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# LINEARIZATION AND IMPLEMENTATION OF VENU MODEL IN SMALL STRAIN THEORY FOR POLYAMIDE 6.6

D.S. Anagnostou[1], G. Chatzigeorgiou[1], J.L. Bouvard[2], Y. Chemisky[1], F. Meraghni[1], N. Billon[2]

[1]LEM3-UMR 7239 CNRS, Arts et Métiers ParisTech Metz-Lorraine, 4 Rue Augustin Fresnel 57078 Metz, France
Emails: dimitrios.anagnostou@ensam.eu, georges.chatzigeorgiou@ensam.eu, yves.chemisky@ensam.eu, fodil.meraghni@ensam.eu
[2]MINES ParisTech, CEMEF-Centre de Mise en Forme des Matériaux, CNRS UMR 7635, BP 207, 06904 Sophia Antipolis Cedex, France
Emails: jean-luc.bouvard@mines-paristech.fr, noelle.billon@mines-paristech.fr

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**Abstract**

The so-called VENU[1] model is a visco-hyperelastic constitutive model, designed for amorphous rubbery polymers, which is based on an original approach initially developed by N. Billon (J. Appl. Polym. Sci. 125:4390-4401, 2012) and extended by A. Maurel-Pantel et al. (Int. J. Plast. 67:102126, 2015) to three-dimensional thermomechanical framework. In the aforementioned references, the constitutive equations and thermodynamical framework are presented within large deformation theory. However, in fatigue tests of polymeric composites significant temperature gradients are noticed despite the fact that the measured strains are within the small strain theory. In addition, well established techniques and tools of micromechanics for polymeric composites are applicable in small deformation regions. These observations render important the reduction of the VENU model in the case of linear strains. Here, a method is proposed for the reduction of the VENU model to small strain theory. A proper numerical scheme is also provided, based on the so-called return-mapping algorithm. The model capabilities are illustrated by comparing numerical calculations with available experimental data for polyamide 66.

## 1. Introduction

New economic data, such as the reduction of world oil reserves, force transport fields to find quick solutions in order to reduce the fuel consumption and $CO_2$ emissions from future vehicles. This makes necessary the use of new materials combining lightness and strength. To this end, amorphous and semi-crystalline polymers are widely used in vehicle industry due to their physical, optical (light transparency), and mechanical properties (toughness). In addition, research in automative industry aims at reducing the need of expensive mechanical tests. Thus the increasing interest for accurate and reliable theoretical models that adequately predict the material behavior and dependence upon time, temperature and loading. In particular, semi-crystalline polymers, such as polyamide 6.6, are well known to exhibit a rate and temperature dependent behavior. With the increase interest for this kind of materials in the automotive industry, a large number of material models were developed in the literature to predict their material

[1]VENU stands for Visco-hyper Elastic Network Unit.

response (e.g., [1–7] and references cited therein).

Here, attention is focused to one particular theoretical model that captures the thermo-mechanical behavior of semi-crystalline polymers; namely the VENU model. The initials stand for visco-hyperastic network unit and as the very name implies, the model is based on the material network desription. VENU model is a visco-hyperelastic constitutive model, based on an an original approach by Billon, [8], initially developed as one-dimensional formulalism and further extended by Maurel-Pantel et al., [7], to three-dimensional thermomechanical framework. It should be noted that very few studies have focused on the full thermomechanical (i.e., non-isothermal) modeling of semi-crystalline polymers. The model accounts for chains network reorganization under external loading through the introduction of an evolution equation for the internal state variable, representing the degree of mobility of the entanglement points. The model captures the visco-elastic behavior of the material, the different stress states (tension and shear), the thermomechanical coupling observed under large deformation, and the material self-heating under large deformation. The thermomechanical model agrees well with the experimental mechanical and temperature measurements under tension and shear conditions. The developed approach may thus open a different way to model the polymer behavior.

In [7] the constitutive equations and thermodynamical framework are presented within large deformation theory. However, in fatigue tests of polymeric composites significant temperature gradients are noticed despite the fact that the measured strains are within the small strain theory. In addition a linearized version of the model permits the application of well established techniques and tools of micromechanics (see, among others, [9–13]). These observations render important the reduction of the VENU model in the case of linear strains. In this note, a method is proposed for the reduction of the VENU model to small strain theory. The method is based on rewriting the Cauchy stress tensor of the original model, in terms of the infinitesimal elastic strain tensor through appropriate series expansions and discarding terms other than the linear ones. Further, the governing equations of the model are reduced in their small-strain counterparts. Three main kinematic assumptions are made regarding the VENU model: (i) the material is incompressible; (ii) the flow is incompressible and (iii) the flow is irrotational. These assumptions still hold in the linearized version of the model. After deriving the small strain theory version of VENU model we provide a numerical scheme for the proper implementation of it. This scheme is based on the so-called *return-mapping algorithm* [14]. This is a robust algorithm which was used for a multitude of applications (see, e.g., [6, 15–18]). Finally, some results from the application of the numerical scheme are compared with the respective experimental results for the infinitesimal regime, available in [7].

## 2. Formulation

According to Billon [8] the Edward-Vilgis free energy of the polymer can be decomposed into two components: (i) the energy due to the polymer network deformation constrained by permanent nodes (crosslinks) and (ii) the energy due to the polymer network deformation constrained by slip links such as entanglement points. In particular, the Edward-Vilgis free energy used in the VENU model reads

$$w\left(I_1^e, I_2^e, I_3^e, \eta, T\right) = (1/2)\left[N_c^* w_c\left(I_1^e, I_2^e, I_3^e\right) + \ N_s^* \, w_s\left(I_1^e, I_2^e, I_3^e, \eta\right)\right] , \tag{1}$$

with $T$ being the absolute temperature and $\eta$ denoting the scalar internal state variable (ISV) representing the degree of mobility of the entanglement points. In addition, $N_c^*$ is related to the density per unit volume of crosslinking, $N_s^*$ is related to the density per unit volume of entanglement points; in general

these parameters are temperature dependent. Moreover,

$$\begin{cases} w_s\left(I_1^e, I_2^e, I_3^e, \eta\right) = & \dfrac{(1+\eta)\left(1-\alpha^2\right)}{\left(1-\alpha^2 I_1^e\right)} \dfrac{\left(I_1^e + 2\eta I_2^e + 3\eta^2 I_3^e\right)}{\left(1+\eta I_1^e + \eta^2 I_2^e + \eta^3 I_3^e\right)} + \ln\left[\left(1-\alpha^2 I_1^e\right)\left(1+\eta I_1^e + \eta^2 I_2^e + \eta^3 I_3^e\right)\right] \\ w_c\left(I_1^e, I_2^e, I_3^e\right) = & w_s\left(I_1^e, I_2^e, I_3^e, 0\right) = \frac{(1-\alpha^2)I_1^e}{(1-\alpha^2 I_1^e)} + \ln\left[\left(1-\alpha^2 I_1^e\right)\right] \end{cases} . \tag{2}$$

In the above expressions $\alpha$ is the limit of chain extensibility. Also, $\{I_1^e, I_2^e, I_3^e\}$ is the standard set of three independent invariants of the elastic Right and Left Cauchy-Green deformation tensors, [19].

Using standard arguments from the seminal work [20], Cauchy stress tensor is written as

$$\boldsymbol{\sigma} = 2(J^e)^{-1}\left[\left(\frac{\partial w}{\partial I_1^e} + \frac{\partial w}{\partial I_2^e} I_1^e\right)\mathbf{B}^e - \frac{\partial w}{\partial I_2^e}(\mathbf{B}^e)^2 + \frac{\partial w}{\partial I_3^e} I_3^e \mathbf{I}\right] . \tag{3}$$

where $\mathbf{C}^e$ is the elastic Left Cauchy-Green deformation tensor. Further, for infinitesimal displacement gradients, taking into account that $\mathbf{B}^e \cong 1 + 2\boldsymbol{\varepsilon}^e$, with $\boldsymbol{\varepsilon}^e$ being the elastic part of the infinitesimal strain tensor[2], (Eq. 3) can be seen to reduce to

$$\boldsymbol{\sigma} = 2(J^e)^{-1}\left[\left(\frac{\partial w}{\partial I_1^e} + 2\left(1+\mathrm{tr}(\boldsymbol{\varepsilon}^e)\right)\frac{\partial w}{\partial I_2^e} + \left(1+2\mathrm{tr}(\boldsymbol{\varepsilon}^e)\right)\frac{\partial w}{\partial I_3^e}\right)\mathbf{I} + 2\left(\frac{\partial w}{\partial I_1^e} + \frac{\partial w}{\partial I_2^e}\right)\boldsymbol{\varepsilon}^e\right] . \tag{4}$$

Next, we express the derivatives of the potential as a function of invariants in a linear form. For this, we first take a Taylor series expansion around the null deformation, i.e. around the values of invariants $\left\{I_1^e = 3,\ I_2^e = 3,\ I_3^e = 1\right\}$ and then replace the invariants by their expressions in the first order. The various involved derivatives were evaluated using the computer algebra system *Mathematica*®.

Finally, the Cauchy stress tensor of the reduced model for an incompressible isotropic viscoelastic material in the case of infinitesimal displacement gradients (i.e. $J^e = 1$ ) is written in the following form

$$\boldsymbol{\sigma} = -p\mathbf{I} + 2\left(N_c^* w_1^c + N_s^*\left(w_1^s + w_2^s\right)\right)\boldsymbol{\varepsilon}^e , \tag{5}$$

where $p$ is a pressure term that must be determined from the equilibrium equations and the boundary conditions of the problem. Note that, as $p$ is undetermined from any constitutive equation the terms multiplying $\mathbf{I}$ may be absorbed into $p$ , and this was done implicitly in writing Eq. (5). This is a common practice in incompressible continuum theories, see [19]. Eq. (5) is similar in form with its large deformation theory counterpart (cf. Eq. (29) of [7]).

In order to facilitate the subsequent numerical analysis and the application of the return mapping algorithm we rewrite Cauchy stress tensor, as follows

$$\boldsymbol{\sigma} = \left(K - \frac{2G}{3}\right)\mathrm{tr}\boldsymbol{\varepsilon}^e\,\mathbf{I} + 2\mathrm{G}\,\boldsymbol{\varepsilon}^{\mathrm{e}} , \tag{6}$$

with

$$\begin{cases} G \equiv N_c^* w_1^c + N_s^*\left(w_1^s + w_2^s\right) \\ w_1^s = \frac{2\alpha^2}{\left(1-3\alpha^2\right)^2} + \frac{1-\eta-\alpha^2(1+\eta)}{(1-3\alpha^2)(1+\eta)^3} \ , \quad w_2^s = \frac{2\eta\left(\alpha^2(\eta+1)-1\right)}{(3\alpha^2-1)(\eta+1)^3} , \quad w_1^c = w_1^s\big|_{\eta=0} \end{cases} . \tag{7}$$

where we took into account that for an incompressible linear elastic material holds $\{I_1^e = 3\,, I_2^e = 3\,, I_3^e = 1\}$. Once evaluating the "shear-like modulus" $G$ the analysis proceed with adopting a very high value for

[2]We assume, as is the standard pracice, that the total strain can be decomposed into two parts, an inelastic part $\boldsymbol{\varepsilon}^v$ and an elastic one $\boldsymbol{\varepsilon}^e$.

the "bulk-like modulus" $K$. In particular, for the results of Section 4, we use a value of Poisson's ratio $\nu = 0.49$ and the value of $K$ is provided by the usual relation $K = (2G(1+\nu))/(3(1-2\nu))$.

The evolution equation for the viscoelastic part of the strain tensor after the reduction of the non-linear counterpart reads

$$\dot{\boldsymbol{\varepsilon}}^{v} = \frac{3}{2}\frac{1}{(1-\beta)}\frac{1}{\left\|\boldsymbol{\sigma}^{D}\right\|}\frac{\partial w}{\partial \eta}\dot{\eta}\left(\frac{\boldsymbol{\sigma}^{D}}{\left\|\boldsymbol{\sigma}^{D}\right\|}\right). \tag{8}$$

where $\beta$ is the so-called Taylor-Quinney coefficient accounting for the assumption that a part of the inelastic energy is stored in the chain network and contributes to the internal energy of the system. The norm $\|\cdot\|$ is defined as $\|\cdot\| = \sqrt{(3/2)\cdot:\cdot}$ and $\boldsymbol{\sigma}^{D}$ is the deviatoric part of the Cauchy stress tensor given by

$$\boldsymbol{\sigma}^{D} = \sigma - (1/3)\,\mathrm{tr}\,(\boldsymbol{\sigma})\,\mathbf{I}\,. \tag{9}$$

The evolution equation for the internal state variable $\eta$ is related to the rate of polymer chains network reorganization when submitted to external loading. More specifically, $\eta$ accounts for the disentanglement of the polymer chains. Adopting the evolution equation of [8] and properly modified in the case of small-strain theory we arrive at

$$\Phi = \Omega - \dot{\eta}, \quad \Omega := \Omega(I_1^e, I_2^e, I_3^e, T, \eta) = z_{0p}^{*}\left[\exp\left(z_{p1}[\psi_s - w_s]\right) - 1\right], \tag{10}$$

with $z_{0p}^{*}$, $z_{p1}$ being material parameters which are given in [7]. The functions inside the exponential are defined by

$$w_s = 1 + \frac{2}{1-3\alpha^2} + \ln\left([1-3\alpha^2][1+\eta]^3\right), \quad \psi_s = \frac{K_a}{K_b} + \ln K_b, \tag{11}$$

with

$$K_a = [1-\alpha^2][1+\eta][I_1^{\mathrm{e}} + 2\eta I_2^{\mathrm{e}} + 3\eta^2 I_3^{\mathrm{e}}], \quad K_b = [1-\alpha^2 I_1^{\mathrm{e}}][1+\eta I_1^{\mathrm{e}} + \eta^2 I_2^{\mathrm{e}} + \eta^3 I_3^{\mathrm{e}}], \tag{12}$$

We have to point out that $w_s$ represents the energy of the incompressible material due to slip links in small deformation formalism, while $\psi_s$ represents the same energy in large deformation formalism. In other words, a slightly perturbed version of $\Omega$ is adopted in which the argument of the exponential is the difference of $\psi_s$ minus $w_s$. This expression was assumed in order to avoid the vanishing of $\Omega$ within small strain theory and in the case of an incompressible material. The perturbed version permits the proper evaluation of the derivatives of $\Omega$.

The "linearized" version of the heat equation is derived from the respective non-linear one [7]. In particular,

$$C_V\dot{T} = (\beta + (1-\beta)\,(w_c^{*} + w_s^{*}))\,\sigma : \dot{\varepsilon}^{v} + \frac{1}{2}\,(w_c^{*} + w_s^{*})\,\sigma : \dot{\varepsilon}^{e} + k\Delta T\,, \tag{13}$$

where the evolution of $\varepsilon^{v}$ is provided by Eq. (8). Moreover, $C_V = \rho_0 C_{\mathrm{Th}}$ with $\rho_0$ denoting the density of the material and $C_{\mathrm{Th}}$ its thermal capacity. Also, $k$ is the thermal conductivity. The terms $w_c^{*}$ and $w_s^{*}$ are temperature dependent and they are going to be identified below through the strain rate equivalence technique.

Some of the variables were assumed to be dependent on time $t$ and temperature $T$ by the use of an equivalent strain rate $a_T\dot{\varepsilon}_{eq}$, defined at the reference temperature $T_{\mathrm{ref}}$ . This equivalent stain rate follows classical time-temperature equivalence principle. The notion of equivalent strain-rate is capable of building master curves and therefore decreases the number of testing needed to built a material database. Details are provided in [7]. The main results are summarized below

$$\begin{cases} \lambda = a_T\dot{\varepsilon}_{eq} = a_T\left(d\varepsilon_{eq}\big/dt\right) = d\varepsilon_{eq}\big/(dt/a_T) = d\varepsilon_{eq}\big/dt_{eq} \\ dt_{eq} = dt/a_T\,, \qquad t_{eq} = \int_0^t dt/a_T(t) \end{cases}, \tag{14}$$

where $t_{\mathrm{eq}}$ is the equivalent time at reference temperature $T_{\mathrm{ref}}$; $\dot{\varepsilon}$ is the experimental applied uniaxial strain; $\dot{\varepsilon}_{eq} = \sqrt{(2/3)\,\dot{\boldsymbol{\varepsilon}} : \dot{\boldsymbol{\varepsilon}}}$ is the equivalent strain rate; $a_{\mathrm{T}}$ is the shift factor from the Williams-Landel-Ferry equation

$$\lambda = a_T \dot{\varepsilon}_{eq} = 10^{-\frac{C_1(T-T_{\mathrm{ref}})}{C_2+(T-T_{\mathrm{ref}})}} \dot{\varepsilon}\,. \tag{15}$$

With the help of the strain rate equivalence variable $\lambda$ one can identify the parameters $N_c^*$ and $N_s^*$ as

$$N_c^* = N_{c0}^* + N_{c1}^* \left[ \frac{2}{1 + [\tau\lambda]^{-2m}} \right], \quad N_s^* = N_{s0}^* + N_{s1}^* \left[ \frac{2}{1 + [\tau\lambda]^{-2m}} \right], \tag{16}$$

where $\{N_{c0}^*, N_{c1}^*, N_{s0}^*, N_{s1}^*, m, \tau\}$ are material constants. Moreover

$$w_c^* = \frac{T}{N_c^*} \frac{\partial a_T}{\partial T} \dot{\varepsilon}_{eq} \frac{\partial N_c^*}{\partial \lambda}, \quad w_s^* = \frac{T}{N_s^*} \frac{\partial a_T}{\partial T} \dot{\varepsilon}_{eq} \frac{\partial N_s^*}{\partial \lambda}\,. \tag{17}$$

## 3. Return Mapping Algorithm

The Return mapping algorithm is probably the most popular mean of numerically solving conventional plasticity equations. It is discussed in full detail in [14]. The main point of the algorithm is as follows: *At each quadrature point, given the stress and the internal variables of the previous steps, as well as a specified strain and temperature increment, determine the values of the stress and the internal variables of the current time increment.*The return mapping algorithm used for the numerical implementation of the VENU model consists of three main steps:

i. In the first step, the internal variables $\boldsymbol{\zeta} := \{\boldsymbol{\varepsilon}^v, \eta\}$ of the material do not evolve and only generation of thermoelastic strains are considered (known as *thermoelastic prediction* step). Thus, during this step $\boldsymbol{\zeta}$ is kept fixed, while $\boldsymbol{\varepsilon}$ and $T$ evolve. This part is taken care of by a global solver (for instance a FE software).
ii. In the second step, the error in the stress is corrected by identifying the actual change in the internal variables (known as *inelastic correction* step). Thus, during this step $\boldsymbol{\varepsilon}$ and $T$ are fixed, while $\boldsymbol{\zeta}$ evolves.
iii. In order to check the validity of the correction, appropriate tangent moduli are required. These are computed by applying small, arbitrary perturbations in $\boldsymbol{\varepsilon}$ and $T$, using the instantaneous response obtained from the second step.

Regarding the basic assumptions of the discretization with respect to the time and per iteration, the reader is referred to [21] and the book of Simo and Hughes [14].

## 4. Results and Discussion

The model and material parameters $\{N_{c0}^*, N_{s0}^*, N_{c1}^*, N_{s1}^*, \alpha, z_{p0}, z_{p1}, \beta, m, \tau, \chi, C_1, C_2, T_{\mathrm{ref}}, \eta_{P0}, k, \rho_0, C_{\mathrm{th}}, h\}$ are summarized in (Table 1). These parameters have been obtained by appropriate parameter identification method, using available experimental data. Figures 1 and 2 illustrate the comparison between experimental and numerical results for three different strain rates and almost constant temperature 21.6°C. The result shows a very good fitting between the model and the experimental curves.


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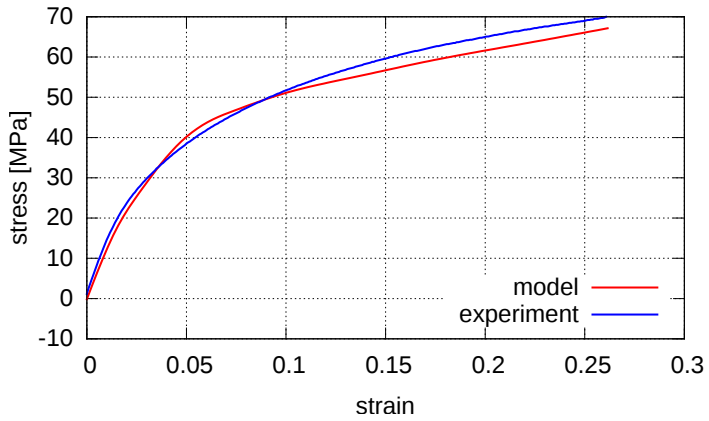


**Figure 1.** Experimental results and model simulation for polyamide 6.6 at strain rate 1 $\times 10^{-3}$ 1/s.

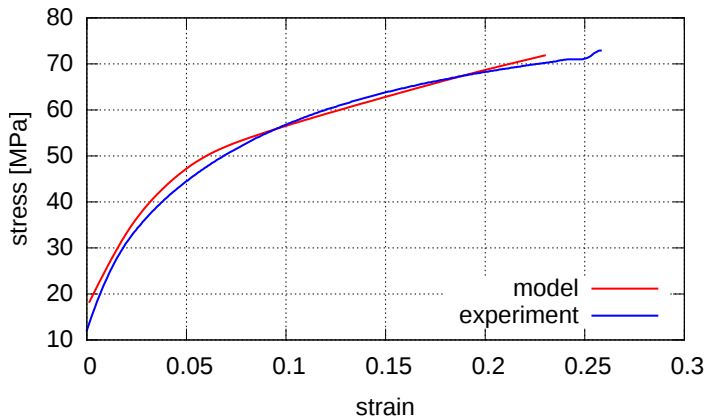


**Figure 2.** Experimental results and model simulation for polyamide 6.6 at strain rate 1 $\times 10^{-2}$ 1/s.

**Table 1.** Material and model parameters

| Parameters | Used values | Units |
|---|---|---|
| $N^*_{c0}$ | 48.6936 | MPa |
| $N^*_{c1}$ | 572.788 | MPa |
| $N^*_{s1}$ | 16.1014 | MPa |
| $N^*_{s2}$ | 18.1213 | MPa |
| $\beta$ | 0.9490 | – |
| $m$; $\tau$ | $3.63 \times 10^{-2}$; 0.7067 | –; s |
| $\zeta_{p0}$ | 5.4599 | – |
| $\zeta_{p1}$ | 1.0 | – |
| $\alpha$; $\chi$ | $8.698 \times 10^{-2}$; $7.522 \times 10^{-1}$ | – |
| $C_1$ | 45.685 | – |
| $C_2$ | 245.06 | °C |
| $T_{\text{ref}}$ | 25 | °C |
| $\eta_0$ | 0.2343 | – |

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