of chain length <5 but H_m^E was overestimated significantly for mixtures involving larger differences. Thus, some discretion must be exercised if the parameters obtained above are used for mixtures other than those studied in the present work.

Acknowledgment

We are indebted to P. J. D'Arcy and C. J. Halpin for technical assistance during this investigation.

Glossary

H_m^E	excess molar enthalpy, J mol^{-1}
h_j	coefficients in eq 1
h_{st}	molar interchange enthalpy per unit relative molecular surface area for contact between surfaces of types s and t
k	skewing factor in eq 1
q_i	relative molecular surface area of component i
x	mole fraction of 2,4-dimethylpentane

Greek Letters

$\alpha_{s,i}$	surface fraction of type s surface in component i

Registry No. 2,4-DMP, 108-08-7; hexane, 110-54-3; heptane, 142-82-5; octane, 111-65-9; dodecane, 112-40-3.

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Vapor Pressures of Binary (H_2O–HCl, –$MgCl_2$, and –$CaCl_2$) and Ternary (H_2O–$MgCl_2$–$CaCl_2$) Aqueous Solutions

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Vapor pressures of binary aqueous solutions containing HCl (1.262–13.435 mol/kg), $MgCl_2$ (1.051–4.104 mol/kg), and $CaCl_2$ (0.9568–5.002 mol/kg) were measured from 320 to 400 K. Furthermore, those of a ternary solution containing both $MgCl_2$ (1.057 mol/kg) and $CaCl_2$ (2.905 mol/kg) were also measured in the same temperature range. The apparatus for measuring vapor pressures was an all-glass Bourdon gauge and the precision of measurement was ±0.1 K or ±70 Pa, whichever was larger. The experimental data for the binary solutions were fitted to the Antoine type of equation adapted to concentrated aqueous solution and were in good agreement with the calculated results for all solutions. On the other hand, the data obtained for the ternary solution were compared with the values estimated by Teruya's method, and they both agreed well.

Introduction

Vapor pressure data of aqueous electrolyte solutions are important for the study of salt effects on vapor–liquid equilibrium. Data for a lot of aqueous electrolyte solutions are available in the literature (*1*, *2*) and the correlation methods (*3*–*5*) have also been reported. In this work, the vapor pressures of three binary (H_2O–HCl, –$MgCl_2$, and –$CaCl_2$) solutions and one ternary (H_2O–$MgCl_2$–$CaCl_2$) solution were measured by using an all-glass Bourdon gauge which was more accurate than that used previously for MgI_2 solution (*6*). The results were correlated by the Antoine type of equation.

Experimental Section

Apparatus and Procedure. Vapor pressure was measured by using an all-glass Bourdon gauge similar to that devised by Ewing (*7*). The form of the gauge is shown in Figure 1. It consisted of a thin-walled glass spiral (A), a pointer 25 cm in length (B), an outer tube 7 cm in diameter (C), a sample container with a volume of about 30 cm^3 (D), and a connecting tube of 0.25-cm inside diameter (F). The sensitivity of the gauge was 40 Pa. The volume occupied by the vapor phase was made as small as possible to minimize the change of liquid-phase composition due to distribution of water or HCl between both phases. For this apparatus in which the volume of the liquid phase was about 25 cm^3 and that of the vapor phase was about 10 cm^3, the change of the composition was negligible (within 0.1%). The temperature was measured within ±0.1 K with a platinum resistance thermometer (E) calibrated against a standard thermometer. A mercury manometer readable to 13.3 Pa and a cylinder of nitrogen gas were connected ahead of tube G. The temperature of the air bath (H) was controlled to within ±0.1 K.

Before each run, the extraneous gas in the solution and apparatus was removed by use of a vacuum pump for 15 min at room temperature. The vapor pressure of a sample was determined by measuring the pressure outside of the spiral which was equal to the vapor pressure inside of the spiral. The position of the pointer in balance of the pressure on both sides was observed by means of a cathetometer. To examine the consistency of the apparatus, the vapor pressures of pure water were measured and they agreed well with the literature values (*8*) (the maximum deviation was 213 Pa at 387.6 K and the mean deviation was 67 Pa).

Materials and Analysis. HCl, $MgCl_2$, and $CaCl_2$ used in this work were of guaranteed reagent grade from Wacô Chemicals Co., and all the reagents were used without further purification. Water, which was deionized and distilled, was used throughout the experimental work.

The composition of the sample was analyzed after vapor pressure measurement. HCl was analyzed by acid–base titra-

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Table I. Vapor Pressures of HCl Aqueous Solutions

T, K	p(exptl), kPa	p(calcd), kPa	dev, kPa
		1.262 mol/kg	
323.4	12.06	12.29	0.23
333.0	18.93	19.39	0.46
344.5	31.55	32.21	0.66
354.1	46.87	47.79	0.92
364.6	70.50	71.62	1.12
372.0	92.62	93.77	1.15
376.9	110.23	111.36	1.13
383.6	138.47	139.72	1.25
387.5	157.57	158.80	1.23
			mean dev = 0.90[a]
		1.782 mol/kg	
323.8	11.90	12.32	0.42
337.7	22.98	23.52	0.54
350.3	39.50	40.16	0.66
360.9	60.20	60.93	0.73
368.4	80.50	80.51	0.01
374.6	100.43	100.38	0.05
380.3	122.74	122.08	0.66
385.8	147.19	146.53	0.66
389.0	163.83	162.52	1.31
			mean dev = 0.56[a]
		3.882 mol/kg	
323.5	10.52	10.33	0.19
336.6	19.52	19.25	0.27
348.7	32.90	32.61	0.29
361.0	53.80	53.47	0.33
370.5	77.15	76.36	0.79
377.0	97.59	96.30	1.29
383.1	120.69	118.77	1.92
386.7	135.94	133.94	2.00
391.9	161.15	158.64	2.51
			mean dev = 1.07[a]
		6.644 mol/kg	
323.8	8.65	8.18	0.47
337.8	16.96	16.53	0.43
351.8	31.54	31.32	0.22
364.6	53.00	53.52	0.52
375.0	79.26	80.20	0.94
381.0	98.86	100.14	1.28
385.5	116.17	117.67	1.50
391.0	140.90	142.51	1.61
394.5	159.89	160.48	0.59
			mean dev = 0.84[a]
		8.567 mol/kg	
324.1	8.10	8.02	0.08
337.7	16.10	16.37	0.27
351.8	31.41	32.05	0.64
363.4	51.80	53.29	1.49
374.2	81.23	82.84	1.61
379.4	100.13	101.40	1.27
383.9	119.28	120.19	0.91
387.5	136.63	137.26	0.63
392.1	162.13	161.99	0.14
			mean dev = 0.78[a]
		10.042 mol/kg	
323.5	8.77	8.85	0.08
338.1	18.88	19.34	0.46
350.9	35.44	36.11	0.67
360.6	55.13	56.03	0.90
368.1	77.51	77.28	0.23
373.2	96.60	95.37	1.23
376.8	112.04	110.20	1.84
383.6	147.62	143.58	4.04
385.6	159.21	154.89	4.32
			mean dev = 1.53[a]
		13.170 mol/kg	
323.7	23.58	23.90	0.32
332.7	38.47	38.65	0.18
339.8	54.96	55.32	0.36
343.9	67.12	67.51	0.39
349.1	85.85	86.25	0.40
353.2	103.29	104.02	0.73
357.6	126.05	126.48	0.43
361.9	151.04	152.31	1.27
			mean dev = 0.51[a]
		13.435 mol/kg	
323.5	27.62	27.16	0.46
332.8	45.06	44.48	0.58
340.4	65.81	65.03	0.78
346.4	87.90	86.59	1.31
350.3	104.67	103.67	1.00
354.2	124.30	123.57	0.73
357.0	140.88	139.79	1.09
359.9	158.75	158.47	0.28
			mean dev = 0.78[a]

[a] Mean dev = $\sum_i |p_i(\text{calcd}) - p_i(\text{exptl})|/(\text{no. of data points})$.

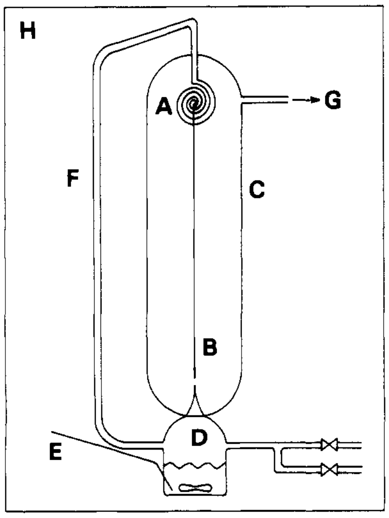

Figure 1. Vapor pressure apparatus: (A) glass spiral, (B) pointer, (C) outer tube, (D) sample container, (E) thermometer, (F) tube connecting A and D, (G) mercury manometer and cylinder of nitrogen gas, (H) air bath.

tion with 100 mol/m^3 sodium hydroxide solution. $MgCl_2$ and $CaCl_2$ were titrated with 10 mol/m^3 EDTA solution. In order to determine the molality of the electrolyte, the density of the sample was measured at 293.2 K by use of a density meter from Antone Paar Co., Ltd. The accuracy of analysis was within 0.3%.

Results and Discussion

Binary Solutions. Vapor pressures of binary aqueous solutions were determined to about 160 kPa for several concentrations of the solutes. The experimental results are given in Tables I–III. Figure 2 shows the relationship between log P and $1/T$ for $MgCl_2$ solution, and most of the points for each solute concentration lie on a line with slight curvature. This fact suggests that the experimental data may be correlated to the Antoine type of equation.

The original Antoine equation was modified to correlate the vapor pressures of aqueous electrolyte solutions. That is, the parameters A and B were assumed to be cubic functions of molality, m, of electrolyte:

$$\log p(\text{kPa}) = A(m) + B(m)/[T(\text{K}) + C] \quad (1)$$

$$A(m) = A_0 + A_1m + A_2m^2 + A_3m^3 \quad (2)$$

$$B(m) = B_0 + B_1m + B_2m^2 + B_3m^3 \quad (3)$$

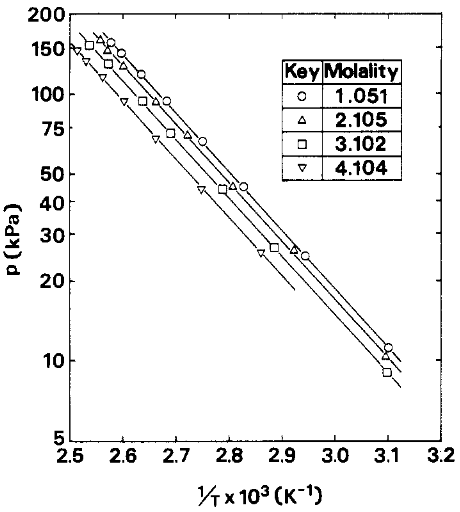

Figure 2. Relation between log p and $1/T$ for $MgCl_2$ aqueous solution.

Table II. Vapor Pressures of $MgCl_2$ Aqueous Solutions

T, K	p(exptl), kPa	p(calcd), kPa	dev, kPa
		1.051 mol/kg	
322.6	11.33	11.23	0.10
339.6	24.82	24.96	0.14
353.5	44.78	44.93	0.15
363.5	66.14	66.44	0.30
373.0	94.35	94.24	0.11
379.5	118.46	118.34	0.12
385.1	142.84	142.98	0.14
388.1	158.15	157.83	0.32
			mean dev = 0.17
		2.105 mol/kg	
323.1	10.30	10.32	0.02
342.9	25.95	26.02	0.07
356.3	45.38	45.51	0.13
367.4	69.96	69.82	0.14
375.4	93.47	93.37	0.10
384.2	126.64	126.52	0.12
388.8	147.80	147.38	0.42
391.1	159.37	158.83	0.54
			mean dev = 0.19
		3.102 mol/kg	
323.0	8.94	8.85	0.09
346.6	26.37	26.58	0.21
358.8	43.66	43.98	0.32
371.7	71.97	71.90	0.07
379.1	93.31	93.70	0.39
388.8	130.61	130.32	0.29
393.8	153.93	153.37	0.56
			mean dev = 0.28
		4.104 mol/kg	
349.7	25.26	25.21	0.05
363.8	44.52	44.66	0.14
375.4	68.93	68.93	0.00
384.1	93.52	93.60	0.08
390.5	116.13	116.08	0.05
394.9	134.46	133.98	0.48
397.8	146.90	146.98	0.08
			mean dev = 0.13

where parameters A_0–A_3 and B_0–B_3 are determined from the experimental data of the vapor pressure by the least-squares method and parameter C is that for pure water (the value of C was chosen to be −45.107 from the experimental results of the vapor pressure of water). Tables I–III give the vapor pressures calculated by eq 1–3, and Table IV summarizes the best-fit parameters in eq 2 and 3. The calculated vapor pressures were in good agreement with the experimental results for all of the solutions.

Table III. Vapor Pressures of $CaCl_2$ Aqueous Solutions

T, K	p(exptl), kPa	p(calcd), kPa	dev, kPa
		0.9568 mol/kg	
339.6	25.23	25.25	0.02
353.5	45.42	45.45	0.03
363.2	65.94	66.45	0.51
372.8	94.31	94.65	0.34
379.3	118.66	118.88	0.22
385.2	144.22	145.11	0.89
388.8	163.13	163.33	0.20
			mean dev = 0.31
		2.059 mol/kg	
323.1	10.62	10.54	0.08
343.1	26.61	26.70	0.09
355.9	45.32	45.46	0.14
367.3	70.21	70.47	0.26
375.4	94.84	94.46	0.38
382.5	121.16	120.72	0.44
386.7	139.89	138.90	0.99
390.5	157.99	157.24	0.75
			mean dev = 0.39
		3.084 mol/kg	
323.1	9.28	9.23	0.05
344.6	25.09	25.13	0.04
357.4	42.55	42.72	0.17
369.9	68.65	68.89	0.24
378.9	95.36	95.06	0.30
386.7	124.64	123.95	0.69
394.5	160.80	159.70	1.10
			mean dev = 0.37
		4.086 mol/kg	
322.7	7.73	7.64	0.09
348.8	25.49	25.81	0.32
363.1	45.51	46.20	0.69
373.5	67.85	68.34	0.49
382.5	93.56	94.05	0.49
388.5	115.49	115.28	0.21
394.4	139.74	139.87	0.13
398.5	159.63	159.38	0.25
			mean dev = 0.33
		5.002 mol/kg	
322.9	6.45	6.41	0.04
348.3	21.37	21.44	0.07
363.7	40.19	40.60	0.41
375.4	63.36	63.38	0.02
384.3	87.38	87.13	0.25
391.4	111.69	110.99	0.70
396.2	130.47	130.01	0.46
402.7	160.61	159.97	0.64
			mean dev = 0.33

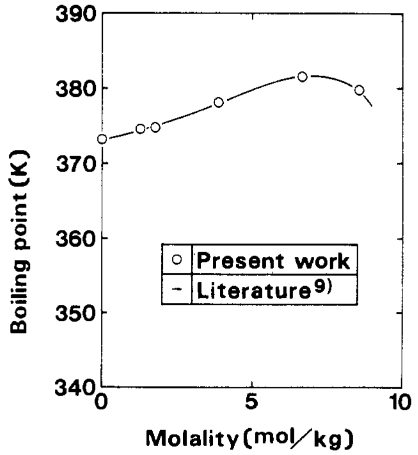

Figure 3. Boiling points of HCl aqueous solution.

To check the reliability of the vapor pressures, the normal boiling points (for HCl solution) and osmotic coefficients (for $MgCl_2$ and $CaCl_2$ solutions) were determined from the experimental results and compared with the literature. The comparison of the boiling points is shown in Figure 3, where the

Table IV. Parameters in Eq 2 and 3

system	A_0	A_1	A_2	10^3A_3	B_0	B_1	B_2	B_3
H_2O–HCl	7.15695	-1.45515×10^{-1}	3.43402×10^{-2}	−1.37832	−1690.97	47.3468	−13.8392	7.04648×10^{-1}
H_2O–$MgCl_2$	7.09562	-5.42893×10^{-2}	2.50527×10^{-2}	−4.02686	−1670.05	10.5198	−9.38085	1.07931
H_2O–$CaCl_2$	7.09453	-2.84935×10^{-2}	-2.76703×10^{-3}	1.48604	−1669.84	3.76316	−1.45301	-4.17236×10^{-1}

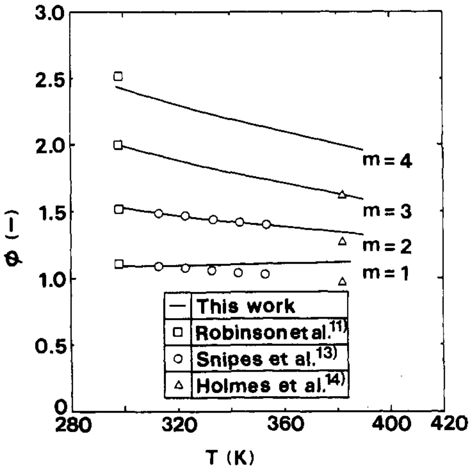

Figure 4. Osmotic coefficient as a function of temperature for $MgCl_2$ aqueous solution.

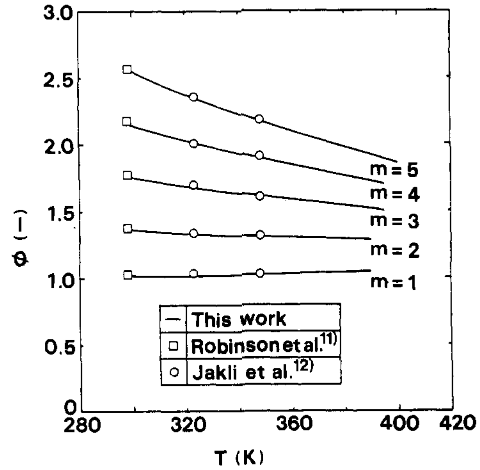

Figure 5. Osmotic coefficient as a function of temperature for $CaCl_2$ aqueous solution.

boiling points of this work were calculated by substituting 101.3 kPa for p in eq 1. The agreement was excellent. The osmotic coefficient, Φ, was computed by using the following equation:

$$\Phi = \frac{1000}{\nu m M_w}\left[\ln\left(\frac{p_w}{p_w^\circ}\right) + \frac{B_T(p_w - p_w^\circ)}{RT}\right] \quad (4)$$

where M_w is the molecular weight of water, ν is the number of moles of ions formed from 1 mol of electrolyte, p_w is the water vapor pressure of the solution and was calculated by eq 1–3, p_w° is that of pure water at the same temperature as the solution, and B_T is the second virial coefficient for water vapor. B_T is a function of temperature and was evaluated from the literature (*10*). The plots of Φ vs. T for various molalities are shown in Figures 4 and 5. In these figures the solid line represents the result obtained in this work, where the line below 323 K was determined by use of the extrapolated value of p_w. For $CaCl_2$ solution, Φ in this work agreed well with the literature (*11*, *12*). On the other hand, there is a discrepancy between the values in this work and the literature for $MgCl_2$ solution (*11*, *13*, *14*) (especially in 1 mol/kg). As pointed out by Holmes et al., the literature values vary widely and the additional measurement may be required to resolve these discrepancies.

Ternary Solution. Experimental results of the vapor pressures of the ternary aqueous solution containing $MgCl_2$ and $CaCl_2$ are given in Table V and were compared with the calculation using the method proposed by Teruya et al. (*5*). According to their method, the water vapor pressure of ternary solution can be calculated as follows:

$$\ln\frac{p_{w(mix)}}{p_w^\circ} = \frac{1}{I_t}\sum_k I_k \ln\left(\frac{p_{w(k)}}{p_w^\circ}\right) \quad (5)$$

where $p_{w(mix)}$ is the water vapor pressure of the ternary solution, $p_{w(k)}$ is that of the binary solution containing only the electrolyte k in the same ionic strength and temperature as the ternary mixture, p_w° is that of pure water at the same temperature as the ternary mixture, I_t is the total ionic strength of the ternary mixture, and I_k is the ionic strength of the electrolyte k in the ternary mixture. For $p_{w(k)}$ in this equation, the experimental data of this work were used. The calculated results are given in Table V and were in good agreement with the experimental ones.

Table V. Vapor Pressures of Ternary Aqueous Solution Containing 1.057 mol/kg $MgCl_2$ and 2.905 mol/kg $CaCl_2$

T, K	p(exptl), kPa	p(calcd), kPa	dev, kPa
322.5	8.38	7.63	0.75
348.5	26.29	25.64	0.65
363.2	48.11	46.63	1.48
373.0	68.89	67.44	1.45
383.1	98.50	96.45	2.05
388.4	117.77	115.39	2.38
393.5	140.00	136.42	3.58
398.2	162.12	158.49	3.63
			mean dev = 2.00

Acknowledgment

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Glossary

A, B, C	constants of the Antoine equation
A_0, A_1, A_2, A_3	parameters in eq 2
B_0, B_1, B_2, B_3	parameters in eq 3
B_T	second virial coefficient for water vapor, cm^3/mol
I	ionic strength, mol/kg
m	molality of electrolyte, mol/kg
M_w	molecular weight of water
p	vapor pressure, Pa
R	gas constant, cm^3·Pa/(mol·K)
T	temperature, K
Φ	osmotic coefficient
ν	number of moles of ions formed from 1 mol of electrolyte

Registry No. HCl, 7647-01-0; $MgCl_2$, 7786-30-3; $CaCl_2$, 10043-52-4.

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Vapor Pressures of the MgI_2–H_2O–I_2 System

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Vapor pressures for the MgI_2–H_2O–I_2 system were measured up to about 150 kPa. The mole ratio of H_2O to MgI_2 and that of I_2 to MgI_2 were varied from 10.862 to 42.432 and from 0.01 to 8.0, respectively. An empirical method was suggested for correlating the vapor pressures in the ternary system. The agreement between the experimental and calculated results was very good.

Introduction

The concentrating of aqueous solutions containing I_2 and MgI_2 is an important process in the magnesium–iodine cycle for thermochemical hydrogen production (*1*). In order to design this evaporation process, the vapor pressures of the MgI_2–H_2O–I_2 system were measured as the fundamental data.

Experimental Section

Vapor pressure data were obtained by means of the equipment and procedures described previously (*2*) except for two modifications: (a) the volume of the sample container was changed from 30 to 100 cm^3 in order to minimize the change of liquid-phase composition during the evacuation of the air from the system and (b) the liquid-phase composition was determined from the charged weights of the dried pure I_2 and MgI_2 aqueous solution of known concentration. The change of the composition owing to evacuation was within 0.4% and that owing to partition of I_2 and H_2O between both phases was within 0.4%. Therefore, the total error of the liquid-phase composition was judged to be within 0.8%.

Results and Discussion

The vapor pressures of the MgI_2–n_1H_2O–n_2I_2 system were measured up to about 150 kPa, where n_1 is the mole ratio of H_2O to MgI_2 and n_2 is that of I_2 to MgI_2. The experiments were divided into three groups: (a) MgI_2–$10.862H_2O$–n_2I_2, (b) MgI_2–$21.301H_2O$–n_2I_2, and (c) MgI_2–$42.432H_2O$–n_2I_2. The experimental results are given in Table I. Figure 1 shows the relationship between p and n_2 at various temperatures, where p is the vapor pressure of the MgI_2–n_1H_2O–n_2I_2 system smoothed by the Antoine equation: $\log p = A + B/(T + C)$. In this figure the y intercept is the vapor pressue of the MgI_2–n_1H_2O system (p_1) and can be calculated by the method proposed by the authors (*3*). Most of the points for each temperature and n_1 lie on a straight line and so p was fitted by the least-squares relation

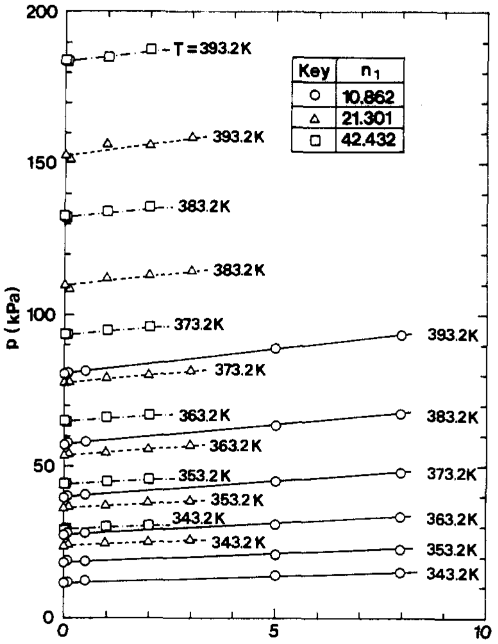

Figure 1. p as a function of n_2 for MgI_2–n_1H_2O–n_2I_2.

$$p = p_1 + \alpha n_2 \qquad (1)$$

where α is the slope of the straight line. Furthermore, the relationship between α and n_1 is shown in Figure 2 from 343.2 to 393.2 K. The values of α were correlated by using the empirical equation

$$\alpha = \alpha_0 + \alpha_1 n_1^{1/2} + \alpha_2 n_1 \qquad (2)$$

The parameters α_0, α_1, and α_2 at various temperatures are

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