

Refractive Index, Excess Molar Volume and Viscometric Study of Binary Liquid Mixture of Morpholine with Cumene at 298.15 K, 303.15 K, and 308.15 K

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Abstract—Experimental data of refractive index, excess molar volume and viscosity of binary mixture of morpholine with cumene over the whole composition range at 298.15 K, 303.15 K, 308.15 K and normal atmospheric pressure have been measured. The experimental data were used to compute the density, deviation in molar refraction, deviation in viscosity and excess Gibbs free energy of activation as a function of composition. The experimental viscosity data have been correlated with empirical equations like Grunberg-Nissan, Herric correlation and three body McAllister's equation. The excess thermodynamic properties were fitted to Redlich-Kister polynomial equation. The variation of these properties with composition and temperature of the binary mixtures are discussed in terms of intermolecular interactions.

Keywords—Cumene, excess Gibbs free energy, excess molar volume, morpholine.

I. INTRODUCTION

THE physicochemical properties of liquid mixtures have attracted much attention from both theoretical and engineering applications point of view. Such results are frequently used in design, process flow, mass transfer or heat transfer calculations in many chemical and industrial processes [1]. In recent years, use of mixed organic solvents has gained importance for the researchers, as some mixed organic solvent may have better properties than a pure solvent. A detailed search in the literature shows that very few measurements have been made on the density, viscosity studies for binary mixtures containing morpholine as one of the components. Awwad et al. [2] measured densities of binary mixtures of morpholine with methanol, benzene, chloroform, acetone at 298.15 K. Sangita Sharma et al. [3] measured the densities and viscosities of morpholine with some aromatic hydrocarbons (benzene, toluene, *m*-Xylene and mesitylene) at temperatures from 303.15 to 313.15 K. As part of our experimental investigation of the thermophysical

properties of liquid mixtures with morpholine as one of the components, we present here the experimental data on refractive index, excess molar volume, and viscosity for binary mixtures of morpholine and cumene over the whole composition range and at temperatures from 298.15 K to 308.15 K and atmospheric pressure.

Morpholine [4] is a chemical used in variety of applications such as rubber industry, corrosion inhibition, pharmaceuticals, dye industries, crop pesticide industries and as a solvent in various manufacturing processes.

Cumene [5], an aromatic hydrocarbon is used in the manufacture of rubber, iron and steel and pulp and paper. Cumene in minor amounts is used as a thinner for paints, enamels and lacquers and as a component of high octane fuels.

II. EXPERIMENTAL METHODOLOGY

A. Chemicals

Morpholine (AR GRADE, Sisco Research Lab. Pvt. Ltd., Mumbai) and Cumene (AR GRADE, Sigma-Aldrich, China) were used in this study, after purification using standard methods [6]. The purity of the chemicals was checked by measuring the refractive indices (n_D) and viscosities (η) of the pure compounds and comparing them with the literature values. The results are reported in Table I. Mercury was obtained from S.D. Fine Chem. Ltd., Mumbai.

B. Apparatus and Procedure

Binary mixtures were prepared by mass in airtight bottles. The mass measurement of pure liquids and liquid mixtures were formed on an electronic balance. The measurement range of weight balance is 220 g to 0.1 mg with an accuracy of ± 0.0001 . The mixtures were well mixed by shaking before use.

Refractive indices were measured using a Bausch and Lomb Abbe-3L refractometer [7]. Refractive index readings were taken on samples with sufficient time allowed for the sample to come to thermal equilibrium at the desired temperature with the help of a circulating type cryostat (Type MK70, MLW, Germany) maintained at a temperature with in ± 0.02 K. Same thermostatic bath was used for maintaining temperatures of samples for measurement of other properties.

Viscosities were measured by using a modified Ubbelohde viscometer. The viscometer was calibrated and two constants

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A and B were obtained by measuring the flow time with distilled toluene and cyclohexane using the equation $\eta/\rho = (A \times t + B/t)$. The flow measurements were made with an electronic stopwatch with a precision of ± 0.01 s. An average of three to four sets of flow times was taken for each liquid mixture. The reproducibility of the viscosity estimates is found to be within ± 0.003 cp.

Molar excess volumes were measured using specially designed V-shaped dilatometer. It consists of two limbs having vacuum tested standard joints at its two ends. The two limbs are interconnected by small bent tubing which is further connected to a thin uniform bored capillary having a reference mark R on it which is fused to one of the limbs. Molar excess volume of the binary mixture was calculated using:

$$V^E = \frac{\pi r^2 \Delta h}{(n_1 + n_2)}$$

where, Δh is the change in height of liquid level in capillary before and after mixing and n_1, n_2 are the number of moles of liquid A and B, respectively.

Area of the capillary tube was evaluated from the weight of mercury column of average length in the capillary. The length of the mercury column at various positions in the capillary was read by a travelling microscope that could read to ± 0.001 cm.

TABLE I
COMPARISON OF EXPERIMENTAL AND LITERATURE VALUES OF REFRACTIVE INDEX AND VISCOSITY FOR PURE COMPONENTS

Component	T(K)	n_D		η (cP)	
		Exp.	Lit.	Exp.	Lit.
Morpholine	298.15	1.452345	1.453	2.0283	2.02
	303.15	1.450095	-	1.7408	1.740
	308.15	1.447864	-	1.6116	-
Cumene	298.15	1.489025	1.488	0.7441	0.739
	303.15	1.486542	-	0.6940	0.6881
	308.15	1.483994	-	0.6497	0.6488

III. TREATMENT OF DATA

The experimental excess molar volume as a function of mole fraction of morpholine for the Morpholine + Cumene binary mixture at 298.15, 303.15 and 308.15 K are presented in Tables II, III and IV respectively. From the experimental excess molar volume data, density of the binary mixture is calculated using:

$$\rho_m = \frac{(M_1 x_1 + M_2 x_2)}{(V^E + x_1 V_1^0 + x_2 V_2^0)}$$

where x_1 and x_2 are the mole fractions of components 1 and 2 respectively; V^E is excess molar volume; ρ_m is the density of binary mixture, V_1^0 and V_2^0 are molar volumes of pure components 1 and 2 respectively.

The experimental refractive index data for the Morpholine + Cumene binary mixture at 298.15, 303.15 and 308.15 K are presented in Tables V, VI and VII respectively. Molar refraction values are calculated from the experimentally

determined refractive index data using Lorentz-Lorentz equation:

$$R_m = \frac{(n_D^2 - 1)}{(n_D^2 + 2)} \times \frac{\sum x_i M_i}{\rho_m}$$

$$R_i = \frac{(n_D^2 - 1)}{(n_D^2 + 2)} \times \frac{M_i}{\rho_i}$$

where x_i is mole fraction of pure component i , M_i is molar mass of pure component i

Deviation in molar refraction (ΔR) is calculated using:

$$\Delta R = R_m - \sum x_i R_i$$

The deviation in molar refraction as a function of mole fraction of morpholine for the binary mixture at 298.15, 303.15 and 308.15 K are also presented in Tables V, VI and VII, respectively.

The experimental value of viscosity (η) and deviation in viscosity ($\Delta\eta$) as a function of mole fraction of morpholine for Morpholine + Cumene binary mixture at 298.15, 303.15 and 308.15 K are also presented in Tables V, VI and VII, respectively. Deviation in viscosity ($\Delta\eta$) is calculated using:

$$\Delta\eta = \eta_m - (x_1 \eta_1 + x_2 \eta_2)$$

where x_1, x_2 are the mole fractions of component 1 and 2, respectively; η_1, η_2 are dynamic viscosities of component 1 and 2, respectively; $\eta_m, \Delta\eta$ are dynamic viscosity of the binary mixture and deviation in viscosity, respectively.

The excess Gibbs free energy of activation for viscous flow, G^E was calculated by:

$$\frac{G^E}{RT} = \ln(\eta V_m) - \sum_{i=1}^2 x_i \ln(\eta_i V_i)$$

where, R is universal gas constant, T is temperature in Kelvin, V_1 and V_2 are the molar volumes of the pure components 1 and 2 and V_m is the molar volume of the mixture.

Excess Gibbs free energy of activation, G^E as a function of mole fraction of morpholine for Morpholine + Cumene binary mixture at 298.15, 303.15 and 308.15 K are presented in Table VIII.

The experimentally determined values of V^E , $\Delta\eta$ and G^E for the binary mixtures were fitted to the Redlich-Kister equation [8]:

$$A = x_1 x_2 \sum_{j=1}^n A_{j-1} (x_1 - x_2)^{j-1}$$

where A is the property under consideration; A_{j-1} is the polynomial coefficient; n is the polynomial degree and x_1, x_2 are the mole fractions of component 1 and 2, respectively.

TABLE II
EXCESS MOLAR VOLUME (V^E) AND DENSITY (ρ) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT 298.15K

x_1	V^E (cm^3/mol)	ρ (g/cm^3)
0.0000	0.0000	0.8574
0.1960	0.1194	0.8749
0.3914	0.2043	0.8954
0.4849	0.1971	0.9070
0.5698	0.1618	0.9186
0.6294	0.1380	0.9273
0.6462	0.1259	0.9299
0.6606	0.1127	0.9322
0.7320	0.0843	0.9437
0.8004	0.0575	0.9555
0.9011	0.0216	0.9746
1.0000	0.0000	0.9955

TABLE III
EXCESS MOLAR VOLUME (V^E) AND DENSITY (ρ) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT 303.15K

x_1	V^E (cm^3/mol)	ρ (g/cm^3)
0.0000	0.0000	0.8530
0.2073	0.1745	0.8713
0.3746	0.2434	0.8890
0.4725	0.2373	0.9010
0.5317	0.2021	0.9090
0.6266	0.1635	0.9227
0.6418	0.1510	0.9250
0.6577	0.1446	0.9275
0.7297	0.1141	0.9391
0.7823	0.0872	0.9483
0.8890	0.0425	0.9684
1.0000	0.0000	0.9920

TABLE IV
EXCESS MOLAR VOLUME (V^E) AND DENSITY (ρ) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT 308.15K

x_1	V^E (cm^3/mol)	ρ (g/cm^3)
0.0000	0.0000	0.8488
0.2206	0.2246	0.8684
0.3848	0.2944	0.8861
0.4568	0.2738	0.8951
0.5368	0.2415	0.9059
0.6294	0.1975	0.9195
0.6485	0.1852	0.9225
0.6902	0.1845	0.9212
0.7307	0.1404	0.9360
0.7922	0.1054	0.9469
0.8825	0.0732	0.9641
1.0000	0.0000	0.9897

The standard deviation (σ) is calculated using equation [9]:

$$\sigma(X) = \left[\frac{\sum (X_{exp} - X_{cal})^2}{N - n} \right]^{1/2}$$

where X is the property under consideration; X_{exp} , X_{cal} are the experimental and calculated values respectively, N is the number of data points and n is the number of coefficients.

TABLE V
REFRACTIVE INDEX (n_D), DEVIATION IN MOLAR REFRACTION (ΔR),
VISCOSITY (η) AND DEVIATION IN VISCOSITY ($\Delta\eta$) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT T=298.15K

x_1	n_D	ΔR	η (cp)	$\Delta\eta$ (cp)
0.0000	1.489025	0.0014	0.7441	0.0000
0.1299	1.485721	0.0127	0.8856	-0.0253
0.1558	1.484975	0.0158	0.9136	-0.0306
0.2825	1.482095	0.0432	1.0551	-0.0517
0.4024	1.478066	0.0558	1.1959	-0.0650
0.5129	1.474101	0.0719	1.3335	-0.0693
0.6159	1.470521	0.0662	1.4721	-0.0629
0.7103	1.466949	0.0551	1.6030	-0.0533
0.7933	1.462932	0.0404	1.7214	-0.0414
0.8686	1.460083	0.0357	1.8324	-0.0271
0.9327	1.455595	0.0132	1.9268	-0.0151
0.9662	1.453569	0.0055	1.9762	-0.0087
1.0000	1.452345	-0.0009	2.0283	0.0000

TABLE VI
REFRACTIVE INDEX (n_D), DEVIATION IN MOLAR REFRACTION (ΔR),
VISCOSITY (η) AND DEVIATION IN VISCOSITY ($\Delta\eta$) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT T=303.15K

x_1	n_D	ΔR	η (cp)	$\Delta\eta$ (cp)
0.0000	1.486542	0.0000	0.6940	0.0000
0.1299	1.483313	0.0221	0.8101	-0.0199
0.1558	1.481681	0.0271	0.8318	-0.0253
0.2825	1.480082	0.0586	0.9445	-0.0452
0.4024	1.476027	0.0745	1.0572	-0.0580
0.5129	1.471720	0.0894	1.1694	-0.0615
0.6159	1.468439	0.0909	1.2823	-0.0565
0.7103	1.464343	0.0741	1.3899	-0.0477
0.7933	1.460601	0.0593	1.4901	-0.0344
0.8686	1.457294	0.0412	1.5816	-0.0217
0.9327	1.453438	0.0239	1.6587	-0.0116
0.9662	1.451684	0.0099	1.7000	-0.0054
1.0000	1.450095	0.0000	1.7408	0.0000

TABLE VII
REFRACTIVE INDEX (n_D), DEVIATION IN MOLAR REFRACTION (ΔR),
VISCOSITY (η) AND DEVIATION IN VISCOSITY ($\Delta\eta$) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT T=308.15K

x_1	n_D	ΔR	η (cp)	$\Delta\eta$ (cp)
0.0000	1.483994	0.0000	0.6497	0.0000
0.1299	1.480651	0.0314	0.7287	-0.0150
0.1558	1.479954	0.0385	0.7493	-0.0202
0.2825	1.476801	0.0770	0.8556	-0.0403
0.4024	1.473685	0.0950	0.9639	-0.0516
0.5129	1.469289	0.1148	1.0705	-0.0552
0.6159	1.466203	0.1200	1.1779	-0.0506
0.7103	1.462681	0.1056	1.2809	-0.0417
0.7933	1.458989	0.0821	1.3751	-0.0303
0.8686	1.455649	0.0714	1.4618	-0.0187
0.9327	1.451799	0.0395	1.5363	-0.0082
0.9662	1.449527	0.0256	1.5757	-0.0022
1.0000	1.447864	0.0000	1.6116	0.0000

The coefficients and their mean standard deviations are listed in Table IX.

To estimate the viscosity of binary liquid mixtures in terms of pure component values, the experimental viscosity data of the mixtures were fitted to the following empirical relations.

TABLE VIII
EXCESS GIBBS FREE ENERGY OF ACTIVATION (G^E) FOR THE MORPHOLINE +
CUMENE BINARY MIXTURE AT 298.15, 303.15 AND 308.15 K

x_1	G^E (J/mol)		
	298.15K	303.15K	308.15K
0.0000	0.0000	0.0000	0.0000
0.1299	0.1417	0.1181	0.0229
0.1558	0.1558	0.1271	0.0389
0.2825	0.2184	0.1754	0.1043
0.4024	0.2436	0.1934	0.1400
0.5129	0.2407	0.1962	0.1572
0.6159	0.2287	0.1889	0.1593
0.7103	0.1974	0.1618	0.1453
0.7933	0.1583	0.1377	0.1248
0.8686	0.1094	0.0982	0.0908
0.9327	0.0583	0.5230	0.0545
0.9662	0.0281	0.0296	0.0327
1.0000	0.0000	0.0000	0.0000

A. Heric's Correlation

Kinematic viscosities were fitted to Heric's Correlation [10],

$$\ln v = x_1 \ln v_1 + x_2 \ln v_2 + x_1 x_2 [\alpha_{12} + \alpha'_{12}(x_1 - x_2)] - \ln M_{\text{mix}} + x_1 \ln M_1 + x_2 \ln M_2$$

where v , v_1 , v_2 are the kinematic viscosity values of mixture and pure components 1 and 2 respectively; x_1 , are the mole fractions of component 1 and 2, respectively and M_1 , M_2 are the molecular weights of pure components 1 and 2 respectively. α_{12} and α'_{12} are coefficients of Heric's correlation.

B. Grunberg and Nissan Relation

Grunberg and Nissan have formulated equation to access molecular interactions leading to viscosity changes. Interaction parameter d , which is a measure of the strength of the interaction between components of the binary mixture can be estimated using the relation [11]:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d$$

where, η is the dynamic viscosity of mixture, η_1 is the viscosity of pure morpholine, η_2 is the viscosity of pure cumene and x_1 , x_2 are the mole fractions of pure components 1 and 2 respectively.

C. McAllister Equation [12]

The experimentally determined values of viscosity were fitted to McAllister model given by:

$$\begin{aligned} \ln v = & x_1^3 \ln v_1 + x_2^3 \ln v_2 + 3 x_1^2 x_2 \ln A_{12} + 3 x_1 x_2^2 \ln A_{21} \\ & - \ln \left[x_1 + x_2 \frac{M_2}{M_1} \right] + 3 x_1^2 x_2 \ln \left[\frac{2 + (M_2/M_1)}{3} \right] \\ & + 3 x_1 x_2^2 \ln \left[\frac{1 + (2M_2/M_1)}{3} \right] + x_2^3 \ln \left[\frac{M_2}{M_1} \right] \end{aligned}$$

where v , v_1 , v_2 are the kinematic viscosity values of mixture and pure component respectively; x_1 , are the mole fractions of pure components 1 and 2 respectively; M_1 , M_2 are the molar

volume of pure components 1 and 2 respectively and A_{12} , A_{21} are the fitting parameters that give viscosity contribution for interaction between unlike components.

The evaluated interaction parameters of Heric's correlation α_{12} and α'_{12} ; Grunberg and Nissan parameter, d ; McAllister interaction parameters A_{12} and A_{21} are presented in Table X.

IV. RESULTS AND DISCUSSION

Fig. 1 reveals that deviation in molar refraction are positive for the Morpholine + Cumene binary mixture over the whole composition range at the selected temperatures i.e. at 298.15, 303.15 and 308.15 K. With increase in temperature, deviation in molar refraction increases.

Experimental viscosity measurements for the Morpholine + Cumene binary mixture over the entire composition range at 298.15 K, 303.15 K and 308.15 K are presented in Tables V, VI and VII respectively. It is observed that with the increase in temperature, viscosity of mixture decreases. From Fig. 2, it is observed that viscosity deviations are negative over the entire composition range at 298.15, 303.15 and 308.15 K. Negative values suggest that the intermolecular interactions become weaker on mixing the two components together, thus indicating that dispersive forces are predominant in this binary mixture.

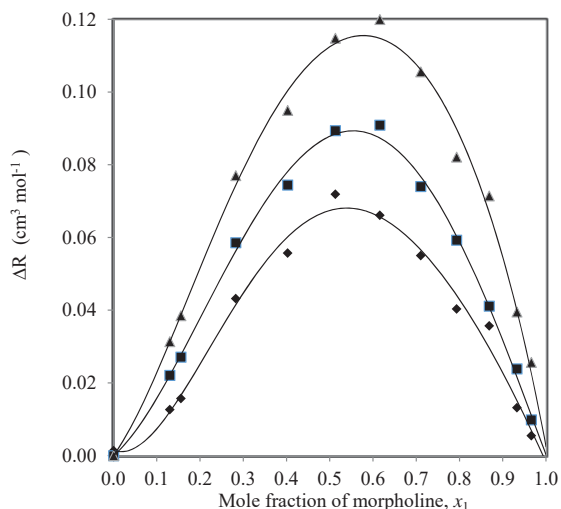


Fig. 1 Deviation in molar refraction (ΔR) vs. mole fraction of Morpholine (x_1) at 298.15 K (\blacklozenge), 303.15 K (\blacksquare) and 308.15 K (\blacktriangle)

Fig. 3 shows the experimental excess molar volume values are positive for the Morpholine + Cumene binary mixture over the whole composition range at 298.15, 303.15 and 308.15 K. The positive values of V^E indicate that there is a volume expansion. This signifies that the mixtures are less compressible than the corresponding ideal mixtures [3]. Positive V^E values suggest the presence of weak dispersive forces.

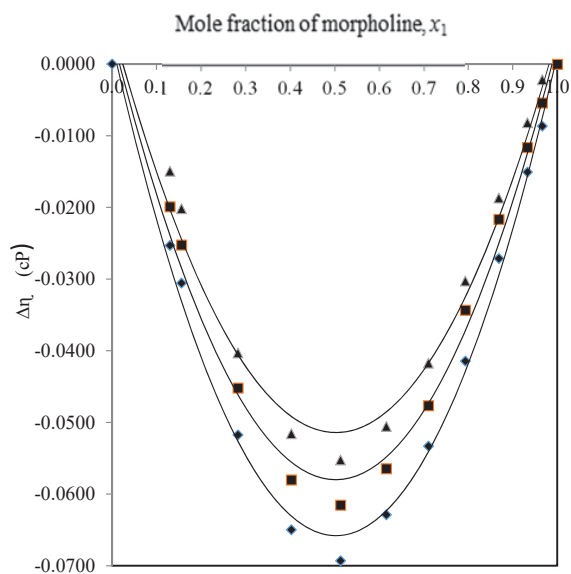


Fig. 2 Deviation in viscosity ($\Delta\eta$) vs. mole fraction of Morpholine (x_1) at 298.15 K (♦), 303.15 K (■) and 308.15 K (▲).

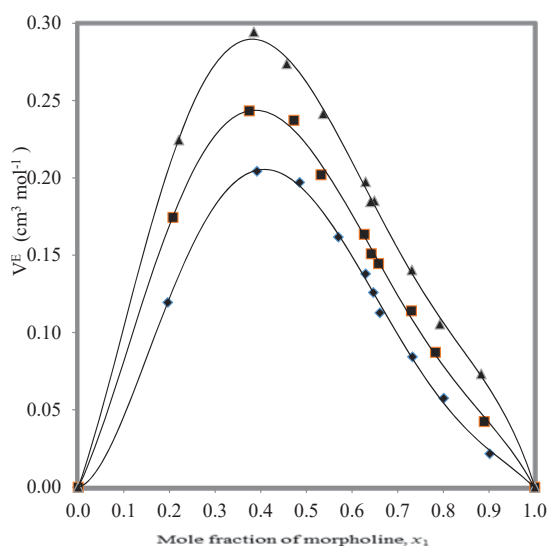


Fig. 3 Excess molar volume (V^E) vs. mole fraction of Morpholine (x_1) at 298.15 K (♦), 303.15 K (■) and 308.15 K (▲)

From Fig. 4, it is observed that the values of excess Gibbs free energy of activation are slightly positive over the entire composition range for all the selected temperatures.

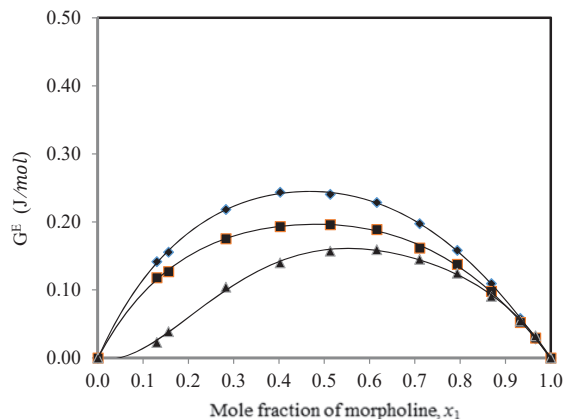


Fig. 4 Excess Gibbs free energy (G^E) vs. mole fraction of Morpholine (x_1) at 298.15 K (♦), 303.15 K (■) and 308.15 K (▲)

TABLE IX
COEFFICIENTS (A_{j-1}) OF REDLICH-KISTER EQUATION AND STANDARD DEVIATION OF V^E , $\Delta\eta$, ΔR AND G^E FOR MORPHOLINE + CUMENE BINARY MIXTURE AT 298.15, 303.15 AND 308.15 K

Excess property	T(K)	A_0	A_1	A_2	A_3	σ
V^E (cm^3/mol)	298.15	0.0015	1.2134	-2.2426	1.0244	0.2179
	303.15	0.0003	1.4161	-2.5525	1.1347	0.2721
	308.15	0.0009	1.6982	-3.0558	1.3643	0.3265
$\Delta\eta$ (cp)	298.15	0.0006	-0.1802	-0.0006	0.1878	0.0571
	303.15	0.0008	-0.1583	-0.0005	0.1667	0.0501
	308.15	-0.0002	-0.1397	-0.0004	0.1489	0.0439
ΔR (cm^3/mol)	298.15	0.0005	0.1391	0.0791	-0.2248	0.0515
	303.15	0.0004	0.2057	0.0473	-0.2583	0.0703
	308.15	0.0001	0.2917	-0.0002	-0.2877	0.0973
G^E (J/mol)	298.15	-0.0101	0.7013	0.0026	-0.7237	0.2253
	303.15	-0.0076	0.5716	0.0019	-0.5838	0.1844
	308.15	0.0035	0.3918	-0.0006	-0.3876	0.1319

TABLE X
INTERACTION PARAMETERS FOR MORPHOLINE + CUMENE BINARY MIXTURE AT 298.15, 303.15 AND 308.15K

Interaction parameters	T(K)			σ		
	298.15K	303.15K	308.15K	298.15K	303.15K	308.15K
Heric's:				0.0042	0.0047	0.0037
α_{12}	0.4133	0.3423	0.2382			
α'_{12}	-0.0661	-0.0329	0.1381			
Grunberg-Nissan: d				0.0066	0.0815	0.0112
McAllister:				0.0042	0.0047	0.0037
A_{12}	1.7007	1.4880	1.4183			
A_{21}	1.3386	1.1781	1.0067			

ACKNOWLEDGMENT

The authors gladly acknowledge financial support by TEQIP-II and DST (PURSE grant).

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