

A viscoelastoplastic approach to model nonlinear polymeric materials

André Kühl¹, Maria A. Polak¹

¹Dept. of Civil Engineering, University of Waterloo 200 University Avenue West, Waterloo/Ontario, Canada somebody1@somewhere.edu, somebody2@elsewhere.com

Abstract. This work presents a new viscoelastoplastic model to describe the nonlinear behavior of polymers. In this paper, an extension of the results published by Kühl et al. [1] and Kühl and Muñoz-Rojas [2] is developed, considering viscoelastic and viscoplastic strains simultaneously. To model the viscoelastic strain, a master curve approach is employed, as proposed by Kühl and Muñoz-Rojas. To account for the viscoplastic deformation, a power law is used, as detailed by Kühl [3]. Therefore, a final viscoelastoplastic model is defined to predict the behavior of polymeric materials. The procedure proposed in this work is compared to the traditional interpolation of properties, and a discussion of the results is provided.

Keywords: polymers, viscoelasticity, viscoplasticity, master curve

1 Introduction

Polymers have been used for many engineering applications, due to their excellent properties, low cost manufacture and worldwide employability and interchangeability. Those advantages make those materials of great interest for the industry and the society. One application of polymers is in pipelines and high density polyethylene (HDPE) is the most widely used thermoplastic material for this purpose [4]. Nevertheless, a new set of knowledge is necessary when dealing with these kinds of structures. Considering the combination of the properties of fluids and solids present in polymers, it is of fundamental importance to account for those characteristics when proposing an idea to describe the behavior of these materials. Nowadays, literature includes models and procedures which can help model viscoelasticity, although, the experimental limitations and mathematical complexity may bring practical difficulties in solving the problem (correctly describing the behavior of polymers). Therefore, straightforward and uncomplicated methods have arisen and allow characterizing the material response easily and with favorable accuracy. Based on this scenario, novel techniques can be found, such as the interpolation of rheological properties proposed by Liu [5] for viscoelastic parameters and extended by Kühl et al. [1] for plasticity and a master curve scheme developed by Kühl and Muñoz-Rojas [2]. HDPE exhibits nonlinear behavior, which means the parameters of this material change with the applied stress. Thus, the models used to represent the experimental behavior must propose a way to include the variation of the coefficients with the load. The two methods cited above, interpolation of rheological properties and the master curve scheme, present, each one in a different way, a whole procedure to incorporate this dependency in the analysis. In the next section, a summary about these procedures will be discussed.

2 Material model review

Several papers were published in the last decades proposing constitutive models to account for the nonlinear behavior of materials. Zhang and Moore [6] investigated the nonlinear behavior of PE and presented a formulation of the material parameters as a function of stress by using phenomenological time-dependent models. Time-dependent behavior of HDPE was studied by Beijer and Spoormaker [7] using integral

formulation. Bae and Cho [8] developed several VE constitutive equations to characterize the nonlinear viscoelastic behavior. Phenomenological creep modeling was proposed by Lu et al. [9] for ABS pipes by including stress and temperature levels in the formulation. However, none of the works above presented a viscoelastoplastic formulation and quantifies the elastic and permanent strains. As introduced above, this work is a discussion and a comparison between two different approaches to model the nonlinear behavior of polymers. The paper published by Kühl et al. [1] details the viscoelastic and viscoplastic strains that are developed during a creep-recovery test. During this test, the stress is kept constant during 24h and after this period, the load is removed (Figure 1). If the strain goes to zero after the unloading, this quantity is purely viscoelastic. On the other hand, if the strain is not fully recovered (a non-zero amount is observed) the total strain is composed by a viscoelastic and viscoplastic [10]. To account for the viscoelastic strain and the nonlinearity of this quantity, the interpolation of rheological coefficients and the master curve approach are employed and compared here. The viscoplastic formulation is based on the work developed by the authors and cited previously and remains unchanged (an equation based on a power law is employed).



Figure 1. Creep-recovery tests. $\varepsilon_c(t)$ is the creep phase, $\varepsilon_c(t_r)$ represents the accumulated viscoplastic strain and $\varepsilon_r(t)$ is the recovery strain.

The procedure introduced by Liu [5] identifies the material parameters at each stress level tested (the model follows the Kelvin-Voigt arrangement) and interpolates the coefficients for the levels which are not tested. For example, creep tests were conducted at 4 and 6 MPa, after the obtaining of the set of the parameters for each stress level, it is possible to find the behavior of the material at 5 MPa calculating the coefficients by interpolating the ones determined at 4 and 6 MPa. The main concern of the interpolation process is that the parameters have to follow a certain tendency of behavior. If the variation of them is extreme and there is no observed rule in them (for example monotonicity), the model may not proper describe the real response of the polymer. The master curve approach, proposed by Kühl and Muñoz-Rojas [2] present a different concept in modeling the nonlinear viscoelasticity. This method is based on finding local master curves and fitting polynomials to each pair of creep curve. In sequence, another polynomial is fitted to these previous ones. The stress dependency is found by inserting splines into the strain-time equation. The main difference between the two methods is that the interpolation evaluates the local response of each test and the master curve analyzes the set of experimental data together. Equation (1) shows the total strain and Eqs. (2) and (3) defines the models used by Kühl [3] and by Kühl and Muñoz-Rojas [2] (now including the viscoplastic part to the master curve developed by them for viscoelastic analysis), respectively.

$$\varepsilon(t) = \varepsilon^{ve}(t) + \varepsilon^{vp}(t) \tag{1}$$

$$\varepsilon(t) = \left[\frac{1}{E_0} + \sum_{i=1}^{nrb} \frac{1}{E_i} \left(1 - e^{-\frac{t}{\tau_i}}\right)\right] \sigma + kt^n$$
(2)

$$\varepsilon(t) = \left\{ A \left[\ln \left(10^{a\sigma^3 + b\sigma^2 + c\sigma + d} t \right) \right]^4 + B \left[\ln \left(10^{a\sigma^3 + b\sigma^2 + c\sigma + d} t \right) \right]^3 + \dots \right\} \sigma + kt^n$$
(3)

The viscoelastic material parameters for the Kelvin-Voigt approach (Eq. 2) are the E_0 , E_i and τ_i . For the master curve, the viscoelastic variables are the polynomial coefficients (*A*, *B*, *C*, etc). The viscoplastic parameters are the same for both cases, *k* and *n*. The material parameters from Eq.(2) E_0 , E_i , *k* and *n* are interpolated (equations developed by Liu [4] and Kühl [3]). The parameter τ_i is defined a priori as $\tau_1 = 500$ s, $\tau_2 = 10000$ s and $\tau_3 = 20000$ s [4]. For Eq. (3), only the coefficients *k* and *n* are interpolated, since the master curve (for the viscoelastic part) does not comprise this type of calculus.

The procedure to obtain the viscoelastic and viscoplastic parameters for the interpolation method can be summarized below:

- 1. Creep tests are conducted for different stress levels $(\sigma_{1,...,\sigma_{n}})$;
- 2. The elastic parameter E_0 is identified from experimental data using the following equation:

$$E_0 = \frac{\sigma}{\varepsilon} \tag{4}$$

where ε is the measure of the strain for one minute. The stress is the one applied to the test.

3. The viscoelastic coefficients E_i ($J_i=1/E_i$) are identified from the recovery curve applying the expression:

$$\mathcal{E}_{r}(t) = \mathcal{E}_{c}(t_{r}) - \left\{ J_{0}\sigma + \sum_{i=1}^{nrb} J_{i}\sigma \left[\exp\left(\frac{t_{r}}{\tau_{i}}\right) - 1 \right] \left[\exp\left(-\frac{t_{r}}{\tau_{i}}\right) - \exp\left(-\frac{t_{r}}{\tau_{i}}\right) \right] \right\}$$
(5)

in which τ_i is defined a priori (more details see Kühl [3]). The whole formulation can be found in [3] also. The viscoelastic parameters provided in this step can be substituted in Eq. (2) to describe the viscoelastic behavior of the material.

4. An approximation of the viscoplastic strains is obtained by subtracting the experimental creep information from the viscoelastic curve given by the material parameters found in the steps 1 and 2.

$$\mathcal{E}_{vp}(t) = \mathcal{E}(t)_{exp} - \mathcal{E}_{ve}(t)_{num} \tag{6}$$

Employing the natural logarithmic in both sides of the power law, the result is

$$\ln\left[\varepsilon_{vp}(t)\right] = \ln(k) + n\ln(t) \tag{7}$$

And the viscoplastic parameters k and n can be found using the Eq. (7) above.

For the master curve, the viscoelastic coefficients are obtained using the viscoelastic data found in step 3. Equation (3) is employed to describe the viscoelastic strains and the material parameters associated to the master curve are identified. The viscoplastic step is the same for both cases.

For the master curve method, the steps applied to obtain the viscoelastic parameters are:

- 1. Obtain local master curves for each pair of creep curves. Those master curves are developed by using polynomials.
- 2. A global master curve is found by fitting a high-order polynomial to the local master curves obtained in the step 1.
- 3. The shifts (amount each curve is moved to build the master curves) are related to the stress levels using splines.
- 4. The function that relates the shifts and the stress is introduced to the global master curve providing an expression that is able to describe the viscoelastic behavior (given by Eq. (3)).

Finally, the viscoelastic and viscoplastic material parameters are interpolated using the equations below (for the master curve, only the viscoplastic parameters are interpolated, the viscoelastic part is given by the master curve procedure). For the viscoelastic parameters, the equations used for interpolation are:

$$E_0(\sigma) = E_0(\sigma_m) + \frac{\sigma - \sigma_m}{\sigma_n - \sigma_m} \left[E_0(\sigma_n) - E_0(\sigma_m) \right]$$
(8)

where σ_m and σ_n are the stresses corresponding to step 1, such that $\sigma_m < \sigma < \sigma_n$. According to Muñoz-Rojas et al. [11] the following expression is developed to interpolate the E_i parameters:

$$E_i(\sigma) = E_i(\sigma_m) + \frac{\sigma - \sigma_m}{\sigma_n - \sigma_m} \left[E_i(\sigma_n) - E_i(\sigma_m) \right]$$
(9)

Kühl [3] expanded the interpolation above to the viscoplastic parameters. Therefore, the equations used for this part of the formulation are:

$$y(\sigma) = \ln(k(\sigma_m)) + \frac{|\sigma| - \sigma_m}{\sigma_n - \sigma_m} [\ln(k(\sigma_n)) - \ln(k(\sigma_m))]$$
(10)

$$k(\sigma) = \exp(y(\sigma))\left(\frac{\sigma}{|\sigma|}\right)$$
(11)

$$n(\sigma) = n(\sigma_m) + \frac{|\sigma| - \sigma_m}{\sigma_n - \sigma_m} [n(\sigma_n) - n(\sigma_m)]$$
(12)

3 Experimental set-up

Experimental tests used for this research were conducted by Kühl [3]. HDPE creep-recovery tests were performed at State University of Santa Catarina at different stress levels. The samples were extracted from pipes and the dimensions follow the ASTM D638 standard. Figure 2 shows the specimen employed for the test.



Figure 2. HDPE specimen employed for creep-recovery test

4 Results

First, the viscoelastic creep phase of the test (not including the viscoplastic part) is modeled using the master curve approach (this part, using multi-Kelvin approach was developed by Kühl before). The results can be seen in Fig. 3. The procedure to uncouple the viscoelastic and viscoplastic strains was already provided by Kühl in 2014 and the same method is used here. In sequence, the viscoelastoplastic models are used to describe the total strain of the experimental tests. Figure 4 illustrates this second case (the recovery does not change from what was published by the authors in previous papers). Finally, stress rate test at three different rates are analyzed and the two approaches are compared. The results for this last case are depicted in Fig. 5, Fig. 6 and Fig. 7.



Figure 3. Viscoelastic creep response for the master curve model.



Figure 4. Viscoelastoplastic creep-recovery response of the master curve and the interpolation procedure.



Figure 5. Stress rate response of the master curve and the interpolation procedure for 1 MPa/s



Figure 6. Stress rate response of the master curve and the interpolation procedure for 0.1 MPa/s



Figure 7. Stress rate response the master curve and the interpolation procedure for 0.01 MPa/s

The material parameters used to apply the interpolation procedure are the same ones obtained from Kühl [3]. The viscoelastoplastic formulation using master curve and power law, proposed in this work, presented the following viscoelastic and viscoplastic coefficients (the viscoplastic coefficients remained unchanged and came from Kühl [3]). Table 1 shows those material parameters for this model.

Constitutive	A	В	С	D
relation/Parameter				
Viscoelastic model	1.18×10^{-8}	8.80 x10 ⁻⁶	1.058×10^{-4}	$7.55 \text{ x}10^{-4}$
Viscoplastic model	k	п		
1.8 [MPa]	-	-		
3.6 [MPa]	$2.33 \text{ x} 10^{-21}$	3.68		
5.5 [MPa]	2.90 x10 ⁻⁷	0.81		
7.2 [MPa]	1.34 x10 ⁻⁹	1.32		
9.1 [MPa]	4.97 x10 ⁻¹⁴	2.26		
11 [MPa]	3.23 x10 ⁻¹⁵	2.51		
13 [MPa]	2.03 x10 ⁻¹⁴	2.41		

Table 1. Viscoelastoplastic coefficients for the master curve approach.

The relation between the shifts and the stress levels are given by splines (as detailed by Kühl and Muñoz-Rojas, 2019). Table 2 shows the splines functions found for those creep results.

Spline	Stress [MPa]	Piecewise cubic polynomials of the spline function
segment		
<i>j</i> = 1	σ < 5.5	$\psi_1 = -1.9424 + 0.5396\sigma - 0.03941(\sigma - 3.6)^3$
<i>j</i> = 2	$5.5 \le \sigma < 7.2$	$\psi_2 = 0.1346 + 0.1128\sigma - 0.2246(\sigma - 5.5)^2 + 0.1343(\sigma - 5.5)^3$
<i>j</i> = 3	$7.2 \le \sigma \le 9.1$	$\psi_3 = -2.7411 + 0.5137\sigma + 0.4604(\sigma - 7.2)^2 - 0.1547(\sigma - 7.2)^3$
<i>j</i> = 4	9.1≤ σ < 11	$\psi_4 = -1.9983 + 0.4985\sigma - 0.4679(\sigma - 9.1)^2 + 0.1213(\sigma - 9.1)^3$
<i>j</i> = 5	otherwise	$\psi_5 = 2.7594 - 0.007493\sigma + 0.1868(\sigma - 11)^2 - 0.03114(\sigma - 11)^3$

Table 2. Spline equations describing the shift-stress relation.

As shown by the results, the viscoelastoplastic model described in this paper, when using the master curve and the power law, can describe the creep behavior of polymers at different stress levels. In addition to that, the same approach is capable of simulating the physical response of polymeric materials in a realistic way. The comparison between the master curve and the interpolation procedure proves that both methods can predict the nonlinear creep behavior properly (the latter one is slightly better, since the analysis is developed curve by curve). Nonetheless, the interpolation of properties presents limitations and it is highly dependent on the values of the material parameters, and this approach may not represent the physical behavior of polymers adequately to describe the stress rate tests.

5 Conclusions

This paper has presented an extension of the viscoelastic model described before in literature. Thus, a viscoelastoplastic formulation is shown to explain the behavior of polymeric materials. According to the results, it was possible to observe that this viscoelastoplastic approach can represent the creep tests as well as the interpolation procedure. Notwithstanding, the model composed by the master curve and the power law reproduces the stress rate cases with more realism than the interpolation of properties. Despite being a practical and simple procedure, the interpolation of properties exhibits some limitations, as mentioned previously. For future analysis, creep-recovery and stress rates tests will be conducted to investigate the formulation proposed with more details.

Acknowledgements. The authors express their sincere gratitude for financial support to Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES), project number 88881.17962/2016-01. Also, the authors would like to thank professor Pablo Andrés Muñoz-Rojas for his enlightened contributions to this work.

Authorship statement. The authors hereby confirm that they are the sole liable persons responsible for the authorship of this work, and that all material that has been herein included as part of the present paper is either the property (and authorship) of the authors, or has the permission of the owners to be included here.

References

[1] A. Kühl, P.A. Muñoz-Rojas, R. Barbieri, and I. J. Benvenutti, "A procedure for modeling the nonlinear viscoelastoplastic creep of HDPE at small strains", *Polymer Engineering and Science*, v. 57, pp. 144-152, 2016.

[2] A. Kühl and P.A. Muñoz-Rojas. "Application of a master curve and the modified superposition principle for modeling creep and loading rate effects on high density polyethylene at small strains". *Mechanics of Advanced Materials and Structures*. 1537-6532, 2019.

[3] A. Kühl, "Uma formulação viscoelastoplástica não-linear aplicada ao polietileno de alta densidade (PEAD) ". Master Thesis, Universidade do Estado de Santa Catarina, Brazil, 2014.

[4] B. Ma, M. Najafi, G. Jiang, and L. Slavin, "Collapse of RTP (reinforced thermoplastic pipe) subjected to external pressure." *ICPTT 2012: Better Pipeline Infrastructure for a Better Life*", American Society of Civil Engineers, pp. 725–743, 2012.

[5] H. Liu, "Material Modeling for Structural Analysis of Polyethylene", PhD Thesis, University of Waterloo, Canada, 2007.
[6] C. Zhang and I. D. Moore, "Nonlinear mechanical response of high density polyethylene. Part I: Experimental investigation and model evaluation", *Polymer Engineering and Science*, v. 37, pp. 404-413, 1997.

[7] J. G. J. Beijer and J. L. Spoormaker, "Modeling of creep behavior in injection-moulded HDPE". *Polymer*, v. 41, pp. 5443-5449, 2000.

[8] J.E. Bae, and K.S. Cho, "Semianalytical methods for the determination of the nonlinear parameter of nonlinear viscoelastic constitutive equations from LAOS data", *Journal of Rheology*, vol. 59, pp. 525–555, 2015.

[9] J. P., Lu, L. S., Burn and B. E., Tiganis, "Creep modeling of ABS pipes at variable temperature". *Polymer Engineering and Science*, v. 40, pp. 2407-2413, 2000.

[10] J. Mui, "Viscoelastic-viscoplastic model to predict creep in random chopped mat thermoplastic composite", University of Waterloo, Canada, 2008.

[11] P. A. Muñoz-Rojas, A. Kühl, P. T. R. de Mendonça, I. J. Benvenutti and G. J. Creus. "Modeling nonlinear viscoelastic behavior of high density polyethylene (hdpe): application of stress-time equivalence versus interpolation of rheological properties". *International Symposium on Solid Mechanics*, Florianópolis, 2011.