Density and Speed of Sound of Diethylene Glycol Monoethyl Ether + Propylene Glycol at T = (288.15–318.15) K

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ABSTRACT: *The density (ρ) and speed of sound (u) of the binary mixture (diethylene glycol monoethyl ether (carbitol) + propylene glycol (PG)) have been reported at T= (288.15, 298.15, 308.15 and 318.15) K. The excess molar volume (V_m^E) and isentropic compressibility deviation ($\Delta\kappa_s$) were calculated using the measured data. The obtained values of (V_m^E) and ($\Delta\kappa_s$) are negative and positive, respectively and become more negative and more positive with increase in carbitol mole fraction. The ρ, u, V_m and κ_s values of the mixture were correlated by the Jouyban-Acree model with high accuracy. Furthermore, the calculated V_m^E and $\Delta\kappa_s$ values were correlated well by the Redlich-Kister equation.*

KEYWORDS: *Density; Speed of sound; Propylene glycol; Diethylene glycol monoethyl ether.*

INTRODUCTION

Glycol ethers with etheric and alcoholic groups as well as hydrocarbon chain can dissolve the wide range of compounds [1, 2]. Due to the attractive properties of these components, such as low vapor pressure, low toxicity, low viscosity, high chemical stability, and low melting point, they are much important in various industrial applications. Glycol ethers are used as scrubbing liquids for absorption of acid gases exhausting from industrial plants, as octane

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number enhancer [1, 2], as the polar additive in anionic polymerization and automotive brake fluid [3, 4], as heat absorbents in pumps and chillers [3, 4] and as a solubilizer in drilling and cutting coolants. They are also used in biomedical processes [5]. Alkanediols are also very important components with applications in industry such as antifreezes, coolants, aircraft deicing fluids, cosmetic, pharmaceutical, food, automotive industries and so on [7-9].

Thermophysical property data and the mixing deviation from ideality of the systems consisting diethylene glycol monoethyl ether (carbitol) is important for designing the engineering process and also studying the molecular interactions [6]. Despite the wide application range of the carbitol, the studies on its physicochemical properties are limited to the binary mixture of carbitol + water presented by Xinxue et al. [2]. Therefore, in this work, the physicochemical properties of binary mixtures of carbitol + propylene glycol (PG) including density (ρ) and speed of sound (u) were measured at T= (288.15, 298.15, 308.15 and 318.15) K and the obtained data were used to calculate the excess molar volume (V_m^E) and isentropic compressibility deviation ($\Delta\kappa_s$). The Jouyban-Acree and Redlich-Kister were used for correlation of (ρ, u, V_m and κ_s) values at T= (288.15 to 318.15) K and (V_m^E and $\Delta\kappa_s$) values at a fixed temperature, respectively.

EXPERIMENTAL SECTION

PG with the mass fraction purity of more than 0.998 was supplied from Chem-lab NV (Belgium) and carbitol with purity of more than 0.980 in mass fraction is purchased from Merck (Germany). A sample description of the used chemicals were given in Table 1.

The solutions were prepared in mole fraction by using an analytical balance (Shimadzu, 321−34553, Shimadzu Co., Japan) with an uncertainty of $\pm 1\times 10^{-7}$ kg. The density ρ and speed of sound u of the mixture or pure components were measured with a vibrating tube densimeter (Anton Para, DSA 5000 densimeter and speed of sound analyzer). The instrument was calibrated with double distilled deionized and degassed water and dry air at atmospheric pressure.

The experimental density ρ and speed of sound u of the pure components accompanied with their comparison with the available data in the literature are given in Table 2.

It is observed from this table, the measured data are almost consistent with those reported in the literature. The small deviations may be related to the different purity of the pure components, uncertainty of the measurement, atmospheric pressure and frequency of sound measurement.

RESULTS AND DISCUSSION

Density and Speed of Sound

The experimental density ρ and speed of sound u of the binary mixture of (carbitol + PG) are given in Table 3 and Figs. 1 and 2.

As observed in these Figs. 1 and 2, the density and speed of sound of the studied mixture decrease with increase in the carbitol mole fraction as well as temperature.

Excess molar volume

The values of excess molar volume, V_m^E of the binary mixture were calculated using the following equations:

$$V_m^E/(m^3mol^{-1}) = V_m - x_1V_1 - x_2V_2 \qquad (1)$$

$$V_m^E/(m^3mol^{-1}) = \left(\frac{x_1M_1 + x_2M_2}{\rho_m}\right) - x_1\left(\frac{M_1}{\rho_1}\right) - x_2\left(\frac{M_2}{\rho_2}\right) \qquad (2)$$

where (V_m, V_1 and V_2) and (ρ_m, ρ_1 and ρ_2) are the molar volumes and densities of the mixture, component 1 and 2, respectively. M_1 and M_2 are the molar masses of component 1 and 2. The excess molar volume of the binary mixture were reported in Table 3 and Fig. 3.

As can be seen in this figure, the excess molar volumes V_m^E of the mixtures are negative and become more negative by increasing the temperature. The minimum value of the V_m^E value (~ -0.08) is observed in the 0.3 mole fraction of carbitol. The negative values of V_m^E indicate the volume contraction in the mixing process. With increase in the carbitol concentration, the self-association in PG are reduced and the formation of the newly interactions between carbitol and PG are increased. The more negative values of V_m^E with rising in temperature indicate that the formation of carbitol-PG interaction needs energy to form.

Isentropic compressibility deviation

The isentropic compressibility κ_s of the pure components and binary mixtures, as well as isentropic compressibility deviation $\Delta\kappa_s$, were calculated using the measured density and speed of sound values according to the following equations [21-24]:

Table 1: A Sample Description of the Used Chemicals.

Chemical name	CAS No.	Source	Mass fraction purity
Carbitol	111-90-0	Merck (Germany)	> 0.980
PG	504-63-2	Chem-lab (Belgium)	> 0.998

Table 2: The Experimental density (ρ) and speed of sound (u) of pure liquids at the specified temperature and p= 0.0868 MPa.[a]

component	T/ K	$10^{-3}\rho$/ kg m^{-3}		u/ m s^{-1}	
		Exp.	Lit.	Exp.	Lit.
carbitol	288.15	0.99217	0.99227 [10]	1410.40	1410.09 [10]
	298.15	0.98311	0.983413 [10] 0.9842 [11] 0.9841[12]	1374.42	1374.62 [10]
	308.15	0.97418	0.974518 [10]	1338.93	1339.51 [10]
	318.15	0.96533	0.965545 [10]	1304.08	1305.13 [10]
PG					
	288.15	1.03935		1537.36	
	298.15	1.03216	1.03258 [13] 1.03252 [14] 1.03262 [15]	1509.10	1510.97 [16]
	308.15	1.02458	1.02508 [14] 1.02513 [13] 1.0231 [17]	1481.16	1483.12 [16]
	318.15	1.01700	1.01752 [13]	1452.53	1454.91 [16]
water					
	288.15	0.99909	0.99910 [18]	1466.56	
	298.15	0.99705	0.99707 [18] 0.997081 [19] 0.997081[20]	1496.91	1497.00 [19] 1496.77 [20]
	308.15	0.99403	0.99406 [19] 0.9950681[20]	1520.00	1521.00 [19] 1507.16 [20]
	318.15	0.99019	0.99024 [18] 0.99024 [19] 0.994194 [20]	1536.63	1537.00 [19] 1520.17 [20]

a) Standard uncertainties (u) for each variables are; u (ρ)= 0.2 kg m^{-3}; u (u)= 0.5 m s^{-1}; u (T)= 0.03 K; u (P)= 0.5 kPa.

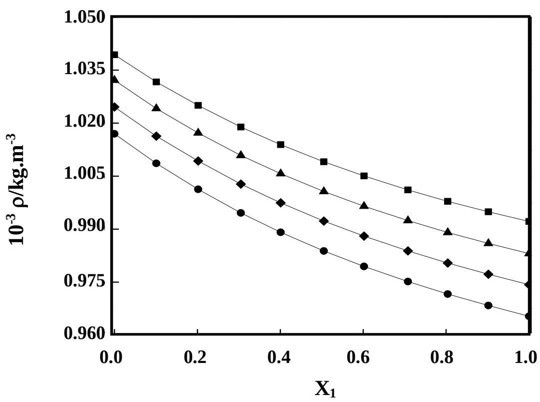

Fig. 1: The density of the binary mixtures of carbitol (1) + PG (2) at temperatures 288.15 (■), 298.15 (▲), 308.15 (♦), and 318.15 K (●).

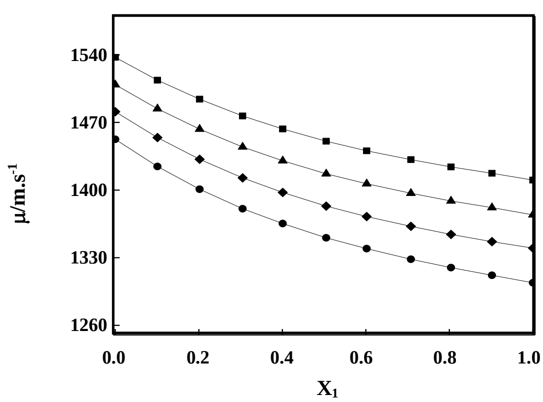

Fig. 2: The speed of sound of the binary mixtures of carbitol (1) + PG (2) at temperatures 288.15 (■), 298.15 (▲), 308.15 (♦), and 318.15 K (●).

Table 3: Experimental density (ρ), molar volume (V_m), excess molar volume (V_m^E), speed of sound (u), isentropic compressibility ($κ_s$) and isentropic compressibility deviation ($Δκ_s$) for the binary mixtures of (carbitol (1) + PG (2)) at the experimental temperatures and carbitol mole fractions and p= 0.0868 MPa[a]

x_1	$10^{-3}ρ$/ kg m^{-3}	$10^6 V_m$/ m^3 mol^{-1}	$10^6 V_m^E$ m^3 mol^{-1}	u/ m s^{-1}	$κ_s$/ (TPa)$^{-1}$	$Δκ_s$/ (TPa)$^{-1}$
carbitol (1) + PG (2)						
T/K = 288.15						
0.0000	1.03935	73.219		1537.36	407.087	
0.1013	1.03165	79.472	-0.033	1513.75	423.018	5.840
0.2020	1.02503	85.693	-0.059	1494.07	437.042	9.836
0.3052	1.01890	92.092	-0.062	1476.74	450.050	12.568
0.4009	1.01392	98.025	-0.063	1463.26	460.633	13.625
0.5054	1.00906	104.517	-0.057	1450.57	470.984	13.565
0.6023	1.00506	110.536	-0.051	1440.63	479.408	12.337
0.7082	1.00111	117.118	-0.040	1431.53	487.435	9.817
0.8043	0.99785	123.094	-0.023	1423.98	494.228	7.044
0.9015	0.99490	129.138	-0.013	1417.37	500.325	3.454
1.0000	0.99217	135.259		1410.40	506.676	
T/K = 298.15						
0.0000	1.03216	73.729		1509.10	425.419	
0.1013	1.02413	80.056	-0.035	1484.22	443.250	6.373
0.2020	1.01723	86.350	-0.063	1463.11	459.225	10.964
0.3052	1.01087	92.823	-0.067	1444.59	474.039	14.111
0.4009	1.00570	98.826	-0.068	1430.40	485.979	15.237
0.5054	1.00066	105.394	-0.064	1416.89	497.785	15.223
0.6023	0.99649	111.485	-0.058	1406.60	507.206	13.685
0.7082	0.99242	118.143	-0.049	1396.73	516.512	11.017
0.8043	0.98905	124.189	-0.034	1388.76	524.238	7.882
0.9015	0.98595	130.311	-0.018	1381.88	531.135	3.782
1.0000	0.98308	136.510		1374.42	538.484	
T/K = 308.15						
0.0000	1.02458	74.275		1481.16	444.889	
0.1013	1.01633	80.670	-0.036	1454.33	465.199	7.461
0.2020	1.00929	87.029	-0.068	1431.88	483.248	12.743
0.3052	1.00276	93.575	-0.071	1412.61	499.758	16.171
0.4009	0.99743	99.646	-0.071	1397.59	513.284	17.569
0.5054	0.99227	106.285	-0.067	1383.40	526.591	17.62

Table 3: Experimental density (ρ), molar volume (V_m), excess molar volume (V_m^E), speed of sound (u), isentropic compressibility (κ_s) and isentropic compressibility deviation ($\Delta\kappa_s$) for the binary mixtures of (carbitol (1) + PG (2)) at the experimental temperatures and carbitol mole fractions and p= 0.0868 MPa[a]. (continued)

x_1	$10^{-3}\rho$/ kg m^{-3}	$10^6 V_m$/ m^3 mol^{-1}	$10^6 V_m^E$ m^3 mol^{-1}	u/ m s^{-1}	κ_s/ (TPa)$^{-1}$	$\Delta\kappa_s$/ (TPa)$^{-1}$
0.6023	0.98803	112.440	-0.063	1372.59	537.214	15.954
0.7082	0.98386	119.172	-0.053	1362.47	547.538	12.851
0.8043	0.98040	125.285	-0.037	1354.09	556.292	9.424
0.9015	0.97723	131.474	-0.022	1346.67	564.262	5.062
1.0000	0.97427	137.744		1339.93	571.683	
			T/K = 318.15			
0.0000	1.01700	74.828		1452.53	466.048	
0.1013	1.00862	81.287	-0.046	1424.42	488.647	8.100
0.2020	1.00128	87.725	-0.071	1400.91	508.889	13.934
0.3052	0.99459	94.343	-0.077	1380.74	527.392	17.673
0.4009	0.98912	100.483	-0.076	1365.34	542.339	18.934
0.5054	0.98381	107.199	-0.071	1350.60	557.231	18.866
0.6023	0.97946	113.425	-0.067	1339.40	569.108	16.876
0.7082	0.97517	120.233	-0.057	1328.43	581.089	13.703
0.8043	0.97164	126.414	-0.041	1319.75	590.899	9.768
0.9015	0.96838	132.675	-0.025	1311.86	600.039	4.990
1.0000	0.96533	139.019		1304.08	609.136	

a) Standard uncertainties (u) for each variables are u (ρ)= 0.2 kg/m^3; u (u)= 0.5 m/s; u (T)= 0.03 K; u (V_m^E 10^6)= 0.1 m^3/mol^1; u (κ_s)= 1 TPa^{-1} and u ($\Delta\kappa_s$)= 0.1 TPa^{-1}.

$$\kappa_s / \mathrm{Pa}^{-1} = (\rho.u^2)^{-1} \quad (3)$$

$$\Delta\kappa_s / \mathrm{Pa}^{-1} = \kappa_s - \left(\sum_{i=1}^{2} x_i k_{si}\right) \quad (4)$$

Where ρ and u are the density and speed of sound of the mixtures, respectively, and κ_{si} is the

isentropic compressibility value for the pure component *i* (see Table 3 and Fig. 4).

As can be seen in Table 3, the κ_s values of the system are positive and become more positive with increase in the temperature. The maximum value of $\Delta\kappa_s$ (19 TPa^{-1}) is located at 0.4 mole fraction of carbitol and at 318.15 K. This phenomenon shows that compressibility of the mixtures is higher than pure components.

Correlation of measured data using Jouyban-Acree model

Measurement of the physicochemical properties is as important as their correlation with respect to solvent composition and temperature, since both are fine tuned in industrial applications. Mathematical representation of density and speed of sound as two crucial physicochemical properties help the engineers and chemists to derive these properties at different mole fraction and temperature since their determinations at all fractions and temperatures are impossible and time consuming. In this regard, one model which can correlate these properties with high accuracy is an essential issue. In our earlier work [17], the Jouyban-Acree model was introduced as a model for correlating the physicochemical properties. This model describes

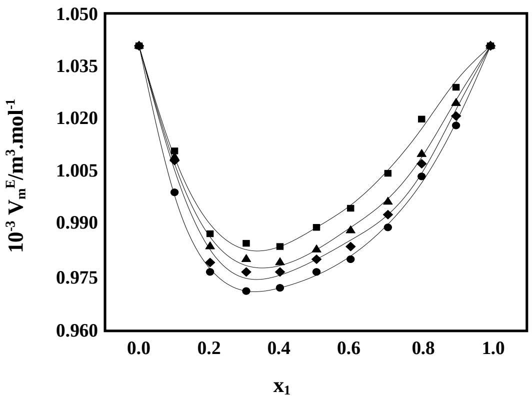

Fig. 3: The excess molar volume of the binary mixtures of carbitol (1) + PG (2) at temperatures 288.15 (■), 298.15 (▲), 308.15 (♦), and 318.15 K (●).

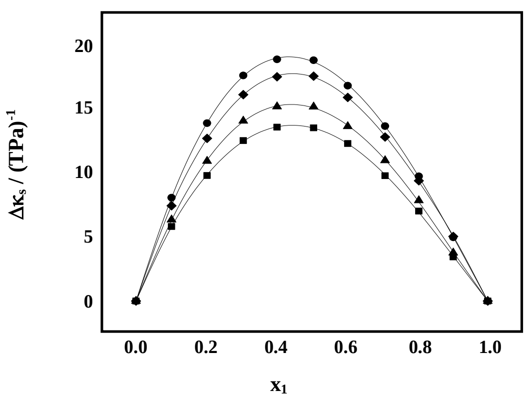

Fig. 4: The isentropic compressibility deviation of the binary mixtures of carbitol (1) + PG (2) at temperatures 288.15 (■), 298.15 (▲), 308.15 (♦), and 318.15 K (●).

the properties of mixtures with respect to the composition and temperature and is expressed as:

$$\ln K_{m,T} = x_1 \ln K_{1,T} + x_2 \ln K_{2,T} + \sum_{i=0}^{2} J_i \left[\frac{x_1 x_2 (x_1 - x_2)^i}{T} \right] \qquad (5)$$

Where $K_{m,T}$, $K_{1,T}$ and $K_{2,T}$ are the values of physicochemical properties such as, density ρ, speed of sound u, molar volume V_m and isentropic compressibility κ_s of the mixture, component 1 (carbitol) and 2 (PG) at temperature T, x_1 and x_2 are the mole fractions of the carbitol and PG in the binary mixture, respectively. The J_i are the model parameters evaluated using no intercept least squares analysis [25, 26].

The accuracy of the model tested on the studied datasets was evaluated by percent average relative deviation (%ARD) [25]:

$$\%ARD = \frac{100}{N} \sum \left| \frac{K_{m,T}^{Calculated}}{K_{m,T}^{Experimental}} - 1 \right| \qquad (6)$$

in which N is the number of data points in each set and K is the physicochemical properties such as density ρ, speed of sound u, molar volume V_m and isentropic compressibility κ_s. The evaluated parameters of Jouyban-Acree at T= (288.15 to 318.15) K along with their %ARDs were listed in Table 4.

As observed in Table 4, the %ARD for ρ, u, κ_s and V_m are 0.12, 0.17, 4.63 and 0.13, respectively. The acceptable %ARDs mean that the Jouyban-Acree model can be used as a simple model with high accuracy to correlate the physicochemical properties at T= (288.15 to 318.15) K.

Application of Redlich-Kister polynomial equation

Redlich-Kister equation is one of the polynomial equation used for correlation of excess properties such as V_m^E and $\Delta\kappa_s$ with respect to the solvent mole fraction [27]:

$$Q = x_1 x_2 \sum_{i=0}^{3} a_i (2x_i - 1)^i \qquad (7)$$

where A represents (V_m^E and $\Delta\kappa_s$) and a_i are the fitting parameters based on a least-squares method. In this work, it was supposed that the a_i parameters are not temperature dependent and correlation was performed at a fixed temperature. Standard deviation, σ (A) was calculated using the relation [27]:

$$\sigma(A) = [\sum Q_{exp} - Q_{cal}]^2 / (p - n)]^{1/2} \qquad (8)$$

where p is the total numbers of experimental points (here is 11) and n is the number of parameters (here is 4). The evaluated parameters a_i values along with their standard deviations σ is given in Table 5.

CONCLUSIONS

The experimental density and speed of sound for the binary system (carbitol + PG) over the temperature range from 288.15 K to 318.15 K has been reported. The excess molar volume (V_m^E) and isentropic compressibility deviation ($\Delta\kappa_s$) of the studied system were calculated using the experimental data. The calculated values of the V_m^E and $\Delta\kappa_s$ are negative and positive, respectively and become

Table 4: Correlation parameters of Jouyban-Acree Equation for carbitol (1) + PG (2) mixtures at T= (288.15 to 318.15) K.

	J_0	J_1	J_2	%ARD
$10^{-3}\rho$/ kg m^{-3}	-9.658	2.297	NS[a]	0.12
u/ m s^{-1}	-2.690	7.070	NS	0.17
$10^6 V_m$/m^3 mol^{-1}	55.088	-10.721	NS	0.13
κ_s/ (TPa)$^{-1}$	-78.671	282.683	NS	4.63

[a]NS is the non-significant correlation coefficient (p value < 0.05)

Table 5: Parameters of the Redlich–Kister Equation for the Excess Molar Volume (V_m^E) and Isentropic Compressibility Deviation ($\Delta\kappa_S$) of carbitol (1) + PG (2) mixtures.

	T/K	a_0	a_1	a_2	a_3	σ
$10^6 V_m^E$ / m^3 mol^{-1}	288.15	-0.234	0.136	-0.056	0.042	0.002
	298.15	-0.261	0.115	-0.086	0.023	0.002
	308.15	-0.276	0.111	-0.114	0.023	0.003
	318.15	-0.290	0.106	-0.17	0.082	0.001
$10^2 \Delta\kappa_s$/ (TPa)$^{-1}$	288.15	54.346	-13.192	-4.146	-3.049	0.063
	298.15	60.884	-15.355	-5.823	-1.378	0.098
	308.15	70.108	-17.213	-1.704	2.736	0.084
	318.15	75.307	-20.951	-3.209	1.207	0.089

more negative and more positive with increase in temperature. The Jouyban-Acree and Redlich-Kister polynomial equations were used for correlation of (ρ, u, η and κ_s) at T= (288.15 to 318.15) K and (V_m^E and $\Delta\kappa_s$) at a fixed temperature, respectively. The used models have good consistency with the experimental data.

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