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Numerical Correlation of Isopiestic Data for Various Aqueous Solutions of Electrolytes

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A simple numerical relation was proposed to correlate the isopiestic data and has been tested over 136 systems with 2679 experimental points. The results show that this relation describes accurately the isopiestic data, and can be applied to thermodynamic calculations such as phase equilibria for the systems containing hydrocarbons, water, and salts, in order to simplify numerical calculation.

The purpose of this paper is to present a relationship designed to correlate the isopiestic data of aqueous saline solutions along with an application of this correlation able to simplify the numerical calculation of the thermodynamic properties of hydrocarbon−aqueous saline solution type systems.

Aqueous Saline Solutions and Isopiestic Data

A large amount of work has been done on the experimental and theoretical study of aqueous saline solutions. Multiphase systems involving water (with or without salts) and hydrocarbons are of increasing interest to the petroleum industry, as such systems are encountered either during oil production, where certain wells contain a not insignificant quantity of water, or during migration or transport of crude oils in the presence of water, or in underground gas storage in water-bearing reservoirs where the gas solubility in the aquifer depends on the degree of salinity of this aquifer.

Among the experimental measurements likely to be used to characterize solutions or to develop thermodynamic models, the following are of particular relevance: vapor pressure, freezing temperature depression, boiling temperature elevation, solubility, isopiestic data, enthalpic data, specific heat, and the electrode potential of solutions. The isopiestic method, which consists in bringing several solutions into equilibrium with a reference solution with known thermodynamic properties by means of the vapor phase in a closed vessel, allies simplicity of handling, accuracy of measurements, and a wide-range experimental domain.

In contrast with the preoccupations of geophysicists and geochemists, the aqueous phase does not represent the main center of interest in the petroleum industry. Aqueous saline solutions are often considered as pseudocomponents since the salts only exist in the aqueous phase. In this case, it is possible to extend studies carried out for a system composed of a specific aqueous saline solution, referred to as the reference solution, and hydrocarbons to all aqueous saline solution−hydrocarbon systems by means of an equivalent solution in the isopiestic sense. This is equivalent, for a given solution, to replacing it by the reference solution which will be in isopiestic equilibrium with it.

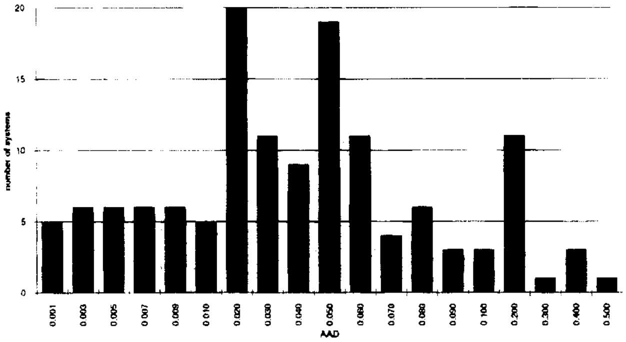

Figure 1. Distribution of the absolute average deviation (AAD) of 136 systems studied.

Isopiestic equilibrium of two solutions is attained when the chemical potentials of the solvent of each one of them are equal, i.e.

$$\mu = \mu^* \qquad (1)$$

in which the asterisk is used to denote the reference solution. Under given pressure and temperature conditions, the equilibrium is characterized in this way by

$$\mu^\circ_{\text{water}} + RT\ln(a) = \mu^\circ_{\text{water}} + RT\ln(a^*) \qquad (2)$$

or

$$a = a^* \qquad (3)$$

in which the quantity a designates activity.

The osmotic coefficient ϕ being defined by

$$\ln(a) = \phi\ln(x) \qquad (4)$$

it follows from the equality of a and a^* that

$$\phi = \phi^*\ln(x^*)/\ln(x) \qquad (5)$$

in which x represents the molar fraction of the solvent. In practice this relationship can be simplified as

$$\nu m\phi = \nu^* m^* \phi^* \qquad (6)$$

where ν is the number of ions generated on complete dissociation.

It is in principle possible to use a thermodynamic model, giving ϕ versus m, to determine the equivalent solution, i.e., m^*. When the quantity of water in different phases is variable, for example, during a flash, it is necessary to perform the calculation each time the composition of the aqueous phase changes. Given the complexity of the thermodynamic model, the number of operations becomes very high, and this represents a fastidious and time-consuming activity. We propose here a simple numerical relationship, correlating the experimental isopiestic data

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Table 1. Results of the Calculation for the Solutions with a Single Electrolyte[a]

solution studied	reference solution	eq	no. of points	T/K	m_{max}/ (mol kg^{-1})	A or A'	A''	B	AAD	ref
KCl	$MgBr_2$	7	8	373.45	6.9769	1.757 643		1.395 191	0.0448	*b*
KBr	$MgBr_2$	7	10	373.45	8.8075	1.647 941		1.436 174	0.0773	
$MgCl_2$	$MgBr_2$	7	15	373.45	6.3789	1.089 425		1.002 802	0.0063	
$CaCl_2$	$MgBr_2$	7	15	373.45	8.8690	0.912 190		1.270 852	0.1799	
$Lu_2(SO_4)_3$	NaCl	7	17	298.15	0.89631	0.879 692		0.747 327	0.0249	*c*
$ZnCl_2$	$CaCl_2$	7	70	298.15	13.134	1.775 805		0.952 485	0.1318	*d*
$ZnCl_2$	NaCl	7	16	298.15	4.2691	0.813 412		1.025 906	0.0320	
NaCl	$CaCl_2$	7	15	373.45	6.5979	1.498 401		1.209 658	0.0397	*e*
LiCl	$CaCl_2$	7	12	373.45	6.3015	1.432 548		1.096 533	0.0118	
$LiNO_3$	$CaCl_2$	7	32	373.45	15.2838	1.365 003		1.243 401	0.0817	
NaCl	KCl	7	12	298.15	3.926	0.964 445		0.937 944	0.0189	*f*
NaCl	$CaCl_2$	7	16	298.15	6.1580	1.600 018		1.234 446	0.0045	*g*
$MgCl_2$	$CaCl_2$	7	59	298.15	3.6424	1.017 622		0.919 965	0.0560	
H_2SO_4	$CaCl_2$	7	23	298.15	5.9623	1.351 831		1.073 797	0.0097	
KCl	NaCl	8	78	382.0−474.0	8.0750	1.212 059	−0.000 470	1.054 856	0.0187	*h*
$MgCl_2$	NaCl	8	78	382.0−474.0	3.5370	0.493 718	0.000 655	0.772 089	0.0316	
$CaCl_2$	NaCl	8	78	382.0−474.0	3.9568	0.348 432	0.000 971	0.824 439	0.0220	
$CaCl_2$	H_2SO_4	7	60	298.15	8.8254	0.661 833		0.989 668	0.0990	*i*
LiBr	LiCl	8	36	373.45−422.65	17.084	1.618 067	−0.000 923	0.834 831	0.2213	*j*
$LiNO_3$	LiCl	8	36	373.45−422.65	36.772	1.197 096	−0.001 166	1.247 049	0.1966	
$Ca(NO_3)_2$	LiCl	8	34	373.45−422.65	23.168	0.890 042	−0.000 493	1.126 714	0.1455	

[a] Total: 21 systems, 720 experimental points. [b] Fanghänel and Grjotheim (1990). [c] Rard (1990). [d] Rard and Miller (1989). [e] Grjotheim et al. (1988). [f] Thiessen and Wilson (1987). [g] Rard and Miller (1981). [h] Holmes et al. (1978). [i] Rard and Spedding (1977). [j] Braunstein and Braunstein (1971).

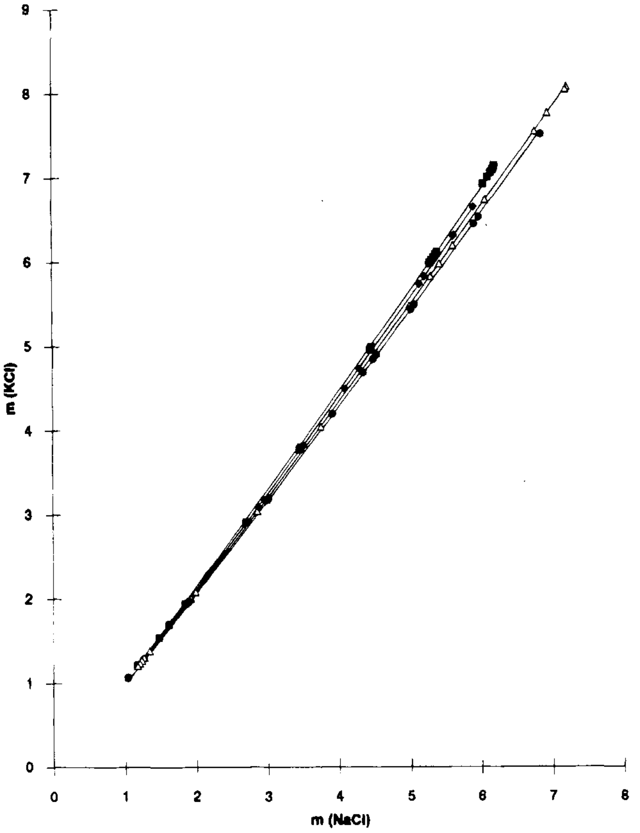

Figure 2. Experimental and calculated isopiestic molalities $m(MgCl_2)$ vs m(NaCl): (■) points at T = 382.0 K, (◇) T = 413.8 K, (△) T = 445.4 K, (●) T = 474.0 K, (solid line) curve calculated using eq 8.

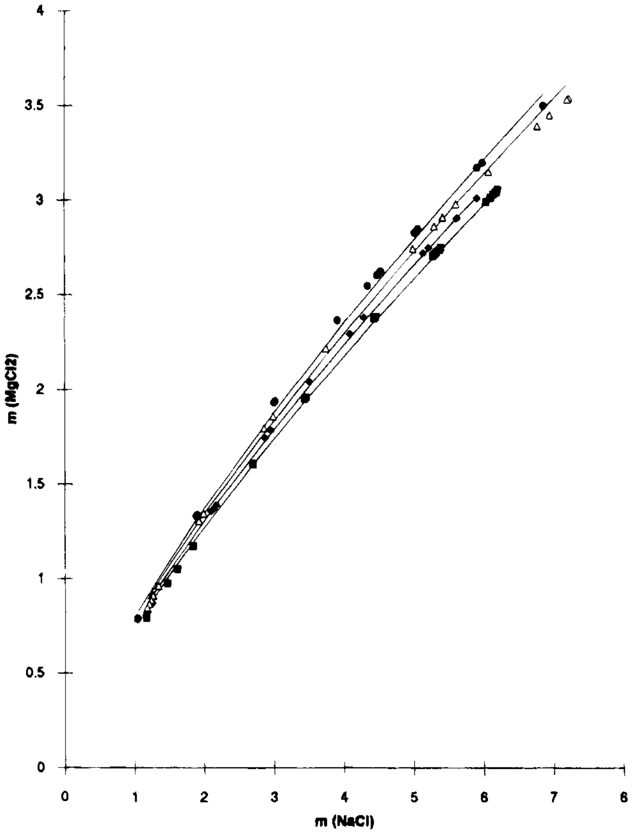

Figure 3. Experimental and calculated isopiestic molalities $m(MgCl_2)$ vs m(NaCl): (■) points at T = 382.0 K, (◇) T = 413.8 K, (△) T = 445.4 K, (●) T = 474.0 K, (solid line) curve calculated using eq 8.

m and m^* and designed to facilitate numerical calculation. This relationship, which is very convenient, reflects precisely all the available experimental data and can also be used for interpolation or extrapolation of isopiestic data.

Numerical Processing of Isopiestic Data

At any given temperature T, the concentration m^* of a reference solution and the concentration m of any other solution in isopiestic equilibrium with it are linked. On the basis of a compilation extended to numerous results provided by the literature (concerning various kinds of solutions containing one or two electrolytes, e.g., NaCl, KCl, LiCl, $CaCl_2$, $MgCl_2$, $Ca(NO_3)_2$, $ZnCl_2$, KBr, LiBr, H_2SO_4, $LiNo_3$, and $Lu_2(SO_4)_3$), we observed, by plotting m versus m^*, that there was a high degree of correlation between these two series of values. The relationship which appeared adequate to represent this dependency allows the

Table 2. Results of the Calculation for the Solutions with Two Electrolytes[a]

solution of 1 + 2	y_1	reference solution	eq	no. of points	*T*/K	m_{max}/(mol kg^{-1})	*A* or *A*′	*A*″	*B*	AAD	ref
KCl + $MgCl_2$	0.579 85	$CaCl_2$	7	14	373.45	6.5781	1.360 956		1.240 870	0.0175	*c*
	0.437 39			14	373.45	6.3459	1.301 971		1.190 190	0.0120	
	0.376 96			13	373.45	5.9861	1.279 208		1.160 298	0.0082	
	0.226 49			15	373.45	5.0377	1.192 714		1.084 790	0.0013	
	0.186 69			15	373.45	4.7796	1.165 599		1.063 281	0.0021	
	0.144 23			15	373.45	4.4954	1.134 046		1.038 063	0.0031	
	0.124 25			15	373.45	4.3593	1.118 299		1.024 773	0.0089	
	0.033 91			15	373.45	3.7422	1.030 810		0.972 416	0.0067	
KBr + $MgBr_2$	0.767 08	$CaCl_2$	7	6	373.45	7.5063	1.353 855		1.289 031	0.0338	*c*
	0.628 94			8	373.45	7.4885	1.305 871		1.246 334	0.0239	
	0.428 32			7	373.45	6.3897	1.231 718		1.159 272	0.0113	
	0.333 89			9	373.45	5.7512	1.183 545		1.113 933	0.0051	
	0.289 75			10	373.45	6.1923	1.162 701		1.087 527	0.0084	
	0.204 56			10	373.45	5.4922	1.107 223		1.042 111	0.0099	
	0.143 87			10	373.45	4.9915	1.060 851		1.008 756	0.0123	
	0.092 67			10	373.45	4.5716	1.015 224		0.980 568	0.0124	
	0.046 89			10	373.45	4.2002	0.970 343		0.955 182	0.0123	
$MgBr_2$ + $MgCl_2$	0.819 10	$CaCl_2$	7	11	373.45	4.2506	0.945 822		0.914 174	0.0236	*c*
	0.680 11			11	373.45	3.9159	0.940 304		0.930 054	0.0135	
	0.525 92			12	373.45	4.3497	0.962 282		0.917 894	0.0237	
	0.347 25			10	373.45	4.4122	0.975 515		0.917 684	0.0246	
	0.141 75			11	373.45	4.4829	0.992 801		0.917 343	0.0265	
KBr + KCl	0.817 17	$CaCl_2$		9	373.45	8.5850	1.417 652		1.360 123	0.0536	
	0.665 75			9	373.45	8.6451	1.423 183		1.362 513	0.0538	
	0.461 16			9	373.45	8.7521	1.430 932		1.367 111	0.0561	
	0.267 28			8	373.45	7.8047	1.466 686		1.346 946	0.0466	
KBr + $MgCl_2$	0.781 04	$CaCl_2$	7	12	373.45	8.0884	1.373 818		1.289 594	0.0491	*c*
	0.593 10			14	373.45	7.4992	1.319 358		1.231 007	0.0323	
	0.433 31			14	373.45	6.5639	1.264 635		1.167 571	0.0185	
	0.346 15			14	373.45	6.0289	1.226 313		1.129 716	0.0107	
	0.250 64			14	373.45	5.4167	1.175 697		1.084 708	0.0050	
	0.182 26			14	373.45	4.9611	1.132 904		1.049 550	0.0041	
	0.116 14			13	373.45	4.5157	1.085 019		1.014 131	0.0073	
	0.092 02			14	373.45	4.3481	1.065 506		1.000 447	0.0075	
	0.045 06			14	373.45	4.0220	1.022 472		0.974 619	0.0095	
KCl + $MgBr_2$	0.725 31	$CaCl_2$	7	14	373.45	7.3923	1.426 554		1.266 072	0.1345	*c*
	0.565 86			14	373.45	6.5575	1.359 858		1.211 696	0.0288	
	0.476 82			14	373.45	6.0543	1.315 302		1.177 065	0.0216	
	0.331 58			14	373.45	5.1713	1.223 596		1.112 712	0.0109	
	0.247 29			12	373.45	4.6497	1.154 836		1.076 160	0.0046	
	0.187 48			14	373.45	4.2670	1.106 141		1.043 618	0.0036	
	0.110 51			14	373.45	3.7764	1.031 155		1.004 360	0.0013	
	0.102 30			14	373.45	3.7251	1.022 842		1.000 178	0.0015	
	0.041 71			13	373.45	3.2113	0.954 721		0.969 055	0.0017	
NaBr + $LiNO_3$	0.000 00	$CaCl_2$	7	15	373.45	12.6058	1.342 536		1.256 873	0.0563	*d*
	0.126 74			13	373.45	12.6304	1.369 476		1.247 826	0.0516	
	0.249 95			15	373.45	12.6270	1.370 705		1.239 433	0.0542	
	0.374 60			15	373.45	12.5762	1.402 640		1.232 970	0.0582	
	0.471 43			15	373.45	12.5314	1.417 992		1.224 535	0.0522	
	0.627 98			15	373.45	12.3930	1.423 673		1.215 752	0.0466	
	0.750 14			15	373.45	12.2545	1.422 670		1.209 460	0.0400	
	0.875 29			15	373.45	12.1147	1.410 846		1.206 184	0.0310	
	1.000 00			10	373.45	11.5935	1.403 357		1.199 948	0.0400	
$NaNO_3$ + LiBr	0.000 00	$CaCl_2$	7	11	373.45	8.306 0	1.427 152		1.023 569	0.0953	*d*
	0.127 86			11	373.45	8.955 1	1.434 649		1.059 975	0.0763	
	0.249 92			11	373.45	9.725 4	1.425 365		1.108 508	0.0619	
	0.374 93			12	373.45	10.709 75	1.419 359		1.163 586	0.0466	
	0.501 67			12	373.45	11.986 4	1.418 437		1.225 786	0.0425	
	0.647 72			12	373.45	13.956 05	1.411 427		1.313 302	0.0646	
	0.749 38			9	373.45	11.385 55	1.421 802		1.376 536	0.0859	
	0.825 16			12	373.45	17.547 05	1.390 355		1.451 149	0.1182	
	1.000 00			9	373.45	15.990 4	1.335 005		1.642 352	0.1843	
$NaNO_3$ + LiCl	0.110 62	$CaCl_2$	7	6	373.45	6.530 8	1.421 546		1.127 914	0.0051	*e*
	0.206 00			8	373.45	6.777 3	1.416 396		1.157 067	0.0091	
	0.328 56			8	373.45	7.162 7	1.413 178		1.198 200	0.0199	
	0.449 71			8	373.45	7.635 5	1.410 614		1.245 190	0.0340	
	0.580 74			7	373.45	8.284 6	1.406 005		1.306 435	0.0543	
	0.716 71			8	373.45	9.163 3	1.403 913		1.381 163	0.0739	
	0.848 03			8	373.45	10.306 0	1.396 443		1.470 681	0.1008	
$NaNO_3$ + LiCl	0.000 00	$CaCl_2$	7	9	373.45	8.237 9	1.473 773		1.067 858	0.0380	*e*
	0.200 82			9	373.45	9.120 1	1.423 347		1.149 425	0.0167	
	0.402 86			9	373.45	10.379 3	1.375 989		1.248 034	0.0155	
	0.601 16			9	373.45	12.166 9	1.330 494		1.365 428	0.0404	
	0.799 51			9	373.45	14.896 6	1.286 206		1.510 435	0.0701	
NaCl + $LiNO_3$	0.124 30	$CaCl_2$	7	9	373.45	7.136 6	1.380 454		1.243 557	0.0454	*e*
	0.240 29			9	373.45	7.262 9	1.391 501		1.250 937	0.0453	
	0.352 25			8	373.45	7.369 0	1.406 963		1.254 530	0.0429	
	0.474 55			10	373.45	7.463 2	1.426 324		1.252 343	0.0485	
	0.595 09			10	373.45	7.526 2	1.435 064		1.253 611	0.0485	
	0.735 05			6	373.45	7.563 6	1.433 266		1.260 315	0.0471	
	0.842 54			10	373.45	7.560 4	1.453 797		1.247 544	0.0450	

Table 2 (Continued)

solution of 1 + 2	y_1	reference solution	eq	no. of points	T/K	m_{max}/(mol kg^{-1})	A or A'	A''	B	AAD	ref
$LiNO_3$ + LiCl	0.199 64	$CaCl_2$	7	17	373.45	10.1237	1.465 061		1.089 492	0.0576	*e*
	0.400 85			17	373.45	10.6468	1.435 043		1.127 789	0.0483	
	0.599 06			17	373.45	11.2394	1.406 229		1.167 842	0.0406	
	0.800 64			17	373.45	11.9582	1.369 207		1.215 604	0.0368	
	0.000 00			17	373.45	9.6330	1.501 250		1.050 421	0.0728	
$NaNO_3$ + NaCl	0.199 58	$CaCl_2$	7	6	373.45	7.1787	1.458 276		1.289 426	0.0434	*e*
	0.412 22			7	373.45	8.7105	1.430 349		1.368 775	0.0697	
	0.599 51			7	373.45	9.4051	1.421 804		1.430 708	0.0878	
	0.799 69			7	373.45	10.3200	1.408 686		1.508 148	0.1072	
$LiNO_3$ + $NaNO_3$	0.190 44	$CaCl_2$	7	8	373.45	12.3572	1.339 238		1.317 302	0.0702	*e*
	0.393 51			8	373.45	13.8925	1.336 877		1.387 370	0.0919	
	0.583 05			8	373.45	15.6716	1.324 371		1.464 287	0.1134	
	0.782 33			4	373.45	18.2597	1.236 170		1.595 612	0.0629	
LiCl + NaCl	0.302 3	NaCl	8	110	383.19−523.17	7.4333	0.939 617	0.000 173	0.965 930	0.0121	*f*
	0.456 5			110	383.19−523.17	7.1577	0.956 471	0.000 150	0.945 319	0.0161	
	0.575 1			110	383.19−523.17	7.0674	0.918 247	0.000 247	0.935 149	0.0228	
	0.714 2			110	383.19−523.17	6.8288	0.931 487	0.000 212	0.920 043	0.0251	
NaCl + $MgCl_2$	0.856 86	NaCl	7	36	298.15	4.5416	0.975 299		0.979 207	0.0033	*g*
	0.712 76			36	298.15	4.2440	0.941 940		0.958 623	0.0053	
	0.573 90			36	298.15	3.9309	0.903 661		0.936 762	0.0074	
	0.431 20			35	298.15	3.7582	0.856 426		0.911 202	0.0097	
	0.285 69			35	298.15	3.3383	0.797 111		0.883 105	0.0110	
	0.145 19			35	298.15	2.8881	0.725 945		0.852 625	0.0121	
NaCl + $MgCl_2$	0.856 86	$CaCl_2$	7	9	298.15	5.8940	1.527 955		1.215 578	0.0007	*g*
	0.712 76			12	298.15	5.5942	1.470 457		1.182 655	0.0012	
	0.573 90			13	298.15	5.2488	1.403 564		1.149 237	0.0008	
	0.431 20			11	298.15	4.8568	1.320 614		1.113 255	0.0009	
	0.285 69			13	298.15	4.5099	1.220 344		1.070 234	0.0008	
	0.145 19			16	298.15	4.2465	1.102 902		1.024 268	0.0008	
$LiNO_3$ + KNO_3	0.5034	LiCl	8	36	373.45−422.65	74.745	1.436 281	−0.002 043	1.534 690	0.3684	*b*
	0.5129			32	373.45−422.65	74.096	1.413 540	−0.002 020	1.538 991	0.3224	
$LiNO_3$ + $CsNO_3$	0.4942	LiCl	8	36	373.45−422.65	82.632	1.532 472	−0.002 339	1.581 482	0.3993	
	0.4783			34	373.45−422.65	84.460	1.552 239	−0.002 390	1.589 948	0.4098	
$Ca(NO_3)_2$ + $CsNO_3$	0.4976	LiCl	8	35	373.45−422.65	59.711	1.519 954	−0.001 841	1.381 215	0.1930	

[a] Total: 115 systems, 1959 experimental points. Remark: $y_1 = I_1/(I_1 + I_2)$, with I = ionic strength, $y_1 = m_1/(m_1 + m_2)$. [b] Braunstein and Braunstein (1971). [c] Fanghänel et al. (1991). [d] Voigt et al. (1990). [e] Voigt et al. (1990). [f] Holmes and Mesmer (1988). [g] Rard and Miller (1987).

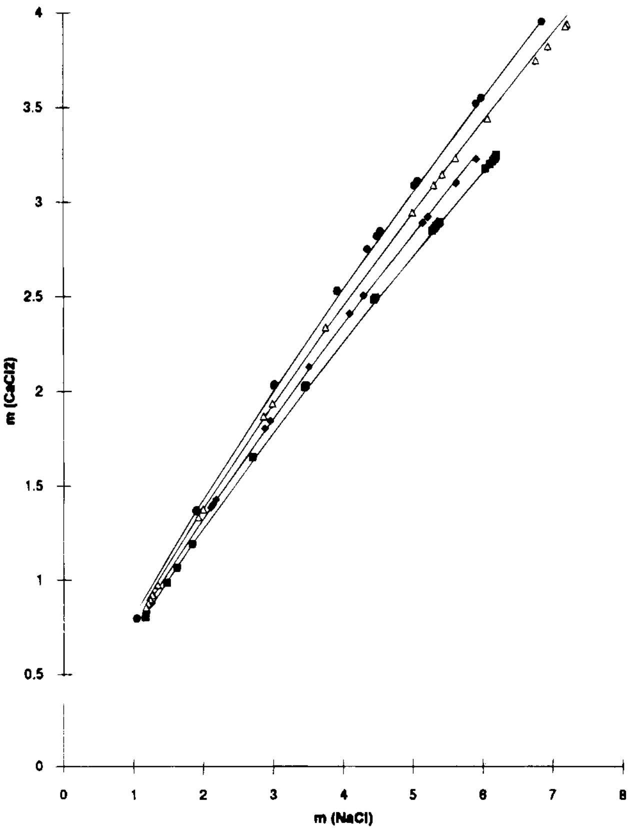

Figure 4. Experimental and calculated isopiestic molalities $m(CaCl_2)$ vs m(NaCl): (■) points at T = 382.0 K, (◇) T = 413.8 K, (△) T = 445.4 K, (●) T = 474.0 K, (solid line) curve calculated using eq 8.

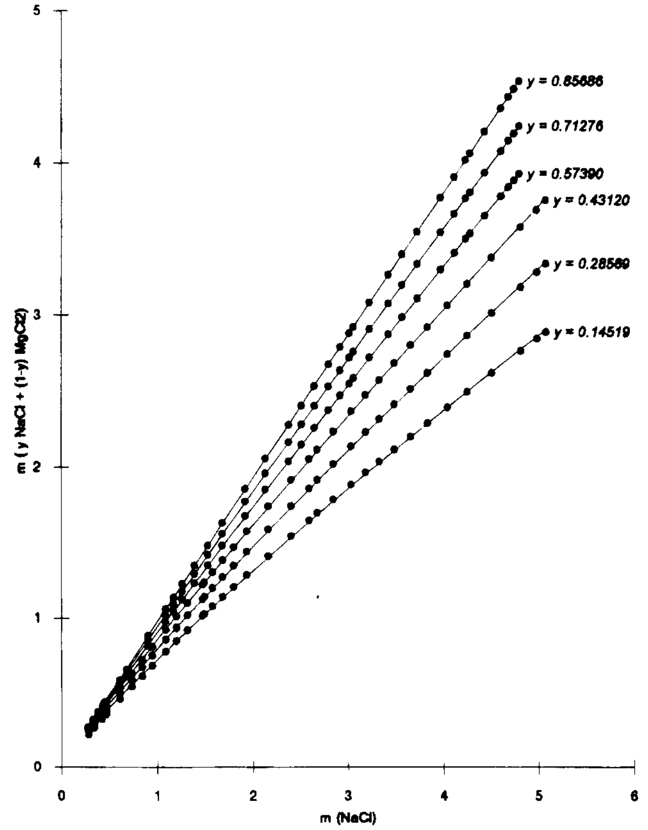

Figure 5. Experimental and calculated isopiestic molalities $m(y$ NaCl + $(1 - y)$ $MgCl_2)$ vs m(NaCl), where y is the ionic strength fraction: (●) experimental points, (solid line) curve calculated using eq 7.

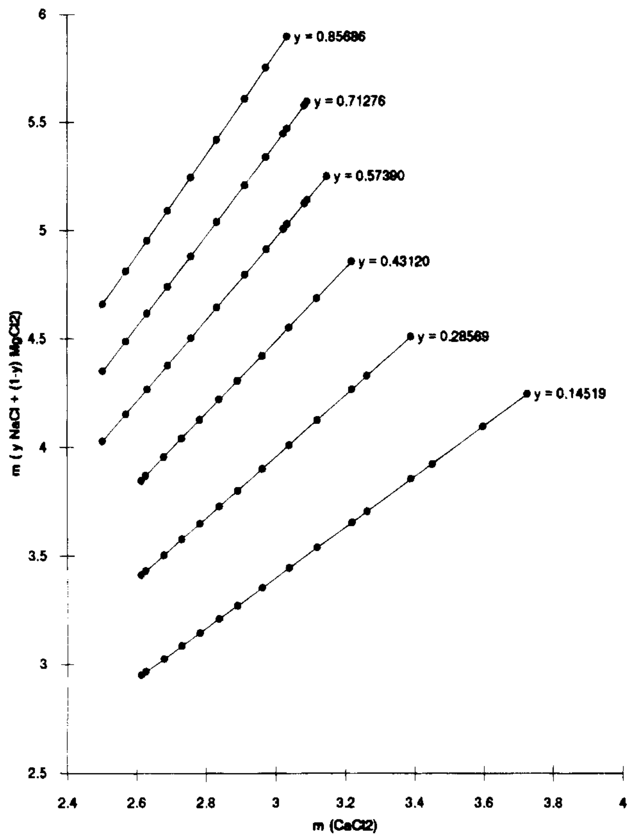

Figure 6. Experimental and calculated isopiestic molalities $m(y$ NaCl + $(1-y)$ $MgCl_2)$ vs $m(CaCl_2)$, where y is the ionic strength fraction: (●) experimental points, (solid line) curve calculated using eq 7.

following form:

$$m/(\text{mol kg}^{-1}) = A\{m^*/(\text{mol kg}^{-1})\}^B \quad (7)$$

in which A and B are numerical parameters. The first is a function of temperature, unlike the second, which may be assimilated to an invariant. Experimental data show that the temperature dependency of A is practically linear, and we therefore propose to adopt the following representation:

$$m/(\text{mol kg}^{-1}) = (A' + A''T/\text{K})\{m^*/(\text{mol kg}^{-1})\}^B \quad (8)$$

However, the influence of temperature proves small and the ratio between the two parameters B and A'' is found to be high, on the order of 1000:

$$|B/A''| \approx 1000 \quad (9)$$

Thus, when isopiestic data are only available along one single isotherm, values for other temperatures can be estimated to an acceptable degree of accuracy on the basis of the features presented above. This possibility is of great practical interest because of the considerable time required to perform the experiments necessary to determine a complete isotherm.

Results

Tables 1 and 2 summarize (1) all the information concerning the systems studied, i.e., 720 experimental points relating to 21 solutions with one single electrolyte and 1959 experimental points relating to 115 solutions with two electrolytes and (2) the results obtained by applying this procedure (numerical values of the parameters A', A'', B or A, B, as well as the mean absolute deviation).

The distribution of average absolute deviations is represented schematically in Figure 1 which makes it possible to verify the validity of the process on a wide variety of saline solutions. The absolute average deviation (AAD) is defined by

$$\text{AAD} = \frac{\sum_n |m_{i,\text{exptl}} - m_{i,\text{calcd}}|}{n} \quad (10)$$

As an example, Figures 2–6 illustrate the very good fit between experimental and calculated values obtained for a few one- or two-electrolyte systems.

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