Table I. Interaction Second Virial Coefficients (cm³/mol) for Sodium-Inert Gas Atom Pairs

T, K	NaHe	NaNe	NaAr	NaKr	NaXe
200	121.14	45.61	−12.31	−57.15	−146.18
300	101.83	44.94	11.83	−9.58	−58.51
400	89.50	43.18	21.74	11.82	−19.67
500	80.69	41.33	26.58	23.68	1.89
600	73.96	39.60	29.15	31.05	15.43
700	68.58	38.02	30.54	35.97	24.60
800	64.16	36.59	31.26	39.42	31.15
900	60.42	35.29	31.59	41.93	36.01
1000	57.22	34.12	31.68	43.80	39.73
1100	54.42	33.04	31.61	45.22	42.63
1200	51.96	32.06	31.44	46.32	44.94
1300	49.76	31.15	31.21	47.17	46.80
1400	47.79	30.31	30.93	47.84	48.33
1500	46.01	29.53	30.63	48.36	49.58
1600	44.38	28.80	30.31	48.77	50.62
1700	42.89	28.12	29.98	49.10	51.49
1800	41.52	27.48	29.65	49.35	52.23
1900	40.25	26.88	29.31	49.54	52.84
2000	39.08	26.31	28.98	49.69	53.37

listed in Table I it is now a straightforward task to evaluate $B(T)$ coefficients for any particular mixture of Na vapor and inert gases using eq 4. Subsequently, *PVT* properties for such mixtures can be easily derived by using a real gas as the model.

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Sechenov Salt-Effect Parameter[†]

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The units used to express the electrolyte and nonelectrolyte concentrations affect the numerical value of the salt-effect parameter. Conversion factors among the different modes of expression of the salt-effect parameter are derived and some typical numerical values are presented. The effects of random errors in the nonelectrolyte concentrations in water and the aqueous electrolyte solution are discussed. A set of evaluated salt-effect parameters are presented for up to 12 gases dissolved in aqueous sodium chloride and potassium hydroxide solutions at 298.15 K.

Introduction

The effect of an electrolyte on the solubility of a nonelectrolyte has been systematically studied since the pioneering work of I. M. Sechenov (J. Setschenow) (*1*) on the solubility of carbon dioxide in blood and in various aqueous electrolyte solutions between 1874 and 1892. A nonelectrolyte will have a different solubility in water and in an aqueous electrolyte solution mainly because of the effect of the electrolyte on the nonelectrolyte activity coefficient. The electrolyte may increase the nonelectrolyte activity coefficient (decrease the nonelec-

trolyte solubility, salt-out) or it may decrease the nonelectrolyte activity coefficient (increase the nonelectrolyte activity coefficient, salt-in). There are many reviews of the salt-effect experimental data and theories (*2–10*). The usual experimental measure of the salt effect is the Sechenov salt-effect parameter.

Sechenov (*11, 12*) proposed the empirical equation $\alpha = \alpha^0 \exp(-k/x)$ where α^0 and α are the Bunsen coefficients of the nonelectrolyte gas solubility in water and aqueous salt solution, x is the dilution, and k is the salt-effect parameter. Sechenov tested the equation by measuring the solubility of carbon dioxide in a concentrated (sometimes saturated) electrolyte solution, a number of dilutions of the solution, and pure water. He used the solubility in the most concentrated solution and water to establish the constant. He then calculated the solubility in the various solutions to compare with the experimental value. The agreement between equation and experiment was usually satisfactory. Many of Sechenov's original Bunsen coefficients for carbon dioxide in water and aqueous electrolyte solution agree within a few percent of modern values.

Rothmund (*13*) was apparently the first to put Sechenov's equation in the form commonly used today. In the notation of this paper that is

$k_{scc}/(dm^3\ mol^{-1}) =$

$\{1/(c_s/(mol\ dm^{-3}))\}\ \log\ \{(c_g^0/(mol\ dm^{-3}))/(c_g/(mol\ dm^{-3}))\}$ (1)

Table I. Conversion Factors for Salt-Effect Parameters for Several Aqueous Electrolyte Solutions

m^a/(mol kg^{-1})	c^b/(mol dm^{-3})	$10^2 F_{1c}$		$10^2 F_{2c}$		$10^2 F_{3c}$		$10^2 F_{1m}$		$10^2 F_{2m}$		$10^2 F_{3m}$	
		10 °C	40 °C	10 °C	40 °C	10 °C	40 °C	10 °C	40 °C	10 °C	40 °C	10 °C	40 °C
					Barium Chloride, BaCl$_2$								
0.0980	0.0977	1.09	1.27	2.35	2.37	1.25	1.09	1.09	1.26	2.34	2.34	1.25	1.08
1.516	1.499	1.35	1.49	2.36	2.39	1.01	0.90	1.29	1.41	2.20	2.26	0.97	0.85
					Sodium Chloride, NaCl								
0.173	0.172	0.68	0.80	1.56	1.58	0.88	0.77	0.68	0.79	1.56	1.56	0.88	0.77
4.278	3.958	0.89	0.96	1.58	1.60	0.69	0.64	0.82	0.88	1.46	1.46	0.63	0.60

m^a/ (mol kg^{-1})	c^b/ (mol dm^{-3})	$10^2 F_{1c}$		$10^2 F_{2c}$		$10^2 F_{3c}$		$10^2 F_{1m}$		$10^2 F_{2m}$		$10^2 F_{3m}$	
		10 °C	50 °C	10 °C	50 °C	10 °C	50 °C	10 °C	50 °C	10 °C	50 °C	10 °C	50 °C
					Magnesium Sulfate, MgSO$_4$								
0.722	0.721	0.14	0.27	1.55	1.57	1.41	1.30	0.14	0.27	1.54	1.54	1.40	1.28
1.352	1.339	0.30	0.39	1.54	1.56	1.24	1.17	0.30	0.38	1.53	1.53	1.23	1.14
					Sodium Sulfate, Na$_2$SO$_4$								
0.782	0.771	0.82	1.07	2.33	2.37	1.52	1.30	0.80	1.04	2.30	2.30	1.49	1.26
1.341	1.301	1.01	1.22	2.34	2.37	1.33	1.16	0.98	1.16	2.27	2.27	1.29	1.10
					Potassium Iodide, KI								
0.821	0.792	1.97	2.19	1.60	1.62	−0.37	−0.57	1.90	2.22	1.54	1.54	−0.36	−0.54
1.322	1.247	2.02	2.08	1.62	1.65	−0.40	−0.57	1.90	2.06	1.53	1.53	−0.30	−0.53

^{a} The molality values are independent of temperature. ^{b} The molarity values are for 10 °C.

where k_{scc} is the salt-effect parameter, c_s is the electrolyte concentration, and c_g^0 and c_g are the gas solubilities in water and electrolyte solution, respectively.

Over the years various workers have expressed the electrolyte concentrations in molarity, c/(mol dm^{-3}), or molality, m/(mol kg^{-1}), and the gas concentration in molarity, molality, or mole fraction, x, where the mole fraction is calculated with respect to the sum of moles of water, moles of all the ions, and moles of dissolved gas. Gas Bunsen coefficients and Ostwald coefficients are proportional to molarity and the Kuenen coefficient and aquamolality (or solvomolality) are proportional to molality.

Many workers assume that the salt-effect parameters calculated from the various concentrations differ negligibly if they differ at all. It is the purpose of this paper to show that there are small, but significant, differences in the values of the salt-effect parameter depending on which units of electrolyte and gas concentration are used.

Discussion

Most of the salt-effect parameters in the literature are one of four types. In addition to the k_{scc} defined in eq 1 there are

$$k_{smm}/(\text{kg mol}^{-1}) = \{1/(m_s/(\text{mol kg}^{-1}))\} \log \{(m_g^0/(\text{mol kg}^{-1}))/(m_g/(\text{mol kg}^{-1}))\} \quad (2)$$

$$k_{scx}/(\text{dm}^3 \text{ mol}^{-1}) = \{1/(c_s(\text{mol dm}^{-3}))\} \log (x_g^0/x_g) \quad (3)$$

$$k_{smx}/(\text{kg mol}^{-1}) = \{1/(m_s/(\text{mol kg}^{-1}))\} \log (x_g^0/x_g) \quad (4)$$

The k_{scc}/(dm^3 mol^{-1}) includes the Bunsen coefficient, α, and the Ostwald coefficient, L, ratios since

$$c_g^0/c_g = \alpha^0/\alpha = L^0/L$$

and the k_{smm}/(kg mol^{-1}) includes the Kuenen coefficients, s, and aquamolality, A, ratios since

$$m_g^0/m_g = s^0/s = A^0/A$$

By the use of the standard definitions of molarity, molality, and mole fractions the following relations can be derived among

the various formulations of the salt-effect parameter; a detailed derivation is available (14):

$$k_{smc} = (c_s/m_s)k_{scc} = (c_s/m_s)k_{scm} + F_{1m} = k_{smm} + F_{1m} \quad (5)$$

$$k_{scm} = k_{scc} - F_{1c} = (m_s/c_s)k_{smc} - F_{1c} = (m_s/c_s)k_{smm} \quad (6)$$

$$k_{scx} = (m_s/c_s)k_{smx} = (m_s/c_s)k_{smm} + F_{2c} \quad (7)$$

$$k_{smm} = k_{smx} - F_{2m} = (c_s/m_s)k_{scx} - F_{2m} \quad (8)$$

$$k_{smx} = (c_s/m_s)k_{scx} = (c_s/m_s)k_{scc} + F_{3m} \quad (9)$$

$$k_{scc} = k_{scx} - F_{3c} = (m_s/c_s)k_{smx} - F_{3c} \quad (10)$$

The three subscripts of the salt-effect-parameter symbol are as follows: first subscript, s for salt effect; second subscript, the electrolyte concentration symbol; and third subscript, the nonelectrolyte (gas) concentration symbol.

The F factors in the equations are

$$F_{1c} = (1/c_s) \log \{(\rho^0/\rho) \times (1000 + m_g M_g + m_s M_s)/(1000 + m_g^0 M_g)\}$$

$$F_{2c} = (1/c_s) \log \{(1000 + m_g M_g + \nu m_s M_s)/(1000 + m_g^0 M_g)\}$$

$$F_{3c} = (1/c_s) \log \{(1000\rho + (M_w - M_g)c_g + (\nu M_w - M_s)c_s)/(1000\rho^0 + (M_w - M_g)c_1^0)\}$$

$$F_{1m} = (1/m_s) \log \{(\rho^0/\rho) \times (1000 + m_g M_g + m_s M_s)/(1000 + m_g^0 M_g)\}$$

$$F_{2m} = (1/m_s) \log \{(1000 + m_g M_w + \nu m_s M_w)/(1000 + m_g^0 M_g)\}$$

$$F_{3m} = (1/m_s) \log \{(1000\rho + (M_w - M_g)c_g + (\nu M_w - M_s)c_s)/(1000\rho^0 + (M_w - M_g)c_g^0)\}$$

In the equations above ρ^0 and ρ are the water and aqueous electrolyte densities (gas saturated), respectively, and M_g, M_s, and M_w are the molecular weights of the gas, electrolyte, and water, respectively. The other symbols are as defined earlier. The terms in the F equations of $m_g M_g$, and $c_g M_g$ can be neglected for the sparingly soluble permanent gases. The

Table II. Salt-Effect Parameters

T/K	k_{smm}[a]/ (kg mol^{-1})	k_{smx}/ (kg mol^{-1})	k_{scc}/ (dm^3 mol^{-1})	k_{scx}/ (dm^3 mol^{-1})
	N_2 + LiCl + H_2O			
285.75	0.125	0.140	0.135	0.142
303.15	0.095	0.110	0.105	0.112
322.55	0.084	0.099	0.095	0.102
344.85	0.092	0.107	0.104	0.110
	N_2 + KF + H_2O			
285.75	0.134	0.149	0.160	0.156
303.15	0.100	0.115	0.126	0.121
322.55	0.086	0.101	0.113	0.107
344.85	0.084	0.099	0.112	0.106
	C_4H_{10} + $BaCl_2$ + H_2O			
285.75	0.250	0.273	0.275	0.281
303.15	0.210	0.233	0.233	0.241
322.55	0.180	0.203	0.184	0.197

[a] Experimental values taken from ref 16.

terms should be included for the soluble gases such as ammonia, sulfur dioxide, etc.

Table I gives some typical values of the F factors for several electrolyte solutions as a function of temperature and electrolyte concentration. All have been calculated by using the density data from the International Critical Tables (15). The factors are small but significant. They change slowly with temperature and concentration so interpolations can be made with confidence.

Table II gives values of the several forms of the salt-effect parameters for several gases and electrolytes. The original data, k_{smm}/(kg mol^{-1}), were taken from Morrison and Billett (16). The effect of the units used is apparent from the values in the table. There is even a difference in the temperature coefficient of the salt-effect parameter with the units used to express the concentrations.

Most of the measurements of the salt-effect parameter are made at an electrolyte concentration of 1 mol dm^{-3} or greater. There are reports of unusually large salt-effect parameters in the very dilute electrolyte range below 0.1 mol dm^{-3} (17-19). These claims need to be treated with some caution. At small electrolyte concentrations the difference in the gas solubility in water and in the electrolyte solution approaches the value of the random error of the gas solubility measurement. At this point, the maximum possible random error in the salt-effect parameter increases to unmanageable size (Figure 1). Thus, it would appear that to have reliable values of the salt-effect parameter at electrolyte concentrations below 0.1 mol dm^{-3} would require a precision of 0.1% or better in the gas solubility determination. Very few gas solubility determinations have been made with this precision until the past few years (20, 21) and
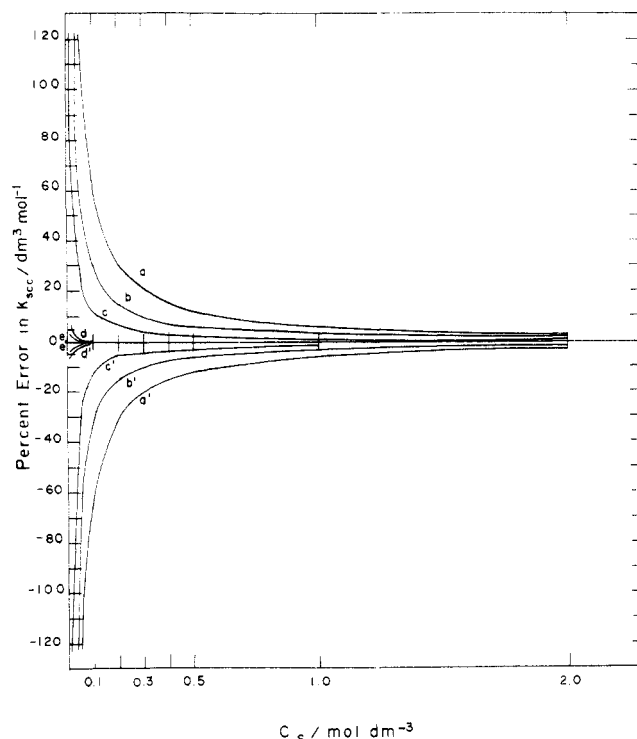

Figure 1. Percent error in the Sechenov salt-effect parameter, k_{scc}/(dm^3 mol^{-1}) vs. electrolyte concentration, c_s/(mol dm^{-3}). The curves were calculated for a gas + electrolyte + water system for which the solubility of the gas in water was 1 × 10^{-3} mol dm^{-3}, and the value of k_{scc} was 0.100 dm^3 mol^{-1}. The entire error was assumed to be due to error in the gas solubility. Percent error in the gas solubility: ±1, curves a and a'; ±0.5, curves b and b'; ±0.2, curves c and c'; ±0.1, curves d and d'; and ±0.05, curves e and e'.

to our knowledge no salt-effect parameters have been measured at this level of precision.

It is important to know if there is a larger than average salt-effect parameter for oxygen for certain lithium salts at room temperature (18) and for sodium bicarbonate and sodium hydroxide at high temperatures (19) as claimed. Confirmation of these claims will require experimental methods of greater accuracy and precision than those employed before.

The selection of the best value of a salt-effect parameter from the many choices in the literature is difficult. Table III summarizes the literature salt-effect parameter values for the hydrogen + sodium chloride + water system. The Bunsen coefficients measured by Braun appear to be considerably in error. The salt-effect parameters appear to be small when compared with the other data. The values obtained in 0.2 M NaCl could be in error because of the random error effect

Table III. Literature Salt-Effect Parameters, k_{scc}/(dm^3 mol^{-1}), for the Hydrogen + Sodium Chloride + Water System at 298.15 K

T/K	1894 (27)	1900 (28)	1952 (16)	1969 (29)	1971 (30)	1974 (31)	smoothed data
273.04						0.127[a]	0.108
278.2		0.103					0.107
283.2		0.095					0.106
283.97						0.101[b]	0.106
285.75			0.107				0.1055
288.15	0.093	0.093			0.107		0.105
289.55						0.115[a]	0.1045
293.2		0.090					0.104
295.34						0.106[b]	0.103
298.15		0.096		0.106			0.103
301.50						0.088,[a] 0.091[b]	0.102
303.15			0.103				0.101
322.55			0.099				0.096
323.15						0.100	0.096
344.85			0.088				0.091

[a] Measurements made in 0.2 M sodium chloride. [b] Measurements made in 0.5 M sodium chloride.

Table IV. Some Recommended Salt-Effect Parameters for Aqueous NaCl and KOH at 298.15 K and 1 mol dm^{-3}

	$k_{scc}/(dm^3 \, mol^{-1})$	
gas	NaCl	KOH
He	0.079	0.14
Ne	0.110	0.17
Ar	0.133	0.17
Kr	0.157	
Xe	0.160	
Rn	0.180	
H$_2$	0.103	0.140
N$_2$	0.134	
O$_2$	0.141	0.178
NO	0.158	
N$_2$O	0.128	0.133
C$_2$H$_6$	0.173	

NaCl	$k_{smx} = 0.977 \, (k_{scc} + 0.0075)$
	$k_{smm} = 0.977 k_{scc} - 0.0080$
	$k_{scx} = k_{scc} + 0.0075$
KOH	$k_{smx} = 0.992(k_{scc} + 0.0124)$
	$k_{smm} = 0.992 k_{scc} - 0.0030$
	$k_{scx} = k_{scc} + 0.0124$

discussed above. All the other data show some consistency. A linear regression of all except Braun's data and the three values from 0.2 M NaCl solutions gives the equation

$$k_{scc}/(dm^3 \, mol^{-1}) = 0.1760 - (2.46 \times 10^{-4})(T/K)$$

The smoothed data were calculated from the equation.

Table IV gives salt-effect parameters for up to 12 gases in aqueous sodium chloride and potassium hydroxide for use at 298.15 K and 1 M electrolyte. It is believed that this is a self-consistent set of useful tentative data for comparison with theories and for use in correlations. The reliability of the data is considered to be about ±5%.

Conclusion

One cannot say with certainty which of the modes of expressing the salt-effect parameter is the best. In the early literature the k_{scc} form was used almost exclusively. Recent workers who have made contributions which have increased our knowledge of salt effects have not been in agreement with each other. Ben-Naim (22) uses the k_{smc} form, and Masterton et al. (23) use the k_{scx} form along with their development of a scaled particle theory. There does seem to be some consensus favoring the k_{smm} form by Desnoyers et al. (24) and others using present-day statistical mechanical theories. This includes the Russian workers Krestov et al. (25), who use solvomolality, A or $m_g^{(s)} = m_g(M_{solvent}/M_w)$, which is proportional to molality, to express the gas solubility. Friedman (26) points out that a thermodynamically defined parameter which he designates as $k_{s,g} = -(\delta \ln m_g/\delta m_s)_{\mu_g}$ would be better than any

of the formulations discussed here. Friedman's parameter is numerically equal to the $k_{smm}/2.303$ form in the limit of infinite dilution. There are still other workers who believe that the k_{scc} form best describes the salt effect on very soluble gases at high pressure.

Although the paper has been written from the view of one interested in gas solubility, the same considerations should apply to salt effects on the solubility of solids. The solubility of liquids may be more complicated because of distribution between the two liquid phases.

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