

## LOW-FREQUENCY SHEAR PARAMETERS VIS-COELASTIC RELAXATION PROCESS

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### ABSTRACT

An experimental study of the shear parameters of viscoelastic liquids is carried out by the acoustic resonance method based on the changes in the natural frequency and  $Q$  factor of a piezoelectric quartz resonator. The liquid to be studied is placed between a stationary quartz strap and the piezoelectric quartz crystal vibrating at the resonance frequency. For a set of drilling mud, the values of the real and imaginary shear module are obtained at a frequency of 74 kHz. The measurements are performed with a liquid layer thickness much smaller than the shear wavelength. It is shown that the shear modulus decreases with increasing strain amplitude. A hole-cluster model based on the Isakovitch-Chaban nonlocal diffusion theory is proposed for explaining the low-frequency viscoelastic relaxation process.

### INTRODUCTION

Many liquids are known to change their viscosity and exhibit viscoelastic properties under an external load. These are the so-called non-Newtonian behavior attract the attention of researchers [1-3]. For studying the mechanical properties of viscoelastic liquids, different acoustic methods were developed. A review of acoustic methods used for measuring viscoelastic properties of materials can be found in [4]. Earlier, we performed experimental studies of low-frequency shear properties of viscoelastic liquids at a constant strain by the acoustic resonance method with the use of a piezoelectric quartz resonator [5].

In the present experiment, we use a method of measuring the complex shear module of liquids to study the shear properties of drilling mud depending on the vibration amplitude of the resonator.

### METHOD

An acoustic resonance method of measuring the complex shear modules of liquid was

described in [6]. This method is basically identical to that proposed by L.I.Mandel'shtam [7] and applied by S.E.Kaikin [8] to study the nature of interaction of forces between two contacting solid bodies. In /1/ a piezoelectric quartz crystal shaped as a rectangular bar ( Fig 1), Length vibration are<sup>3</sup> excited at the fundamental resonance frequency. On the crystal, /3/ a second solid body (a strap) is placed, and the character of the interaction of these two bodies is studied by the changes in the parameters of vibration. Using this method, it was shown that friction forces are of elastic nature when the vibration amplitudes are small. In the resonance method of measuring viscoelastic properties of liquids, /2/ a thin layer of the strap. The working surface of the piezoelectric quartz crystal is its horizontal face perpendicular to the optical axis. When the quartz crystal vibrates, the strap is practically at rest, because the coupling through the liquid layer is weak and cannot transmit the acceleration of the quartz crystal to the strap. The piezoelectric quartz crystal is fixed by steel needles at points lying on the nodal line. The voltage from the output of the of an oscillator is supplied to the first pair of electrodes positioned at one end of the quartz crystal on its two lateral faces. Under the effect of the alternating electric field the quartz crystal performs forced vibrations of compression-extension type. The alternating arising due to the piezoelectric effect is applied to the second pair of electrodes symmetrically positioned at the other end of the lateral face. Vibration of the quartz crystal is determined with an accuracy of 1 Hz. By smoothly varying the modulation frequency it is possible to make one of the side frequencies equal to the natural frequency of the quartz crystal. In this case, the vibration amplitude of the latter increases reaching its maximum. Correspondingly, the voltage supplied to a mill voltmeter also increases. A voltmeter is used to measure the voltage across the quartz crystal, which usually does not exceed several hundreds of mill volts. The resonance frequency shift of the piezoelectric quartz crystal is measured for different thickness of the liquid film. The increase observed in the resonance frequency of the vibratory system proves that the liquid layer exhibits not only viscous but also elastic properties.

If only dissipative forces are present, e.g., internal friction, the resonance frequency can only decrease. The experiments were performed with an 18.5 X-cut quartz crystal whose Poisson ratio was zero. The dimensions of the crystal were 36x12x5 mm , and its resonance frequency was 74 kHz. From the changes in the acoustic parameters of the piezoelectric quartz crystal (the resonance frequency and the width of the resonance curve), the real  $G'$  and imaginary  $G''$  shear module of the viscoelastic liquid were determined. The measuring procedure was as follows. The liquids under investigation and the surfaces of both quartz and strap were carefully cleaned- Immediately after cleaning, the piezoelectric quartz crystal was mounted on the sample holder and the liquid under study was placed on the quartz surface and covered with the strap. The film thickness was determined and monitored by the interference method [6]. After this, the resonance frequency of the vibratory system and the width of the resonance curve were measured. The theory of the resonance method [9] yields the following formulas for the real and imaginary shear module of the liquid layer:

$$G' = \frac{4\pi^2 M f_0' \Delta f' H}{S}$$

$$G'' = \frac{4\pi^2 M f_0'' \Delta f'' H}{S}$$

where  $M$  is the mass of the piezoelectric quartz crystal,  $Af$  is the resonance frequency shift of the piezoelectric quartz crystal,  $\Delta f$  is the change in the half-width of the resonance curve of the quartz crystal,  $f_0$  is its natural resonance frequency,  $H$  is the thickness of the liquid layer, and  $S$  is the area of the strap. Formulas (1) and (2) are valid under the following condition

$$m/M \ll 1, H/\lambda \ll 1, f_1/f_0 \ll 1$$

where  $m$  is the mass of the strap;  $[\lambda]$  is the wavelength of the shear wave propagating in the liquid layer of thickness  $H$  under the effect of the quartz crystal vibrations; and  $f_1$  is the natural frequency of the strap, which is determined by the elastic coupling with the quartz surface through the liquid layer. In the experiment, to satisfy conditions (3), we used  $M=0.74$  g and  $m=0.4$  g. The shear wavelength was of an order of 100  $\mu\text{m}$  at the given frequency, and the values of the film thickness  $H$  were usually no greater than several microns [10]. The behavior of a viscoelastic liquid at low frequencies of external actions can be described by the simple Maxwell rheological model [11]. The viscosity  $\eta$  is calculated by the formula

$$\eta' = \frac{G'^2 + G''^2}{2\pi f_0 G''}$$

From Eqs. (1), (2), and (4), one can calculate the main shear parameters of liquids.

## EXPERIMENTAL RESULTS AND DISCUSSION

We studied the shear visco elastic properties of a set of drilling mud with different dynamic viscosities. The drilling mud were chosen as liquids that exhibit a non-Newtonian behavior under a shear external action. A drilling mud is a water solution of silicates (clays). Drilling mud with different dynamic viscosities were given different numbers: 1,2,3, etc. For all liquids studied, linear dependences of the frequency shifts on the inverse thickness of the liquid layer were obtained at a constant vibration amplitude of the piezo-

electric quartz crystal. Figure 2 shows the dependences of the real resonance frequency shift of the quartz crystal on the thickness of the liquid layer for drilling mud 2 and 3. One can see that the dependences are linear in the whole thickness range under study. From formulas (1), (2) and (4), we determine the real and imaginary parts of the complex shear modulus and the viscosity of the liquids under investigation. The results of measurements are presented in the table. The last column shows the values of dynamic viscosity  $\eta_d$ , which are given for comparison.

Earlier, in [6] it was shown that the shear elasticity of liquids is of nonlinear character, i.e., depends on the amplitude of external action. We performed a study of the drilling mud under increasing angle of shear. Figure 3 presents the experimental dependences of the real resonance frequency shift on the vibration amplitude of the piezoelectric quartz crystal for several drilling mud. The abscissa axis represents the ratio  $A/H$  as a measure of the angle of shear, where  $a$  is the vibration amplitude of the quartz crystal in microns. The amplitude was determined by the method described in [12]. One can see that, up to a certain value of the angle of shear, the real frequency shift (and, hence, the shear modulus) has a constant value; as the angle of shear increases further, the real frequency shift decreases. Based on this observation, we can assume that, in equilibrium, the liquid possesses some equilibrium structure, which is retained in the presence of small strains; as the shear strain increases, the structure is destroyed, which leads to a decrease in the real shear modulus.

Thus, the study of drilling mud showed that they possess a complex shear modulus. This means that, in these liquids, a low frequency visco elastic process due to the interaction of large groups of molecules, i.e., clusters, takes place.

According to the Isokovich-Caban model [13], a highly viscous liquid can be considered as a microinhomogeneous medium consisting of two dynamic components; ordered micro regions, i.e., clusters are rearranged. micro regions, i.e., clusters are rearranged. We assume that such a dynamic components; ordered. micro regions, i.e., clusters are rearranged. We assume that such a dynamic structures microinhomogeneity is a characteristic structural feature of all liquids. No fundamental qualitative difference exists between highly viscous liquids. Only a quantitative difference can be found between them, namely; the cluster lifetime in ordinary liquids is much smaller than that in highly viscous liquids. In terms of this model, the low-frequency visco elastic relaxation of liquids is caused by the decay and recovery of such fluctuation clusters, i.e., dynamic microinhomogeneities of the structure, which collapse under an external dynamic action.

TABLE 1

Table Shear viscoelastic properties of liquids  
Shear viscoelastic properties of liquids

Liquids	$G' \cdot 10^{-5}, \text{Pa}$	$G'' \cdot 10^{-5}, \text{Pa}$	$N, \text{Pas}$	$n_d, \text{Pas}$
Drilling muds 1	1,353	0,271	1,528	0,0192
Drilling muds 2	0,334	0,072	0,353	0,0128
Drilling muds 3	0,201	0,063	0,153	0,0085
Drilling muds 4	0,442	0,099	0,451	0,0156

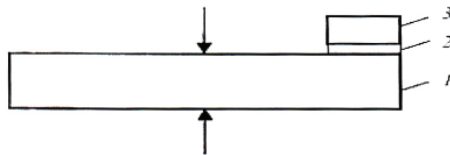


Fig 1. Piezoelectric quartz crystal (1) with a liquid layer (2) and a quartz strap (3)

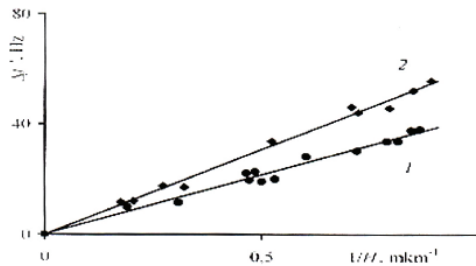
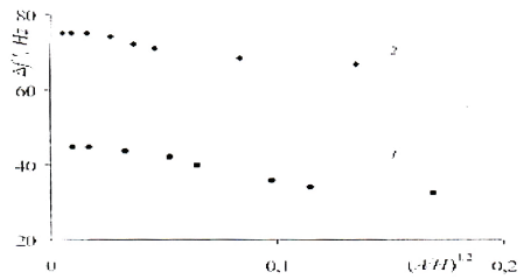


Fig 2. Dependences of the real frequency shift of the resonator on the inverse thickness of liquid layer for drilling muds (1) 3 and (2) 2.  
Key: 1, Hz; 2,  $\text{mkm}^{-1}$



**CONCLUSIONS**

(i) By the acoustic resonance method with the use of a piezoelectric quartz resonator, we obtained the values of the real and imaginary shear module and mechanical loss tangent of drilling mud at a constant vibration amplitude of the quartz crystal. We showed that all liquids studied possess measurable values of the complex shear modulus.

(ii) The study of shear parameters under an increasing vibration amplitude of the quartz crystal showed that shear modulus of the objects studied decreases with increasing angle of shear.

(iii) We proposed a hole-cluster model of liquids on the basis of the Isakovich-Chaban nonlocal diffusion theory. In the framework of the model, we estimated the activation energy of the viscoelastic relaxation process.

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