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M. Hashemi, Postgraduate Student, Y. Zhuk, Dr. Sci., Prof. Taras Shevchenko National University of Kyiv y.zhuk@i.ua, moh.hashemi2013@gmail.com

INFLUENCE OF FREQUENCY AND AMPLITUDE OF HARMONIC LOADING ON COMPLEX MODULI FOR POLYMER MATERIALS

This paper is devoted to the modeling and characterization of cyclic response of polymers subjected to monoharmonic kinematic loading. To predict the time dependent behavior of the polymeric materials, the Goldberg constitutive model is used. To simulate the response in terms of amplitudes, the relations between the amplitudes of main field variables are established with making use of complex moduli concept. Characterization of the complex moduli dependence on frequency 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.

Introduction. Harmonic loading of 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, 11]. 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. Therefore taking account of this effect is important for characterization of a material response at different excitation frequencies and temperatures. Concerning polymeric materials, the effect of hysteretic heating has been clearly shown to dramatically affect the mechanical response of the material [3].

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 [2, 4, 6, 10] apply to identify this relationship. 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.

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 [9, 11]. 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 different 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.

This paper is devoted to investigation of the technique applicability to the typical viscoelastic materials such as PR-520, and to determination the frequency effect on complex moduli for isothermal loading case 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.

Time dependent constitutive relations. To accurately predict an overall performance and lifetime of polymer, it is necessary to model time dependent and inelastic responses. Viscoelastic materials such as polymer materials have the particularity of possessing 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. [5,6] 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 used by Pan and co-workers [8]. First, an inelastic potential function based on the Drucker-Prager yield criterion [7] is defined as

$$f = \sqrt{J_2 + \alpha \sigma_{kk}} , \qquad (1)$$

where J_2 is the second invariant of the deviatoric stress tensor that can be expressed as a function of σ_{ij} . The variable σ_{kk} is the sum of the normal stress components and is equal to three times the hydrostatic stress.

The variable α is a state variable which controls the level of the hydrostatic stress effects. According to this model, the inelastic strain components can be expressed in terms of the deviatoric stress components as follows

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

where, $\dot{\varepsilon}_{ij}^{in}$ is the inelastic strain rate tensor which can be defined as a function of deviatoric stress and Z and α are the state variables.

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

$$\sigma_e = \sqrt{3J_2} + \sqrt{3}\alpha\sigma_{kk}, \qquad (3)$$

the evolution of the internal stress state variable Z and the hydrostatic stress state variable α are defined by the equations $\dot{Z} = q(Z_1 - Z)\dot{e}_e^{in}$,

$$\dot{\alpha} = q(\alpha_1 - Z)\dot{e}_e^{in},\tag{5}$$

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{e}_e^{in} in equations (4) and (5) represents the effective deviatoric inelastic strain rate, which was defined as follows

$$\dot{\varepsilon}_{e}^{in} = \dot{e}_{e}^{in} = \sqrt{\frac{2}{3}} \dot{e}_{ij}^{in} \dot{e}_{ij}^{in}, \qquad (6)$$

$$\dot{e}_{ij}^{in} = \dot{\varepsilon}_{ij}^{in} - \dot{\varepsilon}_{_m}^{in} \delta_{ij}, \qquad (7)$$

where \dot{e}_{e}^{in} is the effective deviatoric inelastic strain rate and $\dot{\varepsilon}_{m}^{in}$ is the mean inelastic strain rate, which matches the effective inelastic strain rate definition given by Pan and co-workers [8].

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.

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 [9,11] 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

$$s_{ij} = 2G(e_{ij} - \varepsilon_{ij}^{in}), \sigma_{kk} = 3K_V(\varepsilon_{kk} - \varepsilon^{\theta}),$$
(8)

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, $\dot{\epsilon}_{kk}^{in} = 0$, i.e. the plastic strain rate is deviatoric: $\dot{\epsilon}_{ii}^{in} = \dot{e}_{ii}^{in}$.

According to this model, if a body as a system subjected to harmonic deformation or loading, then its response is also close to harmonic law

$$e_{ij}(t) = e'_{ij}\cos\omega t - e''_{ij}\sin\omega t, \quad s_{ij}(t) = s'_{ij}\cos\omega t - s''_{ij}\sin\omega t.$$
(9)

The complex amplitudes of the deviator of total strain, \tilde{e}_{ij} , inelastic strain, \tilde{e}_{ij}^{in} , and the stress deviator, 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

$$\tilde{s}_{ij} = 2G\tilde{e}_{ij}, \quad \tilde{e}_{ij}^{in} = \lambda \tilde{e}_{ij}, \quad N = 1, 2, 3, \dots,$$
 (10)

here

 $\tilde{e}_{ii} =$

$$e'_{ij} + ie''_{ij}, \quad \tilde{s}_{ij} = s'_{ij} + is''_{ij}, \quad \tilde{e}^{in}_{ij} = e'^{in}_{ij} + ie''^{nin}_{ij}, \quad \tilde{G} = G'_N + iG''_N, \quad \tilde{\lambda}_N = \lambda'_N + i\lambda''_N, \tag{11}$$

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

$$\tilde{G} = \tilde{G}_N(e_0, \omega, \theta), \quad \tilde{\lambda}_N = \tilde{\lambda}_N(e_0, \omega, \theta),$$
(12)

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

$$G_N'' = \frac{\langle D' \rangle_N}{\omega e_0^2}, \quad \lambda_N'' = \frac{G_N''}{G_0}, \quad \langle (\cdot) \rangle_N = \frac{1}{T} \int_{T(N-1)}^{TN} (\cdot) dt, \quad T = \frac{2\pi}{\omega}, \tag{13}$$

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

$$G'_{N}(e_{0},\omega) = \left[\frac{s_{aN}^{2}(e_{0},\omega)}{4e_{0}^{2}} - G''^{2}(e_{0},\omega)\right]^{1/2},$$

$$\lambda'_{N}(e_{0},\omega) = \left[\frac{s_{aN}^{2}(e_{0},\omega)}{4e_{0}^{2}} - {\lambda''^{2}}(e_{0},\omega)\right]^{1/2},$$
(14)

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 (14). According to equation (12), the complex moduli for isothermal loading case depend on the frequency and amplitude of kinematic loading only. The purpose of this paper is to investigate the influence of these parameters on complex moduli.

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 (2), 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. 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 formulae (13) and (14). 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 (2), (4), (5) and evolutionary equations

$$\dot{\varepsilon}_{12}^{in} = 2D_0 \exp\left(-\frac{1}{2} \left(\frac{Z^2}{3S_{12}^2}\right)^n\right) \left(\frac{S_{12}}{2|S_{12}|}\right),\tag{15}$$

$$\dot{Z} = \frac{2}{\sqrt{3}} q D_0 \left(Z_1 - Z \right) \exp \left(-\frac{1}{2} \left(\frac{Z^2}{3S_{12}^2} \right)^n \right) \left(\frac{S_{12}}{|S_{12}|} \right), \tag{16}$$

$$\dot{\alpha} = \frac{2}{\sqrt{3}} q D_0 \left(\alpha_1 - \alpha \right) \exp \left(-\frac{1}{2} \left(\frac{Z^2}{3S_{12}^2} \right)^n \right) \left(\frac{S_{12}}{|S_{12}|} \right).$$
(17)

The law of strain deviator variation $e = e_0 \sin \omega t$, as well as Hooke law for shear stress

$$s_{12} = 2G(e_{12} - \varepsilon_{12}^{in}), \tag{18}$$

should be added to the system.

The values of material constants for RP-520, which were used for calculations, have been taken from [5]. The list of the values is given below

$$E = 3250$$
 MPa, $D_0 = 10^{\circ}$ 1/sec, $n = 0.92$, $q = 253.6$,

$$Z_0 = 407.5$$
 MPa, $Z_1 = 768.6$ MPa, $\alpha_0 = 0.571$, $\alpha_1 = 0.122$, $\nu = 0.4$.

Numerical results and discussion. The results of transient response simulation and frequency effects (short and long times) on the complex moduli in the frame of modified technique described above are presented. Evolution of stress and inelastic strain for epoxy resin (PR-520) under harmonic loading in pure shear with strain amplitude $e_0 = 5.5 \cdot 10^{-2}$ are shown in Fig. 1 and Fig. 2 respectively for frequency 1 Hz. The material demonstrates cyclically stable response over the whole interval of loading amplitudes and frequencies investigated. According to Fig. 3, stabilization of the response amplitude occurs after the several initial cycles. Relatively slow stabilization is observed only in the vicinity of yield point. Fig. 4 illustrates the mechanical hysteresis phenomenon under cyclic loading in the maximum dissipation condition ($e_0 = 5.5 \cdot 10^{-2}$) at the frequency 1 Hz.

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.



Fig. 1. Stress evolution under harmonic loading

Fig. 2. Inelastic strain evolution under harmonic 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 formulae (14) for $N \to \infty$) are shown in Fig. 5. The curves are calculated for cyclic pure shear for different frequencies (1, 50,100 Hz). 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.



Fig. 3. Stabilized cyclic diagram for PR-520 at 1 Hz

Fig. 4. Hysteresis loops

Using the cyclic diagram and making use of the formulae (13) and (14), 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.6. for 1, 50, 100 Hz by solid, circle-solid and dashed lines, respectively. This figure and cyclic diagram show the inelastic behavior will be started at higher strain amplitude with increase of frequency.

The trend of storage moduls behavior presented in Fig. 6 show that the it's values increase with increase of frequency. The loss modulus varies slightly. The peak values of the modulus increase insignificantly. Within the interval of interest between 1 and 100 Hz, the maximum in loss modulus occurs in the vicinity of 6% of strain intensity. For higher values of strain intensity, the loss modulus decreases.

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.

Results of calculations show evidently that, the strength of material increases with increase of frequency. The sensitivity of cyclic diagrams to frequency variations at the low values is more profound than at the region of higher frequency (see Fig.5). 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.



Fig. 5. Cyclic diagram for PR-520 at 1, 50, 100 Hz



Fig. 6. The real and imaginary parts of complex modulus at various frequencies

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Хашемі М., асп.

Жук Я., д-р фіз.-мат. наук, проф. Київський національний університет імені Тараса Шевченка, Київ

ВПЛИВ ЧАСТОТИ І АМПЛІТУДИ ГАРМОНІЧНОГО НАВАНТАЖЕННЯ

НА КОМПЛЕКСНІ МОДУЛІ ПОЛІМЕРНИХ МАТЕРІАЛІВ

Проведено моделювання і характеризацію реакції полімерного матеріалу на моногармонічне кінематичне навантаження. Для описання нестаціонарної поведінки полімеру використані визначальні рівняння моделі Голдберга. Для моделювання реакції матеріалу в термінах амплітуд встановлені амплітудні співвідношення для основних польових змінних. Для цього використано концепцію комплексних модулів. Досліджено залежність комплексних модулів від частоти та амплітуди інтенсивності деформацій. Показано, що коефіцієнт втрат демонструє низьку залежність від частоти навантаження в усьому дослідженому інтервалі частот.

Хашеми М., асп.

Жук Я., д-р физ.-мат. наук, проф. Киевский национальный университет имени Тараса Шевченко, Киев

ВЛИЯНИЕ ЧАСТОТЫ И АМПЛИТУДЫ ГАРМОНИЧЕСКОГО НАГРУЖЕНИЯ НА КОМПЛЕКСНЫЕ МОДУЛИ ПОЛИМЕРНЫХ МАТЕРІАЛОВ

Проведены моделирование и характеризация реакции полимерного материала на моногармоническое кинематическое нагружение. Для описания нестационарного поведения полимера использованы определяющие уравнения модели Голдберга. Для моделирования реакции материала в терминах амплитуд установлены амплитудные соотношения для основных полевых переменных. Для этого использована концепция комплексных модулей. Исследована зависимость комплексных модулей от частоты и амплитуды интенсивности деформаций. Показано, что коэффициент потерь демонстрирует спабую зависимость частоты нагружения во всем исследованном интервале частот.