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Kinetic Study in Aqueous Solutions of Alcohols with Butyl Group by Ultrasonic Methods

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The ultrasonic absorption coefficients in aqueous solutions of 3-methoxy-1-butanol and 1-methoxy-2-butanol have been measured in the frequency range from 8.5 to 220 MHz at 25 °C. One process associated with a single-relaxational ultrasonic absorption has been found in both solutions. From the concentration dependencies of the relaxation frequency and the amplitude of ultrasonic relaxation, the cause of the relaxation has been attributed to a perturbation of an equilibrium associated with interaction between the solute and the solvent. The rate and thermodynamic parameters have been determined for the reaction and the effect of the isomeric alcohols on water structure has been considered based on these parameters. As results, both alcohols have been found to act as a water structure promoter. The effect of the methoxyl group in the alcohol molecules on the water structure has been speculated to break the water hydrogen bonds, i.e. to act as a structure breaker for water. Also, the results for solutions of other isomers, 2-propoxyethanol and 3-ethoxy-1-propanol, and that of a similar structure, 2-allyloxyethanol, have been compared with those obtained in this study. The effects of substituents in alcohol molecules have been discussed in relation to the water structure. They have also been considered from a comparison with the hydrophobic ability of the alcohols estimated from the concentration dependence of the apparent molar compressibility, along with the result from an ethanol aqueous solution. Both estimations from ultrasonic absorption and compressibility have provided a consistent interpretation for the effects on the water structure. From these present results regarding ultrasonic absorption, it has been speculated that the large ultrasonic absorptions observed near critical points in some alcohol solutions are superimposed with the usual Debye type relaxations.

In a series of ultrasonic studies in aqueous solutions of alcohols, we have noticed that a slight difference in the solute structure gives quite different characteristics of the absorption and velocity.[1-5] When solutes comprising a relatively small hydrophobic group, e.g. 2-propanol and allyl alcohol, a single relaxational absorption is observed in their aqueous solutions in the MHz frequency range. Double relaxation processes are found in the solutions the solutes of which have a relatively large hydrophobic group, e.g. 1-propanol and 2-butoxyethanol. Even if a single relaxation process is observed, the solution characteristics seem to be different. In order to clarify the dynamic characteristics in the solutions more quantitatively, it is necessary to examine in more detail the properties of the aqueous solutions of alcohols with similar structures.

Attention should also be paid to anomalies of the ultrasonic absorption and velocity in alcohol solutions when the solutions are situated close to the phase transition conditions. Some of the aqueous solutions of alcohols exhibit abnormal ultrasonic characters near the critical points.[4,6] In order to clarify the ultrasonic absorption mechanisms in aqueous solutions of alcohols, a distinction of the different relaxation processes in the solutions is very important.

Due to these situations, we have chosen two isomeric alcohols, 3-methoxy-1-butanol and 1-methoxy-2-butanol, as solutes; ultrasonic absorption as well as velocity and density measurements have been carried out as a function of the concentration. These results are compared with those for other solutions of alcohols with similar structures, which have been reported previously in our research series.

Experimental

Chemicals: 1-Methoxy-2-butanol was purchased from Tokyo Kasei Co., Lid. and 3-methoxy-1-butanol was kindly supplied from Daicel Chemical Co., Ltd. They were distilled at normal pressure. The boiling point was 158 °C for 3-methoxy-1-butanol and 135 °C for 1-methoxy-2-butanol. No organic impurities were found by the ^{1}H NMR method. The water contents in both chemicals were shown by the Karl Fisher method to be less than 0.4 wt%. The desired aqueous solutions were made up by doubly distilled water by weight.

Apparatus: Ultrasonic absorption measurement was carried out by a pulse method in the frequency range from 8.5 to 220 MHz; the details were described elsewhere.[4] The sound velocity was measured by an interferometer equipped at 2.5 MHz and a sing-around meter at 1.92 MHz. The density was measured using a conventional Ostwald-type pycnometer, the volume of which was around 5 cm^3 and by Shibayama Density Meter(SS-D-200). All of the measurement cells were immersed in a water bath maintained at a constant temperature within ±0.003 °C. The measurement temperature was 25 °C.

Results and Discussion

Although the solubilities of 1-butanol and 2-butanol into water are limited, the introduction of a methoxyl group into these molecules makes them miscible with water at room temperature. This may be because the methoxyl group has a relatively high hydrophilic ability. Then, the hydrophobic ability of the whole molecule decreases when the methoxyl group is introduced into butanols. The temperature of aqueous solutions of 1-methoxy-2-butanol and 3-methoxy-1-butanol was gradually increased with stirring up to 60 °C in a water bath in order to determine their critical solution temperature. However, no phase separation was observed, and the solutions kept their homogeneous phases. The conditions of the present ultrasonic experiments have been confirmed to be far beyond their critical points.

We first give the ultrasonic absorption results and discuss the relaxation process. In Figs. 1 and 2, the representative ultrasonic absorption spectra are shown at the various concentrations. It should be noticed that the absorption coefficient divided by the square of the measurement frequency, (α/f^2), is almost frequency independent at a frequency less than 35 MHz. In some alcohol solutions, analyses of the frequency dependence of the absorption coefficient were carried out on the bases of the fluctuation of mode coupling models.[7] According to the proposed theories,[8,9] the (α/f^2) values should increase with decreasing frequency. Actually, in solutions of 2-methyl-2-propanol,[10,11] 1-propanol,[5,12] and 2-butoxyethanol,[4] large absorptions were observed in the frequency range less than 10 MHz. Such absorptions, however, have been limited to solutions, the solutes of which have relatively large hydrophobic groups. In solutions of alcohols with a slightly lower hydrophobicity, such as 2-propanol and those studied here, a frequency dependence at the lower frequency range is scarcely found. These phenomena have made us prefer to analyze the absorption coefficient by a conventional Debye type relaxational procedure. The analytical equation is

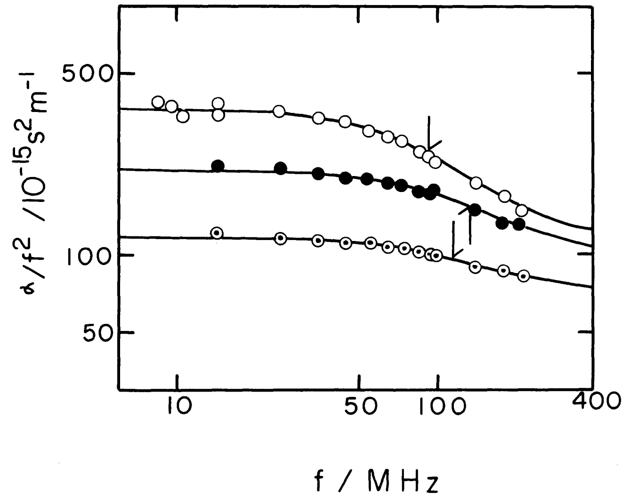

Fig. 1. Representative ultrasonic absorption spectra in aqueous solutions of 1-methoxy-2-butanol and 3-methoxy-1-butanol at 25 °C. The spectra are shown in form by Eq. 1; the arrow indicates the position of the relaxation frequency. ○: 3.57 mol dm^{-3} 1-methoxy-2-butanol solution, ●: 5.00 mol dm^{-3} 3-methoxy-1-butanol solution, ⊙: 3.57 mol dm^{-3} 3-methoxy-1-butanol solution.

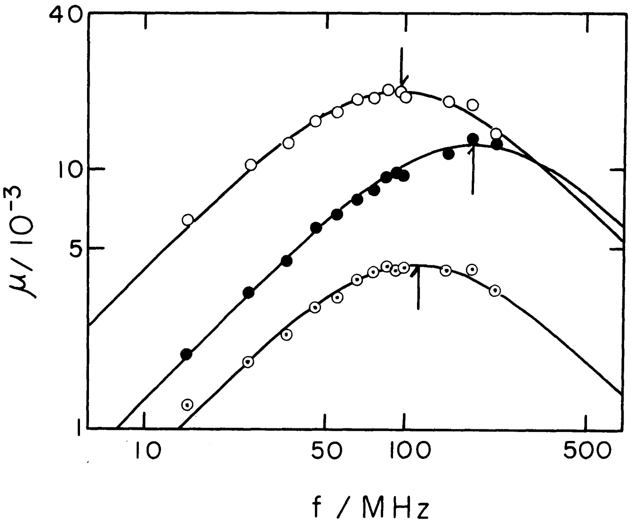

Fig. 2. Representative spectra for the solutions of 1-methoxy-2-butanol and 3-methoxy-1-butanol. They are shown for the excess absorption per wavelength by Eq. 1′. ○: 4.00 mol dm^{-3} 1-methoxy-2-butanol solution, ⊙: 3.57 mol dm^{-3} 3-methoxy-1-butanol solution, ●: 6.75 mol dm^{-3} 3-methoxy-1-butanol solution

$$\alpha/f^2 = A/[1+(f/f_r)^2]+B \qquad (1)$$

or

$$\mu = (\alpha/f^2 - B)fc = Afc/[1+(f/f_r)^2], \qquad (1')$$

where f_r is the relaxation frequency, A the amplitide of the ultrasonic relaxation, B the backgrond absorption, μ the absorption per wavelength, and c the sound velocity. The ultrasonic parameters, (f_r, A and B) were determined so as to obtain the best fit of the experimental data to Eq. 1 by a nonlinear least mean-square method, the details of which are described elsewhere.[4,13] As can be seen in these figures, the obtained absorption coefficients fit well to the calculated curves, which are shown by solid curves. In Table 1, the thus-calculated ultrasonic parameters are listed along with the sound velocity and density data. A complete set (α/f^2, f, sound velocity and density) is available from one of the authors (S.N.) upon request. Measurements of the absorption coefficients by the pulse method were not possible at less than 8.5 MHz, because of the small absorption at lower frequencies. The concentration dependence of the relaxation frequency is shown in Fig. 3, and that of the amplitude of the ultrasonic relaxation in Fig. 4. Although both dependencies of the relaxation frequency and the relaxational amplitude are similar, the magnitudes are quite different, even if the solutes are isomers each other. The trends of the minimum relaxation frequency and the maximum relaxational amplitude are the same as those previously observed on other aqueous solutions of alcohols.[1-3] These obtained ultrasonic parameters are well-known to be related to the dynamic properties associated with chemical reactions occurring in solutions. An analysis by Zana's group for aqueous solutions of alcohols[14] is

Table 1. Ultrasonic and Thermodynamic Parameters for Aqueous Solutions of 3-Methoxy-1-butanol and 1-Methoxy-2-butanol at 25 °C

C_e	c	A	B	f_r	ρ
mol dm^{-3}	m s^{-1}	10^{-15} s^2 m^{-1}		MHz	kg dm^{-3}
(3-Methoxy-1-butanol)					
0.50	1529.1	—	—	—	0.9955
1.25	1572.1	—	—	—	0.9937
2.00	1605.5	—	—	—	0.9922
2.75	1625.2	22.2±2.4	45.0±2.8	213.9±29.4	0.9905
3.00	1626.9	24.4±1.1	55.2±1.6	146.6±16.5	0.9897
3.25	1629.1	31.1±1.0	65.5±1.5	133.4±12.5	0.9890
3.50	1627.4	45.1±1.0	71.2±1.1	115.0±7.0	0.9879
3.57	1626.1	46.9±0.8	72.2±0.8	113.5±5.1	0.9878
4.00	1620.6	65.9±1.1	87.8±1.6	120.0±5.4	0.9855
4.25	1614.2	72.9±1.6	97.2±1.9	119.4±7.2	0.9840
4.51	1607.5	80.2±1.7	102.0±2.2	126.6±7.6	0.9824
5.00	1588.9	116.6±4.9	94.5±6.9	133.2±12.4	0.9785
5.51	1567.1	99.4±3.0	115.8±4.3	146.9±11.1	0.9740
6.00	1549.2	105.6±7.6	114.4±10.5	156.4±25.1	0.9691
6.50	1523.5	107.1±2.6	134.1±3.6	128.5±9.1	0.9635
6.75	1515.5	87.5±6.6	117.0±8.1	183.8±22.4	0.9602
(1-Methoxy-2-butanol)					
0.50	1535.0	—	—	—	0.9962
1.00	1571.4	—	—	—	0.9960
1.50	1602.8	—	—	—	0.9958
2.00	1624.1	32.7±0.7	40.1±1.1	141.6±8.4	0.9953
2.50	1628.0	98.4±2.1	61.1±2.5	117.3±7.1	0.9941
3.00	1621.3	178.8±2.0	85.0±1.8	107.6±3.0	0.9921
3.57	1604.8	256.6±3.6	111.7±2.2	92.6±2.6	0.9892
4.00	1584.5	262.2±5.2	132.1±3.4	95.8±4.0	0.9850
4.50	1558.5	259.5±5.4	134.5±4.2	102.1±5.0	0.9805
5.50	1508.3	231.9±3.7	131.4±2.3	94.9±3.1	0.9694
6.51	1449.4	139.3±2.7	124.8±2.1	101.9±4.6	0.9555
7.50	1385.3	54.0±1.8	103.9±2.3	122.9±11.7	0.9382

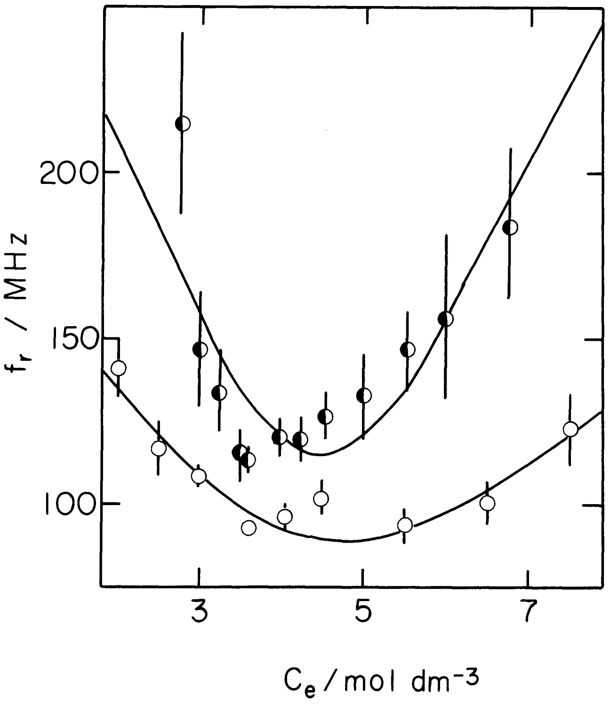

Fig. 3. Concentration dependence of the relaxation frequency for solutions of 1-methoxy-2-butanol (○) and 3-methoxy-1-butanol (◐).

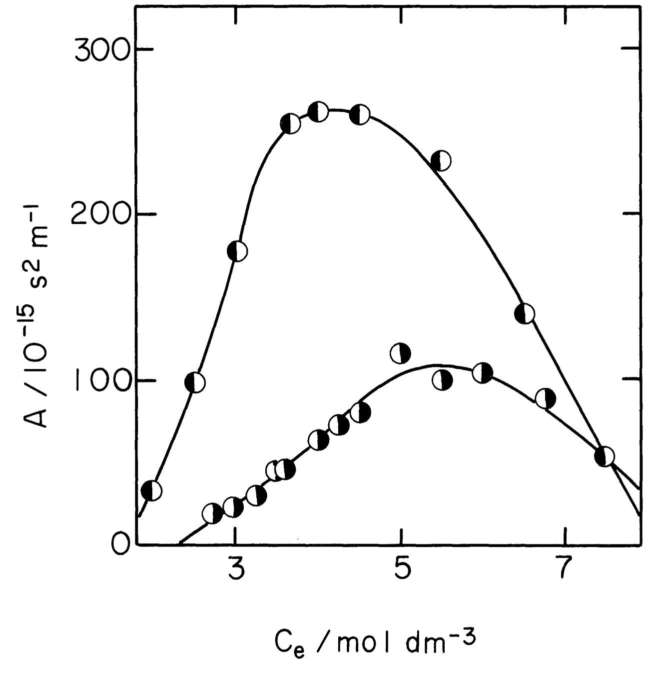

Fig. 4. The concentration dependence of the amplitude of the ultrasonic relaxation for the solutions of 1-methoxy-2-butanol (◐) and 3-methoxy-1-butanol (◑).

worth noticing. The minimum relaxation frequency and the maximum amplitude of ultrasonic relaxation were found in aqueous solutions of 1,2-hexanediol and 1,2,3-octanetriol. The cause of the relaxational absorption was attributed to a molecular aggregation reaction. A careful look at their reported ultrasonic parameters indicates that the concentration dependencies of the relaxation frequency and the amplitude are slightly different from those observed in this study. That is, the amplitude of their results increases dramatically and goes through a maximum and the relaxation frequency shows a discontinuous trend. Such large changes in the ultrasonic parameters have also been observed in other nonelectrolyte aqueous solutions.[15] In such a case, plural relaxations are superimposed. On the other hand, the dependencies for the solutions under this study indicate quite smooth changes with the analytical concentration as can be seen in Figs. 3 and 4. Relaxational absorption was not observed below some certain concentration; there exists a threshold concentration above which relaxation appears. According to a compressibility study for aqueous nonelectrolyte solutions by Endo,[16] the water structure is altered at some certain concentration of the nonelectrolytes. When the concentration of the alcohols is not so high, we consider that the original water structure is not altered very dramatically. Above some threshold concentration of the alcohols, the water structure might be altered in order to maintain uniform solutions. Above such a concentration, the ultrasonic relaxation process is observed. Therefore, the concentration at which the relaxation appears is much dependent on the alcohol structure. Though an aggregation reaction due to the hydrophobic interaction in solutions of the present study may be ruled out as the cause of the relaxation, aggregates formed by hydrogen bonding may be plausible. However, the relaxation process associated with the aggregate by hydrogen bonding has been observed in the GHz frequency range,[17] and is far from our experimental time range. Thus, the most plausible reaction model for relaxational absorption is speculated to be associated with a solute-solvent interaction, presumably, according to their hydrogen bonding abilities. It is simply expressed by

$$\mathrm{AB} \underset{k_b}{\overset{k_f}{\rightleftharpoons}} \mathrm{A} + \mathrm{B}, \qquad (2)$$

where k_f and k_b are the forward backward rate constants, A the solute molecule, B the solvent molecule and AB the complex. We assume here that the solvent water consists of two states, one of which is hydrogen-bonded water and the other is the non-hydrogen-bonded one (it should be expressed as the less-structured water molelcule). The latter may participate in the reaction under consideration. We thus obtain the relation between the analytical concentrations of the solute, C_e, the solvent, C_w, and the relaxation frequency as follows:[5]

$$2\pi f_r = k_b\{(C_e - \beta C_w + K_{12})^2 + 4\beta C_w K_{12}\}^{1/2}. \qquad (3)$$

Here β is the fraction of non-hydrogen-bonded water and K_{12} is defined as $K_{12}=k_f/k_b$. It may be a good approximation that the rate and thermodynamic parameters, k_b, β and K_{12}, are concentration independent. They are determined so as to obtain the best fit of the experimental relaxation frequencies to Eq. 3 using a nonlinear least mean-square method. The solid curves in Fig. 3 represent the calculated values using the thus-obtained rate and thermodynamic parameters. In both solutions, the calculated values fit well to the experimental data. These parameters are listed in Table 2 along with those for solutions of 2-propoxyethanol,[18] 2-allyloxyethanol,[19] and 3-ethoxy-1-propanol.[1] The calculations for 2-propoxyethanol and 2-allyloxyethanol solutions were performed previously by a trial-and-error procedure; a recalculation was carried out using the same computer program. The obtained parameters are almost the same as those reported previously. Taking into account that the fraction of the non-hydrogen-bonded water molecule in liquid water is 0.24,[20] all of the β values in the solutions under study are smaller than that in liquid water. That is, these alcohols in the aqueous solutions act as water structure promoters. However, depending upon the structures of the alcohol molecules, it can clearly be seen that the β values are different. The smaller is the value, the fewer are the non-hydrogen-bonded water molecules; it is considered that the hydrophobicity of the alcohol molecule increases with a decrease of the β value. Thus, the order of the hydrophobicity for the five alcohols may be according the series 2-propoxyethanol, 2-allyloxyethanol, 3-methoxy-1-butanol, or 3-ethoxy-1-propanol, and 1-methoxy-2-butanol. These results indicate that the propyl group has a relatively high hydrophobic ability and that the

Table 2. Rate and Thermodynamic Constants for the Reaction Associated with the Solute Solvent Interaction at 25 °C

Alcohol	k_b / 10^8 mol^{-1} dm^3 s^{-1}	K_{12} / mol dm^{-3}	β	Ref.
3-Methoxy-1-butanol	2.2 ±0.2	0.57±0.14	0.159±0.007	This work
1-Methoxy-2-butanol	0.94±0.06	1.7 ±0.3	0.19 ±0.01	This work
2-Propoxyethanol	1.8 ±0.1	0.48±0.09	0.102±0.004	(18)
2-Allyloxyethanol	3.3 ±0.2	0.09±0.02	0.114±0.002	(19)
3-Ethoxy-1-propanol	2.4 ±0.1	0.27±0.07	0.159±0.004	(1)

methoxyl group assists in the hydrophobicity of the alcohol decrease. These estimations are resonably accepted in terms of the solubility data for some butanols in water.

Another important parameter for sound absorption is a maximum excess absorption per wavelength, $\mu_{\max}$, which is expressed by

$$\mu_{\max} = \pi \rho c^2 \Gamma (\Delta V - \alpha_p \Delta H / \rho C_p)^2 / 2RT. \tag{4}$$

Here, ρ is the solution density, ΔV the standard volume change of the reaction, α_p the thermal expansion coefficient, ΔH the standard enthalpy change of the reaction and C_p the specific heat at constant pressure. The concentration term, Γ, is specified for the reaction under consideration as

$$\Gamma = \{1/[\mathrm{A}] + 1/[\mathrm{B}] + 1/[\mathrm{AB}] - 1/([\mathrm{A}]+[\mathrm{B}]+[\mathrm{AB}])\}^{-1}. \tag{5}$$

From the concentration dependence of the relaxation frequency, it has been possible to determine the equilibrium constant, K_{12}, and the fraction of the less structured water, β. The concentrations of the reactants in Eq. 5 are thus obtainable. In order to interpret the concentration dependence of the maximum excess absorption per wavelength precisely, the specific heat and expansion coefficient data are necessary. However, they are not available at the present stage. If the relaxation process is only due to the volume term, then plots of $\mu_{\max}$ *vs.* $\rho c^2 \Gamma$ provide a straight line with a zero intercept. For solution of 1-methoxy-2-butanol, a straight line was obtained, of which the slope gave the volume change to be 3.8×10^{-6} m^3 mol^{-1}. This value seems to be too small. Further, no such linear relation was obtained in other solutions. These facts may indicate that the observed relaxation process is not only associated with thermal relaxation, but also volume relaxation. However, the most controlling factor affecting on the maximum excess absorption per wavelength is considered to be due to $\rho c^2 \Gamma$. In Fig. 5, both the dependencies of $\mu_{\max}$ and $\rho c^2 \Gamma$ are shown as a function of the analytical concentration of 1-methoxy-2-butanol. Similar trends were also obtained for other solutions, even though the peak positions are not very close to each other.

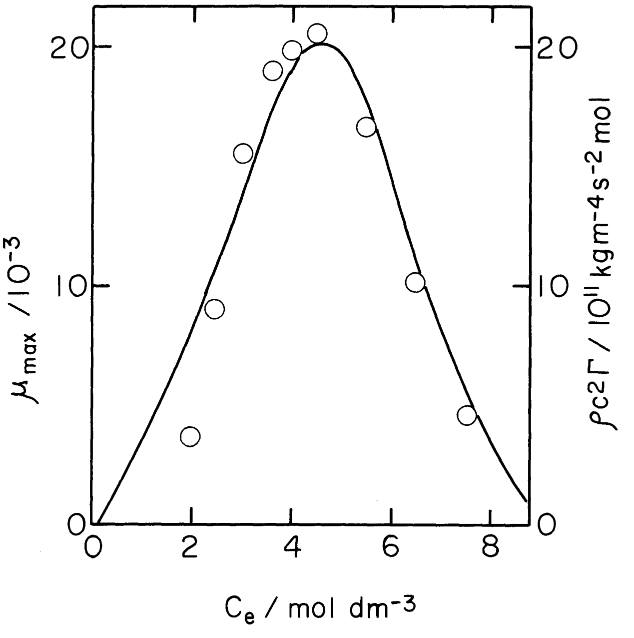

Fig. 5. Comparison of the $\mu_{\max}$ and $\rho c^2 \Gamma$ as a function of the 1-methoxy-2-butanol concentration.

In the second place, we discuss the sound velocity and density data. As can be seen in Table 1, the sound velocity increases with the analytical concentration, and then goes through a maximum, even although the position where the peak is observed depends on the structure of alcohol. The density decreases, on the other hand, monotonously with the concentration. From these two experimental values, it is possible to obtain the apparent molar quantities. The apparent molar volume, ϕ_v, and the apparent molalr compressibility, ϕ_s are calculated using

$$\phi_v = M_e/\rho - 1000(\rho - \rho_0)/(m\rho\,\rho_0) \tag{6}$$

and

$$\phi_s = \kappa_s \phi_v + (\kappa_s - \kappa_w)/m\rho_0, \tag{7}$$

where M_e is the molecular weight of the solute and ρ_0 is the solvent density; κ_s and κ_w are the adiabatic compressibilities for the solution and the solvent water, respectively, and m the molality which is converted from the molarity using the solution density, ρ. The above two compressibilities were obtained from the relation $\kappa = (\rho c^2)^{-1}$. The calculated apparent molar compressibility for these five alcohol solutions decreases with increasing the analytical concentration at a low-concentration range, but then increases with the concentration (a minimum appears). Although profiles are similar to those of 2-alkoxyethanol solutions,[21-23] the differences in these concentration dependencies for four alcohols are not very clear. Figure 6 shows the concentration dependence of the apparent molar compressibility for the solutions of four alcohols under investigation, as well as that of an ethanol solution. The sound velocity and density data for the ethanol solution were taken from the literature values.[24] According to Lara and Desnoyers' work,[23] the more hydrophobic is the alcohol molecule, the sharper is the concentration dependence of the apparent molar compressibility. Following their estimation of the hydrophobicity of the solute in aqueous media from Fig. 6, the hydrophobic ability might be estimated to be in the series of 2-propoxyethanol, 1-methoxy-2-butanol ≈ 2-allyloxyethanol ≈ 3-methoxy-1-butanol and ethanol. However, an estimation according to the sharpness of the curves

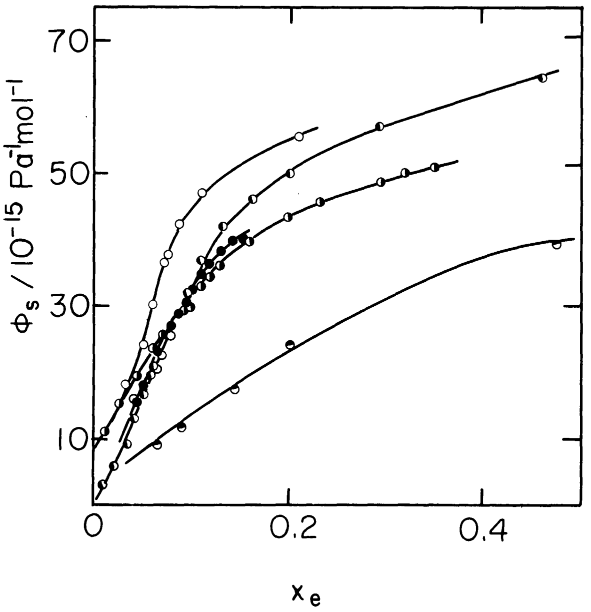

Fig. 6. The concentration dependence of the apparent molar compressibility for aqueous solutions of five alcohols. ◒: ethanol, ○: 2-propoxyethanol, ◐: 1-methoxy-2-butanol, ◑: 3-methoxy-1-butanol, ●: 2-allyloxyethanol.

seems not to be very definite, although clear differences are seen between ethanol and other alcohol solutions. When the difference in the hydrophobicities of alcohol molecules is sufficiently large, an estimation is useful.

We now combine the information obtained from the absorption results with the velocity data and then consider the solution characteristics of alcohols with similar structures. In our previous report,[3] we showed that plots of the fraction of the nonhydrogen-bonded water, β, vs. the solute mole fraction, x_e, where the adiabatic compressibility gets a minimum value, fall on a straight with a zero intercept for aqueous solutions of some isomeric alcohols with six carbons in the molecules. Figure 7 indicates plots for the alcohol solutions under study, in which the results for the four alcohol solutions are almost on one straight line; only the parameter for 1-methoxy-2-butanol solution is seen to deviate from the straight line. This means that the 1-methoxy-2-butanol molecule has less effect as a water structure promoter than that expected from the molecular structure. One of the reasons for this might be because of the intramolecular hydrogen bonding as has been predicted from comparisons of the absorption data in 1-methoxy-2-propanol and 3-methoxy-1-propanol solutions.[1] Such intramolecular hydrogen-bond formation is expected to help the hydrophilic ability of the entire molecule to increase; the stability of a complex with water may not be very stable compared with others. Then, the equilibrium constant, K_{12}, which is a measure of the unstabilization of the complex, is becoming larger than others, as can be seen in Table 2.

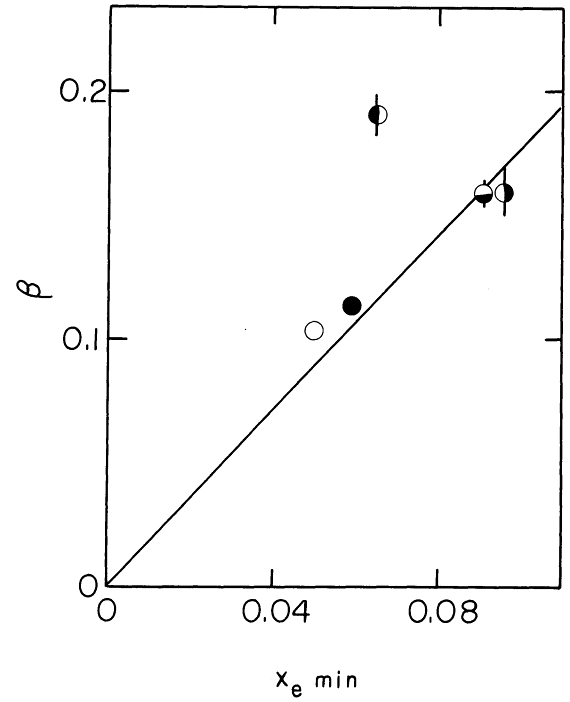

Fig. 7. The correlation between the β and the minimum concentration of the compressibility. ◑: 3-methyl-1-butanol, ◒: 3-ethoxy-1-propanol, ◐: 1-methoxy-2-butanol, ●: 2-allyloxyethanol, ○: 2-propoxyethanol.

An estimation of the hydrophobicity from the profiles of the concentration dependence of the apparent molar compressibility might have indicated that 1-methoxy-2-butanol had its hydrophobic activity between those of 2-propoxyethanol and 2-allyloxyethanol, though this is not very clear. On the other hand, the β parameter clearly reflects the difference in the hydrophobic ability of the alcohols, even for quite similar structures of the solutes, i.e. for the isomeric alcohols.

From the above results for these five solutions of alcohols, it may be stressed that ultrasonic absorption is well-characterized by the usual Debye type single-relaxational equation, and that the absorption in the relatively low frequency range (less than 35 MHz) is almost frequency independent. This means that the ultrasonic absorption in aqueous solutions of alcohols can not be interpreted uniquely by fluctuation of mode-coupling models. The anomaly in the ultrasonic absorption observed near their critical points might involve several processes. We consider that the sound absorption mechanisms in solutions should be systematically clarified from results concerning many similar systems. In our research series of ultrasonic absorption in various alcohol solutions, it has been pointed out that the solute-solvent interaction is one of the causes of relaxation in the solutions.

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References

1) S. Nishikawa and M. Ueda, *Bull. Chem. Soc. Jpn.,* **64**, 1294 (1991).

2) S. Nishikawa, M. Moritsubo, F. Matsuo, and Y. Endo, *Bull. Chem. Soc. Jpn.,* **63**, 26 (1990).

3) S. Nishikawa, G. Tanaka, and R. Shinohara, *Bull. Chem. Soc. Jpn.,* **59**, 827 (1986).

4) S. Nishikawa and K. Kotegawa, *J. Phys. Chem.,* **89**, 2896 (1985).

5) S. Nishikawa, M. Mashima, and T. Yasunaga, *Bull. Chem. Soc. Jpn.,* **48**, 661 (1975).

6) R. Zana and B. Michels, *J. Phys. Chem.,* **93**, 2643 (1989).

7) W. M. Madigosky and R. W. Warfield, *J. Chem. Phys.,* **86**, 1491 (1987).

8) R. A. Ferrel and J. K. Bthattacharjee, *Phys. Rev.,* **31**, 1788 (1985).

9) V. P. Ramanov and V. A. Solov'ev, *Sov. Phys.-Acoust. (Engl. Transl.)*, **11**, 68 (1965).

10) S. Nishikawa, M. Mashima, M. Maekawa, and T. Yasunaga, *Bull. Chem. Soc. Jpn.,* **48**, 2353 (1975).

11) M. J. Blandamer, D. E. Clarke, N. J. Hidden, and M. C. R. Symons, *J. Chem. Soc., Faraday Trans.,* **64**, 2691 (1968), and M. Tabellout, P. Lancelecer, J. R. Emery, D. Hayward, and R. A. Pethrick, *J. Chem. Soc., Faraday Trans.,* **86**, 1493 (1990).

12) M. J. Blandamer, N. J. Hidden, M. C. R. Symons, and N. C. Treloar, *J. Chem. Soc., Faraday Trans.,* **64**, 3242 (1968).

13) S. Nishikawa and F. Matsuo, *J. Phys. Chem.,* **95**, 438 (1991).

14) M. Frindi, B. Michels, and R. Zana, *J. Phys. Chem.,* **95**, 4832 (1991).

15) S. Nishikawa and Y. Harano, *J. Phys. Chem.,* **93**, 7152 (1989).

16) H. Endo, *Bull. Chem. Soc. Jpn.,* **46**, 1106 (1973).

17) K. Oda, R. Hayakawa, and Y. Wada, *Jpn. J. Appl. Phys.,* **14**, 1113 (1975).

18) S. Nishikawa, Y. Yamashita, and M. Mashima, *Bull. Chem. Soc. Jpn.,* **55**, 1 (1982).

19) S. Nishikawa and T. Yamaguchi, *Bull. Chem. Soc. Jpn.,* **56**, 1585 (1983).

20) C. M. Davis Jr. and J. Jarzynski, *Adv. Mol. Relaxation Processes,* **1**, 155 (1968).

21) G. Roux, G. Perron, and J. E. Desnoyers, *J. Solution Chem.,* **7**, 639 (1978).

22) L. Avedikian, G. Perron and J. E. Desnoyers, *J. Solution Chem.,* **4**, 331 (1975).

23) L. Lara and J. E. Desnoyers, *J. Solution Chem.,* **10**, 465 (1972).

24) J. Emery and S. Gasse, *Acustica,* **43**, 205 (1979).