

## ULTRASONIC AND HYPERSONIC INVESTIGATIONS OF STRUCTURAL RELAXATION IN AQUEOUS SOLUTIONS OF HEXAMETHYLPHOSPHORTRIAMIDE

PIOTR MIECZNIK

Chair of Acoustics, Mickiewicz University

(60-769 Poznań, ul. Matejki 48/49)

Measurements of the absorption coefficient of ultrasonic waves in the frequency range 10-100 MHz and of the propagation velocity of ultra- and hypersonic waves in aqueous solutions of hexamethylphosphortriamide (HMPT) were carried out. In addition the density of the solutions and the coefficient of shear viscosity was measured. On the basis of the quantities measured, the coefficient of bulk viscosity, relaxation parameters of the process observed and, on the basis of the theory of compressibility relaxation, change in free energy and volume between two structural states were measured.

The analysis of the results of measurements of the absorption coefficient of ultrasonic waves, depending on the frequency, temperature, and composition of solutions, shows the presence in aqueous solutions of HMPT of a relaxation process related to the formation and disintegration of clathrate structures with the composition  $\text{HMPT} \cdot 17 \text{H}_2\text{O}$ . On the basis of compressibility relaxation theory, it was shown that the process of structural relaxation is related to a change in volume  $\Delta V$  and in free energy  $\Delta F$  between two structural states.

### 1. Introduction

The character of changes in the absorption coefficient of ultrasonic waves depending on the concentration of a solution varies greatly for a great number of liquid mixtures [1, 6, 11, 10, 3]. An interesting group is made of aqueous solutions of non-electrolytes for which the absorption coefficient of ultrasonic waves reaches a maximum for a strictly defined composition [2, 5, 11-14]. The occurring maximum of the absorption coefficient of ultrasound indicates the presence in the solution of a relaxation process related to the formation and disintegration of strictly defined structures (molecular complexes) between water molecules and the molecules of the medium dissolved.

The most systematic investigations which gave most insight into the problem of the character of relaxation occurring in aqueous solutions of non-electrolytes were performed by ANDREAE, EDMONDS, and MCKELLAR [2]. They formulated theses on the ultrasonic relaxation for non-electrolytes dissolved in water, under the assumption that the processes observed in aqueous solutions of some amines consist in the formation and disintegration of free complexes, in which a strictly defined number of water molecules corresponds to one non-electrolyte molecule. Accordingly, these authors relate the additional absorption of acoustic waves to the interaction of the type solvent — dissolved substance. In turn, NISHIKAWA, YASUNAGA, and TAKAHASHI [9] explain the additional absorption of ultrasonic waves in aqueous solutions of amyloamine and butyloamine on the basis of the reaction of association — dissociation of molecules and hydrolysis.

It is also significant whether the relaxation process observed in aqueous solutions of non-electrolytes is mainly conditioned by a change in the volume of the system (structural relaxation) or a process dependent on temperature changes (thermal relaxation). ANDREAE, EDMONDS, and MCKELLAR assume that the relaxation observed in aqueous solutions of non-electrolytes is thermal in character; in theoretical considerations, however, they also assume that the process of sound propagation in aqueous solutions of non-electrolytes is isothermal, obtaining as a result the equation for the maximum coefficient of sound absorption,  $\mu_{\max}$ , dependent on the change in volume of the system  $\Delta V$ , in the course of the reaction. The question remains as to whether the relaxation process observed in aqueous solutions of non-electrolytes is in fact a thermal process or a process of structural (compressibility) relaxation.

The present paper attempts, on the basis of the results of measurements of the absorption coefficient of ultrasonic waves and the velocity of the propagation of hypersonic waves, to determine what sort of relaxation processes occur in aqueous solutions of HMPT.

The previous data on the structure and interactions in aqueous solutions of HMPT [7] permit the conclusion that within the temperature range ( $-30^{\circ}\text{C}$  —  $+50^{\circ}\text{C}$ ) and the concentration of solutions (0.1 molar fraction HMPT), the occurrence of the effect of molecular relaxation process on the absorption of ultrasonic waves can be expected.

## 2. Properties of aqueous solutions of HMPT

On the basis of both the measurements of the propagation velocity of an ultrasonic wave at a frequency of 15 MHz and of density in aqueous solutions of HMPT ( $[(\text{CH}_3)_2\text{N}]_3\text{OP}$ ), depending on temperature and concentration, and on the calculated values of adiabatic compressibility of solutions, partial molar volumes, and partial molar compressibilities of components in the solution,

it was concluded that within the concentration range 0.04 ( $-30^{\circ}\text{C}$ )-0.08 ( $+50^{\circ}\text{C}$ ) molar fraction of HMPT a stabilization of the structure of a solution occurs as a result of the formation of clathrate structures with the composition HMPT · 17 H<sub>2</sub>O [7]. This phenomenon should become clear in measurements of the absorption coefficient of ultrasonic waves, depending on the frequency of the waves and the concentration of solutions investigated.

### 3. Method

The measurements of the absorption coefficient of ultrasonic waves were made using an ultrasonic high-frequency system type US-4 manufactured by the Institute of Fundamental Technological Research, which permitted measurements at frequencies of 10, 13.3, 16.6, 20, 30 and 47 MHz and using a modified high-frequency ultrasonic system which permitted measurements at frequencies of 60 and 100 MHz. The measurements of the absorption coefficient of ultrasonic waves in the range 10-47 MHz were taken using broadband transducers, and at frequencies of 60 and 100 MHz using resonance transducers at a fundamental frequency of 20 MHz. In the detective channel of the signal at frequencies of 60 and 100 MHz attachments, which functioned as preamplifiers of high-frequency signals and transformed frequencies of 60 and 100 MHz to 10 MHz, were additionally used. As a result, detection and amplification in the receiver occurred as low as a frequency of 10 MHz. The high-frequency ultrasonic system was modified at the Chair of Acoustics, Adam Mickiewicz University. The relative error of measurements of the absorption coefficient of ultrasonic waves did not exceed 10 percent.

The measurements of the propagation velocity of hypersonic waves were taken using the Mandelsztam-Brillouin light scattering method. The measurement system used was described in paper [8].

Moreover, additional measurements of the coefficients of shear viscosity, density, and propagation velocity of ultrasonic waves were taken. The coefficient of shear viscosity was measured using a type BH2 Hoeppler viscosimeter, while density was measured by a method using the Archimedes law (of the submerged float) with relative accuracies of 0.5 and 0.02 percent. The measurements of the propagation velocity of ultrasonic waves were taken using an impulse-phase interferometer at a frequency of 12 Mz, with error not exceeding 0.1 percent. The medium investigated was thermostated with an accuracy of up to  $0.05^{\circ}\text{C}$  and controlled using a copper - constantan thermocouple. The investigations used HMPT with purity degree recognized by the producer (E. Merck, Darmstadt) as "pure for analysis" and double distilled water.

The measurements of the frequency of ultrasonic waves were taken using a type CZE-30 numerical frequency meter with an accuracy of 100 Hz over the range 10-100 MHz.

#### 4. Experimental results and their analysis

The measurements of the absorption coefficient of ultrasonic waves for  $T = +15^\circ\text{C}$  were taken over the frequency range 10-100 MHz and showed the stability of the quantity  $\alpha/f^2$  over the whole range investigated. In turn, the character of changes in the quantity  $(\alpha/f^2)_{\text{exp}}$  relative to concentration (Fig. 1) shows the presence of a sharp maximum for a strictly defined concentration  $k_{\text{mo}} \cong 0.1$ . Fig. 1 also shows the plot of the dependence  $(\alpha/f^2)_{\text{cl}}$  on concentration [11], which defines the contribution of the compressible viscosity to the total absorption of acoustic waves. The maximum in the dependence  $(\alpha/f^2)_{\text{ad}} = (\alpha/f^2)_{\text{exp}} - (\alpha/f^2)_{\text{cl}}$  on concentration which occurs for the concentration  $k_{\text{mo}} = 0.1$  shows in turn that in aqueous solutions of HMPT there is some relaxation process related to volume viscosity relaxation. In order to investigate this process the absorption coefficient of ultrasonic waves was measured in the temperature range  $-30^\circ\text{C} - +15^\circ\text{C}$  and in the frequency range 10-100 MHz. Fig. 2 shows the experimental results of the measurement of the absorption coefficient depending on the concentration  $k_{\text{mo}}$  (as an example, for  $T = -30^\circ\text{C}$ ). A sharp maximum occurs for a strictly defined composition and the value of the maximum decreases as the frequency of ultrasonic waves increases which shows that in aqueous solutions of HMPT a relaxation process occurs in the frequency range 10-100 MHz. The maximum of the absorption coefficient corresponds to a concentration close to 0.1 molar fraction of HMPT.

The relation  $(\alpha/f^2)_{\text{cl}} = f(k_{\text{mo}})$  also shows a maximum for a strictly defined concentration (Fig. 3), which indicates that in aqueous solutions of HMPT both the process of volume relaxation and that of shear viscosity occur. However, since the experiment reveals a single relaxation process, it can be expected that both processes have similar values of the relaxation time and cannot be distinguished in the range of measurement error. The calculations of the absorption coefficient related to the so-called classical absorption used the quantities listed in Table 1 (the values of the propagation velocity of ultrasonic waves  $c$ , the shear viscosity  $\eta_s$  and the density of the medium  $\rho$ ).

The concentration dependencies of the absorption coefficient of ultrasonic waves (Fig. 2) show that a relaxation process also occurs in media investigated. However, only the dependence of the quantity  $\alpha/f^2$  on frequency permits the determination of the relaxation time of the above process. The dependencies of  $\alpha/f^2$  on frequency for specific concentration and temperature of the medium are given in Figs. 4-8.

It can be noted that for temperatures below  $-20^\circ\text{C}$  and in the investigated frequency range of ultrasonic waves and in the concentration range of the solution there is the dependence of the quantity  $\alpha/f^2$  on frequency, indicating the presence of a single relaxation process. In order to determine accurately the relaxation frequency, the numerical method proposed by BOCH [4] was used. On the basis of knowledge of the relaxation frequencies  $f_r$  and



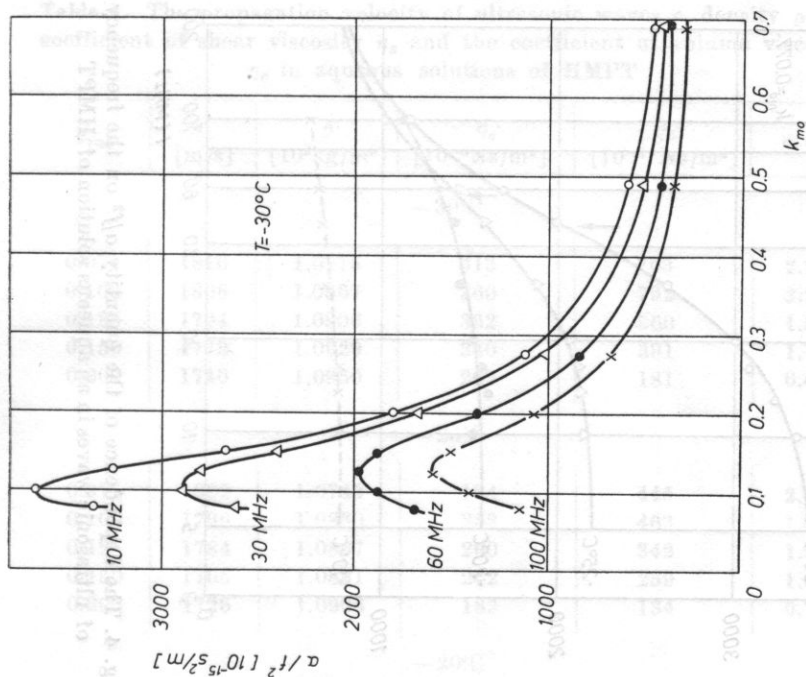


Fig. 2. The dependence of the quantity  $\alpha/f^2$  on concentration in aqueous solutions of HMPT

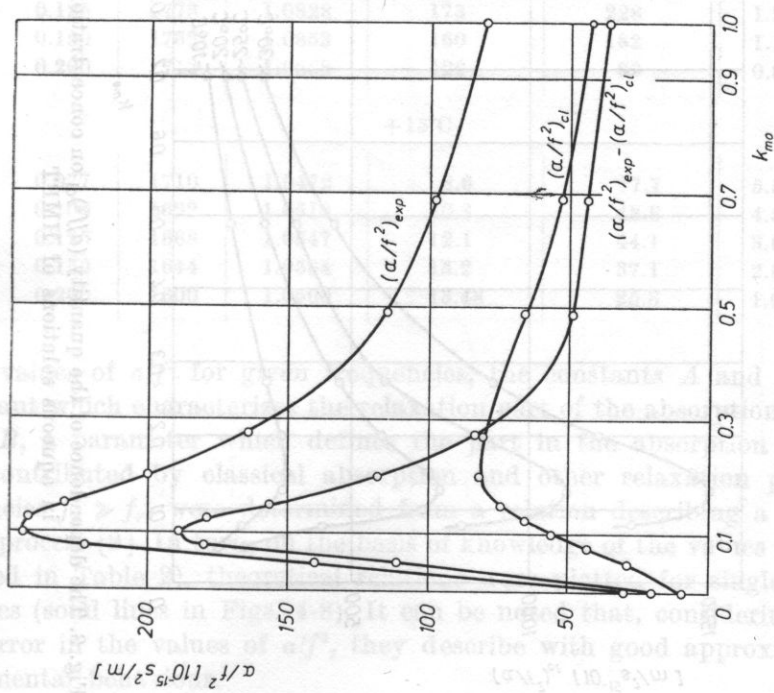


Fig. 1. The dependence of the quantity  $\alpha/f^2$  on concentration in aqueous solutions of HMPT for  $T = +15^\circ\text{C}$

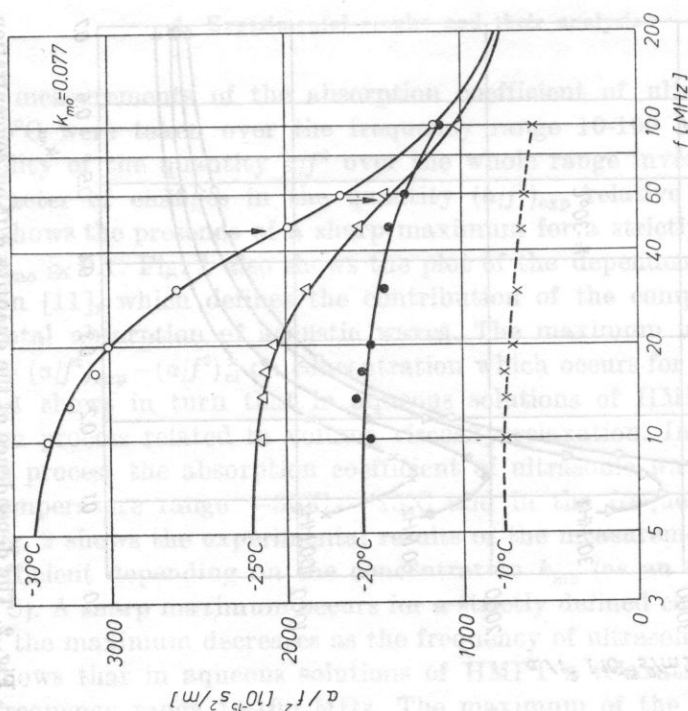


Fig. 4. The dependence of the quantity  $a/f^2$  on the frequency of ultrasonic waves in an aqueous solution of HMPT

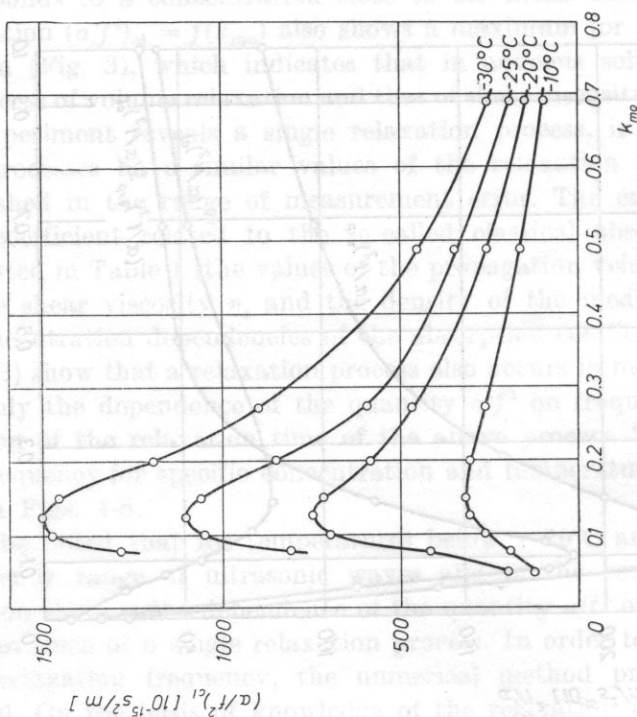


Fig. 3. The dependence of the quantity  $(a/f^2)_{cl}$  on concentration in aqueous solutions of HMPT

**Table 1.** The propagation velocity of ultrasonic waves  $c$ , density  $\rho$ , the coefficient of shear viscosity  $\eta_s$  and the coefficient of volume viscosity  $\eta_v$  in aqueous solutions of HMPT

$k_{mo}$	$c$ [m/s]	$\rho$ [ $10^3 \text{kg/m}^3$ ]	$\eta_s$ [ $10^{-3} \text{Ns/m}^2$ ]	$\eta_v$ [ $10^{-3} \text{Ns/m}^2$ ]	$\eta_v/\eta_s$
-30°C					
0.077	1810	1.0775	313	703	2.25
0.100	1806	1.0867	360	792	2.20
0.125	1794	1.0906	362	560	1.55
0.150	1778	1.0929	340	391	1.15
0.200	1740	1.0950	262	181	0.69
-25°C					
0.077	1802	1.0743	194	445	2.29
0.100	1796	1.0830	252	463	1.83
0.125	1784	1.0867	260	342	1.32
0.150	1765	1.0891	242	259	1.07
0.200	1726	1.0908	183	134	0.73
-20°C					
0.077	1794	1.0711	103	348	3.39
0.100	1786	1.0792	165	561	3.40
0.125	1773	1.0828	173	228	1.32
0.150	1752	1.0853	160	182	1.14
0.200	1713	1.0868	122	99	0.81
+15°C					
0.077	1710	1.0472	8.6	47.7	5.58
0.100	1692	1.0519	10.8	48.8	4.50
0.125	1668	1.0547	12.1	44.1	3.65
0.150	1644	1.0564	13.2	37.1	2.80
0.200	1600	1.0560	13.48	25.8	1.92

of the values of  $a/f^2$  for given frequencies, the constants  $A$  and  $B$  ( $A$  being a constant which characterizes the relaxation part of the absorption of acoustic wave;  $B$ , a parameter which defines the part in the absorption of acoustic wave contributed by classical absorption and other relaxation processes at frequencies  $f_r' \geq f_r$ ) were determined from a relation describing a single relaxation process [2]. In turn, on the basis of knowledge of the values of  $f_r$ ,  $A$  and  $B$  (listed in Table 2), theoretical relations were plotted for single relaxation processes (solid lines in Figs. 4-8). It can be noted that, considering measurement error in the values of  $a/f^2$ , they describe with good approximation the experimental behaviour.

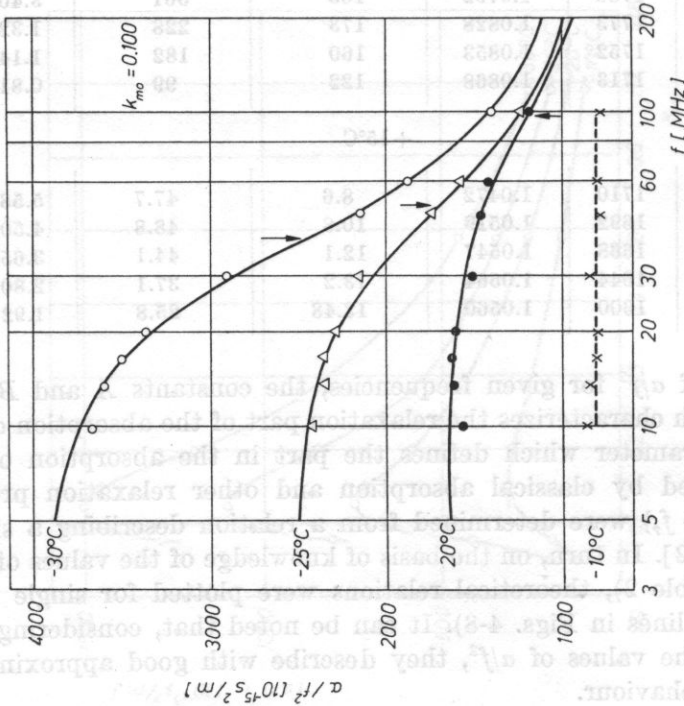


Fig. 5. The dependence of the quantity  $\alpha/f^2$  on the frequency of ultrasonic waves in an aqueous solution of HMPT

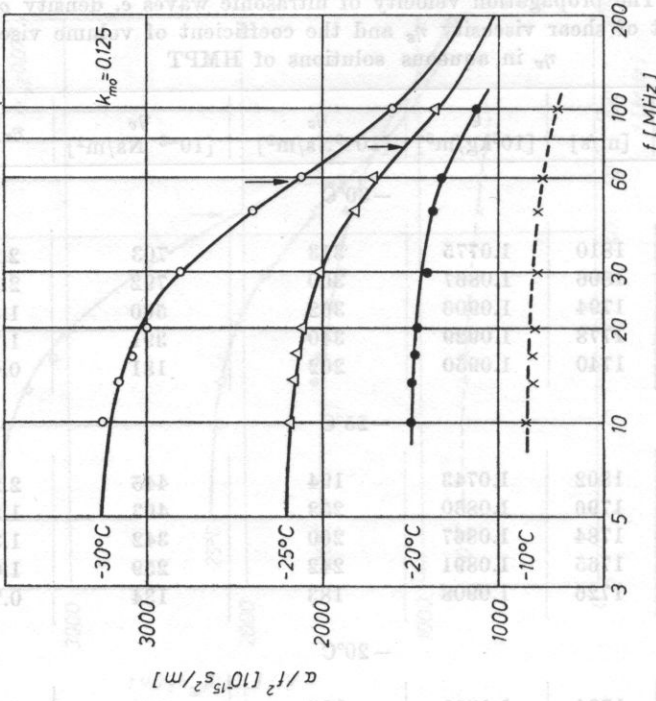


Fig. 6. The dependence of the quantity  $\alpha/f^2$  on the frequency of ultrasonic waves in an aqueous solution of HMPT



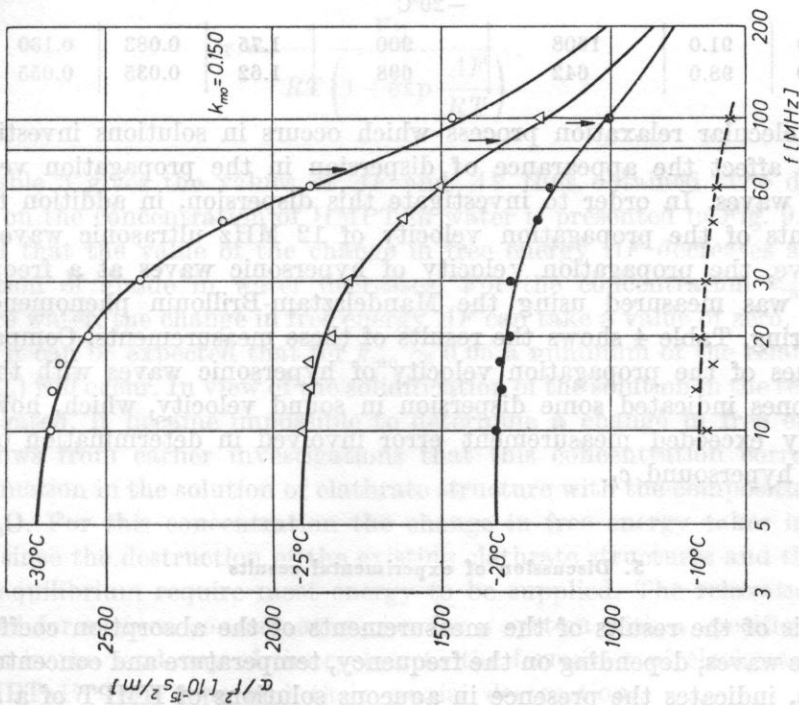


Fig. 7. The dependence of the quantity  $a/f^2$  on the frequency of ultrasonic waves in an aqueous solution of HMPT

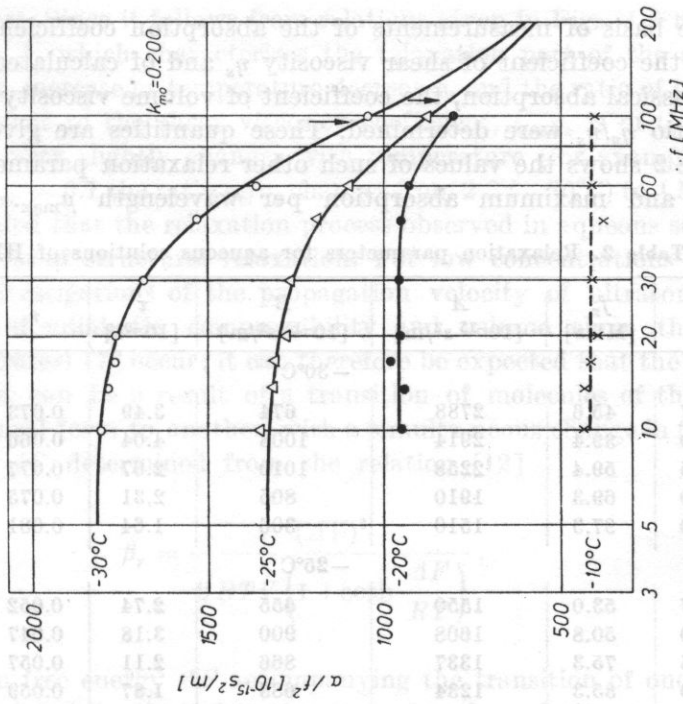


Fig. 8. The dependence of the quantity  $a/f^2$  on the frequency of ultrasonic waves in an aqueous solution of HMPT

On the basis of measurements of the absorption coefficient of ultrasonic waves and the coefficient of shear viscosity  $\eta_s$  and of calculated values of the so-called classical absorption, the coefficient of volume viscosity  $\eta_v$  of solutions and the ratio  $\eta_v/\eta_s$  were determined. These quantities are given in Table 1, while Table 2 shows the values of such other relaxation parameters as relaxation force and maximum absorption per wavelength  $\mu_{\max}$ .

Table 2. Relaxation parameters for aqueous solutions of HMPT

$k_{\text{mo}}$	$f_r$ [MHz]	$A$ [ $10^{-15}$ s <sup>2</sup> /m]	$B$ [ $10^{-15}$ s <sup>2</sup> /m]	$\tau$ [ $10^{-9}$ s]	$r$	$\mu_{\max}$
-30°C						
0.077	45.6	2788	674	3.49	0.073	0.115
0.100	39.4	2914	1008	4.04	0.066	0.104
0.125	59.4	2258	1010	2.67	0.077	0.121
0.150	69.3	1910	805	2.31	0.075	0.117
0.200	97.3	1510	306	1.64	0.081	0.128
-25°C						
0.077	53.0	1550	655	2.74	0.052	0.082
0.100	50.8	1608	900	3.18	0.047	0.074
0.125	75.3	1337	866	2.11	0.057	0.090
0.150	85.3	1234	683	1.87	0.059	0.093
0.200	111.0	1002	328	1.43	0.061	0.096
-20°C						
0.100	91.0	1608	900	1.75	0.083	0.130
0.150	98.0	642	698	1.62	0.035	0.055

The molecular relaxation process which occurs in solutions investigated should also affect the appearance of dispersion in the propagation velocity of acoustic waves. In order to investigate this dispersion, in addition to the measurements of the propagation velocity of 12 MHz ultrasonic waves discussed above, the propagation velocity of hypersonic waves at a frequency of 5 GHz was measured using the Mandelsztam-Brillouin phenomenon of light scattering. Table 4 shows the results of these measurements. Comparison of the values of the propagation velocity of hypersonic waves with that of ultrasonic ones indicated some dispersion in sound velocity, which, however, only slightly exceeded measurement error involved in determination of the velocity of hypersonic  $c_h$ .

### 5. Discussion of experimental results

Analysis of the results of the measurements of the absorption coefficient of ultrasonic waves, depending on the frequency, temperature and concentration of solutions, indicates the presence in aqueous solutions of HMPT of a single

relaxation process. Since it follows from relations given in Figs. 4-8 that values of the constant  $A$ , which characterizes the relaxation part of the absorption of acoustic wave, increase as temperature decreases, and the ratio of the volume viscosity coefficient to the shear viscosity coefficient,  $\eta_v/\eta_s$  is of the order of unity and only very slightly changes with temperature (for example, for the concentration  $k_{mo} = 0.1$  the ratio  $\eta_v/\eta_s$  changes from 2.2 ( $-30^\circ\text{C}$ ) to 4.5 ( $+15^\circ\text{C}$ ); it can be concluded that the relaxation process observed in aqueous solutions of HMPT is a process of structural relaxation. For low concentrations of amides in water, the investigations of the propagation velocity of ultrasonic waves, the coefficient of adiabatic compressibility and volume show that spatial structures (clathrates) [7] occur; it can therefore be expected that the relaxation process observed can be a result of a transition of molecules of the medium from one structural form to another, with a simultaneous change in the volume of the medium  $\Delta V$  determined from the relation [12]

$$\beta_r = \frac{(\Delta V)^2}{2RTV \left(1 + \cosh \frac{\Delta F}{RT}\right)}, \quad (1)$$

and a change in free energy  $\Delta F$  accompanying the transition of one structure into another, related to the relaxation time by the relation

$$\tau = \frac{V\eta_s}{RT \left(1 + \exp \frac{\Delta F}{RT}\right)}. \quad (2)$$

Table 3 gives the values of  $\Delta F$  and  $\Delta V$  thus obtained. The dependence of  $\Delta F$  on the concentration of HMPT in water is presented in Fig. 9. It can be noticed that the value of the change in free energy  $\Delta F$  decreases as the concentration of amide in water decreases. For the concentration  $k_{mo} = 0$ , i.e. for pure water, the change in free energy  $\Delta F$  can take a value of zero, and accordingly it can be expected that for  $k_{mo} \cong 0.05$  a minimum of the relation  $\Delta F = f(k_{mo})$  will occur. In view of the solidification of the solution in the temperature investigated, it became impossible to determine a change in free energy  $\Delta F$ . It follows from earlier investigations that this concentration corresponds to the formation in the solution of clathrate structure with the composition  $\text{HMPT} \cdot 17 \text{H}_2\text{O}$ . For this concentration the change in free energy takes its extreme value, since the destruction of the existing clathrate structures and their return to an equilibrium require most energy to be supplied. The relaxation process observed for a given concentration, i.e. for a system with a specific structure, consists in the tendency of the system to the formation of clathrate structure of  $\text{HMPT} \cdot 17 \text{H}_2\text{O}$  type and their partial destruction.

**Table 3.** Results of calculation of the quantities  $\Delta F$ ,  $\Delta V$ ,  $\beta_r$ ,  $\beta_\infty$  and  $c_\infty$  based on the theory of compressibility relaxation

$k_{mo}$	$\Delta F$ [ $10^6$ J/kmol]	$\Delta V$ [ $10^{-6}$ m <sup>3</sup> /mol]	$\beta_r$ [ $10^{-12}$ m <sup>2</sup> /N]	$\beta_\infty$ [ $10^{-12}$ m <sup>2</sup> /N]	$c_\infty$ [m/s]
-30°C					
0.077	-2.78	2.39	16.2	267	1864
0.100	-1.94	2.32	17.0	265	1863
0.125	0.60	2.22	17.0	268	1850
0.150	1.20	2.20	14.3	275	1823
0.200	1.96	2.17	10.1	292	1770
-25°C					
0.100	-3.19	2.31	11.9	274	1836
0.125	0.19	1.98	13.6	276	1826
0.150	0.75	1.99	12.0	283	1801
0.200	1.29	1.93	8.9	298	1754
-20°C					
0.100	-1.84	2.13	14.1	276	1831
0.150	-0.41	1.84	10.2	290	1782

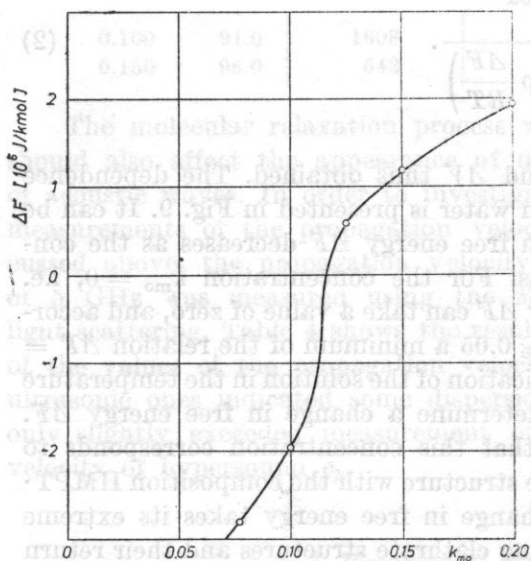


Fig. 9. The dependence of a change in free energy  $\Delta F$  on concentration in aqueous solutions of HMPT for  $T = -30^\circ\text{C}$

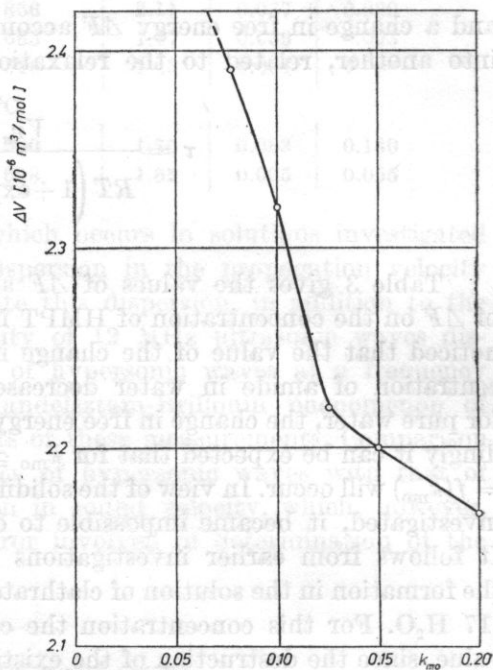


Fig. 10. The dependence of a change in volume  $\Delta V$  between two structural states on concentration in aqueous solutions of HMPT for  $T = -30^\circ\text{C}$

Reaction of this type causes a change in volume of the system  $\Delta V$ , whose magnitude depending on concentration is shown in Fig. 10. It follows from this dependence that for the concentration  $k_{\text{mo}} \cong 0.05$  the change in volume between two states takes an extreme (maximum) value, since for  $k_{\text{mo}} = 0$ , i.e. for pure water,  $\Delta V = 0$ . Another conclusion also follows, that the relaxation process revealed consists in destruction and formation in the solution of clathrate structures with the composition HMPT·17 H<sub>2</sub>O, since the difference in volume between the two structural states takes a maximum value for the characteristic concentration.

Analysis of results of measurements of the velocity of hypersound shows slight dispersion in the velocity of sound propagation. Similar results were, however, obtained for  $T = -30^\circ\text{C}$  on the basis of the theory of compressibility relaxation by determining the value  $c_\infty$ , i.e. sound propagation velocity for

**Table 4.** The velocity of hypersound in aqueous solutions of HMPT. The scattering angle  $\theta = 90^\circ$ ,  $D = 5876 \text{ \AA}$ ,  $\delta\nu_L = 0.079 \text{ cm}^{-1}$ ,  $T = 15^\circ\text{C}$

$k_{\text{mo}}$	$\Delta\nu \text{ [cm}^{-1}\text{]}$	$n_D$	$f \text{ [GHz]}$	$c_h \text{ [m/s]}$	$c_0 \text{ [m/s]}$	$\delta c_h \text{ [m/s]}$
0.050	0.1758	1.38052	5.3	1710	1704	16
0.100	0.1807	1.40492	5.4	1727	1691	16
0.125	0.1788	1.41375	5.4	1698	1669	18
0.200	0.1735	1.42851	5.2	1630	1600	19
0.492	0.1595	1.44641	4.8	1480	1458	19
1.000	0.1437	1.45470	4.3	1326	1366	20

$f \rightarrow \infty$ . Dispersion in this case does not exceed 60 m/s, i.e. 4 percent (Table 3 and 1). This agreement confirms the conclusions drawn above as to the type of relaxation occurring in aqueous solutions of HMPT. In general, for aqueous solutions of non-electrolytes the dispersion of sound velocity takes low values in view of the low value of the relaxation part of the adiabatic compressibility  $\beta_r$ .

## 6. Conclusions

Analysis of results of measurements of the absorption coefficient of ultrasonic waves and of the propagation velocity of ultra- and hypersonic waves, depending on the frequency, temperature and composition of solutions, shows the presence in aqueous solutions of HMPT of a relaxation process with a specific relaxation time, related to the formation and disintegration of strictly defined spatial structures (clathrates with composition HMPT·17 H<sub>2</sub>O).

It was shown on the basis of the theory of compressibility relaxation that the relaxation process observed (structural relaxation) is related to a change in volume between two structural states and a change in free energy involved in the transition of one structure into another. The relaxation parameters of this molecular process were calculated.



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