ON THE CREEP PROPERTIES OF POLYVINYL-CHLORIDE

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1. INTRODUCTION

The mechanical properties of viscoelastic materials may be represented by creep of stress-relaxation functions which enable formulation of the functional constitutive equations. For a particular material, these functions are usually determined by means of creep or relaxation tests. In practice, creep tests have been more popular, as stress relaxation tests, in which the stress is produced by small constant strain, are not so easy to perform.

A creep function of a material is defined as the time-dependent strain under constant or piecewice constant stress. In general, the creep properties are dependent upon the state of stress, and upon the temperature of the material. If the state is considered to be isothermal, then the linear Bolzmann-Volterra representation, or the nonlinear Green-Rivlin theory, are applicable for the description of creep equations.

The behaviour of most polymeric materials is greatly dependent upon time and temperature. This is also the case in the glass-like zone, and particularly near the transition temperature before the rubber-like state. Consequently the theory of viscoelasticity or thermoviscoelasticity offers a common basis for treatment of the stress-deformation problems of polymers.

A great deal of attention has been given to the viscoelastic properties of polymers in recent years. Those putting forward more general representations for different polymeric materials have included Ferry and Tobolsky [1], [12]. The creep of polyvinyl-chloride has been investigated by Onaran and Findley [4], [10], [1], Lai and Findley [7], Leaderman, Mc Cracken and Nakada [8], and others.

In the present paper, the creep functions are considered for some types of Finnish polyvinyl-chloride. To obtain expressions applicable to practical engineering problems, the simplest possible functions have been used for the description of linear and nonlinear creep behaviour. Furthermore, some creep tests have been made, and the corresponding material constants have been solved by application of the method of least squares.

The following restrictive assumptions have been introduced into the treatment.

- The strains are infinitesimal, and the considered creep equations uniaxial
- The state in the material is isothermal
- The ageing of the material is insignificant.

2. MATHEMATICAL REPRESENTATION OF UNIAXIAL CREEP PROPERTIES

The one-dimensional constitutive equation of a linear viscoelastic material is expressible in terms of the known alternative functionals

$$\sigma(t) = \int_{-\infty}^{t} K(t - \tau)\, \dot{\varepsilon}(\tau)\, d\tau \tag{1}$$

$$\varepsilon(t) = \int_{-\infty}^{t} J(t - \tau)\, \dot{\sigma}(\tau)\, d\tau \tag{2}$$

They include the Bolzmann superposition principle (1874), and the principle of fading memory introduced in its original form by Volterra (1930). If the constant strain state

$$\varepsilon = \varepsilon_o H(t) \,, \quad \dot{\varepsilon} = \varepsilon_o \delta(t) \tag{3}$$

and the constant stress state

$$\sigma = \sigma_o H(t) \,, \quad \dot{\sigma} = \sigma_o \delta(t) \tag{4}$$

are chosen, Equations (1) and (2) correspondingly give

$$\sigma(t) = K(t)\varepsilon_o \tag{5}$$

and

$$\varepsilon(t) = J(t)\sigma_o \tag{6}$$

K(t) denotes the stress-relaxation function, and J(t) the creep function of the material (Fig. 1). The uniaxial mechanical properties of a linear viscoelastic material can be characterized either by the relaxation function K(t), or by the creep function J(t).

In what follows below, only the representation of the creep function J(t) is considered. A simple dependence exists between the

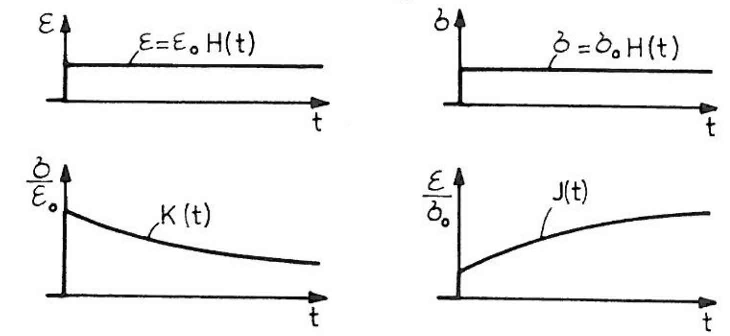

Fig. 1. a) Stress-relaxation function
b) Creep function.

creep and the stress-relaxation function in the linear theory. If the Laplace transforms of K(t) and J(t) are denoted by $\tilde{K}(s)$ and $\tilde{J}(s)$, then

$$\tilde{K}(s)\tilde{J}(s) = \frac{1}{s^2} \tag{7}$$

In the nonlinear theory, this dependence is more complicated. Numerical solution of the stress-relaxation function from the known creep function, or the concerse, is possible under certain conditions of continuity [7], [6 p. 35].

A suitable representation for the constitutive relations of nonlinear viscoelastic materials can be found if, instead of Equations (1) and (2), consideration is given to the more general functionals

$$\sigma(t) = \underset{\tau=-\infty}{\overset{t}{\sigma}} \left[\varepsilon(\tau),\theta\right] \tag{8}$$

and

$$\varepsilon(t) = \underset{\tau=-\infty}{\overset{t}{\varepsilon}} \left[\sigma(\tau),\theta\right] \tag{9}$$

θ is the absolute temperature, and the functionals are restricted by the objectivity requirement.

If the functionals (8) and (9) are Fréchet-differentiable, they can be expanded in series, in powers of ε and σ, which are analogous to the Taylor series expansion of an analytic function. In accord with Green and Rivlin [5], such an expansion is expressible in terms of multiple integrals. By this means, Functional (9) can be written in the form

$$\begin{aligned}\varepsilon(t) = &\int_{-\infty}^{t} J_1(t-\tau_1)\dot{\sigma}(\tau_1)d\tau_1 + \\ &+ \int_{-\infty}^{t}\int_{-\infty}^{t} J_2(t-\tau_1,t-\tau_2)\dot{\sigma}(\tau_1)\dot{\sigma}(\tau_2)\,d\tau_1 d\tau_2 + \qquad (10)\\ &+ \int_{-\infty}^{t}\int_{-\infty}^{t}\int_{-\infty}^{t} J_3(t-\tau_1,t-\tau_2,t-\tau_3)\dot{\sigma}(\tau_1)\dot{\sigma}(\tau_2)\dot{\sigma}(\tau_3)\,d\tau_1 d\tau_2 d\tau_3 + \cdots\end{aligned}$$

$\tau_1, \tau_2, \tau_3, \ldots$ are different time variables in the segment $(-\infty, t)$, and the kernel functions $J_1, J_2, J_3, \ldots$ correspond to the coefficients of the Taylor expansion of an analytic function.

The kernel functions characterize the mechanical properties of nonlinear, viscoelastic materials. They are functions of time and material parameters still dependent upon the temperature θ. If the state of material is free of stresses until time $t = 0$, the integration limit $t = 0$ can be used instead of $t = -\infty$ in Expansion (10). If the initial state is unstrained, $\varepsilon_o = 0$.

The one-step creep case (3) is now obtained by putting

$$\sigma(\tau_1) = \sigma(\tau_2) = \sigma(\tau_3) = \cdots = \sigma_o H(t) \tag{11}$$

of which

$$\dot{\sigma}(\tau_1) = \dot{\sigma}(\tau_2) = \dot{\sigma}(\tau_3) = \cdots = \sigma_o \delta(t) \tag{12}$$

If $\varepsilon_o = 0$, the substitution of Relations (12) into Equations (10) gives

$$\varepsilon(t) = J_1(t)\sigma_o + J_2(t,t)\sigma_o^2 + J_3(t,t,t)\sigma_o^3 + \cdots \tag{13}$$

In accord with Formulae (6) and (13),

$$J(t) = \frac{\varepsilon(t)}{\sigma_o} = J_1(t) + J_2(t,t)\sigma_o + J_3(t,t,t)\sigma_o^2 + \cdots \tag{14}$$

The creep function of a nonlinear viscoelastic material is then dependent upon the corresponding stress (σ_o). J_1 corresponds to the linear creep function (6), and J_2, J_3, ... are called creep functions of higher orders.

In this study, the creep function of polyvinyl-chloride is sought both in the linear form (6)

$$J^o(t) = \varepsilon(t)/\sigma_o \tag{15}$$

and in the nonlinear forms

$$J^*(t) = J_1(t) + J_3(t,t,t)\sigma_o^2 \tag{16a}$$

$$J^*(t) = J_1(t) + J_2(t,t)|\sigma_o| + J_3(t,t,t)\sigma_o^{\,2} \qquad (16b)$$

It is noticeable that for the material Equations (16) indicates the same behaviour in tension and in compression. In the following, some simple expressions are considered for $J^o(t)$ and $J^*(t)$.

The linear creep function $J^o(t)$ is described by the following expressions

- $J_1^o(t) = a_o - a_1 e^{-\alpha t}$ (17)
- $J_2^o(t) = b_o - b_1 e^{-\beta_1 t} - b_2\, e^{-\beta_2 t}$ (18)
- $J_3^o(t) = c_o + c_1 t^n$ (19)

J_1^o (17) corresponds to the creep function of the linear standard material, which has the rheological models in Fig. 2a. For the former model, the coefficients acquire the forms

$$a_o = \frac{1}{E}, \quad a_1 = \frac{E_1 E_2}{E_1 + E_2}, \quad \alpha = \frac{E_1 E_2}{\eta(E_1 + E_2)}$$

J_2^o (18) is obtainable from the five parametric models in Fig. 2b. In the latter case,

$$b_o = \frac{1}{E_1} + \frac{1}{E_2} + \frac{1}{E_3}, \quad b_1 = \frac{1}{E_1}, \quad b_2 = \frac{1}{E_2}, \quad \alpha_1 = \frac{E_1}{\eta_1}, \quad \alpha_2 = \frac{E_2}{\eta_2}$$

The third creep function J_3^o (19) can not be represented by means of simple rheological models. It has been applied for polymers by a number of researchers, probably for the first time by Findley and

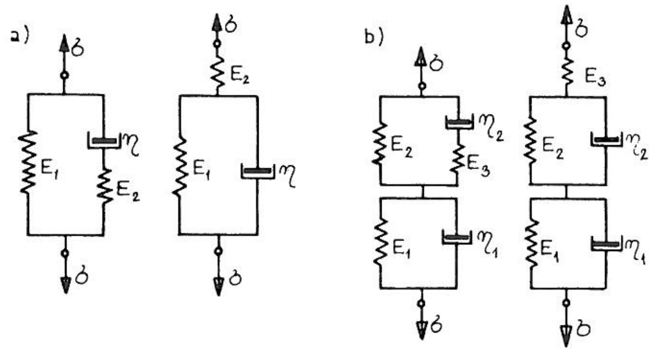

Fig. 2. Rheological models for Equation a) (17) and b) (18).

Khosla [3].

The creep functions in Equations (16) are expressed by the following three equations

$$J_i = a_{oi} - a_{1i}e^{-\alpha_i t} \qquad i = 1,2,3 \tag{20}$$

$$J_i = b_{oi} - b_{1i}e^{-\beta_{1i} t} - b_{2i}e^{-\beta_{2i} t} \qquad i = 1,3 \tag{21}$$

$$J_i = c_{oi} + c_{1i}t^{n_i} \qquad i = 1,2,3 \tag{22}$$

Formulae (20) and (21), which involve exponential functions in the second and third order terms, have earlier been presented by Jumppanen in a more complete form [6 , p. 25]. Expression (22) has been previously applied in studies that include [2], [4], [7], [10] and [11]. In some of them, the second order term $J_2(t,t)\sigma_o$ (14) has been taken into account.

The same types of creep kernel functions are also applicable to two- and three-dimensional constitutive equations. However, in these cases Expression (10) assumes a more complicated form, and the determination of material constants calls for a large number of complex creep tests [5], [6], [9], [10].

3. EXPERIMENTS

The experiments were performed in the Department of Civil Engineering, at Helsinki University of Technology. The materials tested were polyvinyl-chlorides of three slightly different types, developed for sewer-pipe materials by UPO Ltd in Finland. Table 1 indicates the material characterization.

Table 1. Materials tested.

Code No	Type	K	$V^{o}C$	σ_f kp/cm²	ε_f %
04	Pulp polymer	60	81.5	569[1)]	138[1)]
05	Suspension polymer	57	81.5	505[1)]	159[1)]
10	Pulp polymer	68	80.2	590[2)]	117[2)]

1) drawing speed of 3.27 mm/min
2) drawing speed of 11.8 mm/min

K is a characteristic value, proportional to the relative viscosity (ISO R 174) and the average molecular weight of material, and V is the transition temperature determined by the Vicat test (ISO R 306) which almost corresponds to transition temperature T_g between the glass-like and rubber-like zones. σ_f is the ultimate strengh, and ε_f the corresponding ultimate strain at a temperature of 25°C.

The creep testing machine employed in this work is illustrated in Fig. 3, and a single specimen tested in Fig. 4. The strains were measured with strain gauges (ε_{max} = 4-5 %), and registered automatically by a data logger. The results obtained were also punched on tape, and drawn by the digital computer HP 2000A. Creep tests under different combinations of constant stress were made only at the room temperature 22°C. The maximal deviations were ±1°C.

Some of the results obtained in the creep tests have been reproduced in Fig. 5-8. The materials PVC 05 and 04 are more clearly nonlinear than PVC 10. The nonlinearity with respect to stress increases markedly with time (Fig. 7 and 8).

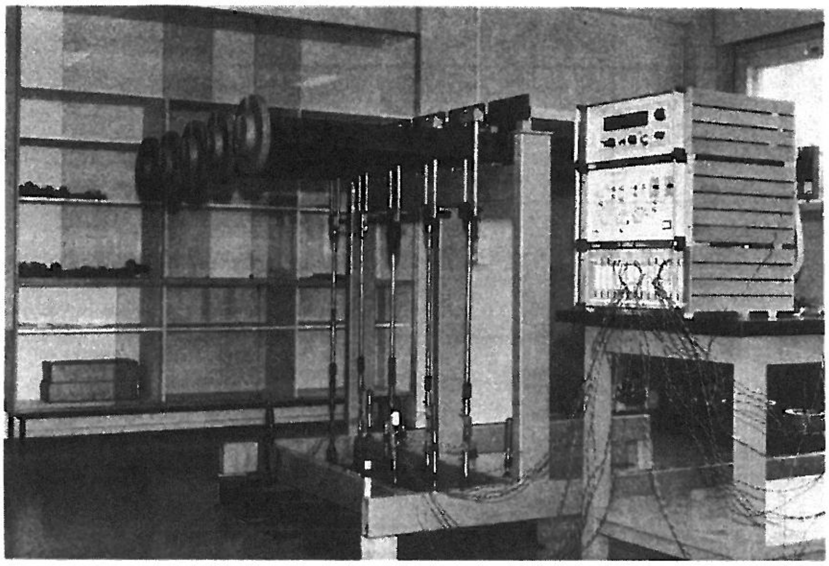

Fig. 3. Testing machine.

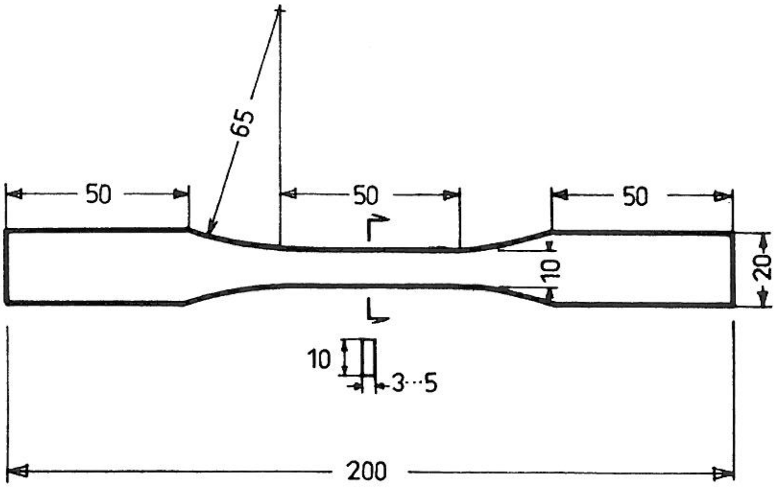

Fig. 4. Single specimen tested.

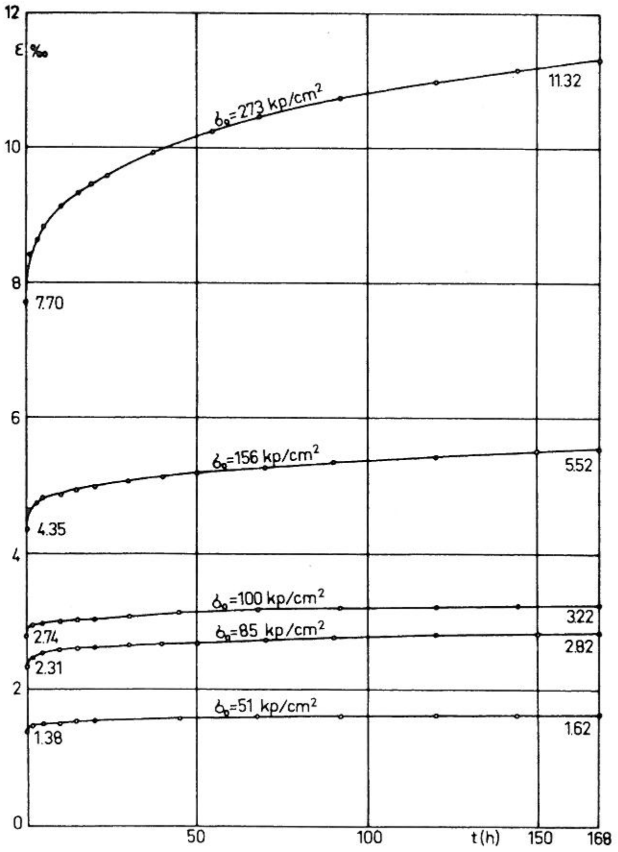

Fig. 5. Creep curves of PVC 04, time range 168 h.

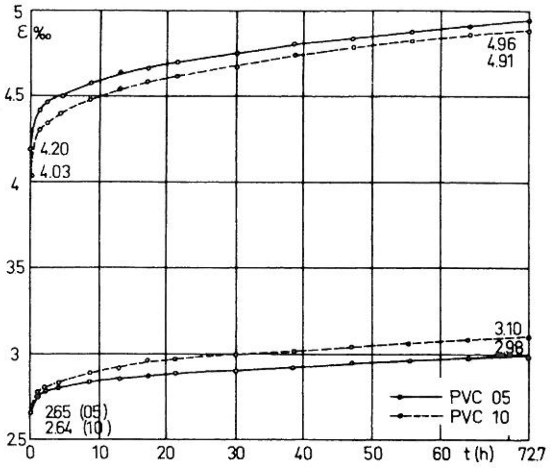

Fig. 6. Creep curves of PVC 05 and PVC 10, time range 72,7 h.

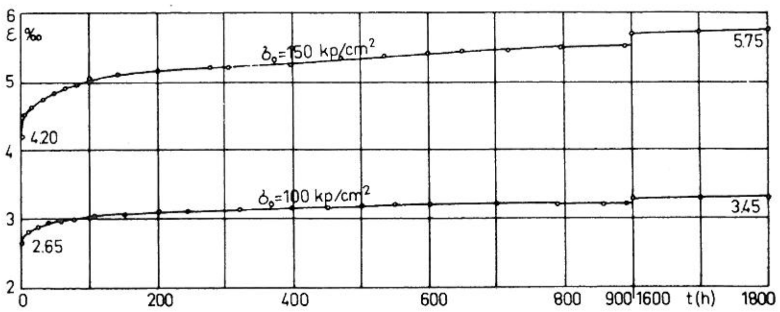

Fig. 7. Creep curves of PVC 05, time range 1800 h.

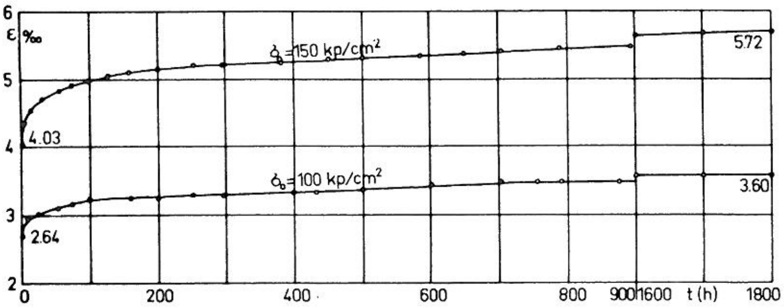

Fig. 8. Creep curves of PVC 10, rime range 1800 h.

4. EVALUATION OF MATERIAL CONSTANTS

For the numerical evaluation of material constants, the least-squares method is employed. In the error functional

$$\Phi = \sum_{i=1}^{r} \left[\overline{J}(t_i) - J_a(t_i) \right]^2 \tag{23}$$

$\overline{J}(t_i)$ is the measured value of creep, and $J_a(t_i)$ the value of an approximative creep function at instant t_i. In the case of linear functions (17) - (19), test values $\overline{J}(t_i)$ have been taken from a creep curve. In the nonlinear cases (20) - (22), the test values have been taken from two or five creep curves, measured at different stress levels. The number of points t_1, t_2, ..., t_r in Equation (23) has been taken to be 14···30 per a creep curve in calculations, so that reliable statistical values have been obtained for material constants. For example, the determination of parameters c_1, c_2, and n in Formula (19) can be effected as follows.

First, three fixed close values are taken for n, and the minimal values of the functional $\Phi(c_1,c_2,n)$ are calculated from the conditions

$$\frac{\partial\Phi}{\partial c_o} = 0, \quad \frac{\partial\Phi}{\partial c_1} = 0 \tag{24}$$

Then, the second order parabola $\Phi(n)$ is put through these three points, and from this the least value of Φ and the corresponding value of n are sought. Furthermore, two close values are taken for this n, and the process is repeated to find a new n. This iterative procedure is repeated as many times as required.

The material constants of $J_1^o(t)$ (17) have been solved in the same way by the use of α in the role of n. This method has also been adopted for creep function (18), by alternate solution of one of the material constants in the exponents. In the nonlinear cases (16), the values of J_1, J_2, and J_3 have been first solved at different instants t_i by the use of two or more creep curves. The material constants of Functions (20), (21), and (22) have been then evaluated by application of the procedure above described. In every case, the covergence of iteration has been good.

For comparison of the applicability of different creep functions, the meen dispersal

$$S = \{\frac{1}{n-1} \sum_{i=1}^{n} [\bar{J}(t_i) - J_a(t_i)]^2\}^{1/2} \tag{25}$$

has been computed for all the solutions. Another suitable measure could be the maximal difference between the tested values and the computed regression curve

$$q = \max_i \left[\bar{J}(t_i) - J_a(t_i)\right] \qquad (26)$$

The numerical values of material constants, s, and q are given in tables 2-7 (for strain ε cm/cm, stress σ_o kp/cm², and time t h).

The initial modulus of elasticity of material at time t = 0 is (cf (6) or (14))

$$E(0) = 1/J(0)$$

For linear creep functions (17) - (19), E(0) obtains the values $1/(a_o - a_1)$, $1/(b_o - b_1 - b_2)$, and $1/c_o$. For nonlinear Equations (20) - (22), the corresponding values are obtainable in the very simple way. The useful values of E(0) for small stresses are obtainable from the linear parts of nonlinear creep equations.

The values of E(∞) (modulus of elasticity in very slow motions) can also be derived simply from Equations (17), (18), (20), and (21). The test period 1800 h is not sufficiently long for the evaluation of E(∞).

Table 2. Material constants of Equation (17) (creep curves in Fig. 5-8)

Material	Stress	Time range	r	$a_o \cdot 10^5$	$a_1 \cdot 10^5$	α	$s \cdot 10^3$	q ⁰/oo
04	51	168 h	14	3,146	0,337	0,0487	0,022	0,04
05	100	72 h	15	2,979	0,253	0,0472	0,026	0,08
10	100	72 h	15	3,069	0,334	0,0658	0,049	0,09
05	100	1800 h	30	3,228	0,444	0,0069	0,060	0,13
10	100	1800 h	30	3,455	0,618	0,0063	0,084	0,20

Table 3. Material constants of Equation (18) (Fig. 5-8)

Material	Stress	Time range	r	$b_o \cdot 10^5$	$b_1 \cdot 10^5$	$b_2 \cdot 10^5$	β_1	β_2	$s \cdot 10^3$	q ‰
04	51	168 h	14	3,153	0,309	0,119	0,0360	15,96	0,008	0,01
05	100	72 h	15	3,057	0,275	0,131	0,0190	1,024	0,004	0,009
10	100	72 h	15	3,132	0,309	0,183	0,0278	0,096	0,008	0,01
05	100	1800 h	30	3,393	0,378	0,273	0,0011	0,026	0,030	0,09
10	100	1800 h	30	3,512	0,545	0,265	0,0033	0,149	0,043	0,08

Table 4. Material constants of Equation (19) (Fig. 5-8)

Material	Stress	Time range	r	$c_o \cdot 10^5$	$c_1 \cdot 10^5$	n	$s \cdot 10^3$	q ‰
04	51	168 h	14	2,721	0,125	0,255	0,012	0,02
05	100	72 h	15	2,654	0,095	0,301	0,007	0,01
10	100	72 h	15	2,639	0,140	0,277	0,009	0,01
05	100	1800 h	30	2,625	0,133	0,229	0,022	0,03
10	100	1800 h	30	2,607	0,184	0,229	0,030	0,05

Table 5. Material constants of Equations (16), (20).

Material →	04		05		10	
Quantity ↓	Eq. 16a	Eq. 16b	Eq. 16a	Eq. 16a	Eq. 16a	Eq. 16a
Time range	168 h	168 h	72 h	1800 h	72 h	1800 h
r	70	70	30	60	30	60
$a_{o1} \cdot 10^5$	3,151	3,013	2,741	2,875	2,953	3,323
$a_{11} \cdot 10^5$	0,313	0,259	0,156	0,263	0,238	0,548
α_1	0,052	0,047	0,040	0,0079	0,049	0,0049
$a_{o2} \cdot 10^7$	-	0,168	-	-	-	-
$a_{12} \cdot 10^7$	-	0,144	-	-	-	-
α_2	-	0,571	-	-	-	-
$a_{o3} \cdot 10^9$	0,133	0,081	0,240	0,355	0,111	0,145
$a_{13} \cdot 10^9$	0,104	0,084	0,098	0,183	0,107	0,094
α_3	0,016	0,017	0,060	0,0057	0,400	0,0035
$s \cdot 10^3$	0,094	0,085	0,042	0,073	0,043	0,065
q ‰	0,51	0,30	0,15	0,30	0,12	0,30
Ref.	Fig. 5	Fig. 5	Fig. 6	Fig. 7	Fig. 6	Fig. 8

Table 6. Material constants of Equation (16a), (21) (Fig. 5)

Material →	04		
Quantity ↓	Eq. 16a	Quantity ↓	Eq. 16a
Time range	168 h	$b_{o3} \cdot 10^9$	0,158
r	70	$b_{13} \cdot 10^9$	0,115
$b_{o1} \cdot 10^5$	3,178	$b_{23} \cdot 10^9$	0,022
$b_{11} \cdot 10^5$	0,257	β_{13}	0,0085
$b_{21} \cdot 10^5$	0,182	β_{23}	0,224
β_{11}	0,021	$s \cdot 10^3$	0,040
β_{21}	0,841	q ‰	0,12

Table 7. Material constants of Equations (16), (22)

Material →	04		05		10	
Quantity ↓	Eq. 16a	Eq. 16b	Eq. 16a	Eq. 16a	Eq. 16a	Eq. 16a
Time range	168 h	168 h	72 h	1800 h	72 h	1800 h
r	70	70	30	60	30	60
$c_{o1} \cdot 10^5$	2,734	2,684	2,534	2,517	2,663	2,643
$c_{11} \cdot 10^5$	0,141	0,098	0,058	0,083	0,042	0,096
n_1	0,228	0,250	0,290	0,221	0,438	0,228
$c_{o2} \cdot 10^7$	-	0,065	-	-	-	-
$c_{12} \cdot 10^7$	-	0,051	-	-	-	-
n_2	-	0,172	-	-	-	-
$c_{o3} \cdot 10^9$	0,0179	-0,0042	0,119	0,109	-0,008	-0,012
$c_{13} \cdot 10^9$	0,0108	0,0042	0,033	0,048	0,067	0,085
n_3	0,459	0,595	0,317	0,242	0,172	0,102
$s \cdot 10^3$	0,038	0,037	0,008	0,025	0,012	0,043
q ‰	0,11	0,10	0,02	0,07	0,03	0,11
Ref.	Fig. 5	Fig. 5	Fig. 6	Fig. 7	Fig. 6	Fig. 8

5. CONCLUSIONS

Tables 2-4 indicate that the linear creep Functions (18) and (19) very well described the creep curves tested. The three-parametric Function (17) is somewhat weaker than the other two.

The application of the creep Functions (21) and (22) in the nonlinear Equations (16) gave satisfactory results (tables 6 and 7), while the results given by Functions (16), (20) were less satisfactory (table 5). The comparison between Equations (16a) and (16b) shows that the second order term $J_2(t,t)$ can be ignored in the case of materials investigated.

The material constants α_{ij}, β_{ij}, and n_i were quite labile in the nonlinear creep functions. In practise average constant values could be taken as these parameters (Onaran and Findley [10],[11]), and the material constants could be then solved from Conditions (24).

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