

The Influence of Strain Amplitude, Temperature and Frequency on Complex Shear Moduli of Polymer Materials under Kinematic Harmonic Loading

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Modeling of the cyclic deformation of viscoelastic materials as the key aspects of analysis of the structural behavior is performed. The approach that uses the complex-value amplitude relations is preferred rather than direct numerical integration of the complete set of constitutive equation for the material. Time dependent transient inelastic response of polymer materials to monoharmonic kinematic loading is simulated by the Goldberg constitutive model. To predict the steady-state response in terms of amplitudes, the relations between the amplitudes of main field variables are established with making use of complex moduli concept. It is performed by making use of equivalent linearization technique. It is shown that this technique leads to overestimation of stress amplitude. To avoid this, the modified equivalent linearization technique is applied. Characterization of the complex moduli dependence on frequency and temperature as well as amplitude of strain intensity is performed. Results demonstrate a weak dependence of loss moduli on the frequency of the loading within the wide interval of it, while variation of storage moduli with increasing temperature is more pronounced.

Keywords: harmonic loading, amplitude relations, inelastic deformation, complex moduli.

1. Introduction

Harmonic loading is one of the most important and widely used types of loading imposed on structural elements. Under cyclic loading a viscoelastic or elasto–plastic material (beyond the elastic domain) yields a hysteresis loop in the stress–strain relationship. Such a loop indicates that part of the strain energy is not recovered but dissipated during the cycle. This phenomenon is usually called the ”dissipative heating” [1, 2]. These materials can exhibit specific time dependent properties and can

be deformed inelastically being exposed to high stress levels. Their viscoelastic responses become more significant under high loading levels and severe environmental conditions and are often accompanied by inelastic deformations. This self-heating effect caused by mechanical energy dissipation in polymer materials subjected to harmonic loads is considered to have a great influence on the residual life of the component. It's important to notice here that, under some conditions, the dissipative heating, being small over one separate cycle of vibration, can lead to overheating for long term processes causing the change of mechanical properties, degradation of performance and durability of the structure. Therefore taking account of this effect is important for characterization of a material response at different excitation frequencies and temperatures.

There are currently two approaches to address this issue. In the first approach, the complex set of constitutive equations governing response of numerous internal parameters is introduced. The relationship between these parameters and the strain and temperature history yields evolution equations, which account for both dynamic recovery, and also creep. For polymers, the constitutive modeling utilizes, either directly or with some modifications, viscoplastic constitutive equations which have been developed for metals. The generalized yield theories of Schapery, Perzyna, Frank and Brockman, Goldberg and others [3–6] apply to identify this relationship. Czechowski in his works [7, 8] utilized apart from the elastic–plastic range of material with isotropic hardening as well as the strain rate effect described by Perzyna model. It is generally admitted that to describe the material time dependent behavior accounting for different features and peculiarities over the cycle of vibration, a direct integration of the set of constitutive equations is necessary. Usually it appears to be time and resource costly for multi–cyclic processes.

Within the second approach, the approximate amplitude relations are used to characterize the cyclic response of the material, i.e. the relations between amplitudes of the main mechanical field parameters over the cycle [9]. Naturally, the application of this technique is justified for the class of problems where there is no need for detailed information on the material response during the cycle (life prediction of the structure, failure due to overheating as a result of internal dissipation etc.). The key point of the amplitude theories is concept of complex moduli [9]. For an inelastic (particularly viscoelastic) material, the modulus governing the relation between strain and stress amplitudes is represented by a complex quantity with real and imaginary parts referred to as storage and loss modulus respectively. The former characterizes elastic response of material and the latter one defines the dissipative ability of the material [1]. In other words, the energy is stored during the loading part of cycle and released under unloading phase, whereas the energy loss occurs during complete cycle due to dissipative properties of the material. The drawback of the approach was the overestimation of stress amplitudes as a result of making use of standard equivalent linearization technique for calculation of both storage and loss moduli. To overcome this difficulty, the modified scheme was proposed in [2, 9, 12]. But applicability of the method should be verified for each particular type of the material.

Considering the importance of examination of self–heating effect under cyclic loading in polymeric materials, researches done on time dependent behavior of polymeric materials are mainly aimed to study the viscoelastic behavior in differ-

ent frequency application over wide ranges of loading amplitudes. These researches show that, the temperature will change with respect to the frequency spectrum of cyclically loading due to the stress relaxation processes in the material, thus it is necessary to determine the dependence of the modal characteristics in a frequency domain on mechanical properties at special steady state of temperature.

This paper is devoted to investigation of the technique applicability to the typical viscoelastic materials such as PR-520, and to determination the frequency and temperature effects on complex moduli for wide range of loading amplitudes. Particular attention will be paid to simulation of cyclic response of pure polymer material (PR-520) to monoharmonic kinematic loading in the frame of the second approach.

2. Time dependent constitutive relations

To accurately predict an overall performance and lifetime of polymer, it is necessary to simulate the time dependent and inelastic responses. Viscoelastic materials such as polymer materials demonstrate viscous, elastic and, under some conditions, plastic behavior. Constitutive material models of viscoelastic solids have been proposed for isotropic materials undergoing small deformation gradients whereas the inelastic strain can be calculated as the difference of the total strain and elastic strain.

Goldberg et al. [5, 10] proposed a model for predicting the viscoplastic response of neat polymers, utilizing a set of state variables as an indication of the resistance of polymeric chains against flow. It should also be mentioned that polymer's mechanical properties and loading/strain rate are the two main parameters that govern the nonlinear response of the polymer. The formulation employed in this model is based on that model which was further developed by Pan and co-workers [11].

According to this model, the inelastic strain components can be expressed in terms of the deviatoric stress components as follows:

$$\dot{\varepsilon}_{ij}^{in} = 2D_0 \exp\left(-\frac{1}{2}\left(\frac{Z}{\sigma_e}\right)\right)^{2n} \left(\frac{s_{ij}}{2\sqrt{J_2}} + \alpha\delta_{ij}\right) \quad (1)$$

where, $\dot{\varepsilon}_{ij}^{in}$ is the inelastic strain rate tensor which can be defined as a function of deviatoric stress s_{ij} and the state variables Z and α . The state variable α controls the level of the hydrostatic stress effects. In equation (1), J_2 is the second invariant of the deviatoric stress tensor that can be expressed as a function of σ_{ij} . Moreover, D_0 and n are material constants; D_0 represents the maximum inelastic strain rate and n controls the rate dependency of the material. The equivalent (effective) stress, also be defined as a function of the mean stress, such that the summation of the normal stress components σ_{kk} is three times of the mean stress, as follows:

$$\sigma_e = \sqrt{3J_2} + \sqrt{3}\alpha\sigma_{kk} \quad (2)$$

the evolution of the internal stress state variable Z and the hydrostatic stress state variable α are defined by the equations:

$$\dot{\alpha} = q(\alpha_1 - \alpha)\dot{\varepsilon}_e^{in} \quad (3)$$

$$\dot{Z} = q(Z_1 - Z)\dot{\varepsilon}_e^{in} \quad (4)$$

where q is a material constant representing the "hardening" rate, and Z_1 and α_1 are material constants representing the maximum values of Z and α , respectively. The initial values of Z and α are defined by the material constants Z_0 and α_0 . The term $\dot{\epsilon}_e^{in}$ in equations (3) and (4) represents the effective deviatoric inelastic strain rate, which was defined as follows:

$$\dot{\epsilon}_e^{in} = \dot{\epsilon}_e^{in} = \sqrt{\frac{2}{3} \dot{\epsilon}_{ij}^{in} \dot{\epsilon}_{ij}^{in}} \quad (5)$$

$$\dot{\epsilon}_{ij}^{in} = \dot{\epsilon}_{ij}^{in} - \dot{\epsilon}_m^{in} \delta_{ij} \quad (6)$$

where $\dot{\epsilon}_m^{in}$ is the mean inelastic strain rate, which matches the effective inelastic strain rate definition given by Pan and co-workers [11]. The material constants Z_0 , Z_1 , α_1 , α_0 , n and D_0 can be determined using the shear stress-strain and tensile or compression stress-strain curves, obtained by experiments conducted under constant strain rates on neat polymers. Empirically, it has been shown that the value of D_0 , quantitatively, can be set equal to 10^6 times the maximum applied total strain rate; qualitatively, it is the restricting (controlling) value of the inelastic strain rate. The values of Z_1 and n can be identified using the shear stress-strain curves constructed under various strain rates. The plateau region of the effective stress under a uniaxial tensile loading at a particular strain rate corresponds to the saturation region of the effective stress obtained under pure shear loading.

3. Complex moduli approach

Harmonic loading is one of the most widely used and important types of loadings imposed upon a mechanical structure. In this investigation, approximate model of inelastic behavior developed in [2, 9] for the case of proportional harmonic loading has been used. In this case, the cyclic properties of the material are described in terms of complex moduli. It is important to notice that the inelastic deformation is considered to be incompressible and thermal expansion is dilatational, it may be more convenient in some applications to separate the isotropic stress-strain relations into deviatoric and dilatational components that can be shown by equations as:

$$s_{ij} = 2G(e_{ij} - \varepsilon_{ij}^{in}), \quad \sigma_{kk} = 3K_V(\varepsilon_{kk} - \varepsilon^\theta) \quad (7)$$

where G is the shear modulus, K_V is the bulk modulus, $i, j, k = 1, 2, 3$ and repeated index implies a summation over. Due to incompressibility of plastic deformation, the rate $\dot{\varepsilon}_{kk}^{in} = 0$, i.e. the plastic strain rate is deviatoric one: $\dot{\varepsilon}_{ij}^{in} = \dot{\epsilon}_{ij}^{in}$.

According to this model, if a material is subjected to harmonic deformation or loading, then its response is also close to harmonic law:

$$e_{ij}(t) = e'_{ij} \cos \omega t - e''_{ij} \sin \omega t, \quad s_{ij}(t) = s'_{ij} \cos \omega t - s''_{ij} \sin \omega t. \quad (8)$$

The complex amplitudes of the deviator of total strain, \tilde{e}_{ij} , inelastic strain, $\tilde{\epsilon}_{ij}^{in}$, and the stress deviator, \tilde{s}_{ij} , are related in the N^{th} cycle by the complex shear modulus, \tilde{G}_N , and plasticity factor, $\tilde{\lambda}_N$, as shown below:

$$\tilde{s}_{ij} = 2\tilde{G}_N \tilde{e}_{ij}, \quad \tilde{\epsilon}_{ij}^{in} = \tilde{\lambda}_N \tilde{e}_{ij}, \quad N = 1, 2, 3, \dots \quad (9)$$

where:

$$\begin{aligned}\tilde{e}_{ij} &= e'_{ij} + ie''_{ij} & \tilde{s}_{ij} &= s'_{ij} + is''_{ij} & \tilde{e}_{ij}^{in} &= e_{ij}^{in} + ie_{ij}^{in} \\ \tilde{G}_N &= G'_N + iG''_N & \tilde{\lambda}_N &= \lambda'_N + i\lambda''_N\end{aligned}$$

and N is the cycle number, $(\cdot)'$ and $(\cdot)''$ denote the real and imaginary parts of complex quantities. The shear modulus and plasticity factor are functions of the intensity of the strain-range tensor, frequency and temperature:

$$\tilde{G}_N = \tilde{G}_N(e_0, \omega, \theta), \quad \tilde{\lambda}_N = \tilde{\lambda}_N(e_0, \omega, \theta) \quad (10)$$

where the square of the intensity of strain-range tensor is calculated as:

$$e_0^2 = e'_{ij}e'_{ij} + e''_{ij}e''_{ij}$$

The imaginary parts of the complex moduli are determined from the condition of equality of the energies dissipated over a period and are calculated according to the formula:

$$G''_N = \frac{\langle D' \rangle_N}{\omega e_0^2} \quad \lambda''_N = \frac{G''_N}{G_0} \quad \langle (\cdot) \rangle_N = \frac{1}{T} \int_{T(N-1)}^{TN} (\cdot) dt \quad T = \frac{2\pi}{\omega} \quad (11)$$

where D' is the rate of dissipation of mechanical energy, G_0 is the elastic shear modulus.

The real parts are found with making use of the condition that generalized cyclic diagrams $s_{aN} = s_{aN}(e_0, \omega)$ and $e_{paN} = e_{paN}(e_0, \omega)$, which relate the ranges of the stress and plastic-strain intensities in the N th cycle, coincide in the frame of the complete and approximate approaches:

$$\begin{aligned}G'_N(e_0, \omega) &= \left[\frac{s_{aN}^2(e_0, \omega)}{4e_0^2} - G_N''^2(e_0, \omega) \right]^{1/2} \\ \lambda'_N(e_0, \omega) &= \left[\frac{e_{paN}^2(e_0, \omega)}{4e_0^2} - \lambda_N''^2(e_0, \omega) \right]^{1/2}\end{aligned} \quad (12)$$

where G' and λ' are the sought-for real part of shear modulus and plasticity factor. In spite of the fact that the single-frequency approximation based on harmonic linearization has a well agreement with precise model of nonlinear behavior, it's necessary to analyze its practical accuracy for specific classes of problems.

As mentioned in the introduction, the second approach is based on the concept of complex moduli, which are determined by standard and modified techniques of equivalent linearization. It is important to notice that, the imaginary parts of complex moduli are defined by the exact expression for rate of dissipation averaged over the period of cyclic loading while to improve the accuracy of real parts of complex moduli the modified approach is proposed as shown in equation (12). According to equation (10), the complex moduli for isothermal loading case depend on the frequency, temperature and amplitude of kinematic loading only. The purpose of this paper is to investigate the influence of these parameters on complex moduli.

4. Numerical technique and the material properties

In the present work, as it was mentioned above, due to significant nonlinearity of the stiff type, the numerical integration of Goldberg equations was adopted. To solve the implicit equation (1), one should utilize an appropriate numerical discretization technique. Three step scheme of attacking the problem of complex moduli determination was designed. At the first step, the elastic-viscoplastic response of the material to harmonic deformation was calculated by numerical technique for different amplitudes of loading strain at different frequencies and temperature. At the second step, the stabilized cyclic stress-strain and inelastic-strain-strain diagrams were obtained for the whole set of calculated data. At the final step, the complex moduli were calculated by the averaging over the period of vibration of the results of direct integration and making use of cyclic diagrams and equations (11) and (12). The system of nonlinear ordinary differential equations that describes the polymer response to harmonic loading in the case of pure shear consists of the one-dimensional equations of Goldberg model comprising equations (1), (3) and (4) and evolutionary equations [12]:

$$\varepsilon_{12}^{in} = 2D_0 \exp\left(-\frac{1}{2}\left(\frac{Z^2}{3S_{12}^2}\right)^n\right) \left(\frac{S_{12}}{2|S_{12}|}\right) \quad (13)$$

$$\dot{Z} = \frac{2}{\sqrt{3}}qD_0(Z_1 - Z) \exp\left(-\frac{1}{2}\left(\frac{Z^2}{3S_{12}^2}\right)^n\right) \left(\frac{S_{12}}{|S_{12}|}\right) \quad (14)$$

$$\dot{\alpha} = \frac{2}{\sqrt{3}}qD_0(\alpha_1 - \alpha) \exp\left(-\frac{1}{2}\left(\frac{Z^2}{3S_{12}^2}\right)^n\right) \left(\frac{S_{12}}{|S_{12}|}\right) \quad (15)$$

The law of strain deviator variation $e = e_0 \sin \omega t$, as well as Hooke law for shear stress should be added to the system.

$$s_{12} = 2G(e_{12} - \varepsilon_{12}^{in}) \quad (16)$$

The values of material constants for RP-520, which were used for calculations, have been taken from [10]. The list of the values is given in Tab. 1.

Table 1 The values of material constants for RP-520

ν	α_1	α_0	q	n	Z_1 MPa	Z_0 MPa	D_0 Sec ⁻¹	E MPa	Temp. °C
0.4	0.122	0.571	253.6	0.92	768.6	407.5	10 ⁶	3250	25
0.4	0.085	0.316	226.1	0.94	616.4	267.9	10 ⁶	2980	50
0.4	0.064	0.087	273.4	0.88	564.9	195.4	10 ⁶	2520	80

5. Numerical results and discussion

The results of transient response simulation and effects of frequency and temperature on the complex moduli in the frame of modified technique described in Section 3 are presented.

In Fig. 1, the stress–strain curve was obtained for PR-520 under monotonic loading in pure shear at different temperature before glass transition temperature. In this figure, the numerical predictions of the model are generated for strain rate $5 \cdot 10^{-5} \text{sec}^{-1}$ at 25, 50, 80°C. As can be seen, this figure demonstrates a very good correspondence with the results presented in [10]. Evolution of stress and inelastic strain for epoxy resin (PR-520) under kinematic harmonic loading in pure shear with strain amplitude $e_0 = 5.5 \cdot 10^{-2}$ are shown in Fig. 2 and Fig. 3 respectively for frequency 1 Hz at different temperatures.

Fig. 4 illustrates the actual loop under cyclic loading in the maximum dissipation condition ($e_0 = 5.5 \cdot 10^{-2}$) at the frequency 1Hz for 25, 50 and 80°C. The material demonstrates cyclically stable response over the whole interval of loading amplitudes and frequencies investigated. According to Fig. 5, stabilization of the response amplitude occurs after several initial cycles. Relatively slow stabilization is observed only in the vicinity of yield point. Fig. 6 illustrates the actual loop at the frequency 1Hz. As it was mentioned earlier, this actual loop can be approximated with making use of either standard or modified equivalent linearization scheme. In the same figure, the actual loop (line 1) is shown along with the loops calculated in the frame of standard (line 2) and modified (line 3) equivalent linearization techniques.

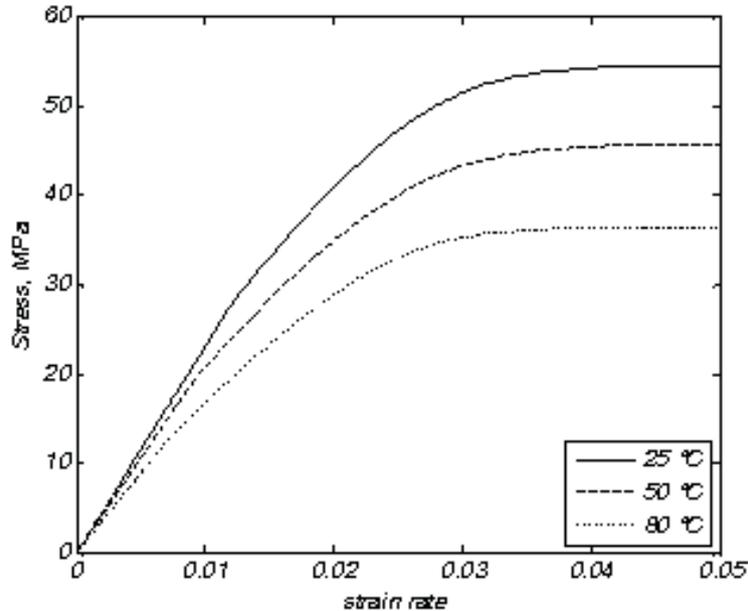


Figure 1 The stress–strain curve under monotonic loading in pure shear

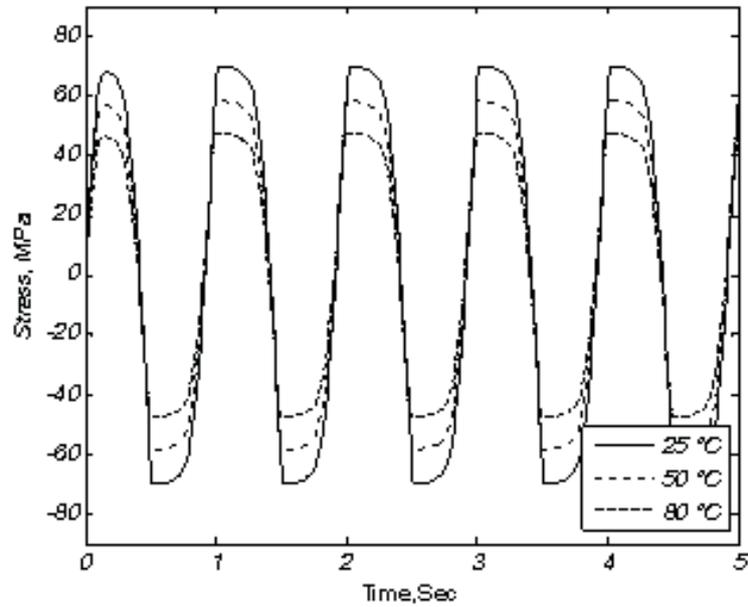


Figure 2 Stress evolution under kinematic harmonic loading

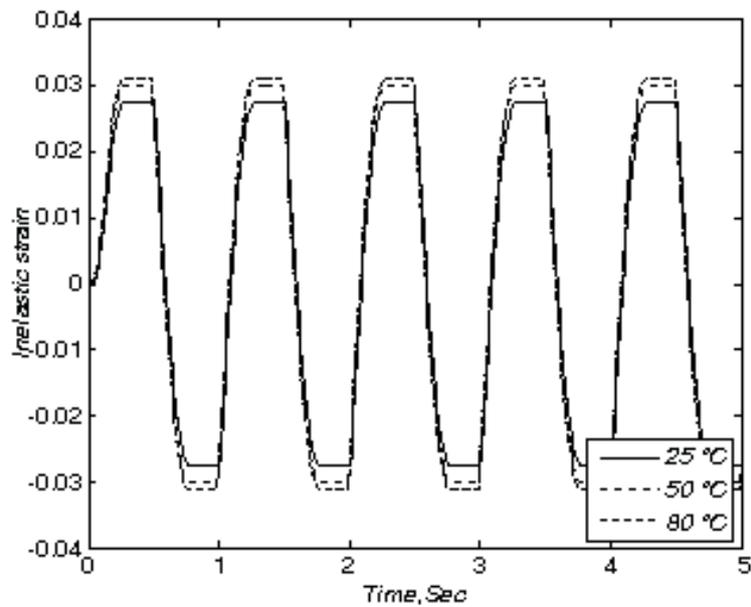


Figure 3 Inelastic strain evolution under kinematic harmonic loading

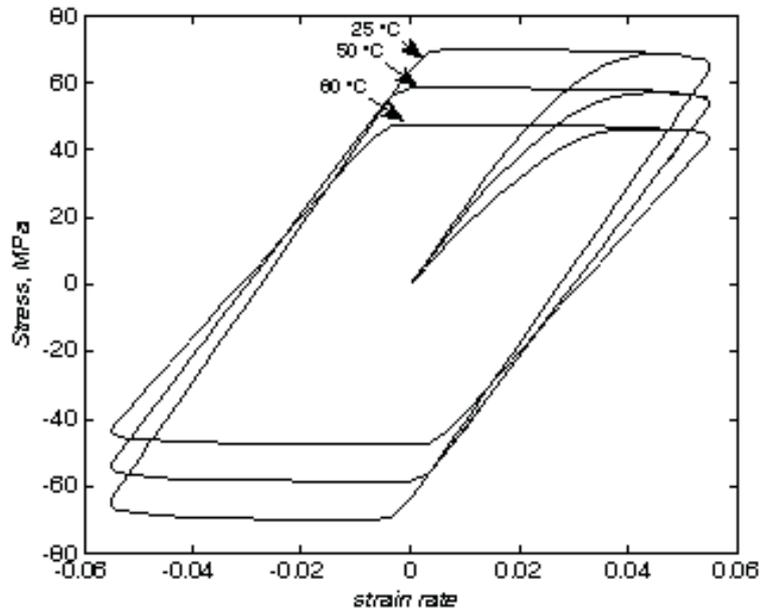


Figure 4 Actual loops under cyclic loading

The cyclic diagrams at stabilized stage of the vibration $s_a = s_a(e_0)$ (i.e. concretization of general cyclic diagram $s_{aN} = s_{aN}(e_0, \omega)$ used in the equation (12) for $N \rightarrow \infty$) are shown in Fig. 7. The curves are calculated for cyclic pure shear for different frequencies (1, 50, 100 Hz) at 25°C. The effect of frequency is easily observable. Two order of magnitude variation in frequency leads to approximate 20% change in stress amplitude at the stage of advanced deformation.

Using the cyclic diagram and making use of the equations (11) and (12), the imaginary and real parts of the complex moduli (the loss moduli G'' and λ'' , storage moduli G' and λ') in the frame of modified equivalent linearization scheme are determined. The improved values of G' and G'' have been found according to the modified scheme for different frequencies at steady-state cyclic regime and constant temperature. Dependency of storage modulus, G' , and loss modulus, G'' , on the amplitude of strain, e_0 , and frequency for the PR-520 are shown in Fig. 8 for 1, 50 and 100 Hz by solid, circle-solid and dashed lines, respectively. This figure and cyclic diagram show the inelastic behavior occurs at higher strain amplitude with increase of frequency. The trend of storage modulus behavior presented in Fig. 8 show that its values increase with increase of frequency while, the loss modulus decrease slightly as frequency increases before the peak of strain intensity.

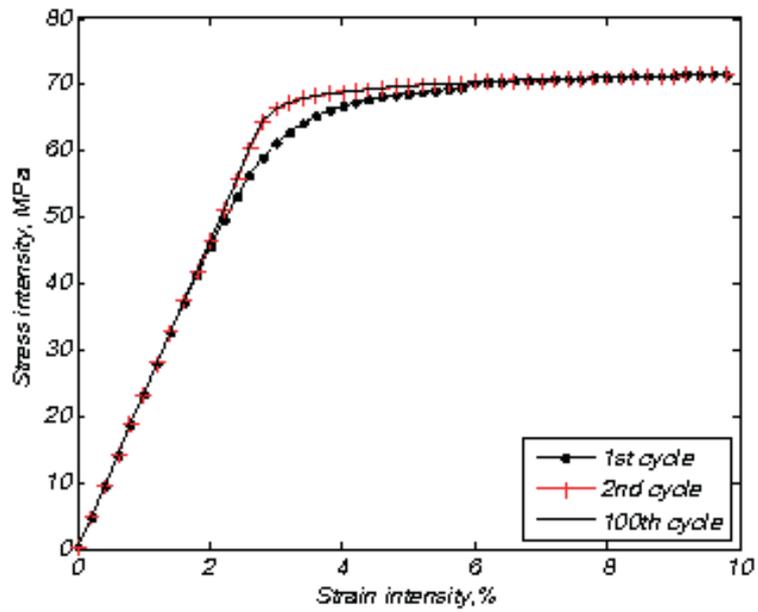


Figure 5 Stabilized cyclic diagram for PR-520 at 1 Hz

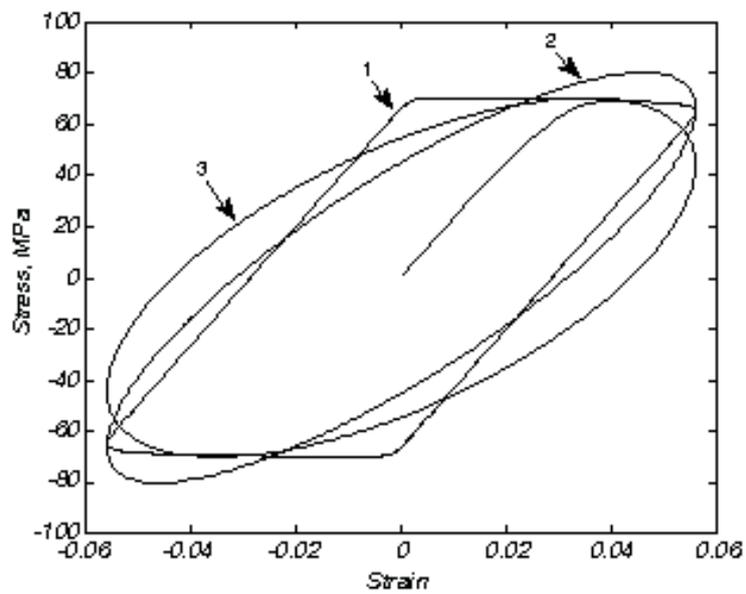


Figure 6 Hysteresis loops at the frequency 1 Hz

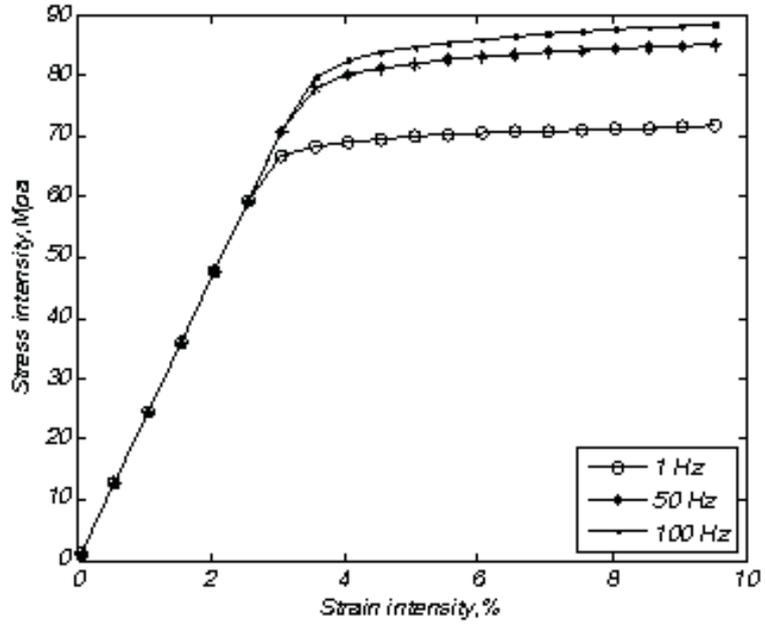


Figure 7 Cyclic diagram for PR-520 at 1, 50 and 100 Hz

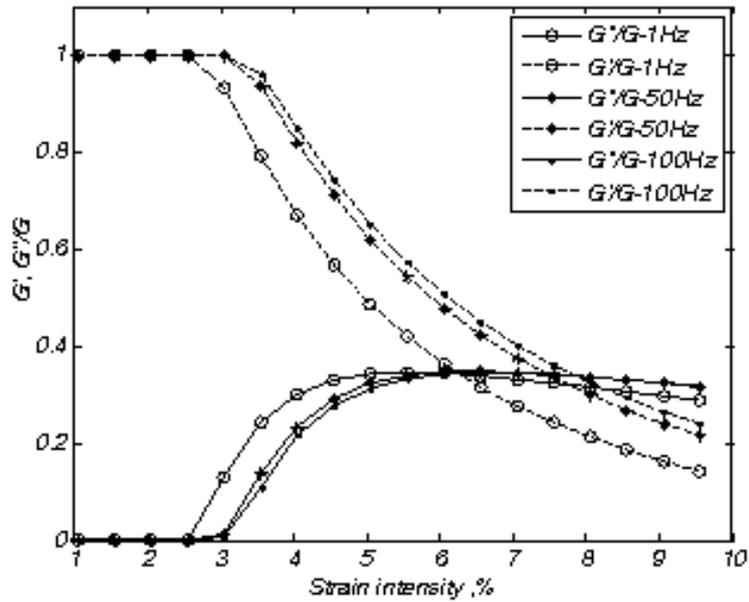


Figure 8 The real and imaginary parts of complex modulus at various frequencies

The peak values of the modulus increase insignificantly and occur later with frequency increase. Within the interval of interest between 1 and 100 Hz, the maximum in loss modulus occurs in the vicinity of 7% of strain intensity at 100 Hz. For higher values of strain intensity, the loss modulus decreases.

The cyclic diagrams at stabilized stage of the vibration at 1Hz for different temperatures are shown in Fig. 9. The effect of temperature on behavior of material is observed clearly. This figure show the inelastic behavior occurs at lower strain amplitude with increasing of temperature.

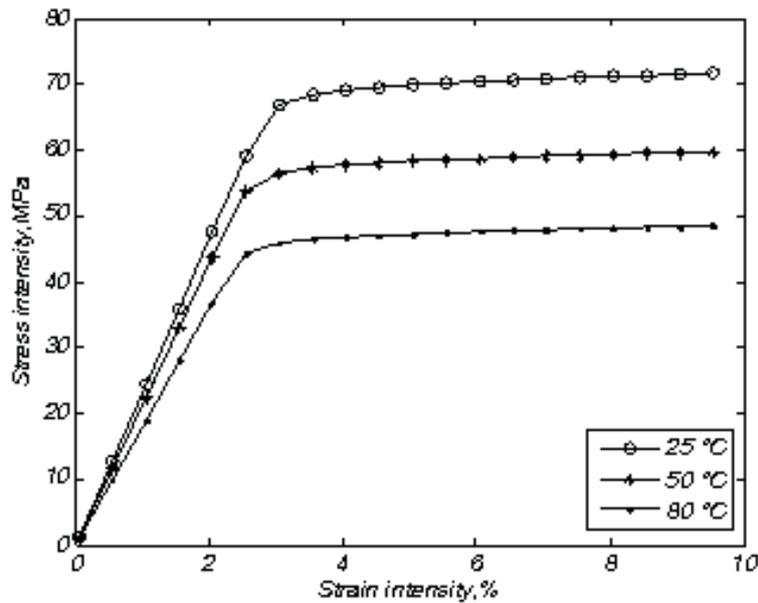


Figure 9 Cyclic diagram for 1 Hz at 25, 50 and 80°C

According to the procedure mentioned above, dependency of storage modulus, G' , and loss modulus, G'' , on the amplitude of strain, e_0 , and temperature are shown in Fig. 10 for 25, 50 and 80°C by solid, circle–solid and dashed lines, respectively. As it is seen in this figure, the inelastic behavior occurs at lower strain amplitude with increasing of temperature. The trend of storage modulus behavior presented in Fig. 10 show that its values decrease with increase of temperature while the loss modulus increase slightly before the peak of loss modulus in the vicinity of 6% of strain intensity.

The peak values of the loss modulus increase insignificantly and occur earlier with increasing temperature. Within the interval of interest between 25 and 80°C, the maximum in loss modulus occurs in the vicinity of 4.5% of strain intensity at 80°C. For higher values of strain intensity for all temperatures, the loss modulus decreases.

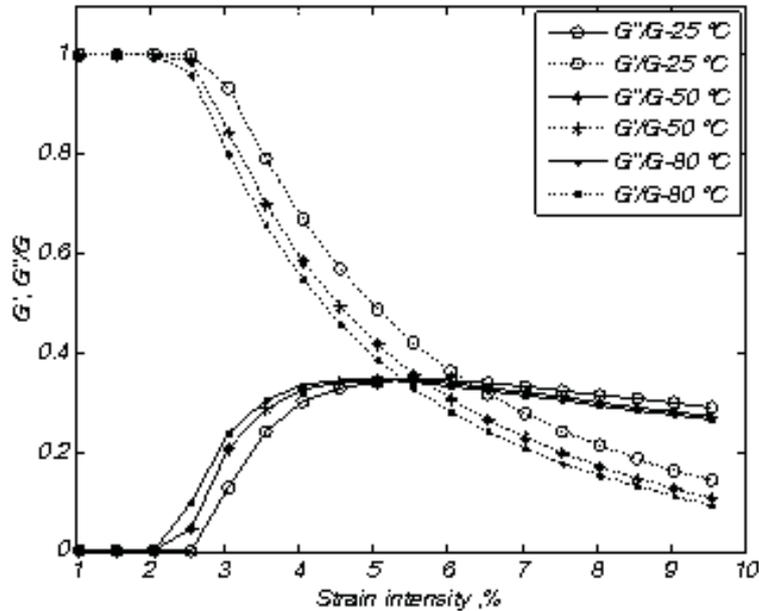


Figure 10 The real and imaginary parts of complex modulus at various temperatures

6. Conclusions

In this paper, Goldberg model was used to simulate the time dependent response of PR-520. Obtained histories of main field variables evolution were used to find the stress-strain cyclic diagram and real as well as imaginary parts of complex shear modulus with making use of both standard and modified equivalent linearization techniques over wide range of frequency and amplitude at different temperature.

Results of calculations show evidently that, the strength of material increases as the frequency increases while decreases with increasing temperature. The sensitivity of cyclic diagrams to frequency variations at the low values is more profound than at the region of higher frequency (see Fig. 7). It's important to notice that with the increase of strength of material the sensitivity to frequency is reduced. Therefore the behavior of saturation type is clearly exhibited. In general, it is possible to conclude that complex moduli demonstrate the weak dependence on the frequency within the interval investigated.

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