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A RHEOLOGICAL MODEL OF POLYMERIC MATERIALS AND IDENTIFICATION OF ITS PARAMETERS

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A new rheological model of polymeric materials has been proposed, based on experimental data concerning creep of tensile samples. A mechanical model of the material consists of four elements connected in series, formulated by Hooke, Wilczyński, Kelvin and Newton. Constitutive equations of rheology of the polymeric material have been formulated, both in uncoupled and coupled form, with the analytical generating functions well-fitting the experiments. A method for the identification of the elastic, viscoelastic and viscous material constants, based on the creep of the tensile samples, has been developed and computerised. The introductory experiments have been carried out on samples made of POLIMAL 109 polyester resin and the results of identification of the material constants have been given.

Key words: polymers, rheology, constitutive equations, combined numerical and experimental methods

1. Introduction

Recent experiments showed that creep of polymeric materials contains three main components, i.e. elastic, viscoelastic and viscous ones, e.g. Wilczyński (1968) and Ochelski (1997). The experiments also pointed out that the viscoelastic component differs substantially from the Kelvin model, Wilczyński (1968).

In order to illustrate the proposed description of the polymeric material, the classic mechanical models will be used. Three well-known rheological models of polymeric materials, treated as isotropic solid media, are shown in

Fig.1a,b,c. The Hooke-Kelvin-Newton (H-K-N) model, presented in Fig.1a, consists of the Hooke, Kelvin and Newton elements, connected in series, cf Wilczyński (1984). The H-nK-N model, drawn in Fig.1b, consists of the Hooke element (the elastic part), n elements formulated by Kelvin (the viscoelastic part) and the Newton element (the viscous part), cf Wilczyński (1984). The H-W-N model, shown in Fig.1c, contains the Wilczyński element, described by a fractional exponent generating function. That means the viscoelastic properties of the Wilczyński element vary in time. The use of the fractional exponent generating functions to describe viscoelastic properties of polymers has first proposed by Rabotnov (1966). Wilczyński (1978, 1996) developed Rabotnov's approach via transforming the fractional exponent function series, weakly converging, to an integral form as well as via uncoupling the distortional and volumetric creep. On the other hand, Wilczyński neglected pure viscous part of the creep of polymers.

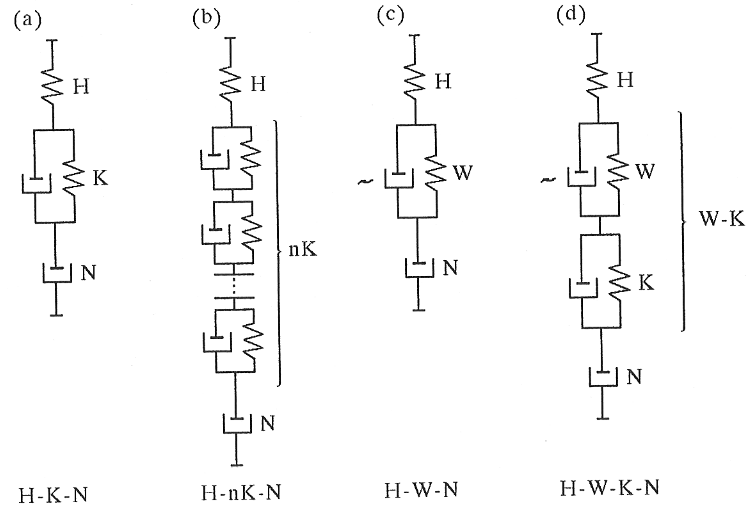

Fig. 1. Rheological models of polymers

Taking into account the experimental data, one can observe that the H-K-N model poorly fits the viscoelastic part of the creep of polymeric materials. The H-nK-N model slightly better approximates that part of the creep, but incorporates some qualitative errors. The H-W-N model states a substantial progress as it fits well the viscoelastic part of the creep in a large time interval, except for relatively shorter and longer times.

In this study, a new rheological model of polymeric materials will be proposed, denoted by the H-W-K-N symbol, and illustrated in Fig.1d, in a mechanical form. The viscoelastic strains result from the Wilczyński and Kelvin elements. This model will be formulated in the successive sections of this study. The result of experimental verification of the model will be given as well.

2. Uncoupled constitutive equations of elasticity and rheology for an isotropic material

Let x, y, z denote directions of a Cartesian coordinate system, assumed for an isotropic material. Constitutive equations of linear elasticity of a given material can be written after uncoupling shear (distortional) and bulk (volumetric) strains. Using the matrix notation, the uncoupled equations have the following form, e.g. Wilczyński and Klasztorny (2000)

$$\boldsymbol{\varepsilon}_s = J_s \boldsymbol{\sigma}_s \qquad\qquad \varepsilon_b = J_b \sigma_b \tag{2.1}$$

where

$$\begin{aligned}
&\boldsymbol{\varepsilon}_s = \left[\varepsilon_{xx} - \varepsilon_b, \varepsilon_{yy} - \varepsilon_b, \varepsilon_{zz} - \varepsilon_b, \varepsilon_{yz}, \varepsilon_{xz}, \varepsilon_{xy}\right]^\top \\
&\boldsymbol{\sigma}_s = \left[\sigma_{xx} - \sigma_b, \sigma_{yy} - \sigma_b, \sigma_{zz} - \sigma_b, \sigma_{yz}, \sigma_{xz}, \sigma_{xy}\right]^\top \\
&\varepsilon_b = \frac{1}{3}(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \qquad\qquad \sigma_b = \frac{1}{3}(\sigma_{xx} + \sigma_{yy} + \sigma_{zz}) \\
&J_s = \frac{1}{2G} \qquad\qquad J_b = \frac{1}{3B} \\
&G = \frac{E}{2(1+\nu)} \qquad\qquad B = \frac{E}{3(1-2\nu)}
\end{aligned} \tag{2.2}$$

and where

ε_{jk} – components of the strain tensor, $j,k = x,y,z$

σ_{jk} – components of the stress tensor, $j,k = x,y,z$

$\boldsymbol{\varepsilon}_s$ – strain vector creating the deviator of the strain tensor

ε_b – strain defining the isotropic part of the strain tensor

$\boldsymbol{\sigma}_s$ – stress vector creating the deviator of the stress tensor

σ_b – stress defining the isotropic part of the stress tensor

E, ν, G, B – elasticity constants (Young's modulus, Poisson's ratio, Kirchhoff's modulus, Helmholtz's modulus)

J_s, J_b – shear and bulk elastic compliances.

The following assumptions have been adopted:

- The polymeric material (a high-molecular plastic) is an isotropic body, with linear elastic-viscoelastic-viscous properties.
- The mechanical model of the material is presented in Fig.1d.
- Processes of the deformation of the material are isothermal.

The uncoupled constitutive equations of rheology, describing an isotropic material, can be formulated with the Boltzmann integral operators (cf Wilczyński. 1996, 1997; Wilczyński and Klasztorny, 2000)

$$\tilde{\boldsymbol{\varepsilon}}_s(t) = \tilde{J}_s(t) \otimes \tilde{\boldsymbol{\sigma}}_s(t) \qquad \tilde{\varepsilon}_b(t) = \tilde{J}_b(t) \otimes \tilde{\sigma}_b(t) \tag{2.3}$$

where

$$\begin{gathered}
\tilde{\boldsymbol{\varepsilon}}_s = [\tilde{\varepsilon}_{xx} - \tilde{\varepsilon}_b, \tilde{\varepsilon}_{yy} - \tilde{\varepsilon}_b, \tilde{\varepsilon}_{zz} - \tilde{\varepsilon}_b, \tilde{\varepsilon}_{yz}, \tilde{\varepsilon}_{xz}, \tilde{\varepsilon}_{xy}]^\top \\
\tilde{\boldsymbol{\sigma}}_s = [\tilde{\sigma}_{xx} - \tilde{\sigma}_b, \tilde{\sigma}_{yy} - \tilde{\sigma}_b, \tilde{\sigma}_{zz} - \tilde{\sigma}_b, \tilde{\sigma}_{yz}, \tilde{\sigma}_{xz}, \tilde{\sigma}_{xy}]^\top \\
\tilde{\varepsilon}_b = \frac{1}{3}(\tilde{\varepsilon}_{xx} + \tilde{\varepsilon}_{yy} + \tilde{\varepsilon}_{zz}) \qquad \tilde{\sigma}_b = \frac{1}{3}(\tilde{\sigma}_{xx} + \tilde{\sigma}_{yy} + \tilde{\sigma}_{zz})
\end{gathered} \tag{2.4}$$

and where

$\tilde{J}_s(t), \tilde{J}_b(t)$ – shear and bulk time-dependent compliances
$\tilde{\varepsilon}_{jk}(t)$ – time-dependent components of the stress tensor, $j, k = x, y, z$
$\tilde{\sigma}_{jk}(t)$ – time-dependent components of the strain tensor, $j, k = x, y, z$
$\otimes$ – convolution operator
t – time.

The quantities $\tilde{\boldsymbol{\varepsilon}}_s(t)$, $\tilde{\boldsymbol{\sigma}}_s(t)$, $\tilde{\varepsilon}_b(t)$, $\tilde{\sigma}_b(t)$ are defined in a similar way like $\boldsymbol{\varepsilon}_s$, $\boldsymbol{\sigma}_s$, ε_b, σ_b, with the following relationships

$$\begin{aligned}
\boldsymbol{\varepsilon}_s &= \tilde{\boldsymbol{\varepsilon}}_s(0) \qquad & \boldsymbol{\sigma}_s &= \tilde{\boldsymbol{\sigma}}_s(0) \\
\varepsilon_b &= \tilde{\varepsilon}_b(0) \qquad & \sigma_b &= \tilde{\sigma}_b(0)
\end{aligned}$$

3. Coupled (general) constitutive equations of linear elasticity and rheology for an isotropic material

Uncoupled equations (2.1) can be rewritten in the form

$$\boldsymbol{\varepsilon} - \mathbf{A}\boldsymbol{\varepsilon} = J_s(\boldsymbol{\sigma} - \mathbf{A}\boldsymbol{\sigma}) \qquad \mathbf{A}\boldsymbol{\varepsilon} = J_b\mathbf{A}\boldsymbol{\sigma} \tag{3.1}$$

where

$$\boldsymbol{\varepsilon} = \begin{bmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \\ \varepsilon_{yz} \\ \varepsilon_{xz} \\ \varepsilon_{xy} \end{bmatrix} \qquad \boldsymbol{\sigma} = \begin{bmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \\ \sigma_{yz} \\ \sigma_{xz} \\ \sigma_{xy} \end{bmatrix} \qquad \mathsf{A} = \frac{1}{3}\begin{bmatrix} 1 & 1 & 1 & 0 & 0 & 0 \\ 1 & 1 & 1 & 0 & 0 & 0 \\ 1 & 1 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}$$

Summing Eqs $(3.1)_1$ and $(3.1)_2$, one obtains the general constitutive equations of the elasticity of an isotropic material, in the form of coupled linear equations, cf Daniel (1994), i.e.

$$\boldsymbol{\varepsilon} = \mathsf{J}\boldsymbol{\sigma} \tag{3.2}$$

where

$$\mathsf{J} = J_s(\mathsf{I} - \mathsf{A}) + J_b\mathsf{A} = \begin{bmatrix} J_1 & J_2 & J_2 & 0 & 0 & 0 \\ J_2 & J_1 & J_2 & 0 & 0 & 0 \\ J_2 & J_2 & J_1 & 0 & 0 & 0 \\ 0 & 0 & 0 & J_s & 0 & 0 \\ 0 & 0 & 0 & 0 & J_s & 0 \\ 0 & 0 & 0 & 0 & 0 & J_s \end{bmatrix} \tag{3.3}$$

and

$$\begin{aligned} J_1 &= \frac{2}{3}J_s + \frac{1}{3}J_b = \frac{1}{E} \\ J_2 &= -\frac{1}{3}J_s + \frac{1}{3}J_b = -\frac{\nu}{E} \end{aligned} \tag{3.4}$$

Symbol $\mathsf{I} = \mathrm{diag}(1,1,1,1,1,1)$ denotes the identity matrix.

In a similar way, Eqs (2.3) can be rewritten in the following form

$$\widetilde{\boldsymbol{\varepsilon}} - \mathsf{A}\widetilde{\boldsymbol{\varepsilon}} = \widetilde{J}_s \otimes (\widetilde{\boldsymbol{\sigma}} - \mathsf{A}\widetilde{\boldsymbol{\sigma}}) \qquad \mathsf{A}\widetilde{\boldsymbol{\varepsilon}} = J_b \otimes \mathsf{A}\widetilde{\boldsymbol{\sigma}} \tag{3.5}$$

with

$$\begin{aligned} &\widetilde{J}_s \otimes (\widetilde{\boldsymbol{\sigma}} - \mathsf{A}\widetilde{\boldsymbol{\sigma}}) = \widetilde{J}_s \otimes (\mathsf{I} - \mathsf{A})\widetilde{\boldsymbol{\sigma}}) = \widetilde{J}_s(\mathsf{I} - \mathsf{A}) \otimes \widetilde{\boldsymbol{\sigma}} \\ &\widetilde{J}_b \otimes \mathsf{A}\widetilde{\boldsymbol{\sigma}} = \widetilde{J}_b\mathsf{A} \otimes \widetilde{\boldsymbol{\sigma}} \end{aligned} \tag{3.6}$$

Inserting Eqs (3.6) into Eqs (3.5) and summing Eqs $(3.5)_1$ and $(3.5)_2$, one obtains the coupled (general) constitutive equations of rheology for an isotropic material, in the form

$$\widetilde{\boldsymbol{\varepsilon}}(t) = \widetilde{\mathbf{J}}(t) \otimes \widetilde{\boldsymbol{\sigma}}(t) \tag{3.7}$$

where

$$\widetilde{\boldsymbol{\varepsilon}} = \begin{bmatrix} \widetilde{\varepsilon}_{xx} \\ \widetilde{\varepsilon}_{yy} \\ \widetilde{\varepsilon}_{zz} \\ \widetilde{\varepsilon}_{yz} \\ \widetilde{\varepsilon}_{xz} \\ \widetilde{\varepsilon}_{xy} \end{bmatrix} \qquad \widetilde{\boldsymbol{\sigma}} = \begin{bmatrix} \widetilde{\sigma}_{xx} \\ \widetilde{\sigma}_{yy} \\ \widetilde{\sigma}_{zz} \\ \widetilde{\sigma}_{yz} \\ \widetilde{\sigma}_{xz} \\ \widetilde{\sigma}_{xy} \end{bmatrix}$$

and

$$\widetilde{\mathbf{J}}(t) = \widetilde{J}_s(\mathbf{I} - \mathbf{A}) + \widetilde{J}_b\mathbf{A} = \begin{bmatrix} \widetilde{J}_1 & \widetilde{J}_2 & \widetilde{J}_2 & 0 & 0 & 0 \\ \widetilde{J}_2 & \widetilde{J}_1 & \widetilde{J}_2 & 0 & 0 & 0 \\ \widetilde{J}_2 & \widetilde{J}_2 & \widetilde{J}_1 & 0 & 0 & 0 \\ 0 & 0 & 0 & \widetilde{J}_s & 0 & 0 \\ 0 & 0 & 0 & 0 & \widetilde{J}_s & 0 \\ 0 & 0 & 0 & 0 & 0 & \widetilde{J}_s \end{bmatrix}$$

$$\widetilde{J}_1(t) = \frac{2}{3}\widetilde{J}_s(t) + \frac{1}{3}\widetilde{J}_b(t) \qquad \widetilde{J}_2(t) = -\frac{1}{3}\widetilde{J}_s(t) + \frac{1}{3}\widetilde{J}_b(t) \tag{3.8}$$

Equations $(3.8)_2$ give the following inverse relationships

$$\widetilde{J}_s(t) = \widetilde{J}_1(t) - \widetilde{J}_2(t) \qquad \widetilde{J}_b(t) = \widetilde{J}_1(t) + 2\widetilde{J}_2(t) \tag{3.9}$$

Hence, the shear and bulk time-dependent compliances are obtained as simple transforms of two well-conditioned time-dependent compliances $\widetilde{J}_1$, $\widetilde{J}_2$ termed here as directional compliances.

4. A new rheological model of polymeric materials

As Poisson's ratio for polymeric materials is greater than 0.4, see Wilczyński (1996), the creep of a unidirectionally tensioned sample results in relatively small bulk strains, when compared with the shear strains (see Eqs (2.2), (2.4)).

Time histories of the bulk creep of the polymeric samples exhibit some irregularities, as mentioned in point 8, hence they are difficult for direct analytical description. On the other hand, the time histories of the directional creep $\widetilde{\varepsilon}_{xx}(t)$, $\widetilde{\varepsilon}_{yy}(t)$ are monotone and well measurable, thus they have been selected for direct approximation. The generating functions are formulated for the directional time compliances $\widetilde{J}_1(t)$, $\widetilde{J}_2(t)$. The shear and bulk time compliances can be calculated from simple Eqs (3.9).

The directional compliances, occurring in the compliance matrix $\widetilde{\mathbf{J}}$, corresponding to the H-W-K-N rheological model, have the following form

$$\widetilde{J}_i(t) = J_i\Big(1+\omega_i\int\limits_0^t [\gamma_i\Phi_i(t-\vartheta)+(1-\gamma_i)F_i(t-\vartheta)]\;d\vartheta+\beta_i\int\limits_0^t d\vartheta\Big) \qquad i=1,2 \tag{4.1}$$

where

Φ_1/Φ_2 – fractional exponent functions, related to the first/second directional compliance (load history functions for the W element)

F_1/F_2 – exponent functions, related to the first/second directional compliance (load history functions for the K element)

ω_1/ω_2 – viscoelastic constants determining the first/second long-lasting compliance

γ_1/γ_2 – fractions determining the contribution of the elements W and K to the total viscoelastic strains, related to the first/second directional compliance

τ_1^N/τ_2^N – retardation times of the Newton element, related to the first/second directional compliance, with

$$\beta_i = \frac{1}{\tau_i^N} \qquad i=1,2 \tag{4.2}$$

The load history functions are defined by the following formulae (see Wilczyński, 1978, 1997; Wilczyński and Klasztorny 2000)

$$\Phi_i(t) = \alpha_i\int\limits_0^\infty \mathrm{e}^{-\alpha_i\xi t}\xi\Lambda_i(\xi)\;d\xi \qquad\qquad F_i(t) = \kappa_i\mathrm{e}^{-\kappa_i t} \qquad i=1,2 \tag{4.3}$$

where $(i=1,2)$

$$\Lambda_i = \frac{\sin\pi\mu_i}{\pi}\frac{\xi^{\mu_i-1}}{1+2\xi^{\mu_i}\cos\pi\mu_i+\xi^{2\mu_i}} \qquad \alpha_i = \frac{1}{\tau_i} \qquad 0 < \mu_i < 1 \qquad \kappa_i = \frac{1}{\tau_i^K} \tag{4.4}$$

with

τ_1/τ_2 – retardation times of the W element, related to the first/second directional compliance

μ_1/μ_2 – fractions defining the fractional exponent functions, related to the first/second directional compliance

τ_1^K/τ_2^K – retardation times of the K element, related to the first/second directional compliance.

Summing up, the H-W-K-N model of a polymeric material is described by 14 material constants, i.e. 2 elasticity constants (E,ν), 10 viscoelasticity constants $(\omega_1,\omega_2,\gamma_1,\gamma_2,\tau_1,\tau_2,\mu_1,\mu_2,\tau_1^K,\tau_2^K)$ and 2 viscosity constants (τ_1^N,τ_2^N).

5. Creep of polymeric materials

The material constants that describe the H-W-K-N model will be identified from simple experiments concerning the creep of samples in pure tension. The experiments are carried out on cylindrical samples. Each sample is under a uniform normal stress $\sigma_{xx} = \text{const}$, as shown in Fig.2a. The longitudinal and transverse strains, $\widetilde{\varepsilon}_{xx}(t)$ and $\widetilde{\varepsilon}_{yy}(t)$, are measured for $t \geqslant 0$.

The remaining components of the stress and strain tensor satisfy the following conditions

$$\begin{aligned} &\sigma_{yy} = \sigma_{zz} = 0 && \sigma_{xy} = \sigma_{yz} = \sigma_{xz} = 0 \\ &\widetilde{\varepsilon}_{yy}(t) = \widetilde{\varepsilon}_{zz}(t) && \widetilde{\varepsilon}_{xy}(t) = \widetilde{\varepsilon}_{yz}(t) = \widetilde{\varepsilon}_{xz}(t) \equiv 0 \end{aligned} \tag{5.1}$$

Inserting Eqs (5.1) into Eqs (3.7), taking into consideration Eqs (4.1) ÷ (4.4), one obtains the following functions describing the directional strains

$$\begin{aligned} \widetilde{\varepsilon}_{xx}(t) &= [1+\omega_1\varphi_1(t)+\beta_1 t]\varepsilon_{xx} \\ \widetilde{\varepsilon}_{yy}(t) &= [1+\omega_2\varphi_2(t)+\beta_2 t]\varepsilon_{yy} \end{aligned} \tag{5.2}$$

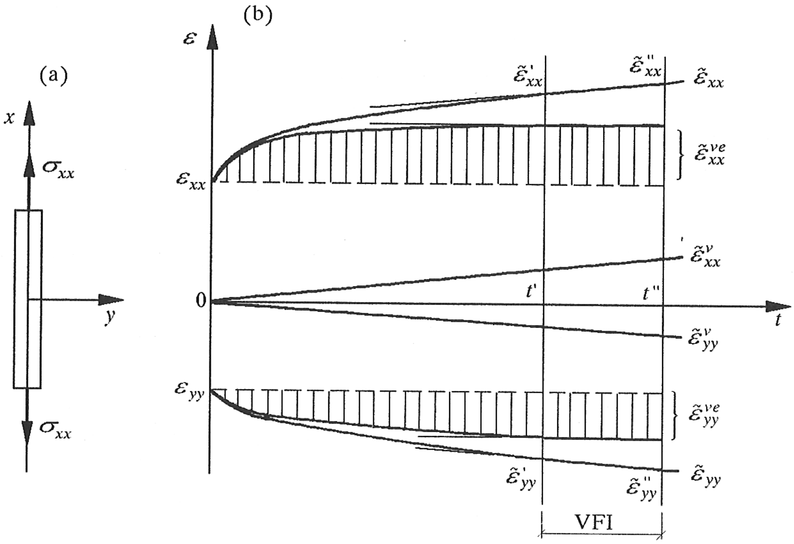

Fig. 2. Longitudinal and transverse creeps of a tensile polymeric sample

where $(i = 1, 2)$

$$\begin{aligned} \varphi_i(t) &= \gamma_i \int\limits_0^t \Phi_i(t-\vartheta)\, d\vartheta + (1-\gamma_i) \int\limits_0^t F_i(t-\vartheta)\, d\vartheta = \\ &= \gamma_i \Big[1 - \int\limits_0^t \exp\Big(\frac{-\xi t}{\tau_i}\Big) \Lambda_i(\xi)\, d\xi\Big] + (1-\gamma_i)\Big[1 - \exp\Big(\frac{-t}{\tau_i^K}\Big)\Big] \end{aligned} \tag{5.3}$$

are the directional creep functions. Eqs (5.3) can be rewritten in the form

$$\varphi_i(t) = 1 - \gamma_i \int\limits_0^t \exp\Big(\frac{-\xi t}{\tau_i}\Big) \Lambda_i(\xi)\, d\xi - (1-\gamma_i) \exp\Big(\frac{-t}{\tau_i^K}\Big) \qquad i = 1, 2 \tag{5.4}$$

Subsequent components in Eqs (5.2) correspond to elements H, W, K, N in the material model. Hence, the total strains contain the elastic, viscoelastic and viscous parts, i.e.

$$\begin{aligned} \tilde{\varepsilon}_{xx}(t) &= \varepsilon_{xx} + \tilde{\varepsilon}^{ve}_{xx}(t) + \tilde{\varepsilon}^{v}_{xx}(t) \\ \tilde{\varepsilon}_{yy}(t) &= \varepsilon_{yy} + \tilde{\varepsilon}^{ve}_{yy}(t) + \tilde{\varepsilon}^{v}_{yy}(t) \end{aligned}$$

where

$$\varepsilon_{xx} = J_1\sigma_{xx} \qquad \widetilde{\varepsilon}^{ve}_{xx}(t) = \varepsilon_{xx}\omega_1\varphi_1(t) \qquad \widetilde{\varepsilon}^{v}_{xx}(t) = \varepsilon_{xx}\beta_1 t$$

$$\varepsilon_{yy} = J_2\sigma_{xx} \qquad \widetilde{\varepsilon}^{ve}_{yy}(t) = \varepsilon_{yy}\omega_2\varphi_2(t) \qquad \widetilde{\varepsilon}^{v}_{yy}(t) = \varepsilon_{yy}\beta_2 t$$

6. An algorithm for identification of the material constants

In the experiment concerning the creep of a sample in pure tension, the directional strains $\widetilde{\varepsilon}_{xx}(t)$, $\widetilde{\varepsilon}_{yy}(t)$, $t \in < 0, t'' >$ are measured in a high-density discrete set of time instants. The elasticity constants can be estimated experimentally, using the classic formulae resulting from Eqs (3.2), (3.4) and (5.1), i.e.

$$E = \frac{\sigma_{xx}}{\varepsilon_{xx}} \qquad \nu = -\frac{\varepsilon_{yy}}{\varepsilon_{xx}}$$

The general shapes of the creep functions $\widetilde{\varepsilon}_{xx}(t)$, $\widetilde{\varepsilon}^{ve}_{xx}(t)$, $\widetilde{\varepsilon}^{v}_{xx}(t)$, $\widetilde{\varepsilon}_{yy}(t)$, $\widetilde{\varepsilon}^{ve}_{yy}(t)$, $\widetilde{\varepsilon}^{v}_{yy}(t)$ are shown in Fig.2b. The time histories of the directional creep strains contain the VFI zone, in which the pure viscous flow of the material decides about the variation in time (Fig.2b). The experiments show that the directional strains in the VFI zone are approximately linearly dependent on time. From Eqs (5.2), (5.4) one obtains the approximate formulae $t \in < t', t'' >$

$$\begin{aligned} \widetilde{\varepsilon}_{xx}(t) &\approx \varepsilon_{xx}(1+\omega_1) + \varepsilon_{xx}\beta_1 t \\ \widetilde{\varepsilon}_{yy}(t) &\approx \varepsilon_{yy}(1+\omega_2) + \varepsilon_{yy}\beta_2 t \end{aligned} \tag{6.1}$$

since $\varphi_1(t) \to 1$, $\varphi_2(t) \to 1$ for $t \to \infty$.

The experimental strains in the VFI zone are approximated with straight lines, i.e.

$$\widetilde{\varepsilon}_{xx}(t) = A_1 + B_1 t \qquad \widetilde{\varepsilon}_{yy}(t) = A_2 + B_2 t \qquad t \in < t', t'' > \tag{6.2}$$

The simplest estimation results in the directional coefficients calculated from the formulae (Fig.2b)

$$B_1 = \frac{\varepsilon''_{xx} - \varepsilon'_{xx}}{t'' - t'} \qquad B_2 = \frac{\varepsilon''_{yy} - \varepsilon'_{yy}}{t'' - t'}$$

and the coefficients A_1, A_2 derived from the condition of collocation at the termined instant

$$A_1 = \varepsilon''_{xx} - B_1 t'' \qquad A_2 = \varepsilon''_{yy} - B_2 t''$$

Comparing Eqs (6.1) and (6.2), one obtains the following formulae for the second group of the material constants

$$\omega_1 = \frac{A_1}{\varepsilon_{xx}} - 1 \qquad\qquad \omega_2 = \frac{A_2}{\varepsilon_{yy}} - 1$$
$$\tau_1^N = \frac{1}{\beta_1} = \frac{\varepsilon_{xx}}{B_1} \qquad\qquad \tau_2^N = \frac{1}{\beta_2} = \frac{\varepsilon_{yy}}{B_2}$$

The formulae for the experimental creep functions result from Eqs (5.2) and have the following form

$$\varphi_1^e(t) = \frac{1}{\omega_1}\Big[\frac{\widetilde{\varepsilon}_{xx}(t)}{\varepsilon_{xx}} - 1 - \beta_1 t\Big] \qquad\qquad \varphi_2^e(t) = \frac{1}{\omega_2}\Big[\frac{\widetilde{\varepsilon}_{yy}(t)}{\varepsilon_{yy}} - 1 - \beta_2 t\Big]$$

The material constants γ_1, τ_1, μ_1, τ_1^K can be derived from the experimental curve $\varphi_1^e(t)$, whereas the constants γ_2, τ_2, μ_2, τ_2^K – from the experimental curve $\varphi_2^e(t)$. The optimisation problem is nonlinear and, in general, can only be solved numerically. The authors propose an algorithm of systematic searching, in the set of four decision variables in each optimisation problem, with the optimum conditions

$$H_i = \sum_{k=1}^{n} |\varphi_i(t_k) - \varphi_i^e(t_k)| = \min \qquad\qquad i = 1,2$$

where $\varphi_i(t)$, $i = 1,2$ denote the theoretical (approximate) creep functions, calculated from Eqs (5.4). The collocation instants $t_1, t_2, ..., t_n$ must be distributed uniformly over the logarithmic scale, within the time interval $< \log(0.1\ [\mathrm{min}]),\ \log t'' >$.

7. Separation of the shear and bulk strains

According to Eqs (2.2), (2.4), (5.1) one obtains

$$\widetilde{\varepsilon}_b = \frac{1}{3}(\widetilde{\varepsilon}_{xx} + 2\widetilde{\varepsilon}_{yy}) \qquad\qquad \sigma_b = \frac{1}{3}\sigma_{xx}$$
$$\widetilde{\varepsilon}_{xx} - \varepsilon_b = \frac{2}{3}(\widetilde{\varepsilon}_{xx} - \widetilde{\varepsilon}_{yy}) \qquad\qquad \widetilde{\varepsilon}_{yy} - \varepsilon_b = -\frac{1}{3}(\widetilde{\varepsilon}_{xx} - \widetilde{\varepsilon}_{yy})$$
$$\sigma_{xx} - \sigma_b = \frac{2}{3}\sigma_{xx} \qquad\qquad \sigma_{yy} - \sigma_b = -\frac{1}{3}\sigma_{xx}$$

i.e. the first three equations in matrix equation $(2.3)_1$ are proportional one to another, and the remaining three equations are trivial. The considerations can be then limited to the following equalities

$$\begin{aligned}
\sigma_s &= \frac{2}{3}\sigma_{xx} & \qquad \varepsilon_s &= \frac{2}{3}(\varepsilon_{xx} - \varepsilon_{yy}) & \qquad \widetilde{\varepsilon}_s(t) &= \frac{2}{3}[\widetilde{\varepsilon}_{xx}(t) - \widetilde{\varepsilon}_{yy}(t)] \\
\sigma_b &= \frac{1}{3}\sigma_{xx} & \qquad \varepsilon_b &= \frac{1}{3}(\varepsilon_{xx} + 2\varepsilon_{yy}) & \qquad \widetilde{\varepsilon}_b(t) &= \frac{1}{3}[\widetilde{\varepsilon}_{xx}(t) + 2\widetilde{\varepsilon}_{yy}(t)]
\end{aligned}$$

The symbols ε_s, ε_b denote respectively the shear and bulk elastic strains, whereas $\widetilde{\varepsilon}_s(t)$, $\widetilde{\varepsilon}_b(t)$ – the shear and bulk rheological strains.

Inserting Eqs (4.1) into Eqs (3.9), one obtains the following final formulae describing the shear and bulk time-dependent compliances

$$\begin{aligned}
\widetilde{J}_s(t) &= J_s\Big[1 + \int\limits_0^t \varPhi_s(t-\vartheta)\;d\vartheta + \beta_s\int\limits_0^t d\vartheta\Big] \\
\widetilde{J}_b(t) &= J_b\Big[1 + \int\limits_0^t \varPhi_b(t-\vartheta)\;d\vartheta + \beta_b\int\limits_0^t d\vartheta\Big]
\end{aligned}$$

where

$$J_s = J_1 - J_2 \qquad\qquad J_b = J_1 + 2J_2$$

$$\begin{aligned}
\varPhi_s(t) = {} & \frac{J_1\omega_1}{J_1 - J_2}[\gamma_1\varPhi_1(t) + (1-\gamma_1)F_1(t)] + \\
& -\frac{J_2\omega_2}{J_1 - J_2}[\gamma_2\varPhi_2(t) + (1-\gamma_2)F_2(t)]
\end{aligned}$$

$$\begin{aligned}
\varPhi_b(t) = {} & \frac{J_1\omega_1}{J_1 + 2J_2}[\gamma_1\varPhi_1(t) + (1-\gamma_1)F_1(t)] + \\
& +\frac{2J_2\omega_2}{J_1 + 2J_2}[\gamma_2\varPhi_2(t) + (1-\gamma_2)F_2(t)]
\end{aligned}$$

$$\beta_s = \frac{J_1\beta_1 - J_2\beta_2}{J_1 - J_2} \qquad\qquad \beta_b = \frac{J_1\beta_1 + 2J_2\beta_2}{J_1 + 2J_2}$$

The generating functions $\varPhi_s(t)$, $\varPhi_b(t)$ are related to the shear and bulk strains of the bielement W-K, while β_s, β_b are the shear and bulk viscosity coefficients, corresponding to the Newton element in the H-W-K-N material model.

8. An example of the identification of the material constants

The experiments have been carried out on three cylindrical samples, made of the POLIMAL 109 polyester resin, prepared in accordance with the standard procedures. The computer aided experiments have been realised in Laboratory of Strength of Materials, Military University of Technology, Warsaw[1]. The samples were under normal stress $\sigma_{xx} = 0.3R_m = 10.5\,\mathrm{MPa}$, where R_m is the material strength in tension. The experiments have been conducted on the DST 5000 creep-testing machine, in constant temperature of 18°C, using 10 mm long bonded wire strain gauges. The time histories of the longitudinal and transverse strains were measured in the discrete set of time instants from the interval $t \in < 0, 60000\mathrm{min} >$. The static stress level was reached linearly during 1 minute, and from that moment the strains were registered.

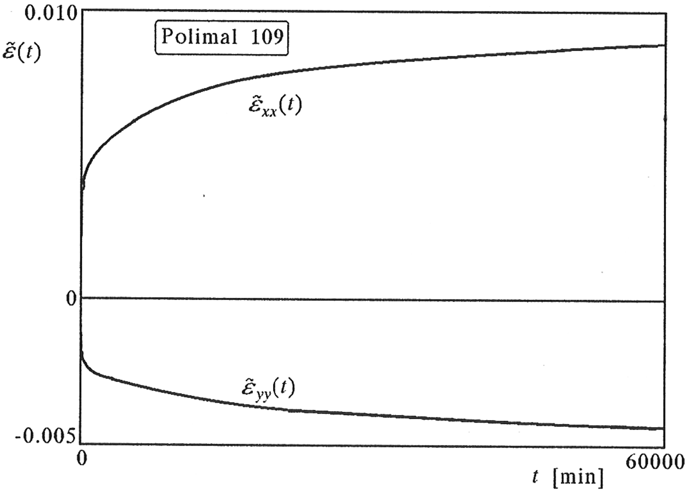

Fig. 3. Experimental time histories of longitudinal and transverse strains $\varepsilon_{xx}(t)$, $\varepsilon_{yy}(t)$ for the tensile sample made of the POLIMAL 109 polyester resin

The results of the experiments and fitting the H-W-K-N model to the real data are presented in Fig.3 ÷ Fig.5, for the selected sample. The experimental, longitudinal and transverse strains versus time are plotted in Fig.3. Fig.4a,b show the viscoelastic creep curves, both the experimental and theoretical, respectively for the longitudinal and transverse strains. Fig.5 positively verifies the H-W-K-N model, as the shear and bulk strain curves, both the

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[1] Dr. Roman Gieleta, from Military University of Technology, Warsaw, had made the samples and performed the experiments.

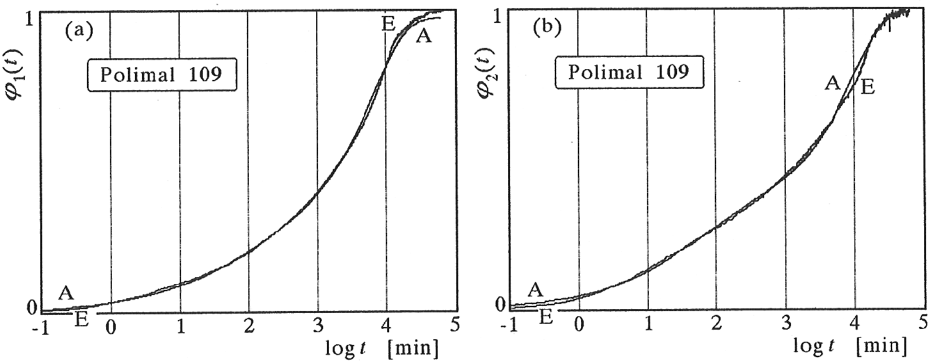

Fig. 4. Creep function φ_1 and φ_2 for the sample made of the POLIMAL 109 polyester resin, in a semi-logarithmic scale; E –experiment, A – approximation with the H-W-K-N model

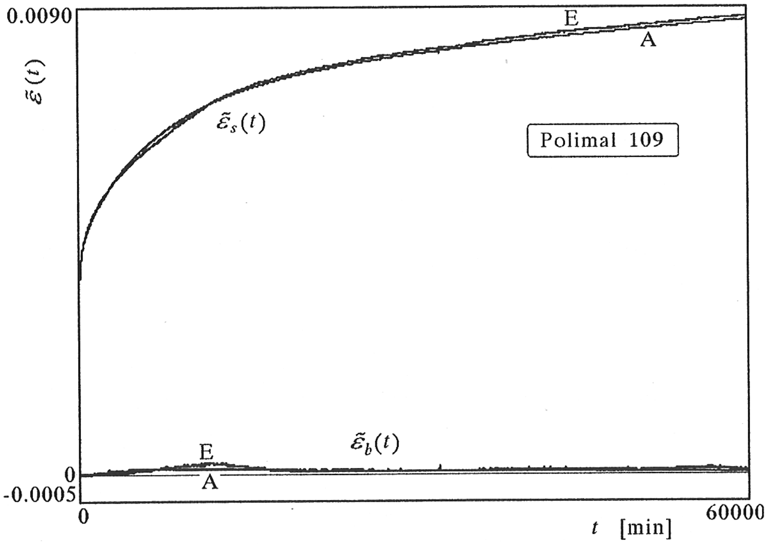

Fig. 5. Shear ε_s and bulk ε_b creep for the tensile sample made of the POLIMAL 109 polyester resin, in a semi-logarithmic scale; E – experiment, A – approximation with the H-W-K-N model

experimental and theoretical ones, are very closed. The creep of the remaining samples and their approximation were of similar character and the same level of accuracy.

The identification algorithm was programmed on a PC. In each optimisation problem only one minimum was detected, and low sensitivity of the optimum conditions was found. Systematic searching with steps $\Delta\gamma_i = 0.01$, $\Delta\mu_i = 0.01$, $\Delta\tau_i = 0.1\,\mathrm{h}$, $\Delta\tau_i^K = 1\,\mathrm{h}$, $i = 1,2$ yielded the functions $\varphi_1^e(t)$, $\varphi_2^e(t)$ with extremely high accuracy. The relative error measuring the deviation of the theoretical curve from the experimental one was 2.0% for $\varphi_1(t)$ and 2.5% for $\varphi_2(t)$, provided that 23 collocation points were used.

The results of the identification of the material constants, based on the selected sample, are collected below:

$E = 3.58$ GPa $\qquad \nu = 0.423$

$\gamma_1 = 0.43 \qquad \omega_1 = 1.62 \qquad \tau_1 = 4.1$ h

$\mu_1 = 0.48 \qquad \tau_1^K = 115$ h

$\gamma_2 = 0.45 \qquad \omega_2 = 2.08 \qquad \tau_2 = 1.3$ h

$\mu_2 = 0.55 \qquad \tau_2^K = 158$ h

$\tau_1^N = 2270$ h $\qquad \tau_2^N = 2375$ h

9. Conclusions

The authors formulated a new rheological model of polymeric materials, composed of the Hooke, Wilczyński, Kelvin and Newton elements. The Wilczyński element is described by fractional exponent generating functions, whereas the remaining elements are understood classically, i.e. the Kelvin and Newton elements are expressed by ordinary exponent generating functions.

The H-W-K-N model, proposed in the study, is governed by the consitutive equations formulated in both the uncoupled and coupled form. The generating functions have been formulated analytically for the directional compliances. The generating functions for the shear and bulk compliances are then derived by symbolic transforms of the directional functions.

The introductory experiments show that the H-W-K-N model of polymers predicts approximate time-dependent strains closely to the experimental values. The computer algorithm for the identification of the material constants, based on monotone and easy-measurable directional strains, appeared to be very stable.

The main advantages of the material model proposed in the study are as follows:

- very good approximation of realistic strains of polymeric materials, within the considered time interval
- possibility of determining, in an analytical way, Laplace's transforms of the generating functions.

The last advantage enables application of the H-W-K-N model to predict rheological properties of fibrous polymeric composites, Klasztorny, Wilczyński, Witemberg-Perzyk (2000).

A. Appendix – description of functions $\Phi(t)$, $\Lambda(\xi)$, $\varphi(t)$ related to the fractional exponent function

Each component function in the pair $\Phi_1(t)$, $\Phi_2(t)$ is formulated in the same form. The pairs $\Lambda_1(\xi)$, $\Lambda_2(\xi)$, and $\varphi_1(t)$, $\varphi_2(t)$ have analogous character. Hence, when analysing these functions one can neglect the subscripts.

The functions of the load history, retardation time distribution and creep are described by the following formulae (see Wilczyński, 1978, 1996; Wilczyński and Klasztorny, 2000)

$$\Phi(t) = \alpha \int\limits_0^\infty \mathrm{e}^{-\alpha\xi t} \xi \Lambda(\xi)\, d\xi \qquad \alpha = \frac{1}{\tau}$$

$$\Lambda(\xi) = \frac{\sin \pi\mu}{\pi} \frac{\xi^{\mu-1}}{1 + 2\xi^{\mu} \cos \pi\mu + \xi^{2\mu}} \qquad 0 < \mu < 1$$

$$\varphi(t) = 1 - \int\limits_0^\infty \mathrm{e}^{-\alpha\xi t} \Lambda(\xi)\, d\xi$$

where τ is the retardation time describing the comparable Kelvin model.

These functions satisfy the following conditions

$$
\begin{array}{llll}
\Phi(t) > 0 & \text{for} \quad t > 0 & \lim\limits_{t\to 0} \Phi(t) = \infty & \lim\limits_{t\to\infty} \Phi(t) = 0 \\
\Lambda(\xi) \geqslant 0 & \text{for} \quad \xi \geqslant 0 & \displaystyle\int\limits_0^\infty \Lambda(\xi)\, d\xi = 1 & \lim\limits_{\xi\to 0} \Lambda(\xi) = \infty \\
\varphi(0) = 0 & \varphi(\infty) = \lim\limits_{t\to\infty} \varphi(t) = 1 & &
\end{array}
\tag{A.1}
$$

Formulae (A.1) give theoretically an infinite value of the initial creep velocity, i.e.

$$
\dot{\varphi}(t) = \frac{d\varphi}{dt} = \alpha \int\limits_0^\infty \mathrm{e}^{-\alpha\xi t} \xi \Lambda(\xi)\, d\xi = \Phi(t)
$$

$$
\dot{\varphi}(0) = \lim_{t\to 0} \Phi(t) = \infty
$$

On the other hand, the velocity $\dot{\varphi}(t)$ drops rapidly to a small value after passing the singular point $t = 0$.

The function $\Lambda(\xi) = \Lambda(\xi, \mu)$ only depends on the constant μ. After introducing the dimensionless parameter $\eta = \alpha t = t/\tau$, related to time, one can analyse the normalised load history function

$$
\Phi_u(\eta, \mu) = \int\limits_0^\infty \mathrm{e}^{-\xi\eta} \xi \Lambda(\xi, \mu)\, d\xi \tag{A.2}
$$

and the creep function

$$
\varphi(\eta, \mu) = 1 - \int\limits_0^\infty \mathrm{e}^{-\xi\eta} \Lambda(\xi, \mu)\, d\xi \tag{A.3}
$$

with

$$
\Phi(t) = \frac{1}{\tau} \Phi_u(\eta, \mu) \qquad \varphi(t) = \varphi(\eta, \mu) \qquad t = \eta\tau
$$

Assuming $\mu = 1$, one obtains the comparable Kelvin model, for which

$$
\Lambda(\xi, 1) = \delta(\xi - 1) \qquad \Phi_u(\eta, 1) = \mathrm{e}^{-\eta} \qquad \varphi(\eta, 1) = 1 - \mathrm{e}^{-\eta}
$$

where δ is Dirac's delta function.

Diagrams of the functions $\Lambda(\xi, \mu)$, $\Phi_u(\eta, \mu)$, $\varphi(\eta, \mu)$ for the selected values of the fraction μ are presented in Fig.6 ÷ Fig.8.

Integrals (A.2), (A.3) were calculated using the Gauss quadratures. The interval $\xi \in < 0, \infty)$ was divided into appropriate subintervals. The singularity in the point $\xi = 0$ has been eliminated classically.

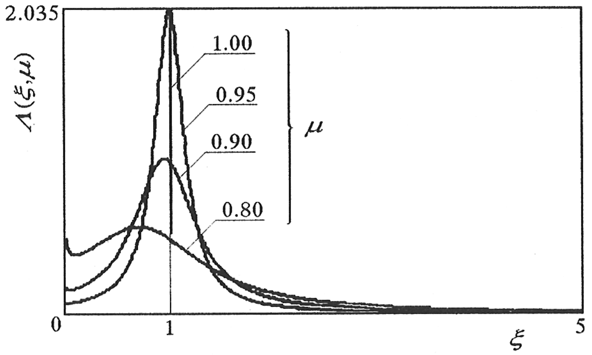

Fig. 6. Exemplary diagrams of function $\Lambda(\xi,\mu)$

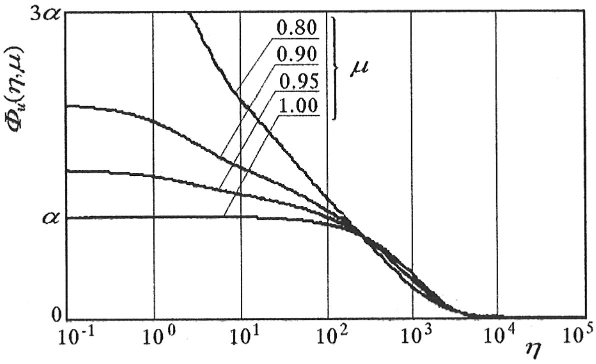

Fig. 7. Exemplary diagrams of function $\Phi_u(\eta,\mu)$

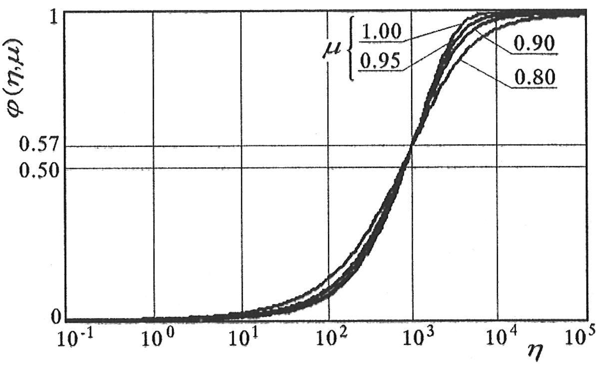

Fig. 8. Exemplary diagrams of function $\varphi(\eta,\mu)$

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Model reologiczny materiałów polimerowych oraz identyfikacja jego parametrów

Streszczenie

Zaproponowano nowy model reologiczny materiałów polimerowych, oznaczony symbolem H-W-K-N. Model mechaniczny materiału składa się z czterech elementów połączonych szeregowo, sformułowanych przez Hooke'a, Wilczyńskiego, Kelvina i Newtona. Sformułowano równania konstytutywne reologii opisujące model H-W-K-N, w postaci niesprzężonej i sprzężonej, z funkcjami tworzącymi dobrze aproksymującymi eksperymentalne przebiegi pełzania. Opracowano metodę identyfikacji 14 stałych materiałowych, opisujących model H-W-K-N. Algorytm identyfikacji zaprogramowano w języku PASCAL. Podano wyniki identyfikacji stałych materiałowych żywicy poliestrowej (POLIMAL 109), bazujące na pomiarach pełzania wzdłużnego i poprzecznego próbek przy rozciąganiu jednokierunkowym.

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