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Ultrasonic Study in Aqueous Solutions of Alcohols with Methoxyl and Propoxyl Groups

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Ultrasonic absorption, velocity and density measurements were carried out in aqueous solutions of 1-methoxy-2-propanol and 3-(3-methoxypropoxy)propanol as a function of their concentrations at 25 °C. A single ultrasonic relaxational phenomenon was observed in both solutions in the frequency range from 6.5 to 220 MHz. The cause was attributed to a perturbation of an equilibrium associated with the solute solvent interaction, $AB \rightleftharpoons A+B$, from the concentration dependences of the relaxation frequency and the amplitude of the relaxational absorption. The rate and thermodynamic parameters were determined for the above process. It was found that the methoxyl group introduced in alcohol molecule acted as a water structure breaker while the propoxyl group promoted the water structure. Speculation of excess ultrasonic absorption in various monohydroxy alcohol aqueous solutions was carried out from the present data. In order to further confirm the participation of the solvent water molecule in the observed relaxation process, absorption measurements were carried out in methanol, dimethyl sulfoxide, and their mixed solvents with water for 3-(3-methoxypropoxy)-propanol.

It is well established that the solvent water structure may be altered by the addition of solutes. Such characteristics have been studied by many static experiments in electrolyte and nonelectrolyte aqueous solutions.[1,2] The application of ultrasonic absorption techniques, as one type of relaxation method, provides useful information concerning the dynamic properties of the solutions and also gives structural characteristics of the solvents.[3,4]

The present authors have reported on the kinetic properties in aqueous solutions of various alcohols studied using ultrasonic methods.[5–7] The characteristics of sound absorption are strongly dependent upon the hydrophobicities of the solute molecules. In aqueous solutions of 2-butoxyethanol, 1-propanol, and 2-methyl-2-propanol, two relaxation processes have been found, while a single relaxation process has been found in those of 2-propanol and allyl alcohol with smaller hydrophobicities. One of the relaxation processes observed in the frequency range from 10 to 120 MHz has been associated with an interaction between the solute and the solvent. In a previous report,[8] we showed that the ethoxyl group in alcohols slightly affects the water structure as the promoter. It is therefore desirable to examine the effects of other groups which have ether oxygen in solutes. For this purpose we have chosen two solutes, 1-methoxy-2-propanol and 3-(3-methoxypropoxy)propanol, and the ultrasonic absorption coefficients have been measured in their aqueous solutions as functions of the frequency and concentrations.

On the other hand, excess absorption in aqueous solutions of alcohols has now been interpreted using fluctuation models.[9–11] According to the theories proposed so far, the absorption coefficient divided by the square of the measured frequency should increase monotonously with decreasing frequency. In this paper, we also report on some results which are frequency independent in the low-frequency range. Further, it is speculated from the present study that excess sound absorption in methanol and ethanol aqueous solutions may appear at very high-frequency regions.

Experimental

1-Methoxy-2-propanol and 3-(3-methoxypropoxy)propanol [dipropylene glycol monomethyl ether] were of purest grade from Tokyo Kasei Co., Ltd. and were distilled once. Methanol and dimethyl sulfoxide [DMSO] were purchased from Wako Chemical and were used without further purification. The desired solutions were prepared with doubly distilled water, methanol, and DMSO either by weighing or volumetrically.

The sound absorption coefficient, α, in the frequency range between 6.5 and 220 MHz was measured by a computerized pulse apparatus, details of which are described elsewhere.[12] The sound velocity was measured using an interferometer at 2.5 MHz. Density measurements were carried out using a conventional pycnometer (about 4 cm^3). The measurement cells for all equipments were immersed in a water bath which was thermostated within ±0.002 °C. All of the measurements were carried out at 25 °C.

Results and Discussion

Figures 1 and 2 show some representative ultrasonic absorption spectra in aqueous solutions of 1-methyloxy-2-propanol and 3-(3-methoxypropoxy)propanol at 25 °C. All of the spectra are well-expressed by a single relaxational equation,

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

or

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

In the above, μ is the ultrasonic excess absorption per wavelength, f the frequency, c the sound velocity, A the amplitude of the relaxational absorption, f_r the relaxation frequency and *B* the background absorption. The ultrasonic absorption parameters, *A*, *B*, and f_r were determined by a microcomputer connected to a pulse apparatus employing a non-linear least-mean-square method. The solid curves in Figs. 1 and 2 are those of the calculated values. It should be noticed that in both solutions, α/f^2 values less than 10 MHz are almost frequency independent. In Table 1, the obtained ultrasonic parameters are listed as a functions of the analytical concentration of the solutes, along with sound velocity and density data. The concentration dependences of the ultrasonic parameters are quite similar to those for other alcohol solutions so far reported.[5-9] That is, the relaxation frequency shows a minimum and the amplitude of the relaxational absorption a maximum, though their values depend on the structures of alcohols. Also, the sound velocity goes through a maximum at 3.75 mol dm^{-3} for a 1-methoxy-2-propanol solution and at 2.30 mol dm^{-3} for a 3-(3-methoxypropoxy)propanol solution. These are characteristic of aqueous solutions of nonelectrolytes with a hydroxyl group. As has been described in detail

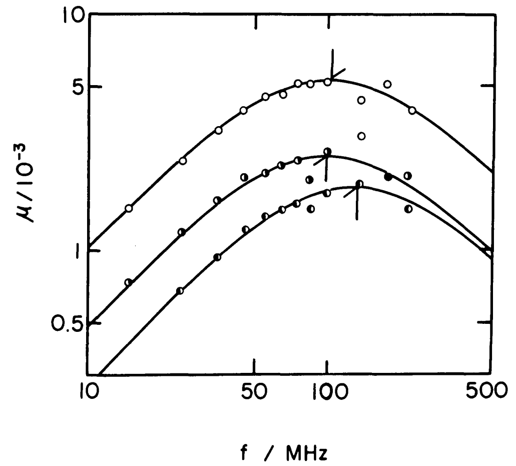

Fig. 1. Representative ultrasonic absorption spectra in aqueous solutions of 1-methoxy-2-propanol at 25 °C. The arrow shows the position of the relaxation frequency. ◐: 3.22 mol dm^{-3}, ◑: 3.94 mol dm^{-3}, ○: 4.97 mol dm^{-3}.

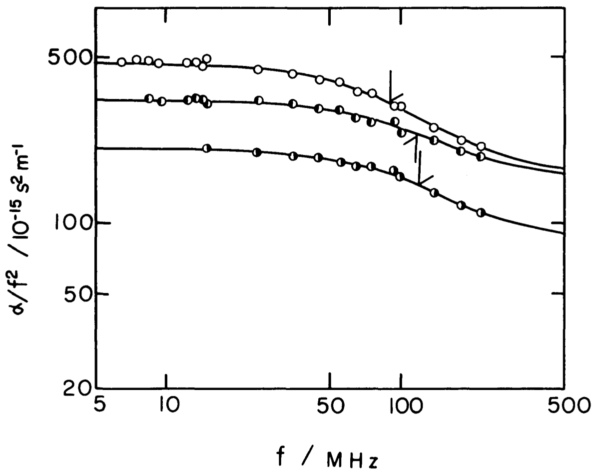

Fig. 2. Representative ultrasonic absorption spectra in aqueous solution of 3-(3-methoxypropoxy)propanol at 25 °C. ◑: 2.40 mol dm^{-3}, ○: 3.50 mol dm^{-3}, ◐: 5.00 mol dm^{-3}.

Table 1. Ultrasonic and Thermodynamic Parameters for Two Aqueous Soutions at 25 °C

Concn	f_r	*A*	*B*	*c*	ρ
mol dm^{-3}	MHz	10^{-15} s^2 m^{-1}		m s^{-1}	kg dm^{-3}
	1-Methoxy-2-propanol aqueous solution				
2.98	191±30	21±2	31±3	1644.5	0.9975
3.22	134±22	17±1	46±1	1648.1	0.9975
3.48	93±13	24±2	54±1	1651.6	0.9972
3.94	102±26	30±3	67±3	1651.2	0.9964
4.48	89±13	58±5	74±2	1646.4	0.9948
4.97	103±12	64±3	91±3	1627.6	0.9925
5.57	103±30	62±8	105±6	1598.8	0.9884
6.05	95±15	70±5	110±3	1579.2	0.9848
6.90	126±17	58±3	111±4	1526.5	0.9761
	3-(3-Methoxypropoxy)propanol aqueous solution				
1.80	132±20	42±2	49±3	1638.9	1.0001
1.96	143±14	57±2	53±3	1643.3	1.0076
2.40	120±9	120±3	83±4	1647.7	1.0085
2.60	99±9	165±7	88±5	1640.2	1.0087
3.00	96±10	202±9	149±12	1624.4	1.0076
3.41	89±6	291±8	153±8	1598.4	1.0066
3.50	90±3	313±5	160±3	1592.7	1.0061
3.86	98±5	306±6	177±7	1566.3	1.0034
4.00	106±7	305±8	166±8	1553.3	1.0011
4.40	111±6	276±6	159±6	1526.9	0.9976
5.00	119±10	178±5	151±6	1469.5	0.9875

in previous reports,[5,7] only a perturbation of the equilibrium associated with the solute solvent interaction has remained as a possible cause of relaxational absorption, in order to interpret the concentration dependence of the ultrasonic parameters. It can be expressed by the following model:

$$\mathrm{AB} \underset{k_{21}}{\overset{k_{12}}{\rightleftharpoons}} \mathrm{A} + \mathrm{B}, \qquad (2)$$

where A is the solute molecule, B the solvent molecule, and AB the complex formed by the solute and solvent molecules. It is assumed that the reactant, B, is a water molecule which is a non-hydrogen-bonded, or less structured water molecule: precisely, it should be expressed as water molecule which has the ability to interact with the solutes. The relation between the relaxation frequency and the reactant concentrations is given by

$$2\pi f_r = k_{21}([\mathrm{A}] + [\mathrm{B}]) + k_{12}. \qquad (3)$$

From the structures of alcohols and water, it might be possible to say that more complicate complexes by plural hydrogen-bondings between them may be constructed. However, the most plausible or main complex seems to be a 1:1 type, since the observed absorption can clearly be expressed by a single relaxational equation and the concentration dependence of the relaxation frequency is not well-interpreted by a model like $\mathrm{AB_2} \rightleftharpoons \mathrm{A}+2\mathrm{B}$.[13] Further, the interaction process between water-water[14] has been estimated to be around 160 GHz, and that for alcohol-alcohol[15] has been found to be around 8 GHz. Therefore, the coupling effect from the fast process to the observed one may arise from a reaction associated with an alcohol-alcohol interaction. Then, the reaction scheme may be expressed as

$$\mathrm{AB} \underset{k_{21}}{\overset{k_{12}}{\rightleftharpoons}} \underset{\searrow \mathrm{B}}{\mathrm{A}} \underset{k_{32}}{\overset{k_{23}}{\rightleftharpoons}} \mathrm{A}^*, \qquad (4)$$

where A* is an alcohol molecule in another state, i.e. the molecule participating in aggregates by hydrogen-bonding with each other. Then, the relaxation frequency associated with the observed process is derived as

$$2\pi f_r = k_{21}([\mathrm{A}] + [\mathrm{B}])\{1 - k_{23}/(k_{23} + k_{32})\} + k_{12}. \qquad (5)$$

If k_{32} is much larger than k_{23}, the effect from the faster process may be neglected. Even if the effect is not ignored, the concentration dependence of the relaxation frequency is the same as that of scheme (2). Therefore, the rate constant, k_{21}, used in the following analysis might include the term, $k_{23}/(k_{23}+k_{32})$. The relation between the relaxation frequency and the analytical concentrations of alcohol and water is derived as follows,[5] (on the assumption that the concentration of the hydrogen-bonded alcohol is small):

$$2\pi f_r = k_{21}[(C_e - \beta C_w + K_{12})^2 + 4\beta C_w K_{12}]^{1/2}, \qquad (6)$$

where C_e and C_w are the analytical concentrations of alcohol and water, respectively, K_{12} the equilibrium constant defined as $K_{12}=k_{12}/k_{21}$ and β the fraction of the non-hydrogen-bonded water molecules. The three parameters, k_{21}, K_{12}, and β, which were assumed to be concentration independent, were determined so as to obtain the best fit of the exprimental relaxation frequency to Eq. 6 by means of a non-linear least-mean-square. Figure 3 shows plots of the observed relaxation frequencies and theoretical curves which were calculated continuously, using the values in Table 2. For aqueous solutions of 1-propanol and 2-propanol,[5,6] the calculation was carried out using a trial-and-error method. Therefore, the recalculation was carried out using a similar computer program; almost the same values were determined (Table 2).

The concentration dependence of a maximum excess absorption per wavelength, μ_m, should also be explained in order to confirm the cause of the relaxation phenomenon. It is given by[5]

$$\mu_m = \pi \rho c^2 \Gamma(\Delta V - \alpha_p \Delta H/\rho C_p)^2/2RT, \qquad (7)$$

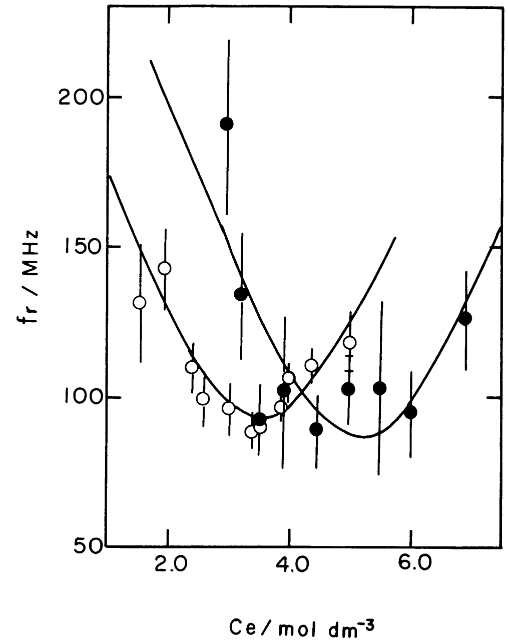

Fig. 3. Concentration dependence of the relaxation frequency for aqueous solutions of 1-methoxy-2-propanol and 3-(3-methoxypropoxy)propanol. The solid curves are the calculated ones. ●: 1-methoxy-2-propanol solution, ○: 3-(3-methoxpropoxy)propanol solution.

Table 2. Comparison of the Rate and Thermodynamic Constants for Aqueous Solutions of 1-Propanol, 2-Propanol, 1-Methoxy-2-propanol, and 3-(3-Methoxypropoxy)propanol at 25 °C

Solute	k_{21} / 10^8 mol^{-1} dm^3 s^{-1}	K_{12} / mol dm^{-3}	β	Reference
1-Propanol	0.51±0.05	4.5 ±1.0	0.141±0.013	5
2-Propanol	0.92±0.03	1.5 ±0.1	0.155±0.004	6
1-Methoxy-2-propanol	1.9 ±0.3	0.41±0.20	0.177±0.011	This work
3-(3-Methoxypropoxy)propanol	1.7 ±0.1	0.79±0.13	0.133±0.005	This work

where 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, C_p the specific heat at a constant pressure and Γ the concentration term which is dependent on the reaction. It is expressed by the following equation for the reaction under consideration:

$$\Gamma = \{1/[AB] + 1/[A] + 1/[B] - 1/([AB] + [B] + [A])\}^{-1} \tag{8}$$

In order to interpret quantitatively the concentration dependence of μ_m, the specific heat and the thermal expansion coefficient data at each concentration are necessary. However, such data are not available in the literature and the former value, unfortunately, can not be determined in our laboratory. We reluctantly examine the trends of the maximum excess absorption per wavelength qualitatively, since the most dominant term controlling the dependence may be $\rho c^2 \Gamma$. In Fig. 4, the dependences of the maximum excess absorption per wavelength and $\rho c^2 \Gamma$ on the analytical concentration are shown for 3-(3-methoxypropoxy)propanol solution. A similar profile was also found for a 1-methoxy-2-propanol solution. These trends of μ_m and $\rho c^2 \Gamma$ were quite similar and they confirmed to us that the cause of the relaxation was associated with an interaction between the alcohols and water. When the ultrasonic relaxation was assumed to be mainly due to a volume relaxation, the standard volume change of the reaction was calculated to $\Delta V \cong 4.8$ cm^3 mol^{-1} for 3-(3-methoxypropoxy)propanol solution. However, it increased monotonously with the analytical concentration. This means that the contribution from thermal relaxation can not be ignored.

It may still be questioned whether the water molecules participate in the observed relaxation process. This may be clarified by changing the water concentration at a fixed solute concentration. In the MHz frequency range, there is no relaxational absorption in methanol and DMSO. Also, their mixtures with water indicate no relaxational absorption up to 220 MHz.[16–18] These solvents may therefore be suitable for the above experiments. Figure 5 shows the absorption spectra in a methanol and a methanol–water mixed 3-(3-methoxypropoxy)propanol at 3.50 mol dm^{-3}. In the methanol solution of 3-(3-methoxypropoxy)propanol, no relaxational absorption was

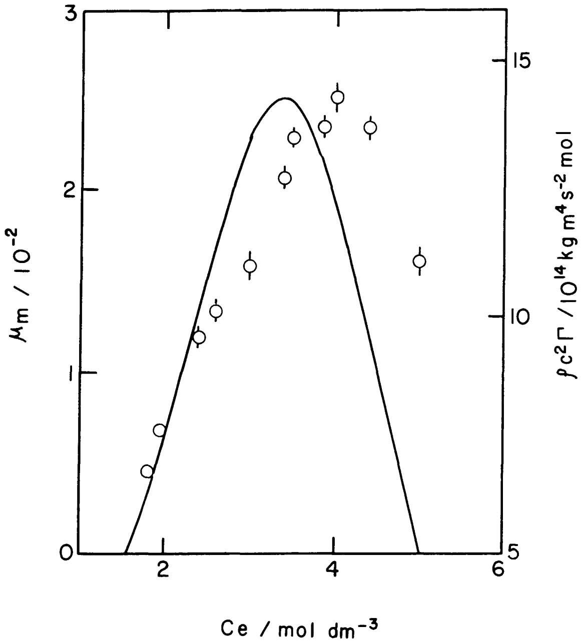

Fig. 4. The plots of the experimental μ_m (ф) and the calculated $\rho c^2 \Gamma$ (solid line) as a function of the analytical concentration for 3-(3-methoxpropoxy)-propanol solution at 25 °C.

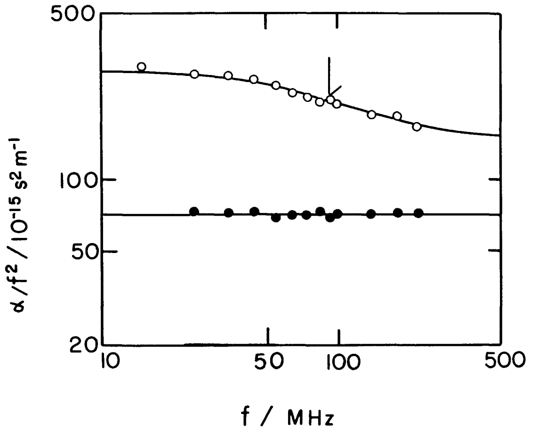

Fig. 5. Ultrasonic absorption spectra for the methanol solution (●) and for the solution with 25 vol% methanol as solvent (○) of 3-(3-methoxypropoxy)-propanol at 3.50 mol dm^{-3}.

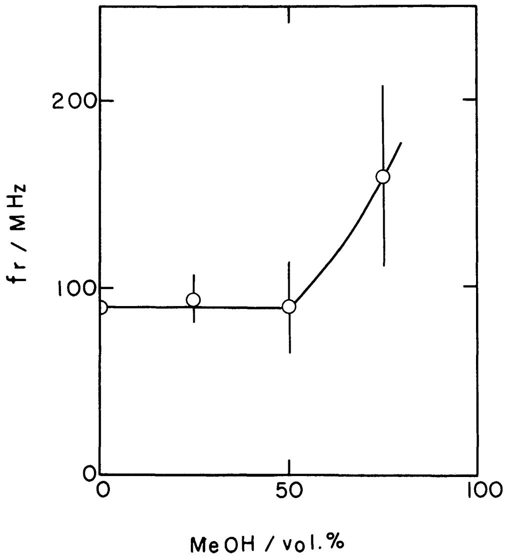

Fig. 6. The methanol concentration dependence of the relaxation frequency for 3-(3-methoxypropoxy)-propanol solution at 3.50 ml dm^{-3}. The concentration of the methanol is shown as the volume% of the mixed solvent.

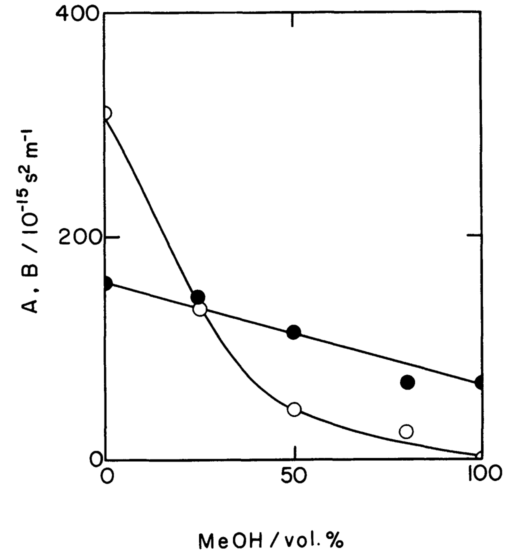

Fig. 7. The methanol concentration dependences of the amplitude of the relaxational absorption (○) and the background absorption (●) for 3-(3-methoxy-propoxy)propanol at 3.50 mol dm^{-3}.

observed. Upon adding the water, the relaxational absorption appeared. Figure 6 shows the trend of the relaxation frequency and Fig. 7 those of the amplitude of the relaxational absorption and the background absorption in the methanol–water mixed solvent at a fixed concentration of 3-(3-methoxypropoxy)propanol. With increasing the methanol content, the relaxation frequency may be approximately constant up to 50 vol% of methanol and then increases while the amplitude of the relaxational absorption decreases, as can be seen in these figures. These results may be because the water structure is broken when methanol is added, and the number of non-hydrogen-bonded water molecules may still be maintained, even if the analytical concentration of water decreases. A further addition of methanol would change the water structure even more. Figure 8 shows a similar ultrasonic absorption spectra in a DMSO and DMSO–water mixed solvent of 3-(3-methoxypropoxy)propanol. No relaxational absorption was found in the DMSO solution, though a relaxational phenomenon was observed in the mixed solvent. Figures 9 and 10 show the dependences of the relaxation frequency, the amplitude of the relaxational absorption and the background absorption on the DMSO content. The relaxation frequency showed a minimum, as can be seen in Fig. 9, and the amplitude of the relaxational absorption indicated a maximum. These trends are quite different from those found in a methanol–water mixed solvent. This may be because the hydrogen-bonding ability of DMSO (aprotic solvent) with water is not the same as that of methanol. A quantitative analysis for these mechanisms is beyond our objective in this paper.

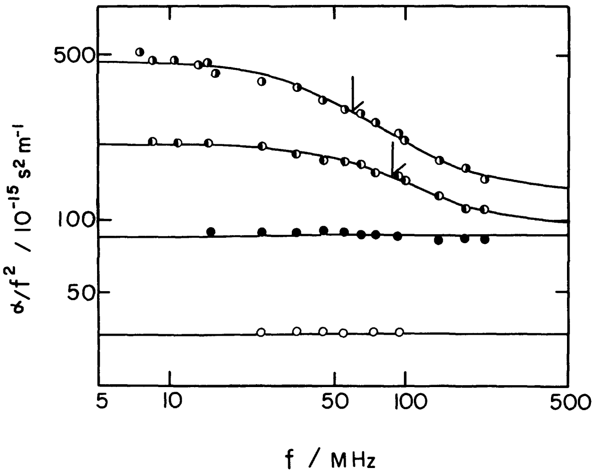

Fig. 8. Ultrasonic absorption spectrum of DMSO (○) and those of DMSO solution (●) 25 vol% DMSO solution (◐), and the 50 vol% DMSO solution (◑) at 3.50 mol dm^{-3} 3-(3-methoxypropoxy)propanol. The concentration of DMSO is the volume% of the mixed solvent.

The above results regarding organic and mixed solvents indicate that water molecules are surely associated with the observed relaxation process in aqueous media.

It is interesting to compare the obtained results with those of various alcohol solutions. We first compare the results for 1-methoxy-2-propanol and 2-propanol solutions. As can be seen in the Table 2, the β parameter for a 1-methoxy-2-propanol solution is

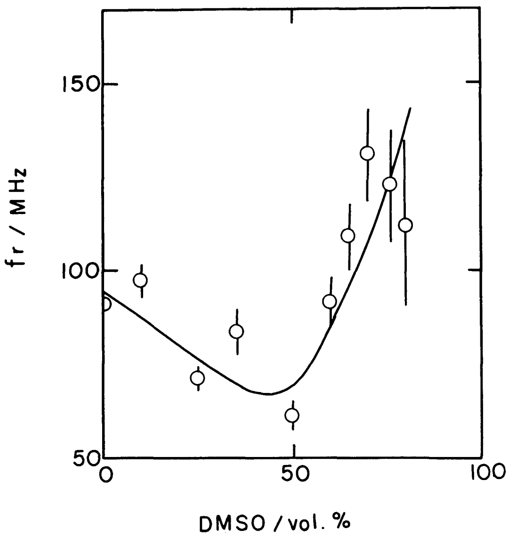

Fig. 9. The DMSO concentration dependence of the relaxation frequency for 3-(3-methoxypropoxy)propanol at 3.50 mol dm^{-3}.

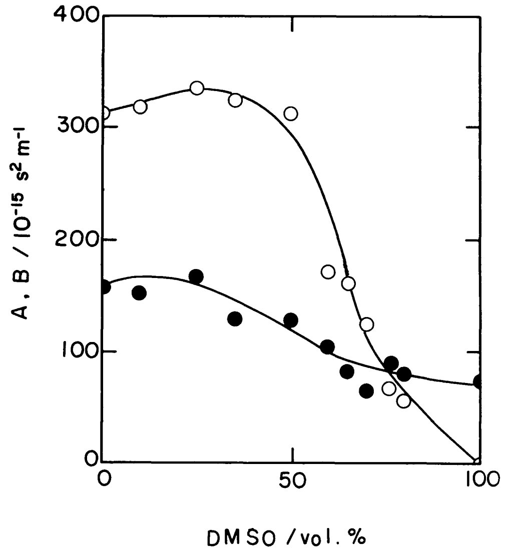

Fig. 10. The DMSO concentration dependence of the amplitude of the relaxational absorption (○) and the background absorption (●) for 3-(3-methoxypropoxy)propanol at 3.50 mol dm^{-3}.

slightly larger than that for a 2-propanol solution. This means that the number of hydrogen-bonded water molecules decreases in an aqueous solution when a methoxyl group is introduced in 2-propanol. That is, a 1-methoxy-2-propanol molecule may have more hydrophilicity than that of 2-propanol. Then, the methoxyl group is estimated to act as a water structure breaker. This may be consistent with the interpretation regarding the result in a mixed solvent of water and methanol for 3-(3-methoxypropoxy)-propanol. It is noteworthy that the association rate constant k_{21}, for the 1-methoxy-2-propanol solution is larger than that of the 2-propanol solution. With increasing the hydrophilicity of the solute molecule with a similar functional group, the association rate constant tends to increase, as can be seen from a comparison of the β values and the rate constants in solutions of 2-propanol and 2-methyl-2-propanol.[7] However, this does not always hold, especially for molecules which greatly differ in sizes.

Secondly, the results for 1-methoxy-2-propanol and 3-(3-methoxypropoxy)propanol are considered. The difference between the β values for these two solutions is about 0.04 lower, but much larger when it is compared with that between 2-propanol and 1-methoxy-2-propanol solutions (it is about 0.02 increase). These differences show that the introduction of the propoxyl group allows the hydrogen-bonded water molecule to increase: that is, the propoxyl group is estimated as acting as a water structure promoter.

We have been speculating that the ethoxyl group acts as a slight water structure promoter due to the small differences in the β parameters in aqueous solutions of three alcohols with different numbers of oxyethylene group.[8] It may therefore be concluded from the magnitudes of the β parameters that the methoxyl group acts as a water structure breaker, the ethoxyl group acts very slightly as the promoter and the propoxyl group acts as a strong structure promoter.

It should be noticed that two relaxational absorptions are observed in the aqueous solution of 1-propanol. One of them is due to a perturbation of the equilibrium, as in Eq. 2. The other may be associated with a molecular aggregation reaction due to a hydrophobic interaction.[5] This experimental evidence might indicate that 1-propanol acted as a better water structure promoters than did 1-methoxy-2-propanol, 3-(3-methoxypropoxy)propanol and 2-propanol. Actually, the β parameter for 1-methoxy-2-propanol and 2-propanol is larger than that for 1-propanol, which is consistent with the above prediction. However, the β value for 3-(3-methoxypropoxy)propanol is smaller than that for 1-propanol, and in the former solution, only a single relaxational process associated with the alcohol water interaction was observed. This situation might be because the ether oxygens in 3-(3-methoxypropoxy)propanol prevent the formation of an aggregate due to a hydrophobic interaction by their electrical repulsion. However, the ability as a water structure former of 3-(3-methoxypropoxy)propanol is still higher than that of 1-propanol, since the β value indicates the characteristic of water as a whole in aqueous media.

These conclusions led us to speculate concerning the appearance of an ultrasonic relaxational absorption in aqueous solutions of familiar alcohols. In the frequency range below 200 MHz, no excess absorption has been found in a methanol aqueous solution, even at 0 °C.[16,17] In an ethanol aqueous solution, the relaxation process with a relaxation frequency around 100 MHz has been reported below room temperature.[19] On the other hand, a clear relaxation process is observed in the range around 50 MHz for aqueous solutions of propanol.[3,5,6] From speculation concerning the present study, the relaxation process in the aqueous solution of methanol would be expected at a very high frequency range since the methanol breaks the water structure, a lot of non-hydroen-bonded water molecules exist and the value of the rate constant for the interaction may be high. In an ethanol solution, the rate associated with the interaction process may decrease compared with that for methanol, and the water structure is still remained, or slightly promoted. The relaxation process may thus appear in a lower frequency range than that in a methanol solution. For propanol solutions, the solute creates the water structure and the rate decreases further. Then, the ultrasonic relaxational process appears in the MHz frequency range. This kind of correlation between the hydrophobicity of the solutes and the water structure has been speculated in static studies.[1,20] The present investiation has supported such consideration dynamically. Zana et al. have recently reported that in the MHz frequency range, there are three relaxation processes in aqueous solutions of alcohol when the solute consists of a relatively large hydrophobic group (1-butanol).[21] We consider that the present excess absorption in this study is one of them and that the aggregation process for 1-methoxy-2-propanol and 3-(3-methoxypropoxy)propanol may not be found, due to the smaller hydrophobicity of the molecules and the existence of the ether oxygen.

Finally, it should be stated why we do not apply the fluctuation model in order to interpret the ultrasonic absorption mechanisms in aqueous solutions of alcohols. According to the proposed theories, the α/f^2 values would increase with decreasing frequency. However, the present results indicate that an almost frequency-independent region is observed at less than 10 MHz, as can be seen in Fig. 2. In order to use the fluctuation model, it seems to be necessary to measure the absorption coefficients over very wide frequency range (more than 3 orders), especially in the range less than 5 MHz. However, the absolute values of the absorption coefficients may not be precisely determined in a such low-frequency region.

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