.0634	3.045	0.2610	62.7600	2.592	0.2100	59.0000	2.242	0.1730	56.4200	1.731	0.1200	49.5900
0.1322	3.200	0.4940	117.3400	2.732	0.4070	112.3100	2.365	0.3410	108.7300	1.834	0.2370	94.6900
0.2070	3.321	0.6990	164.9400	2.841	0.5780	157.8900	2.463	0.4880	153.4500	1.926	0.3440	133.4700
0.2888	3.397	0.8670	204.8600	2.915	0.7210	196.0000	2.530	0.6090	190.3300	1.998	0.4330	164.5300
0.3786	3.413	0.9840	235.1500	2.940	0.8200	224.5500	2.558	0.6950	217.8500	2.045	0.4980	186.6400
0.4775	3.349	1.0320	253.0500	2.900	0.8630	240.8000	2.530	0.7320	233.2400	2.058	0.5300	198.1300
0.5870	3.181	0.9860	253.2500	2.775	0.8290	240.1400	2.433	0.7060	232.5000	2.023	0.5170	194.6500
0.7090	2.882	0.8250	227.7500	2.540	0.6960	214.6900	2.243	0.5960	207.6700	1.925	0.4430	171.3600
0.8457	2.401	0.4980	155.3400	2.153	0.4230	145.2700	1.921	0.3640	139.8100	1.727	0.2720	111.3500
1.0000	1.730	0.0000	0.0000	1.602	0.0000	0.0000	1.456	0.0000	0.0000	1.424	0.0000	0.0000
diethyl malonate + 2-propanol												
0.0000	1.801	0.0000	0.0000	1.562	0.0000	0.0000	1.361	0.0000	0.0000	1.191	0.0000	0.0000
0.0531	1.748	-0.0490	-8.1200	1.480	-0.0850	-25.7900	1.266	-0.1000	-39.1800	1.069	-0.1350	-67.0100
0.1120	1.696	-0.0970	-17.7600	1.419	-0.1480	-45.2500	1.193	-0.1780	-71.7500	1.010	-0.2080	-103.2900
0.1778	1.645	-0.1430	-28.5500	1.361	-0.2080	-66.2700	1.145	-0.2330	-95.3200	0.946	-0.2860	-146.7800
0.2518	1.599	-0.1840	-39.9700	1.310	-0.2620	-87.1000	1.093	-0.2920	-123.5900	0.892	-0.3570	-188.8100
0.3354	1.559	-0.2180	-50.5500	1.269	-0.3060	-105.6100	1.053	-0.3400	-148.1700	0.854	-0.4160	-224.3000
0.4309	1.530	-0.2400	-58.8900	1.244	-0.3350	-118.7500	1.031	-0.3710	-164.8000	0.838	-0.4530	-245.8700
0.5408	1.520	-0.2430	-62.2800	1.240	-0.3430	-123.9400	1.037	-0.3750	-167.3600	0.859	-0.4580	-244.7100

0.6687	1.537	-0.2170	-57.1300	1.284	-0.3050	-109.0400	1.086	-0.3380	-147.7700	0.934	-0.4130	-210.0400
0.8196	1.598	-0.1450	-38.3200	1.385	-0.2100	-73.0100	1.213	-0.2260	-93.0100	1.093	-0.2890	-134.7100
1.0000	1.730	0.0000	0.0000	1.602	0.0000	0.0000	1.456	0.0000	0.0000	1.424	0.0000	0.0000
diethyl malonate + 2-butanol												
0.0000	2.585	0.0000	0.0000	2.154	0.0000	0.0000	1.724	0.0000	0.0000	1.541	0.0000	0.0000
0.0630	2.753	0.2220	58.2600	2.294	0.1750	54.8400	1.838	0.1310	50.9400	1.639	0.1050	46.2700
0.1314	2.910	0.4380	112.3100	2.430	0.3490	106.3900	1.950	0.2620	98.7600	1.738	0.2130	89.9700
0.2060	3.040	0.6310	159.4500	2.546	0.5060	150.6800	2.053	0.3850	140.7800	1.834	0.3170	129.5500
0.2875	3.123	0.7840	196.9800	2.628	0.6330	186.1000	2.131	0.4840	173.5200	1.918	0.4100	162.9000
0.3771	3.156	0.8930	225.3400	2.670	0.7240	212.3600	2.181	0.5580	197.6100	1.970	0.4730	184.4000
0.4759	3.118	0.9400	241.3900	2.657	0.7660	226.6400	2.190	0.5940	209.7400	1.989	0.5030	194.4700
0.5855	2.989	0.9050	240.6900	2.572	0.7410	224.7500	2.146	0.5790	206.8500	1.966	0.4930	190.8300
0.7077	2.741	0.7610	215.1300	2.394	0.6300	200.4300	2.031	0.4970	183.2700	1.883	0.4250	167.9800
0.8449	2.320	0.4580	144.2500	2.068	0.3810	132.7200	1.799	0.3020	119.4000	1.699	0.2570	107.1500
1.0000	1.730	0.0000	0.0000	1.602	0.0000	0.0000	1.456	0.0000	0.0000	1.424	0.0000	0.0000

Excess Gibbs Energy Linked to the Activation Mechanism Underlying Fluid Viscosity

When assessing the kind and intensity of intermolecular interactions in liquid mixtures, the excess Gibbs free energy of activation for viscous flow ΔG^E is a very sensitive indication. The composition dependence of ΔG^E for diethyl malonate–based binary systems at 303.15–318.15 K is illustrated in Fig.-2(a–d), with values derived from experimental viscosity data. Positive ΔG^E values generally reflect strong attractive interactions, indicating efficient accommodation of one component within the structural environment of the other. In binary mixtures, ΔG^E arises from both chemical interactions and physical or structural contributions. Large positive values are commonly observed in systems containing polar components, where associative interactions dominate, and in mixtures with significant molecular size mismatch.[30] Thus, ΔG^E analysis provides valuable insight into mixing behaviour and supports the evaluation of transport properties and theoretical interaction models.[31] Moreover, positive excess Gibbs free energy values indicate specific intermolecular forces like dipole–dipole interactions and hydrogen bonding, whereas negative values suggest weak dispersive interactions between unlike molecules.[32,33]

Investigating Viscosity of Fluid Mixtures Through Semi-Experimental Representations

Numerous empirical and semi-empirical correlations have been established to explain how viscosity and intermolecular interactions in liquid mixtures are related. The current study used the Grunberg-Nissan, Katti-Chaudhari, Heric-Brewer, Hind *et al.* McAllister, Kendall, Monroe, and Frenkel formulations derived from Eyring's theory to interpret the viscosity data of binary systems comprising diethyl malonate and branched alcohols, specifically 3-methyl-1-butanol, 2-methyl-1-propanol, 2-propanol, and 2-butanol. The experimentally measured and theoretically calculated viscosity (η) values for these mixtures at 303.15, 308.15, 313.15, and 318.15 K are reported in Tables-4(a) to 4(d). Based on the methodology described in Reference[25], the adjustable interaction parameters are presented in Tables 5(a) and 5(b) for the binary systems compared at the same temperatures. The viscosity (η) values determined experimentally and those predicted using the theoretical models for the investigated mixtures at 303.15, 308.15, 313.15, and 318.15 K are compiled in Tables-4(a) to 4(d). Tables-5(a) and 5(b) provide a summary of the fitted interaction parameters derived from the reported literature[25] over the same temperature range for all binary systems. According to Fort and Moore, the degree of interaction between components in a mixture can be accurately determined using the interaction parameter G12.[25]

Table-3: Redlich–Kister Coefficients (A_{i-1}) along with the Associated Standard Deviations (σ), Derived from Correlating the Excess/Deviation Thermodynamic Properties of Binary Systems Containing Diethyl Malonate and Branched-Chain Alcohols Across a Range of Temperatures

Property	Temp(K)	A_0	A_1	A_2	A_3	A_4	σ
Diethyl malonate + 3-methyl 1-butanol							
Δη (m Pa s)	303.15	4.5584	-0.0518	0.3133	-0.2603	0.0353	0.0071
	308.15	3.4762	-0.3548	0.2382	-0.3976	-0.0681	0.0056
	313.15	2.9649	-0.3376	0.1144	-0.0754	-0.1724	0.0018

	318.15	2.4004	-0.4055	0.1815	0.3154	-0.271	0.0056
ΔG^{*E}(cal mol^{-1})	303.15	1055.413	-198.548	285.4709	-175.18	93.9479	1.4956
	308.15	979.3819	-150.59	245.5097	-192.548	42.6652	1.6193
	313.15	955.5277	-140.898	204.2526	-88.5992	-3.1169	0.5685
	318.15	861.2374	-122.728	184.145	61.489	-39.8081	1.9259
Diethyl malonate + 2-methyl 1-propanol							
Δη (m Pa s)	303.15	4.1253	0.2875	-0.2535	0.1212	0.2269	0.0034
	308.15	3.4516	0.1618	-0.1082	0.0946	-0.0628	0.0017
	313.15	2.9297	0.0991	0.0107	0.0551	-0.2536	0.0013
	318.15	2.1256	-0.0409	0.0146	0.0752	-0.2238	0.0012
ΔG^{*E}(cal mol^{-1})	303.15	1019.785	-95.966	147.1411	-15.282	10.5426	0.3846
	308.15	969.3139	-95.966	147.1411	-15.282	10.5426	0.3846
	313.15	938.3677	-89.8646	166.2038	-10.0967	-51.0693	0.3157
	318.15	793.9669	-31.5975	118.8616	17.8459	-57.9741	0.5022
Diethyl malonate + 2-propanol							
Δη (m Pa s)	303.15	-0.9781	0.0038	-0.0104	-0.0045	0.0184	0.0002
	308.15	-1.3846	-0.0224	0.2483	0.0551	-0.8204	0.0028
	313.15	-1.5239	-0.1332	0.4713	0.2082	-1.3649	0.0075
	318.15	-1.8727	-0.2745	0.9816	0.6807	-2.8786	0.0141
ΔG^{*E}(cal mol^{-1})	303.15	-247.416	45.3351	49.736	10.1495	-2.4741	0.0614
	308.15	-498.746	19.7485	196.3611	29.5527	-328.733	1.1619
	313.15	-680.798	-56.2018	355.9968	95.0426	-627.478	3.4823
	318.15	-1009.68	-236.578	792.7115	405.4589	-1620.77	7.7771
Diethyl malonate + 2-butanol							
Δη (m Pa s)	303.15	3.0615	0.0598	0.1037	0.1246	-0.5354	0.0016
	308.15	3.0615	0.0598	0.1037	0.1246	-0.5354	0.0016
	313.15	2.379	-0.0316	0.0752	0.099	-0.4392	0.0018
	318.15	2.0303	-0.0289	-0.1371	-0.04	-0.1634	0.0037
ΔG^{*E}(cal mol^{-1})	303.15	970.0973	-92.7708	203.4033	13.1205	-113.865	0.3168
	308.15	909.6781	-65.7754	180.3068	7.3576	-113.192	0.5122
	313.15	841.3832	-37.8271	157.6675	9.7456	-124.306	0.5968
	318.15	782.9975	-6.695	70.8165	-25.2559	-52.6796	1.3132

A strong molecular connection is indicated by positive values of G12G12, whereas weaker intermolecular interactions within the system are often implied by negative values. Furthermore, investigations on binary and ternary liquid mixtures have led to the establishment of specific criteria for improving the understanding and classification of molecular interactions occurring within these systems.[34-36] When $\Delta\eta > 0$ and $G_{12} > 0$, with both values being large, strong specific interactions are likely to occur.

When $\Delta\eta < 0$ and $G_{12} > 0$, the system indicates the presence of weak interactions. When $\Delta\eta < 0$ and $G_{12} < 0$, and both magnitudes are significant, dispersion forces are considered to be dominant. The interaction parameters Wvis/RTWvis/RT and G_{12} display closely comparable variation patterns. While both parameters describe similar interaction behaviour, minor differences arise because their defining expressions involve logarithmic relationships. The G_{12} values in Table-5(a) are positive for the binary mixes of diethyl malonate with branched alcohols, specifically 3-methyl-1-butanol, 2-methyl-1-propanol, and 2-butanol. The corresponding viscosity deviations ($\Delta\eta > 0$) are similarly positive. The diethyl malonate + 2-propanol combination is the sole exception. All of these findings point to the presence of robust and targeted intermolecular interactions in most of the mixes under investigation.

Table-4(a): Experimental and Calculated Viscosity Values, η (mPa·s), for Binary Mixtures of Diethyl Malonate with 3-methyl-1-butanol at different Temperatures

X_1	η Expt.	ηGN.	η KC.	η HB.	η H.	η Mc.	η KM.	η F.
303.15K								
0.0000	2.920	2.920	2.920	2.920	2.920	2.920	2.920	2.920
0.0733	3.145	3.182	3.174	3.174	3.146	3.154	2.818	3.452
0.1510	3.325	3.411	3.399	3.400	3.332	3.334	2.713	3.979
0.2337	3.462	3.584	3.572	3.573	3.469	3.458	2.604	4.446
0.3217	3.526	3.677	3.667	3.669	3.545	3.522	2.492	4.780

0.4157	3.531	3.662	3.658	3.659	3.547	3.522	2.375	4.904
0.5163	3.459	3.518	3.521	3.521	3.459	3.448	2.254	4.750
0.6241	3.254	3.234	3.244	3.242	3.261	3.272	2.129	4.288
0.7400	2.936	2.818	2.830	2.827	2.928	2.952	2.000	3.551
0.8649	2.460	2.299	2.308	2.306	2.430	2.440	1.867	2.645
1.0000	1.730	1.730	1.730	1.730	1.730	1.730	1.730	1.730
308.15K								
0.0000	2.055	2.055	2.055	2.055	2.055	2.055	2.055	2.055
0.0733	2.230	2.259	2.254	2.254	2.257	1.810	2.019	2.386
0.1510	2.405	2.449	2.440	2.441	2.431	1.580	1.982	2.715
0.2337	2.538	2.610	2.601	2.602	2.570	1.391	1.942	3.015
0.3217	2.642	2.725	2.718	2.719	2.666	1.254	1.901	3.248
0.4157	2.698	2.773	2.771	2.772	2.709	1.177	1.858	3.372
0.5163	2.696	2.736	2.739	2.739	2.687	1.168	1.812	3.345
0.6241	2.613	2.597	2.605	2.604	2.586	1.231	1.764	3.137
0.7400	2.430	2.353	2.363	2.361	2.387	1.366	1.713	2.747
0.8649	2.130	2.012	2.019	2.018	2.068	1.537	1.659	2.210
1.0000	1.602	1.602	1.602	1.602	1.602	1.602	1.602	1.602
313.15K								
0.0000	1.775	1.775	1.775	1.775	1.775	1.775	1.775	1.775
0.0733	1.930	1.948	1.944	1.944	1.951	1.573	1.750	2.087
0.1510	2.079	2.111	2.104	2.104	2.103	1.391	1.724	2.404
0.2337	2.200	2.251	2.243	2.244	2.225	1.241	1.697	2.700
0.3217	2.292	2.353	2.347	2.349	2.312	1.133	1.668	2.937
0.4157	2.349	2.402	2.400	2.401	2.354	1.072	1.637	3.074
0.5163	2.353	2.380	2.384	2.383	2.342	1.063	1.605	3.068
0.6241	2.293	2.275	2.282	2.281	2.263	1.110	1.571	2.887
0.7400	2.146	2.079	2.089	2.087	2.103	1.216	1.535	2.528
0.8649	1.879	1.800	1.807	1.805	1.842	1.359	1.497	2.026
1.0000	1.456	1.456	1.456	1.456	1.456	1.456	1.456	1.456
318.15K								
0.0000	1.529	1.529	1.529	1.529	1.529	1.529	1.529	1.529
0.0733	1.673	1.675	1.671	1.671	1.683	1.363	1.521	1.763
0.1510	1.791	1.814	1.808	1.809	1.819	1.220	1.513	1.999
0.2337	1.909	1.938	1.932	1.933	1.931	1.106	1.504	2.220
0.3217	1.999	2.036	2.031	2.033	2.015	1.023	1.495	2.402
0.4157	2.053	2.095	2.093	2.094	2.064	0.976	1.485	2.518
0.5163	2.075	2.100	2.104	2.103	2.070	0.968	1.474	2.537
0.6241	2.051	2.040	2.047	2.046	2.023	1.006	1.463	2.436
0.7400	1.956	1.905	1.914	1.913	1.910	1.096	1.451	2.204
0.8649	1.741	1.697	1.704	1.702	1.717	1.241	1.438	1.854
1.0000	1.424	1.424	1.424	1.424	1.424	1.424	1.424	1.424

Table-4(b): Measured and Predicted Viscosity Values, η (mPa·s), for Binary Mixtures of Diethyl Malonate with 2-methyl-1-propanol at different Temperatures

X_1	η Expt.	ηGN.	η KC.	η HB.	η H.	η Mc.	η KM.	η F.
303.15K								
0.0000	2.855	2.855	2.855	2.855	2.855	2.855	2.855	2.855
0.0634	3.045	3.050	3.043	3.038	3.030	3.061	2.772	3.303
0.1322	3.200	3.229	3.217	3.210	3.183	3.235	2.684	3.764
0.2070	3.321	3.374	3.362	3.356	3.304	3.369	2.591	4.203
0.2888	3.397	3.466	3.459	3.454	3.383	3.457	2.491	4.564
0.3786	3.413	3.481	3.481	3.480	3.406	3.490	2.385	4.770
0.4775	3.349	3.392	3.399	3.404	3.354	3.456	2.271	4.737
0.5870	3.181	3.173	3.188	3.196	3.202	3.323	2.149	4.390
0.7090	2.882	2.813	2.829	2.839	2.915	3.039	2.019	3.707
0.8457	2.401	2.318	2.329	2.336	2.446	2.524	1.880	2.760
1.0000	1.730	1.730	1.730	1.730	1.730	1.730	1.730	1.730
308.15K								
0.0000	2.435	2.435	2.435	2.435	2.435	2.435	2.435	2.435
0.0634	2.592	2.599	2.592	2.589	2.587	2.521	2.375	2.780
0.1322	2.732	2.750	2.741	2.735	2.721	2.594	2.311	3.132
0.2070	2.841	2.877	2.867	2.861	2.830	2.653	2.243	3.464

0.2888	2.915	2.962	2.956	2.952	2.904	2.697	2.170	3.738
0.3786	2.940	2.988	2.987	2.987	2.932	2.721	2.092	3.900
0.4775	2.900	2.930	2.937	2.941	2.899	2.715	2.008	3.887
0.5870	2.775	2.768	2.781	2.789	2.784	2.655	1.918	3.643
0.7090	2.540	2.488	2.502	2.511	2.557	2.497	1.821	3.143
0.8457	2.153	2.090	2.100	2.106	2.181	2.168	1.716	2.423
1.0000	1.602	1.602	1.602	1.602	1.602	1.602	1.602	1.602
313.15K								
0.0000	2.111	2.111	2.111	2.111	2.111	2.111	2.111	2.111
0.0634	2.242	2.250	2.245	2.242	2.243	2.182	2.065	2.384
0.1322	2.365	2.380	2.372	2.367	2.360	2.245	2.015	2.660
0.2070	2.463	2.489	2.481	2.476	2.455	2.300	1.962	2.920
0.2888	2.530	2.566	2.561	2.557	2.522	2.343	1.905	3.133
0.3786	2.558	2.594	2.593	2.593	2.550	2.371	1.844	3.259
0.4775	2.530	2.553	2.559	2.562	2.527	2.371	1.778	3.253
0.5870	2.433	2.426	2.437	2.444	2.435	2.324	1.707	3.070
0.7090	2.243	2.198	2.211	2.219	2.249	2.192	1.630	2.686
0.8457	1.921	1.869	1.877	1.883	1.938	1.920	1.547	2.121
1.0000	1.456	1.456	1.456	1.456	1.456	1.456	1.456	1.456
318.15K								
0.0000	1.623	1.623	1.623	1.623	1.623	1.623	1.623	1.623
0.0634	1.731	1.732	1.728	1.725	1.735	1.723	1.610	1.835
0.1322	1.834	1.837	1.831	1.827	1.838	1.817	1.596	2.055
0.2070	1.926	1.933	1.927	1.923	1.927	1.904	1.580	2.270
0.2888	1.998	2.012	2.008	2.006	1.997	1.980	1.564	2.460
0.3786	2.045	2.063	2.063	2.063	2.042	2.038	1.546	2.597
0.4775	2.058	2.072	2.077	2.081	2.052	2.070	1.526	2.646
0.5870	2.023	2.025	2.034	2.041	2.015	2.059	1.504	2.568
0.7090	1.925	1.907	1.917	1.925	1.915	1.977	1.480	2.333
0.8457	1.727	1.706	1.714	1.720	1.729	1.780	1.454	1.938
1.0000	1.424	1.424	1.424	1.424	1.424	1.424	1.424	1.424

Table-4(c): Experimental and Calculated Viscosity Values, η (mPa·s), for Binary Mixtures of Diethyl Malonate with 2-propanol at different Temperatures

X_1	η Expt.	ηGN.	η KC.	η HB.	η H.	η Mc.	η KM.	η F.
303.15K								
0.0000	1.801	1.801	1.801	1.801	1.801	1.801	1.801	1.801
0.0531	1.748	1.850	1.805	1.750	1.748	1.747	1.797	2.027
0.1120	1.696	1.899	1.812	1.706	1.696	1.695	1.793	2.275
0.1778	1.645	1.946	1.821	1.671	1.645	1.645	1.788	2.538
0.2518	1.599	1.989	1.832	1.644	1.599	1.599	1.783	2.799
0.3354	1.559	2.022	1.841	1.626	1.559	1.559	1.777	3.030
0.4309	1.530	2.040	1.848	1.618	1.530	1.531	1.770	3.184
0.5408	1.520	2.035	1.847	1.622	1.520	1.519	1.762	3.194
0.6687	1.537	1.993	1.834	1.640	1.537	1.536	1.753	2.980
0.8196	1.598	1.899	1.799	1.674	1.598	1.599	1.743	2.483
1.0000	1.730	1.730	1.730	1.730	1.730	1.730	1.730	1.730
303.15K								
0.0000	1.562	1.562	1.562	1.562	1.562	1.562	1.562	1.562
0.0531	1.480	1.644	1.605	1.550	1.492	1.488	1.564	1.751
0.1120	1.419	1.728	1.651	1.545	1.424	1.420	1.566	1.959
0.1778	1.361	1.813	1.701	1.544	1.360	1.359	1.569	2.179
0.2518	1.310	1.893	1.751	1.550	1.303	1.308	1.572	2.400
0.3354	1.269	1.963	1.796	1.561	1.257	1.268	1.575	2.599
0.4309	1.244	2.012	1.831	1.575	1.229	1.244	1.579	2.740
0.5408	1.240	2.023	1.846	1.592	1.229	1.244	1.584	2.767
0.6687	1.284	1.977	1.826	1.608	1.272	1.282	1.589	2.613
0.8196	1.385	1.845	1.752	1.615	1.383	1.384	1.595	2.223

1.0000	1.602	1.602	1.602	1.602	1.602	1.602	1.602	1.602
313.15K								
0.0000	1.361	1.361	1.361	1.361	1.361	1.361	1.361	1.361
0.0531	1.266	1.457	1.425	1.374	1.285	1.304	1.366	1.513
0.1120	1.193	1.559	1.493	1.392	1.212	1.239	1.371	1.680
0.1778	1.145	1.663	1.565	1.415	1.143	1.171	1.378	1.856
0.2518	1.093	1.764	1.638	1.443	1.083	1.106	1.385	2.032
0.3354	1.053	1.855	1.705	1.473	1.035	1.052	1.392	2.193
0.4309	1.031	1.921	1.757	1.503	1.008	1.019	1.401	2.310
0.5408	1.037	1.943	1.781	1.527	1.014	1.023	1.412	2.342
0.6687	1.086	1.894	1.755	1.538	1.069	1.084	1.424	2.236
0.8196	1.213	1.740	1.656	1.522	1.202	1.230	1.439	1.944
1.0000	1.456	1.456	1.456	1.456	1.456	1.456	1.456	1.456
318.15K								
0.0000	1.191	1.191	1.191	1.191	1.191	1.191	1.191	1.191
0.0531	1.069	1.317	1.290	1.242	1.105	1.098	1.203	1.335
0.1120	1.010	1.455	1.398	1.299	1.023	1.014	1.216	1.495
0.1778	0.946	1.602	1.515	1.364	0.947	0.941	1.230	1.668
0.2518	0.892	1.752	1.635	1.433	0.882	0.885	1.247	1.846
0.3354	0.854	1.893	1.751	1.503	0.834	0.848	1.266	2.014
0.4309	0.838	2.005	1.845	1.568	0.813	0.838	1.288	2.146
0.5408	0.859	2.056	1.896	1.616	0.833	0.865	1.314	2.203
0.6687	0.934	2.004	1.868	1.628	0.915	0.942	1.344	2.131
0.8196	1.093	1.802	1.721	1.576	1.094	1.080	1.380	1.877
1.0000	1.424	1.424	1.424	1.424	1.424	1.424	1.424	1.424

Table-4(d): Observed and Estimated Viscosity Values, η (mPa·s), for Binary Mixtures of Diethyl Malonate with 2-Butanol at different Temperatures

X_1	η Expt.	ηGN.	η KC.	η HB.	η H.	η Mc.	η KM.	η F.
303.15K								
0.0000	2.585	2.585	2.585	2.585	2.585	2.585	2.585	2.585
0.0630	2.753	2.763	2.756	2.752	2.753	2.757	2.679	3.177
0.1314	2.910	2.929	2.918	2.912	2.901	2.911	2.600	3.615
0.2060	3.040	3.069	3.059	3.053	3.022	3.036	2.516	4.035
0.2875	3.123	3.168	3.162	3.157	3.107	3.122	2.426	4.383
0.3771	3.156	3.202	3.203	3.201	3.143	3.156	2.330	4.590
0.4759	3.118	3.148	3.156	3.159	3.113	3.120	2.227	4.574
0.5855	2.989	2.981	2.996	3.003	2.994	2.991	2.116	4.263
0.7077	2.741	2.684	2.701	2.709	2.756	2.736	1.997	3.627
0.8449	2.320	2.258	2.268	2.275	2.354	2.322	1.868	2.727
1.0000	1.730	1.730	1.730	1.730	1.730	1.730	1.730	1.730
303.15K								
0.0000	2.154	2.154	2.154	2.154	2.154	2.154	2.154	2.154
0.0630	2.294	2.301	2.296	2.293	2.299	2.298	2.245	2.614
0.1314	2.430	2.441	2.432	2.428	2.428	2.430	2.193	2.943
0.2060	2.546	2.564	2.555	2.550	2.537	2.543	2.138	3.257
0.2875	2.628	2.656	2.651	2.647	2.618	2.627	2.078	3.520
0.3771	2.670	2.700	2.702	2.700	2.660	2.670	2.014	3.685
0.4759	2.657	2.678	2.686	2.688	2.649	2.660	1.944	3.693
0.5855	2.572	2.567	2.581	2.586	2.568	2.574	1.869	3.489
0.7077	2.394	2.352	2.366	2.374	2.392	2.388	1.787	3.043
0.8449	2.068	2.025	2.034	2.041	2.086	2.071	1.699	2.380
1.0000	1.602	1.602	1.602	1.602	1.602	1.602	1.602	1.602
313.15K								
0.0000	1.724	1.724	1.724	1.724	1.724	1.724	1.724	1.724

0.0630	1.838	1.842	1.838	1.835	1.845	1.841	1.812	2.076
0.1314	1.950	1.957	1.950	1.946	1.956	1.951	1.784	2.320
0.2060	2.053	2.061	2.055	2.051	2.052	2.050	1.754	2.556
0.2875	2.131	2.146	2.142	2.139	2.126	2.130	1.722	2.759
0.3771	2.181	2.198	2.199	2.198	2.173	2.181	1.687	2.896
0.4759	2.190	2.203	2.210	2.212	2.180	2.193	1.649	2.927
0.5855	2.146	2.144	2.155	2.160	2.135	2.148	1.607	2.809
0.7077	2.031	2.004	2.016	2.023	2.018	2.026	1.562	2.513
0.8449	1.799	1.773	1.781	1.788	1.804	1.802	1.511	2.043
1.0000	1.456	1.456	1.456	1.456	1.456	1.456	1.456	1.456
318.15K								
0.0000	1.541	1.541	1.541	1.541	1.541	1.541	1.541	1.541
0.0630	1.639	1.644	1.641	1.638	1.650	1.642	1.625	1.852
0.1314	1.738	1.746	1.740	1.736	1.750	1.741	1.610	2.072
0.2060	1.834	1.840	1.834	1.831	1.838	1.833	1.593	2.287
0.2875	1.918	1.920	1.917	1.914	1.910	1.912	1.575	2.478
0.3771	1.970	1.975	1.976	1.975	1.958	1.969	1.556	2.615
0.4759	1.989	1.994	2.000	2.002	1.975	1.993	1.534	2.664
0.5855	1.966	1.961	1.971	1.976	1.949	1.969	1.511	2.584
0.7077	1.883	1.862	1.872	1.879	1.864	1.878	1.485	2.346
0.8449	1.699	1.683	1.691	1.697	1.700	1.701	1.456	1.946
1.0000	1.424	1.424	1.424	1.424	1.424	1.424	1.424	1.424

Table-5(a): A Binary Mixture of Diethyl Malonate and Branched Alcohols at Various Temperatures and their Interaction Parameters

T/K	G12	σ	Wvis/RT	σ	Δ12	σ	H12	σ
diethyl malonate + 3-methyl 1-butanol								
303.15	1.8278	0.1036	1.8544	0.0965	2.0067	0.0976	4.6341	0.0131
308.15	1.6605	0.0651	1.6777	0.0597	1.8398	0.0606	3.5625	0.031
313.15	1.5845	0.0472	1.5943	0.0423	1.7645	0.0432	3.0808	0.0251
318.15	1.4176	0.0313	1.418	0.0273	1.5985	0.0281	2.668	0.0231
diethyl malonate + 2-methyl 1-propanol								
303.15	1.649	0.0517	1.7432	0.0457	1.9564	0.043	4.3697	0.0213
308.15	1.5431	0.0371	1.6266	0.032	1.8508	0.0298	3.7459	0.0127
313.15	1.473	0.0298	1.5474	0.0253	1.7807	0.0231	3.244	0.0076
318.15	1.2301	0.0125	1.2925	0.0109	1.5393	0.0117	2.5739	0.0048
diethyl malonate + 2-propanol								
303.15	0.5797	0.3651	0.3699	0.2276	0.1166	0.0645	1.2763	0.0003
308.15	0.9871	0.5516	0.7849	0.424	0.4737	0.2427	0.8671	0.0095
313.15	1.2873	0.6384	1.0943	0.5233	0.7682	0.3444	0.6062	0.0162
318.15	1.8101	0.8338	1.6306	0.7235	1.2897	0.531	0.3325	0.0191
diethyl malonate + 2-butanol								
303.15	1.5559	0.0367	1.6556	0.0314	1.864	0.0288	4.0325	0.0149
308.15	1.4378	0.025	1.5269	0.0211	1.7462	0.0189	3.3978	0.0084
313.15	1.3053	0.0148	1.3875	0.0122	1.6142	0.0111	2.7601	0.0076
318.15	1.1839	0.0095	1.2552	0.0062	1.493	0.0059	2.4645	0.0114

Table-5(b): Evaluation of Binary Systems with Diethyl Malonate and Branched-Chain Alcohols at different Temperatures and their Interaction Coefficients

T/K	McAllister four body model			σ	KM	σ	F	σ
	Z1112	Z1122	Z2221					
diethyl malonate + 3-methyl 1-butanol								
303.15	3.8967	3.4295	4.0583	0.0045	0.5489	0.301	0.4029	0.162
308.15	3.1026	2.7213	2.9474	0.0048	0.4512	0.204	0.2378	0.057
313.15	2.6471	2.4343	2.5258	0.0019	0.4051	0.164	0.2728	0.074

318.15	2.3148	2.1661	2.1733	0.0025	0.3417	0.117	0.1914	0.037
diethyl malonate + 2-methyl 1-propanol								
303.15	3.3932	3.5131	3.9249	0.0005	0.4884	0.239	0.4204	0.177
308.15	2.9373	3.0705	3.3319	0.0003	0.4295	0.185	0.3298	0.109
313.15	2.5696	2.6903	2.8705	0.0007	0.3854	0.148	0.2636	0.069
318.15	2.1266	2.2124	2.2236	0.0009	0.2942	0.087	0.2387	0.057
diethyl malonate + 2-propanol								
303.15	1.5441	1.4537	1.6614	0.001	0.1237	0.015	0.6381	0.407
308.15	1.2748	1.1682	1.3085	0.0025	0.1861	0.035	0.627	0.393
313.15	1.2139	0.7789	1.2035	0.021	0.2189	0.048	0.5863	0.344
318.15	1.1307	0.6725	0.8513	0.0121	0.2781	0.077	0.6254	0.391
diethyl malonate + 2-butanol								
303.15	3.1451	3.3302	3.5645	0.001	0.3423	0.117	0.5324	0.283
308.15	2.7003	2.8569	2.9662	0.0013	0.8216	0.675	0.419	0.176
313.15	2.2483	2.3763	2.3723	0.0014	0.2556	0.065	0.332	0.11
318.15	2.0385	2.199	2.0906	0.002	0.232	0.054	0.232	0.054

CONCLUSION

The density and dynamic viscosity of binary liquid systems including diethyl malonate and the branched alcohols 3-methyl-1-butanol, 2-methyl-1-propanol, 2-propanol, and 2-butanol are experimentally measured in this work at four different temperatures: 303.15K, 308.15K, 313.15K, and 318.15 K. In order to understand the nature of intermolecular interactions in the mixtures, viscosity deviations ($\Delta\eta$) and the excess Gibbs free energy of activation for viscous flow (ΔG^{*E}) were assessed using these data. A variety of semi-empirical correlations based on Eyring's absolute rate theory, such as the Grunberg-Nissan, Katti-Chaudhari, Heric-Brewer, Hind *et al.* McAllister, Kendall, Monroe, and Frenkel models were used to examine the experimental viscosity values. The two that agreed with the measured data the best were the Hind *et al.* and McAllister four-body approaches. Strong specific interactions between different molecules, including hydrogen bonds and dipole–dipole attractions, are indicated by the majority of systems' positive $\Delta\eta$, ΔG^{*E}, and interaction parameter (G_{12}) values. The diethyl malonate + 2-propanol mixture was an exception, showing considerably weaker intermolecular interactions.

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CONFLICT OF INTERESTS

The authors affirm that there are no known financial or personal relationships that could have influenced the work reported in this study.

AUTHOR CONTRIBUTIONS

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