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# A constitutive model for semi-crystalline polymer deformation involving lamellar fragmentation

## *Une loi de comportement pour les polymères semi-cristallins se déformant par fragmentation*

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### A B S T R A C T

The mechanism of lamellar fragmentation in the semi-crystalline polymers with spherulitic structure, is observed at the beginning of plastic flow. It causes significant damage. This elementary mechanism is considered here as a result of plastic deformation coupled with damage, in the framework of generalized standard materials. The simplicity and the efficiency of the proposed approach come from the fact that the semi-crystalline polymers are considered as a macromolecular network bridled by intra-lamellar cohesive forces. Tensile tests and relaxation tests demonstrate the usefulness of a damage-plasticity coupled model.

### R É S U M É

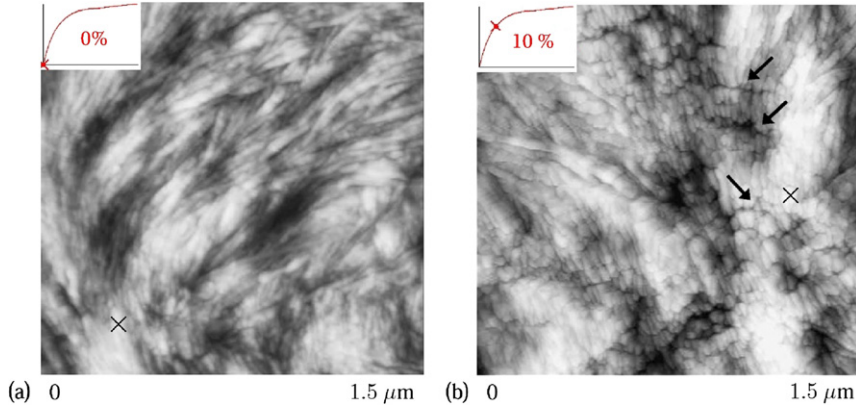
Le mécanisme de fragmentation lamellaire, observé lors de la déformation des polymères semi-cristallins à structure sphérolitique, apparaît et endommage considérablement la microstructure, dès le début de l'écoulement plastique. Dans cette Note, consacrée à la modélisation du comportement de ces matériaux, le mécanisme élémentaire de fragmentation est pris en compte à l'aide d'un couplage plasticité/endommagement. Les développements présentés reposent sur la considération d'un réseau macromoléculaire bridé par les forces cohésives intra-lamellaire. Des essais de traction cyclique et de relaxation, sur le polybutène, valident l'intérêt de l'approche proposée.

## 1. Introduction

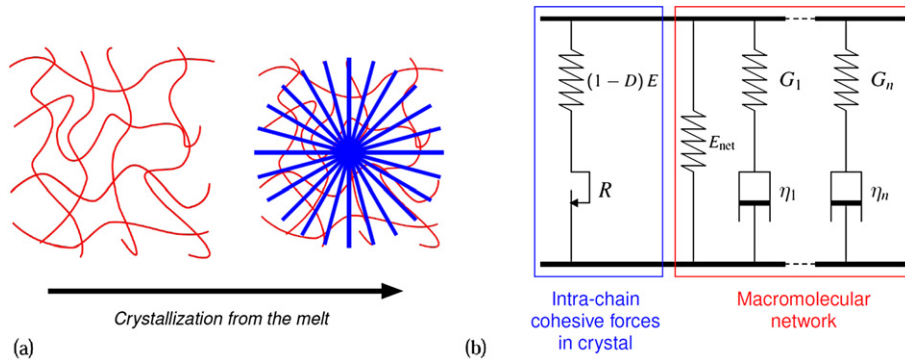
Different models that have been proposed in the literature to describe the mechanical behavior spherulitic semi-crystalline polymers are not suitable for loading/unloading tensile tests. Important coupling between damage and plasticity has not yet been taken into account. The mechanism of the lamellae fragmentation is the key to understand how the plasticity and damage are coupled. In fact, this mechanism has always been overlooked not only in the thermodynamics-based approaches [1] but also in the micromechanical [2,3].

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**Fig. 1.** (a) AFM height image of undeformed PB ( $\Delta l/l_0 = 0\%$ ); (b) AFM height image of deformed PB  $\Delta l/l_0 = 10\%$  ( $Z_{range} = 120$  nm) [9].



**Fig. 2.** (a) Structural model for spherulitic semi-crystalline polymers; (b) rheological model.

The main purpose of this Note is to derive a new, physically based, constitutive model for describing lamellae fragmentation mechanisms that occur during the deformation process of semi-crystalline polymer. This approach is developed within the framework of the generalized standard materials model [4].

The fragmentation mechanism in semi-crystalline polymer corresponds to the splitting into pieces of the crystalline lamellae. The cracks occur perpendicular to the crystal growth direction. *In situ* tensile tests were performed under atomic force microscopy (AFM). Fig. 1a shows the undeformed state of the polybutene (PB) and Fig. 1b shows the evolution of polybutene microstructure under deformation. The lamellae (in white Fig. 1) oriented along the tensile direction cracked during the deformation process (black arrows Fig. 1).

## 2. Mechanism of lamellae fragmentation in semi-crystalline polymers

Classically, plasticity in spherulitic semi-crystalline polymers has been theoretically described using crystal slip [2]. The modelling of damage has scarcely been addressed in consideration through cavitation [5]. It is yet important to point out that mechanical damage in semi-crystalline polymers may appear well before necking, *i.e.* before cavitation can be detected [6].

Furthermore, AFM observations have provided better insight on the lamellae fragmentation process in polypropylene [7] and polyamide 6 [8]. More recently, *in situ* AFM examination of PB under tensile drawing revealed that lamellae fragmentation takes place at early stages of plastic deformation [9] (Fig. 1). This mechanism of gradual destruction of the spherulitic structure is not only accompanied with permanent strain but also with stiffness loss.

We assume semi-crystalline polymers to consist of a macromolecular network bridled by the intra-lamellar cohesive forces. The rheological model shown in Fig. 2 is used to determine the mechanical behavior of semi-crystalline polymers. Starting from a rubber-like macromolecular network in the melt, the structural model (Fig. 2a) assumes that the spherulitic growth of the crystalline lamellae during solidification leads to a bridling of the entangled chain coils. Indeed it is well known that everything macromolecules of the entangled network run across several crystalline lamellae and amorphous layer so that network and crystal structure are deeply interconnected.

Accordingly, the new model represents the material resistance to deformation with two parallel elements: the first one models the intra-lamellar cohesive forces and the second describes the stretching of entropic network (Fig. 2). The specific free energy could then be split into two terms:

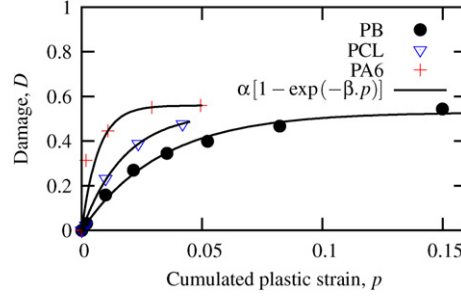


Fig. 3. Damage versus plastic strain for polybutene (PB), polycaprolactone (PCL), polyamide 6 (PA6).

- an energy  $\varphi^{cs}$  of a hyperelasto-plastic type with damage variable  $D$ ;
- an energy  $\varphi^{net}$  of visco-hyperelastic type.

The same separation into two terms for the free energy  $\varphi$  and for the dissipation potential  $\phi$  is applied:

$$\varphi = \varphi^{cs} + \varphi^{net} \quad \text{and} \quad \phi = \phi^{cs} + \phi^{net} \quad (1)$$

where the superscripts  $cs$  and  $net$  denote, respectively, the intra-lamellar cohesive forces in crystalline structure and the entropic network.

The contribution of cohesive forces decreases with an increasing deformation, which is due to lamellae fragmentation. This unbridled effect is taken into account by an isotropic damage, variable  $D$ . Moreover, we have proposed a phenomenological relation coupling damage  $D$  and cumulated plastic strain  $p$  (see Eq. (13)), based on previous experimental investigations on several semi-crystalline polymers [10] (Fig. 3), given by:

$$D = \alpha(1 - e^{-\beta p}) \quad \text{where } \alpha \text{ and } \beta \text{ are material parameters} \quad (2)$$

This law represents the coupling between the stiffness loss and the permanent strain during the lamellar fragmentation process. The strain rate dependence of semi-crystalline polymer is assumed to be only supported by the dissipation in the macromolecular network.

### 3. Cohesive forces in crystalline structure

#### 3.1. Kinematics

For elastic–plastic constitutive description of cohesive forces in the crystalline structure, the deformation gradient tensor  $\mathbf{F} = \mathbf{F}^e \mathbf{F}^p$  is decomposed into a plastic and an elastic component, denoted by the superscripts  $p$  and  $e$ . The arbitrary rotation of this decomposition is eliminated by keeping the microstructure orientation constant in the intermediate configuration [11], i.e. the plastic flow is assumed to preserve the microstructure orientation.

#### 3.2. Thermodynamics potential and state laws

Let  $\mathcal{Z}^{cs} = \{\mathbf{C}^e, r, D\}$  be a system of state variables.  $\mathbf{C}^e = \mathbf{F}^{eT} \cdot \mathbf{F}^e$  represents the elastic right Cauchy–Green strain tensor,  $r$  is an isotropic hardening variable and  $D$  is a scalar damage variable.

The free energy  $\varphi^{cs}$  per unity of mass is defined as:

$$\varphi^{cs} = (1 - D)[\varphi^e(\mathbf{C}^e) + \varphi^p(r)] \quad (3)$$

Such an approach has been initially developed in small deformation by Saanouni [12]. The same damage factor  $(1 - D)$  is introduced in the stored free energy via elasticity  $\varphi^e$  and the stored free energy via hardening  $\varphi^p$ . The elastic free energy is characterized by an isotropic fourth-order elastic modulus  $\underline{\underline{\mathbb{A}}}$

$$\varphi^e = \frac{1}{2\rho^e} \mathbf{E}^e : \underline{\underline{\mathbb{A}}} : \mathbf{E}^e \quad \text{with } \mathbf{E}^e = \frac{1}{2}(\mathbf{C}^e - \mathbf{I}) \text{ and } \rho^e = \rho \det \mathbf{F}^e \quad (4)$$

where  $\rho$  and  $\rho_e$  are mass density measured in the current configuration and the intermediate configuration respectively.  $\underline{\underline{\mathbb{A}}}$  is defined by the Young modulus ( $E$ ) and Poisson ration ( $\nu$ ).

The hardening free energy  $\varphi^p = \frac{1}{2\rho^e} Q \cdot b \cdot r^2$  is fully determined by two constants ( $Q$ ,  $b$ ). The state laws are derived from the free energy

$$\underline{\underline{\mathfrak{S}}}^e = \rho^e \frac{\partial \varphi^{cs}}{\partial \underline{\underline{\mathbf{C}}}^e} = \rho^e (1-D) \frac{\partial \varphi^e}{\partial \underline{\underline{\mathbf{C}}}^e} = (1-D) \underline{\underline{\mathbf{A}}}: \underline{\underline{\mathbf{E}}}^e \quad (5)$$

$$R = -\rho^e \frac{\partial \varphi^{cs}}{\partial r} = -\rho^e (1-D) \frac{\partial \varphi^p}{\partial r} = -(1-D) b \cdot Q \cdot r \quad (6)$$

$$Y = -\rho^e \frac{\partial \varphi^{cs}}{\partial D} = \rho^e (\varphi^e(\underline{\underline{\mathbf{C}}}^e) + \varphi^p(r)) \quad (7)$$

The Cauchy stress tensor in crystalline structure  $\underline{\underline{\sigma}}^{cs}$  is obtained by transport of  $\underline{\underline{\mathfrak{S}}}^e$  through the current configuration

$$\underline{\underline{\sigma}}^{cs} = \frac{1}{J^e} \underline{\underline{\mathbf{F}}}^e \cdot \underline{\underline{\mathfrak{S}}}^e \cdot \underline{\underline{\mathbf{F}}}^{eT} \quad \text{with } J^e = \det \underline{\underline{\mathbf{F}}}^e \quad (8)$$

### 3.3. Evolution laws

The approach of Lemaitre [13,14] is adopted as the fragmentation governs both the plasticity and damage. This approach involves a single plastic multiplier  $\lambda$  with a single dissipation potential and a single rule of normality. The dual dissipation potential  $\phi_\star^{cs}$ , obtained by Legendre–Fenchel’s transform of the dissipation potential  $\phi^{cs}$ , is written as:

$$\phi_\star^{cs} = \phi_\star^p(\underline{\underline{\mathfrak{Z}}}, R; D) + \phi_\star^D(Y; D) \quad \text{with } \underline{\underline{\mathfrak{Z}}} = \frac{1}{\rho} \underline{\underline{\mathbf{F}}}^{eT} \cdot \underline{\underline{\sigma}}^{cs} \cdot \underline{\underline{\mathbf{F}}}^{e-T} \quad (9)$$

where  $\underline{\underline{\mathfrak{Z}}}$  is the state force associated with plastic flow. The dissipation dual potentials are chosen as:

$$\phi_\star^D(Y; D) = \frac{\alpha}{\beta} Y \quad \text{and} \quad \phi_\star^p(\underline{\underline{\mathfrak{Z}}}, \tilde{R}) = f(\underline{\underline{\mathfrak{Z}}}, \tilde{R}) + \frac{\tilde{R}^2}{2Q} \quad \text{with } f(\underline{\underline{\mathfrak{Z}}}, \tilde{R}) = J(\underline{\underline{\mathfrak{Z}}}) - \tilde{R} - R_0 \quad (10)$$

where  $\underline{\underline{\mathfrak{Z}}} = \underline{\underline{\mathfrak{Z}}}/(1-D)$  and  $\tilde{R} = R/(1-D)$  are effective state forces. They are defined as the state forces obtained for the same internal state variable  $\underline{\underline{\mathbf{C}}}^e$  and  $r$  but with a damage-free material, *i.e.*  $D = 0$  [15]. The function  $f(\underline{\underline{\mathfrak{Z}}}, \tilde{R})$  defines the elastic domain.  $R_0$  is the limit of elasticity and  $J(\bullet)$  is the von Mises norm. The normality rule gives:

$$\underline{\underline{\nabla}}^p = \dot{\underline{\underline{\mathbf{F}}}}^p \cdot \underline{\underline{\mathbf{F}}}^{p-1} = \dot{\lambda} \frac{\partial \phi_\star^{cs}}{\partial \underline{\underline{\mathfrak{Z}}}} = \dot{\lambda} \frac{\partial \phi_\star^p}{\partial \underline{\underline{\mathfrak{Z}}}}, \quad \dot{r} = \dot{\lambda} \frac{\partial \phi_\star^{cs}}{\partial R} = \dot{\lambda} \frac{\partial \phi_\star^p}{\partial R}, \quad \dot{D} = \dot{\lambda} \frac{\partial \phi_\star^{cs}}{\partial Y} = \dot{\lambda} \frac{\partial \phi_\star^D}{\partial Y} \quad (11)$$

The gradient tensor of plastic rate is

$$\underline{\underline{\nabla}}^p = \dot{p} \underline{\underline{\mathbf{n}}}^e \quad \text{with } \underline{\underline{\mathbf{n}}}^e = \frac{\partial \phi^p}{\partial \underline{\underline{\mathfrak{Z}}}} = \frac{3}{2} \frac{\text{dev } \underline{\underline{\mathfrak{Z}}}}{J(\underline{\underline{\mathfrak{Z}}})} \quad (12)$$

because the derivative of accumulated plastic strain is:

$$\dot{p} = \sqrt{\frac{2}{3} \underline{\underline{\nabla}}^p : \underline{\underline{\nabla}}^p} = \frac{\dot{\lambda}}{1-D} \quad (13)$$

The expression of  $\dot{p}$  is obtained using the flow conditions:  $f(\underline{\underline{\mathfrak{Z}}}, \tilde{R}) = 0$  and  $\dot{f}(\underline{\underline{\mathfrak{Z}}}, \tilde{R}) = 0$  [16]. It is important to point out that the experimental coupling law (2) is obtained by integrating evolution equation (11-3).

## 4. Macromolecular network

### 4.1. Kinematics

Regarding the visco-elastic constitutive description of the macromolecular network, the deformation gradient tensor  $\underline{\underline{\mathbf{F}}} = \underline{\underline{\mathbf{F}}}_i^{ev} \underline{\underline{\mathbf{F}}}_i^v$  is decomposed into an elastic and a viscous component in the  $i$  Maxwell’s element, denoted respectively by the superscripts  $ev$  and  $v$ . The arbitrary rotation of this decomposition is eliminated by fixing the viscous spin tensor  $\underline{\underline{\Omega}}_i^v = \underline{\underline{\mathbf{0}}}$  [17], *i.e.* the viscous flow is assumed irrotational. The strain rate tensor  $\underline{\underline{\mathbf{D}}}$  is the sum of two terms:

- viscous strain rate tensor  $\underline{\underline{\mathbf{D}}}_i^v = \underline{\underline{\mathbf{L}}}_i^v = \underline{\underline{\mathbf{F}}}_i^{ev} \cdot \dot{\underline{\underline{\mathbf{F}}}}_i^v \cdot \underline{\underline{\mathbf{F}}}_i^{ev-1} \cdot \underline{\underline{\mathbf{F}}}_i^{ev-1}$ ;
- elastic strain rate tensor  $\underline{\underline{\mathbf{D}}}_i^{ev} = \frac{1}{2} (\underline{\underline{\mathbf{L}}}_i^{ev} + \underline{\underline{\mathbf{L}}}_i^{evT})$  with  $\underline{\underline{\mathbf{L}}}_i^{ev} = \dot{\underline{\underline{\mathbf{F}}}}_i^{ev} \cdot \underline{\underline{\mathbf{F}}}_i^{ev-1}$ .

#### 4.2. Thermodynamics potential and state laws

Let  $\mathcal{Z}^{rm} = \{\mathcal{C}, \mathcal{C}_1^{ev}, \dots, \mathcal{C}_i^{ev}, \dots\}$  be the set of internal state variables in the macromolecular network.  $\mathcal{C} = \mathbf{F}^T \cdot \mathbf{F}$  is the right Cauchy–Green strain tensor and  $\mathcal{C}_i^{ev} = \mathbf{F}_i^{evT} \cdot \mathbf{F}_i^{ev}$  is the right Cauchy–Green tensor in the relaxed configuration. The free energy is defined as

$$\varphi^{net} = \varphi^0(\mathcal{C}) + \sum_{i=1}^N \frac{1}{2\rho_i} \mathbf{E}_i : \mathbf{A}_i : \mathbf{E}_i \quad \text{with } \mathbf{E}_i = \frac{1}{2}(\mathcal{C}_i^{ev} - \mathbf{I}) \quad (14)$$

where  $\rho_i = \rho \det \mathbf{F}_i^{ev}$  is the mass density. The network behavior is assumed to be isotropic and deviatoric for simplicity. The compressibility of the materials is assumed to be governed only by the cohesive forces in crystalline structure. In other words, the objective derivative of Cauchy stress tensors  $\hat{\boldsymbol{\sigma}}_i$  is given by

$$\hat{\boldsymbol{\sigma}}_i = G_i(\mathbf{J} : \mathbf{D}_i^{ev}) \quad \text{with } \hat{\boldsymbol{\sigma}}_i = \dot{\boldsymbol{\sigma}}_i - \mathbf{L}_i^{ev} \cdot \boldsymbol{\sigma}_i - \boldsymbol{\sigma}_i \cdot \mathbf{L}_i^{evT} \quad \text{and } \mathbf{J} : \mathbf{A} = \text{dev } \mathbf{A} \quad (15)$$

so only one parameter  $G_i$  is needed to define the coefficients of the elasticity tensor  $\mathbf{A}_i$  by the transport.

$$G_i \mathbf{J} = \frac{1}{\det \mathbf{F}_i^{ev}} (\mathbf{F}_i^{ev} \otimes \mathbf{F}_i^{ev}) : \mathbf{A}_i : (\mathbf{F}_i^{evT} \otimes \mathbf{F}_i^{evT}) \quad \text{with } (\mathbf{A} \otimes \mathbf{B})_{ijkl} = A_{ik} B_{jl} \quad (16)$$

An Arruda–Boyce potential [18] defined by two scalars ( $n, \mu$ ) is adopted for the hyperelastic contribution of free energy  $\varphi^0$ . The state laws are derived from the free energy

$$\mathbf{S}^0 = \rho_0 \frac{\partial \varphi^0}{\partial \mathcal{C}} \quad \text{and} \quad \mathbf{S}^i = \mathbf{A}_i : \mathbf{E}_i \quad (17)$$

The Cauchy stress tensor in the entropic network  $\boldsymbol{\sigma}^{net}$  are obtained by transport of state forces  $\mathbf{S}^i$  through current configuration

$$\boldsymbol{\sigma}^{net} = \frac{1}{J} \mathbf{F} \cdot \mathbf{S}^0 \cdot \mathbf{F}^T + \sum_{i=1}^n \mathbf{F}_i^{ev} \cdot \mathbf{S}^i \cdot \mathbf{F}_i^{evT} \quad \text{with } \boldsymbol{\sigma}^i = \mathbf{F}_i^{ev} \cdot \mathbf{S}^i \cdot \mathbf{F}_i^{evT} \quad (18)$$

#### 4.3. Evolution laws

The dissipation potential is written as:

$$\phi^{net} = \sum_{i=1}^N \frac{1}{2} \mathbf{D}_i^v : \boldsymbol{\eta}_i : \mathbf{D}_i^v \quad \text{with } \boldsymbol{\eta}_i = \tau_i \cdot G_i \cdot \mathbf{J} \quad (19)$$

where  $\boldsymbol{\eta}_i$  is fourth order viscous tensor in  $i$  Maxwell element. It is defined just by the relaxation time  $\tau_i$  and the modulus  $G_i$ .

The evolution law for the Cauchy stress tensor  $\boldsymbol{\sigma}_i = \frac{\partial \phi^{net}}{\partial \mathbf{D}_i^v}$  is rewritten using Eq. (15)

$$\hat{\boldsymbol{\sigma}}_i + \frac{1}{\tau_i} \boldsymbol{\sigma}_i = G_i(\mathbf{J} : \mathbf{D}) \quad (20)$$

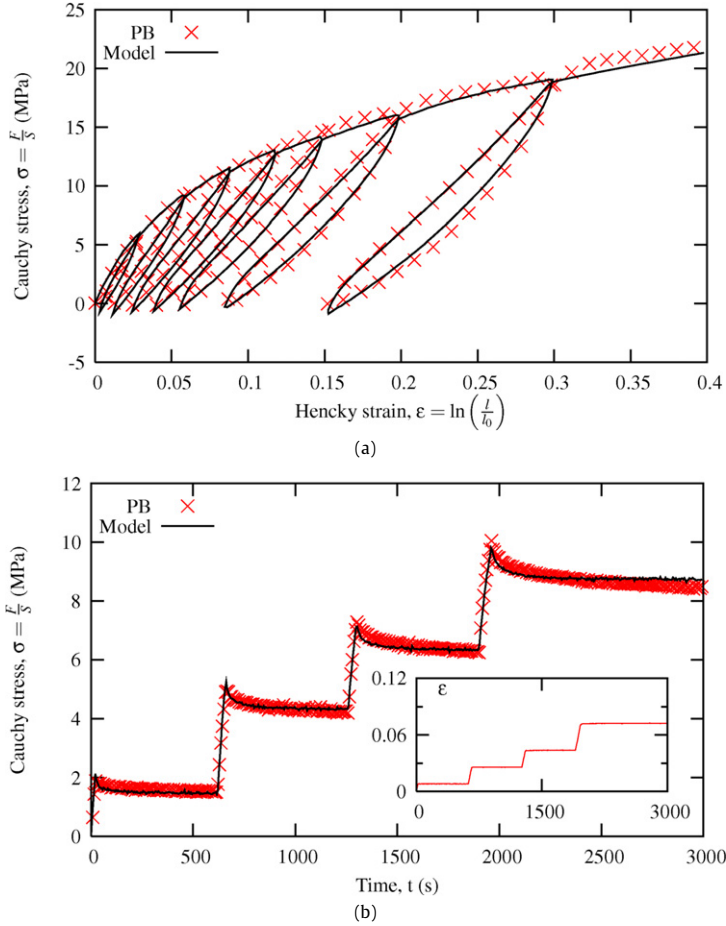
To conclude the model part, Table 1 summarizes the current model and gives the different material parameters. A fourth order Runge–Kutta method has been used in the numerical integration using the finite element code, ZeBuLon.

### 5. Uniaxial response of the model: Case of polybutene

Two relaxation times  $\tau_1 = 10$  s and  $\tau_2 = 100$  s have been considered. Initially, the viscosity parameters ( $G_1 = 101$  MPa,  $G_2 = 21$  MPa) are identified on a relaxation test before plastic deformation. The remaining material parameters ( $E = 167$  MPa,  $\nu = 0.27$ ,  $Q = 28.6$  MPa,  $b = 20.8$ ,  $R_0 = 3.24$  MPa,  $\alpha = 0.59$ ,  $\beta = 41.7$ ,  $n = 4.5$ ,  $\mu = 3.58$  MPa) are identified using monotonic and loading/unloading tensile tests. Good agreement between the experimental cyclic tensile test and numerical simulation have been obtained (Fig. 4a) with fairly good predictions of the relaxation behavior (Fig. 4b). The introduction of the damage/plasticity coupling law can account for both the slope variation at the beginning of the relaxation and the increase in the stress drop with maximal stress.

**Table 1**  
Visco-hyperelastic and plastic model coupled with isotropic damage.

	Relevant equations	Material parameters
Kinematic	$\underline{\mathbf{F}} = \underline{\mathbf{F}}^e \cdot \underline{\mathbf{F}}^p$ $\underline{\mathbf{F}} = \underline{\mathbf{F}}_i^{ev} \cdot \underline{\mathbf{F}}_i^v$	(Multiplicative decompositions)
Elasticity	$\underline{\boldsymbol{\sigma}} = \underline{\boldsymbol{\sigma}}^{cs} + \underline{\boldsymbol{\sigma}}^{net}$	(Stress partition) $E; \nu; \mu; N; G_i$
Plasticity	$\underline{\nabla}^p = \dot{\underline{\mathbf{F}}}^p \cdot \underline{\mathbf{F}}^{p-1} = \dot{p} \underline{\mathbf{n}}^e$ $\dot{R} = \frac{R}{1-D} = Q(1 - e^{-b \cdot p})$ $f \leq 0$ and $\dot{p} \geq 0$ and $\dot{p} \cdot f = 0$	(Plastic flow) (Isotropic hardening) (Plastic criterion) $b; Q$ $R_0$
Damage	$D = \alpha(1 - e^{-\beta \cdot p})$	(Coupling law) $\alpha; \beta$
Viscosity	$\underline{\mathbf{D}}_i^v = \underline{\mathbf{F}}_i^{ev} \cdot \dot{\underline{\mathbf{F}}}_i^v \cdot \underline{\mathbf{F}}_i^{v-1} \cdot \underline{\mathbf{F}}_i^{ev-1} = \frac{1}{\eta_i} \underline{\boldsymbol{\sigma}}_i$	(Viscous flow) $\tau_i = \frac{\eta_i}{G_i}$



**Fig. 4.** (a) Load/unload tensile curve ( $\dot{\epsilon} = 5 \times 10^{-4} \text{ s}^{-1}$ ,  $T = 20^\circ\text{C}$ ). (b) Multi-relaxation curve ( $T = 20^\circ\text{C}$ ).

## 6. Conclusion and outlook

In this Note, the introduction of a plasticity/damage coupling has allowed us to model the loading/unloading tensile tests of semi-crystalline polymers. Moreover, relaxations behavior has been predicted by the proposed formulation.

First, an isotropic damage has been considered. However, AFM observations has revealed an anisotropic fragmentation inside the spherulites. It could be relevant to introduce a second order tensor to represent damage  $\underline{\mathbf{d}}$  coupled with a kinematic hardening.

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