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DAMPING OF THE MECHANICAL VIBRATIONS OF A PIEZOELECTRIC ELEMENT BY SENSITIVE COATINGS

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A method of calculating the viscoelastic properties of moisture-sensitive polymer films deposited on the piezoelectric element of a quartz resonator is proposed.

In quartz atmospheric humidity sensors that employ the energy measuring principle the role of moisture-sensitive element is played by thin polymer films deposited on the piezoelectric element of the quartz resonator [1, 2]. The change in the damping properties of the polymer film as a result of the sorption of water vapor from the air leads to a change in the dissipation of the mechanical energy of the quartz resonator and its equivalent electrical resistance. Sensors of this kind have been called dissipative quartz mechanical energy transducers (DQMET).

The sensitivity of DQMET to a nonelectric quantity such as atmospheric humidity depends on the damping capacity of the sensitive coatings deposited on the piezoelectric element of the quartz resonator and its dependence on the parameter being monitored. The output parameter of the DQMET is its equivalent resistance R [3]:

$$R = K_0 \eta_{\mathbf{f}} \Delta_{\mathbf{f}},\tag{1}$$

where  $K_0$  is the conversion coefficient of the piezoelectric element, ohm sec/kg, and  $\eta_f$  and  $\Delta_f$  are the viscosity, Pa sec, and thickness, m, of the sensitive film.

The viscosity of the film (internal friction) varies with the parameter monitored, for example the humidity of the air, affecting the dissipation of the elastic vibration energy of the piezoelectric element and causing a corresponding change in the equivalent resistance of the quartz resonator. If the viscosity of the sensitive polymer is too great, there will be a decrease in its damping capacity expressed in a decrease in the equivalent resistance of the DQMET with increase in viscosity, which limits the range of application of expression (1). In our opinion, this effect is attributable to relaxation processes in the polymer film.

For DQMET intended to serve as atmospheric humidity sensors it is proposed to use a polyamide (Kapron, nylon-6) coating [2]. We will consider the variation of the damping properties of a polyamide filament with relative humidity when vibrations are excited in the filament at a frequency of 100 Hz. Figure 1 shows the loss modulus E" =  $\omega\eta$  and the modulus of elasticity E' of the filament as functions of the relative humidity  $\phi$  [4]. The loss modulus has a maximum at 60% relative humidity. With further increase in humidity the loss modulus decreases. We will explain this effect from the standpoint of relaxation theory.

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Fig. 1. Modulus of elasticity E', GPa, and loss modulus E'' =  $\omega \tau$ , GPa, of a polyamide filament as functions of the relative humidity  $\phi$ , %, at a frequency of 100 Hz and a temperature of 35°C.

Fig. 2. Relative increment of equivalent resistance  $\Delta R/R$ , %, of a quartz resonator with a polyamide film as a function of the relative humidity  $\phi$ , %, at a temperature of 36°C for the 1st, 3rd, and 5th harmonics: 1) 40; 2) 120; 3) 200 kHz.

The dry filament has a high relaxation frequency  $f_r$  exceeding the vibration frequency of the filament  $f_F$ , i.e.,  $f_r > f_F$ . When the filament sorbs water vapor, the mass of the side chains of the macromolecules responsible for the  $\beta$ -relaxation process increases and their motion is slowed. This leads to a fall in the relaxation frequency of the filament, which approaches its vibration frequency. When these frequencies become equal, a maximum of the loss modulus is observed. A continued increase in the moisture content of the filament lowers the relaxation frequency of the polymer still further and it fall below the vibration frequency of the filament, which is characterized by a decrease in the loss modulus and, moreover, the loss tangent:

$$\lg \delta = \frac{E''}{E'} \,. \tag{2}$$

The conditions for maximum dissipation of the vibration energy are given by the relations

$$tg \delta = \omega \tau, \quad f_r = Q_f f_F. \tag{3}$$

The increase in the viscosity of the filament continues beyond the relaxation peak (E" maximum). This is confirmed by experiments on rosin [5], whose viscosity varies with increase in temperature without an extremum, whereas its damping capacity has a maximum corresponding to the maximum dissipation of the vibration energy of a quartz resonator with rosin on the surface of the piezoelectric element. In the polymer film on the surface of the piezoelectric element. In the polymer film on the surface of the piezoelectric element of a DQMET relaxation processes similar to those in the polyamide filament take place. This is confirmed by the dependence of the relative increase in the equivalent resistance of a quartz resonator with a polyamide film on the surface of the piezoelectric element on the relative humidity of the air at three frequencies obtained by exciting the quartz resonator at the harmonics. As may be seen from Fig. 2, as the vibration frequency increases, the energy dissipation maximum is displaced towards higher humidities, which confirms the relaxational nature of the processes in the polyamide film. An increase in vibration frequency reduces the viscosity of the film in accordance with the law [6]

$$\eta = t(\omega)^{-n}.$$

The more precise determination of the frequency dependence of the viscosity of the polyamide film relative to the viscosity of the polyamide filament leads to the expression



Fig. 3. Dependence of the viscosity  $\eta_f$ , Pa·sec, modulus of elasticity  $E_f$ , GPa. figure of merit  $Q_f$ , and mechanical resistance  $N_f$ , mN·sec/m, of polyamide film on the relative humidity  $\phi$ ,  $\mathcal{Z}$ , at various frequences.

$$\eta_{f} = \eta_{F} \left( \frac{f_{q}}{f_{F}} \right)^{-\alpha}, \ \alpha = 1,18.$$
(5)

The viscosity of the polyamide filament as a function of the relative humidity can be determined by means of the interpolation equation obtained using the dependence given in Fig. 1:

$$\eta_{\mathbf{F}} = \frac{1}{2\pi} \left( 231\phi^2 + 2, 12 \cdot 10^4\phi + 6, 8 \cdot 10^5 \right),\tag{6}$$

where  $\phi$  is the relative humidity of the air, %.

We will consider the laws of variation of the modulus of elasticity of the polyamide film. According to our investigations [7], in a polyamide film deposited on the surface of the piezoelectric element of a quartz resonator it is possible to observe "antiplasticization" characterized by an increase in the modulus of elasticity with increase in the moisture content of the polymer. Taking into account the theory of antiplasticization [8], we can express the modulus of elasticity of the film in the form:

$$\frac{1}{E} = \frac{\xi}{E_1} + \frac{1 - \xi}{E_2},$$
(7)

where

$$\xi = \frac{u}{u_{\text{max}}} \,. \tag{8}$$

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Fig. 4. Calculated dependence of the equivalent resistance of the DQMET R, ohm, on the relative humidity  $\phi$ , %, for various polyamide film thickness: 1)  $\Delta_{\rm F}$  = 1, 2) 3, 3) 5 µm.

The moisture content of the polymer at various relative humidities can be found from the interpolation formula obtained from [9]:

$$u = 6,0625 \cdot 10^{-4} \varphi^2 + 0,0185 \varphi + 0,863.$$
(9)

The frequency dependence of the modulus of elasticity of the polyamide film is determined as

$$E_{f_1} = E_{f_2} \left( \frac{f_1}{f_2} \right)^{\alpha}, \ \alpha = 0,06625.$$
(10)

The viscoelastic properties of the polyamide film calculated from expressions (5)-(10) enable us to estimate the range of application of expression (1) from the condition

$$2\pi f_{\alpha} \eta < E. \tag{11}$$

The damping properties of the coatings can be estimated from the mechanical resistance of the polymer  $N_f$  or its figure of merit  $Q_f$ :

$$N_{f} = \frac{\pi^{2} \eta_{f} m_{f}}{2 \left( l_{0} + 2\Delta_{f} \right)^{2} \rho_{f}} ; \qquad (12)$$

$$Q_{\mathbf{f}} = \frac{\omega m_{\mathbf{f}}}{2N_{\mathbf{f}}} \,. \tag{13}$$

In Fig. 3 we have plotted the dependence, calculated from the above expressions, of the mechanical parameters of the polyamide coating of the piezoelectric element on the relative humidity of the air for several frequencies. For DQMET acting as humidity sensors it is advisable to employ a 300 kHz quartz resonator with a DT cut piezo-element having a width-to-length ratio of 0.39. The expression for calculating the output parameter of the DQMET - its equivalent resistance R, ohm - is as follows:

$$R = \Delta_{f} (1,63 \cdot 10^{4} \varphi^{2} + 1,50 \cdot 10^{6} \varphi + 4,8 \cdot 10^{7}).$$
(14)

In Fig. 4 we have plotted the conversion characteristics of the quartz humidity sensor obtained using expression (14). A comparison with experiment gives satisfactory results with a discrepancy of the order of  $\pm 10\%$ , which is good enough for practical purposes.

The absence from the literature of data on the viscoelastic properties of moisturesensitive polymer films made it necessary to carry out special investigations, as a result of which we established the frequency dependence of the viscosity, modulus of elasticity, figure of merit, and mechanical resistance of a thin polymer film used as the moisturesensitive coating of a DQMET. The physical parameters of the moisture-sensitive coating thus obtained made it possible to calculate the moisture sensitivity of a quartz humidity sensor operating on the energy measuring principle and widely used for monitoring and regulating the humidity in industrial environments.

## NOTATION

R is the equivalent resistance of the DQMET;  $K_0$  is the conversion coefficient of the piezoelectric element;  $\eta_f$  and  $\eta_F$  are the viscosities of the polymer film and filament respectively;  $\Delta_f$  is the thickness of the film; tan  $\delta$  is the loss tangent of the polymer; E" and E' are the loss modulus and the modulus of elasticity of the polyamide filament;  $E_f$  is the modulus of elasticity of the polyamide film;  $E_1$  and  $E_2$  are the moduli of elasticity of the polymer; u and with the maximum moisture content;  $\xi$  is the relative moisture content of the polymer;  $f_r$  is the relaxation frequency of the polymer;  $f_F$  and  $f_q$  are the vibration frequencies of the polyamide filament and the DQMET with a polyamide film;  $\beta$  is the type of relaxation process;  $\omega$  is the angular frequency;  $\tau$  is the relaxation time of the polymer;  $Q_f$  and  $N_f$  are the figure of merit and the mechanical resistance of the polymer film; n and  $\alpha$  are constant coefficients;  $E_{f_1}$  and  $E_{f_2}$  are the moduli of elasticity of the polyamide film is the polyamide film; n and  $\alpha$  are constant coefficients;  $E_{f_1}$  and  $E_{f_2}$  are the moduli of elasticity of the polyamide film is the polyamide film in  $\beta$  is the polyamide film at the frequencies  $f_1$  and  $f_2$  respectively;  $m_f$  and  $\rho_f$  are the mass and density of the polymer film; and  $\ell_0$  is the width of the piezoelectric element of the DQMET.

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