Experimentální Analýza Napětí 2011
Experimental Stress Analysis

Viscoelastic Behaviour of Carbon Black Filled Rubber

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Abstract: The rate-dependent behaviour of carbon-black filled rubber is investigated in tensile tests with different loading rates and in relaxation tests. The viscosity-induced rate-dependent effects are described. The storage and loss moduli and phase angle δ dependency on different amplitudes and frequencies are determined by strain controlled dynamical mechanical analysis. The temperature dependence of dynamic and tribological behaviour of SBR is also investigated.

Keywords: Rubber, Carbon-black filled, Viscoelastic, DMA

1. Introduction

In order to determine rate-dependent properties of examined rubber we previously performed experimental measurements of the time dependent response and of damping properties of rubber materials consisting of uniaxial creep and stress relaxation tests which were convenient for studying material response at long times. The behaviour at different strain levels was examined in detail through quasistatic cyclic tests and in simple and multistep relaxation tests. The viscosity-induced rate-dependent effects were described and parameters of the material model were determined. The model was implemented into FE code [1]. Next we focused on the dynamic mechanical analysis of filler-reinforced rubber. The dependence of the storage and dissipation moduli on the static pre-strain, on the deformation amplitude and on the frequency was investigated [2].

The dynamic mechanical analysis (DMA) is well suited for the identification of the short-time range of rubber response. DMA consists of dynamic tests, in which the force resulting from a sinusoidal strain controlled loading is measured.

Payne [3] first pointed out that the moduli of carbon black filled rubber decrease with increasing deformation amplitudes. By means of further tests he reached the conclusion that this behaviour has to be attributed to a thixotropic change. Lion [4] observed that both the storage and the dissipation modulus depend on the frequency of the deformation process. This variation is weakly pronounced and it is of power series type approximately. In terms of the theory of linear viscoelasticity this behaviour corresponds to a continuous relaxation time distribution. With increasing temperatures Lion [4] observed both a decrease in moduli and a lessening of the frequency dependence. The dependence of the dynamic moduli on the filler content and the static pre-strain is investigated in detail by Namboodiri and Tripathy [5].

When a viscoelastic material is subjected to a sinusoidally varying strain after some initial transients the stationary stress-response will be reached in which the resulting stress is also sinusoidal, having the same angular frequency but advanced in phase by an angle δ. Then

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the strain lags the stress by the phase angle δ. The axial displacement $u(t)$ consists of a static pre-strain u_0 under tension which is superimposed by small sinusoidal oscillations:

$$u(t) = u_0 + \Delta u \, sin(2\pi ft).$$

Stresses and strains are calculated with respect to the reference geometry [6] of the pre-deformed specimen

$$\varepsilon_0 = u_0 / (L_0 + u_0), \quad \Delta\varepsilon = \Delta u / (L_0 + u_0), \tag{1}$$

where L_0 is the undeformed length of the specimen. The force response $F(t)$ of the specimen is a harmonic function and can be written as :

$$F(t) = F_0 + \Delta F \sin(2\pi ft + \delta). \tag{2}$$

F_0 is the static force depending only on the pre-deformation u_0. The force amplitude ΔF and the phase angle δ depend, in general, on the pre-deformation, the frequency and the strain amplitude [6, 7]. If the incompressibility of the rubber is assumed $A_0 L_0 = A(L_0+u_0)$, where A_0 is the cross-sectional area of the undeformed specimen, we can relate the force to the cross-sectional area A of the pre-deformed specimen:

$$\begin{aligned}\sigma(t) &= \frac{F(t)}{A} = \sigma_0 + \\ &+\Delta\sigma\left[cos(\delta) sin(2\pi ft) + sin(\delta) cos(2\pi ft)\right].\end{aligned} \tag{3}$$

The dynamic stress-response σ(t) normalized by the deformation amplitude $\Delta\varepsilon$ can be written:

$$\begin{aligned}\sigma(t) &= \sigma_0 + \Delta\varepsilon\left[E'(\varepsilon_0, f, \Delta\varepsilon) sin(2\pi ft) + \right.\\ &\left.+ E''(\varepsilon_0, f, \Delta\varepsilon) cos(2\pi ft)\right],\end{aligned} \tag{4}$$

where

$$E'(\varepsilon_0, f, \Delta\varepsilon) = \frac{\Delta\sigma}{\Delta\varepsilon} cos(\delta), \tag{5}$$

and

$$E''(\varepsilon_0, f, \Delta\varepsilon) = \frac{\Delta\sigma}{\Delta\varepsilon} sin(\delta) \tag{6}$$

are the storage and dissipation moduli respectively and δ is the phase angle. In general, carbon black-reinforced rubber has fairly weak frequency dependence in conjunction with a pronounced amplitude dependence [7]. If the strain amplitude $\Delta\varepsilon$ increases, the storage modulus E' lessens and the dissipation modulus E" shows a more or less pronounced sigmoidal behaviour - Payne effect. If the material is linear viscoelastic, then these two moduli depend neither on the deformation amplitude nor on the static pre-strain. The damping factor or loss tangent (tan δ) which is the ratio E"/ E' is the measure of mechanical energy dissipated as heat during the dynamic cycle. If the dynamic strain amplitude is constant in time, we can observe time-independent moduli [4]. These phenomena are frequently interpreted as a dynamic state of equilibrium between breakage and recovery of physical bonds linking adjacent filler clusters. The most common model of this state is Kraus model [8, 9] which describes the amplitude dependence of dynamic moduli. The influence of static pre-deformation ε_0 is included in different models [10, 11] and the uniaxial form of the frequency, amplitude and pre-strain dependent dynamical moduli is proposed by Lion [12].

The purpose of present paper is to summarize the results of experimental research of the behaviour of rubber under dynamic loading conditions in harmonic strain-controlled tests under tension and to show the dependence of the storage and dissipation moduli on the frequency, on the deformation amplitude, on the static pre-strain and different temperatures.

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2. Experimental measurements

DMA tests under sinusoidal tension mode were carried out on an electro-dynamic testing machine Instron ElectroPuls E3000 equipped with an environmental chamber and with WaveMatrix software. The specimens were thin rectangular strips of length 160 mm, width 25 mm and thickness 2.75 mm. The basic sampling frequency was 100 Hz and was increased up to 500 Hz when needed according to the testing frequency used.

The dynamic properties of SBR were investigated at different temperature levels. The tensile loading was strain-controlled. Every test was performed on a virgin specimen. At the chosen static pre-deformation ε_0, the frequency and the strain amplitude $\Delta\varepsilon$ were changed in order to determine their influence to the storage (SM) and loss moduli (LM) and to hysteretic losses. Before each test the virgin specimens were preconditioned in order to exclude the Mullins effect. The preloading process started on the static pre-strain ε_0 and consisted of 10 cycles with the maximum strain amplitude to be reached in the subsequent experiment. After that specimens relaxed 15 min at static pre-strain ε_0. After this preconditioning, the mean stress σ0 changed only little in the subsequent cyclic loading.

Raw test data were recorded by a PC and evaluated in the Matlab Signal Processing Toolbox. The discrete Fourier transform was used to determine the frequency content of force and displacement signals and to calculate the phase delay δ between them. Furthermore, we determined the complex dynamic modulus as the ratio between the amplitudes of stress and strain and dynamic moduli were calculated according to the Eqn. (5-6).

2.1. Testing at ambient temperature

In order to determine the dependence of the dynamic moduli we carried out the tests at the temperature 22°C with five frequencies and amplitudes $\Delta\varepsilon$ and with three static pre-strains ε_0 as shown in Tab. 1.

Table 1. Parameters of testing at 22°C

$\Delta\varepsilon$	0.014	0.028	0.042	0.056	0.070
f [Hz]	1.0	2.5	5.0	7.5	10.0
ε_0	0.17	0.17	0.21	0.21	0.25

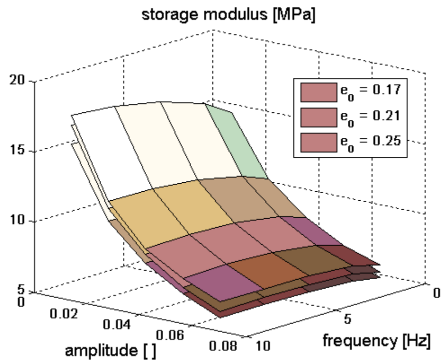

Fig. 1. SM amplitude and frequency dependence

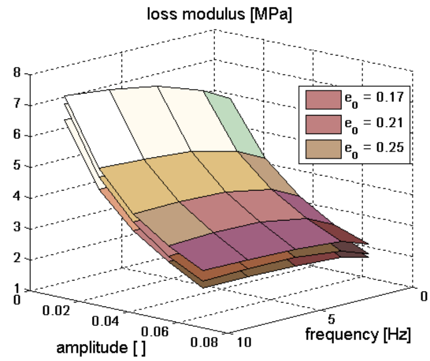

Fig. 2. LM amplitude and frequency dependence

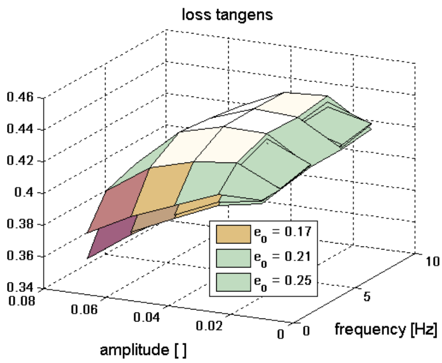

Fig. 3. LA amplitude and frequency dependence

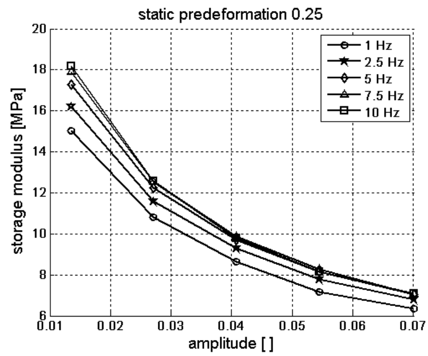

Fig. 4. SM amplitude and frequency dependence

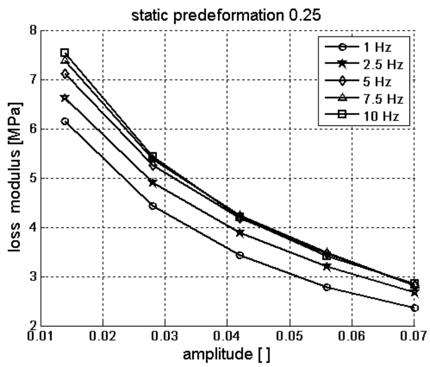

Fig. 5. LM amplitude and frequency dependence

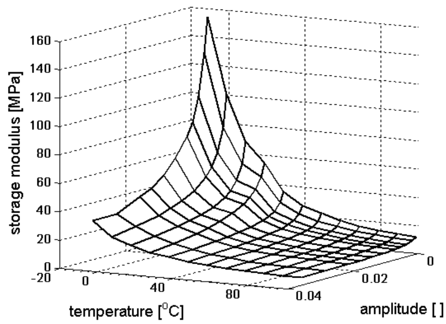

Fig. 6. SM amplitude and temperature dependence

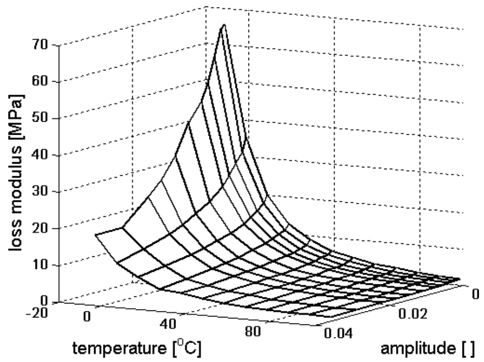

Fig. 7. LM amplitude and temperature dependence

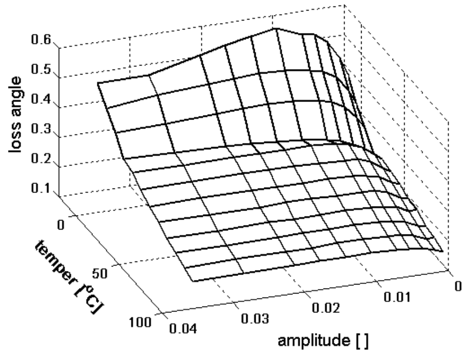

Fig. 8. LA amplitude and temperature dependence

After the preconditioning at the given pre-strain ε_0 the test started at the smallest amplitude and the frequency sweep in the chosen range was performed then the amplitude was raised to the next value. The number of cycles executed at each frequency step was between 200 and 300 and was adapted to achieve a steady state. The results of tests are represented by synoptic graphs in Fig. 1-3 where the storage and dissipation moduli and the loss angle (LA) are plotted as a function of frequency and amplitude for the static pre-deformation ε_0 as a parameter. We can make the three following essential conclusions:

- The storage and dissipation moduli increase and the loss angle decreases with increasing static pre-strain ε_0.
- The storage modulus and the loss modulus increase slightly with increasing frequency i.e. increasing frequencies lead to an increase in stiffness and an increase in energy loss. The graph of the loss angle has a convex shape and shows a slight maximum in the range of applied frequencies and amplitudes.
- Both moduli show a pronounced decrease with an increasing strain amplitudes – so called Payne effect.

The Payne effect is explained by a concept [4, 6, 13] that during cyclic deformations the weak physical bonds between molecules of rubber and clusters of filler are breaking and recovering continually. The rate of breakage is assumed to be an increasing function of the strain amplitude and the rate of recovery is a decreasing function. The storage modulus is assumed to be proportional to the total number of intact bonds and the dissipation modulus to the rate of breakage per unit of time.

The detailed dependence of the storage and dissipation moduli on amplitude for different frequencies is shown in Fig. 4-5. The both moduli decrease monotonically with increasing strain amplitudes. In our range of amplitudes the loss modulus does not show any sigmoidal behaviour which was reported by Lion and Kardelky [6].

2.2. Temperature Dependency of Dynamic Properties

In order to determine the influence of temperature on dynamic properties of examined SBR rubber another series of tests was carried on with the temperature sweep for different frequencies and amplitudes and with a sole static pre-strain ε_0 = 0.29. The tests were accomplished in the Instron 3119 Environmental Chamber suitable for a temperature range from -70 to +250°C.

Influence of Strain Amplitude. The first series of tests was lead with the temperature sweep from -10°C to 100°C with a step 10°C. The frequency of the strain controlled loading was fixed at 5 Hz. After each temperature step the specimens relaxed 15 min at the static pre-strain. Then the cyclic loading started and the amplitude sweep was performed in a range from 0.001 to 0.036. The number of cycles was 200 at each amplitude.

Results of the tests are shown in Fig. 6.-8. We see a similar dependency of the storage and loss moduli on the temperature and strain amplitude. Both moduli are rising sharply with decreasing temperature but their dependence on the strain amplitude (Payne effect) is much more pronounced in the low temperature zone as can also be seen on the detailed graphs in Fig. 9 and 10 where the amplitude dependency is displayed with the temperature as a parameter.

The loss angle in Fig. 8 increases initially with the increasing amplitudes and at low temperatures it decreases after reaching a summit or it remains at approximately the same values at higher temperatures.

All dynamic properties deteriorate considerably at higher temperatures. Rubber loses its elasticity and damping properties. Similar results are reported by other authors [15].

Described tests were conducted also at frequencies 2.5 Hz and 7.5 Hz. The values of investigated quantities did not show substantial differences in this frequency range.

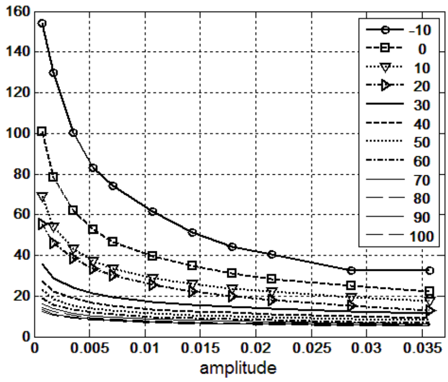

Fig. 9. SM amplitude and temperature dependence

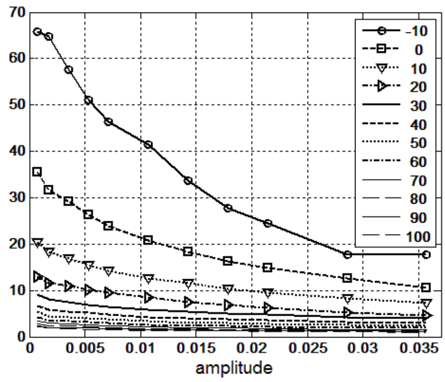

Fig. 10. LM amplitude and temperature depend

3. Conclusion

In this experimental essay we present the results of dynamic testing of SBR rubber under different conditions In order to investigate the internal damping of rubber a complex experimental research of dynamic properties was lead by DMA at different frequencies, strain amplitudes and temperatures. The dependency of storage and loss moduli and of loss angle on these quantities was identified and displayed synoptically.

We should emphasize that material properties of rubber are affected by the temperature to a great extent. Results show that the response of rubber changes at temperatures even slightly different than the ambient temperature which leads to a drastic change in the material properties.

Another salient property of rubber is its phenomenal memory. Rubber remembers almost all that happened to it from the beginning and the slightest change in the experimental procedure leads to the scattering of results.

Acknowledgement

This work was supported by a grant from Ministry of Education of Czech Republic FRVŠ 2889/2011.

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