ELSEVIER

Mechanics of Materials 34 (2002) 795–808

MECHANICS
OF
MATERIALS

www.elsevier.com/locate/mechmat

Endochronic viscoplastic material models for filled PTFE

Thomas Kletschkowski [a,*], Uwe Schomburg [a], Albrecht Bertram [b]

[a] *Engineering Mechanics, Universität der Bundeswehr Hamburg, Holstenhofweg 85, D-22043 Hamburg, Germany*
[b] *Institut für Mechanik, Otto-von-Guericke-Universität Magdeburg, Universitätsplatz 2, D-39106 Magdeburg, Germany*

Received 18 June 2002

Abstract

To describe the nonlinear material behaviour of polytetrafluoroethylene (PTFE), a viscoplastic material model of the overstress type is proposed based on experimental data. The approach starts with a rheological model without an elastic range, using a rate-independent elastoplastic model with an endochronic flow rule and a nonlinear elastic law in parallel connection with a nonlinear Maxwell model. The generalization to three dimensions is possible for both small and finite strains under the assumption that changes of the density are purely elastic. Numerical simulations show that the model can describe the short and long time material behavior of filled PTFE.

Keywords: Viscoplasticity; Endochronic theory of plasticity; PTFE compound

1. Introduction

Due to their tribological characteristics, chemical inertness and temperature stability, polytetrafluoroethylene (PTFE) compounds are increasingly used for rotary shaft seals in engineering. In contrast to classical rotary shaft seals, which are made of elastomeric materials, the engineer has to take into account the inelastic material behaviour of PTFE for the design process.

In order to prepare a numerical simulation of the sealing system, uniaxial experiments have been carried out to study the material behaviour of a typical PTFE compound that consists of 90% PTFE, 5% short cylindrical glass fibres and 5% MoS_2. The glass fibres are introduced to reduce the wear of the seal. The MoS_2 is used to decrease the friction between the seal and the shaft in the contact zone. The material behaviour has been studied in uniaxial relaxation tests, uniaxial tension tests with different strain rates, and retardation tests after unloading (see Pohl, 1999; Kletschkowski et al., 2000). The tension and relaxation tests have been performed with respect to the mechanical straining of the PTFE seal during the mounting and service life on the shaft of the engine. The hoop strain of the seal during the mounting results in a pressure between the seal and the shaft, which is required to prevent leakage. This pressure is caused by the inner hoop tension in the sealing element.

In the past, engineers used different constitutive laws for the numerical simulation of different

---

[*] Corresponding author. Tel.: +49-40-6541-2282; fax: +49-40-6541-2822.

E-mail address: kletsch@unibw-hamburg.de (T. Kletschkowski).

0167-6636/02/$ - see front matter © 2002 Elsevier Science Ltd. All rights reserved.
PII: S0167-6636(02)00197-7

successive processes. The mounting (short time process) of the seal on the shaft was usually calculated using an elastoplastic (Sui, 1998; Pohl, 1999), or a viscoplastic approach with a strain rate-dependent yield stress (Pohl, 1999). To compute the stress relaxation curves after loading, Olbrich et al. (2000) used experimental data as initial values instead of the calculated data from the loading curve. Moreover the numerical results of Sui (1998) and Pohl (1999) show that material models with linear or quasi-linear elastic ranges are not adequate to describe the nonlinearities of PTFE during unloading. Furthermore all these models were unable to separate the inelastic effects by the decoupling of the total stress response of the material into a rate-dependent overstress and a rate-independent equilibrium stress.

An additive decomposition of the total stress into a rate-independent equilibrium part and a rate-dependent overstress can be found in the calculations of Wüstenhagen (1995) and Lin (1999). Wüstenhagen (1995) used the linear theorie of viscoelastic solids in the form of generalized Maxwell models. Hence his approach is limited to very small strains. The more sophisticated theory of finite linear viscoelasticity in combination with a nonlinear elastic law for the equilibrium stress, used by Lin (1999), allows to regard physical and geometrical nonlinearities during the computation, but the rate-independent plastic deformation, which can be clearly observed during the tests, is completely ignored by the use of a purely viscoelastic approach.

In order to simulate PTFE shaft seals in a unified approach, this paper presents viscoplastic material models for PTFE based on the results of uniaxial experiments at ambient temperature. Such material behaviour can be described by a rheological model consisting of a Maxwell model and an endochronic elastoplastic model in parallel. In contrast to other models of overstress type (see Reese, 1998) and due to the continuous evolution of the plastic deformation during the tests, an endochronic approach for the rate-independent equilibrium stress is used. An elastic range limited by a yield stress is not needed in our model. In the present paper a rheological model is proposed and its three-dimensional generalization is motivated for both small and finite strains. Numerical simulations show that the proposed model is capable to describe the nonlinear material behaviour of the analyzed PTFE compound.

2. Uniaxial experiments

In order to characterize the material behaviour of the investigated PTFE compound, uniaxial tension tests with different strain rates, relaxation tests at different strain levels, and retardation tests after unloading have been performed at ambient temperature.

2.1. Quasi-static loading and succeeding stress relaxation

The quasi-static tensile tests and the relaxation tests have been carried out by Pohl (1999) on a tensile test system. Plain tensile test specimens with a relevant working range of 10 mm in the undeformed state have been prepared for the strain controlled tests using engineering stress and engineering strain as measurable quantities. In every test the specimen have been initially loaded with strain rates of 1%, 10% or 100% min^{-1} until the top strain of 8% was reached. Then the succeeding stress relaxation has been measured over 10 h.

After this time period the stress is reduced to nearly 50% of its initial value at the end of the loading process. This is equivalent to the time dependent reduction of the pressure between a PTFE shaft seal and the shaft in the contact zone that is needed to prevent sharp wear of the seal during the engine run in. The measurement of the quasi-static loading curves during the investigations on the plastic memory effect of PTFE compounds (see Kletschkowski et al., 2000) have been used to test the repeatability of Pohl's experiments. The strain rate-dependence of the analyzed compound is illustrated in Fig. 1. Fig. 2 shows the stress relaxation after loading with a strain rate of 10% min^{-1} at 3%, 5% and 8% engineering strain. The results of this test show that

- the stress response in isothermal uniaxial tension tests shows a nonlinear stress–strain behav-

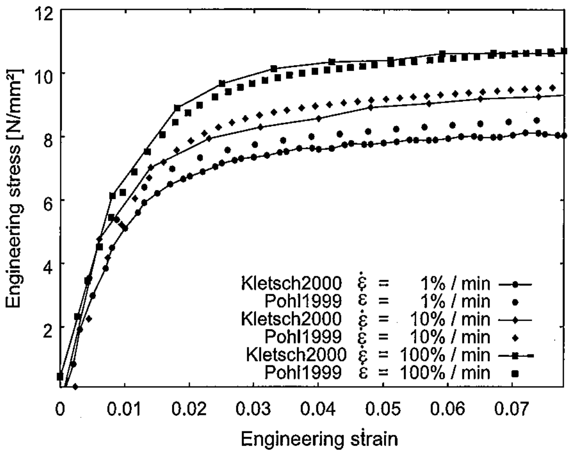

Fig. 1. Strain rate-dependence (Kletschkowski, Pohl).

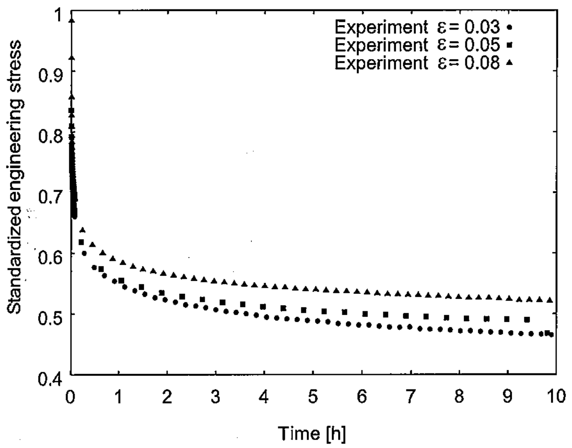

Fig. 2. Stress relaxation.

iour, which depends strongly on the strain rate. An increase in strain rate of one decade results in a stress increase of nearly 14% at the end of the test.

- the relaxation curves, showing the time dependent decrease of the measured stress, are not congruent when using a normalization based on the respective maximum value of the performed strain level.

2.2. Strain recovery and plastic deformation

Fig. 3 illustrates the effects of strain recovery and plastic deformation of an unloaded tension test specimen. In Fig. 3 the engineering strain is plotted over the time for a test piece, which has been first strained with a strain rate of 10% min^{-1} to a total engineering strain of 10% at ambient temperature. After reaching 10% engineering strain, the strain was kept constant over 30 min, and a stress relaxation process occurred in the test pieces. The next step was to switch from the strain to a force controlled test by unloading the test pieces quite quickly with a rate of 0.5 N/s. Then a temperature cycle to accelerate the process of viscous strain recovery has been performed. Therefore the temperature was increased in 15 min from room temperature up to 100 °C, and cooled down in the same time. The result of this unloading process was an inelastically deformed test piece. The instantaneous inelastic deformation after unloading, documented in Fig. 3, is nearly 50% of the total technical strain and a sum of plastic and viscous parts of the deformation. The final permanent inelastic deformation is independent of the strain rate of the loading phase, as reported in Kletschkowski et al. (2000), and therefore called plastic. The uniaxial experiments show that the analysed PTFE compound is a viscoplastic material without a purely elastic domain.

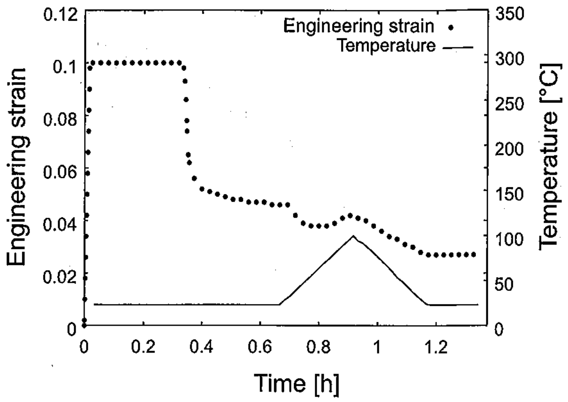

Fig. 3. Strain recovery and plastic deformation.

3. A viscoplastic rheological model for PTFE

In order to simulate inelastic material behaviour of polymers via the example of the investigated

PTFE compound, a small strain viscoplastic theory based on overstress motivated by the rheological model shown in Fig. 4 is used. In contrast to previous models and with respect to the experimental data that indicate a continuous growth of plastic deformations during tension tests without crossing a significant flow limit, the endochronic theory of plasticity is applied.

Morphological changes in polymers under load, like the breakup of crystalline structures and their recrystallization, depend on the existence of a nonzero state of stress, but in contrast to metallic materials the state of stress does not need to reach a critical limit to activate morphological changes. The endochronic theory of plasticity was developed by Valanis (1971) and is discussed in detail in Haupt (1977) and Krawietz (1986). In the case of linear elasticity and the absence of hardening effects, such material behaviour can be described by a differential equation of the saturation-type (see Sedlan and Haupt, 1999). Its integration leads to the formula shown in Fig. 5, which describes the elastoplastic endochronic material behaviour during tension tests. E is the Young's modulus, ε the total strain, and Y defines the maximum stress that can be reached during an elastoplastic deformation process.

A general concept of modeling viscoplastic material behavior with elastic domains can be found in Haupt (2000).

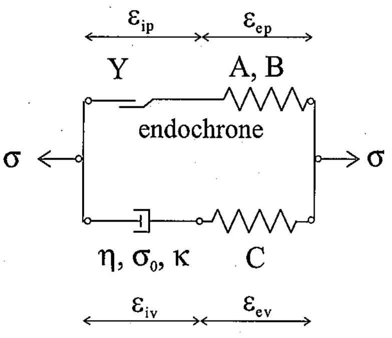

Fig. 4. Rheological model.

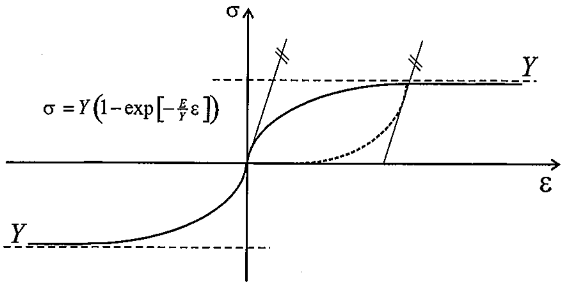

Fig. 5. Endochronic plasticity—stress–strain behaviour.

In the present paper, and with respect to the nonlinear material characteristics of the analysed PTFE compound, the development of the constitutive equations starts with the additive decomposition of the total strain into elastic and inelastic parts.

$$\varepsilon = \varepsilon_{ep} + \varepsilon_{ip} = \varepsilon_{ev} + \varepsilon_{iv}, \tag{1}$$

$$\sigma = \sigma_\infty + \sigma_{ov}, \tag{2}$$

$$\sigma_\infty = A\ln(1 + B\langle\varepsilon_{ep}\rangle) - A\ln(1 - B\langle-\varepsilon_{ep}\rangle) \quad \text{with } \langle x\rangle := \frac{x+|x|}{2}, \tag{3}$$

$$\sigma_{ov} = C\varepsilon_{ev}, \tag{4}$$

$$\dot{\varepsilon}_{ip} = \frac{1}{Y}\sigma_\infty|\dot{\varepsilon}|, \tag{5}$$

$$\dot{\varepsilon}_{iv} = \frac{1}{\eta}\left[\sinh\left(\frac{\sigma_{ov}}{\sigma_0}\right)\right]^\kappa. \tag{6}$$

The stress response is the sum of the equilibrium stress (Eq. (3)) and the overstress (Eq. (4)). Hence the proposed model separates the rate-dependent and rate-independent effects on the stress. The rate of the plastic deformation is given by the endochronic flow rule (Eq. (5)). The development of the plastic deformation is forced only by the equilibrium stress. The viscous deformation is determined by a nonlinear rate equation of the Garofalo type (Eq. (6)) caused by the overstress. For very small deformations Eq. (3) reduces to Hooke's law. The proposed model is geometrically linear, but physically nonlinear and describes a rate-dependent material behaviour with equilibrium hysteresis at ambient temperature using seven material parameters, summarized in Table 1.

Numerical simulations and experimental data are shown in Figs. 5–8. The model parameters

Table 1
Parameters of the rheological model for filled PTFE

A (N/mm^2)	Y (N/mm^2)	η (1/s)	κ	B	C (N/mm^2)	σ_0 (N/mm^2)
1.6	10.5	9.0×10^5	2.4	440.0	304.0	1.3

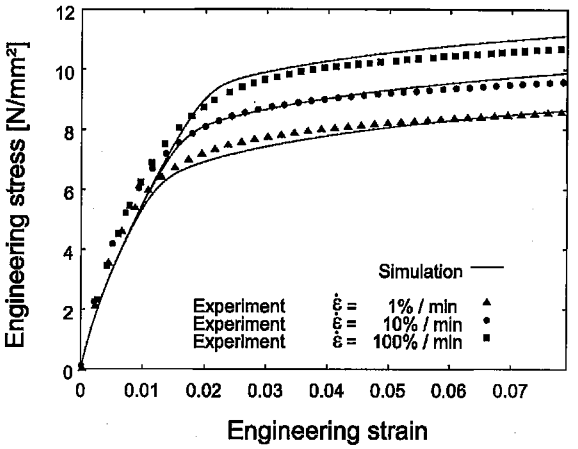

Fig. 6. Strain controlled simulations—strain rate-dependence.

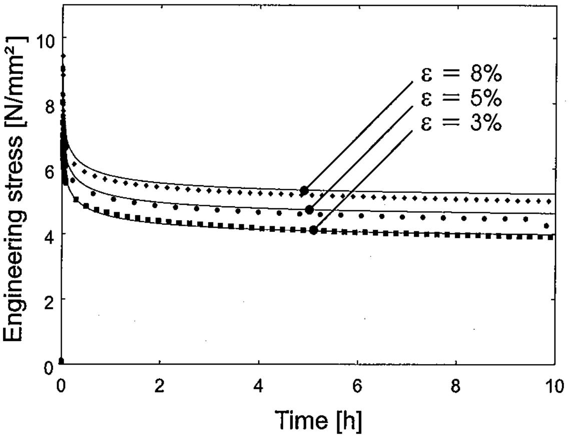

Fig. 7. Strain controlled simulations—stress relaxation.

have been determined by an indirect identification process using the loading curve of a tension test with a strain rate of 10% min^{-1} up to an engineering strain of 8% and the succeeding stress relaxation. With respect to Fig. 3, the amount of the plastic deformation was expected to be 2.4%. In Figs. 6 and 7 the correlation between experimental data from strain controlled tests and numerical simulations with the rheological model are shown.

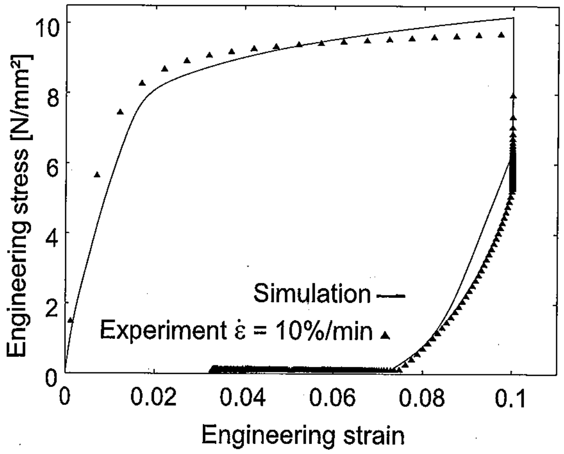

Fig. 8. Stress controlled simulations and experiments—strain recovery.

Fig. 6 illustrates that the proposed model is suitable to compute the strain rate-dependence during loading with different strain rates. The model is also very precise in the calculation of the long-time material behaviour during the stress relaxation processes (see Fig. 7).

For the verification of the rheological model the creep experiments performed by Oppermann et al. (1995) and the strain recovery experiments reported in Kletschkowski et al. (2002) have been used. The correlation between these stress controlled experiments (see Figs. 8 and 9) and the numerical simulations using the same material parameters is of good quality, too.

The results of the previous section can be summarized in the form of the DEICE-scheme, which contains the general steps of material modeling.

- *Delineate general characteristics:* The analysed PTFE compound shows a viscoplastic material behaviour in uniaxial experiments at ambient temperature.

Table 7
Constitutive equations at finite strains

Isochoric part of the deformation	$\overline{\mathbf{C}} = (\det \mathbf{C})^{-1/3}\mathbf{C}$
Decomposition of stress	$\mathbf{S} = S_{\mathrm{vol}}\mathbf{C}^{-1} + \mathbf{S}_{\infty}^{\mathrm{DEV}} + \mathbf{S}_{\mathrm{ov}}^{\mathrm{DEV}}$
Elastic law for the volumetric stress contribution	$S_{\mathrm{vol}} = K \ln\{\sqrt{\det \mathbf{C}}\}$
Overstress in the initial elastic range of the Maxwell model	$h_{\mathrm{V0}}(\overline{\mathbf{C}}_{\mathrm{V}}) = G_{\mathrm{ov}}(\overline{\mathbf{C}}_{\mathrm{V}} - \mathbf{I})$ with $\overline{\mathbf{C}}_{\mathrm{V}} = \mathbf{P}_{\mathrm{V}}^{\mathrm{T}}\overline{\mathbf{C}}\mathbf{P}_{\mathrm{V}}$
Isomorphic condition for the overstress	$\mathbf{S}_{\mathrm{ov}}^{\mathrm{DEV}} = (\mathbf{P}_{\mathrm{V}} h_{\mathrm{V0}}(\overline{\mathbf{C}}_{\mathrm{V}})\mathbf{P}_{\mathrm{V}}^{\mathrm{T}})^{\mathrm{DEV}}$
Equilibrium stress in the initial elastic range of the endochronic model	$h_{\mathrm{P0}}(\overline{\mathbf{C}}_{\mathrm{P}}) = G_{\infty} \sum_{i=1}^{3} \{\ln(1 + \widehat{B}\langle\lambda_i\rangle) - \ln(1 - \widehat{B}\langle-\lambda_i\rangle)\}\mathbf{n}_i \otimes \mathbf{n}_i$ with $(\lambda_i, \mathbf{n}_i) = \mathrm{eigensystem}\{\overline{\mathbf{C}}_{\mathrm{P}} - \mathbf{I}\}$ and $\overline{\mathbf{C}}_{\mathrm{P}} = \mathbf{P}_{\mathrm{P}}^{\mathrm{T}}\overline{\mathbf{C}}P_{\mathrm{P}}$
Isomorphic condition for the equilibrium stress	$\mathbf{S}_{\infty}^{\mathrm{DEV}} = (\mathbf{P}_{\mathrm{P}} h_{\mathrm{P0}}(\overline{\mathbf{C}}_{\mathrm{P}})\mathbf{P}_{\mathrm{P}}^{\mathrm{T}})^{\mathrm{DEV}}$
Evolution equation for the viscous transformation	$\dot{\mathbf{P}}_{\mathrm{V}}\mathbf{P}_{\mathrm{V}}^{-1} = -(3/2\eta)[\sinh(\sigma_{\mathrm{VV}}/\sigma_0)]^{\kappa}((\mathbf{S}_{\mathrm{ov}}^{\mathrm{DEV}}\mathbf{C})/\sigma_{\mathrm{VV}})$
Equivalent material overstress	$\sigma_{\mathrm{VV}} = \sigma_{\mathrm{VV}}(\mathbf{S}_{\mathrm{ov}}^{\mathrm{DEV}}\mathbf{C}) := \sqrt{(3/2)\mathrm{tr}\{(\mathbf{S}_{\mathrm{ov}}^{\mathrm{DEV}}\mathbf{C})(\mathbf{S}_{\mathrm{ov}}^{\mathrm{DEV}}\mathbf{C})\}}$
Evolution equation for the plastic transformation	$\dot{\mathbf{P}}_{\mathrm{P}}\mathbf{P}_{\mathrm{P}}^{-1} = -(3/2Y)\sigma_{\mathrm{VP}}\dot{\varepsilon}_{\mathrm{V}}((\mathbf{S}_{\infty}^{\mathrm{DEV}}\mathbf{C}))/\sigma_{\mathrm{VP}})$
Equivalent material equilibrium stress	$\sigma_{\mathrm{VP}} = \sigma_{\mathrm{VP}}(\mathbf{S}_{\infty}^{\mathrm{DEV}}\mathbf{C}) := \sqrt{(3/2)\mathrm{tr}\{(\mathbf{S}_{\infty}^{\mathrm{DEV}}\mathbf{C})(\mathbf{S}_{\infty}^{\mathrm{DEV}}\mathbf{C})\}}$
Equivalent strain rate	$\dot{\varepsilon}_{\mathrm{V}} := \sqrt{(2/3)\mathrm{tr}\{\mathbf{D}^{\mathrm{dev}}\mathbf{D}^{\mathrm{dev}}\}}$

elastic and inelastic parts is not needed. The inelastic spin is included in this concept.

The set of constitutive equations for the description of the viscoplastic material behaviour of the analysed PTFE compound at finite strains and at ambient temperature is summarized in Table 7.

Fig. 10 illustrates the capability of the model to describe the inelastic effects, observed during the tests. It presents the measured data and calculated loading curve of a tension test with a strain rate of $\dot{\varepsilon} = 10\%\ \mathrm{min}^{-1}$ and the succeeding stress relaxation.

The capability of the model to describe finite deformations of viscoplastic materials is tested with material parameters identified for the PTFE compound. Therefore the stress response of the material model has been calculated due to simple shear.

During loading the deformation gradient

$$\mathbf{F} = \mathbf{I} + \tan(\dot{\delta}t)\mathbf{e}_1 \otimes \mathbf{e}_2 \quad \text{with } \dot{\delta} = \mathrm{const} \tag{52}$$

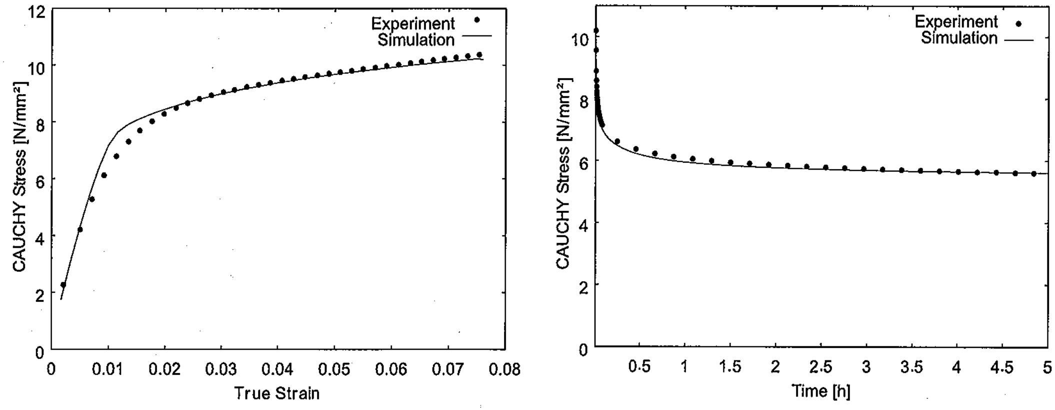

Fig. 10. Loading and stress relaxation (parameters: $K = 3020.8$ N/mm², $G_{\infty} = 0.86$ N/mm², $G_{\mathrm{ov}} = 171.2$ N/mm², $\widehat{B} = 380.0$, $\eta = 9 \times 10^5$ 1/s, $\kappa = 2.2$, $\sigma_0 = 1.2$ N/mm², $Y = 9.0$ N/mm²).

has been used. For the calculation of the succeeding stress relaxation after loading, the deformation has been kept constant. Fig. 11 illustrates the capability of the model to describe the rate-dependent effects during loading. An increase of the loading rate $\dot{\delta}$ of one decade results in an increase of the true shear stress T_{12} that is lower than one decade in the end of the test.

Fig. 12 presents the calculated loading curves and the curves of the succeeding stress relaxation processes after loading with a constant loading rate of $\dot{\delta} = 1/600\ \mathrm{s}^{-1}$.

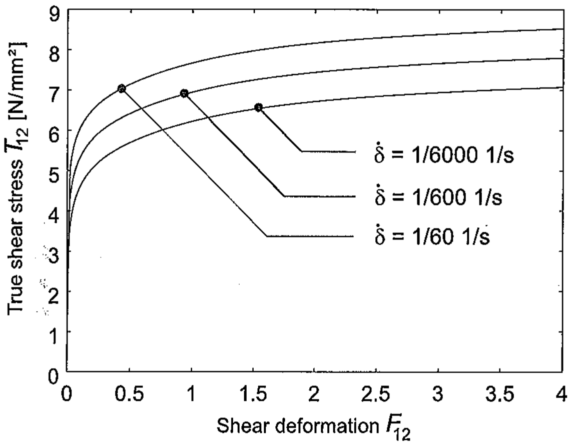

Fig. 11. Rate-dependent loading.

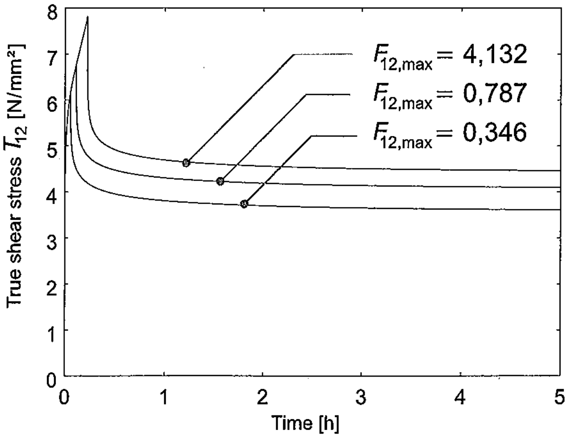

Fig. 12. Stress relaxation.

6. Summary

This paper presents an endochronic viscoplastic material model for the description of the inelastic material behaviour of a typical PTFE compound for isothermal deformation processes at ambient temperature at both small and finite strains. The model is based on the results of uniaxial experiments. An additive decomposition of the total stress into a rate-independent equilibrium stress and a rate-dependent overstress is applied. The three-dimensional constitutive equations are obtained by a generalization of a rheological model, which consists of an endochronic elastoplastic model coupled with a nonlinear Maxwell model in parallel. The numerical simulations show the capability of the model to describe the inelastic phenomena observed in the tests. The description of the inelastic material behaviour of the investigated PTFE compound at finite strains is successfully using an approach to finite inelasticity based on material isomorphisms.

References

Bertram, A., 1998. An alternative approach to finite plasticity based on material isomorphisms. Int. J. Plast. 52, 353–374.

Bertram, A., 1999. The dissipation-inequality of rate-independent thermo-plasticity. ZAMM (Zeitschrift Angew. Math. Mech.) 79, 195–198.

Haupt, P., 1977. Viskoelastizität und Plastizität. Springer, Berlin.

Haupt, P., 2000. Continuum Mechanics and Theory of Materials. Springer, Berlin.

Holzapfel, G.A., 2000. Nonlinear Solid Mechanics A Continuum Approach for Engineering. John Wiley and Sons, Ltd.

Kletschkowski, T., Schomburg, U., Bertram, A., 2002. An endochronic viscoplastic material model for filled PTFE. In: Khan, A., Pamies, O.L. (Eds.), Plasticity Damage and Fracture at Micro- and Nanoscales, Fulton, USA, pp. 135–137.

Kletschkowski, T., Subramanian, S.P., Schomburg, U., 2000. Experimental investigation on the plastic memory effect of PTFE compounds. J. Mater. Process. Manufact. Sci. 9.

Kletschkowski, T., 1998. Numerische Untersuchnung des Verfestigungsverhaltens bei grossen plastischen Verformungen. Diplomarbeit Technische Universität, Berlin.

Kraska, M., 1998. Textursimulation bei grossen inelastischen Verformungen mit der RVE-Technik, Dissertation, Technische Universität, Berlin.

Krawietz, A., 1986. Materialtheorie Mathematische Beschreibung des phänomenologischen thermomechanischen Verhaltens. Springer, Berlin.

Lin, R., 1999. A new thermoviscoelastic constitutive model in finite strain domain. In: Wang, C.M. et al. (Eds.), Computational, Mechanics for the Next Millennium, Proceedings of APCOM' 99, Singapore. Elsevier Science, pp. 911–916.

Olbrich, M., Reichert, M., Haas, W., 2000. Ein Überspannungsmodell zur Beschreibung des mechanischen Verhaltens des Dichtungswerkstoffs Polytetrafluorethylene. III. Hamburger Dichtungstechnisches Kolloquium, TU Hamburg-Harburg.

Oppermann, A., Sui, H., Pohl, H., Schomburg, U., Upper, G., Hatch, F., 1995. Material and Computational Analysis of PTFE Shaft Seals. SAE Technical Paper Series 951055, Society of Automotive Engineers, Warrendale, PA, USA.

Pohl, H., 1999. Computergestützte und experimentelle Untersuchungen von Manschettendichtungen aus glasfaserverstärktem PTFE-Compound, Dissertation, Universität der Bundeswehr Hamburg.

Reese, S., 1998. Multiplicative thermo-viscoplasticity: a thermodynamic model and its finite element implementation. Techn. Mechanik 18, 209–216.

Sedlan, K., Haupt, P., 1999. Nichtlineare Viskoelastizität von gummiartigen Werkstoffen: Experimentelle Untersuchung, Modellierung und Identifikation. Techn. Mechanik 19.

Sui, H., 1998. Beitrag zur Simulation von PTFE-Wellendichtmanschetten in Verbrennungsmotoren, Dissertation, Universität der Bundeswehr Hamburg.

Valanis, K., 1971. A theory of viscoplasticity without a yield surface, Part I: general theory. Arch. Mech. 23.

Wüstenhagen, G., 1995. Beitrag zur Optimierung des entlasteten Wellendichtrings, Dissertation, Universität Stuttgart.