



Study of Thermophysical Properties of Binary Mixtures of 1,4-Butanediol + Cresols at Different Temperatures

J.V. SRINIVASU^{1,*}, K. NARENDRA², CH. KAVITHA³ and B. SUBBARAO⁴

¹Department of Physics, Shri Vishnu Engineering College for Women, Bhimavaram-534202, India

²Department of Physics, V.R. Siddhartha Engineering College, Vijayawada-520007, India

³Department of Chemistry, V.R. Siddhartha Engineering College, Vijayawada-520007, India

⁴Department of Physics, S.K.B.R. Government Degree College, Macherla-522426, India

*Corresponding author: E-mail: srinivas.jaddu@gmail.com

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Density (ρ) and ultrasonic velocity (u) of liquid binary mixtures of 1,4-butanediol (1,4-BD), *o*-cresol (OC), *m*-cresol (MC) and *p*-cresol (PC) have been measured at four distinct temperatures (303.15 K, 308.15 K, 313.15 K and 318.15 K) as a function of composition. Using data, significant thermodynamic and thermophysical parameters *viz.*, internal pressure-IP (π), free volume-FV (V_f), enthalpy-EH (H), entropy-ET (Ts) and their excess counterparts have been evaluated. The excess parameter functions EFV (V_f^E), EEH (H^E), EFE (G^E) and EET (T_s^E) were observed to be primarily negative, while π^E is positive indicating that the constituent molecules in these mixtures have discernible interactions with one another.

Keywords: Thermophysical properties, Internal pressure, Excess free volume, Molecular interactions, 1,4-Butanediol, Cresol.

INTRODUCTION

The interactions among molecules investigate between the components of binary liquid mixtures many workers [1-4] have obtained significant thermodynamic parameters from density measurements in conjunction with speed of sound. 1,4-Butanediol (1,4-BD) is a transparent liquid with high viscosity that readily solvate in water and also majority of organic polar solvents. This diol is considered as a excellent intermediate chemical, serving as a crucial component in the fabrication of various chemical products due to hydroxyl groups at vicinal locations 1 and 4. THF is the main byproduct of 1,4-butanediol and about 80% of THF is used for the production of polytetramethylene ether glycol (PTMEG) and some type of plastic, elastic fibers and polyurethanes [5]. The production of bio based succinic acid and the synthesis of bio-1,4-BD from sustainable sources have garnered significant investment from the 1,4-butanediol sector. As a result, organizations are keen on utilizing 1,4-BD as a means to reduce the usage of petroleum-derived feedstocks [6].

Cresols, classified as second generation bio-oil/fuel sources, are increasingly being used as alternatives to fossil fuels. Their

utilization helps in reducing global warming by emitting little amounts of ozone-depleting gasses [7,8]. The growing interest in bio-oil produced through pyrolysis has motivated us to investigate the thermophysical properties of the aforementioned materials.

We have conducted the experimental determination of the densities and velocity of sounds in the combined liquids of 1,4-butanediol (1,4-BD), *o*-cresol (OC), *m*-cresol (MC) and *p*-cresol (PC) including their pure forms, as function of composition at temperatures ranging between 303.15 and 318.15 K. In order to get the knowledge to investigate the nature and extent of interactions, the values of IP (π), FV (V_f), EH (H), ET (Ts), EIP (π^E), EFV (V_f^E), EEH (H^E), EFE (G^E) and EET (T_s^E) have been assessed with the help of experimental measurements. The polynomial equation of Redlich-Kister is used to match the results. It has been observed that the qualitative data derived from the variation of measured parameters with mixture composition provides insight into the kind and strength of molecular interactions. Excess functions also play crucial and considerable role in finding the extent and character of interactions between the molecules existing in binary liquid mixtures.

EXPERIMENTAL

The experimental approach employed in this work for the calculation of different parameters is the same as reported earlier [9]. The measured densities and sound velocities of the pure liquids were compared with those documented in the literature (Table-1) in order to verify the chemical purity [10-24].

RESULTS AND DISCUSSION

Table-2 listed values of density (ρ) and ultrasonic velocity (u) for 1,4-butanediol (1,4-BD), *o*-cresol (OC), *m*-cresol (MC), *p*-cresol (PC) binary mixtures as a function of 1,4-butanediol mole fraction (x_1), at observed temperatures. Table-3 shows the

values of excess internal pressure (EIP) (π_i^E), excess free volume (V_f^E), excess enthalpy (H^E), excess entropy (T_s^E) and excess free energy (G^E) for the binary mixtures at observed temperatures, whereas Table-4 displays the coefficient values (A_i) and corresponding standard deviations (s) for the excess functions.

Table-2 clearly indicates that the ultrasonic velocities show a decreasing trend with raise in the temperature, a trend that is also replicated with the density values. The highest values are demonstrated by 1,4-BD and the *o*-cresol mixture as compared to its *m*- and *p*-counterparts. These values would find reflection in the k_s values of pure components and their mixtures.

Excess internal pressure (EIP): The importance of the internal pressure (IP) was recognized by Hildebrand & Scott

TABLE-1
COMPARISON OF DENSITY (ρ) AND SPEED OF SOUND (u) OBTAINED IN THIS
WORK WITH LITERATURE AVAILABLE AT DIFFERENT TEMPERATURES

Compounds	Temp. (K)	ρ (kg m ⁻³)		u (m s ⁻¹)	
		Measured	Literature	Measured	Literature
1,4-Butanediol	303.15	1009.4	1009.67 [10]	1590.4	1590.19 [11]
	308.15	1006.8	1006.70 [10]	1580.5	1578.49 [11]
	313.15	1003.8	1003.73 [10]	1569.2	1566.90 [11]
	318.15	1000.4	1000.76 [10]	1555.2	1555.41 [11]
<i>o</i> -Cresol	303.15	1036.6	1036.90 [12]	1485.3	1487.00 [12]
			1036.20 [25]		1485.26 [25]
			1037.05 [13]		1488.19 [13]
	308.15	1030.9	1031.60 [12]	1466.9	1470.00 [12]
			1031.00 [25]		1466.84 [25]
			1032.64 [13]		1470.21 [13]
			1032.73 [14]		
			1032.73 [18]		
	313.15	1026.3	1026.00 [25]	1452.2	1452.11 [25]
			1028.25 [13]		1452.65 [13]
			1028.10 [15]		1453.35 [15]
			1026.00 [15]		
1029.00 [19]					
318.15	1021.4	1028.20 [20]	1437.2	1437.06 [25]	
		1028.00 [21]			
		1021.10 [25]			1435.18 [13]
		1023.82 [13]			
<i>m</i> -Cresol	303.15	1025.7	1022.90 [16]	1465.5	1465.00 [12]
			1026.10 [12]		1464.21 [25]
			1025.80 [25]		1466.16 [13]
			1025.96 [13]		
	308.15	1021.9	1026.00 [22]	1450.0	1450.00 [12]
			1022.10 [12]		1449.27 [25]
			1021.50 [25]		1450.55 [13]
			1021.99 [13]		
			1022.10 [22]		
	313.15	1017.4	1021.64 [25]	1435.5	1436.32 [25]
			1017.00 [25]		1434.97 [13]
			1017.99 [13]		1434.20 [15]
			1018.00 [15]		
			1016.00 [17]		
			1018.40 [19]		
	318.15	1013.6	1018.10 [21]	1424.9	1424.21 [25]
1017.63 [24]					
1013.50 [25]			1419.45 [13]		
1013.99 [13]					
1014.10 [22]					
1006.50 [23]					
		1013.60 [24]			

<i>p</i> -Cresol	303.15	1026.6	1026.30 [12]	1469.5	1471.00 [12]
			1026.50 [25]		1468.43 [25]
			1026.39 [13]		1471.38 [13]
	308.15	1022.4	1022.40 [12]	1455.6	1455.00 [12]
			1022.00 [25]		1455.79 [25]
			1022.50 [13]		1455.63 [13]
	313.15	1019.0	1022.50 [21]	1440.5	1443.68 [25]
			1018.10 [25]		1439.97 [13]
			1018.59 [13]		1439.56 [15]
			1018.80 [14]		
			1018.50 [15]		
	318.15	1014.3	1016.80 [17]	1433.1	1432.73 [25]
			1018.60 [19]		1424.29 [13]
			1013.90 [25]		
1014.66 [13]					

$u(T) = \pm 0.02$ K, $u(\rho) = \pm 0.6$ kg m⁻³, $u(u) = \pm 0.67$ m s⁻¹ are the standard uncertainties.

TABLE-2
VALUES OF u AND ρ FOR THE BMS OF 1,4-BD + OC, 1,4-BD + MC AND 1,4-BD + PC
AS FUNCTION OF MOLE FRACTION (x_1) OF 1,4-BD AT MEASURED TEMPERATURES

x_1	u (m s ⁻¹)				ρ (Kg m ⁻³)			
	303.15 K	308.15 K	313.15 K	318.15 K	303.15 K	308.15 K	313.15 K	318.15 K
1,4-BD + OC								
0.0000	1485.3	1466.9	1452.2	1437.2	1036.6	1030.9	1026.3	1021.4
0.1001	1502.1	1484.6	1470.1	1455.4	1035.7	1030.0	1025.3	1020.3
0.2001	1515.8	1499.2	1485.3	1470.5	1034.2	1028.7	1023.9	1019.1
0.3002	1527.5	1511.7	1498.2	1483.5	1032.3	1027.0	1022.4	1017.7
0.4002	1538.1	1523.2	1510.0	1495.4	1030.0	1025.1	1020.7	1016.0
0.5002	1548.7	1534.6	1521.7	1507.3	1027.5	1022.8	1018.6	1014.2
0.6002	1559.9	1546.7	1534.2	1519.8	1024.6	1020.4	1016.3	1012.0
0.7002	1571.1	1558.7	1546.5	1532.3	1021.6	1017.6	1013.7	1009.5
0.8001	1581.5	1570.0	1558.1	1544.0	1018.2	1014.4	1010.8	1006.7
0.9001	1589.2	1578.5	1566.9	1552.9	1014.3	1010.9	1007.4	1003.7
1.0000	1590.4	1580.5	1569.2	1555.2	1009.4	1006.8	1003.8	1000.4
1,4-BD + MC								
0.0000	1465.5	1450.0	1435.5	1425.0	1025.7	1021.9	1017.4	1013.6
0.0992	1485.4	1469.0	1453.6	1442.0	1025.6	1021.6	1017.0	1013.0
0.1986	1504.4	1487.6	1471.7	1459.3	1024.9	1020.9	1016.3	1012.2
0.2982	1521.2	1504.9	1487.7	1475.0	1024.1	1020.1	1015.4	1011.3
0.3979	1536.0	1519.9	1503.0	1489.9	1022.9	1019.0	1014.4	1010.2
0.4979	1549.2	1533.1	1517.1	1503.7	1021.4	1017.6	1013.1	1009.0
0.5979	1562.1	1545.8	1529.3	1515.6	1019.7	1016.0	1011.7	1007.6
0.6982	1573.6	1557.4	1541.1	1527.0	1017.8	1014.2	1010.1	1006.1
0.7986	1582.4	1568.1	1552.7	1538.1	1015.5	1012.1	1008.3	1004.4
0.8992	1588.1	1575.6	1562.7	1547.9	1012.7	1009.6	1006.2	1002.6
1.0000	1590.4	1580.5	1569.2	1555.2	1009.4	1006.8	1003.8	1000.4
1,4-BD + PC								
0.0000	1469.5	1455.6	1440.5	1433.1	1026.6	1022.4	1019.0	1014.3
0.0961	1478.5	1464.9	1450.6	1443.2	1026.3	1022.0	1018.3	1013.5
0.1931	1489.5	1475.8	1462.0	1454.7	1025.7	1021.3	1017.5	1012.7
0.2909	1501.5	1488.5	1475.6	1467.8	1024.7	1020.4	1016.6	1011.7
0.3895	1513.5	1502.1	1490.7	1482.3	1023.5	1019.3	1015.4	1010.6
0.4890	1526.0	1516.1	1505.2	1497.0	1022.0	1017.9	1014.0	1009.3
0.5894	1538.0	1528.4	1517.6	1508.6	1020.2	1016.3	1012.5	1007.9
0.6907	1550.1	1539.4	1528.3	1518.2	1018.2	1014.4	1010.7	1006.3
0.7929	1562.0	1551.0	1539.9	1528.9	1015.8	1012.2	1008.6	1004.5
0.8960	1575.2	1564.2	1553.1	1541.0	1012.8	1009.7	1006.4	1002.6
1.0000	1590.4	1580.5	1569.2	1555.2	1009.4	1006.8	1003.8	1000.4

TABLE-3
CALCULATED VALUES OF EXCESS PARAMETERS EIP (π_i^E), EFV (V_f^E), EEH (H^E), EET (T_s^E) AND EFE (G^E) FOR THE
BMs OF 1,4-BD + OC/MC/PC AS FUNCTION OF MOLE FRACTION (x_1) OF 1,4-BD AT DIFFERENT TEMPERATURES

x_1	$\pi_i^E \times 10^8$ (Nm ⁻²)	$V_f^E \times 10^{-6}$ (m ³ mol ⁻¹)	H^E (kJ mol ⁻¹)	$T_s^E \times 10^{-2}$ (J mol ⁻¹)	G^E (kJ mol ⁻¹)	x_1	$\pi_i^E \times 10^8$ (Nm ⁻²)	$V_f^E \times 10^{-6}$ (m ³ mol ⁻¹)	H^E (kJ mol ⁻¹)	$T_s^E \times 10^{-2}$ (J mol ⁻¹)	G^E (kJ mol ⁻¹)
1,4-BD + OC											
T = 303.15 K						T = 308.15 K					
0.0000	0.000	0.000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.000	0.000
0.1001	0.084	-0.213	-1.035	-0.085	-0.951	0.1001	0.074	-0.144	-0.863	-0.063	-0.800
0.2001	0.139	-0.391	-1.742	-0.151	-1.591	0.2001	0.122	-0.258	-1.435	-0.111	-1.324
0.3002	0.171	-0.534	-2.177	-0.201	-1.976	0.3002	0.150	-0.345	-1.774	-0.144	-1.630
0.4002	0.186	-0.642	-2.395	-0.235	-2.160	0.4002	0.163	-0.406	-1.937	-0.166	-1.771
0.5002	0.189	-0.714	-2.449	-0.254	-2.195	0.5002	0.167	-0.444	-1.979	-0.178	-1.801
0.6002	0.184	-0.745	-2.377	-0.258	-2.119	0.6002	0.165	-0.458	-1.939	-0.180	-1.758
0.7002	0.167	-0.721	-2.136	-0.243	-1.894	0.7002	0.153	-0.440	-1.771	-0.169	-1.601
0.8001	0.136	-0.621	-1.710	-0.203	-1.507	0.8001	0.129	-0.377	-1.453	-0.142	-1.311
0.9001	0.085	-0.406	-1.029	-0.128	-0.901	0.9001	0.082	-0.245	-0.903	-0.090	-0.812
1.0000	0.000	0.000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	0.000	0.000
T = 313.15 K						T = 318.15 K					
0.0000	0.000	0.000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.000	0.000
0.1001	0.064	-0.097	-0.702	-0.046	-0.656	0.1001	0.058	-0.070	-0.600	-0.035	-0.564
0.2001	0.107	-0.171	-1.167	-0.080	-1.087	0.2001	0.095	-0.118	-0.960	-0.059	-0.901
0.3002	0.131	-0.223	-1.421	-0.103	-1.318	0.3002	0.115	-0.148	-1.146	-0.073	-1.073
0.4002	0.143	-0.256	-1.533	-0.116	-1.417	0.4002	0.125	-0.165	-1.218	-0.081	-1.137
0.5002	0.146	-0.274	-1.560	-0.123	-1.438	0.5002	0.128	-0.174	-1.234	-0.085	-1.150
0.6002	0.147	-0.282	-1.545	-0.124	-1.421	0.6002	0.130	-0.178	-1.237	-0.086	-1.150
0.7002	0.139	-0.270	-1.438	-0.117	-1.320	0.7002	0.125	-0.172	-1.177	-0.083	-1.094
0.8001	0.120	-0.233	-1.215	-0.100	-1.115	0.8001	0.111	-0.151	-1.028	-0.072	-0.956
0.9001	0.079	-0.153	-0.785	-0.065	-0.720	0.9001	0.075	-0.102	-0.690	-0.048	-0.642
1.0000	0.000	0.000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	0.000	0.000
1,4-BD + MC											
T = 303.15 K						T = 308.15 K					
0.0000	0.000	0.000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.000	0.000
0.0992	0.062	-0.145	-0.661	-0.055	-0.606	0.0992	0.050	-0.087	-0.507	-0.037	-0.470
0.1986	0.113	-0.269	-1.192	-0.101	-1.091	0.1986	0.093	-0.163	-0.935	-0.068	-0.867
0.2982	0.147	-0.365	-1.534	-0.135	-1.399	0.2982	0.125	-0.223	-1.241	-0.092	-1.148
0.3979	0.165	-0.431	-1.704	-0.157	-1.547	0.3979	0.141	-0.258	-1.379	-0.106	-1.273
0.4979	0.168	-0.466	-1.724	-0.166	-1.558	0.4979	0.144	-0.271	-1.381	-0.110	-1.272
0.5979	0.165	-0.477	-1.670	-0.167	-1.503	0.5979	0.139	-0.268	-1.313	-0.108	-1.205
0.6982	0.149	-0.451	-1.490	-0.154	-1.336	0.6982	0.124	-0.247	-1.158	-0.098	-1.061
0.7986	0.117	-0.372	-1.147	-0.124	-1.022	0.7986	0.101	-0.205	-0.926	-0.080	-0.846
0.8992	0.067	-0.227	-0.647	-0.074	-0.573	0.8992	0.058	-0.123	-0.528	-0.047	-0.480
1.0000	0.000	0.000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	0.000	0.000
T = 313.15 K						T = 318.15 K					
0.0000	0.000	0.000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.000	0.000
0.0992	0.040	-0.054	-0.384	-0.024	-0.360	0.0992	0.033	-0.039	-0.312	-0.017	-0.295
0.1986	0.076	-0.103	-0.734	-0.046	-0.688	0.1986	0.064	-0.076	-0.612	-0.033	-0.579
0.2982	0.098	-0.133	-0.930	-0.059	-0.871	0.2982	0.085	-0.099	-0.800	-0.044	-0.756
0.3979	0.114	-0.154	-1.061	-0.068	-0.992	0.3979	0.099	-0.114	-0.920	-0.050	-0.870
0.4979	0.120	-0.162	-1.098	-0.072	-1.026	0.4979	0.105	-0.119	-0.962	-0.053	-0.909
0.5979	0.112	-0.153	-1.007	-0.068	-0.940	0.5979	0.099	-0.109	-0.882	-0.049	-0.833
0.6982	0.099	-0.135	-0.872	-0.060	-0.813	0.6982	0.087	-0.095	-0.760	-0.043	-0.718
0.7986	0.081	-0.112	-0.707	-0.049	-0.657	0.7986	0.070	-0.076	-0.605	-0.035	-0.570
0.8992	0.051	-0.071	-0.437	-0.031	-0.406	0.8992	0.044	-0.047	-0.370	-0.022	-0.349
1.0000	0.000	0.000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	0.000	0.000
1,4-BD + PC											
T = 303.15 K						T = 308.15 K					
0.0000	0.000	0.000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.000	0.000
0.0961	0.036	-0.159	-0.563	-0.059	-0.504	0.0961	0.025	-0.091	-0.368	-0.037	-0.331
0.1931	0.072	-0.309	-1.092	-0.113	-0.979	0.1931	0.050	-0.177	-0.723	-0.071	-0.652
0.2909	0.102	-0.441	-1.521	-0.160	-1.361	0.2909	0.077	-0.256	-1.067	-0.103	-0.965
0.3895	0.120	-0.545	-1.796	-0.194	-1.602	0.3895	0.099	-0.322	-1.334	-0.128	-1.206
0.4890	0.129	-0.618	-1.925	-0.215	-1.710	0.4890	0.113	-0.367	-1.494	-0.144	-1.350
0.5894	0.123	-0.644	-1.856	-0.218	-1.638	0.5894	0.108	-0.376	-1.437	-0.145	-1.292
0.6907	0.104	-0.614	-1.613	-0.202	-1.411	0.6907	0.086	-0.343	-1.184	-0.128	-1.056
0.7929	0.074	-0.509	-1.191	-0.162	-1.030	0.7929	0.058	-0.271	-0.835	-0.098	-0.737
0.8960	0.038	-0.314	-0.646	-0.096	-0.550	0.8960	0.027	-0.158	-0.424	-0.055	-0.369
1.0000	0.000	0.000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	0.000	0.000

T = 313.15 K						T = 318.15 K					
0.0000	0.000	0.000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.000	0.000
0.0961	0.018	-0.048	-0.235	-0.021	-0.213	0.0961	0.013	-0.024	-0.144	-0.012	-0.133
0.1931	0.037	-0.095	-0.470	-0.042	-0.428	0.1931	0.029	-0.049	-0.310	-0.024	-0.287
0.2909	0.061	-0.142	-0.744	-0.063	-0.681	0.2909	0.049	-0.077	-0.515	-0.037	-0.478
0.3895	0.086	-0.187	-1.013	-0.083	-0.930	0.3895	0.071	-0.106	-0.739	-0.051	-0.687
0.4890	0.100	-0.214	-1.156	-0.094	-1.062	0.4890	0.089	-0.129	-0.909	-0.062	-0.846
0.5894	0.096	-0.214	-1.097	-0.093	-1.004	0.5894	0.084	-0.125	-0.845	-0.060	-0.785
0.6907	0.073	-0.184	-0.858	-0.078	-0.780	0.6907	0.063	-0.100	-0.626	-0.048	-0.579
0.7929	0.047	-0.138	-0.573	-0.057	-0.516	0.7929	0.040	-0.070	-0.399	-0.033	-0.367
0.8960	0.021	-0.075	-0.273	-0.030	-0.243	0.8960	0.018	-0.036	-0.182	-0.016	-0.165
1.0000	0.000	0.000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	0.000	0.000

TABLE-4
COEFFICIENTS (A_i) FOR EXCESS PARAMETERS EIP (π_i^E), EFV (V_i^E), EEH (H^E), EET (T_s^E) AND EFE (G^E) AND THEIR STANDARD DEVIATIONS (σ) FOR BMS OF 1,4-BD + OC, 1,4-BD + MC AND 1,4-BD + PC AT VARIOUS TEMPERATURES

Parameter	Temp. (K)	A_1	A_2	A_3	A_4	A_5	σ
1,4-BD + OC							
π_i^E (10^8 Nm^{-2})	303.15	0.7563	-0.0331	0.3123	0.0578	-0.0482	0.0004
	308.15	0.6695	0.0063	0.3374	0.0800	-0.0465	0.0004
	313.15	0.5869	0.0285	0.3606	0.1129	-0.0570	0.0005
	318.15	0.5147	0.0449	0.3639	0.1085	-0.0193	0.0004
V_i^E ($10^{-6} \text{ m}^3 \text{ mol}^{-1}$)	303.15	-2.8561	-1.0444	-0.7947	-0.4505	-0.1811	0.0007
	308.15	-1.7765	-0.5230	-0.5543	-0.2794	-0.0711	0.0006
	313.15	-1.1002	-0.2498	-0.4477	-0.2135	-0.0135	0.0006
	318.15	-0.6969	-0.1189	-0.3923	-0.1558	-0.0221	0.0005
H^E (kJ mol^{-1})	303.15	-9.8143	0.2690	-2.9305	-0.3346	0.5605	0.0037
	308.15	-7.9336	0.0813	-3.2363	-0.5432	0.5037	0.0039
	313.15	-6.2583	0.0225	-3.5433	-0.8755	0.6567	0.0051
	318.15	-4.9580	-0.0686	-3.6158	-0.8473	0.2850	0.0042
T_s^E ($10^{-2} \text{ J mol}^{-1}$)	303.15	-1.0166	-0.2349	-0.2491	-0.1000	-0.0132	0.0002
	308.15	-0.7123	-0.1367	-0.2189	-0.0813	0.0027	0.0003
	313.15	-0.4916	-0.0766	-0.2062	-0.0812	0.0160	0.0003
	318.15	-0.3405	-0.0451	-0.1954	-0.0687	0.0010	0.0002
G^E (kJ mol^{-1})	303.15	-8.7978	0.5047	-2.6808	-0.2379	0.5730	0.0035
	308.15	-7.2212	0.2181	-3.0174	-0.4619	0.5010	0.0036
	313.15	-5.7666	0.0987	-3.3395	-0.7931	0.6456	0.0048
	318.15	-4.6173	-0.0233	-3.4224	-0.7800	0.2854	0.0040
1,4-BD + MC							
π_i^E (10^8 Nm^{-2})	303.15	0.6765	0.0010	0.2078	0.0361	-0.2314	0.0006
	308.15	0.5756	-0.0279	0.1428	0.1419	-0.1586	0.0009
	313.15	0.4733	-0.0297	-0.0021	0.1599	0.0987	0.0013
	318.15	0.4154	-0.0158	-0.0361	0.1347	0.0882	0.0010
V_i^E ($10^{-6} \text{ m}^3 \text{ mol}^{-1}$)	303.15	-1.8724	-0.4720	-0.4843	-0.1333	0.3001	0.0009
	308.15	-1.0846	-0.1099	-0.2504	-0.2164	0.1919	0.0013
	313.15	-0.6407	0.0301	-0.0138	-0.2138	-0.1346	0.0017
	318.15	-0.4686	0.0589	0.0326	-0.1657	-0.0959	0.0013
H^E (kJ mol^{-1})	303.15	-6.9322	0.3945	-1.9505	-0.3322	2.2889	0.0063
	308.15	-5.5406	0.6988	-1.3667	-1.3746	1.6346	0.0084
	313.15	-4.3257	0.6572	-0.0218	-1.4817	-0.6939	0.0134
	318.15	-3.7906	0.5049	0.3603	-1.3239	-0.6745	0.0100
T_s^E ($10^{-2} \text{ J mol}^{-1}$)	303.15	-0.6662	-0.1024	-0.1569	-0.0332	0.1308	0.0004
	308.15	-0.4405	-0.0190	-0.0954	-0.0883	0.0882	0.0005
	313.15	-0.2846	0.0146	-0.0018	-0.0928	-0.0598	0.0008
	318.15	-0.2098	0.0197	0.0182	-0.0770	-0.0466	0.0006
G^E (kJ mol^{-1})	303.15	-6.2661	0.4967	-1.7925	-0.2992	2.1566	0.0059
	308.15	-5.1000	0.7172	-1.2700	-1.2860	1.5427	0.0078
	313.15	-4.0413	0.6422	-0.0175	-1.3868	-0.6383	0.0126
	318.15	-3.5808	0.4845	0.3423	-1.2448	-0.6274	0.0094
1,4-BD + PC							
π_i^E (10^8 Nm^{-2})	303.15	0.5143	-0.0091	-0.1531	-0.0048	-0.0195	0.0004
	308.15	0.4507	0.0477	-0.4219	-0.0750	0.2638	0.0008
	313.15	0.4008	0.0576	-0.5537	-0.0660	0.4205	0.0008
	318.15	0.3516	0.0773	-0.5871	-0.0853	0.4964	0.0017

V_f^E (10^{-6} m ³ mol ⁻¹)	303.15	-2.4904	-0.9152	-0.1897	-0.0715	0.0008	0.0005
	308.15	-1.4779	-0.4695	0.3253	0.1122	-0.2595	0.0010
	313.15	-0.8607	-0.2231	0.5499	0.1052	-0.4328	0.0008
	318.15	-0.5104	-0.1286	0.5946	0.1035	-0.5036	0.0017
H^E (kJ mol ⁻¹)	303.15	-7.6916	-0.1535	1.4877	-0.1127	0.0908	0.0045
	308.15	-5.9600	-0.4988	4.0579	0.5264	-2.5658	0.0081
	313.15	-4.6178	-0.4677	5.4439	0.4763	-4.1809	0.0076
	318.15	-3.5700	-0.5558	5.7829	0.6354	-4.8872	0.0160
T_s^E (10^{-2} J mol ⁻¹)	303.15	-0.8652	-0.2150	0.0147	-0.0032	0.0087	0.0002
	308.15	-0.5793	-0.1356	0.1936	0.0484	-0.1325	0.0005
	313.15	-0.3786	-0.0784	0.2815	0.0442	-0.2189	0.0004
	318.15	-0.2469	-0.0558	0.3037	0.0485	-0.2563	0.0008
G^E (kJ mol ⁻¹)	303.15	-6.8269	0.0610	1.4759	-0.1091	0.0792	0.0042
	308.15	-5.3807	-0.3630	3.8651	0.4767	-2.4358	0.0077
	313.15	-4.2392	-0.3904	5.1649	0.4346	-3.9674	0.0072
	318.15	-3.3233	-0.4998	5.4831	0.5865	-4.6359	0.0151

[26] in liquid-solution thermodynamics. Several researchers [27-29] have made numerous attempts to demonstrate the significance of this trait in comparison to other properties. From Fig. 1, when carefully observed, it reveals that all mixtures excess IP increase as the mole fraction of 1,4-BD increase, reaching its highest levels near the equimolar area before beginning to fall. Also the excess IP levels decrease as temperature increase.

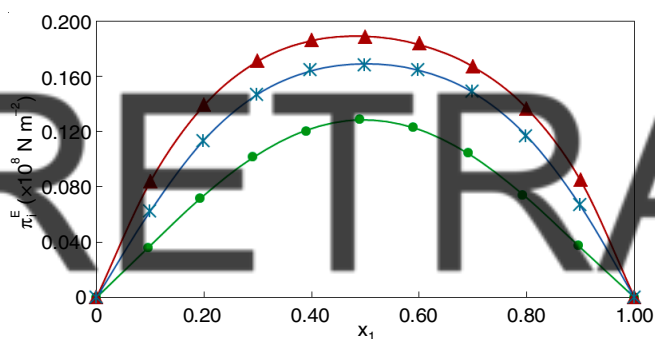


Fig. 1. Change of excess IP, π_i^E with 1,4-BD mole fraction for the BMs 1,4-BD + OC (\blacktriangle), 1,4-BD + MC (\times) and 1,4-BD + PC (\bullet) at temperature 303.15 K. Using the coefficients listed in Table-4, the solid lines have been drawn

According to Fig. 1, the combinations of three binary mixtures, the π_i^E values are observed to be positive values for all the composition ranges and tested temperatures. It is generally recognized that the magnitude and sign of excess functions can provide a good indication of the degree of the unlike interactions in a binary mixtures. The positive π_i^E values happen when the molecules are densely packed, resulting in a decrease in the volume of the mixture and an increase in its internal pressure. The following order shows the magnitude of values: 1,4-BD + OC > 1,4-BD + MC > 1,4-BD + PC, which represents the interactions in order in the observed.

Excess free volume (EFV): For all the three binary mixtures, the change in EFV (V_f^E) as a function of mole fraction (x_1) of 1,4-BD at 303.15 K is shown in Fig. 2. As shown in the figure, the observed EFV (V_f^E) values for the three mixtures are negative. The observed V_f^E values can also be shown to be in the following order: 1,4-BD + OC > 1,4-BD + PC > 1,4-BD + MC. With increase in temperature, the EFV (V_f^E) values are similar found to show a decreasing tendency (Table-3). The declined values of V_f^E points to specific interactions in between

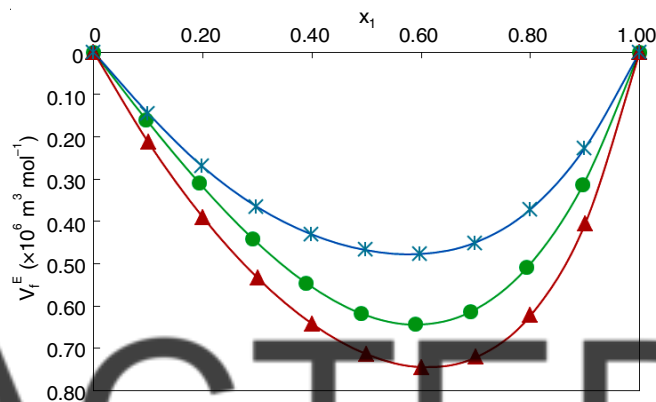


Fig. 2. Change of EFV (V_f^E) with mole fraction of 1,4-BD for the BMs 1,4-BD + OC (\blacktriangle), 1,4-BD + *m*-cresol (\times) and 1,4-BD + *p*-cresol (\bullet) at temperature 303.15 K. Using the coefficients listed in Table-4, the solid lines have been drawn

the mixture's components. The negative excess free volume values of mixtures indicate that volume decrease is caused by the combined effects of the components, and *vice-versa* [30]. The reason behind this could be the negative values of excess free volume, which cause the ion to dipole interaction and packing effect of solvent molecules with the 1,4-BD in these mixtures [31,32].

Excess enthalpy of mixing (EEH): The measured EEH (H^E) values are negative (Fig. 3) for all the three binary mixtures. It can also be showed that with the increase in temperature, the H^E values show a decreasing trend from Table-3. The H^E values of mixing are generally determined by the relative enthalpies of actions, such as endothermic and exothermic, which arise from the mixing of the components. It is evident that the mixing of the 1,4-BD with with *o*- or *m*- or *p*-cresol systems results in an exothermic process, as indicated by the negative values of EEH (H^E) [19]. The negative H^E values are dominant over ion to dipole interactions among unlike molecules in solvent molecules and 1,4-BD.

Excess entropy of mixing: The T_s^E values of three mixtures are found to be negative as observed from Fig. 4. The results obtained from the π_i^E and V_f^E values are confirmed by the negative T_s^E values observed in the binary mixtures. This indicates that the volume is reduced due to the close packing of molecules, which is facilitated by the formation of hydrogen bonds between 1,4-BD and cresol molecules. As a result, the

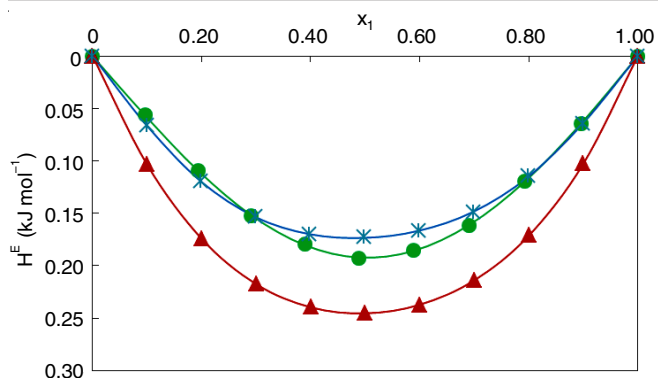


Fig. 3. Change of EEH (H^E) with mole fraction of 1,4-BD for the BMs 1,4-BD + OC (\blacktriangle), 1,4-BD + MC (\times) and 1,4-BD + PC (\bullet) at temperature 303.15 K. Using the coefficients listed in Table-4, the solid lines have been drawn

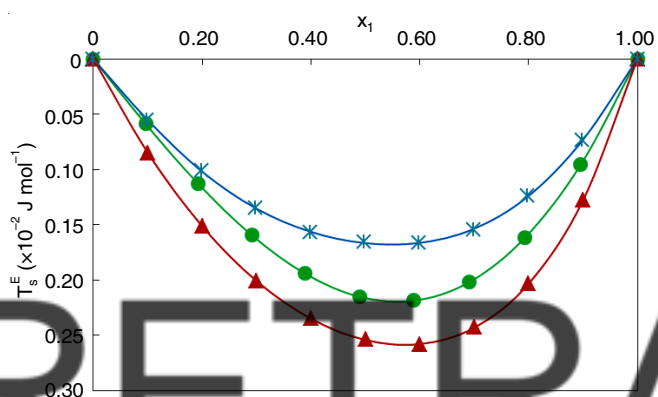


Fig. 4. Change of EET (T_s^E) with mole fraction of 1,4-BD for the BMs 1,4-BD + OC (\blacktriangle), 1,4-BD + MC (\times) and 1,4-BD + PC (\bullet) at temperature 303.15 K. Using the coefficients listed in Table-4, the solid lines have been drawn

entropy values obtained for the mixtures exhibit a negative value of T_s^E .

Excess free energy of mixing: For three three mixtures, the excess free energy (G^E) values were found to be negative (Fig. 5). It can be observed from Table-3 that the G^E values decrease as the temperature increase. Considerable geometrical effect of packing may be responsible for the negative values of G^E .

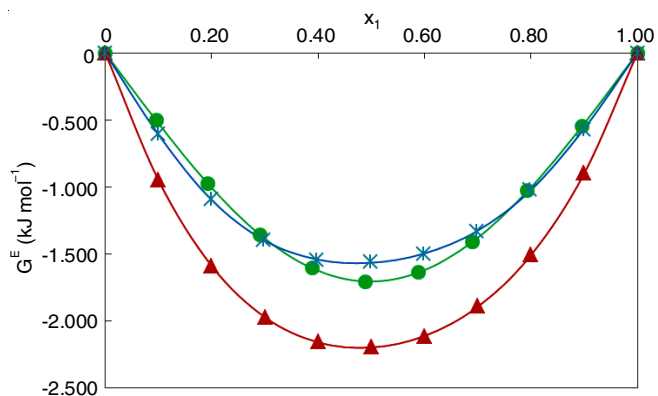


Fig. 5. Change of EFE (G^E) with the mole fraction of 1,4-BD for the BMs 1,4-BD + OC (\blacktriangle), 1,4-BD + MC (\times) and 1,4-BD + PC (\bullet) at temperature 303.15 K. Using the coefficients listed in Table-4, the solid lines have been drawn

Conclusion

In this study, densities (ρ) and sound velocities (u) of pure forms (*o*-, *m*- and *p*-cresol) as well as those of pure forms at $T = (303.15 \text{ to } 318.15) \text{ K}$ and 0.1 MPa atmospheric pressure have been reported for binary mixtures of 1,4-butanediol (1,4-BD). The observed negative values of EVF (V_f^E), EEH (H^E), EFE (G^E) and EET (T_s^E) become less negative as temperature increases. Strong interactions between ion-ion/dipole-dipole and simply cresols occupy the voids of 1,4-butanediol molecule may be responsible for the general negative behaviour of V_f^E , H^E , G^E and T_s^E .

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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