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Rubber ageing at elevated temperature – model calibration

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1. Introduction

The dynamic network model by Naumann and Ihlemann [4] is considered in order to capture mechanical behavior of rubber subjected to mechanical and thermal loading. The model has been adapted to account for fatigue damage instead of Mullins effect [3].

The model is based on a kinematic split such that the deformation gradient

$$\mathbf{F} = \mathbf{F}_2\,\mathbf{F}_1\,, \tag{1}$$

where $\mathbf{F}_1$ corresponds to a stress-free state of deformation and $\mathbf{F}_2$ is the elastic deformation. In its simplest form (neo-hookean), the strain energy density is

$$W = C_{10}\,(1 - D)\,\mu\,\nu\,\left(\mathbf{C}_2 : \mathbf{I} - 3\right), \tag{2}$$

where $\mathbf{C}_2 = \mathbf{F}_2^{\mathrm{T}}\,\mathbf{F}_2$ is the right Cauchy-Green deformation tensor corresponding to the elastic part of deformation. The internal variables μ and ν reflect increase and decrease in material stiffness due to chemical ageing respectively and under the assumption of homogeneous oxygen distribution, valid, e.g., in thin rubber samples, are driven by the following evolution equations:

$$\dot{\mu} = k_{\mathrm{R}}\,\mu\,, \quad \dot{\nu} = -k_{\mathrm{S}}\,\nu\,, \quad \mu(0) = \nu(0) = 1\,, \tag{3}$$

where the coefficients are usually assumed to behave according to the Arrhenius law

$$k_{\{\mathrm{R,S}\}} = k_{\{\mathrm{R,S}\}_0} \exp\left[-\frac{E_{\{\mathrm{R,S}\}}}{RT}\right], \tag{4}$$

$k_{\{\mathrm{R,S}\}_0}$ and $E_{\{\mathrm{R,S}\}}$ being material parameters, $R = 8.314\,\mathrm{J\,mol^{-1}\,kg^{-1}}$ the gas constant, and T absolute temperature. In the case of thick samples, the diffusion equation with an added reaction term would be used to model oxygen transport and both k_{R} and k_{S} would depend on oxygen concentration.

Fatigue damage is described by a power law [1]

$$\dot{D} = (-A\,Y)^a\,, \quad Y = \frac{\partial W}{\partial D}\,, \tag{5}$$

A and a being its parameters.

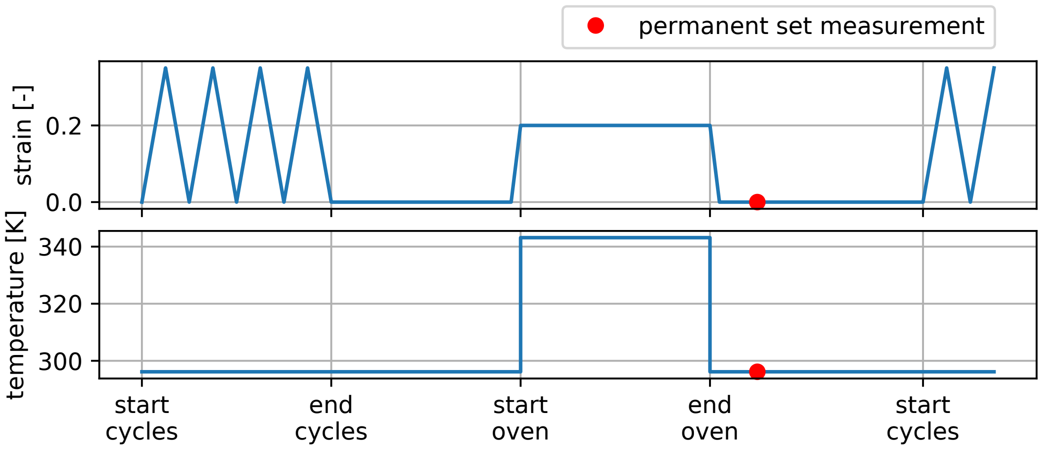

Fig. 1. Single block of prescribed strain and temperature

2. Experimental procedure

The test sequence, used for calibration of the model, consists of repeated cyclic loading and an ageing period of constant deformation at elevated temperature, see Fig. 1. The measured data contain both stress-strain curves from the cyclic loading and permanent set measured before and after each ageing period. All cycles were performed up to $35\,\%$ nominal deformation in compression whereas the constant deformation during the ageing periods was one of 0, $15\,\%$ or $25\,\%$. The ageing temperature was either $23 \pm 2\,°\text{C}$ or $70 \pm 2\,°\text{C}$.

3. Numerical modeling and model calibration

The numerical simulations were implemented with the assumption of uniaxial stress and homogeneous distribution of oxygen and temperature. The system of partial differential equations therefore changes into ordinary differential equations. Moreover, the ageing variables μ and ν become independent of strain history.

The calibration procedure was formulated as a nonlinear least squares problem with the objective function to be minimized

$$f(\mathbf{x}) = \sum_i \frac{\sum_j \left(\sigma_{ij} - \sigma_i(t_j)\right)^2}{\sum_j \sigma_{ij}^2}\,, \tag{6}$$

where i denotes different experiments (e.g., cyclic stretch, permanent set, different stretch values), t_j are values of the independent variable (i.e., time), σ_{ij} are measured values, and $\sigma_i(\cdot)$ is the numerical simulation of the i-th experiment, and the vector $\mathbf{x} = [C_{10}, k_\mathrm{R}, E_\mathrm{R}, k_\mathrm{S}, \ldots]^\mathrm{T}$ contains the unknown parameters of the model.

It is known that calibration of the Arrhenius-type relation might lead to multicollinearity, but several techniques exist that overcome this issue [5]. However, in the case of the experimental data considered here, the principal obstacle is insufficient data regarding temperature, i.e. only two temperature values being measured. The solution to this problem is to discard relation (4) completely and consider separate reaction rates for each temperature: $k_{\mathrm{R}23}$, $k_{\mathrm{R}70}$, $k_{\mathrm{S}23}$, and $k_{\mathrm{S}70}$. Should sufficient temperature-data be available, the parameters of (4) may be fitted to the values of the independent reaction rates.

Based on numerical examples [2], a combination of relaxation and permanent-set measurements is suggested in order to identify the time- and history-related parameters of the dynamic network model under constant temperature. The data available here (Section 2) contain cyclic

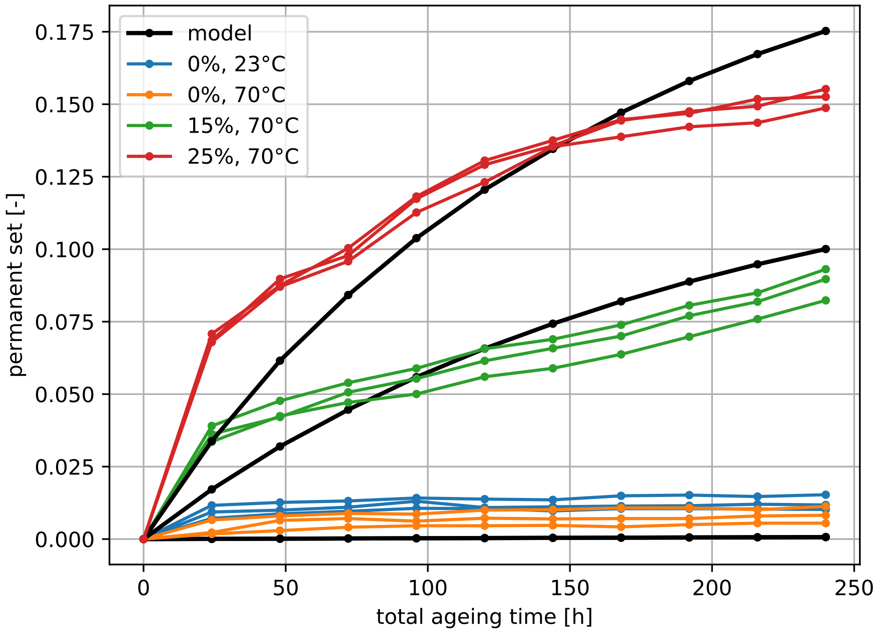

Fig. 2. Comparison of measured and predicted permanent-set

stretch instead of relaxation. Similar numerical examples with these data used here suggest that the reaction rates are identifiable in the absence of damage. The effects of damage compete with those of network-scission, which might lead to non-uniqueness of model calibration.

Nevertheless, an attempt to calibrate the model to real-world data was made. The identified parameters are:

$$C_{10} = 6.69\,\mathrm{MPa}\,, \qquad k_{\mathrm{R23}} = 6.36 \cdot 10^{-7}\,\mathrm{s}^{-1}\,, \qquad k_{\mathrm{R70}} = 1.12 \cdot 10^{-6}\,\mathrm{s}^{-1}\,,$$
$$k_{\mathrm{S23}} = 5.90 \cdot 10^{-7}\,\mathrm{s}^{-1}\,, \qquad k_{\mathrm{S70}} = 8.32 \cdot 10^{-7}\,\mathrm{s}^{-1}\,, \qquad A = 1.18 \cdot 10^{-10}\,\mathrm{Pa}^{-1}\,,$$
$$a = 2.25\,.$$

A comparison of measured and predicted permanent-set is shown in Fig. 2. Comparison of stress-strain curves is shown in Fig. 3.

4. Summary and conclusions

Results of the calibration clearly show several discrepancies between the model and real-world data. The simplifications considered of lesser importance are uniaxial stress assumption and neglecting of oxygen diffusion.

One of the most obvious (and expected) source of disagreement is that only the simplest form of the dynamic network model was used, i.e., only the neo-hookean term with the coefficient C_{10}. Should other terms be included, better description of the stress-strain curves would be expected.

Another shortcoming of the model, when compared to the real-world data, is probably omission of mechanisms other than chemical ageing. This can be shown from the histories of permanent-set, where the blocks of cyclic stretch induce some permanent deformation in the experimental data, as opposed to the numerical model where the impact of cyclic stretch

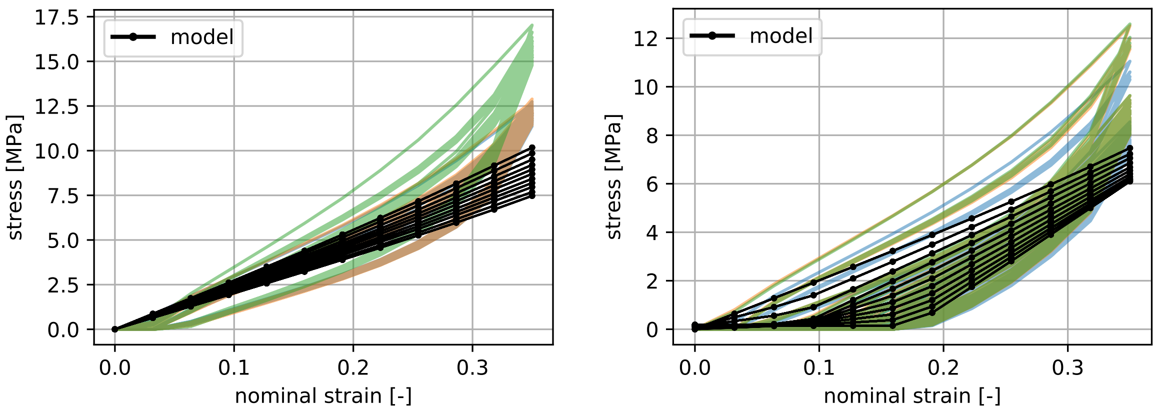

Fig. 3. Stress-strain curves at 0 % (*left*) and 25 % (*right*) strain during the ageing blocks

blocks on permanent-set seems to be negligible. This behavior might be corrected by adding mechanisms such as plasticity, viscoelasticity or physical ageing to the model.

Finally there is the issue of sufficient data and proper selection of loading histories that would enable the identification of a unique combination of parameters. This is already subject of an ongoing research [2], however, the results of this work are essential regarding further modifications of the material model, as discussed above.

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References

[1] Ayoub, G., Naït-Abdelaziz, M., Zaïri, F., Gloaguen, M., Charrier, P., A continuum damage model for the high-cycle fatigue life prediction of styrene-butadiene rubber under multiaxial loading, International Journal of Solids and Structures 48 (18) (2011) 2458-2466.

[2] Heczko, J., Kottner, R., Identification of parameters in the case of material model of chemical ageing and damage, Proceedings of the 11th European Conference on Constitutive Models for Rubber – Constitutive Models for Rubber XI, Nantes, CRC Press/Balkema, 2019, pp. 593-598.

[3] Heczko, J., Kottner, R., Modelling of ageing and fatigue under large strains, Proceedings of the conference Computational Mechanics 2017, University of West Bohemia, Špičák, 2017, pp. 31-32.

[4] Naumann, C., Ihlemann, J., A dynamic network model to simulate chemical aging processes in elastomers, Proceedings of the 9th European Conference on Constitutive Models for Rubber – Constitutive Models for Rubber IX, Prague, CRC Press/Balkema, 2015, pp. 39-45.

[5] Rodionova, O. E., Pomerantsev, A. L., Estimating the parameters of the Arrhenius equation, Kinetics and Catalysis 46 (3) (2005) 305-308.