J. Chem. Eng. Data **2001,** *46,* 51−55

Heat Capacity of Aqueous Mixtures of Monoethanolamine with *N*-Methyldiethanolamine

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Heat capacities of aqueous mixtures of monoethanolamine with *N*-methyldiethanolamine were measured from 30 to 80 °C with a differential scanning calorimeter (DSC). The binary systems studied were *N*-methyldiethanolamine + water and monoethanolamine + *N*-methyldiethanolamine. For mole fractions of water ranging from 0.2 to 0.8, 16 concentrations of the monoethanolamine + *N*-methyldiethanolamine + water systems were studied. The estimated uncertainty of the measured heat capacities is ±2% including the effect of up to 1% impurities in a substance. An excess molar heat capacity expression using the Redlich−Kister equation for the composition dependence is used to represent the measured C_p of aqueous alkanolamine solutions. The overall average absolute percentage deviation of the calculation of the molar heat capacity for a total of 176 data points for the monoethanolamine + *N*-methyldiethanolamine + water system is 0.5%. The heat capacities of aqueous mixtures of monoethanolamine with *N*-methyldiethanolamine presented in this study are, in general, of sufficient accuracy for most engineering-design calculations.

Introduction

Acid gas impurities (CO_2 and H_2S) are commonly removed from natural gas and synthetic and refinery gas streams by absorption into aqueous alkanolamine solutions. Alkanolamines such as monoethanolamine (MEA), diglycolamine (DGA), diethanolamine (DEA), di-2-propanolamine (DIPA), triethanolamine (TEA), *N*-methyldiethanolamine (MDEA), 2-amino-2-methyl-l-propanol (AMP), and 2-piperidineethanol (2-PE) can be used as an absorbent for acid gas removal processes.[1]

Heat capacity data for aqueous alkanolamine solutions are required for the design of the heat-exchanger equipment used in gas-treating processes. The heat capacities of pure alkanolamines have been reported in the literature.[2−6] Hayden et al.[7] have studied heat capacities of aqueous 23 wt % and 50 wt % MDEA solutions at 25, 50, and 75 °C. Presented in graphical form, the C_P values of aqueous MEA solutions (15, 30, and 50 wt %),[8] aqueous DGA solutions (50, 60, 70, 80, and 90 wt %),[9] and aqueous DEA solutions (15, 30, and 50 wt %)[8] have been reported for temperature ranges from 10 to 110 °C.[1] Chiu and Li[10] have recently reported the heat capacities of eight aqueous alkanolamine solutions (MEA, DGA, DEA, DIPA, TEA, MDEA, AMP, 2-PE) at temperatures ranging from 30 to 80 °C. At 25 °C, heat capacities of CO_2-loaded, aqueous solutions of MEA, DEA, MDEA, aqueous MDEA + MEA, and MDEA + DEA were also studied.[11] For the heat capacity of aqueous blend−alkanolamine solutions, little information is available in the literature. The purpose of this study is to measure the heat capacities of MEA + MDEA + H_2O by using a differential scanning calorimeter (DSC). An excess molar heat capacity expression, using the Redlich−Kister equation for the concentration dependence, will be applied to represent the measured C_p data.

Experimental Section

Chemicals. MEA is Riedel-de Haën reagent grade with the purity min. 99% (water max. 0.5%), and MDEA is Acros Organics reagent grade with the purity 99+%. The water is liquid chromatography grade from Alps Chemical Co.

Apparatus and Procedures. The differential scanning calorimeter used for this study consists of a DSC-2010 and a thermal analysis controller from TA Instruments. The DSC operating range is from the room temperature to 725 °C. Both the temperatures and the heat flow associated with transitions in materials can be easily and rapidly measured with the system. The DSC operates with a temperature repeatability of ±0.1 °C. Calorimetric sensitivity is 1 μW (rms). The calorimetric precision is ±1%, based on measurements of metal samples. The purge gas is nitrogen with a flow rate of 40 $mL\cdot min^{-1}$. The heating rate is set to be 5 $°C\cdot min^{-1}$. By using the sample encapsulating press, the liquid sample was prepared within a hermetic sample pan. The internal volume of the hermetic pan is approximately 10 mm^3. Sample weight is in the range 15−20 mg. Normally four to five runs are carried out for each measurement. The accuracy of the C_p measurements is estimated to be ±2% on the basis of the measurement of the C_p of liquid water.[6] The apparatus and the experimental procedure are the same as those described by Chiu et al.[6]

Results and Discussion

The values of C_p of MDEA have been measured for temperatures from 30 to 80 °C and are presented in Table 1. In Figure 1, comparisons between our measurement and the available values[3,5,6,12,13] are shown. The values of C_p of Missenard[12] and Chueh and Swanson[13] are estimated from group contribution methods. Probably due to higher MDEA purity (with 99+%) while only 98.5% MDEA purity for Chiu et al.,[6] the values of C_p obtained in this study are slightly higher that those of Chiu et al.[6] As shown in Figure 1, an

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10.1021/je0000367 CCC: $20.00

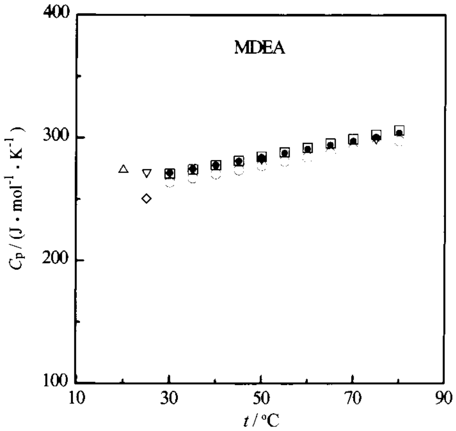

Figure 1. Heat capacity of MDEA: ●, this study; ◇, Missenard;[12] ▲, Chueh and Swanson;[13] □, Lee;[3] ▽, Maham et al.;[5] ○, Chiu et al.[6]

Table 1. Heat Capacities of MDEA

t/°C	C_p/(J·mol^{-1}·K^{-1})	t/°C	C_p/(J·mol^{-1}·K^{-1})
30	272	60	291
35	275	65	295
40	278	70	298
45	281	75	301
50	285	80	304
55	288		

Table 2. Heat Capacities of MDEA (1) + H_2O (2)

	molar heat capacity C_p and excess molar heat capacity C_p^E/(J·mol^{-1}·K^{-1})							
	$x_1 = 0.2$		$x_1 = 0.4$		$x_1 = 0.6$		$x_1 = 0.8$	
t/°C	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E
30	117	2.38	155	1.35	191	−1.71	229	−3.23
35	118	2.91	157	2.00	194	−1.22	232	−2.96
40	119	3.45	159	2.63	196	−0.77	235	−2.65
45	121	4.00	161	3.25	199	−0.28	238	−2.39
50	122	4.53	163	3.88	201	0.21	241	−2.12
55	123	5.07	165	4.53	204	0.67	244	−1.85
60	124	5.60	167	5.16	206	1.16	246	−1.58
65	125	6.15	169	5.78	209	1.65	249	−1.31
70	127	6.69	171	6.43	211	2.14	252	−1.00
75	128	7.22	173	7.06	213	2.60	255	−0.73
80	129	7.77	175	7.68	216	3.09	258	−0.47

excellent agreement between our measurements and those of Lee[3] and Maham et al.[5] is shown. The obtained values of C_p of MDEA are expressed as a function of temperature as

$$C_p/(\mathrm{J\cdot mol^{-1}\cdot K^{-1}}) = 72.7155 + 0.6557(T/\mathrm{K}) \qquad (1)$$

Since the values of C_p of MDEA appear to be linear for the temperatures ranging from 30 to 80 °C, the result (in AAD %, average absolute percentage deviation) of the calculation of C_p for MDEA from eq 1 is only 0.2%.

The values of C_p for the binary systems MDEA (1) + H_2O (2) and MEA (1) + MDEA (2) were also measured for temperatures from 30 to 80 °C and are presented in Tables 2 and 3.

An excess molar heat capacity C_p^E for a mixture is defined as[14]

$$C_p^E = C_p - \sum_i x_i C_{p,i} \qquad (2)$$

Table 3. Heat Capacities of MEA (1) + MDEA (2)

	molar heat capacity C_p and excess molar heat capacity C_p^E/(J·mol^{-1}·K^{-1})							
	$x_1 = 0.2$		$x_1 = 0.4$		$x_1 = 0.6$		$x_1 = 0.8$	
t/°C	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E
30	246	−4.34	223	−6.91	201	−7.56	182	−5.86
35	249	−4.47	225	−7.02	203	−7.68	184	−5.96
40	252	−4.60	228	−7.13	206	−7.79	186	−6.06
45	255	−4.73	230	−7.27	208	−7.87	187	−6.16
50	258	−4.81	233	−7.38	210	−7.99	189	−6.26
55	260	−4.94	235	−7.49	212	−8.10	191	−6.37
60	263	−5.06	238	−7.60	214	−8.21	193	−6.47
65	266	−5.19	240	−7.75	216	−8.33	194	−6.57
70	269	−5.27	242	−7.86	218	−8.44	196	−6.67
75	272	−5.40	245	−7.96	220	−8.56	198	−6.77
80	274	−5.53	247	−8.07	222	−8.67	200	−6.87

Table 4. Parameters of Excess Molar Heat Capacity for Binary Systems

		parameters		no. of data	AAD %[b]	
binary system	i	$a_{i,0}$	$a_{i,1}$	points	C_p^E	C_p
MEA (1) + H_2O (2)[a]	1	−146.65	0.4898	44	4.6	0.2
	2	22.60	−0.0790			
	3	10.88	−0.0532			
MDEA (1) + H_2O (2)	1	−134.61	0.4416	44	3.5	0.2
	2	49.21	−0.2633			
	3	−70.10	0.2149			
MEA (1) + MDEA (2)	1	−2.7567	−0.0896	44	0.4	0.1
	2	−12.655	0.0163			
	3	34.083	−0.1304			
		overall		132	2.8	0.2

[a] Data of Chiu and Li.[10] [b] AAD %: average absolute percentage deviation.

where $C_{p,i}$ is the molar heat capacity of the pure component i. In this study, the molar heat capacities of water, MEA, and MDEA were from Osborne et al.,[15] Chiu et al.,[6] and the values in Table 1, respectively. The obtained C_p^E values from eq 2 are also presented in Tables 2 and 3. The values of C_p^E for MDEA (1) + H_2O (2) exhibit negative values at both concentrations $x_1 = 0.6$ and 0.8 for temperatures 30 to 45 °C. For MEA (1) + MDEA (2), however, the values of C_p^E appear to be all negative values for the entire concentration and temperature regions.

To represent the compositional dependence of the excess molar heat capacity for a binary mixture, a Redlich−Kister equation was applied

$$C_{p,12}^E/(\mathrm{J\cdot mol^{-1}\cdot K^{-1}}) = x_1 x_2 \sum_{i=1}^{n} A_i (x_1 - x_2)^{i-1} \qquad (3)$$

The temperature dependence of A_i is assumed to follow the equation

$$A_i = a_{i,0} + a_{i,1}(T/\mathrm{K}) \qquad (4)$$

The number of terms (A_i), in eq 3, which should be used to represent the excess molar heat capacity depends on the degree of complexity of the binary systems. Plotting the values of $C_p^E/(x_1x_2)$ versus x_1 for three binary systems for temperatures 30 to 80 °C, curves were found for all three binary systems: MEA + H_2O, MDEA + H_2O, and MEA + MDEA. Thus, three terms in eq 3 are required for these three binary systems. The parameters $a_{i,0}$ and $a_{i,1}$ determined for MEA (1) + H_2O (2), MDEA (1) + H_2O (2), and MEA (1) + MDEA (2) are presented in Table 4. The data of $C_{p,12}^E$ for MEA (1) + H_2O (2) were taken from Chiu and Li.[10] The results (in AAD %) of the calculations for the excess molar heat capacity are 4.6%, 3.5%, and 0.4% for MEA (1) + H_2O (2), MDEA (1) + H_2O (2), and MEA (1) + MDEA (2), respectively. For the MDEA (1) + H_2O (2)

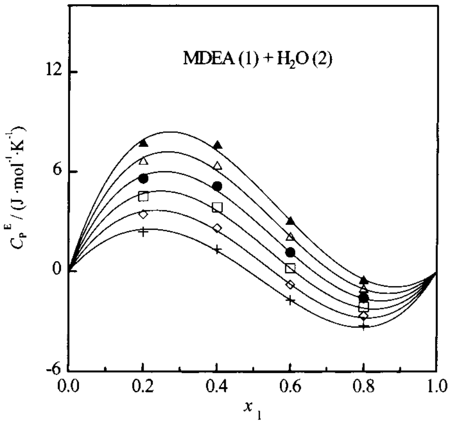

Figure 2. Excess molar heat capacity of MDEA (1) + H_2O (2): ▲, 80 °C; △, 70 °C; ●, 60 °C; □, 50 °C; ◇, 40 °C; +, 30 °C; lines, calculated using eq 3.

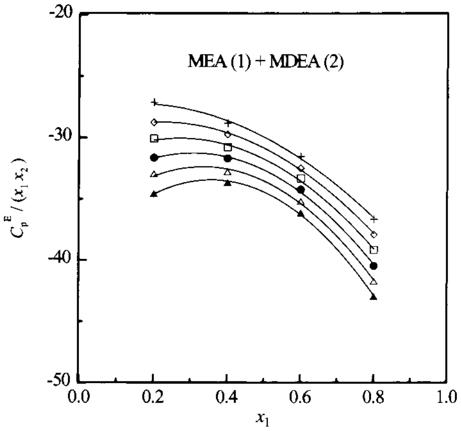

Figure 3. Plot of $C_p^E/(x_1x_2)$ versus x_1 for MEA (1) + MDEA (2): ▲, 80 °C; △, 70 °C; ●, 60 °C; □, 50 °C; ◇, 40 °C; +, 30 °C; lines, calculated using eq 3.

system, a comparison between the calculated and experimental values is shown in Figure 2. The values of $C_{p,12}^E$ show both positive and negative values for various mole fractions of MDEA, as shown in Figure 2. Figure 3 shows the plot of $C_p^E/(x_1x_2)$ versus x_1 for MEA (1) + MDEA (2); a quite good fit (AAD % = 0.4%) was obtained. Also shown in Figure 3, it indicates that the values of $C_{p,12}^E$ for MEA (1) + MDEA (2) cannot be satisfactory represented by using only two terms in eq 3.

On the basis of eq 3 and the parameters in Table 4, the calculation of C_p for MDEA (1) + H_2O (2) for mole fraction of MDEA ranging from 0.0432 to 0.1848 are performed and compared with the literature values of Hayden et al.[7] and Weiland et al.[11] The results are presented in Table 5. The AAD % values of the calculations are 1.8 and 3.7% for data of Hayden et al.[7] and Weiland et al.,[11] respectively. As shown in Figure 4, there is a good agreement between the calculated values and the data of Hayden et al.[7] for temperatures up to 75 °C. The calculated heat capacities are slightly lower than those values of C_p of Weiland et al.[11] at 25 °C.

The values of C_p of MEA (1) + MDEA (2) + H_2O (3) were also measured for temperatures ranging from 30 to 80 °C and for various mole fractions of water from 0.2 to 0.8. The results are presented in Tables 6−9. The calculated C_p^E values from eq 2 for MEA (1) + MDEA (2) + H_2O (3) are also presented in Tables 6−9. At the low mole fraction of water, that is, $x_3 = 0.2$, the values of C_p^E for MEA (1) + MDEA (2) + H_2O (3) systems exhibit all negative values for temperatures 30 °C to 80 °C, as shown in Table 9.

Table 5. Comparison of the Calculated and the Literature Values for Molar Heat Capacity of MDEA (1) + H_2O (2

	mole fraction	C_p/(J·mol^{-1}·K^{-1})			
t/°C	x_1	ref 7	ref 11	calc using eq 3	dev/%
25	0.0432	83.59		84.28	+0.8
	0.1313	105.77		102.26	−3.3
50	0.0432	84.44		86.03	+1.9
	0.1313	107.27		106.70	−0.5
75	0.0432	84.91		87.79	+3.4
	0.1313	110.37		111.14	+0.7
				AAD %[a]	1.8
25	0.0608		91.51	87.94	−3.9
	0.0915		97.76	94.24	−3.6
	0.1313		106.61	102.26	−4.1
	0.1848		116.5	112.82	−3.2
				AAD %[a]	3.7

[a] AAD %: average absolute percentage deviation.

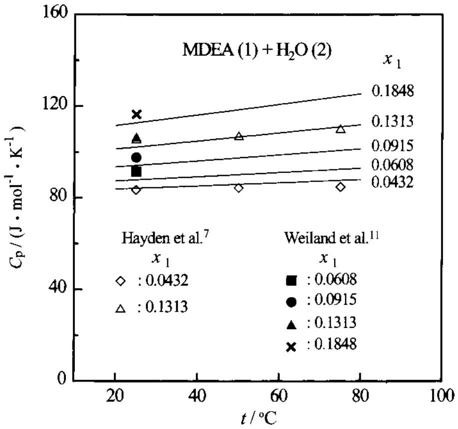

Figure 4. Molar heat capacity of MDEA (1) + H_2O (2) as a function of temperature: points, experimental data; lines, calculated using eq 3.

Table 6. Heat Capacities of MEA (1) + MDEA (2) + H_2O (3) for $x_3 = 0.8$

	molar heat capacity C_p and excess molar heat capacity C_p^E/(J·mol^{-1}·K^{-1})							
	x_1/x_2 = 0.04/0.16		x_1/x_2 = 0.08/0.12		x_1/x_2 = 0.12/0.08		x_1/x_2 = 0.16/0.04	
t/°C	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E
30	112	2.07	108	1.63	103	1.21	98.1	0.32
35	114	2.66	109	2.17	104	1.69	99.0	0.80
40	115	3.23	110	2.72	105	2.16	99.8	1.28
45	116	3.80	111	3.27	106	2.63	101	1.74
50	117	4.36	112	3.81	107	3.08	102	2.20
55	118	4.93	113	4.34	108	3.53	102	2.66
60	119	5.48	114	4.87	109	3.97	103	3.11
65	121	6.02	115	5.39	110	4.41	104	3.55
70	122	6.57	116	5.89	111	4.85	105	4.00
75	123	7.10	117	6.40	111	5.28	106	4.43
80	124	7.63	118	6.91	112	5.71	107	4.85

For a ternary system, the compositional dependence of the excess molar heat capacity is assumed to be given by

$$C_p^E/(\mathrm{J{\cdot}mol^{-1}{\cdot}K^{-1}}) = C_{p,12}^E + C_{p,13}^E + C_{p,23}^E + \{b_0 + b_1(T/\mathrm{K})\}x_1x_2x_3 \quad (5)$$

The ternary parameters, b_i, were introduced and determined from the excess molar heat capacities of ternary systems as $b_0 = -68.2623$ and $b_1 = 0.1451$. In eq 5, the compositions x_i in the ternary system were used in calcula-

Table 7. Heat Capacities of MEA (1) + MDEA (2) + H_2O (3) for $x_3 = 0.6$

	molar heat capacity C_p and excess molar heat capacity C_p^E/(J·mol^{-1}·K^{-1})							
	$x_1/x_2 =$ 0.08/0.32		$x_1/x_2 =$ 0.16/0.24		$x_1/x_2 =$ 0.24/0.16		$x_1/x_2 =$ 0.32/0.08	
t/°C	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E
30	147	1.41	138	0.70	128	−0.86	119	−1.05
35	149	2.13	140	1.36	129	−0.19	121	−0.40
40	151	2.83	141	2.03	131	0.48	122	0.26
45	153	3.55	143	2.69	133	1.14	124	0.92
50	154	4.24	145	3.31	134	1.79	125	1.55
55	156	4.95	146	3.97	136	2.45	126	2.19
60	158	5.65	148	4.60	137	3.07	128	2.82
65	160	6.32	150	5.24	139	3.71	129	3.45
70	162	7.02	151	5.88	140	4.35	130	4.08
75	164	7.70	153	6.48	142	4.98	132	4.69
80	166	8.37	155	7.11	143	5.61	133	5.30

Table 8. Heat Capacities of MEA (1) + MDEA (2) + H_2O (3) for $x_3 = 0.4$

	molar heat capacity C_p and excess molar heat capacity C_p^E/(J·mol^{-1}·K^{-1})							
	$x_1/x_2 =$ 0.12/0.48		$x_1/x_2 =$ 0.24/0.36		$x_1/x_2 =$ 0.36/0.24		$x_1/x_2 =$ 0.48/0.12	
t/°C	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E
30	179	−1.07	166	−2.02	153	−2.43	141	−2.11
35	182	−0.57	168	−1.50	155	−1.89	142	−1.68
40	184	−0.03	170	−0.97	157	−1.34	144	−1.25
45	186	0.47	172	−0.45	159	−0.80	145	−0.82
50	189	0.95	174	0.06	160	−0.29	147	−0.43
55	191	1.48	176	0.59	162	0.25	149	−0.00
60	193	1.96	178	1.09	164	0.77	150	0.41
65	195	2.46	180	1.59	166	1.30	152	0.82
70	198	2.95	182	2.11	168	1.83	153	1.22
75	200	3.45	185	2.60	170	2.32	155	1.62
80	202	3.93	187	3.10	172	2.84	156	2.02

Table 9. Heat Capacities of MEA (1) + MDEA (2) + H_2O (3) for $x_3 = 0.2$

	molar heat capacity C_p and excess molar heat capacity C_p^E/(J·mol^{-1}·K^{-1})							
	$x_1/x_2 =$ 0.16/0.64		$x_1/x_2 =$ 0.32/0.48		$x_1/x_2 =$ 0.48/0.32		$x_1/x_2 =$ 0.64/0.16	
t/°C	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E	C_p	C_p^E
30	211	−4.14	193	−5.94	176	−6.09	162	−3.84
35	214	−3.97	195	−5.74	178	−5.83	163	−3.61
40	217	−3.78	198	−5.49	180	−5.55	165	−3.36
45	219	−3.60	200	−5.25	182	−5.29	167	−3.11
50	221	−3.47	202	−5.02	184	−5.04	169	−2.88
55	224	−3.29	204	−4.77	186	−4.74	170	−2.64
60	227	−3.13	207	−4.55	188	−4.49	172	−2.41
65	229	−2.96	209	−4.32	190	−4.24	174	−2.19
70	232	−2.79	211	−4.08	192	−3.97	175	−1.92
75	234	−2.66	213	−3.89	194	−3.73	177	−1.71
80	237	−2.49	216	−3.66	196	−3.47	179	−1.48

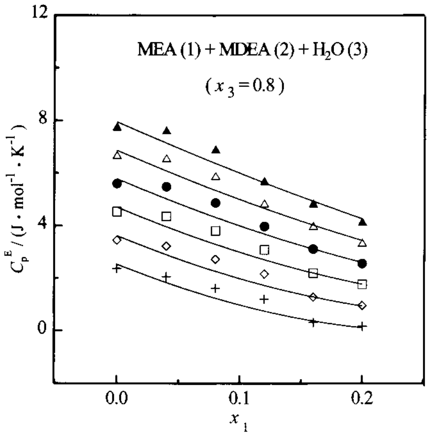

Figure 5. Excess molar heat capacity of MEA (1) + MDEA (2) + H_2O (3) as a function of x_1 at $x_3 = 0.8$: ▲, 80 °C; △, 70 °C; ●, 60 °C; □, 50 °C; ◇, 40 °C; +, 30 °C; lines, calculated using eq 5.

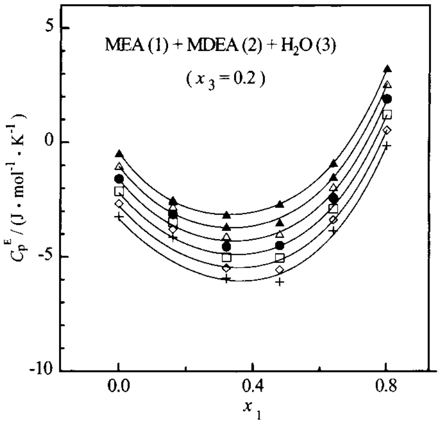

Figure 6. Excess molar heat capacity of MEA (1) + MDEA (2) + H_2O (3) as a function of x_1 at $x_3 = 0.2$: ▲, 80 °C; △, 70 °C; ●, 60 °C; □, 50 °C; ◇, 40 °C; +, 30 °C; lines, calculated using eq 5.

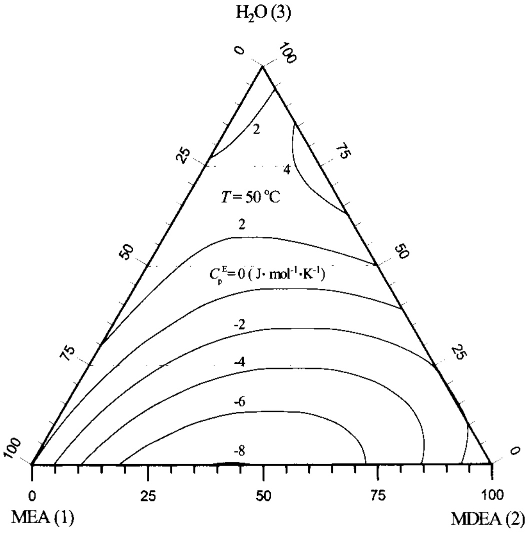

Figure 7. Excess molar heat capacity of MEA (1) + MDEA (2) + H_2O (3) at 50 °C: lines are constant excess molar heat capacities calculated using eq 5.

tion of the $C_{p,ij}^E$ on the the right-hand-side of eq 5. For any x_i approaching zero, eq 5 reduces to the expression for the binary system. Using eq 5 and the binary parameters $a_{i,0}$ and $a_{i,1}$ in Table 4 for the pairs MEA (1) + H_2O (3), MDEA (2) + H_2O (3), and MEA (1) + MDEA (2), the result (AAD %) for the calculation of the excess molar heat capacity for MEA (1) + MDEA (2) + H_2O (3) is 39%. Since some values of C_p^E are quite small, the corresponding AAD % for the C_p^E calculation may appear to be rather large while this deviation does not contribute to a large AAD % for the calculation of C_p. For a total of 176 data points, the overall AAD % for C_p calculations for MEA (1) + MDEA (2) + H_2O (3) is 0.5%, which is a quite satisfactory result. Thus, the excess molar heat capacity for MEA (1) + MDEA (2) + H_2O (3) is satisfactorily represented by eq 5.

Plots of C_p^E for MEA (1) + MDEA (2) + H_2O (3) for x_3 = 0.8 and 0.2 are shown in Figures 5 and 6, respectively. The solid lines in Figures 5 and 6 are calculated values from eq 5. As shown in Figure 5, the value of C_p^E for MEA (1) +

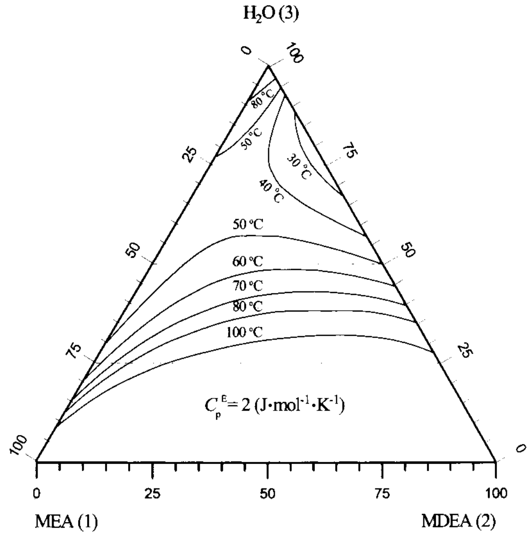

Figure 8. Excess molar heat capacity of MEA (1) + MDEA (2) + H_2O (3) at constant $C_p^E = 2.0$ J·mol^{-1}·K^{-1} for temperatures from 30 to 100 °C: lines are calculated using eq 5.

MDEA (2) + H_2O (3) increases as the temperature increases for a constant x_1; the value of C_p^E decreases as x_1 increases at a constant temperature. As in Figure 6, the value of C_p^E decreases as x_1 increases up to 0.4 for a constant temperature and increases as x_1 increases for $x_1 > 0.4$.

As in a triangle plot in Figure 7, various values of C_p^E for MEA (1) + MDEA (2) + H_2O (3) calculated by eq 5 at 50 °C are shown. It indicates that the positive values of C_p^E occur at the region with large values of x_3 and the negative values at the region with small values of x_3. The isotherms for C_p^E with a constant value of 2.0 J·mol^{-1}·K^{-1} for MEA (1) + MDEA (2) + H_2O (3) were calculated from eq 5 and are shown in Figure 8. As shown in Figure 8, the calculated isotherms distribute systematically along the path of the various concentrations; the lowest isotherm appears in the region with small values of x_1.

Conclusions

Heat capacities of aqueous mixtures of monoethanolamine with *N*-methyldiethanolamine were measured from 30 to 80 °C with a differential scanning calorimeter (DSC). The estimated uncertainty of the measured heat capacities is ±2% including the effect of up to 1% impurities in a substance. An excess molar heat capacity expression using the Redlich−Kister equation for the composition dependence is used to represent the measured C_p of aqueous alkanolamine solutions. For a total of 176 data points, the calculation of the molar heat capacity for MEA (1) + MDEA (2) + H_2O (3) gives an overall AAD % of 0.5%. The heat capacities of aqueous mixtures of monoethanolamine with *N*-methyldiethanolamine presented in this study are, in general, of sufficient accuracy for most engineering-design calculations.

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Received for review January 31, 2000. Accepted September 8, 2000. This research was supported by a grant, NSC 89-2214-E033-011, of the National Science Council of the Republic of China.

JE0000367