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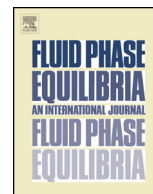
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Effect of solvation with salting effect on solubilities of fluorocarbons in alcohols

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ABSTRACT

In our previous work, we found that salting-out effect of a fluorocarbon in an alcohol–NaOH solution brought solubility decrease which could be expressed in terms of the Setchenov coefficient. In this work, a group contribution method for the Setchenov coefficients was made, for dichlorodifluoromethane (CFC-12), pentafluoroethane (HFC-125), difluoromethane (HFC-32), 1,1,1,2-tetrafluoroethane (HFC-134a) and 1,1-difluoroethane (HFC-152a) in methanol–NaOH and ethanol–NaOH solvents. Moreover, as a simple model, we derived the relationship between the Setchenov coefficients and solvation number of alcohols around a NaOH molecule. Therefore, once information of the Setchenov coefficients was obtained, the solvation numbers could be predicted based on the model. Prediction of the Setchenov coefficients for VOC solubilities in water–salt system is possible with the new method.

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1. Introduction

Fluorocarbons containing chlorine atoms deplete the ozone layer, so that restrictions on production and transportation of chlorofluorocarbons (CFCs) were decided by the Montreal Protocol (1987) and hydrochlorinated fluorocarbons (HCFCs) are expected to be final in 2020. Massive wastes of CFCs and HCFCs will be treated in the near future which will be a serious problem.

We proposed a simple decomposition process of chlorinated fluorocarbons in alcohol containing NaOH at room temperature and atmospheric pressure with or without UV irradiation to produce other fluorocarbons, unsaturated compounds or fluoroethers with sodium chloride [1]. Especially, fluoroethers are expected to be the third generation refrigerants, because it decomposes rapidly in the air.

The dechlorination of the CFC or HCFC occurs after dissolving the gas in an alcohol–NaOH solution with or without UV irradiation. Being much slower than mass transfer rate, the reaction rate is governed by the solubility of fluorocarbons in alcohol–NaOH solutions. To design a reactor, solubility is the most important factor.

To meet the requirement, we measured the Henry's law constants of fluorocarbons in alcohols at atmospheric pressure [2,3].

However, we found that solubility of a fluorocarbon in an alcohol–NaOH solvent decreased with NaOH concentration in a solution, i.e. salting-out effect. Reaction rate is proportional to both the concentration of the dissolved fluorocarbon and NaOH concentration in a solution [4]. The increase of NaOH brings the accelerator-brake effects that the one decreases due to salt-effect and the other increases. It is very interesting to know how concentration of NaOH affects the reaction rate. Therefore, we need the data on salting effects.

In a previous paper [5], we measured the solubilities of dichlorodifluoromethane (CFC-12, CCl₂F₂), pentafluoroethane (HFC-125, C₂HF₅), difluoroethane (HFC-152a, C₂H₄F₂), difluoromethane (HFC-32, CH₂F₂), 1,1,1,2-tetrafluoroethane (HFC-134a, C₂H₂F₄) in NaOH–methanol, NaOH–ethanol and NaOH–1-propanol solvents, ranging from 283 to 313 K at atmospheric pressure. As a result, the solubility of fluorocarbons decreases exponentially with the concentration of NaOH due to salting effects. This is known as the Setchenov relation and the Setchenov coefficients are typically independent of temperature.

In this work, we estimated the Setchenov (salting effect) coefficients in these systems with a group contribution method and the estimated values agreed very well with the experimental data. Moreover, according to a simple model, the Setchenov coefficient

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is related with solvation number of alcohols, and is expressed with a function of concentration of NaOH and partial pressure of fluorocarbon. Moreover, the group contribution method of the Setchenov coefficients were tried to apply this model to the water–VOC–salts systems.

2. Setchenov coefficient

The concentration of a fluorocarbon in an alcohol solution at temperature T , C_i^0 [mol/L] is expressed as the Henry's law equation:

$$C_i^0 = \frac{Py_i}{H_{ij}^0} \quad (1)$$

where P is the total pressure (= atmospheric pressure), y_i is the mole fraction of a fluorocarbon i in a vapor phase, superscript 0 means without NaOH and H_{ij}^0 is the Henry's law constant of a fluorocarbon i in an alcohol j solution without NaOH at temperature T [K].

When NaOH is added, the concentration of a fluorocarbon C_i [mol/L] in an alcohol–NaOH solution at experimental temperature T and at partial pressure Py_i is described as follows:

$$C_i = \frac{Py_i}{H_{ij}} \quad (2)$$

Since the addition of NaOH to an alcohol solvent has no effect on the partial pressure of fluorocarbon in the gas phase at the same experimental temperature and the same initial partial pressure [5], the following relationship is obtained from Eqs. (1) and (2),

$$\frac{C_i}{C_i^0} = \frac{H_{ij}^0}{H_{ij}} \quad (3)$$

Effects of electrolyte concentration on the solubilities of gases in solutions are known as salting-out or salting-in effects. The influence of the dissolved salt in the electrolyte solution is given by the Setchenov relation described as an exponential function of electrolyte concentration [6]. Although NaOH does not dissociate in alcohols, the effect of NaOH concentration on the solubility of 1,1,1,2-tetrafluoroethane (HFC-134a) in ethanol–NaOH at different temperatures is shown in Fig. 1. It shows that salting effect vary much less with temperature, however the solubilities of fluorocarbons decreased exponentially with NaOH concentration. Relation between solubility due to salting effect and C_{NaOH} is expressed as follows,

$$\frac{C_i}{C_i^0} = \exp(-h_{ij}C_{\text{NaOH}}) \quad (4)$$

where h_{ij} is the Setchenov coefficient (salting effect coefficient) of component i in alcohol j expressed in L/mol. It shows that the values of h_{ij} are proper for a system independent of temperature.

The Setchenov coefficients h_{ij} for the systems of dichlorodifluoromethane (CFC-12, CCl_2F_2), pentafluoroethane (HFC-125, C_2HF_5),

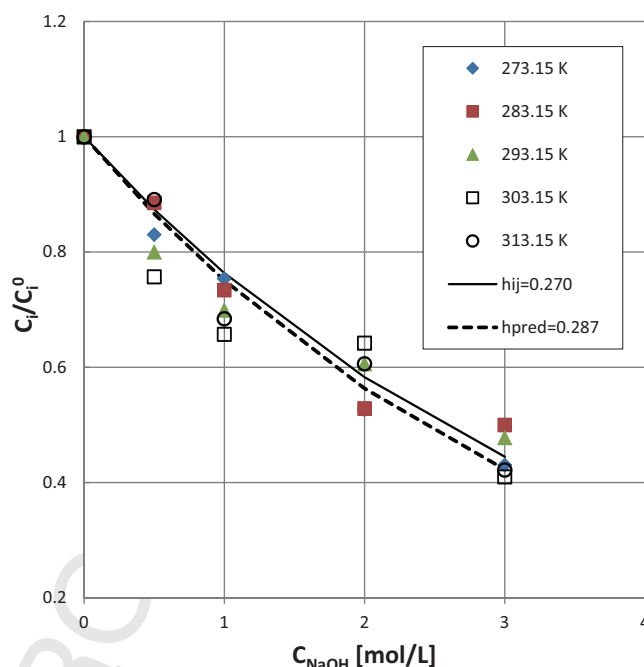


Fig. 1. Effect of NaOH concentration on solubility of HFC-134a in ethanol with changing temperature at atmospheric pressure of a solute gas ranging from 273.15 to 313.15 K: experimental [5].

difluoromethane (HFC-32, CH_2F_2), 1,1,1,2-tetrafluoroethane (HFC-134a, $\text{C}_2\text{H}_2\text{F}_4$) in methanol–NaOH, ethanol–NaOH solvents are listed in Table 1 in [5]. Table 1 shows the values of h_{ij} independent of temperature.

3. Estimation of Setchenov coefficient with group contribution method

We applied a group contribution method to the Setchenov coefficients obtained in the previous work [5]. The results show that contributions were divided into a solute (fluorocarbon) part and a solvent (alcohol) part. Four kinds of fluorocarbons, CH_2F_2 (CFC-12), C_2HF_5 (HFC-125), CH_2F_2 (HFC-32) and $\text{C}_2\text{H}_2\text{F}_4$ (HFC-134a), are composed of four elements of C–Cl, C–F, C–H and C–C. We assumed that alcohols were characterized by bond numbers of C–H_n ($\text{CH}_2=$ or CH_3-) and C–C.

Contributions estimated by the least squares method are listed in Table 1. The Setchenov coefficients with the group contribution method are described as h_{pred} . The estimation h_{pred} agrees well with the experimental h_{ij} [5] as shown in Table 1. Comparison of solubilities of HFC-134a in ethanol–NaOH solvent using h_{pred} are shown also in Fig. 1. Differences between experimental values

Table 1
Group contribution correlation of Setchenov coefficients for fluorocarbons in methanol–NaOH or ethanol–NaOH solution at atmospheric pressure of fluorocarbon gas.

Contribution	0.0449	0.0635	0.0657	–0.0962	0.0585	–0.1195	h_{ij} [5] (L/mol)	h_{pred} (L/mol)	Dev (%)	
Solute	C–Cl	C–F	C–H	C–C	Solvent	C–H _n ^a	C–C ^a			
CFC-12	2	2	0	0	MeOH	1	0	0.267	0.275	3.2
HFC-125	0	5	1	1		1	0	0.356	0.346	2.9
HFC-32	0	2	2	0		1	0	0.300	0.317	5.7
HFC-134a	0	4	2	1		1	0	0.362	0.348	3.9
CFC-12	2	2	0	0	EtOH	2	1	0.223	0.215	3.9
HFC-125	0	5	1	1		2	1	0.276	0.285	3.1
HFC-32	0	2	2	0		2	1	0.273	0.256	6.3
HFC-134a	0	4	2	1		2	1	0.270	0.287	6.4

h_{ij} : from a literature [5]; h_{pred} : estimated by the group contribution method

^a C–H_n: $\text{CH}_2=$ or CH_3- number of alcohol, C–C: for alcohol.

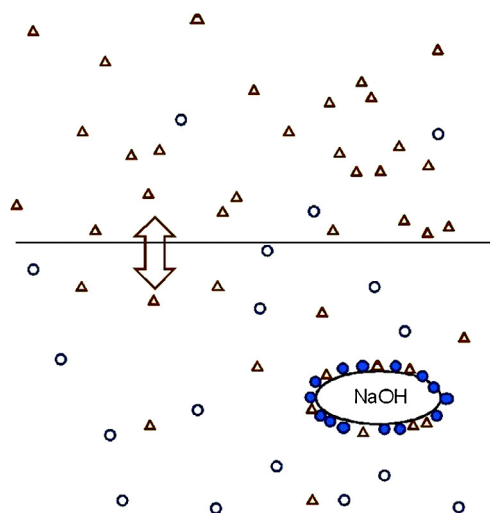


Fig. 2. Solvation model with salting effect. Solvated alcohols have no effect on vapor-liquid equilibria. (○) free alcohol, (●) solvated alcohol, (△) fluorocarbon.

and predicted ones are very small. Table 1 enables to predict the Setchenov coefficients of fluorocarbons in methanol or ethanol. The results show that our method has predictability of fluorocarbons in methanol or ethanol. At present we have no other experimental data for the system. Further predictability will be considered in the future.

In the previous work [5], we found that addition of NaOH in higher alcohols produced gelatinization. For 1-propanol as a solvent, we could not estimate the precise values of h_{ij} with the group contribution method. In the higher alcohols, viscosity of a fluid must be considered.

4. Solvation

4.1. Solvation number of alcohol to NaOH molecule

Solubility of a fluorocarbon in an alcohol solvent without NaOH is expressed as,

$$S = \frac{C_i^0}{C_{\text{alc}}^0} \quad (5)$$

where superscript 0 means the concentration of a fluorocarbon without NaOH. Instead of j script alc is used for emphasis.

Salting-out effects can be explained as the solvation between salt (NaOH) and solvent (alcohol) and/or solute (fluorocarbon) as shown in Fig. 2.

Assuming fluorocarbons maintain vapor-liquid equilibrium with free alcohols excluding solvated alcohols, solubility of a solute (fluorocarbon) in a solvent (alcohol), S , is defined as the mole ratio of fluorocarbons to free alcohols in a liquid phase, as follows,

$$S = \frac{C_i}{C_{\text{alc}} - N_s C_{\text{NaOH}}} \quad (6)$$

where N_s is the solvation number of alcohols around a NaOH molecule. C_{alc} [mol/L] is the total concentration of an alcohol based on the solution volume. C_i is the concentration of fluorocarbon in a solution.

Comparing Eqs. (5) and (6) gives:

$$C_i = C_i^0 \left(1 - \frac{N_s}{C_{\text{alc}}} C_{\text{NaOH}} \right) \quad (7)$$

According to the above equation, the solvation number of alcohols to a NaOH molecule were estimated by fitting to experimental solubility data as shown in the previous paper [5].

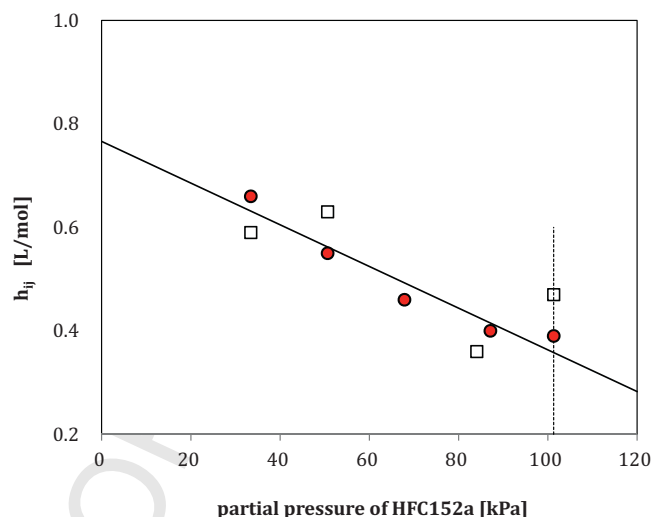


Fig. 3. Effect of partial pressures of fluorocarbons with the Setchenov coefficients in methanol. (●) HFC-152a, (□) HFC-134a.

In this paper, we take another approach. Applying the Maclaurin series to Eq. (4), the following expression is obtained,

$$C_i = C_i^0 (1 - h_{ij} C_{\text{NaOH}} + \dots) \quad (8)$$

Comparing Eqs. (7) with (8), the following simple relation between the solvation number N_s and the Setchenov coefficient h_{ij} is obtained as follows:

$$N_s = h_{ij} C_{\text{alc}} \quad (9)$$

Eq. (9) shows that the solvation number can be expressed as the product of the Setchenov coefficient h_{ij} and the concentration of a solvent alcohol, C_{alc} . Solvation numbers N_s at atmospheric pressure of fluorocarbon gas estimated from Eq. (9) are listed in Table 2. The values of N_s estimated from Setchenov coefficients h_{ij} [5] were in good agreement with N_s estimated based on the solvation theory from Eq. (9). Once the Setchenov coefficients are obtained, then the solvation numbers can be calculated. The more alcohols would be solvated, the more Setchenov coefficient increases.

4.2. Effect of solute (fluorocarbon) with solvation number of solvent (alcohol) to NaOH molecule

Unless fluorocarbons solvate, the Setchenov coefficients should not be affected by fluorocarbons. Since the experimental Setchenov coefficients were found to depend on the fluorocarbons, it means that both alcohol and fluorocarbon molecules solvate to a NaOH molecule. We cannot measure the concentration of solvated fluorocarbons directly. However, we can detect the change of the Setchenov coefficients with partial pressure of fluorocarbons.

The partial pressure effect of 1,1-difluoroethane (HFC-152a) and 1,1,1,2-tetrafluoroethane (HFC-134a) is shown in Fig. 3 for methanol and in Fig. 4 for ethanol. In every figure, the values of the Setchenov coefficients h_{ij} decrease linearly with the partial pressure of fluorocarbons. This means that increase of fluorocarbon concentration in solution expels the solvated alcohol molecule relative to a NaOH molecule. So, the increase of free alcohol molecules increases solubility. A linear relation can be written as follows,

$$h_{ij} = h_j^0 - s_{ij} P_i \quad (10)$$

where h_{ij} is the Setchenov coefficients under a partial pressure P_i of fluorocarbon i . The intersection, h_j^0 shows the Setchenov coefficients when all sites of a NaOH molecule were occupied by alcohol molecules. At room temperature, the values of h_j^0 dependent on

Table 2
Comparison of solvation numbers N_s of alcohols around NaOH estimated from Eq. (4) with ones from Eq. (9) at atmospheric pressure of fluorocarbon gas.

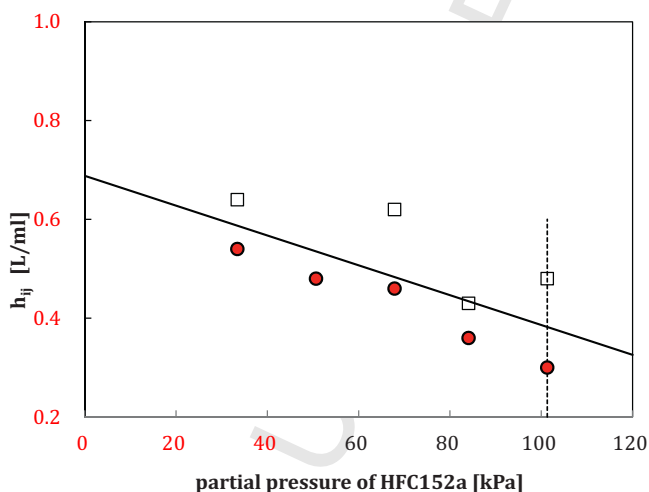
	Methanol			Ethanol		
	N_s [5] (Eq. (4))	N_s (Eq. (9))	AAD (%)	N_s [5] (Eq. (4))	N_s (Eq. (9))	AAD (%)
CFC-12	8.09	6.61	18.3	4.19	3.82	8.7
HFC-125	9.71	8.81	9.3	4.83	4.73	2.0
HFC-32	6.77	7.42	9.6	4.66	4.68	0.5
HFC-134a	7.91	8.96	13.2	5.19	4.63	10.8

Table 3
Group contribution method of Setchenov coefficients D for VOC in water– Na_2SO_4 solution at atmospheric pressure of a solute gas.

Contribution	0.0479 C–H	−0.0564 C–C	0.0163 C–O	0.0163 O–H	0.0843 C=O	−0.0188 C–S	0.0500 S–S	Experimental D^a	Prediction	Dev (%)
Methanol	3	0	1	1	0	0	0	0.24	0.18	26.5
Ethanol	5	1	1	1	0	0	0	0.18	0.22	19.9
1-Propanol	7	2	1	1	0	0	0	0.24	0.26	6.4
1-Butanol	9	3	1	1	0	0	0	0.29	0.29	1.6
1-Pentanol	11	4	1	1	0	0	0	0.33	0.33	1.3
1-Hexanol	13	5	1	1	0	0	0	0.37	0.37	1.0
2-Propanone	6	2	0	0	1	0	0	0.23	0.26	12.7
2-Butanone	8	3	0	0	1	0	0	0.29	0.30	3.0
2-Pentanone	10	4	0	0	1	0	0	0.34	0.34	0.6
2-Hexanone	12	5	0	0	1	0	0	0.40	0.38	5.6
2-Heptanone	14	6	0	0	1	0	0	0.43	0.42	3.0
Dimethyl sulfide	6	0	0	0	0	2	0	0.25	0.25	0.0
Dimethyl disulfide	6	0	0	0	0	2	1	0.30	0.30	0.0

 $\Delta D_{ij} = h_{ij} \times \text{density in moles of solution/liter of solution.}$ **Table 4**
Group contribution method of Setchenov coefficients D for VOC in water–NaCl solution at atmospheric pressure of a solute gas.

Contribution	0.0300 C–H	−0.0067 C–C	0.0100 C–O	0.1067 C–Cl	0.0100 O–H	0.0400 C=C	0.1027 A	0.0547 C=O	0.0609 A–H ^a	0.3693 A–CH ₂ ^a	0.1859 A–CH ₃ ^a	Experimental D^a	Prediction	Dev (%)
Methanol	3	0	1	0	1	0	0	0	0	0	0	0.11	0.11	0.0
2-Propanone	6	2	0	0	0	0	0	1	0	0	0	0.22	0.22	0.6
2-Butanone	8	3	0	0	0	0	0	1	0	0	0	0.26	0.27	5.6
2-Pentanone	10	4	0	0	0	0	0	1	0	0	0	0.34	0.33	3.5
2-Hexanone	12	5	0	0	0	0	0	1	0	0	0	0.40	0.38	4.7
2-Heptanone	14	6	0	0	0	0	0	1	0	0	0	0.42	0.43	3.5
Toluene	0	0	0	0	0	0	1	0	5	0	1	0.67	0.59	11.4
<i>o</i> -Xylene	0	0	0	0	0	0	1	0	4	0	2	0.68	0.72	5.6
Ethylbenzene	3	1	0	0	0	0	1	0	5	1	0	0.86	0.86	0.0
Benzene	0	0	0	0	0	0	1	0	6	0	0	0.43	0.47	8.9
Trichloroethylene	1	0	0	3	0	1	0	0	0	0	0	0.39	0.39	0.0
Chloroform	1	0	0	3	0	0	0	0	0	0	0	0.35	0.35	0.0
Dichloromethane	2	0	0	2	0	0	0	0	0	0	0	0.28	0.27	2.4
1,2-Dichloroethane	4	1	0	2	0	0	0	0	0	0	0	0.32	0.33	2.1

 $\Delta D_{ij} = h_{ij} \times \text{density in moles of solution/liter of solution.}$ **Fig. 4.** Effect of partial pressures of fluorocarbons with the Setchenov coefficients in ethanol: (●) HFC-152a, (□) HFC-134a.

alcohol are 0.766 and 0.688 for methanol and ethanol, respectively. Similarly, the values of C_{alc} are 24.7 and 17.1 mol/L, respectively. So, according to Eq. (9), we can estimate that maximum solvation number of methanol and ethanol to a NaOH molecule is about 19 and 12.

The gradient s_{ij} can be obtained from the value of h_{ij} at atmospheric pressure, $P_i = 101.3$ kPa whose values can be predicted with Table 1 or experimental values [5]. Generally speaking, there should be different straight lines through the same intersection h_j^0 . However, for HFC-152a and HFC-134a, the values of h_{ij} at atmospheric pressure of fluorocarbon gas are very similar, i.e. 0.35 and 0.29 for the systems containing methanol and ethanol. So, correlation lines are the same as shown in Figs. 3 and 4.

Substituting Eq. (10) in (4), we obtain a solubility expression applicable to wider range of partial pressures of fluorocarbons as follows,

$$C_i = C_i^0 \exp[(h_j^0 - s_{ij} P_i) \times C_{\text{NaOH}}] \quad (11)$$

Above expression shows the salting effect on solubility of fluorocarbons in alcohol–NaOH solution.

Table 5Group contribution method of Setchenov coefficients D for VOC in water–TEAB solution at atmospheric pressure of a solute gas.

Contribution	0.023683 C–H	-0.10708 C–C	0.118921 C=O	Experimental D^a	Prediction	Dev (%)
2-Propanone	6	2	1	0.05	0.05	6.3
2-Pentanone	10	4	1	-0.08	-0.07	9.3
2-Hexanone	12	5	1	-0.13	-0.13	1.8
2-Heptanone	14	6	1	-0.19	-0.19	1.1

TEAB: tetraethylammonium bromide

 $D_{ij} = h_{ij} \times \text{density in moles of solution/liter of solution.}$ **Table 6**Group contribution method of Setchenov coefficients D for VOC in water–TMAB solution at atmospheric pressure of a solute gas.

Contribution	0.019259 C–H	-0.09352 C–C	0.103148 C=O	Experimental D^a	Prediction	Dev (%)
2-Pentanone	10	4	1	-0.08	-0.08	2.0
2-Hexanone	12	5	1	-0.13	-0.13	2.6
2-Heptanone	14	6	1	-0.19	-0.19	0.9

TMAB: tetramethylammonium bromide

 $D_{ij} = h_{ij} \times \text{density in moles of solution/liter of solution.}$

5. Application of group contribution method to solubility of VOC gases in water with salts

Falabella and Teja reported the Henry's constants of volatile organic compounds (VOCs) in water–salt solutions [6]. Adopting their functional relationship for these solutions, the following relationship is obtained,

$$D_{ij} = h_{ij} \cdot \rho \quad (12)$$

where D_{ij} [6] and h_{ij} in this paper are the Setchenov coefficients in different units and ρ is the molar density of the solution in moles/liter.

Salts dissociate in water. So, it is doubtful that our model can be applied strictly to these system. We applied the contribution method to these 58 systems. Tables 3 and 4 seem to show good results except a few systems for the systems containing salt Na_2SO_4 or NaCl, respectively. Comparing the solvation results of fluorocarbon to NaOH, interesting results were obtained. According to the model in this paper, solvation number of water on the salt is the same as D , i.e. about 0.3. However experimental data covers wide temperatures, the change in partial pressures were not well described [6]. So, the solvation of VOC on salts cannot be discussed.

Tables 5 and 6 show the results of salting-in systems. The contribution method can be applied superficially. So, from the view point of engineering, the model is useful, but we cannot apply our model strictly to these systems because structural complexities should be considered.

Tables 3–6 enable to predict the Setchenov coefficients of VOC gases in water with salts, i.e. NaCl, Na_2SO_4 , tetraethylammonium bromide and tetramethylammonium bromide.

6. Conclusions

Based on our solubility measurements we succeeded in estimating the Setchenov coefficient h_{ij} by a group contribution method.

Correlated values of h_{ij} were in good agreement with experimental ones.

The results with the group contribution method enable to predict the Setchenov coefficients of fluorocarbons in methanol or ethanol with Table 1 and VOC gases in water with salts, i.e. NaCl, Na_2SO_4 , tetraethylammonium bromide and tetramethylammonium bromide with Tables 3–6.

We set up a simple model that both solute and solvent solvate to a NaOH molecule. We obtained a relationship between the Setchenov coefficient h_{ij} and the solvation number N_s . Once information of the Setchenov coefficients are obtained from experiments or a group contribution method, the solvation numbers can be predicted. Moreover, solubility expressions including both solute and solvent solvation were developed. We showed that our model can be superficially applied to the solubility of VOC gases in water with salts in spite that electrolyte systems contain many structural complexities.

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