computer-generated oblique views of the $P-x$ and $G^E_{123} - x$ surfaces, provide more graphic representations of the behavior of this ternary system.

Glossary

A_{ij}, A_{ji}	parameters in eq 1
B_{ij}	second virial coefficient
C	parameter in eq 2
G^E	excess Gibbs function, liquid phase
g	G^E/RT
P	total pressure
P_i^{sat}	vapor pressure of pure i
R	universal gas constant
T	absolute temperature
x	mole fraction, liquid phase
y	mole fraction, vapor phase
ΔP	difference between calculated and experimental pressures

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Enthalpies of Solution of Some Salts in Water

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Using the apparatus and procedure described in a recent report, with the energy equivalent of the calorimeter system determined with standard substances, enthalpies of solution in water at 298.15 K were measured for the following salts, as ΔH in kJ mol^{-1}: RbCl(c), in 1500 mol of H_2O, 17.225 ± 0.019; CsCl(c), in 800 mol of H_2O, 17.438 ± 0.014; CsBr(c), in 900 mol of H_2O, 26.146 ± 0.043; CsI(c), in 1500 mol of H_2O, 33.239 ± 0.030; Li_2CO_3(c), in 725 mol of H_2O, −14.800 ± 0.021.

1. Introduction

Measurements of the enthalpies of solution of several salts in water have been made, using the apparatus and procedure recently described, with the energy equivalent of the calorimeter being determined with standard substances (*12*).

The molecular weights used in this work are as follows (*3*): CsBr, 212.8094; CsCl, 168.3584; CsI, 259.8099; H_2O, 18.0152; Li_2CO_3, 73.8912; RbCl, 120.9208.

2. Experimental Section

Materials. The salts measured in this investigation are described as follows.

Rubidium chloride, National Bureau of Standards, Washington, D.C., Reference Material No. 984. The stated purity was 99.90 ± 0.02%. Prior to measurement, this material was dried at 873 K for 24 h.

Cesium chloride, Sample A, Harshaw Chemicals Co., Cleveland, Ohio. The purity was given as 99.96% (Gerald K. Johnson, Argonne National Laboratory, Argonne, Illinois, provided the sample and the assay of impurities) with the impurities being, in ppm, as follows: Mg, 5; Rb, 200; Na, 60; K, 5; Li, 4; Si, 30; Cs_2CO_3, 120. Prior to measurement, this material was dried at 645 K for 4 h.

Cesium chloride, Sample B, Alfa Inorganics, Beverly, Mass., Lot No. 87640. The purity was determined (Analysis provided by Gerald K. Johnson, Argonne National Laboratory, Argonne, Ill.) to be 99.99%, with the impurities being as follows, in ppm: Mg, 12; Rb, 10; Na, 5; K, 5; Cs_2CO_3, 56. Prior to measurement, this material was dried at 823 K for 24 h.

Cesium bromide, Alfa Inorganics, Beverly Mass., Lot No. 31672. The impurities were reported as follows, in ppm: Ba, 10; Sr, 5; Rb, 10; Na, 5; K, 5. Prior to measurement, this material was dried at 523 K for 8 h.

Cesium iodide, Alfa Inorganics, Beverly Mass., Lot No. 82473. The impurities were given as follows, in ppm: Ba, 50; Sr, 5; Na, 5; K, 5; Rb, 10. Prior to measurement, this material was dried at 523 K for 8 h.

Lithium carbonate, J. T. Baker, Ultrex, Phillipsburg, N.J. The purity was given as 99.98%, with total metallic impurities being 22 ppm. This material was heated at 523 K for 140 h. Within ±0.01% there was no loss in weight of the material from the 4th, to the 140th h. Prior to measurement, this material was heated again to 523 K for 22 h.

Potassium chloride, National Bureau of Standards, Standard Reference Material 918.

Tris(hydroxymethyl)aminomethane, National Bureau of Standards, Standard Reference Material 724a.

Following the drying procedure, the salts to be measured were placed in ampules in a dry atmosphere, except for lithium

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carbonate and cesium chloride, sample B, which were transferred in the air of the room.

Calorimetric Apparatus and Procedure. The calorimetric apparatus (LKB calorimeter and HP quartz thermometer assembly) and procedure, and methods of calculation, are described in the previous report (*12*).

For the experiments on lithium carbonate, the "reaction" period contained 80 observations of temperature at intervals of 14.5 s, while for the experiments with the other salts, the "reaction" period contained 30 observations at intervals of 14.5 s.

The method of calculating the energy equivalent of the standard calorimeter system, ϵ_s, is described in the preceding report (*12*). The energy equivalent of the actual calorimeter system is

$$\epsilon = \epsilon_S + \Delta\epsilon \tag{1}$$

where $\Delta\epsilon$ is evaluated as given by eq 2 of the previous report.

The enthalpy of solution of a given salt, in a given amount of water, is calculated with the following equation:

$$\Delta H = -(\epsilon(\Delta t_c) + q_v + q_{im})/(m/(\mathrm{MW})) \tag{2}$$

Here ΔH is the enthalpy of solution of the given compound for the final temperature of the "reaction" period; ϵ is the energy equivalent of the actual calorimeter system at the start of the experiment for the mean temperature of the "reaction" period; Δt_c is the corrected rise of temperature in the experiment; q_v is the energy absorbed on the vaporization of water to fill the space made available on the breaking of the ampule (usually barely significant); q_{im} is the correction for impurities in the sample; m and MW are, respectively, the mass of sample and the molecular weight of the compound measured.

Correction of the enthalpy of solution from the observed average final temperature of the experiments to 298.15 K is made with the following equations:

$$\Delta H(298.15\ \mathrm{K}) - \Delta H(T\ \mathrm{K}) = \Delta C_p(298.15 - T) \tag{3}$$

$$\Delta C_p = \phi_c - C_p \tag{4}$$

Here ϕ_c is the apparent molal heat capacity (*15*) of the dissolved salt at the given concentration in water and C_p is the heat capacity of the salt before its solution. For most chemical reactions, the value of ΔC_p is relatively small, with the sum of the heat capacities of the products tending to cancel the sum of the heat capacities of the reactants. In the case of the process of dissolving salts of strong electrolytes in water, however, the situation is quite different. Here the apparent molal heat capacity of the strong electrolyte in dilute solution is usually negative, with a resulting relatively large value of ΔC_p.

The values of the apparent molal heat capacity, ϕ_c, used to make the small correction involved (as per eq 4) in the present experiments, were taken (*13*) as follows, for the four alkali halide solutes in the given number of moles of water, in J K^{-1} mol^{-1}: RbCl, n = 1500, -118; CsCl, n = 800, -122; CsBr, n = 900, -128; CsI, n = 1500, -129.

The values used for the heat capacity of the salts measured were taken (*8*, *16*) as follows, rounded to three significant figures, in J K^{-1} mol^{-1}: RbCl(c), 51.5; CsCl(c), 52.4; CsBr(c), 51.9; CsI(c), 51.9; Li_2CO_3(c), 97.4.

In the case of the lithium carbonate, the correction required by eq 3 was not significant because the value of 298.15 − T was only 0.01 K. This was fortunate, as there are no extant values for the apparent molal heat capacity of lithium carbonate in solution in water and its estimate would have a large uncertainty.

The values of the enthalpy of dilution or the relative apparent molal enthalpy (*15*) of the solute in water at the given concentration, which were used to correct the results to the same concentration, were taken from ref 13. In the absence of other data on the enthalpy of dilution of lithium carbonate, a linear slope of the measured values of ΔH vs. the square root of the molality, by least squares, was used to correct the measured values to a common concentration.

The energy equivalent of the standard calorimeter system was determined as described in the preceding report. For the endothermic experiments, potassium chloride, NBS Standard Reference Material 918, was used as the standard substance, with its enthalpy of solution at 298.15 K and a final concentration of 0.05 m, taken as ΔH = (+17 536 ± 9) J mol^{-1}. For the exothermic experiments, tris(hydroxymethyl)aminomethane, NBS Standard Reference Material 724a, in 0.100 N hydrochloric acid solution, was used as the standard substance, with its enthalpy of solution at 298.15 K and a concentration of 5 g/dm^3 of solution, taken as ΔH = (-245.76 ± 0.26) J g^{-1}. As explained in the preceding report (*12*), the weighted mean value from a number of different investigations for the enthalpy of solution of tris(hydroxymethyl)aminomethane under specified conditions is numerically less than the NBS certified value by 0.09%. If this value were used instead of the value certified for the NBS Standard Reference Material, the values of the exothermic enthalpy of solution would be numerically less by 0.09%.

Whenever more reliable values become available for the standard substances used in the present investigation to determine the energy equivalent of the standard calorimeter system, the reported results can be changed in accordance with the new values.

3. Results

Present Investigation. Tables I–VI give the results of the experiments on the enthalpy of solution in water of the five salts: rubidium chloride; cesium chloride, sample A; cesium chloride, sample B; cesium bromide; cesium iodide; lithium carbonate. In these tables, n is the number of moles of water per mole of solute, and the other symbols are the same as in the preceding report.

Figures 1–5 give a plot of the results of the foregoing experiments as a function of the square root of the molality, $m^{1/2}$, with the results for the two samples of cesium chloride appearing on the same plot. The lines in Figures 1–4 were drawn using the relative apparent molal enthalpies from ref 13, and the line in Figure 5 is a linear least-squares fit of the data shown.

The overall uncertainty assigned to the final value for each compound measured was taken as twice the propagated standard deviation of the mean, including the uncertainties of the following: (a) value for the standard substance used to determine the energy equivalent of the standard calorimeter system, potassium chloride, s.d.m. = 0.025%, and tris(hydroxymethyl)aminomethane, s.d.m. = 0.05%; (b) experiments to determine the energy equivalent of the standard calorimeter system; (c) experiments to determine the enthalpy of solution.

Table VII summarizes the results obtained in the present investigation. The value for cesium chloride is the mean of the 12 experiments.

Previous Investigations. Table VIII gives a comparison of the existing data on the enthalpy of solution of four alkali halides measured, with the results of the present investigation.

For lithium carbonate, there appears to be only one recorded measurement of its enthalpy of solution (*2*), ΔH(298.15 K) = (-3.383 ± 0.030) kcal mol^{-1}, with the concentration being 1 mol of solute in 1900 mol of water. The present measurements were made to a concentration of 1 mole of solute in 725 mol of water, yielding ΔH(298.15 K) = (-3.537 ± 0.005) kcal mol^{-1}. Because of the lack of data on the enthalpy of dilution of lithium carbonate, precise comparison of the two values cannot be made at this time.

Table I. Results of Six Experiments on the Enthalpy of Solution of Rubidium Chloride in Water. For these Experiments, $\epsilon_s = 445.72 \pm 0.13$ (sdm) J K^{-1}

									At 298.15 K	
m_{sample}/g	n	10^4k/s^{-1}	10^4u/K min^{-1}	Δt/K	Δt_c/K	ϵ/J K^{-1}	q_v/J	$[\Delta H(298.15\ K) - \Delta H(T)]$/kJ mol^{-1}	$[\Delta H(n = 1500) - \Delta H(n)]$/kJ mol^{-1}	$\Delta H(298.15\ K)$/kJ mol^{-1} at $n = 1500$
0.36406 to 0.60097 0.44199^a	1113 to 1843 1553^a	1.215 to 1.411	4.18 to 4.97	−0.1170 to −0.1917 −0.1418^a	−0.11609 to −0.19225 −0.14128^a	443.94 to 447.24 445.71^a	0.011 to 0.021 0.015^a	−0.0003 to 0.0064 +0.0038^a	−0.0166 to +0.0115 +0.0011^a	17.2025 to 17.2439 17.2245^{a,b}

^{a} Mean. ^{b} Standard deviation of the mean ±0.0071.

Table II. Results of Six Experiments on the Enthalpy of Solution of Cesium Chloride, Sample A, in Water. For these Experiments, $\epsilon_s = 445.60 \pm 0.13$ (sdm) J K^{-1}

										At 298.15 K	
m_{sample}/g	n	10^4k/s^{-1}	10^4u/K min^{-1}	Δt/K	Δt_c/K	ϵ/J K^{-1}	q_v/J	q_{im}/J	$[\Delta H(298.15\ K) - \Delta H(T)]$/kJ mol^{-1}	$[\Delta H(n = 800) - \Delta H(n)]$/kJ mol^{-1}	$\Delta H(298.15\ K)$/kJ mol^{-1} at $n = 800$
1.01388 to 1.20952 1.12925^a	774 to 922 832^a	1.248 to 1.358	4.16 to 5.32	−0.2324 to −0.2783 −0.2591^a	−0.23463 to −0.28072 −0.26156^a	445.60 to 448.27 446.97^a	0.039 to 0.042 0.040^a	−0.085 to −0.101 −0.094^a	0.0007 to 0.0013 0.0009^a	0.0010 to 0.0042 0.0022^a	17.4323 to 17.4541 17.4398^{a,b}

^{a} Mean. ^{b} Standard deviation of the mean ±0.0032.

Table III. Results of Six Experiments on the Enthalpy of Solution of Cesium Chloride, Sample B, in Water. For these Experiments, $\epsilon_s = 445.72 \pm 0.13$ (sdm) J K^{-1}

										At 298.15 K	
m_{sample}/g	n	10^4k/s^{-1}	10^4u/K min^{-1}	Δt/K	Δt_c/K	ϵ/J K^{-1}	q_v/J	q_{im}/J	$[\Delta H(298.15\ K) - \Delta H(T)]$/kJ mol^{-1}	$[\Delta H(n = 800) - \Delta H(n)]$/kJ mol^{-1}	$\Delta H(298.15\ K)$/kJ mol^{-1} at $n = 800$
0.91290 to 1.28885 1.06633^a	724 to 1022 888^a	1.280 to 1.447	4.30 to 4.74	−0.2121 to −0.2992 −0.2477^a	−0.21146 to −0.29879 −0.24706^a	445.20 to 448.40 446.55^a	0.008 to 0.018 0.012^a	−0.088 to −0.124 −0.103^a	−0.0013 to +0.0008 −0.0002^a	0.0005 to 0.0050 0.0036^a	17.4150 to 17.4462 17.4367^{a,b}

^{a} Mean. ^{b} Standard deviation of the mean ±0.0047.

Table IV. Results of Four Measurements on the Enthalpy of Solution of Cesium Bromide in Water. For these Experiments, $\epsilon_s = 445.68 \pm 0.24$ (sdm) J K^{-1}

									At 298.15 K	
m_{sample}/g	n	10^4k/s^{-1}	10^4u/K min^{-1}	Δt/K	Δt_c/K	ϵ/J K^{-1}	q_v/J	$[\Delta H(298.15\ K) - \Delta H(T)]$/kJ mol^{-1}	$[\Delta H(n = 900) - \Delta H(n)]$/kJ mol^{-1}	$\Delta H(298.15\ K)$/kJ mol^{-1} at $n = 900$
1.23398 to 1.36024 1.31421^a	864 to 955 896^a	1.261 to 1.347	4.16 to 4.84	−0.3380 to −0.3728 −0.3605^a	−0.33977 to −0.37515 −0.36275^a	444.41 to 446.45 445.23^a	0.039 to 0.041 0.040^a	−0.0002 to +0.0014 +0.0005^a	−0.0007 to +0.0007 +0.0002^a	26.1184 to 26.1833 26.1457^{a,b}

^{a} Mean. ^{b} Standard deviation of the mean ±0.0146.

Table V. Results of Six Experiments on the Enthalpy of Solution of Cesium Iodide in Water. For these Experiments, $\epsilon_S = 445.60 \pm 0.13$ (sdm) J K^{-1}

									At 298.15 K	
m_{sample}/g	n	10^4k/s^{-1}	10^4u/K min^{-1}	Δt/K	Δt_c/K	ϵ/J K^{-1}	q_V/J	$[\Delta H(298.15\ K) - \Delta H(T)]$/kJ mol^{-1}	$[\Delta H(n = 1500) - \Delta H(n)]$/kJ mol^{-1}	$\Delta H(298.15\ K)$/kJ mol^{-1} at $n = 1500$
0.86283 to 1.14447	1258 to 1672	1.257 to 1.325	3.70 to 4.18	−0.2453 to −0.3265	−0.24720 to −0.32792	445.84 to 447.33	0.042 to 0.045	−0.0017 to −0.0001	0.0000 to 0.0000	33.2191 to 33.2672
0.96604[a]	1504[a]			−0.2752[a]	−0.27700[a]	446.36[a]	0.044[a]	−0.0008[a]	0.0000[a]	33.2387[a,b]

[a] Mean. [b] Standard deviation of the mean ±0.0076.

Table VI. Results of Six Experiments on the Enthalpy of Solution of Lithium Carbonate in Water. For these Experiments, $\epsilon_S = 446.58 \pm 0.05$ (sdm) J K^{-1}

									At 298.15 K	
m_{sample}/g	n	10^4k/s^{-1}	10^4u/K min^{-1}	Δt/K	Δt_c/K	ϵ/J K^{-1}	q_V/J	$[\Delta H(298.15\ K) - \Delta H(T)]$/kJ mol^{-1}	$[\Delta H(n = 725) - \Delta H(n)]$/kJ mol^{-1}	$-\Delta H(298.15\ K)$/kJ mol^{-1} at $n = 725$
0.42283 to 0.57276	715 to 972	1.231 to 1.319	3.81 to 4.80	0.2025 to 0.2731	0.18813 to 0.25600	447.93 to 449.62	0.014 to 0.016	0.000 to 0.000	−0.021 to +0.001	14.781 to 14.827
0.51299[a]	812[a]			0.2432[a]	0.22882[a]	448.77[a]	0.015[a]	0.000[a]	−0.008[a]	14.800[a,b]

[a] Mean. [b] Standard deviation of the mean ±0.007.

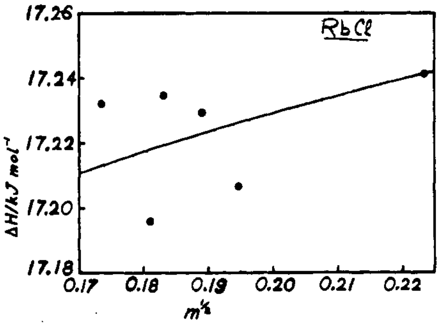

Figure 1. Plot of the values from the six experiments on the enthalpy of solution of rubidium chloride in water at 298.15 K, as a function of the square root of the molality.

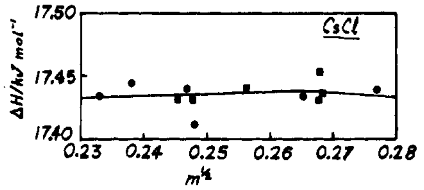

Figure 2. Plot of the values from the 12 experiments on the enthalpy of solution of cesium chloride in water at 298.15 K, as a function of the square root of the molality: sample A, ■; sample B, ●.

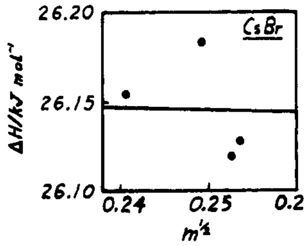

Figure 3. Plot of the values from the four experiments on the enthalpy of solution of cesium bromide in water at 298.15 K, as a function of the square root of the molality.

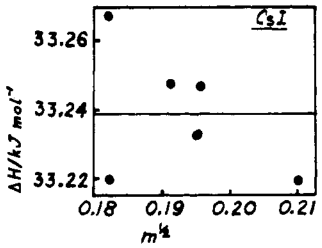

Figure 4. Plot of the values from the six experiments on the enthalpy of solution of cesium iodide in water at 298.15 K, as a function of the square root of the molality.

Table VII. Summary of the Results Obtained in the Present Investigation on the Enthalpies of Solution in Water for the Five Salts Measured

	Concn		ΔH(298.15 K)	
Compd	m	n	kJ mol^{-1}	kcal mol^{-1}
RbCl	0.03701	1500	+17.225 ± 0.019	+4.117 ± 0.005
CsCl	0.06939	800	+17.438 ± 0.014	+4.168 ± 0.003
CsBr	0.06168	900	+26.146 ± 0.043	+6.249 ± 0.010
CsI	0.03701	1500	+33.239 ± 0.030	+7.944 ± 0.007
Li_2CO_3	0.07656	725	−14.800 ± 0.021	−3.537 ± 0.005

4. Discussion

It had been our plan to measure, with this apparatus and method, a large number of salts, selected so as to (a) provide new data in place of scarce or nonexisting data, (b) provide useful information for chemically related compounds, and (c) obtain new data on enthalpies of dilution for those salts for which the existing data are meager or nonexistent, by measuring to

Table VIII. Comparison of the Existing Data on the Enthalpy of Solution in Water of the Four Alkali Halides Measured with the Data of the Present Investigation

Reference		ΔH(298.15 K)/kcal mol^{-1}			
No.	Year	RbCl $n = 1500$	CsCl $n = 800$	CsBr $n = 900$	CsI $n = 1500$
1	1904				8.003 ± 0.150
4	1906	4.085 ± 0.100	4.357 ± 0.100		
24	1910	4.184 ± 0.100			
5, 6	1911	4.157 ± 0.100	4.377 ± 0.100	6.468 ± 0.100	8.013 ± 0.100
7	1912	4.057 ± 0.050	4.134 ± 0.100		
10	1936	4.235 ± 0.050	4.177 ± 0.050		
11	1937			6.258 ± 0.100	
18	1956	4.169 ± 0.050	4.311 ± 0.100		
14	1958		4.237 ± 0.100		
17	1960	4.208 ± 0.075	4.237 ± 0.100		
22	1966	4.107 ± 0.030	4.257 ± 0.040	6.400 ± 0.030	
20	1969	4.128 ± 0.021	4.303 ± 0.022		7.954 ± 0.040
19	1970	4.193 ± 0.080	4.163 ± 0.040		
21	1973			6.304 ± 0.031	
9	1974	4.093 ± 0.006	4.167 ± 0.005		
Present work		4.117 ± 0.005	4.168 ± 0.003	6.249 ± 0.010	7.944 ± 0.007

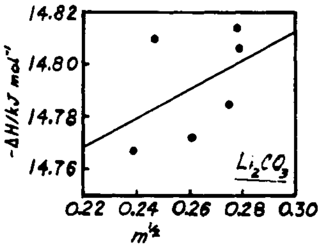

Figure 5. Plot of the values from the six experiments on the enthalpy of solution of lithium carbonate in water at 298.15 K, as a function of the square root of the molality.

significantly different concentrations. However, the development and detailed checking of the method for determining the energy equivalent of the calorimeter system with a standard substance required much more time and work than originally estimated. With all of the checking completed, the method is now arranged in a simple routine form, and it can be used easily by other investigators to obtain much needed data.

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