

Thermodynamic Excess Properties and Intermolecular Interaction Using Fourier Transform Infrared for the Dodecylbenzenesulfonic Acid-Acetone Binary Liquid Mixture

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Abstract—Density, viscosity, and surface tension for the dodecylbenzenesulfonic acid (DBSA)-acetone system at the temperatures (293.15°K, 298.15°K, and 301.15°K) have been investigated experimentally. Excess molar volume, viscosity deviation, surface tension deviation, and molar excess Gibbs free energy deviation are calculated at (293.15°K, 298.15°K, and 301.15°K). All the non-ideal properties calculated are fitted to Redlich-Kister equation. The Fourier transform infrared (FTIR) spectrum of the DBSA-acetone mixture has been characterized for studying the intermolecular interaction. The result of molar excess Gibbs free energy deviation and FTIR studies showed a weak hydrogen bonding for the mixture solution. The intra-molecular interaction is strong in the case of pure DBSA and acetone due to the self-association of DBSA molecule or decreasing the dipole-dipole of acetone molecule. The result of FTIR supported the result of thermodynamic excess, where the excess molar volumes were positive. The calculated values of excess molar volume are positive for all the temperatures ranging from 293.15 to 301.15°K. It was found that the values of viscosity deviation, surface tension deviation, and molar excess Gibbs free energy deviation are negative for all the temperatures studied.

Index Terms—Acetone, Density, Dodecylbenzenesulfonic acid, Excess properties, Fourier Transform infrared spectral analysis, Hydrogen bonding, Intermolecular interaction, Surface tension, Viscosity.

I. INTRODUCTION

Surfactants are widely used in the chemical industries, and the dodecylbenzenesulfonic acid (DBSA) is one of the very important anionic surfactant which can be used as a brønsted acid-catalyst in a wide range of chemical reactions. DBSA had the dual role of surfactant and acid

catalyst (Shrikhande, 2007). In many studies, the role of DBSA has been investigated as an acid catalyst for the esterification (Jing, et al., 2008), dehydration (Chai, et al., 2007), Mannich type (Shiri and Zolfigol, 2009), and thia-Michael addition reactions in water (Manabe, et al., 2002). The hydrophobic property of DBSA has also been studied (Manabe, et al., 2001). As a dopant, DBSA has been tested in the synthesis of poly-aniline (Haba, et al., 1999; Machado, et al., 2006). Moreover, the large alkyl groups of DBSA can improve the melting and solution processability for the poly-aniline (Han, et al., 2002). The study of the thermodynamic properties and the intermolecular interactions for the binary mixture of DBSA-acetone system is very important in the modeling, design, and optimization of the chemical industrial processes. The behavior of the solution and the type of interaction has been explained through the present study, and the data provided may play a vital role in the industrial synthesis of many organic products, solvent extraction, and many other mass transfer phenomena. Acetone has been chosen as a solvent in the present work because of the high polar and self-associative nature of it. Many of the spectroscopic studies have focused on the acetone (Schindler, et al., 1982; Ancian, et al., 1983; Knözinger and Wittenbeck, 1984; Jalilian and Zahedi-Tabrizi, 2008; Shikata, et al., 2012; Kollipost, et al., 2014; Srivastava, et al., 2003). In a recent research, the shift in the frequency of C=O and CH₃ group has been interpreted (Arivazhagan, et al., 2015). A combination of brønsted acid+ hydroxymethyl group binary system was discussed in a recent paper (Liu, et al., 2017). To the best of our knowledge, no work has been focused on the DBSA-acetone mixture which is used industrially and deviate from the ideality of the pure liquid state leading to the errors in the equation of state calculation and the other thermodynamic calculations. The deviation from the ideality can affect the design calculation for the industrial units, which contains a binary liquid system of DBSA-acetone. The novelty of this paper depends on the analysis thermodynamic properties and interactions between the molecules of DBSA and acetone. The objective of the present work is to provide

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an experimental data of density, viscosity, and surface tension for the pure compound and the thermodynamic excess volume, viscosity deviation, surface tension deviation, and excess Gibbs free energy of activation for the binary mixture of DBSA-acetone system. Moreover, the intermolecular interaction has been investigated using the Fourier transform infrared (FTIR) spectroscopy method.

II. MATERIALS AND PROCEDURE

A. Materials

DBSA (>0.99) was procured from Shanghai Hanhong Scientific Co.,Ltd., and acetone (>0.997 GC) of a high chromatographic purity was procured from Sigma-Aldrich. No further purification has been done for the reagents. All the chemical materials used were analytical reagents.

B. Measuring and Procedure

An electronic balance with a precision of $\pm 10^{-4}$ (OHAUS) has been used in the preparation of (0.1-0.9 mole percent) of DBSA-acetone binary mixtures. The experimental uncertainty of mole fractions were $\leq \pm 0.0003$. Density of the pure and DBSA-acetone binary mixtures were measured using Anton-Paar dens meter (DSA111 5000) with uncertainty in density of $\pm 2 \times 10^{-3} \text{ kg m}^{-3}$. Anton-Paar viscometer has been used for measuring the dynamic viscosity (η) of the pure and binary liquid mixtures for DBSA-acetone with uncertainty of $\pm 0.06 \text{ mPa.s}$. The pendant drop method (KRÜSS GmbH) was used for testing the surface tension of the pure and the binary liquid mixtures. Each test was conducted in triplicate. The FTIR spectra for the pure and binary liquid mixtures for DBSA-acetone are recorded using (Tensor 37, Bruker) in the range $400\text{-}4000 \text{ cm}^{-1}$, a resolution of 4 cm^{-1} and average of 16 scans were recorded.

III. THEORY

The experimental density (ρ) data were used for calculating the excess molar volumes (VE) according to the following equation,

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho_m} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (1)$$

Where, x , M , and ρ the mole fraction, molecular weight, and density, respectively. The subscript 1, 2, and m denoted DBSA, acetone, and the DBSA-acetone mixture, respectively.

The deviation in viscosity ($\Delta\eta$) was calculated from the experimental data of viscosity (η) as the following equation,

$$\Delta\eta = \eta_m - (x_1 \eta_1 + x_2 \eta_2) \quad (2)$$

Where, x , η the mole fraction, and viscosity, respectively. The subscript 1, 2, m denoted DBSA, acetone, and the DBSA-acetone mixture, respectively.

The surface tension deviation ($\Delta\sigma$) was calculated from the experimental data of surface tension (σ) as the following equation,

$$\Delta\sigma = \sigma_m - (x_1 \sigma_1 + x_2 \sigma_2) \quad (3)$$

Where, x , σ the mole fraction, and surface tension, respectively. The subscript 1, 2, m denoted DBSA, acetone, and the DBSA-acetone mixture, respectively.

Molar excess Gibbs free energy of activation (ΔG^{*E}) was calculated from the experimental molar volume (V) and viscosity (η) data as the following equation,

$$\Delta G^{*E} = RT[\ln(\eta_m V_m) - (x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2))] \quad (4)$$

Where, x , η , R , T , V the mole fraction, viscosity, universal gas constant, absolute temperature, and molar volume, respectively. The subscript 1, 2, m denoted DBSA, acetone, and the DBSA-acetone mixture, respectively.

Excess molar volumes, deviations in the viscosity, surface tension, and molar excess Gibbs free energy for the mixtures of DBSA-acetone were fitted to Redlich-Kister (RK) equation as follow (Tahery, 2017):

$$RK^E = x_1 x_2 \sum_{i=0}^2 A_i (x_1 - x_2)^i \quad (5)$$

Where, x_1 , x_2 , and A_i are the mole fraction of DBSA, mole fraction of acetone, and the binary coefficients, respectively. The coefficients of RK equation are calculated using least squares method and the standard deviation (SD) for the data are calculated using the following relation:

$$SD_{F^E} = \left(\frac{\sum_{i=1}^M (F_{\text{exp}}^E - F_{\text{cal}}^E)^2}{M - N} \right)^{1/2} \quad (6)$$

Where, M , N are the number of experimental data and the number of polynomial coefficients, respectively.

IV. RESULTS AND DISCUSSION

A. Thermodynamic Properties

The data of density, viscosity, and surface tension for the pure DBSA and acetone were compared with the literature data and presented in Table I. For the binary mixtures of DBSA-acetone, at three temperatures (293.15°K , 298.15°K , and 301.15°K), the density, viscosity, surface tension, excess molar volume, viscosity deviation, surface tension deviation, and molar excess Gibbs free energy were presented in Table II. All the data showed a nonlinear increase or decrease in the property with mole fraction due to the non-ideal behavior of DBSA-acetone binary mixture. The intermolecular and intramolecular interactions between the components were concluded from the non-ideal behavior of the binary system (Gnanakumari, et al., 2007). The calculated values of the excess molar volumes for the DBSA-acetone binary system exhibited a positive values for all mole fractions and temperatures, as presented in the Table II and Fig. 1. Moreover, the values of excess molar volumes increase with increasing temperature. The maximum value of excess molar volume is positioned at 0.4 mole fraction of DBSA.

The same scenario was concluded for the viscosity deviation, but with negative values for all mole fractions and

TABLE I
EXPERIMENTAL AND LITERATURE VALUES OF DENSITY ρ , DYNAMIC VISCOSITY η , AND SURFACE TENSION σ FOR THE PURE DBSA AND ACETONE AT 298.15 K

Component	$10^{-3}\rho$, (kg.m ⁻³)		η , (mPa.s)		σ , (mN. m ⁻¹)	
	Experimental	Literature	Experimental	Literature	Experimental	Literature
DBSA	1.05343	1.05354 (Kumar, et al., 2016) [24]	1056	1055.9 (Kumar, et al., 2016) [24]	39.91	39.89 (Kumar, et al., 2016) [24]
Acetone	0.74924	0.74879 (Qian, et al., 2011) [25]	0.33	0.329 (Qian, et al., 2011) [25]	24.21	24.19 (Dash and Mohanty) [26]

DBSA: Dodecylbenzenesulfonic acid

TABLE II
DENSITY (ρ), EXCESS MOLAR VOLUME (VE), DYNAMIC VISCOSITY (η), VISCOSITY DEVIATION ($\Delta\eta$), SURFACE TENSION (σ), SURFACE TENSION DEVIATION ($\Delta\sigma$), EXCESS MOLAR GIBBS FREE ENERGY OF ACTIVATION ΔG^{*E} FOR THE BINARY MIXTURE OF DBSA-ACETONE AT TEMPERATURES (293.15-303.15) K AND $P=1$ ATM

X_1	$10^{-3}\rho$, (kg.m ⁻³)	$10^6 VE$, (m ³ .mol ⁻¹)	η , (mPa.s)	$\Delta\eta$	σ , (mN. m ⁻¹)	$\Delta\sigma$	ΔG^{*E} , J.mol ⁻¹
293.15°K							
0.0	0.75352	0.000000	000.340	0.000	24.28	0.000	000.0
0.1	0.80465	4.784837	104.785	-1.112	25.53	-0.249	-648.2
0.2	0.86753	4.838747	209.987	-1.477	26.78	-0.568	-922.7
0.3	0.89942	6.873256	315.483	-1.548	27.89	-1.027	-989.2
0.4	0.93247	6.955084	421.097	-1.501	29.23	-1.256	-950.3
0.5	0.95703	7.205812	526.856	-1.309	30.34	-1.715	-865.3
0.6	0.97796	7.108916	632.721	-1.011	32.64	-0.984	-654.6
0.7	0.99687	6.540582	738.583	-0.716	34.48	-0.713	-597.8
0.8	1.01588	5.101230	844.343	-0.523	36.26	-0.502	-396.9
0.9	1.04048	1.304930	950.112	-0.321	38.19	-0.141	-198.7
1.0	1.05937	0.000000	1056.87	0.000	39.97	0.000	000.0
298.15°K							
0.0	0.74924	0.000000	000.330	0.000	24.21	0.000	000.0
0.1	0.80164	5.180777	104.985	-0.912	25.12	-0.659	-612.4
0.2	0.85996	5.972145	210.087	-1.377	26.43	-0.918	-878.3
0.3	0.89579	7.497479	315.498	-1.533	27.63	-1.287	-928.5
0.4	0.92602	8.190561	421.197	-1.401	29.23	-1.256	-897.8
0.5	0.95103	8.473149	526.956	-1.209	30.1	-1.955	-815.6
0.6	0.97326	8.190809	632.821	-0.911	32.41	-1.214	-610.4
0.7	0.99207	7.734298	738.683	-0.616	34.36	-0.833	-556.8
0.8	1.01388	5.630941	844.353	-0.513	36.02	-0.742	-365.8
0.9	1.04021	1.379681	950.122	-0.311	37.96	-0.371	-167.9
1.0	1.05343	0.000000	1056.00	0.000	39.91	0.000	000.0
303.15°K							
0.0	0.74435	0.000000	000.325	0.000	24.01	0.000	000.0
0.1	0.79164	6.517810	105.085	-0.812	25.01	-0.769	-576.4
0.2	0.84996	7.500318	210.287	-1.177	26.33	-1.018	-837.9
0.3	0.89279	8.017197	315.698	-1.333	27.53	-1.387	-878.7
0.4	0.92428	8.526806	421.397	-1.201	29.13	-1.356	-847.6
0.5	0.94903	8.899156	527.056	-1.109	30.01	-2.045	-765.8
0.6	0.97108	8.696178	632.921	-0.811	32.31	-1.314	-571.4
0.7	0.99001	8.250151	738.783	-0.516	34.26	-0.933	-517.2
0.8	1.01088	6.429437	844.453	-0.413	35.92	-0.842	-334.7
0.9	1.04001	1.435077	950.222	-0.211	37.86	-0.471	-136.8
1.0	1.05021	0.000000	1055.60	0.000	39.51	0.000	000.0

DBSA: Dodecylbenzenesulfonic acid

temperatures. The negative values of viscosity deviation are reduced with increasing temperature. The maximum value of viscosity deviation positions at nearly 0.3 mole fraction of DBSA, as presented in the Table II and Fig. 2. The deviation of surface tension data for the DBSA-acetone binary system, presented in the Table II and Fig. 3, are negative at all mole fractions and temperatures and the maximum value is located at about 0.6 mole fraction of DBSA. In addition, the negative values of surface tension are reduced with increasing temperature. As stated in the literature, the molar excess Gibbs free energy of activation had a vital role in the study of the molecular interaction (Bajić, et al., 2013a; 2013b). For

the interpretation of inter- and intra-molecular interaction in the DBSA-acetone binary system, the molar excess Gibbs free energy of activation is calculated and the data are presented in Table II and Fig. 4. All the calculated values of molar excess Gibbs free energy values are negative for all the mole fractions and temperatures range and have a maximum negative value at a mole fraction of 0.3. The negative values of the molar excess Gibbs free energy increase with increasing temperature. The coefficients and results of the R-K equation for all functions are presented in Table III and for all the functions in the Figs. 1-4. Moreover, the values of SDs are given in Table III as calculated using the equation (6).

TABLE III

CALCULATED RK COEFFICIENTS FOR EXCESS MOLAR VOLUME (V^E), VISCOSITY DEVIATION ($\Delta\eta$), SURFACE TENSION DEVIATION ($\Delta\sigma$), EXCESS MOLAR GIBBS FREE ENERGY OF ACTIVATION ΔG^{*E} FOR THE BINARY MIXTURE OF DBSA-ACETONE AT TEMPERATURES (293.15-303.15) K AND P=1 ATM

Function	Temperature ($^{\circ}\text{K}$)	A_0	A_1	A_2	A_3	A_4	SD
V^E	293.15	0.332	19.06	52.318	-147.2	75.266	0.00315
	298.15	0.282	26.106	24.154	-103.23	52.285	0.00321
	301.15	0.4073	61.655	-121.3	95.805	-36.865	0.00322
$\Delta\eta$	293.15	-0.0251	-13.592	38.113	-38.953	14.452	0.03674
	298.15	0.0185	-12.805	35.188	-35.026	12.602	0.02956
	301.15	0.0017	-10.576	27.469	-25.139	8.2314	0.02187
$\Delta\sigma$	293.15	0.0151	-1.453	-15.592	36.552	-19.551	0.00020
	298.15	-0.0141	-6.7685	9.5825	-2.2591	-0.5536	0.00015
	301.15	-0.0282	7.9855	15.744	-12.801	5.0699	0.00010
ΔG^{*E}	293.15	-9.6839	-8170.4	22183	-22658	8667.2	7.5
	298.15	-10.113	-7692.2	20750	-20916	7882.6	7.4
	301.15	-9.4252	-7261.1	19425	-19248	7109.3	7.2

RK: Redlich-Kister

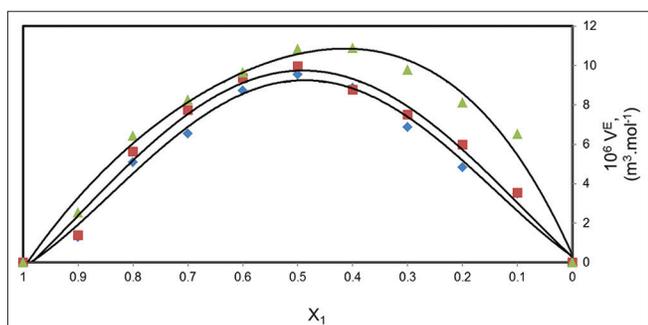


Fig. 1. Excess molar volume V^E as a function of X_1 for the binary system dodecylbenzenesulfonic acid-acetone for the temperature as following (♦293.15, ■298.15, ▲303.15) $^{\circ}\text{K}$.

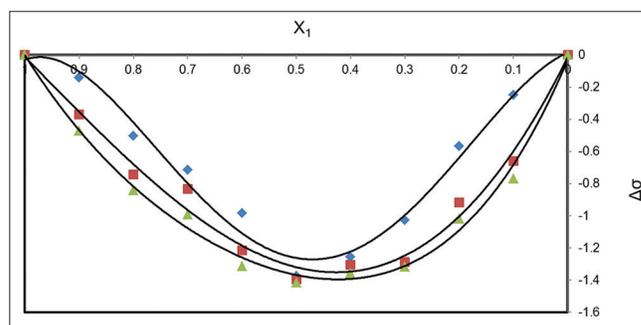


Fig. 3. Surface tension deviation $\Delta\sigma$ as a function of X_1 for the binary system dodecylbenzenesulfonic acid-acetone for the temperature as following (♦293.15, ■298.15, ▲303.15) $^{\circ}\text{K}$.

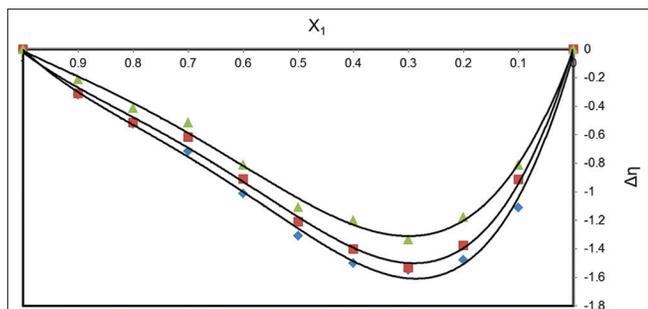


Fig. 2. Viscosity deviation $\Delta\eta$ as a function of X_1 for the binary system dodecylbenzenesulfonic acid-acetone for the temperature as following (♦293.15, ■298.15, ▲303.15) $^{\circ}\text{K}$.

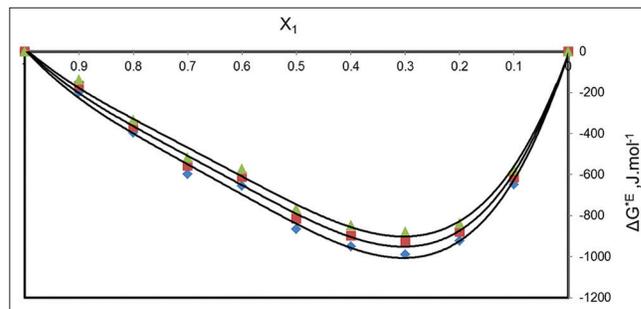


Fig. 4. Excess molar Gibbs free energy of activation ΔG^{*E} as a function of X_1 for the binary system dodecylbenzenesulfonic acid-acetone for the temperature as following (♦293.15, ■298.15, ▲303.15) $^{\circ}\text{K}$.

B. FTIR and Intermolecular Interaction

For the investigation of functional group, molecular structure, inter- and intra-molecular interaction, FTIR play a crucial role (Wei, et al., 2013). The binary mixture of DBSA-acetone system showed a deviation from the ideality and was explained in the previous section that the nonideality and the deviation in the thermodynamic properties was explained due to the intermolecular interaction in hydrogen bonding. Two types of hydrogen bonding can be concluded from the broadening and sharpening of the hydrogen beaks, broadening referred to intermolecular bonding and sharpening referred to intramolecular bonding. The intensity of hydrogen beaks due

to the intermolecular bonding was decreased with increasing the dilution until totally it disappears. The FTIR spectrum for the pure DBSA showed S=O stretch at 1345 cm^{-1} , broad O-H stretch at 2900 cm^{-1} , =C-H stretch (aromatic) in benzene ring at 3100 cm^{-1} , 3070 cm^{-1} , and 3050 cm^{-1} , C-H stretch (alkyl) at 2950 cm^{-1} , C-C stretch in ring (aromatic) at 1590 cm^{-1} , 1478 cm^{-1} , and 1400 cm^{-1} due to vibration in the benzene ring, C-H in-plane bending at $1100, 1050\text{ cm}^{-1}$ (weak band), C-H out-of-plane bending at 750 cm^{-1} , Overtones bands in the region $2000-1665\text{ cm}^{-1}$, C=C stretch at 1644 cm^{-1} .

The FTIR spectrum for the pure Acetone showed C=O at 1715 cm^{-1} , CH_3 asymmetric stretch at 3014 cm^{-1} , CH_3

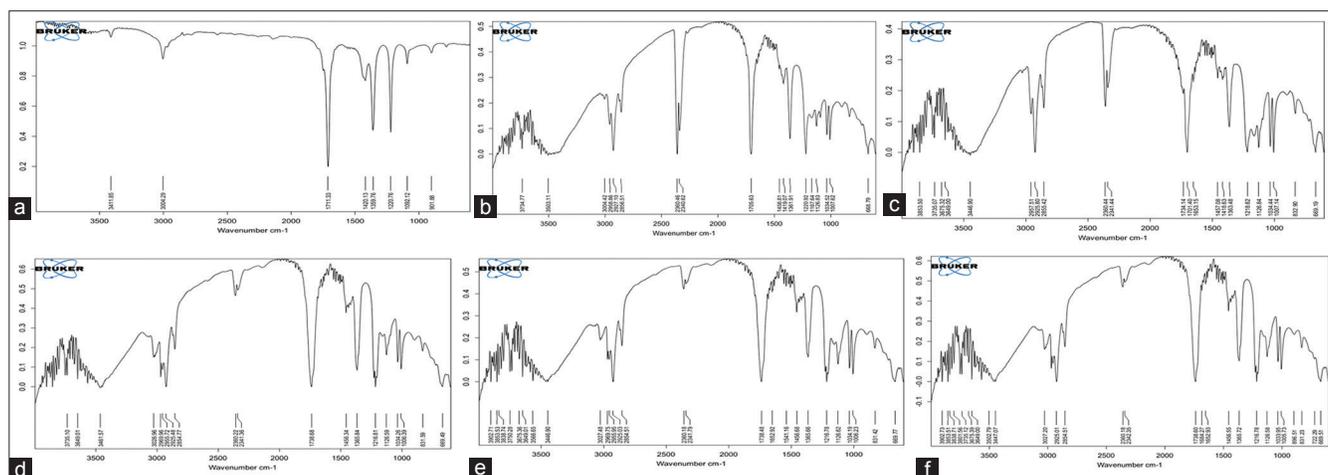


Fig. 5. Fourier transform infrared spectrum for the (a) pure acetone, dodecylbenzenesulfonic acid (DBSA)-acetone mixture of (b) $X_1 = 0.2$, (c) $X_1 = 0.4$, (d) $X_1 = 0.6$, (e) $X_1 = 0.8$, (f) pure DBSA at (298.15°K and P=1 atm)

symmetric stretch at 2971 cm^{-1} , C-C at 1218 cm^{-1} . Clear broad O-H beak at 2900 cm^{-1} in the pure DBSA is the marked sign of it. With increase the dilution of DBSA, it is clear from the Fig. 5 that the OH stretching vibration beak is shifted to a higher wavenumber in the mixtures due to the intermolecular interaction in hydrogen bonding. Moreover, for pure acetone it has been recognized a strong C=O stretch beak at 1715 cm^{-1} . After mixing with DBSA there is a slightly shifting in the C=O stretch beak to a higher wave number. The FTIR characterization for the binary mixture of DBSA-acetone showed a weakness in all the present hydrogen bond in the mixture means that the intermolecular interaction between (O-H) of DBSA and (C=O) of acetone are weak (Hasan, et al., 2011). In the pure case of DBSA and acetone, the intramolecular interaction due to the dispersion is stronger than in the case of their mixtures due to the self-association of DBSA or decreasing the dipole-dipole of the acetone molecules. The result of FTIR supported the result of thermodynamic excess where the excess molar volumes were positive.

V. CONCLUSION

The experimental values of density, viscosity, and surface tension for the pure case of DBSA and acetone liquids at the temperatures (293.15°K, 298.15°K, 301.15°K) and atmospheric pressure are presented in this paper. Excess molar volume, viscosity deviation, surface tension deviation, and molar excess Gibbs free energy deviation are calculated at the above-mentioned temperatures and correlated to the RK equation. FTIR spectrum of the DBSA-acetone mixture shows a weak intermolecular hydrogen bonding interaction between the molecules of DBSA and acetone. The result of molar excess Gibbs free energy deviation supports the above conclusion of FTIR where the excess molar volume is positive. The intramolecular interaction is strong in the case of pure DBSA and acetone due to the self-association of DBSA molecule or decreasing the dipole-dipole of acetone molecule. The calculated values of excess molar volume are

positive for all the temperatures ranging from 293.15°K to 301.15°K. while, the values of viscosity deviation, surface tension deviation, and molar excess Gibbs free energy deviation are negative for all the temperatures studied.

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