# ACOUSTICAL INVESTIGATION OF SOME VISCOELASTIC MATERIALS AT SHEAR DEFORMATIONS

**B. Damdinov, B. Badmaev, and T. Dembelova**
Buryat Scientific Center of RAS, 670047, Sakhyanovoy 8, Ulan-Ude, RUSSIA
dababa@rambler.ru

## Abstract

A research work based on measurements of shear properties has been carried out to study low frequency viscoelastic relaxation in liquids. The liquid shear parameters have been obtained using the resonance dynamic method with piezoelectric quartz crystal. The method are based on frequency shift and attenuation change of piezoquartz. The shear modulus and shear viscosity for different liquids (polymers, solutions, oils) at frequency 74 kHz have been measured. The real and imaginary parts of the complex shear modulus have been measured. It was shown that all tested viscoelastic materials (polymer liquids, glycol's, oils, clay solutions) possesses the measurable shear elasticity modulus at small shear deformations angles (not exceed 10 minutes). The experiments show the existence of relaxation process, which is caused apparently by interaction of large groups of molecules. The relaxation time of viscoelastic process has been estimated. Cluster model of low-frequency shear elasticity has been suggested.

## Introduction

Some liquids can change viscosity at shear deformation. Investigation of shear mechanical properties of such liquids is one of the direct approaches to the study of the rearrangement processes of intermolecular structure. In the studies of the problem of structural relaxation of liquids, the deviation from Newtonian behavior is of prime interest [1,2]. Earlier it was shown that all liquids, regardless of their viscosity and polarity, exhibit complex shear elasticity at the frequencies of shear oscillations about $10^5$ Hz [3]. According to the existing concept of the nature of liquids, this effect must be observed only at the frequencies of $10^{10}$ – $10^{12}$ Hz evaluated by the frequency of liquid particle jumps from the temporary equilibrium states [4]. It is also assumed that the shear elasticity of liquid should be of magnitude of the order of the shear elasticity of a corresponding solid. Investigations of low-frequency shear elasticity of liquids by the resonance method using of a piezoelectric quartz device showed that this phenomenon is unrelated to the properties of the boundary layers of liquid, since there is no dependence of the shear modulus on the thickness of the liquid interlayer [3]. The fact that the revealed phenomenon is a bulk property of liquid confirms the possibility of the shear wave propagation in liquid.

## Method

Studying of dynamical properties of liquids was carried out by resonance method with using a piezoquartz. The resonance method for measuring the shear elasticity consists in the following: a piezoquartz crystal performs longitudinal (axial) oscillations at the basic resonance frequency, and its horizontal surface performing tangential oscillations supports a liquid interlayer covered by a strap. The cover plate with the liquid film is disposed on the one end of the piezoquartz crystal. In this case, the liquid interlayer undergoes a shear deformation, and standing shear waves should be induced in it. Parameters of the resonance curve of the piezoelectric quartz will depend on the thickness of the liquid layer. Equating piezoquartz and liquid impedances it is possible to find the following expressions for a complex shift of resonance frequency of an oscillatory system [3]:

$$\Delta\omega = \frac{SG^*\kappa}{4\pi^2 M f_0}\cdot\frac{1+\cos(2\kappa H-\varphi)}{\sin(2\kappa H-\varphi)}, \qquad (1)$$

where $G^* = G' + iG''$ is a complex shear modulus of liquid, $S$ is the area of the cover-plate, $H$ is the thickness of a liquid interlayer, $M$ is the mass of piezoquartz, $f_0$ is its resonance frequency, $\kappa^* = \beta - i\alpha$ is the attenuation coefficient, $\varphi^*$ is the complex shift of a phase when the wave is reflected from the liquid - strap interface. Separating Eq. (1) into the real and imaginary parts, we obtain two expressions for the frequency shifts:

$$\Delta f' = \frac{SG'\beta}{4\pi^2 M f_0\cos\theta}\cdot\frac{\sin 2\beta H - \tan\frac{\theta}{2}\, sh(2\beta H\cdot\tan\frac{\theta}{2})}{ch(2\beta H\cdot\tan\frac{\theta}{2}) - \cos 2\beta H},$$

$$\Delta f'' = \frac{SG'\beta}{4\pi^2 M f_0\cos\theta}\cdot\frac{\sin 2\beta H\cdot\tan\frac{\theta}{2} + sh(2\beta H\cdot\tan\frac{\theta}{2})}{ch(2\beta H\cdot\tan\frac{\theta}{2}) - \cos 2\beta H},$$

where $\tan\theta = G''/G'$ is the tangent of mechanical loss angle. These expressions show that the real and imaginary shifts of frequencies are the functions of liquid interlayer thickness. Figure 1 shows the theoretical dependence of $\Delta f'$ and $\Delta f''$ on the thickness of the liquid interlayer for $G' = 10^5$ Pa and $\tan\theta = 0.3$. As can be seen from the figure, these curves perform damped oscillations, and, with the increase in the thickness of the liquid interlayer $H\rightarrow\infty$, they tend to the limit $\Delta f_\infty'$ и $\Delta f_\infty''$, respectively.

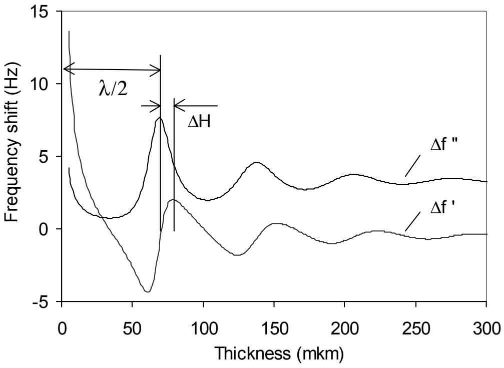


Figure 1. Theoretical dependence of $\Delta f'$ and $\Delta f''$ versus $H$

Thus, the oscillations of the piezoelectric plate generate a damped shear wave propagating through the thickness of the liquid interlayer.

The analysis of these expressions gives three methods of determination the storage shear modulus $G'$ and mechanical loss factor $\tan\theta$.

The first method is connected with the wave propagation with the length of wave

$$\lambda = \frac{1}{f_0 \cos(\theta/2)}\sqrt{\frac{G'}{\rho\cos\theta}}\ . \qquad (2)$$

The maximum attenuation in piezoquartz, hence, maximum of imaginary frequency shift, occurs at antiphase of waves, and the smaller the mechanical loss angle the higher and sharper maximum. It is possible to obtain the maximum attenuation values in the following form:

$$H = \frac{\lambda}{2} n\ .$$

Hence, the first maximum of attenuation should be observed at interlayer thickness equal to half of length of the shear wave.

The second method is based on the determination of the limit shift of frequencies $\Delta f_\infty'$ and $\Delta f_\infty''$ at $H>>\lambda$:

$$\Delta f_\infty'' = \frac{SG'}{2\pi M f_0 \lambda\cos\theta}\ ,\quad \Delta f_\infty' = -\Delta f_\infty''\cdot\tan(\theta/2)\ .$$

where $S$ is the area of all horizontal surface of piezoquartz, $\rho$ is the liquid density.

The third method is realized at $H<<\lambda$, and equations for real $\Delta f'$ and imaginary $\Delta f''$ shifts of the resonance frequency become simplified:

$$\Delta f' = \frac{SG'}{4\pi^2 M f_0 H}\ ,\quad \Delta f'' = \frac{SG'}{4\pi^2 M f_0 H}\tan\theta = \Delta f'\tan\theta\ . \qquad (3)$$

From these expressions follows, that at presence of a liquid with a bulk shear modulus the $\Delta f'$ and $\Delta f''$ should be proportional to the inverse value of the thickness of the liquid interlayer. The imaginary frequency shift is equal to change of attenuation of vibrating system $\Delta f'' = \Delta\alpha/2$, where $\Delta\alpha$ is the changing of a width of resonance curve of piezoquartz.

All these methods give satisfactory agreement of results. Comparison the values of shear moduli obtained by the three methods [5,6] is shown that the third method is more precise in determination of shear modulus.

**Experimental results**

Investigation of shear elastic modulus and the tangent of the mechanical loss angle of different liquids was carried out using the third method when the thickness of the liquid interlayer being much less than the shear wavelengths ($H<<\lambda$). The experiments have shown the linearity of $\Delta f'$ $(1/H)$ and $\Delta f''(1/H)$ dependencies. The piezoelectric crystal was of $X$-18.5° cut with the mass $M$ = 6.28 g. Poison's ratio at its working surface is equal to zero. The resonance frequency of the quartz is 74 kHz. The strap area is $S$ = 0.2 cm$^2$. The experiment temperature $t$ °C, values of the real shear modulus $G'$, tangents of the mechanical loss angle $\tan\theta$ and liquid viscosities $\eta$ at 74 kHz are presented in Table 1. $G'$ and $\tan\theta$ were calculated by formula (3).

Table 1 : Shear properties of tested liquids at 74 kHz

| | $t$, °C | $G'\cdot10^{-5}$ Pa | $\tan\theta$ | $\eta$, P |
|---|---|---|---|---|
| Ethylene glycol | 23 | 0.91 | 0.24 | 0.19 |
| Diethylene glycol | 24 | 1.22 | 0.31 | 0.32 |
| Triethylene glycol | 24 | 3.45 | 0.87 | 0.14 |
| Tridecane | 23 | 0.68 | 0.10 | 0.19 |
| Polyethyl-siloxane-1 | 23 | 9.83 | 0.11 | 0.25 |
| Clay solution | 22 | 1.35 | 0.20 | 0.02 |
| Diesel oil | 24 | 0.42 | 0.28 | 0.02 |

Table 1 shows that all tested liquids possess the measurable shear elasticity modulus at frequency 74 kHz. The tangent of mechanical-loss angle is smaller than unity for low-viscosity liquids. Thus the relaxation frequency of liquids is less than experimental one (74 kHz).

Also we investigate the non-linear visco-elastic properties of liquids [7]. The object of our research was tridecane. Figure 2 presents the dependence of $G'$ and $G''$ on the angle of shear deformation for tridecane. It is visible, that linear shear elasticity of liquid observed at small angles of shear deformation (less than 10 '). In this area shear stress are proportional to shear deformation and liquid have a

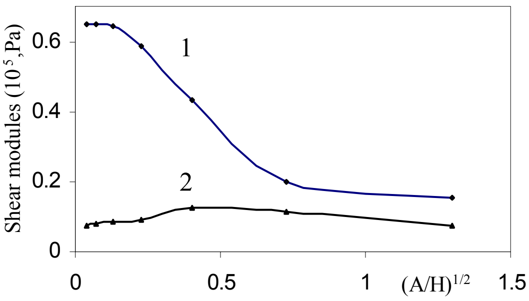


Figure 2 : Experimental dependence of $G'$ (1) and $G''$ (2) on amplitude of shear deformation for tridecane

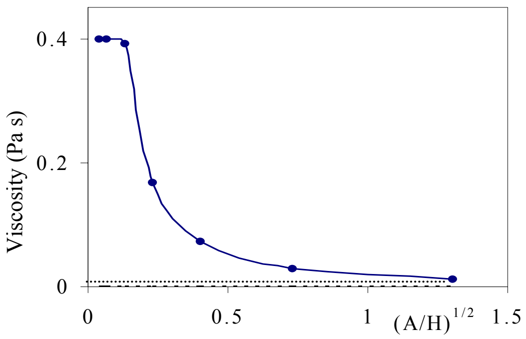


Figure 3 : Dependence of tridecane viscosity on amplitude of shear deformation

constant shear modulus. The linearity of curve is break at some critical value of deformation angle. Figure 3 presents the dependence of $\eta$ on the angle of shear deformation for tridecane. It is visible that $\eta$ decrease with deformation angle increasing and tend to tabular viscosity (dotted line). It can be supposed that liquid has some structure at small amplitude of shear deformation. This structure is breaking with increasing of deformation. Tabular viscosity corresponds to liquid with destroyed structure.

## Model

We suggest a hypothesis that dynamic structural micro-inhomogeneity is characteristic not only for high-viscous liquids and glasses, but also for simple liquids with low viscosity [8]. According to our cluster model, any liquid present micro-inhomogeneous medium, consisting of two dynamic components: ordered areas (clusters) and inhomogeneous disorder matrix. Under deformation, the clusters subject to reorganization that is expressed in diffusion exchange by fluctuation holes (exited kinetic units). The free fluctuation volume is concentrated in the disorder matrix as holes. A break-off of the kinetic unit (atom or group of atoms) from the cluster indicates the formation of a fluctuation hole, and its addition to the cluster means the hole slamming. The low-frequency viscoelastic relaxation of liquids is caused by dissociation of relatively long-living clusters, which nature is fluctuation: they form and break up with time. The lifetime is great because of the large number of the connected molecules, included in cluster. Thus, the preliminary result of estimation is in the following – the low-frequency relaxation process in liquids concerns to low energy processes and explained by interactions of clusters.

## Conclusions

It was shown that the visco-elastic properties of liquids are non-linear, but liquid possesses the constant shear modulus at small angles of shear deformation.

Liquids have cluster structure at small angles of deformation, which break with deformation increasing.

The low-frequency relaxation process in liquids concerns to low energy processes and explained by interactions of large molecule groups (clusters).

## References

[1]. H.A. Barnes, "A review of the slip of polymer solution, emulsion, and particle suspensions in viscometers: its cause, character and cure," J.Non-Newtonian Fluid Mech., vol.56, pp. 221-251, 1995.

[2]. S. Granick, "Soft matter in a tight spot," Physics Today, July 1999, pp. 26-31.

[3]. U.B. Bazaron, Low frequency shear elasticity of liquids, Sib.Div. of RAS, Ulan-Ude, 2000.

[4]. Ya.I. Frenkel', *Kineticheskaya teoriya zhidkostei*, Leningrad: Nauka, 1975.

[5]. B.B. Badmaev, O.R. Budaev, and T.S. Dembelova, "Propagation of shear waves in polymer liquids," Acoustical Physics, vol.45, pp. 541-544, 1999.

[6]. B.B. Badmaev, T.S. Dembelova, M.N. Ivanova, Ye.R. Ochirova, "Impedance method of measurement of liquid shear elasticity," in Proceedings of the X session of Russian Acoustical Society, Moscow, Russia, November 2000, pp. 44-47.

[7]. B.B. Damdinov, B.B. Badmaev, M.N. Ivanova, "Experimental acoustical research of nonlinear shear properties of visco-elastic materials" in Proceedings of 16th ISNA, Moscow, Russia, August 2002, vol.2, pp. 831-834.

[8]. B.B. Damdinov, D.S. Sanditov, B.B. Badmaev, "Cluster model of low-frequency liquid viscoelastic relaxation", in Proceedings of 17th ICA, Rome, Italy, September 2001 (available on CD-ROM).