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Surface Tension of *N*-Methyldiethanolamine in Methanol or in Methanol Aqueous Solutions as a Solvent at Temperatures from 293.15 to 323.15 K

Estrella Álvarez,* Fernando Cerdeira, José Manuel Navaza, and Francisco Javier Tamajón

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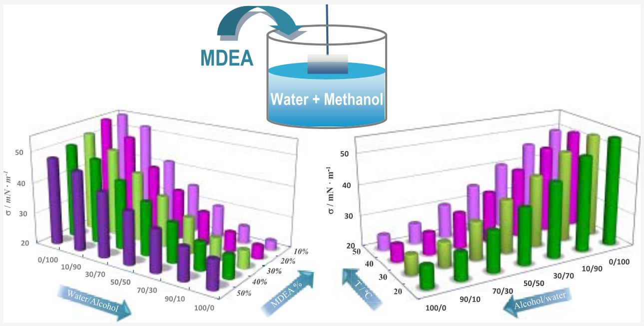
ABSTRACT: The surface tension of *N*-methyldiethanolamine in methanol, or in methanol aqueous solutions as a solvent, was measured at temperatures from 293.15 to 323.15 K. In ternary mixtures, five methanol + water blends with concentrations between 10 and 90% methanol were used as solvents, and the amine concentration varied from 0 to 50% mass, while in the methanol + *N*-methyldiethanolamine (MDEA) system, the amine concentration varied between 0 and 100% mass, at intervals of 10%. Experimental data were correlated with temperature and concentration by means of the Jasper equation, and the Jouyban−Acree and FLW models, respectively.



■ INTRODUCTION

Absorption processes are a reliable and well-known technology in the chemical industry, where there has been a widely remarkable experience in commercial CO_2 capture applications using alkanolamine solutions since many decades.[1] Nevertheless, the high energy demand required for the solvent regeneration has a negative effect on the operating costs of the process, and therefore, it is necessary to do further research on solvents that may have more advantages, processing larger quantities of CO_2 with lower energy requirements in the regeneration stage.[2,3]

Nowadays, the use of *N*-methyldiethanolamine (MDEA) aqueous solutions in CO_2 absorption is gaining importance due to its high performance.[4,5] Several advantages of MDEA with respect to other primary and secondary amines (low vapor pressure, low reaction heat with acid gases, high resistance to thermal and chemical degradation, low corrosiveness) will result in savings in operational costs.[6,7] Also, attention has been paid to the possibility of combining alkanolamines in mixtures of aqueous and nonaqueous solvents by several research groups.[8−11] The addition of an organic compound (physical solvent) such as methanol to a tertiary amine (chemical solvent) in a CO_2 absorption process may lead to favorable effects on the mass transfer.[12−17]

The physical and chemical properties of the liquid phase such as density, viscosity, and surface tension have a significant influence on the gas−liquid mass-transfer processes. Therefore, these properties are essential for the design, optimization, and control of gas treatment processes. The complete characterization of the physicochemical behavior of new solvents to improve the thermodynamic models requires accurate data available on the relevant chemical and physical properties. In the literature, references reporting on physicochemical properties in organic-aqueous solutions of *N*-methyldiethanolamine and methanol are limited[18−20] or even not available.

Due to the limited bibliographic data, in the present work, the surface tension of different *N*-methyldiethanolamine mixtures in organic-aqueous solvents was measured, in a wide range of concentrations and temperatures. Concentrations and temperatures were selected according to their applicability in CO_2 removal processes. For this reason, the MDEA % varied between 0 and 50% mass, methanol % in the solvent varied between 10 and 90% mass, in 20% steps, and the temperature varied from 293.15 to 323.15 K.

2. EXPERIMENTAL SECTION

2.1. Materials. All blends were prepared from pure components, by weight, using a Scaltec SBA31 balance with a precision of $\pm 10^{-4}$ g. Pure solutes are commercial products supplied by Sigma-Aldrich and were used without any further purification. Specifications for all reagents used in this study are shown in Table 1. First, we prepared five aqueous mixtures of methanol by varying the percent of methanol between 10 and 90% mass, in steps of 20% mass. These mixtures were

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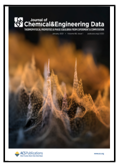

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Table 1. Sample Description

chemical name	CAS number	source[a]	purification method	purity
methanol	67-56-1	Sigma-Aldrich	none	>99.8[b]
N-methyldiethanolamine	105-59-9	Sigma-Aldrich	none	>99[b]
water	7732-18-5	UVIGO's central research support services	distillation, Milli-Q purification	electrical resistivity = 18.2 MΩ·cm at T = 298.15 K

[a] UVIGO refers to the University of Vigo. [b] Purity given in mole fraction, as stated by the supplier.

Table 2. Experimental Values of Surface Tension σ, at Temperature T, Mole Fraction x, and Ambient Pressure for the Liquid Mixture Methanol (1) + *N*-Methyldiethanolamine (2)[a]

	σ/mN·m^{-1}						
x_1	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
0.0000	39.26[c]	38.90[c]	38.41[c]	37.92[c]	37.51[c]	37.16[c]	36.88[c]
0.2923	38.35	38.00	37.56	37.14	36.75	36.40	36.08
0.4817	36.51	36.17	35.81	35.40	35.05	34.69	34.37
0.6147	34.41	34.09	33.75	33.41	33.04	32.71	32.38
0.7126	32.37	32.03	31.72	31.39	31.02	30.69	30.34
0.7878	30.46	30.11	29.78	29.45	29.08	28.71	28.38
0.8479	28.70	28.32	27.97	27.62	27.26	26.86	26.49
0.8965	27.06	26.67	26.28	25.92	25.54	25.13	24.73
0.9369	25.56	25.16	24.76	24.34	23.95	23.51	23.10
0.9709	24.20	23.77	23.33	22.87	22.48	22.00	21.60
1.0000	22.95[b]	22.51[b]	22.01[b]	21.52[b]	21.13[b]	20.61[b]	20.21[b]

[a] Standard uncertainties u are $u(T)$ = 0.01 K, $u(x)$ = 0.0002, and $u(p)$ = 2 kPa. Expanded uncertainty for the surface tension $U(\sigma)$ = 0.11 mN·m^{-1} (0.95 level of confidence). [b] Surface tensions determined in our previous work: Ref 21. [c] Surface tensions determined in our previous work: Ref 22.

prepared with water from a MILLI-Q Advantage A10 purification system (electrical resistivity = 18.2 MΩ·cm at T = 298.15 K). Subsequently, each of these solutions was used as a solvent in the preparation of the ternary mixtures, in which the MDEA % is varied between 0 and 50% mass. For each amine concentration, we have prepared five mixes that correspond to the different methanol/water ratios in the solvent. In addition, we also prepared nine *N*-methyldiethanolamine + methanol solutions in which the mass % of MDEA is varied between 0 and 100%, at constant intervals of 10%.

2.2. Methods. The surface tension of pure components was determined in previous papers[21,22] using a prolabo tensiometer, which employs the Wilhelmy plate method, while the surface tension of mixtures and pure MDEA at 283.15 K was measured with a Krüs K-11 tensiometer, which also employs the Wilhelmy plate method. The experimental measure procedure has been described in a previous paper;[23,24] therefore, now, only specific information for this work is provided.

For each sample, the surface tension was measured at seven different temperatures between 293.15 and 323.15 K, with an uncertainty of ±0.05 mN·m^{-1}. Before each measurement, the sample was thermostated in a closed vessel to prevent evaporation, and the temperature was controlled by a thermostat−cryostat bath, with a precision of ±0.01 K. Finally, each value reported is an average of 10 consecutive measurements.

The surface tensions of pure methanol and MDEA at working temperatures are compared with the values obtained by other authors.[25−32] In addition, the average relative deviation (RD) between our data and those available in the literature is calculated, resulting less than 1.5%.

3. RESULTS AND DISCUSSION

Experimental surface tensions of methanol + *N*-methyldiethanolamine binary mixtures and ternary mixtures of MDEA in aqueous solutions of methanol, at the different temperatures tested, are reported in Tables 2 and 3, respectively. In Table 3, the surface tensions corresponding to the water/alcohol ratio of 100/0, i.e., to aqueous solutions of *N*-methyldiethanolamine, have been determined in previous work.[22] Surface tensions of MDEA aqueous solutions were compared with the values found in the literature,[32−35] and the average relative deviation (RD) was calculated. As shown in Figure 1, the RD values are always less than 2%.

On analyzing the experimental data, it is observed that, in all of the studied solutions, the surface tension decreases with increasing temperature. As an example of this behavior, Figure 2 shows the surface tension versus temperature, both for the MDEA + methanol binary mixtures (internal plot) and the ternary mixture with a methanol/water ratio of 50/50 (main plot). In both cases, the surface tension varies linearly with temperature, this behavior being similar to the one observed for other systems.[36,37]

For this reason, the experimental data have been correlated with temperature using the equation developed by Jasper.[38]

$$\sigma_m/\text{mN}\cdot\text{m}^{-1} = K_1 - K_2 \cdot T/\text{K} \tag{1}$$

where σ_m represents the surface tension of the mixture, T is the temperature, and K_1 and K_2 are two fitted parameters that vary with the composition of the mixture. The values of both parameters are listed in Tables 4 and 5, with the standard deviation (σ_{st}) between experimental and calculated values, for blends of MDEA with methanol or aqueous solutions of methanol, respectively.

In relation to the variation of surface tension with the concentration, it was noticed that for the *N*-methyldiethanol-

Table 3. Experimental Surface Tension σ, at Temperature T, Mole Fraction x, and Ambient Pressure for the Liquid Mixtures Water (1) + Methanol (2) + *N*-Methyldiethanolamine (3)[a]

			σ/mN·m^{-1}						
solvent (water/methanol)	x_1	x_2	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
			Total Amine Concentration = 10% mass						
100/0	0.9835	0.0000	62.52[b]	61.82[b]	61.11[b]	60.23[b]	59.41[b]	58.62[b]	58.00[b]
90/10	0.9248	0.0579	54.13	53.41	52.89	52.27	51.67	51.05	50.40
70/30	0.7905	0.1905	43.25	42.76	42.28	41.79	41.30	40.84	40.34
50/50	0.6268	0.3520	36.65	36.14	35.72	35.27	34.84	34.45	34.07
30/70	0.4235	0.5524	31.32	30.87	30.47	30.11	29.64	29.26	28.88
10/90	0.1620	0.8110	26.79	26.32	25.88	25.48	25.06	24.68	24.32
0/100	0.0000	0.9709	24.20	23.77	23.33	22.87	22.48	22.00	21.60
			Total Amine Concentration = 20% mass						
100/0	0.9636	0.0000	58.53[b]	57.74[b]	56.96[b]	56.10[b]	55.30[b]	54.54[b]	53.73[b]
90/10	0.9053	0.0567	52.13	51.55	50.97	50.35	49.76	49.20	48.56
70/30	0.7725	0.1858	43.36	42.88	42.42	41.94	41.45	41.01	40.57
50/50	0.6124	0.3417	37.21	36.78	36.35	35.94	35.54	35.11	34.75
30/70	0.4109	0.5372	32.03	31.62	31.22	30.82	30.42	30.02	29.63
10/90	0.1562	0.7850	27.53	27.12	26.73	26.35	25.96	25.57	25.17
0/100	0.0000	0.9369	25.56	25.16	24.76	24.34	23.95	23.51	23.10
			Total Amine Concentration = 30% mass						
100/0	0.9392	0.0000	54.44[b]	53.63[b]	52.84[b]	52.05[b]	51.33[b]	50.52[b]	49.74[b]
90/10	0.8759	0.0611	50.22	49.53	48.81	48.14	47.46	46.79	46.16
70/30	0.7500	0.1806	43.94	43.37	42.84	42.27	41.81	41.26	40.76
50/50	0.5914	0.3321	37.95	37.47	37.02	36.59	36.17	35.73	35.34
30/70	0.3960	0.5187	33.01	32.61	32.18	31.82	31.42	31.02	30.65
10/90	0.1499	0.7536	28.67	28.28	27.92	27.52	27.11	26.75	26.37
10/100	0.0000	0.8967	27.06	26.67	26.28	25.92	25.54	25.13	24.73
			Total Amine Concentration = 40% mass						
100/0	0.9085	0.0000	52.45[b]	51.65[b]	50.82[b]	50.11[b]	49.35[b]	48.59[b]	47.80[b]
90/10	0.8515	0.0532	49.30	48.62	47.90	47.23	46.55	45.89	45.17
70/30	0.7500	0.1737	43.80	43.28	42.69	42.09	41.54	40.98	40.42
50/50	0.5670	0.3185	38.96	38.44	37.85	37.38	36.86	36.34	35.85
30/70	0.3783	0.4949	34.52	34.00	33.52	33.01	32.56	32.12	31.70
10/90	0.1425	0.7150	30.53	30.08	29.63	29.19	28.74	28.32	27.96
0/100	0.0000	0.8480	28.70	28.32	27.97	27.62	27.26	26.86	26.49
			Total Amine Concentration = 50% mass						
100/0	0.8686	0.0000	50.41[b]	49.59[b]	48.86[b]	48.04[b]	47.32[b]	46.50[b]	45.70[b]
90/10	0.8122	0.0514	47.67	47.00	46.31	45.66	44.97	44.34	43.66
70/30	0.6861	0.1656	42.79	42.20	41.62	41.10	40.58	40.09	39.56
50/50	0.5370	0.3010	38.62	38.11	37.65	37.18	36.70	36.28	35.83
30/70	0.3626	0.4604	35.08	34.65	34.24	33.78	33.38	33.00	32.62
10/90	0.1324	0.6681	31.87	31.43	31.11	30.76	30.38	30.00	29.68
0/100	0.0000	0.7881	30.46	30.11	29.78	29.45	29.08	28.71	28.38

[a] Standard uncertainties u are $u(T) = 0.01$ K, $u(x) = 0.0002$, and $u(p) = 2$ kPa. Expanded uncertainty for the surface tension $U(\sigma) = 0.11$ mN·m^{-1} (0.95 level of confidence). [b] Surface tensions determined in Ref 22.

amine + methanol mixtures, the surface tension increases nonlinearly when the alkanolamine percentage increases (see Figure 3). This behavior is similar to that observed in previous work for aqueous solutions of MDEA[22] or solutions of MDEA with ethanol.[39] For this reason, first, we have correlated the experimental data by eq 2, which we have previously used for this type of system

$$\frac{\sigma_m - \sigma_1}{\sigma_2 - \sigma_1} = X_2\left(1 + \frac{aX_1}{1 - bX_1}\right) \tag{2}$$

In this equation, $\mathbf{x_1}$ and $\mathbf{x_2}$ are, respectively, the mole fractions of the solvent (methanol) and solute (MDEA), and $\boldsymbol{\sigma_m}$, $\boldsymbol{\sigma_1}$, and $\boldsymbol{\sigma_2}$ represent the surface tensions of the mixture, pure solvent, and pure solute, respectively. Finally, **a** and **b** are two temperature-dependent adjustment parameters whose values are shown in Table 6, along with the standard deviation (σ_{st}).

Second, experimental data has also been correlated using the FLW[40] and Jouyban−Acree[41] (JAM) models, which are applicable to binary and ternary systems. The first model has recently been used to correlate the surface tension of alkanolamine + alcohol binary mixtures,[28] and ternary mixtures containing monoethanolamine, water, and alcohols,[42] with satisfactory results in both cases, while the Jouyban−Acree (JAM) model is widely used by several researchers to correlate the surface tension,[41] density,[43] viscosity,[44] or solubility[45] of binary mixtures with respect to composition. Furthermore, several of these authors have found that this model, applied to aqueous binary solutions, predicts more accurately the

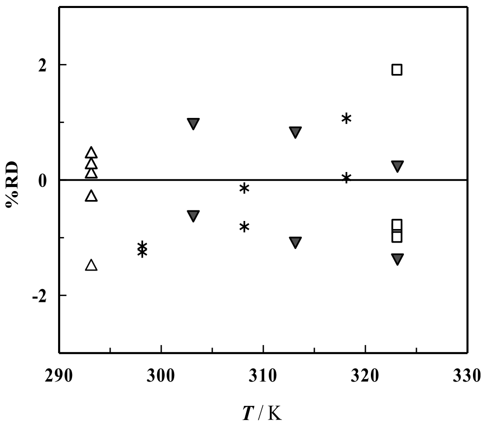

Figure 1. Relative percentage error (% RD) between our surface tensions and the literature, for MDEA aqueous solutions with a concentration between 10 and 50% mass: ▼, ref 32; △, ref 33; □, ref 34; and ∗, ref 35.

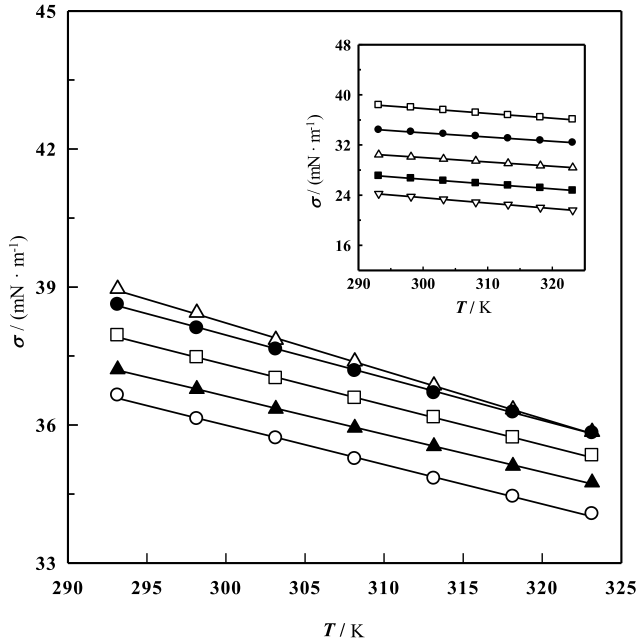

Figure 2. Temperature dependence of surface tensions σ of MDEA mixtures with methanol aqueous solutions and MDEA with methanol (inset plot). Main plot: Solvent with 50% methanol/50% water and ○, 10% MDEA; ▲, 20% MDEA; □, 30% MDEA; △, 40% MDEA; and ●, 50% MDEA. Inset plot: □, 10% MDEA; ●, 30% MDEA; △, 50% MDEA; ■, 70% MDEA; and ▽, 90% MDEA. In both plots, lines represent values calculated by eq 1.

experimental surface tensions than other existing models, such as Lee's model.

For binary mixtures, the general equation of the FLW model for multicomponent mixtures

$$\sigma_m = \sum_{i=1}^{n} \frac{X_i\sigma_i}{\sum_{j=1}^{n} x_j f_{ij}} - \sum_{i=1}^{n}\sum_{j=1}^{n} \frac{x_i x_j(\sigma_i - \sigma_j)}{\sum_{q=1}^{n} x_q f_{iq}\,\sum_{r=1}^{n} x_r f_{ir}} \qquad (3)$$

is reduced to

$$\sigma_m = \frac{x_1\cdot\sigma_2}{x_1 + x_2\cdot f_{12}} + \frac{x_2\cdot\sigma_2}{x_2 + x_1\cdot f_{21}} + \frac{x_1x_2(\sigma_1 - \sigma_2)}{(x_1 + x_2\cdot f_{12})(x_2 + x_1\cdot f_{21})} \qquad (4)$$

where $\mathbf{f_{12}}$ and $\mathbf{f_{21}}$ are the model fit parameters.

On the other hand, the Jouyban–Acree model represents the surface tension of binary mixtures as

$$\ln \sigma_m = x_1\cdot\ln \sigma_1 + x_2\cdot\ln \sigma_2 + x_1\cdot x_2 \sum_{i=0}^{2} B_i(x_1 - x_2)^i \qquad (5)$$

where $\mathbf{B_i}$ is the model constant. In eqs 4 and 5, $\boldsymbol{\sigma_m}$ represents the surface tension of the binary mixture, $\boldsymbol{\sigma_i}$ represents the surface tension of the pure ith component, and $\mathbf{x_i}$ is the mole fraction of component i in the mixture.

Applying both models to our system, the results shown in Table 6 were obtained. From these, it is deduced that the FLW model reproduces the experimental values better than the Jouyban–Acree model and that both models give worse results than eq 2. As an example, Figure 4 shows the relative differences $\Delta\sigma/\sigma = (\sigma_{exp} - \sigma_{cal})/\sigma_{cal}$ between experimental data and those calculated from the three models, noting that their values are significantly lower for eq 2 (Figure 4a) than for the JAM and FLW models (Figure 4b,c).

For ternary mixtures, first, it is noticed that for a given concentration of amine, the surface tension decreases as the methanol % in the solvent increases (see Figure 5). This behavior agrees with the one observed by other authors,[20] for methanol mass fractions between 0 and 0.15 and amine contents of 20–40% mass. Second, it is observed that for methanol/water ratios lower than 30/70, the surface tension decreases with MDEA %, while at ratios higher than 50/50, it decreases with increasing MDEA content. For the methanol/water ratios 30/70 and 50/50, the surface tension initially increases and later decreases with the % MDEA. This change in the trend occurs for 40 and 50% MDEA values, respectively (see Figure 6), and is attributable to two factors: first, at low methanol/water ratios, $\sigma_{solvent} < \sigma_{solute}$ while for ratios greater than 30/70, the solvent surface tension is less than that of the

Table 4. Adjustable Parameters K_1 and K_2 (in eq 1) with the Standard Deviations, σ_{st}, for the Liquid Mixture Methanol (1) + *N*-Methyldiethanolamine (2)[a]

x_1	K_1/mN·m^{-1}	K_2/mN·m^{-1}·K^{-1}	σ_{st}	x_1	K_1/mN·m^{-1}	K_2/mN·m^{-1}·K^{-1}	σ_{st}
0.0000	63.362	0.0823	0.081	0.8479	50.186	0.0733	0.017
0,2923	60.999	0.0773	0.044	0.8965	49.698	0.0772	0.016
0.4817	57.738	0.0724	0.023	0.9369	49.630	0.0821	0.016
0.6147	54.441	0.0683	0.014	0.9709	49.724	0.0871	0.021
0.7126	52.210	0.0676	0.022	1.0000	49.957	0.0921	0.036
0.7878	50.868	0.0696	0.016				

[a] $\sigma_{st} = \left[\sum(\sigma_{cal} - \sigma_{exp})^2/(N - n)\right]^{1/2}$ where σ represents the surface tension, N is the number of data, and n is the number of parameters.

Table 5. Adjustable Parameters K_1 and K_2 (in eq 1) with the Standard Deviations, σ_{st}, for the Ternary Mixtures of N-Methyldiethanolamine in Water (1) + Methanol (2) Solutions[a]

solvent (water/methanol)	x_1	x_2	K_1/mN·m^{-1}	K_2/mN·m^{-1}·K^{-1}	σ_{st}
Total Amine Concentration = 10% mass					
100/0	0.9835	0.0000	107.9195	0.1547	0.074
90/10	0.9248	0.0579	91.8433	0.1269	0.015
70/30	0.7905	0.1905	71.5713	0.0966	0.006
50/50	0.6268	0.3520	61.7058	0.0857	0.042
30/70	0. 4235	0.5524	55.0870	0.0812	0.032
10/90	0.1620	0.8110	50.8480	0.0822	0.044
0/100	0.0000	0.9709	49.6373	0.0868	0.019
Total Amine Concentration = 20% mass					
100/0	0.9636	0.0000	105.5643	0.1604	0.026
90/10	0.9053	0.0567	86.9055	0.1186	0.019
70/30	0.7725	0.1858	70.7082	0.0933	0.023
50/50	0.6124	0.3417	61.3437	0.0824	0.022
30/70	0.4109	0.5372	55.5001	0.0801	0.007
10/90	0.1562	0.7850	50.4702	0.0783	0.010
0/100	0.0000	0.9369	49.4519	0.0815	0.015
Total Amine Concentration = 30% mass					
100/0	0.9392	0.0000	100.1280	0.1559	0.023
90/10	0.8759	0.0611	89.9665	0.1357	0.025
70/30	0.7500	0.1806	74.8783	0.1057	0.035
50/50	0.5914	0.3321	63.4402	0.0871	0.030
30/70	0.3960	0.5187	56.0351	0.0786	0.016
10/90	0.1499	0.7536	51.1787	0.0768	0.014
0/100	0.0000	0.8967	49.6553	0.0771	0.017
Total Amine Concentration = 40% mass					
100/0	0.9085	0.0000	97.5211	0.1539	0.035
90/10	0.8515	0.0532	89.5204	0.1372	0.018
70/30	0.7500	0.1737	77.1136	0.1136	0.021
50/50	0.5670	0.3185	69.3538	0.1038	0.033
30/70	0.3783	0.4949	62.0499	0.0941	0.043
10/90	0.1425	0.7150	55.9051	0.0866	0.035
0/100	0.0000	0.8480	50.3485	0.0738	0.018
Total Amine Concentration = 50% mass					
100/0	0.8686	0.0000	96.1534	0.1561	0.029
90/10	0.8122	0.0514	86.8584	0.1337	0.015
70/30	0.6861	0.1656	74.0627	0.1069	0.044
50/50	0.5370	0.3010	65.7972	0.0928	0.025
30/70	0.3626	0.4604	59.1852	0.0823	0.030
10/90	0.1324	0.6681	53.1532	0.0727	0.028
0/100	0.0000	0.7881	51.1018	0.0704	0.026

[a] $\sigma_{st} = \left[\sum (\sigma_{cal} - \sigma_{exp})^2/(N - n)\right]^{1/2}$ where σ represents the surface tension, N is the number of data, and n is the number of parameters.

solute. Consequently, in the water-rich region, the MDEA addition causes a decrease in surface tension, while in the water-poor region, it increases. Second, the MDEA effect is very strong at low MDEA concentrations (<40–50% mass) and less pronounced at high concentrations. The combination of these two factors results in a transition between water-rich and water-poor behavior, at concentrations around 50% MDEA.

Now, we have correlated the experimental data using the Jouyban–Acree model, which, for ternary systems, takes the form

$$\ln \sigma_m = x_1 \ln \sigma_1 + x_2 \ln \sigma_2 + x_3 \ln \sigma_3 + x_1x_2 \sum_{j=0}^{2} A_j (x_1 - x_2)^j + x_1x_3 \sum_{j=0}^{2} B_j (x_1 - x_3)^j + x_2x_3 \sum_{j=0}^{2} C_j (x_2 - x_3)^j + x_1x_2x_3 \sum_{j=0}^{2} D_j (x_1 - x_2 - x_3)^j \quad (6)$$

where $\mathbf{x_i}$ represents the molar fraction of component i in the mixture, $\boldsymbol{\sigma_i}$ represents the surface tension of the ith pure component, and $\boldsymbol{\sigma_m}$ is the mixture surface tension. In eq 6, A_j, B_j, and C_j represent the binary correlation parameters, and D_j are the ternary correlation parameters whose values are

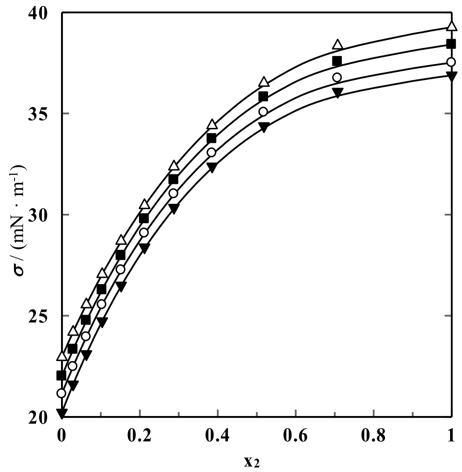

Figure 3. Variation of the surface tension σ with the concentration for binary mixtures of methanol (1) + *N*-methyldiethanolamine (2): △, 293.15 K; ■, 303.15 K; ○, 313.15 K; and ▼, 323.15 K. Lines represent values calculated by eq 2.

reported in Table 7. A_j and C_j parameters for water + methanol and water + *N*-methyldiethanolamine systems, respectively, were determined from experimental data obtained in previous work,[21,22] and their values are presented in Table 8, while B_j values for the methanol + *N*-methyldiethanolamine system are presented in Table 6.

The values calculated based on the JAM model are summarized in Table S1 of the Supporting Information. In this table, we also provide the relative percent deviations (% RD) between the experimental and calculated values. Examination of the numerical entries reveals that the deviation between the experimental and calculated values varies from 0.1 to 6.5%, with the highest values for mixtures with a water/methanol ratio greater than 70/30, i.e., in mixtures with a high difference between the surface tension of the solute and solvent (see Figure 4). In any case, from the % RD obtained, we can conclude that the JAM model equation reliably reproduces our experimental results.

On the other hand, the FLW model was used to predict the surface tension of all of the ternary blends studied. In this case, the general equation of the model is reduced to

$$\begin{aligned}\sigma_m = {} & \frac{x_1\cdot\sigma_2}{x_1 + x_2\cdot f_{12} + x_3\cdot f_{13}} + \frac{x_2\cdot\sigma_2}{x_2 + x_1\cdot f_{21} + x_3\cdot f_{23}} \\ & + \frac{x_3\cdot\sigma_3}{x_3 + x_1\cdot f_{31} + x_2\cdot f_{32}} \\ & - \frac{x_1x_2(\sigma_1 - \sigma_2)}{(x_1 + x_2\cdot f_{12} + x_3\cdot f_{13})(x_2 + x_1\cdot f_{21} + x_3\cdot f_{23})} \\ & - \frac{x_1x_3(\sigma_1 - \sigma_3)}{(x_1 + x_2\cdot f_{12} + x_3\cdot f_{13})(x_3 + x_1\cdot f_{31} + x_2\cdot f_{32})} \\ & - \frac{x_1x_3(\sigma_1 - \sigma_3)}{(x_2 + x_1\cdot f_{21} + x_3\cdot f_{23})(x_3 + x_1\cdot f_{31} + x_2\cdot f_{32})}\end{aligned} \tag{7}$$

where $\boldsymbol{\sigma}_{\mathbf{m}}$ is the surface tension of the mixture, $\mathbf{x}_{\mathbf{i}}$ represents the mole fraction of the *i*th pure component, and $\mathbf{f}_{\mathbf{ij}}$ and $\mathbf{f}_{\mathbf{ji}}$ are the fitting parameters for binary systems, i.e., for water (1) + methanol (2), water (1) + MDEA (3), and methanol (2) + MDEA (3) systems. In this equation, the f_{12}, f_{21}, f_{13}, and f_{31} values were determined by applying the FLW model to the experimental data obtained in previous work,[21,22] and their values are shown in Table 8, while f_{ij} and f_{ji} for the methanol + *N*-methyldiethanolamine system are shown in Table 6.

The values predicted by the FLW model are shown in Table S2 of the Supporting Information. It also includes the relative percentage error values (% RD) between the experimental values, σ_{exp}, and those predicted by the FLW model, σ_{cal}, that are calculated as

$$\%\,\text{RD} = \left|\frac{\sigma_{\text{exp}} - \sigma_{\text{cal}}}{\sigma_{\text{exp}}}\right| \times 100 \tag{8}$$

The results of Table S2 show that the highest values of % RD are obtained for water/methanol ratios between 30/70 and 70/30, reaching the maximum values when the solvent consists of 50% water and 50% methanol, while for ratios below 30/70 or above 70/30, the FLW model successfully reproduces the experimental values. That is, the model predicts worse the experimental data for the systems in which a trend change occurs in the surface tension variation as the amine percentage increases.

To illustrate this behavior, Figure 7 shows the relative percentage deviations between the experimental data and those predicted by the FLW model, noting that their values are

Table 6. Adjustable Parameters *a* and *b* (in eq 2), f_{12} and f_{21} (in eq 4), and B_i (in eq 5) with the Standard Deviations, σ_{st}, for the Methanol (1) + *N*-Methyldiethanolamine (2) Binary Mixtures[a]

	T/K						
	293.15	298.15	303.15	308.15	313.15	318.15	323.15
a	1.0034	1.0055	1.0099	1.0136	1.0152	1.0164	1.0176
b	0.4123	0.4214	0.4522	0.4785	0.4822	0.4940	0.4892
σ_{st}	0.012	0.006	0.010	0.009	0.013	0.008	0.014
f_{12}	1.3629	1.3742	1.4204	1.4654	1.4596	1.4961	1.5004
f_{21}	0.7386	0.7318	0.7070	0.6847	0.6840	0.6701	0.6704
σ_{st}	0.018	0.013	0.015	0.018	0.016	0.016	0.009
B_0	0.7474	0.7685	0.7964	0.8215	0.8367	0.8661	0.8834
B_1	0.3290	0.3507	0.3821	0.4066	0.4303	0.4577	0.4780
B_2	0.2420	0.2636	0.2898	0.3175	0.3347	0.3659	0.3813
σ_{st}	0.047	0.050	0.057	0.060	0.062	0.064	0.065

[a] $\sigma_{st} = \left[\sum(\sigma_{\text{cal}} - \sigma_{\text{exp}})^2/(N - n)\right]^{1/2}$ where σ represents the surface tension, N is the number of data, and n is the number of parameters.

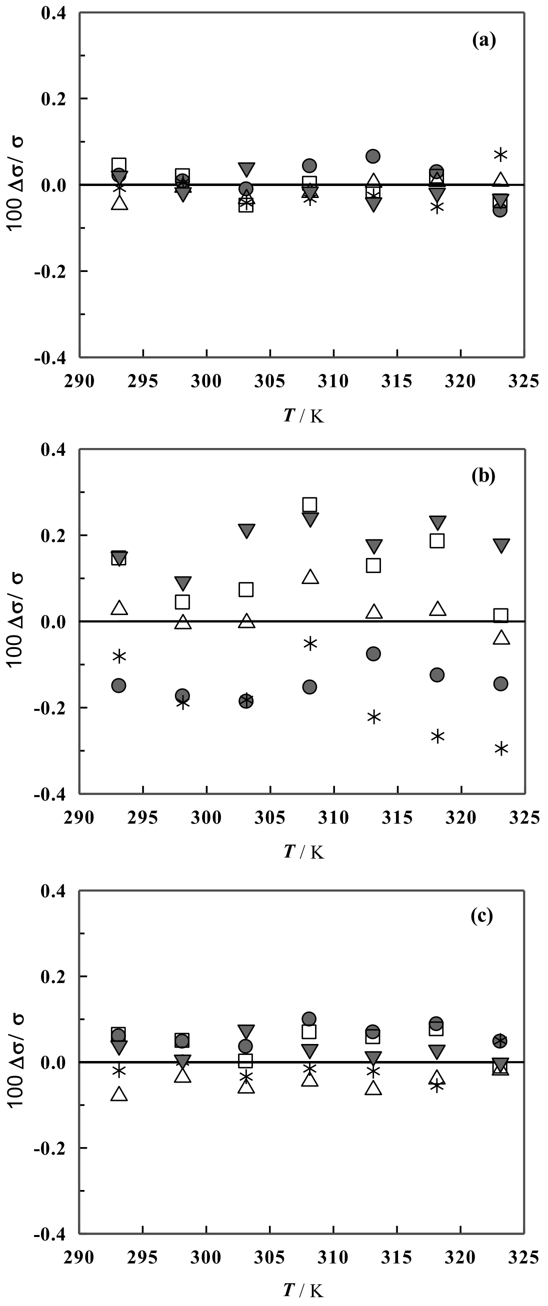

Figure 4. Relative difference $\Delta\sigma/\sigma$ of the experimental surface tensions, from the values calculated from (a) eq 2, (b) the Jouyban–Acree model, and (c) the LFW model, at different temperatures. Methanol + *N*-methyldiethanolamine binary system: ●, 10% mass MDEA; △, 30% mass MDEA; ∗, 50% mass MDEA; ☐, 70% mass MDEA; and ▼, 90% mass MDEA.

always higher for a methanol/water ratio in the solvent of 50% methanol/50% water. A higher percentage deviation is also observed for systems with a lower % MDEA, regardless of the methanol/water ratio. In any case, the effect of temperature is not significant.

Finally, surface thermodynamic properties, such as surface entropy and enthalpy, were calculated from the temperature dependence of the mixture's surface tension at a constant concentration, using

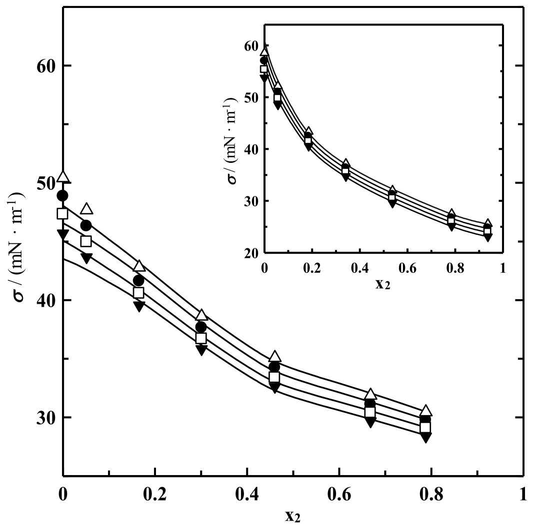

Figure 5. Variation of the surface tension, σ, with the concentration for ternary mixtures of MDEA in methanol aqueous solutions. Main plot: 20% MDEA and △, 293.15 K; ●, 303.15 K; ☐, 313.15 K; and ▼, 323.15 K. Inset plot: 50% MDEA and △, 293.15 K; ●, 303.15 K; ☐, 313.15 K; and ▼, 323.15 K. In both plots, lines represent values calculated by eq 6.

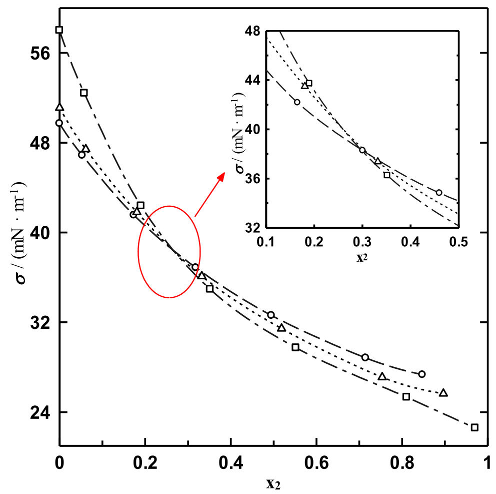

Figure 6. Variation of the surface tension, σ, with the MDEA % for ternary mixtures of MDEA in methanol aqueous solutions, to a fixed temperature and different methanol/water ratios in the solvent. Main plot: T = 313.15 K and ☐, 10% MDEA; △, 30% MDEA; and ○, 50% MDEA. Inset plot: magnification of the zone in which the three curves are crossed.

$$S^{s} = -\left(\frac{\partial \sigma_{m}}{\partial T}\right)_{p} \tag{9}$$

and

$$H^{s} = \sigma_{m} - T\left(\frac{\partial \sigma_{m}}{\partial T}\right)_{p} \tag{10}$$

where $\boldsymbol{\sigma}_{m}$ is the surface tension of the mixture, $\mathbf{S}^{S}$ is the surface entropy per unit surface area due to the formation of the interface, $\mathbf{H}^{S}$ is the surface enthalpy, i.e., the sum of the surface free energy required to extend the surface and the latent heat required to maintain isothermal conditions.[46] These equations are widely used to calculate the surface thermodynamics

Table 7. Ternary Correlation Parameters D_j (in eq 6) for Ternary Mixtures of *N*-Methyldiethanolamine in Aqueous Solutions of Methanol

	T/K						
	293.15	298.15	303.15	308.15	313.15	318.15	323.15
			Total Amine Concentration = 10% mass				
D_1	17.7361	17.9792	18.7251	19.2510	19.4065	20.4785	20.9436
D_2	−6.8469	−8.6467	−9.5005	−9.6804	−10.6673	−11.7799	−12.7135
D_3	13.6111	14.0470	14.9067	17.2511	17.6224	19.7875	21.7753
			Total Amine Concentration = 20% mass				
D_1	8.9956	9.2515	9.5650	9.7203	9.9436	10.3272	10.5560
D_2	−9.2345	−9.6649	−10.1222	−10.3121	−10.8847	−11.6684	−12.4832
D_3	9.8182	10.2342	11.1278	12.7077	12.7218	14.1108	14.8717
			Total Amine Concentration = 30% mass				
D_1	5.6826	5.8532	6.0466	6.1406	6.2941	6.5432	6.7170
D_2	−8.7483	−8.9138	−9.0837	−9.1728	−9.6402	−9.9688	−10.3768
D_3	12.0808	12.0262	12.3956	12.9477	12.6785	13.2298	13.5031
			Total Amine Concentration = 40% mass				
D_1	4.3423	4.3041	4.3192	4.1718	4.2279	4.2987	4.2161
D_2	−5.3974	−5.7925	−5.9983	−6.2045	−6.4822	−6.8135	−6.5740
D_3	12.7070	12.9756	13.1121	13.7119	13.3997	13.8261	14.5082
			Total Amine Concentration = 50% mass				
D_1	2.4436	2.4207	2.5377	2.3835	2.5374	2.6356	2.7268
D_2	−1.7250	−2.1384	−2.0971	−2.1795	−2.1332	−2.3900	−2.4227
D_3	12.5059	12.4792	12.7971	13.4585	13.3711	13.7575	14.0773

Table 8. Binary Correlation Parameters A_j and C_j (in eq 6) and f_{ij} and f_{ji} (in eq 7) for Water (1) + Methanol (2) and Water (1) + *N*-Methyldiethanolamine (3) Systems, Respectively

	T/K						
	293.15	298.15	303.15	308.15	313.15	318.15	323.15
A_1	−1.2837	−1.2874	−1.2886	−1.2846	−1.2886	−1.2901	−1.2851
A_2	0.9467	0.9557	0.9658	0.9768	0.9925	1.0068	1.0197
A_3	−0.9258	−0.9377	−0.9589	−0.9893	−0.9863	−1.0158	−1.0396
C_1	−0.6455	−0.6379	−0.6340	−0.6369	−0.6355	−0.6611	−0.6923
C_2	−0.4767	−0.2671	−0.2316	−0.2416	−0.2586	−0.2746	−0.2736
C_3	−4.8424	−4.6680	−4.6889	−4.8048	−4.8658	−4.9741	−4.9820
f_{12}	2.1333	2.103	2.0737	2.0295	2.0137	1.9524	1.9526
f_{21}	0.1207	0.1196	0.1178	0.1151	0.1041	0.1147	0.1111
f_{13}	11.7025	11.9914	12.3055	12.693	12.9265	13.2774	14.2002
$10 \cdot f_{31}$	0.2724	0.2702	0.2621	0.2526	0.2508	0.2401	0.2364

properties of binary mixtures, aqueous[35,47] or nonaqueous,[46,48] and more recently of ternary mixtures.[49,50]

Since surface tension varies linearly with temperature (eq 1), for a solution of constant composition, the values of K_1 and K_2 in eq 1 correspond to H^S and S^S, respectively. Therefore, in Tables 4 and 5, the K_1 values also represent the surface enthalpy, expressed in $mJ \cdot m^{-2} \cdot K^{-1}$, while the K_2 values represent the surface entropy, in $mJ \cdot m^{-2}$. As shown in Figure 8, for the MDEA + methanol system, the surface entropy remains almost constant along with the MDEA concentration, although at MDEA % lower than 60% ($x_{amine} > 0.2874$), S^S decreases slightly with the concentration, while it increases at higher concentrations. On the other hand, surface enthalpy remains almost constant until a low concentration ($x_{amine} < 0.1521$) and it increases rapidly for the rest of the solutions.

Figure 9 shows the influence of methanol content in the solvent on surface entropy and enthalpy, indicating that both properties decrease with increasing $x_{methanol}$ at a given x_{MDEA}. The high H^S values are an indication of strong bulk interactions, less important as the solute and solvent surface tensions approach, i.e., as the methanol percent in the solvent increases. On the other hand, decreasing S^S values are indicative of more ordered surfaces in the water-poor region, with S^S remaining nearly constant for methanol/water ratios greater than 70/30 and low MDEA %. However, at high concentrations of MDEA (>40% mass), the S^E decreases over the entire concentration range, without a steady value. Finally, the influence of MDEA on surface properties is shown in Figure 10. We observe that for methanol/water ratios > 30/70, enthalpy and surface entropy increase with MDEA % until reaching a maximum value for solutions with 40% MDEA and then both properties decrease. This change in trend coincides with that observed in Figure 6 for surface tension and corresponds to solutions in which $\sigma_{solvent} < \sigma_{solute}$ and the influence of temperature are more important at low MDEA concentrations (less than 40%).

4. CONCLUSIONS

Surface tensions of *N*-methyldiethanolamine in methanol or in methanol aqueous solutions as a solvent were measured at temperatures from 293.15 to 323.15 K. In the ternary systems, the percentage of methanol in the solvent was varied between

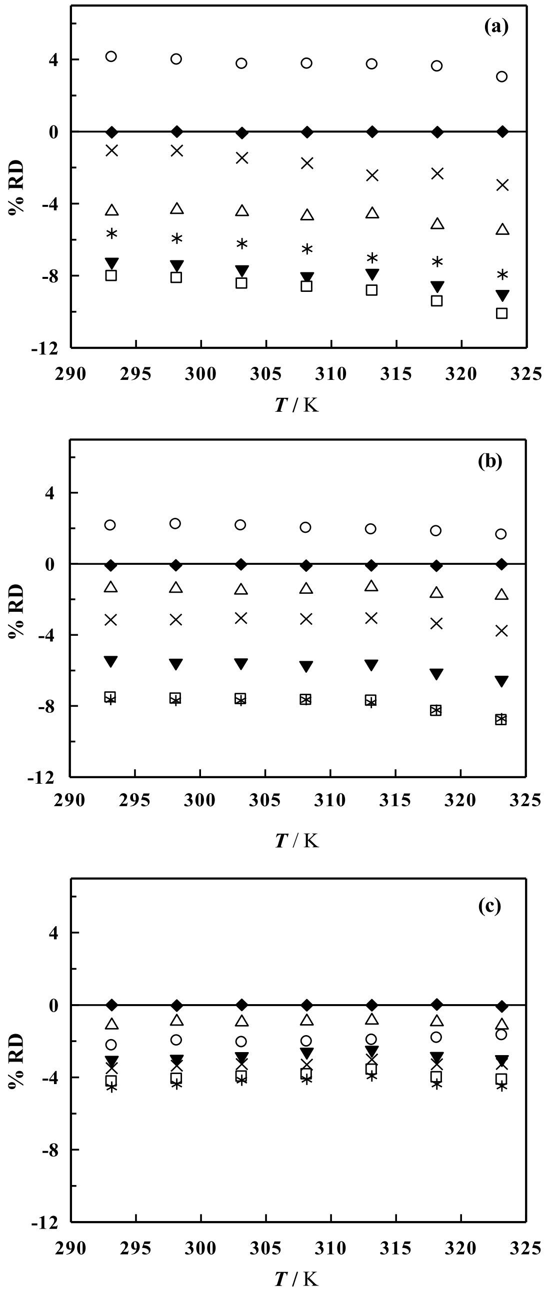

Figure 7. Relative percentage error (% RD) between experimental surface tensions and predicted LFW values, for the methanol + water + *N*-methyldiethanolamine ternary system and (a) 10% MDEA, (b) 30% MDEA, and (c) 50% MDEA, at different temperatures: ◆, % methanol/% water = 100/0; △, % methanol/% water = 90/10; ▼, % methanol/% water = 70/30; □, % methanol/% water = 50/50; ∗, % methanol/% water = 30/70, +, % methanol/% water = 10/90; and ○, % methanol/% water = 0/100.

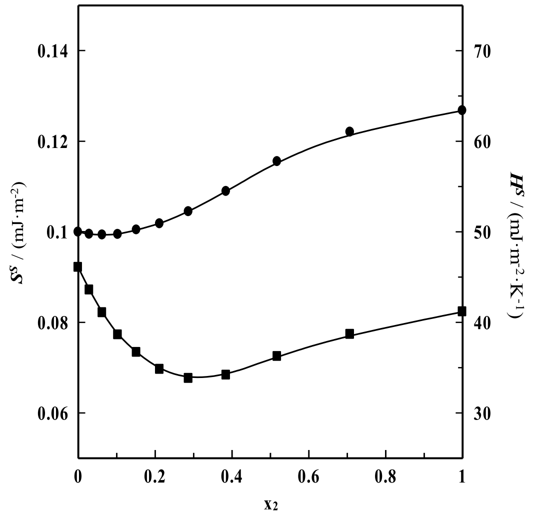

Figure 8. Surface thermodynamic properties as a function of MDEA mole fraction for the binary mixtures of methanol (1) + *N*-methyldiethanolamine (2): ●, surface enthalpy; ■, surface entropy.

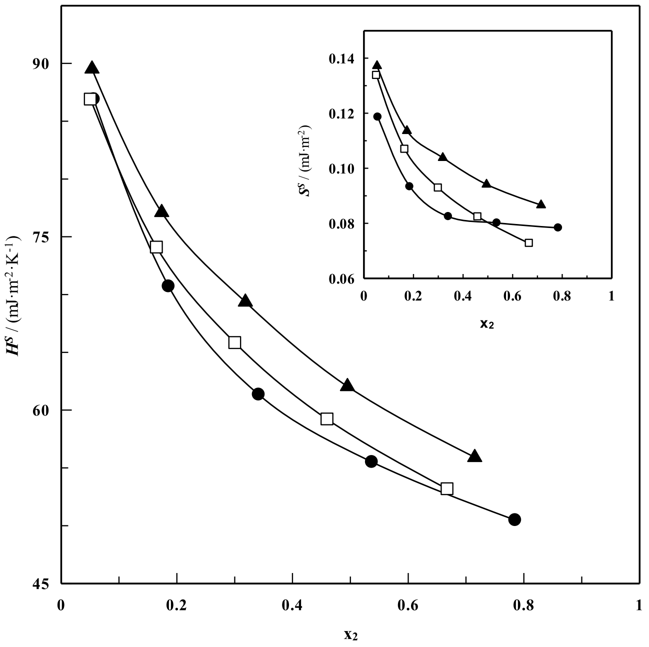

Figure 9. Influence of methanol content in the solvent on surface enthalpy (main plot) and surface entropy (insert plot) for ternary mixtures of *N*-methyldiethanolamine in water (1) + methanol (2) solutions, at different MDEA mass %: ●, 20% MDEA; ▲, 40% MDEA; and □, 50% MDEA.

10 and 90% mass, at steps of 20%, and for each solvent, the concentration of MDEA was varied between 0 and 50% mass, while in the methanol + MDEA system, the amine concentration was varied between 0 and 100% mass, at constant intervals of 10%.

From the experimental data, it is concluded that, first, all mixtures show a temperature-dependent behavior, and their surface tension decreases linearly with temperature. For this reason, the Jasper equation was used to correlate all experimental data with temperature. Second, all mixtures show nonlinear dependence on concentration for their surface tension: in all cases, the surface tension gradually decreases when the methanol % in the mixture increases. However, in ternary blends, the amine concentration effect on surface tension varies depending on the methanol/water ratio in the solvent: for ratios lower than 30/70, the surface tension decreases with increasing MDEA %, but for ratios greater than 70/30, its value increases with increasing MDEA %. For intermediate methanol/water ratios, the surface tension first increases and later decreases with the MDEA %, with the change in trend to an MDEA % > 40%.

From the values of surface entropy and enthalpy, it is concluded that the surface is more orderly as the methanol

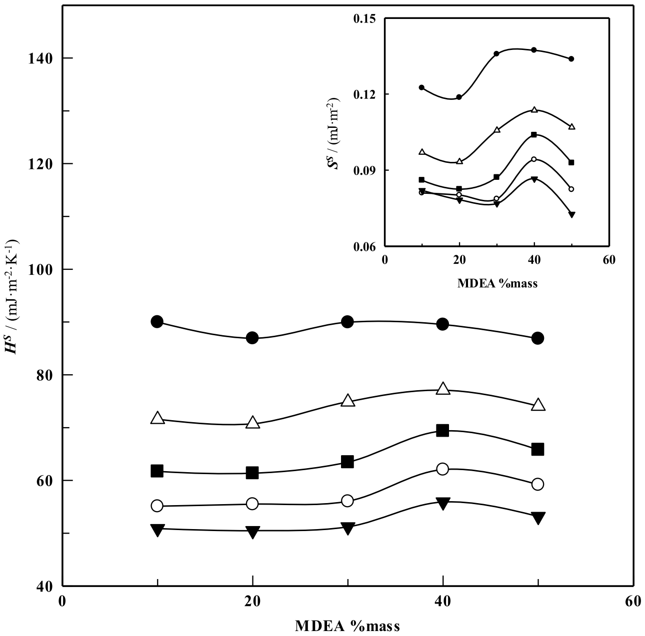

Figure 10. Surface enthalpy (main plot) and surface entropy (insert plot) as a function of MDEA mass percent for ternary mixtures of MDEA in methanol aqueous solutions, at different methanol/water ratios in the solvent: ●, % methanol/% water = 10/90; △, % methanol/% water = 30/70; ■, % methanol/% water = 50/50; ○, % methanol/% water = 70/30; and ▼, % methanol/% water = 90/10.

content increases and that there are strong bulk interactions, more important as the surface tension of solvent and solute approaches.

Finally, different models were used to correlate the experimental data. For binary mixtures, the FLW model reproduces the experimental data better than the Jouyban–Acree model, while in ternary ones, both methods give similar results. However, when the results obtained by both models for the same methanol/water ratio are compared, it is observed that the FLW model predicts the experimental data more poorly at methanol/water ratios between 30/70 and 70/30, while the Jouyban–Acree model gives worse results at ratios lower than 30/70, i.e., when there is a large difference between the solute and solvent surface tension.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jced.0c00846.

Surface tensions calculated by FLW and Jouyban–Acree models for ternary mixtures at different temperatures (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Estrella Álvarez – *Chemical, Thermal and Environmental Engineering Research Group, School of Industrial Engineering, University of Vigo, E-36310 Vigo, Spain;* orcid.org/0000-0003-3657-3495; Phone: +34 986 812213; Email: ealvarez@uvigo.es

Authors

Fernando Cerdeira – *Chemical, Thermal and Environmental Engineering Research Group, School of Industrial Engineering, University of Vigo, E-36310 Vigo, Spain*

José Manuel Navaza – *Department of Chemical Engineering, ETSE, University of Santiago de Compostela, E-15782 Santiago de Compostela, Spain*

Francisco Javier Tamajón – *Chemical, Thermal and Environmental Engineering Research Group, School of Industrial Engineering, University of Vigo, E-36310 Vigo, Spain*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.jced.0c00846

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