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Surface and Calorimetric Behavior of *N,N*-Dimethylacetamide with *n*-Alkanols or 2-Alkanols

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ABSTRACT: The surface tension at the liquid−air interface of eight binary mixtures formed by *N,N*-dimethylacetamide and *n*-alkanols (from methanol to 1-pentanol) or 2-alkanols (from 2-propanol to 2-pentanol) was studied at three different temperatures (283.15, 298.15, and 313.15 K) and a pressure of 0.1 MPa. Surface tension deviations were also calculated. Moreover, the excess molar enthalpies for all of these mixtures were measured at T = 298.15 K and a pressure of 0.1 MPa. Both deviation and excess functions were correlated with the mole fraction by means of the Redlich−Kister equation. The surface tension deviations are found to be positive for the system containing methanol, sigmoidal for ethanol and negative for the rest of the alkanols. With regard to excess molar enthalpies, negative values are found for the system containing methanol, sigmoidal values for ethanol, and positive values for the rest of the alkanols.
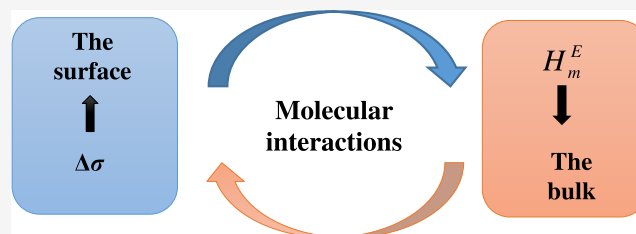
1. INTRODUCTION

Knowledge of the thermophysical properties of liquid mixtures is significant for many purposes. The chemical industries mainly deal with mixtures, and their thermophysical properties are indispensable. Mixture properties, however, show nonideal behavior for most systems. Moreover, these properties can sometimes be complicated and unpredictable, and they cannot be determined simply from the properties of the pure components comprising the mixture.[1,2] Therefore, rigorous data describing the behavior of liquid mixtures must be established.

One of the most important thermophysical properties is the surface tension, which plays an important role in industrial applications as well as in fundamental research. The interface composition of a mixture usually differs, to a certain extent, from that of the bulk phase. An estimate of surface properties is required in many industrial processes, including separation, extraction, and distillation.[3,4]

Surface tension is also related to and influenced by the interactions that take place among molecules in the bulk region.[5−7] Therefore, it is usual to report these kinds of information together. The mixtures presented in this study include *N,N*-dimethylacetamide, a dipolar unassociated solvent widely used in the adhesive industry and in chemical synthesis, and alcohols, which are polar liquids strongly self-associated through hydrogen bonding. Alcohol is a versatile solvent mainly used in chemical and organic processes. Studying these kinds of mixtures through the thermophysical properties contributes to a better understanding of the nature of the intermolecular interactions and facilitates their exploitation for practical applications.

In this sense, surface tension deviation was calculated to understand its dependence on temperature, composition, length of the alcohol chain, and the position of the hydroxyl group.

Previous studies from our laboratory[8−11] include the experimental determination of some bulk properties of these binary mixtures. Here, we focus on the excess molar enthalpy, which is the most relevant property to obtain information about the nature and strength of molecular interactions.

The purpose of this current work is to gain information about the interfacial behavior in mixtures by measuring the surface tension at several temperatures (283.15, 298.15, and 313.15 K) and at a pressure of 0.1 MPa. To the best of our knowledge, the surface tension of the mixtures studied here is not known. There are more data available regarding the excess molar enthalpies for these mixtures at temperatures of 298.15[12−15] and 313.15 K.[16]

2. MATERIALS AND METHODS

The materials in this study were used without further purification. Mass purities were provided by the suppliers, and their water content was determined by Karl−Fischer titration using an automatic device (Crison KF 1S-2B). The corresponding information is presented in Table 1.

A CP225-D Sartorius Semimicro mass balance, with an uncertainty of $\pm 1 \times 10^{-5}$ g, was employed to prepare the mixtures. The uncertainty in the mole fraction is estimated to be 0.001.

The surface tensions, σ, at the liquid−air interface of both the pure liquids and their mixtures were measured using a drop volume tensiometer (Lauda TVT-2). The surface tensions were determined at temperatures of 283.15, 298.15, and 313.15 K and

Table 1. Provenance and Purity of Liquid Compounds

chemical name	CAS number	source	purity[a] (mass fraction)	water content (mass fraction)
N,N-dimethylacetamide	127-19-5	Sigma-Aldrich	0.999	0.000305
methanol	67-56-1	Sigma-Aldrich	0.995	0.000165
ethanol	64-17-5	Acros	0.998	0.000144
1-propanol	71-23-8	Sigma-Aldrich	0.998	0.000195
1-butanol	71-36-3	Sigma-Aldrich	0.999	0.000175
1-pentanol	71-41-0	Sigma-Aldrich	0.998	0.000270
2-propanol	67-63-0	Sigma-Aldrich	0.995	0.000212
2-butanol	78-92-2	Sigma-Aldrich	0.995	0.000188
2-pentanol	6032-29-7	Sigma-Aldrich	0.998	0.000232

[a]As stated by the supplier by GC analysis.

$p = 0.1$ MPa. This tensiometer measures the volume of a drop detached from a capillary of known diameter joined to a syringe containing the sample. The drop volume V is related to the surface tension σ by

$$\sigma = \frac{V \Delta \rho g}{2 \pi r_{cap} f} \tag{1}$$

where g is the gravitational acceleration constant, $\Delta\rho$ is the difference in density between liquid and air, r_{cap} is the radius of the capillary, and f is a correction factor[17,18] that is automatically calculated by the tensiometer. The density differences needed to determine the surface tension were calculated from our previous papers,[8–11] and these calculated densities for the binary mixtures N,N-dimethylacetamide with alkanols are given in Table S1. The temperature was regulated within ± 0.01 K using a thermostat (Lauda E-200). The volumes of at least 50 drops were averaged for each surface tension determination. The uncertainty in the surface tension measurements was 0.2 mN·m^{-1}.

The thermal events associated with the mixing process of the studied mixtures were measured by means of a Thermometric 2277 TAM, thermostated within $\pm 2 \times 10^{-4}$ K, working under
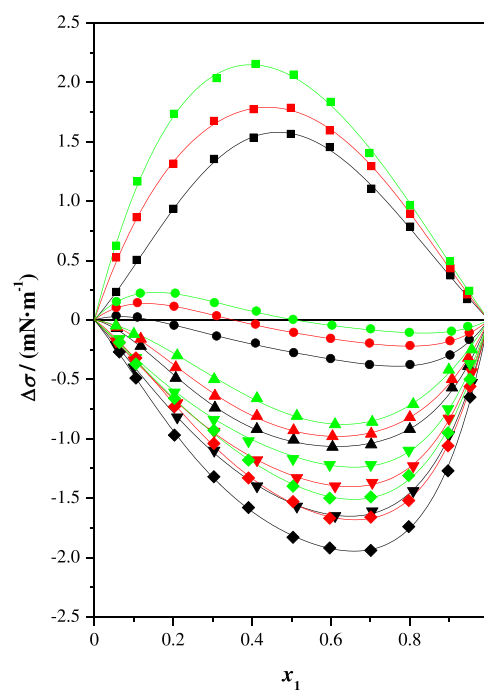

Figure 1. Surface tension deviations, $\Delta\sigma$, for N,N-dimethylacetamide (1) + n-alkanol (2) at $T = 283.15$ K (black symbols), $T = 298.15$ K (red symbols), $T = 313.15$ K (green symbols), and at $p = 0.1$ MPa as a function of mole fraction. x_1: □, methanol; ○, ethanol; Δ, 1-propanol; ▽, 1-butanol; ◇, 1-pentanol; (black, red, green line), Redlich–Kister equation.

constant flow conditions.[19] The pure liquids were supplied using two Shimadzu LC-10ADVP HPLC pumps, previously calibrated, with an uncertainty of 0.01 in the mole fraction of the mixtures. The details for all of these procedures are described in a previous paper.[20] The uncertainty in the mole fractions of the mixtures obtained by taking into account the uncertainty of the liquid flows is 0.01. The uncertainty in the determination of the excess molar enthalpy can be expected to be 0.02.

3. RESULTS AND DISCUSSION

The experimental values for the surface tension of pure components at working temperatures and atmospheric pressure are collected in Table 2. The surface tension values for the pure

Table 2. Surface Tensions, σ, of Pure Compounds at Working Temperatures and Atmospheric Pressure, $p = 0.1$ MPa, and Comparison with the Literature Data[a]

compound	σ (mN·m^{-1})					
	$T = 283.15$ K		$T = 298.15$ K		$T = 313.15$ K	
	exptl.	lit.	exptl.	lit.	exptl.	lit.
N,N-dimethylacetamide	36.91		35.17	35.43[19] 36.43[20]	33.57	31.38[19] 30.92[21]
methanol	23.52	23.5[22] 23.5[23]	22.16	22.14[24] 22.19[43]	20.88	20.9[25] 20.89[43]
ethanol	23.07	23.17[23] 23.1[26]	21.87	21.86[23] 21.9[27]	20.61	20.6[27] 20.62[28]
1-propanol	24.55	24.57[23] 24.5[29]	23.39	23.34[23] 23.31[31]	22.25	22.18[23] 22.11[30]
1-butanol	25.48	25.4[22] 25.36[23]	24.22	24.20[33] 24.18[34]	23.12	23.0[22] 23.07[24]
1-pentanol	26.55	25.5[26] 26.59[32]	25.30	25.29[31] 25.36[35]	24.08	24.05[31] 24.06[36]
2-propanol	22.04	21.98[24] 22.0[37]	20.84	20.90[38] 20.85[39]	19.63	19.71[30] 19.6[38]
2-butanol	24.28	23.54[40]	23.06	23.46[34] 22.74[36]	21.87	20.98[40]
2-pentanol	24.85	24.96[35]	23.64	23.45[41] 23.30[42]	22.37	22.04[36] 21.95[42]

[a]Standard uncertainties u are $u(T) = 0.01$ K, $u(p) = 0.0025$ MPa, and the combined expanded uncertainties U_c are $U_c(\sigma) = 0.2$ mN·m^{-1} with a 0.95 level of confidence ($k = 2$).
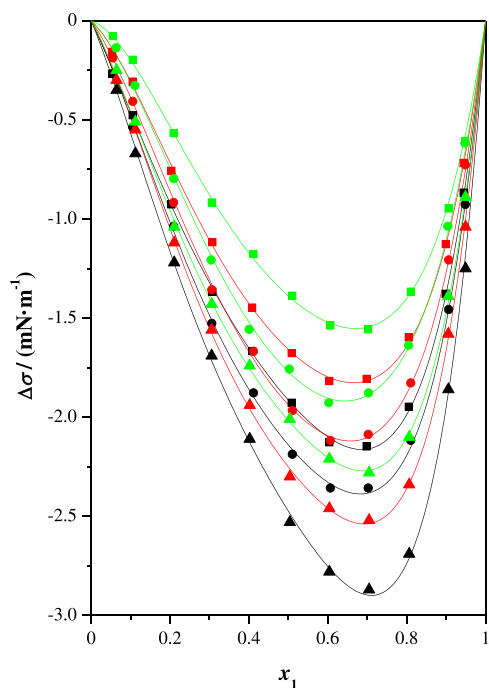
Figure 2. Surface tension deviations, $\Delta\sigma$, for N,N-dimethylacetamide (1) + 2-alkanol (2) at T = 283.15 K (black symbols), T = 298.15 K (red symbols), T = 313.15 K (green symbols), and at p = 0.1 MPa as a function of mole fraction. x_1: □, 2-propanol; ○, 2-butanol; △, 2-pentanol; (black, red, green line), Redlich−Kister equation.

compounds found in the literature[21−45] are also included in this table. In general, the accordance between the experimental and literature data is good, particularly for the case of alcohols. For N,N-dimethylacetamide, there are much less surface tension data available in the literature, and our experimental data are similar to the values reported by Shukla et al.,[21] especially at T = 298.15 K; with respect to the works of Kinart et al.[22] and Gopal and Rizvi,[23] our surface tensions are somewhat different.

Experimental surface tensions, σ, and calculated surface tension deviations, $\Delta\sigma$, and excess molar enthalpies, H_m^E, are collected in Tables 3 and 4, respectively. The surface tension deviations and excess molar enthalpies are graphically represented in Figures 1−4.

The surface tension deviation with respect to a linear dependence on the mole fraction, $\Delta\sigma$, was calculated as follows

$$\Delta\sigma = \sigma - \sum_i x_i\sigma_i \tag{2}$$

where σ, σ_i, and x_i are the surface tension of the mixture, surface tension of component i, and mole fraction of component i, respectively.

Taking into account both the compensating heating power, $\dot{Q}$, which is the heat of mixing per second, and the molar flux of the components, $\dot{n}_i$, the excess molar enthalpy, H_m^E, can be calculated using the following equation[46]

$$H_m^E = \frac{\dot{Q}}{\dot{n}_1 + \dot{n}_2} \tag{3}$$

Table 3. Surface Tensions, σ, and Surface Tension Deviations, $\Delta\sigma$, for the Binary Mixtures N,N-Dimethylacetamide (1) + Alkanol (2) at Working Temperatures and Atmospheric Pressure, p = 0.1 MPa[a]

x_1	T = 283.15 K		T = 298.15 K		T = 313.15 K	
	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})
			N,N-Dimethylacetamide (1) + methanol (2)			
0.000	23.52		22.16		20.88	
0.057	24.51	0.23	23.42	0.52	22.22	0.62
0.109	25.48	0.50	24.43	0.86	23.42	1.16
0.202	27.15	0.93	26.10	1.31	25.17	1.73
0.305	28.95	1.35	27.80	1.67	26.78	2.03
0.405	30.48	1.53	29.20	1.77	28.17	2.15
0.500	31.77	1.56	30.44	1.78	29.28	2.06
0.599	32.99	1.45	31.54	1.59	30.31	1.83
0.704	34.05	1.10	32.61	1.29	31.21	1.40
0.802	35.04	0.78	33.49	0.89	32.02	0.96
0.905	36.00	0.37	34.36	0.43	32.85	0.49
0.947	36.36	0.17	34.69	0.21	33.13	0.24
1.000	36.91		35.17		33.57	
			N,N-Dimethylacetamide (1) + Ethanol (2)			
0.000	23.07		21.87		20.61	
0.057	23.89	0.03	22.73	0.10	21.50	0.15
0.111	24.62	0.02	23.48	0.14	22.26	0.22
0.204	25.84	−0.05	24.69	0.11	23.47	0.22
0.311	27.24	−0.14	26.04	0.03	24.78	0.14
0.410	28.54	−0.20	27.28	−0.04	25.99	0.07
0.507	29.80	−0.28	28.50	−0.11	27.18	0.00
0.601	31.05	−0.33	29.70	−0.16	28.34	−0.05
0.699	32.37	−0.38	30.97	−0.20	29.59	−0.08
0.802	33.79	−0.38	32.32	−0.22	30.89	−0.11
0.903	35.27	−0.30	33.70	−0.18	32.22	−0.10
0.952	36.07	−0.17	34.42	−0.11	32.89	−0.06
1.000	36.91		35.17		33.57	

Table 3. continued

x_1	T = 283.15 K		T = 298.15 K		T = 313.15 K	
	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})
N,N-Dimethylacetamide (1) + 1-Propanol (2)						
0.000	24.55		23.39		22.25	
0.057	25.19	−0.07	24.00	−0.06	22.85	−0.05
0.118	25.79	−0.22	24.62	−0.16	23.46	−0.12
0.207	26.61	−0.49	25.42	−0.40	24.29	−0.30
0.306	27.59	−0.74	26.35	−0.64	25.21	−0.50
0.412	28.72	−0.92	27.44	−0.81	26.26	−0.66
0.505	29.79	−1.01	28.41	−0.93	27.16	−0.81
0.604	30.95	−1.07	29.53	−0.98	28.21	−0.88
0.702	32.17	−1.05	30.70	−0.96	29.33	−0.86
0.800	33.52	−0.92	31.99	−0.82	30.60	−0.71
0.907	35.19	−0.57	33.57	−0.50	32.10	−0.42
0.948	35.88	−0.39	34.24	−0.32	32.73	−0.25
1.000	36.91		35.17		33.57	
N,N-Dimethylacetamide (1) + 1-Butanol (2)						
0.000	25.48		24.22		23.12	
0.055	25.90	−0.21	24.64	−0.18	23.53	−0.16
0.100	26.23	−0.40	24.98	−0.34	23.85	−0.32
0.210	27.06	−0.82	25.82	−0.70	24.71	−0.61
0.304	27.85	−1.10	26.61	−0.94	25.45	−0.84
0.413	28.79	−1.40	27.56	−1.18	26.41	−1.02
0.514	29.78	−1.57	28.52	−1.33	27.32	−1.17
0.610	30.81	−1.65	29.50	−1.40	28.28	−1.22
0.705	31.93	−1.61	30.57	−1.37	29.27	−1.22
0.809	33.28	−1.44	31.85	−1.23	30.47	−1.10
0.899	34.78	−0.97	33.23	−0.83	31.76	−0.75
0.957	35.89	−0.53	34.26	−0.44	32.75	−0.37
1.000	36.91		35.17		33.57	
N,N-Dimethylacetamide (1) + 1-Pentanol (2)						
0.000	26.55		25.30		24.08	
0.063	26.94	−0.27	25.74	−0.19	24.49	−0.19
0.105	27.15	−0.49	26.02	−0.32	24.71	−0.37
0.203	27.68	−0.97	26.57	−0.73	25.35	−0.66
0.303	28.37	−1.32	27.25	−1.04	26.02	−0.93
0.391	29.02	−1.58	27.83	−1.33	26.61	−1.18
0.504	29.94	−1.83	28.74	−1.53	27.46	−1.40
0.597	30.81	−1.92	29.52	−1.67	28.24	−1.50
0.701	31.87	−1.94	30.56	−1.66	29.24	−1.49
0.797	33.07	−1.74	31.65	−1.52	30.34	−1.31
0.897	34.57	−1.27	33.09	−1.06	31.64	−0.95
0.952	35.77	−0.65	34.14	−0.56	32.62	−0.50
1.000	36.91		35.17		33.57	
N,N-Dimethylacetamide (1) + 2-propanol (2)						
0.000	22.04		20.84		19.63	
0.055	22.58	−0.27	21.46	−0.16	20.31	−0.08
0.107	23.15	−0.48	22.06	−0.31	20.92	−0.20
0.205	24.16	−0.93	23.02	−0.76	21.92	−0.57
0.309	25.26	−1.37	24.15	−1.12	23.02	−0.92
0.410	26.46	−1.67	25.26	−1.45	24.16	−1.18
0.510	27.69	−1.93	26.47	−1.68	25.35	−1.39
0.605	28.91	−2.13	27.70	−1.82	26.53	−1.54
0.700	30.30	−2.15	29.06	−1.81	27.83	−1.56
0.807	32.08	−1.95	30.80	−1.60	29.50	−1.37
0.901	34.05	−1.38	32.61	−1.13	31.23	−0.95
0.946	35.24	−0.87	33.67	−0.72	32.21	−0.61
1.000	36.91		35.17		33.57	
N,N-Dimethylacetamide (1) + 2-Butanol (2)						
0.000	24.28		23.06		21.87	
0.056	24.72	−0.27	23.55	−0.19	22.39	−0.14
0.107	25.09	−0.54	23.94	−0.41	22.79	−0.33
0.211	25.90	−1.04	24.69	−0.92	23.53	−0.80

Table 3. continued

x_1	T = 283.15 K		T = 298.15 K		T = 313.15 K	
	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})	σ (mN·m^{-1})	$\Delta\sigma$ (mN·m^{-1})
0.307	26.63	−1.53	25.42	−1.36	24.26	−1.21
0.413	27.62	−1.88	26.39	−1.67	25.14	−1.56
0.512	28.55	−2.19	27.28	−1.97	26.09	−1.76
0.608	29.60	−2.36	28.30	−2.12	27.05	−1.93
0.704	30.81	−2.36	29.49	−2.09	28.22	−1.88
0.812	32.41	−2.12	31.06	−1.83	29.72	−1.64
0.908	34.28	−1.46	32.84	−1.21	31.45	−1.04
0.950	35.35	−0.93	33.83	−0.73	32.36	−0.62
1.000	36.91		35.17		33.57	
			N,N-Dimethylacetamide (1) + 2-Pentanol (2)			
0.000	24.85		23.64		22.37	
0.065	25.28	−0.35	24.08	−0.30	22.84	−0.25
0.112	25.53	−0.67	24.38	−0.55	23.12	−0.51
0.211	26.17	−1.22	24.95	−1.12	23.69	−1.04
0.306	26.84	−1.69	25.60	−1.56	24.36	−1.43
0.402	27.59	−2.11	26.34	−1.94	25.13	−1.74
0.504	28.40	−2.53	27.15	−2.30	26.00	−2.01
0.604	29.35	−2.78	28.14	−2.46	26.92	−2.21
0.705	30.48	−2.87	29.25	−2.52	27.98	−2.28
0.806	31.88	−2.69	30.60	−2.34	29.30	−2.10
0.906	33.91	−1.86	32.50	−1.58	31.12	−1.39
0.949	35.04	−1.25	33.54	−1.04	32.11	−0.89
1.000	36.91		35.17		33.57	

[a]Standard uncertainties u are $u(T)$ = 0.01 K, $u(p)$ = 0.0025 MPa, and $u(x_1)$ = 0.001, and the combined expanded uncertainties U_c are $U_c(\sigma)$ = 0.2 mN·m^{-1} with a 0.95 level of confidence (k = 2).

Table 4. Excess Molar Enthalpies, H_m^E, for the Binary Mixtures *N,N*-Dimethylacetamide (1) + Alkanol (2) at T = 298.15 K and Atmospheric Pressure, p = 0.1 MP[a]

x_1	H_m^E (J·mol^{-1})	x_1	H_m^E (J·mol^{-1})	x_1	H_m^E (J·mol^{-1})
		N,N-Dimethylacetamide (1) + Methanol (2)			
0.04	−97.8	0.35	−679.7	0.80	−422.0
0.10	−278.4	0.51	−731.8	0.89	−231.1
0.20	−475.9	0.60	−680.0	0.95	−105.1
0.28	−615.0	0.70	−572.5		
		N,N-Dimethylacetamide (1) + Ethanol (2)			
0.04	7.1	0.31	−4.1	0.70	−69.7
0.09	16.3	0.40	−25.6	0.79	−56.8
0.19	16.6	0.50	−54.2	0.84	−41.7
0.24	8.1	0.60	−67.4	0.93	−16.1
		N,N-Dimethylacetamide (1) + 1-Propanol (2)			
0.05	67.0	0.47	174.2	0.80	68.8
0.09	117.6	0.58	139.7	0.90	35.9
0.20	187.9	0.67	105.4	0.95	18.1
0.32	203.6	0.80	68.8		
		N,N-Dimethylacetamide (1) + 1-Butanol (2)			
0.03	70.0	0.32	359.3	0.70	256.4
0.08	167.2	0.42	362.8	0.80	189.3
0.16	270.0	0.50	348.0	0.90	104.8
0.20	308.0	0.61	311.9	0.95	56.3
		N,N-Dimethylacetamide (1) + 1-Pentanol (2)			
0.05	108.3	0.38	457.0	0.79	288.3
0.09	193.9	0.49	469.1	0.89	161.5
0.19	331.2	0.60	436.1	0.95	82.2
0.29	417.5	0.69	376.3		
		N,N-Dimethylacetamide (1) + 2-propanol (2)			
0.06	172.2	0.39	551.5	0.79	259.1
0.09	260.9	0.49	528.0	0.89	132.7
0.19	440.3	0.60	462.9	0.95	65.3
0.29	532.9	0.68	372.1		

Table 4. continued

| x_1	H_m^E (J·mol^{-1})	x_1	H_m^E (J·mol^{-1})	x_1	H_m^E (J·mol^{-1})
		N,N-Dimethylacetamide (1) + 2-Butanol (2)			
0.05	159.3	0.39	664.2	0.78	351.4
0.09	290.6	0.49	647.9	0.88	187.1
0.19	505.7	0.59	584.3	0.95	86.0
0.29	616.4	0.69	483.7		
		N,N-Dimethylacetamide (1) + 2-Pentanol (2)			
0.05	174.0	0.40	707.4	0.80	407.2
0.09	311.0	0.49	712.7	0.88	247.2
0.20	521.3	0.60	666.0	0.95	115.8
0.30	651.7	0.69	566.5		

[a]Standard uncertainties u are $u(T)$ = 0.0002 K, $u(p)$ = 0.0025 MPa, and $u(x_1)$ = 0.01, and the expanded uncertainty U_r is $U_r(H_m^E)$ = 0.02 with a 0.95 level of confidence (k = 2).

For each binary mixture, the deviation or excess properties as a function of composition were correlated with a Redlich−Kister polynomial equation[47]

$$Y = x_i(1 - x_i) \sum_{i=0} A_i(2x_i - 1)^i \tag{4}$$

where Y refers to $\Delta\sigma$ or H_m^E and x_i is the mole fraction. The values of coefficients A_i were obtained by the least-squares method, and the number of these coefficients was chosen to minimize the standard deviation for the fit. These parameters, A_i, and the corresponding standard deviation, $\sigma(Y)$, are listed in Table 5.

The surface tension deviations, $\Delta\sigma$, for our mixtures as a function of the composition of *N,N*-dimethylacetamide show three different behaviors (Figures 1−6). Most mixtures show negative deviation at all studied temperatures. The minima for
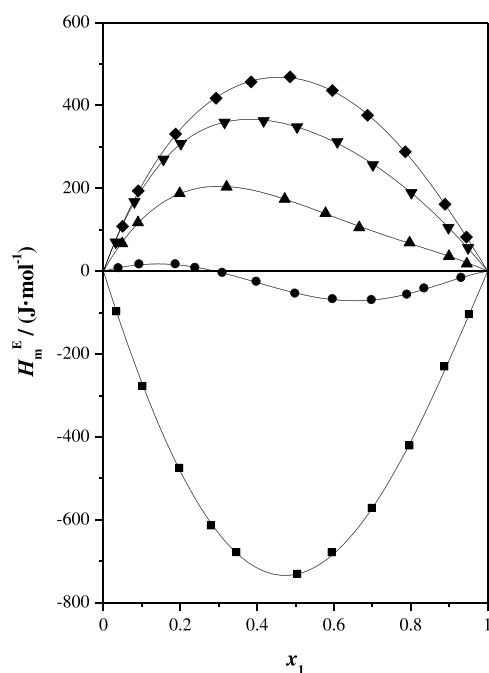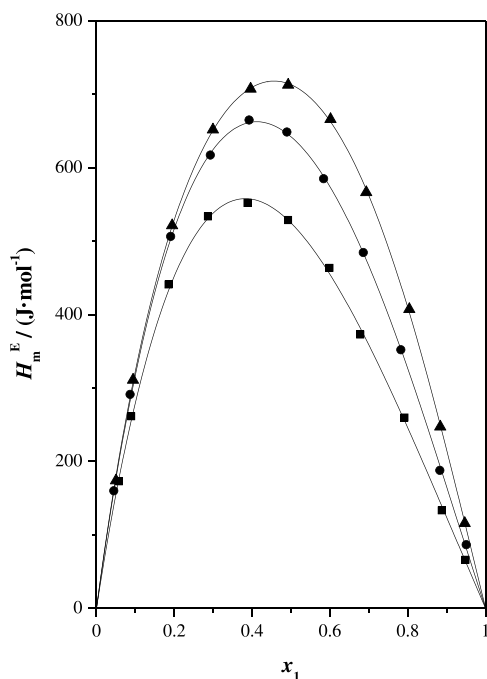
Figure 3. Excess molar enthalpies, H_m^E, for N,N-dimethylacetamide (1) + n-alkanol (2) at T = 298.15 K and p = 0.1 MPa as a function of mole fraction. x_1: ■, methanol; ●, ethanol; ▲, 1-propanol; ▼, 1-butanol; ◆, 1-pentanol; —, Redlich−Kister equation.

the surface tension deviation occur in the amide-rich region ($0.6 \leq x_1 \leq 0.7$). The $\Delta\sigma$ values are more negative in the following order: 1-propanol < 1-butanol < 1-pentanol < 2-propanol < 2-butanol < 2-pentanol. We can remark that the obtained $\Delta\sigma$ values are less negative at higher temperatures and that the absolute values for the mixtures containing 2-alkanol are much higher than for 1-alkanol. However, the N,N-dimethylacetamide + ethanol mixture presents a sigmoidal shape, which is more noticeable at temperatures of 298.15 and 313.15 K. Surface tension deviations are positive in the alcohol-rich region, and they become negative in the amide-rich region. An increase in temperature leads to a shift of the surface tension deviations toward positive values. The N,N-dimethylacetamide + methanol system is the only mixture that shows positive deviation at all temperatures investigated. The curves are nearly symmetrical, and the maxima are found at $x_1 \approx 0.45$. When the temperature increases, the absolute value for the surface tension deviation increases.

The bulk and surface properties are intimately related and influenced by each other. In the mixing process, changes in the structure of the components and new interactions, repulsive or attractive, between unlike molecules can take place in the bulk and affect the surface behavior. Moreover, the surface migration of molecules from the bulk depends on their interaction in the bulk.[48] Therefore, when we analyze the surface behavior, we should consider the intermolecular interactions occurring in the liquid phase.[6,7]

Table 5. Adjusted Coefficients and Standard Deviations, $\sigma(Y)$, for the Fitting Equation

function	T (K)	A_0	A_1	A_2	A_3	$\sigma(Y)$
			N,N-Dimethylacetamide (1) + Methanol (2)			
	283.15	6.27	−1.23	−2.65	1.13	0.02
$\Delta\sigma$ (mN·m^{-1})	298.15	7.05	−1.80	−0.23	−1.19	0.02
	313.15	8.21	−3.79	0.55		0.03
H_m^E (J·mol^{-1})	298.15	−2927.1	371.4	372.4		4.8
			N,N-Dimethylacetamide (1) + Ethanol (2)			
	283.15	−1.08	−1.25	−0.71	−1.49	0.01
$\Delta\sigma$ (mN·m^{-1})	298.15	−0.41	−1.19	0.23	−1.47	0.01
	313.15	0.02	−1.17	0.93	−1.51	0.00
H_m^E (J·mol^{-1})	298.15	−210.5	−441.5	253.0	248.3	1.5
			N,N-Dimethylacetamide (1) + 1-Propanol (2)			
	283.15	−4.09	−1.52	−0.58	−2.42	0.01
$\Delta\sigma$ (mN·m^{-1})	298.15	−3.72	−1.75	−0.04	−1.60	0.01
	313.15	−3.21	−2.23	0.26	−0.18	0.01
H_m^E (J·mol^{-1})	298.15	659.6	−634.5	363.1		2.3
			N,N-Dimethylacetamide (1) + 1-Butanol (2)			
	283.15	−6.17	−2.79	−2.27	−2.04	0.02
$\Delta\sigma$ (mN·m^{-1})	298.15	−5.23	−2.39	−2.01	−1.69	0.01
	313.15	−4.57	−2.20	−2.01	−1.32	0.01
H_m^E (J·mol^{-1})	298.15	1396.6	−551.3	429.6	−152.6	2.5
			N,N-Dimethylacetamide (1) + 1-Pentanol (2)			
	283.15	−7.25	−3.12	−3.18	−3.18	0.02
$\Delta\sigma$ (mN·m^{-1})	298.15	−6.16	−3.27	−2.03	−2.61	0.02
	313.15	−5.51	−3.03	−1.97	−1.47	0.02
H_m^E (J·mol^{-1})	298.15	1859.9	−295.5	161.2	−198.2	3.0
			N,N-Dimethylacetamide (1) + 2-propanol (2)			
	283.15	−7.66	−4.54	−3.96	−2.83	0.02
$\Delta\sigma$ (mN·m^{-1})	298.15	−6.63	−3.73	−2.05	−3.06	0.02
	313.15	−5.56	−3.53	−1.37	−2.73	0.02
H_m^E (J·mol^{-1})	298.15	2098.8	−1076.9	228.6		6.4
function	T / K	A_0	A_1	A_2	A_3	$\sigma(Q)$
			N,N-Dimethylacetamide (1) + 2-Butanol (2)			
	283.15	−8.56	−4.72	−4.07	−3.91	0.03

Table 5. continued

$\Delta\sigma$ (mN·m^{-1})	298.15	−7.75	−4.28	−2.26	−2.89	0.02
	313.15	−7.08	−3.89	−1.24	−2.56	0.02
H_m^E (J·mol^{-1})	298.15	2568.5	−906.7	184.0	−240.6	4.5
			N,N-Dimethylacetamide (1) + 2-Pentanol (2)			
	283.15	−9.90	−6.39	−6.39	−5.00	0.04
$\Delta\sigma$ (mN·m^{-1})	298.15	−9.01	−5.09	−4.59	−4.82	0.02
	313.15	−8.07	−4.55	−4.16	−3.96	0.02
H_m^E (J·mol^{-1})	298.15	2852.0	−460.2	226.4	−455.2	3.9

Figure 4. Excess molar enthalpies, H_m^E, for *N,N*-dimethylacetamide (1) + 2-alkanol (2) at T = 298.15 K and p = 0.1 MPa as a function of mole fraction. x_1: ■, 2-propanol; ●, 2-butanol; ▲, 2-pentanol; —, Redlich−Kister equation.

Based on our calorimetric results (Figures 3 and 4), we can say that for the case of *N,N*-dimethylacetamide + methanol, $H_m^E < 0$, the heteroassociation between alcohol and amide overcomes the structure breaking effects associated with mixing. Molecular complexes are formed between the alcohol and amide in the liquid phase, which maintain the smaller surface tension of molecules in the bulk. This effect and the small difference between the surface tension values of pure components lead to positive surface tension deviation values.[48,49]

For *N,N*-dimethylacetamide + ethanol, the $\Delta\sigma$ behavior exhibits an inversion of signs such as H_m^E, indicating competition between two phenomena: heteroassociation between alcohol and amide, which prevails in the alcohol-rich region, and the breaking of hydrogen bonds, lead to the migration of alcohol to the free surface of the liquid, which dominates in the amide-rich region.[49]

For the rest of the mixtures, positive H_m^E values correspond to negative surface tension deviations and follow the same order. Since alcohols show lower surface tensions than *N,N*-dimethylacetamide, they are displaced to the surface, whereas the amide will tend to remain in the bulk. Consequently, the surface tension of the mixture decreases, and the $\Delta\sigma$ values should be negative.[48] We can conclude that as the alcohol chain length increases, the possibility of establishing specific interactions between alcohol and amide decreases. This is reflected by a more enriched surface with alcohol and consequently more negative $\Delta\sigma$ values. Additionally, the position of the OH group can influence this property, which
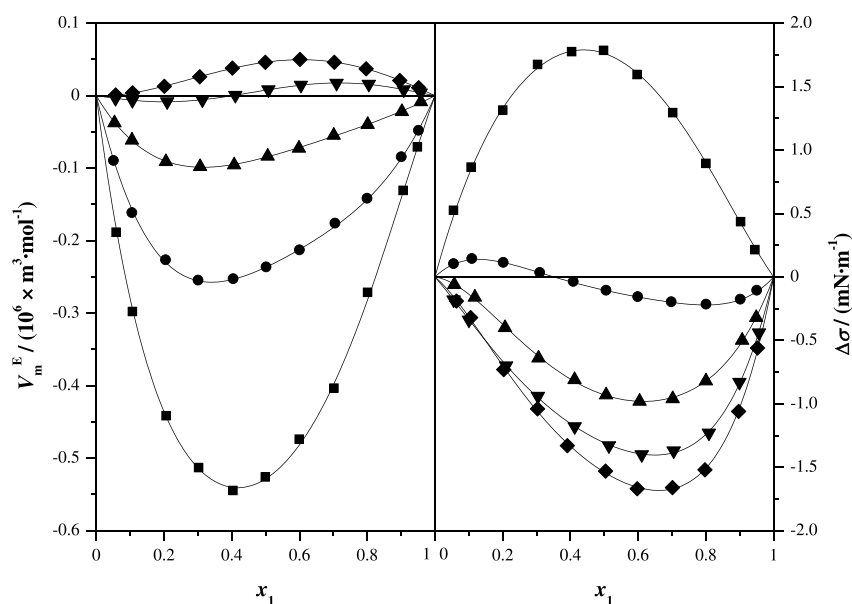

Figure 5. Comparison of excess molar volumes, V_m^E, with surface tension deviations, $\Delta\sigma$, for *N,N*-dimethylacetamide (1) + *n*-alkanol (2) at T = 298.15 K and p = 0.1 MPa as a function of mole fraction. x_1: ■, methanol; ●, ethanol; ▲, 1-propanol; ▼, 1-butanol; ◆, 1-pentanol.

Table 6. Mean Absolute Deviations, MAD, between our Experimental and Literature H_m^E Data

| reference	MAD (J·mol^{-1})
N,N-Dimethylacetamide (1) + Methanol (2)	
Oba et al.[12]	7
Iloukhani and Zarei[14]	65
N,N-Dimethylacetamide (1) + Ethanol (2)	
Oba et al.[12]	4
Iloukhani and Zarei[14]	288
N,N-Dimethylacetamide (1) + 1-Propanol (2)	
Oba et al.[12]	3
Iloukhani and Zarei[14]	24
N,N-Dimethylacetamide (1) + 1-Butanol (2)	
Oba et al.[12]	6
Chao et al.[13]	10
Iloukhani and Zarei[14]	7
N,N-Dimethylacetamide (1) + 1-Pentanol (2)	
Iloukhani and Zarei[14]	44
N,N-Dimethylacetamide (1) + 2-Propanol (2)	
Iloukhani and Zarei[15]	51
N,N-Dimethylacetamide (1) + 2-Butanol (2)	
Chao et al.[13]	22
Iloukhani and Zarei[15]	52

can explain the higher hydrophobicity of secondary alcohols versus primary alcohols.[49]

Excess molar enthalpy, H_m^E, is directly related to energetic effects, and its variation shows a transition from negative to positive as the alcohol chain length increases. An exothermic behavior is marked for the *N,N*-dimethylacetamide + methanol mixture. The H_m^E curve is nearly symmetrical with a minimum $(-730 \text{ J·mol}^{-1})$ at approximately $x_1 \approx 0.47$. Sigmoid behavior is obtained with ethanol, while endothermic behavior is observed for the rest of the alcohols. The curves are slightly shifted to the region rich in alcohol, and the maxima occur at $0.3 \leq x_1 \leq 0.45$. Maxima are obtained at 200, 360, 470, 550, 660, 730 J·mol^{-1}, and they follow the sequence of 1-propanol < 1-butanol < 1-pentanol < 2-propanol < 2-butanol < 2-pentanol.

Negative H_m^E values reflect the existence of strong interactions during the mixing process. This trend can be explained by the predominance of attractive forces between different molecules through dipole−dipole and hydrogen bonding leading to complex formation.[12,16,50] Positive H_m^E values can be interpreted by the preponderance of dissociation of the aggregates formed in the pure state (alcohols through hydrogen bonding and *N,N*-dimethylacetamide through dipole−dipole interaction).[15,16,50] It is noticed that secondary alcohols exhibit much higher H_m^E values than primary alcohols and that the H_m^E values increase with increasing alcohol chain length.

The influence of molecular interactions on the deviation of the surface tension has been commented on above, since the calorimetric behavior is due to these interactions. On the other hand, the influence of structural factors is more related to the volumetric behavior. Figures 5 and 6 show a graphical comparison of excess molar volumes[8−11] with surface tension deviations for the systems studied here at $T = 298.15$ K and $p = 0.1$ MPa; for the rest of the temperatures, the graphical comparisons are similar. In fact, for these systems, the surface tension deviation clearly correlates with the excess molar volume. These last two properties are presented in general terms with opposite signs. The excess molar volumes for *N,N*-dimethylacetamide + 1-alkanols change from slightly positive values for 1-pentanol to negative values for methanol. However, the surface tension deviations change from positive values for methanol to negative values for 1-pentanol. With regard to the systems containing 2-alkanols, they present positive excess molar volumes, and 2-alkanols exhibit unfavorable packing with *N,N*-dimethylacetamide due to steric hindrance with respect to 1-alkanols. On the other hand, their surface tension deviations are negative.

Finally, we compare our H_m^E results with those reported in the literature. Our excess molar enthalpies agree very well with those reported by Oba et al.[12] and Chao et al.,[13] although for the *N,N*-dimethylacetamide + 2-butanol system, the mean absolute deviation rises to 22 J·mol^{-1}. On the other hand, the differences from the results reported by Iloukhani and Zarei[14,15] are higher except for the systems containing the longer *n*-alkanols (C3−C5), and the mean absolute deviation is especially
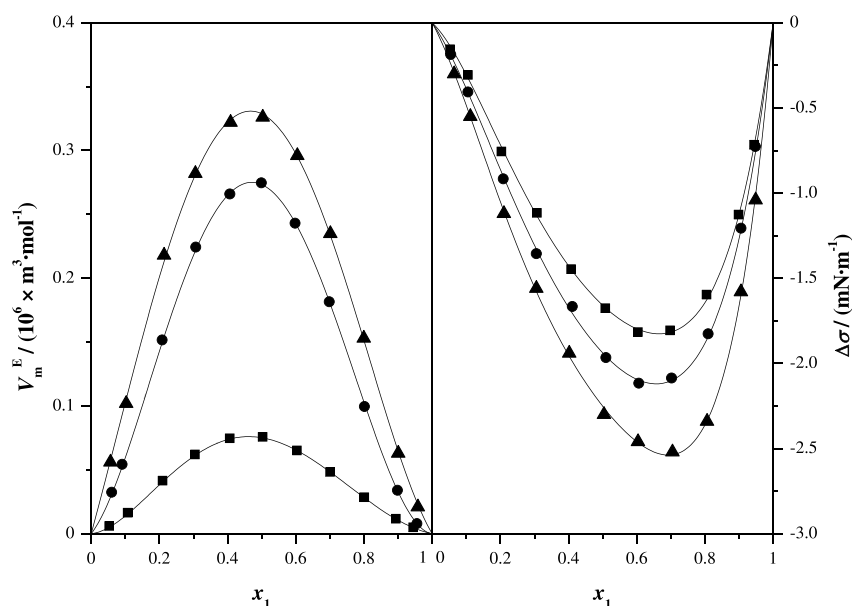
Figure 6. Comparison of excess molar volumes, V_m^E, with surface tension deviations, $\Delta\sigma$, for *N,N*-dimethylacetamide (1) + 2-alkanol (2) at $T = 298.15$ K and $p = 0.1$ MPa as a function of mole fraction. x_1: ■, 2-propanol; ●, 2-butanol; ▲, 2-pentanol.

elevated for the mixtures containing ethanol, 2-propanol, and 2-butanol. For the N,N-dimethylacetamide + ethanol system, it must be taken into account that our excess molar enthalpies show a sigmoidal behavior with a composition similar to that of Oba et al.,[12] while the H_m^E values of Iloukhani and Zarei[15] are negative over the whole composition range. This fact leads to very large mean absolute deviations. Table 6 shows a comparison of our experimental excess molar enthalpy results with previously reported values in terms of mean absolute deviations, MAD. The deviations between our experimental values and those reported in the literature are graphically represented in Figures S1−S7.

4. CONCLUSIONS

This study presents experimental values for the surface tension of binary mixtures of methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 1-pentanol, 2-pentanol, and N,N-dimethylacetamide over the whole composition range at several temperatures (283.15, 298.15, and 313.15 K) and $p = 0.1$ MPa. The results show that the $\Delta\sigma$ values exhibit an inversion in sign from positive to negative for increasing alcohol chain length and that the most negative values are reached in the mixtures containing 2-alkanol. Additional experimental data for the excess enthalpy H_m^E are provided at $T = 298.15$ K, with the aim of understanding the molecular interactions occurring in the bulk phase that can affect the interfacial behavior. The results obtained show that surface and calorimetric behaviors are intimately related and that the H_m^E behavior is opposite to the $\Delta\sigma$ behavior.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jced.2c00265.

> Calculated densities, ρ, from our previous works used for the determination of surface tensions and figures showing the deviations between the experimental and literature excess molar enthalpies (PDF)

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Notes

The authors declare no competing financial interest.

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