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Simulation of the Long Term Behavior of Polymers on the Basis of Short Term Data

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The mechanical long term behavior of polymers is usually determined by long term creep experiments up to 10.000 h which are very time and cost consuming. Therefore these data are often not available for the engineer. But even if long term data is available FEA-tools used in industrial practice usually can not sufficiently represent the nonlinear time, temperature and load-dependency of the mechanical long term behaviour. Hence the dimensioning of polymer parts is still a rather difficult engineering task. As a consequence of this, additional time consuming and expensive component tests in combination with several iterations are often necessary to design a polymer part for long term behaviour.

This paper develops a method for the modeling of the load and temperature-dependent mechanical behaviour of polymers over a wide time, load and temperature range by means of finite element analysis. The method includes a material model as well as the determination of material parameters to calibrate the model. As a special feature of this method the model is calibrated by using creep data generated from short term experiments. The procedure improves the simulation of the long term behaviour of plastic-components and reduces the experimental effort significantly. The simulation results are finally validated by creep experiments performed on an example part.

1 Introduction

FEA-tools used in practice usually offer linear viscoelastic models to simulate the long term behaviour of polymers. These models cannot sufficiently represent the load-dependence of the mechanical long term behavior of plastics that even occurs for non-finite strains. Therefore the dimensioning of plastics parts is still difficult for engineers. An adequate material model has to represent the mechanical long term behavior over a wide time and load range. Most of the existing nonlinear viscoelastic material models in literature need additional experiments to be calibrated (e.g. Blaese, 1999; J.H.J. Beijer, 2002; Schapery, 1969; Schmachtenberg, 1985; Yazici, 2004). The main focus of the following method is to calibrate a nonlinear viscoelastic model with uniaxial tensional creep data that are commonly available in data bases. So the additional experimental effort can be minimized.

Besides the load-dependency the temperature-dependency has to be considered, too. Since in many cases temperature-dependent creep data is not available in material data bases, the question appears how to simulate the long term behaviour if the temperature-dependent creep curves are not available. As there are usually creep experiments over at least 10000 h at different temperatures necessary, the determination of the material data is time and cost intensive. For this purpose the time temperature superposition principle has to be modified. With this modification the temperature-dependent long term behaviour can be described over a wide time and temperature range just on the basis of short term creep data.

2 Experiments

One of the most crucial issues in developing the nonlinear viscoelastic material model is to find an adequate parameter describing the load-dependency. In many publications it is shown that the free volume effects the mechanical behaviour of polymers significantly (e.g. McKenna, Leterrier and Schultheisz, 1995a; McKenna, Leterrier and Schultheisz, 1995b; P. A. OConnell and G. B. McKenna, 1997; Struik, 1980). Furthermore the concept of free volume has also proved to be useful in describing viscosity, glass transition and physical ageing (Doolittle, 1951; L. C. E. Struik, 1977; Robertson, Simha and Curro, 1984; Simha and Roberston, 2006; Williams, Landel and Ferry, 1955). This parameter seems to be appropriate to consider load-dependency as well as influences like temperature and ageing effects. So the influence of three different effects can be determined by only one parameter. In (Ferry and Stratton, 1960; Knauss and Emri, 1987; Kola řík, 2003; Kolarík and Pegoretti, 2006; Losi and Knauss, 1992) volumetric strain is said to constitute a significant contribution to the free volume in the region of reversible strains beyond the yield strain. This assumption has to be proven by experiments.

The influence of the free volume on the creep behaviour of polymers is confirmed by the experimental outcome shown in Figure 1-3. In Figure 1 a result of a dilatometer test is shown. A polycarbonate specimen is heated above its glass transition temperature (160°C) for 20 minutes and afterwards quenched to 90°C. Although the temperature is kept constant for 48 h and no load is applied, the volume decreases with increasing time. This phenomenon is known as physical ageing or volume relaxation (Struik, 1966, 1977; Struik, 1978).

In a second run short term creep experiments were conducted at different ageing times. As represented in Figure 2 the creep modulus increases with increasing ageing time and accordingly decreasing volume. When the short term creep modulus is plotted against the volume a nearly linear correlation can be stated for the investigated polycarbonate (see Figure 3).

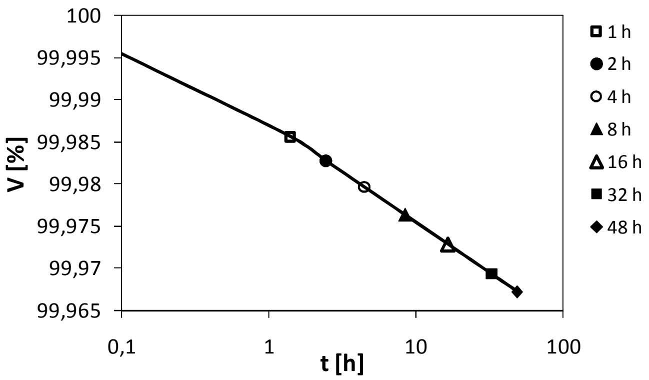


Figure 1. Volume relaxation of a polycarbonate at 90°C

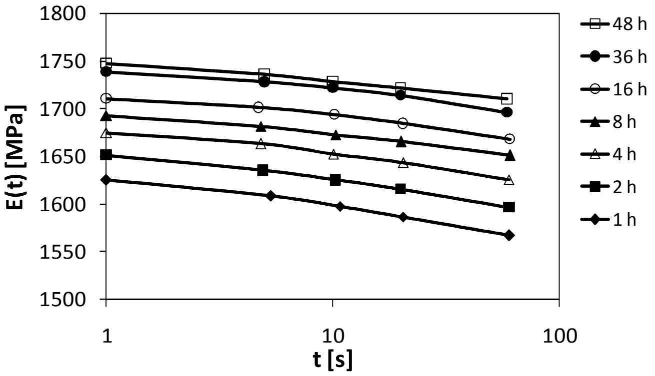


Figure 2. Increase of creep modulus with increasing ageing time

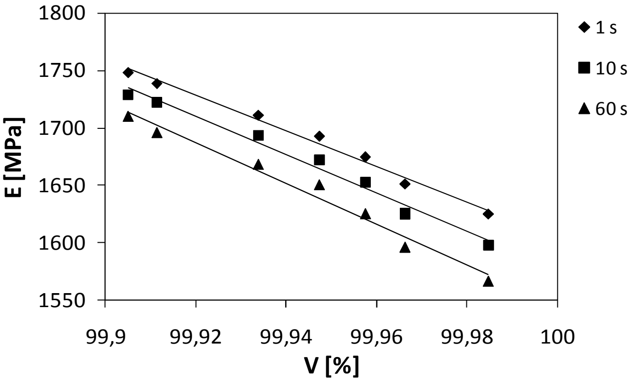


Figure 3. Correlation of volume and short term creep modulus at 1 second, 10 seconds, and 60 seconds

The observation of a dependency between volume and creep modulus leads to the assumption that the creep modulus is a function of the hydrostatic pressure and the volumetric strain respectively. To verify this assumption creep tests under shear and tension are conducted. In a tension test the stress tensor can be separated in a hydrostatic part and an isochoric part

$$\sigma = pI + S_{dev} \tag{1}$$

The hydrostatic part is

$$p = \frac{1}{3}\sigma_{11} \tag{2}$$

In a shear test the hydrostatic part becomes zero and the stress tensor reduces to

$$\sigma = S_{dev} \tag{3}$$

The creep curves obtained from these two tests are compared to determine if the creep behavior is influenced by the hydrostatic pressure.

The shear creep curves measured in a shear creep test on an Iosipescu specimen (Figure 4) are shown in Figure 5. Figure 6 shows the shear creep curves calculated according to (Krevelen, 2008) from tensile creep data with

$$G(t) \approx \frac{E(t)}{2(1+\nu(t))} \tag{4}$$

G(t) shear creep modulus
E(t) tension creep modulus
v(t) Poisson’s ratio

with

$$\upsilon(t) = -\frac{\varepsilon_{22}}{\varepsilon_{11}} \tag{5}$$

and

$$E(t) = \frac{\sigma}{\varepsilon(t)} \tag{6}$$

The creep experiments are conducted at a constant temperature with a temperature tolerance of ±2°C according to (DIN, 1997). A comparison of the resulting curves in Figure 5 and Figure 6, shows that the load dependency in tension is much higher as in shear. Furthermore the error of measurement for these shear tests at the same load are about 10 %, which shows that the occuring load-dependency in the shear creep test could partially be a result of measuring inaccuracy. This confirms the assumption that the creep modulus is significantly influenced by hydrostatic pressure.

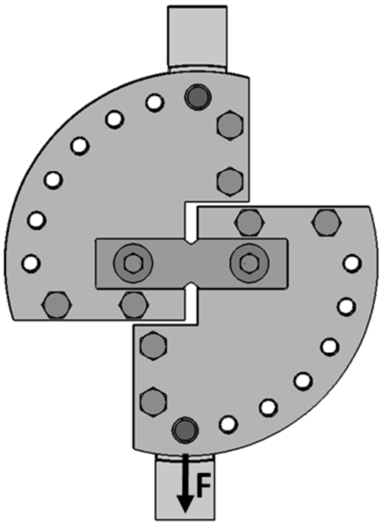


Figure 4. Experimental set up - shear test on a Iosipescu shear specimen

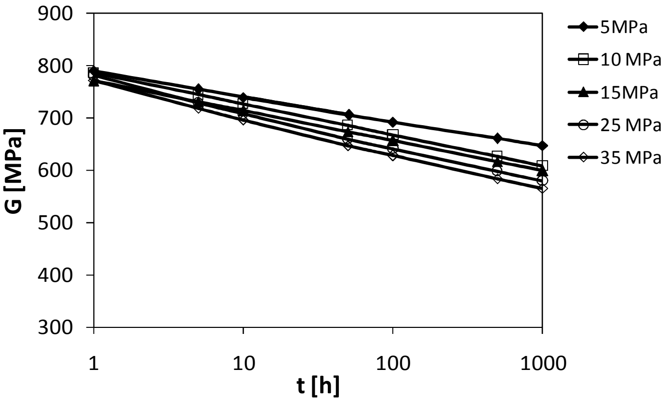


Figure 5. Shear creep curves measured in shear creep test

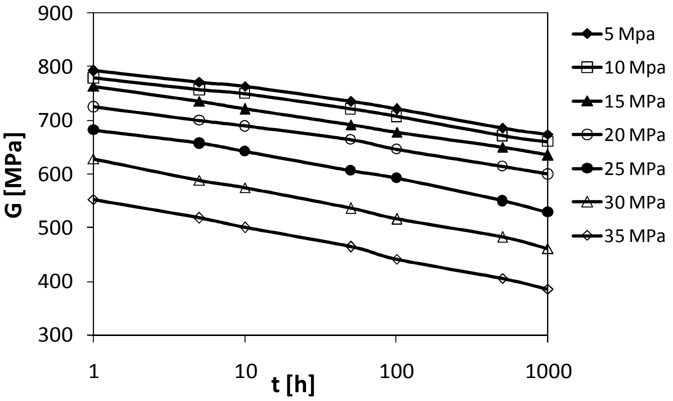


Figure 6. Shear creep curves calculated from tensile creep curves

A further investigation of the dependency of the creep modulus on the hydrostatic pressure is carried out by experiments in a special test device for conducting creep experiments under hydrostatic pressure up to 300 bar (Stommel and Zimmermann, 2011).The specimen is placed in a metal cylinder filled with degassed water. The increase in pressure is realised by a reduction of volume due to screwing in a spindle of a choke valve. The specimen's elongation is applied by another spindle in the head and a nut. The resulting force can be indirectly measured via a compression spring combined with a path sensor. The known spring stiffness enables the determination of the resulting tensile stress. The experiments proof the significant influence of the hydrostatic pressure on the creep behaviour. For the three investigated materials an increase in the creep modulus is obtained for increasing pressure (Figure 8-10).

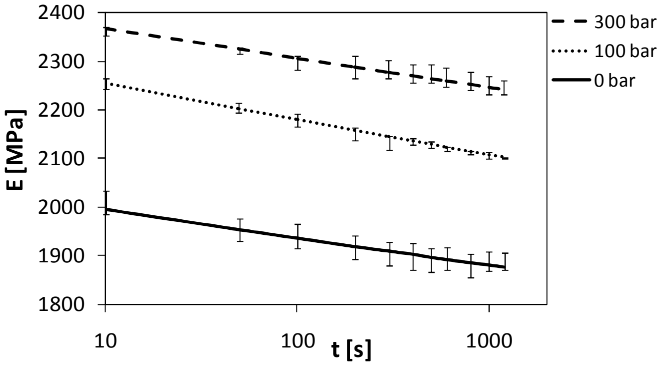


Figure 7. Creep tests under different hydrostatic pressures for a Polycarbonate (PC)

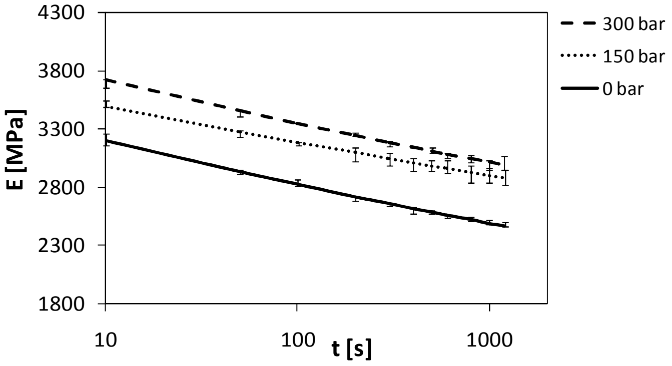


Figure 8. Creep tests under different hydrostatic pressure for a Polymethylmethacrylate (PMMA)

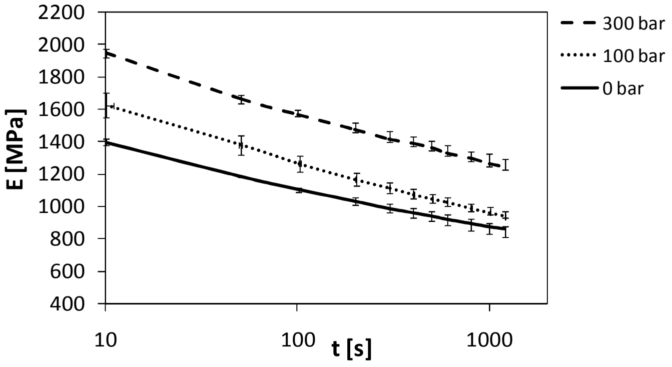


Figure 9. creep tests under different hydrostatic pressures for a Polypropylene (PP)

The conducted experiments show that the creep behaviour is significantly influenced by hydrostatic pressure or volumetric strain, respectively. Both the two amorphous polymers PC and PMMA and the semicrystalline polymer PP show this behaviour. These experimental results can now be used to develop a material model with regard to the real load-dependent long term behaviour of thermoplastic polymers.

3 Material Model

There exist different material models in the literature to describe the nonlinear viscoelastic behaviour of polymers (e.g. Drapaca, Sivaloganathan and Tenti, 2007; Aniskevich, Khristova and Jansons, 2000; Caruthers, Adolf, Chambers and Shrikhande, 2004; Cho and Kim, 2000; Spathis and Kontou, 2001; Wineman, 2009). Many models need additional experimental results (e.g. Blaese, 1999; J.H.J. Beijer, 2002; Schapery, 1969; Schmachtenberg, 1985; Yazici, 2004) or focus on the mechanical behaviour of special polymers (e.g. C.P. Lowe, 2004; Diab and Wu, 2007; Drozdov, 1999). Furthermore there are a lot of nonlinear viscoelastic material models that do not focus on long term behaviour as Buckley and Jones, 1994; Buckley and Lew, 2011; Lin, 2001. In contrast to this, the aim of the approach presented here is to simulate the load-dependency of the long term behaviour of polymers just on the basis of tensile creep data and short term data so that the additional experimental effort can be minimized.

The developed model bases on Wiechert model, that consists of a series of Maxwell elements and an single elastic spring in parallel and is well known for a good description of the linear viscoelastic behavior of polymers (Ferry, 1980; Findley, Lai and Onaran, 1976). Even in actual publications the model is frequently used to describe the linear viscoelastic material behavior of polymers (e.g. Yazici, 2004 ;F. Ellyin, 2007; Pittman and Farah, 1997; Schmachtenberg, Krumpholz and Brandt, 2005). The shear modulus G(t) is time-dependent and load-independent. The time-dependency of the bulk modulus is often not available in material databases. Hence the simplification is made to assume the bulk modulus K as time-independent or to take the same time-dependency for both the shear and the bulk modulus.

$$\sigma_{ij}(t) = \delta_{ij}\sigma_{vol} + S_{ij} = \delta_{ij}K(t)\varepsilon_{vol} + \int_0^t 2G(t-t')\,\dot{e}_{ij}dt \tag{7}$$

σ_{vol}	volumetric stress
S	deviatoric stress
K	bulk modulus
ε_{vol}	volumetric strain
G	shear modulus

Based on the experimental results presented in the last Section, the change in free volume is taken as parameter to describe the load-dependency. It correlates with volumetric strain and can be calculated by equation (7). As described and validated in (Naumann and Stommel, 2010a, b, c) further assumptions in our model are a load-dependent but time independent bulk modulus and a time and load-dependent Poisson's ratio which results in a time and load-dependent shear modulus. Besides these assumptions are also confirmed by different literature sources like (Krevelen, 2008) where the bulk modulus is said to be the only time-independent modulus and (Frank, 1984; Kolupaev, Moneke and Becker, 2005; Kolupaev, Moneke, Becker and Amberg, 2004; Lewen, 1991 Wieser, 1998a) where the time-dependency of the Poisson's ratio is proven. Furthermore the creep behaviour is seen as load-independent for $\Delta f < 0$. This assumption results from the observations of (Sarabi, 1984; Wieser, 1998) who show that the load dependency under compression appears only at very high loads. With these assumptions the material model develops from (7) to its final state

$$\sigma_{ij}(t) = \delta_{ij}\,K(\Delta f)\,\varepsilon_{vol} + \int_0^t 2\left(G_\infty(\Delta f) + \sum_{k=1}^{n} G_k(\Delta f)\,e^{(t'-t)/(\tau_k)}\right)\dot{e}_{ij}\;dt' \tag{8}$$

$$\Delta f = (1-2\upsilon(t)) = \varepsilon_{vol} \tag{9}$$

$$K(\Delta f < 0) = K(\Delta f = 0) \tag{10}$$

$$G(\Delta f < 0) = K(\Delta f = 0) \tag{11}$$

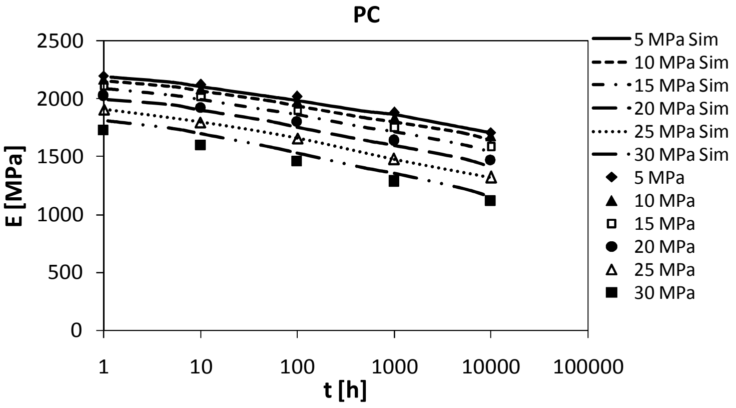
σ_{ij}	Cauchy Stress
K	bulk modulus
ε_{vol}	volumetric strain
G_∞	equilibrium fraction of prony series for shear modulus
G_k	prony coefficients
t	Poisson ratio
Δf	change of free volume

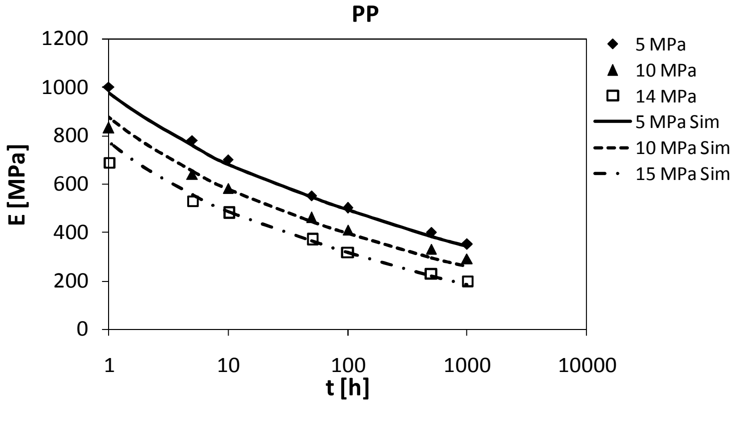
As mentioned before it is one main objective that calibrating the material model should not cause additional experimental effort. In the present case this requirement is fulfilled by making some reasonable assumptions (see Naumann and Stommel, 2010b, c) . So it is possible to calibrate the model only with tensional creep data, which are in most cases accessible in material data bases. Moreover a routine was developed, which allows a completely automated determination of the model parameters.

With this model it is possible to cover the nonlinear load-dependency and deformations up to approximately 2 %. Finite deformations and other geometric nonlinearities are momentarily not included in the model. But this is not a severe restriction as most technical thermoplastics are used at long term deformations beyond 2% technical strain as there occurs plasticity at higher deformations.

4 Simulation Results – Load-Dependency

Figure 10 shows the simulation results of an uniaxial tensile creep test for 5 different materials. For every investigated materials a parameter set is determined with the developed routine. As it is shown the model can represent the creep behaviour over a wide load range for all 5 materials.





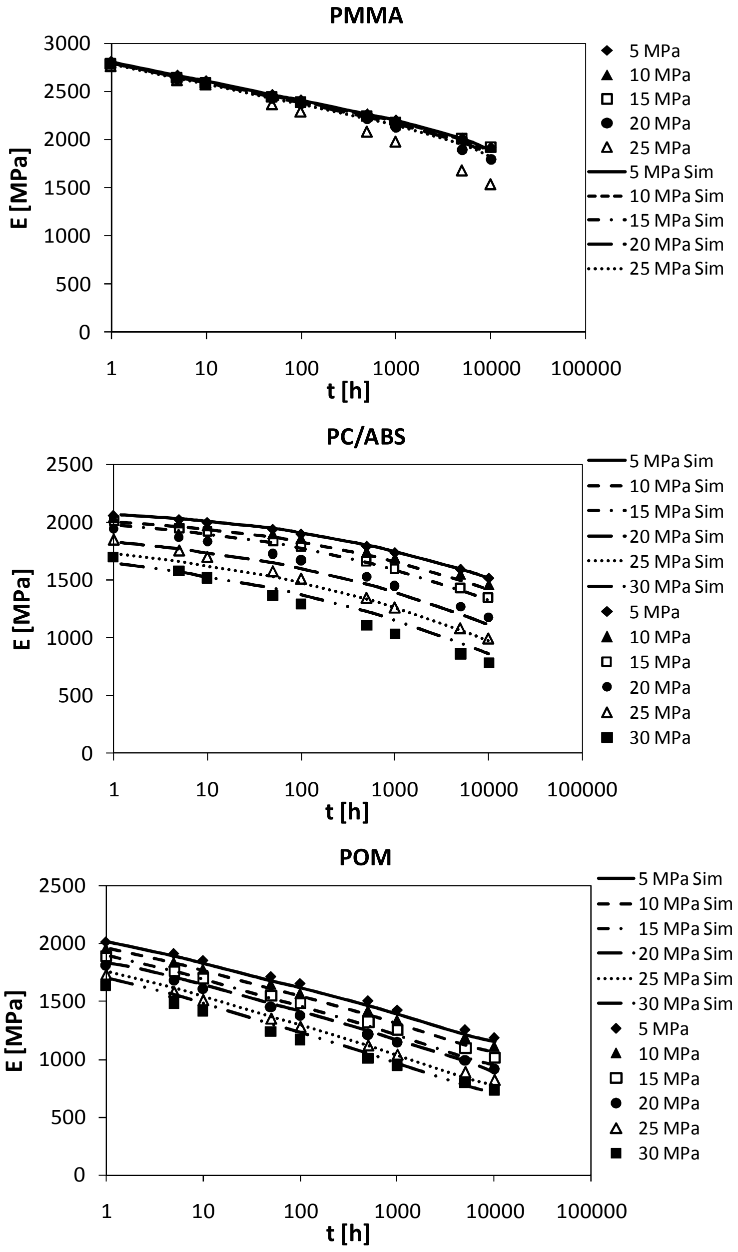


Figure 10. Uniaxial creep behaviour - comparison of simulation and creep data

Since most plastics components are usually exposed to multiaxial loadings, the model is verified by experiments on a multiaxially loaded part. The part is simultaneously loaded by bending and torsion. The torsional deformation and the deflection of the part are determined by an optical measurement system. The simulations are conducted with Abaqus CAE. The model was implemented as an Abaqus user-subroutine. The model consists of 33490 quadratic tetrahedron solid elements. The experimental set-up and the simulation are shown in Figure 11.

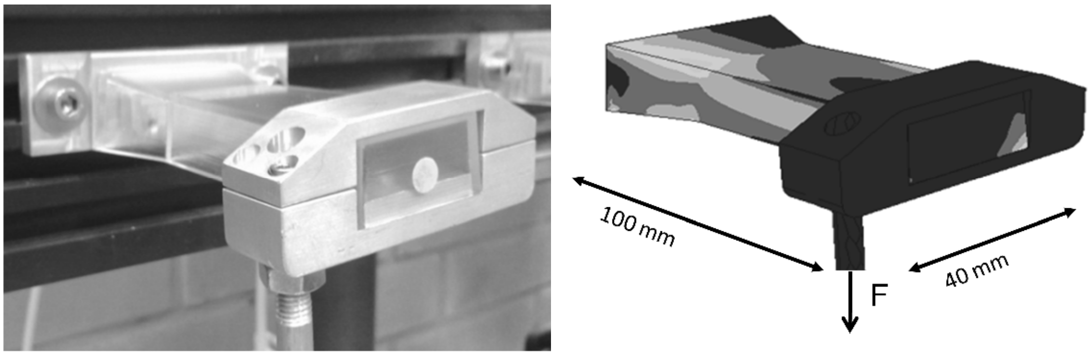


Figure 11. Creep test on an example part - experimental set up (l) and simulation (r)

In Figure 12 and Figure 13 the average value of the torsion angle α and the deflection of the part for applied loads of 200 N, 450 N over 1000 h are shown. The simulation results describe the real deformation behavior of the part for all load levels satisfactory.

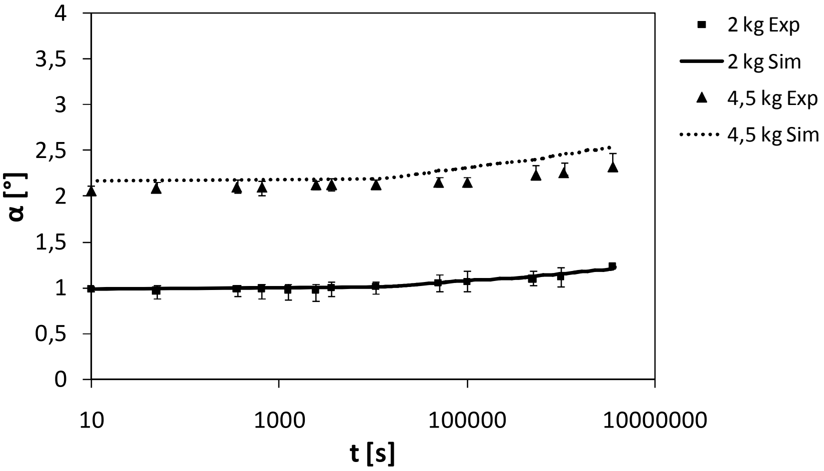


Figure 12. Torsional angle - comparison of experiment and simulation

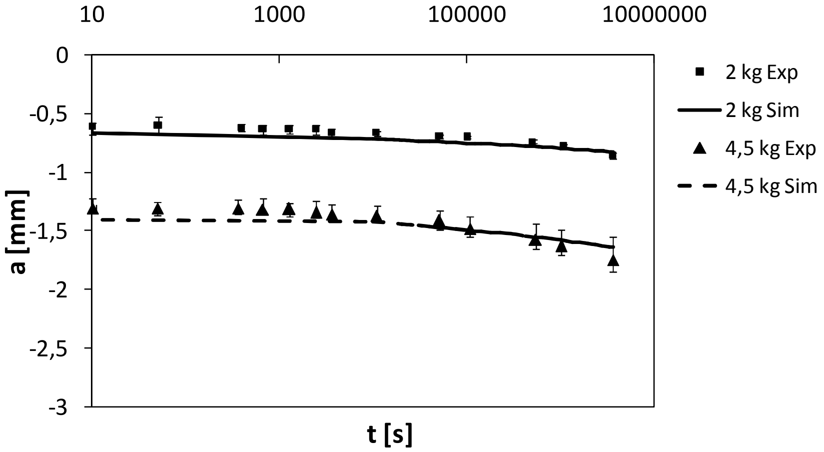


Figure 13. Deflection - comparison of experiment and simulation

5 Simulation of the Temperature-Dependency

Besides the load-dependency of the long term behavior the temperature-dependency has to be considered also. Since in many cases the temperature-dependent creep data is not available in material data bases, creep tests at different temperatures have to be conducted. As the main aim of our approach is to minimize the additional experimental effort, the description of temperature-dependency is introduced in this approach on the basis of short term creep data.

The time temperature superposition principle is often used in the literature for this purpose and is well known to deliver satisfying simulation results. This approach is used here to shift the mastercurve at a reference temperature along the time axis to a short term creep curve at another temperature, Figure 14.

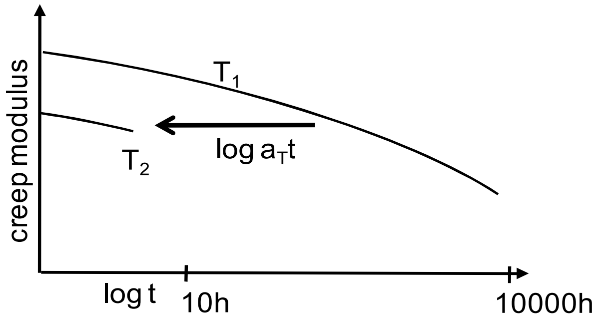


Figure 14.Horizontal shift at time temperature superposition principle

With this modification the model in (3) develops to the following equation:

$$\sigma_{ij}(t) = \delta_{ij} K(\Delta f)\varepsilon_{vol} + \int_0^t 2\left(G_\infty(\Delta f) + \sum_{k=1}^{n} G_k(\Delta f)\, e^{(t'-t)/(a_T\cdot\tau_k)}\right)\dot{e}_{ij}\; dt' \tag{12}$$

The introduction of the time temperature shift factor a_t results in a temperature-dependent scaling of the relaxation times and can be determined from approximately 2h short term creep tests. This approach is based on the iso free volume model. This model was proposed by (Fox Jr and Flory, 1950; Simha and Boyer, 1962) and claims that the free volume fraction below T_g is constant and temperature-independent. Therefore the free volume fraction below T_g is still just load-dependent

$$\Delta f = (1 - 2\upsilon(t)) = \varepsilon_{vol} \tag{13}$$

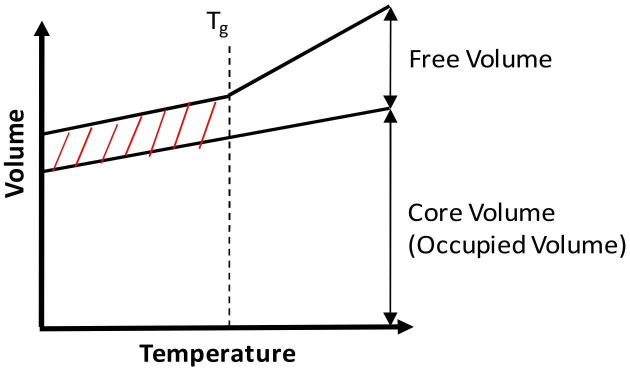


Figure 15. Iso free volume approach

The simulation offers a good description of the temperature-dependent creep curves for a reference temperature of 23°C. The curves are shifted to the left side with increasing temperature. Hence the well described area is getting smaller with increasing temperature, the creep behaviour at 90°C cannot be described with this approach.

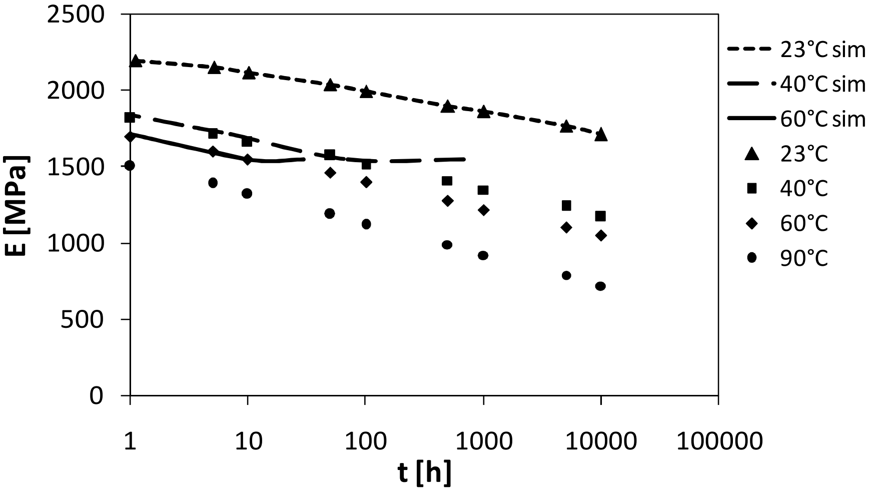


Figure 16. Simulation of the temperature-dependent creep behaviour with TTSP

Although it is the usual practice to apply only a horizontal shift (e.g. Jazouli, Luo, Bremand and Vu-Khanh, 2006; Knauss and Zhu, 2002; Zhao, Luo, Li and Chen, 2008) as it is done above, many publications describe a necessary vertical shift (Ferry, 1980; H. Markovitz, 1975; P.A. OConnell and G.B. McKenna, 1997). This vertical shift is caused by a change in density below the glass transition temperature. As it can hardly be quantified, this vertical shift is often ignored which can cause unrealistic simulation results as described in (Brinson and Brinson, 2008; O'Connell and McKenna, 2002). Therefore the model is further modified to apply also a vertical shift.

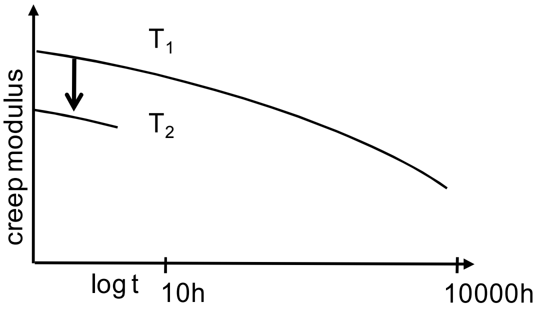


Figure 17. Vertical shift at time temperature superposition principle

The iso core model (Boyer and Spencer, 1944, 1945; Spencer and Boyer, 1946) claims that the whole thermal expansion below T_g is caused by free volume Figure 18. In this case the change in free volume below T_g becomes from equation (13) to

$$\Delta f = (1 - 2\upsilon(t)) + \alpha_{fv}\Delta T \tag{14}$$

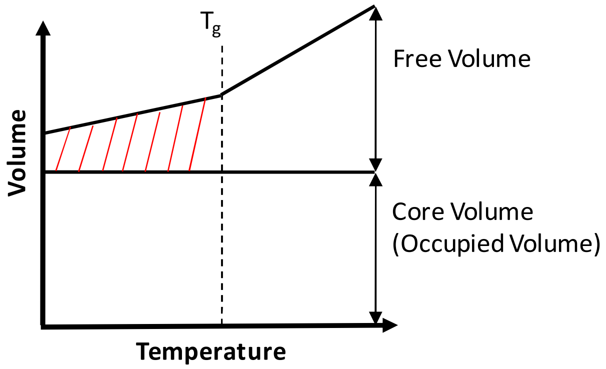


Figure 18. Iso core volume approach

With this modification, a change in temperature results in a change of shear modulus and a horizontal shift of the mastercurve, respectively. According to the iso core volume approach the coefficient of thermal expansion of the free volume correlates to the coefficient of thermal expansion of the material. This coefficient is available in material data bases for most materials, so no further experimental effort is necessary. For the present polycarbonate the linear coefficient of expansion is given in (CampusPlastics, 2011) as $\beta=65\cdot10^{-6}$ 1/K. For an isotropic material follows the volumetric coefficient of expansion $\alpha_{fv}\approx3\cdot\beta=19{,}5\cdot10^{-5}$ 1/K (Schwarzl, 1990). The simulation results for this approach are shown in Figure 19.

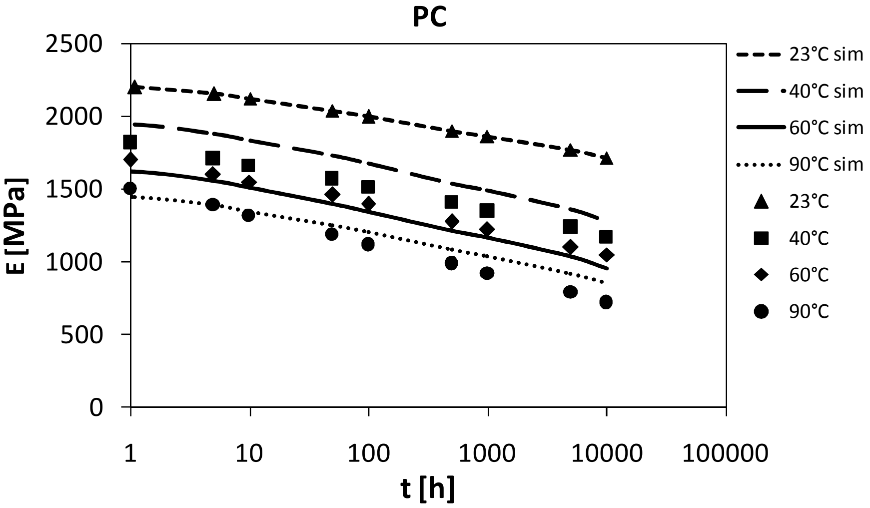


Figure 19. Simulation of the temperature dependent creep behaviour (iso core volume approach)

The change in free volume due to a variation in temperature proposed by this approach is too large and no reasonable additional horizontal shift can be applied (Figure 19). This is caused by the fact that the coefficient of thermal expansion is too large. Nevertheless, the iso core volume approach can be used to get a first estimation of the temperature-dependent long term behaviour, if no temperature-dependent creep data is available. But for an exact determination of the temperature-dependent long term behaviour a different approach has to be used so that both horizontal and vertical shift can be applied.

The thermal expansion of the material is supposed to be caused by thermal expansion of free volume as well as by thermal expansion of the occupied volume. In (Dlubek, Pionteck and Kilburn, 2004; Hagiwara, Ougizawa, Inoue, Hirata and Kobayashi, 2000) the real thermal expansion behaviour of free volume for different polymers is measured by positron annihilation lifetime spectroscopy (PALS) and it is shown that the free volume as well as the occupied volume contribute to thermal expansion (Figure 21). This thermal expanision behaviour results in a smaller vertical shift as with the iso core volume model and an additional horizontal shift.

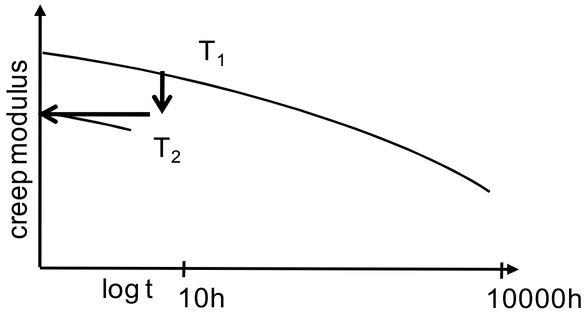


Figure 20. Combination of vertical and horizontal shift at time temperature superposition principle

In a further simulation the thermal coefficient of expansion of free volume below T $_g$ for a polycarbonate is used. This coefficient is determined in (Hagiwara, Ougizawa, Inoue, Hirata and Kobayashi, 2000) by PALS as α_{fv}=10,5·10^{-5} 1/K. The simulation results using this value are shown in Figure 22.

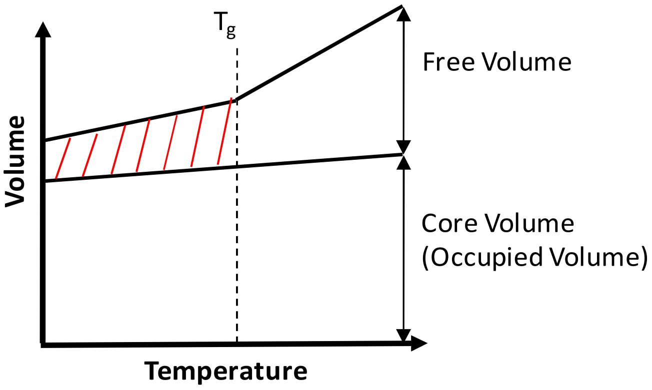


Figure 21. Real thermal expansion of free volume

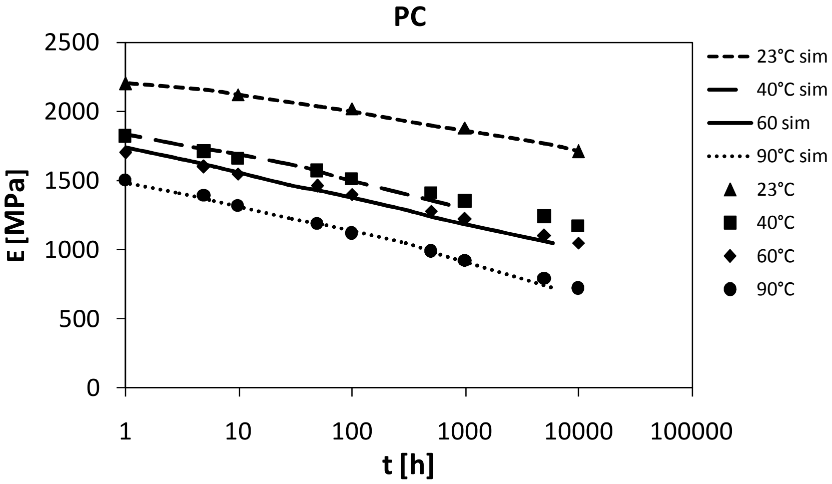


Figure 22. Simulation of the temperature-dependent creep behaviour

The temperature-dependency of the creep behaviour can be described in a good approximation for all temperatures. Furthermore no time consuming creep experiments at different temperatures are needed to calibrate the model. Just the thermal expansion coefficient of the free volume is needed. This coefficient can be determined by Positron Annihilation Lifetime Spectroscopy (PALS). For several polymers information about this expansion coefficient already exists in the literature and can be used as well (e.g. Dlubek, Pionteck and Kilburn, 2004; Hagiwara, Ougizawa, Inoue, Hirata and Kobayashi, 2000).

7 Conclusion and Objectives

The present paper describes a material model to simulate the nonlinear load-dependency of the long term behavior of real plastics components over a wide load range. By integrating a modified time temperature-superposition-principle, the long term behavior over a wide temperature range can be simulated as well. This is shown on simulations on a multiaxially loaded part. The special feature of the derived method is that only creep curves and short term tests are necessary to calibrate the model. Hence the additional experimental effort for the engineer is minimized.

A special advantage of the model is that the load-dependency as well as the temperature-dependency can be described with the same parameter, the change of free volume. This parameter can also be used to describe other influences on long term behaviour such as physical ageing. Further research will be necessary in modifying the model so that also these influences can be regarded by the model.

Moreover this approach will be further validated by experiments. In these experiments more different materials as well as other example parts will be investigated. In addition the assumption that there is nearly no load-dependency under compression will be investigated as this assumption is only based on literature sources (Krumpholz, 2005; Sarabi, 1984; Wieser, 1998a) at the moment.

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