Large-strain viscoelastic–viscoplastic constitutive modeling of semi-crystalline polymers and model identification by deterministic/evolutionary approach

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ABSTRACT

Above the glass transition temperature, a semi-crystalline polymer can behave like an elastomer or a stiff polymer according to the crystal content. For a reliable design of such polymeric materials, it is of prime importance to dispose a unified constitutive modeling able to capture the transition from thermoplastic-like to elastomeric-like mechanical response, as the crystal content changes. This work deals with poly-ethylene materials containing a wide range of crystal fractions, stretched under large strains at room temperature and different strain rates. A large-strain viscoelastic–viscoplastic approach is adopted to describe the mechanical response of these polymers. In order to identify the model parameters, an ana-lytical deterministic scheme and a practical, "engineering-like", numerical tool, based on a genetic algo-rithm are developed. A common point of manipulated constitutive models is that the elementary deformation mechanisms are described by two parallel resistances; one describes the intermolecular interactions and the other deals with the molecular network stretching and orientation process. In a first approach, the semi-crystalline polymers are considered as homogeneous media; at each crystal content, the semi-crystalline polymer is thus considered as a new material and a new set of model parameters is provided. In a second approach, the semi-crystalline polymer is seen as a two-phase composite, and the effective contribution of the crystalline and amorphous phases to the overall mechanical response is inte-grated in the constitutive model, which allows simulating the transition from thermoplastic-like to elas-tomeric-like mechanical response. In this case, one set of model parameters is needed, the only variable being the crystal volume fraction. The identification results obtained using deterministic and numerical methods are discussed.

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1. Introduction

Nowadays, thermoplastic polymers are widely used for engi-neering applications. However, the prediction of their mechanical behavior remains a complex task since these materials have a highly non-linear stress–strain relationship depending on several external (strain rate, temperature) and structural (entanglement, cross-linking, crystal content, crystalline lamellae size, lamellae distribution) factors. Besides, large plastic strains may be locally reached in polymer components. Over decades many constitutive

models have emerged to predict the stress–strain behavior of amorphous thermoplastic polymers both in the glassy and rubbery states and many works have been done to improve the constitutive models to adapt and respond to different material types under dif-ferent conditions, as reviewed in many works [1–22]. The physi-cally-based constitutive models for the glassy amorphous polymers are inspired from the early work of Haward and Thackray [23] founded on the observation of a large recoverable extension under glass transition points. In these constitutive models, a visco-plastic dashpot for the intermolecular interactions is connected to a non-linear spring to simulate the alignment of the polymer chains at large strains. To predict the stress–strain behavior of semi-crystalline polymers, many authors [24–33] used purely phe-nomenological constitutive models. Inherent to the structure of these models is the absence of linking to the microstructure which prevents the understanding and prediction of crystal content effects on the overall mechanical response [34–42]. Recently,

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Ayoub et al. [42] proposed a physically-based constitutive model for semi-crystalline polymers integrating explicitly the crystal content in the mathematical formulation. The constitutive model is based on the Boyce et al. [7] approach, which was later extended by Ahzi et al. [36] to capture the strain-induced crystallization of the initially amorphous polyethylene terephthalate. Inherent to the model structure is the assumption that the resistance in the semi-crystalline polymer to deformation is the sum of elastic–viscoplastic crystalline and amorphous intermolecular resistances and a visco-hyperelastic network resistance. The Ayoub et al. [42] constitutive model was able to capture the transition from thermoplastic-like to elastomeric-like mechanical response of polyethylene, as the crystal content changes. The authors [42] identified the model parameters using an analytical deterministic scheme. The deterministic procedure uses a precise algorithm of identification and calculates a unique set of model parameters [17]. However, the application of this method demands an advanced expertise in model formulation and wide experience in the understanding of experimental material behavior. Moreover, during the cycle of model-experiments comparison and model (re)formulation, the complexity of material response leads often to an excessive number of parameters which do not necessarily all have the reason to be in the model.

The identification procedure is the main barrier of handling the constitutive models. The difficulty of model parameters identification is proportional with the complexity of the constitutive model itself; it is time wasting. An efficient technique of parameters identification is vital to a utility of constitutive models. Thus, developing a software solution for model parameters identification allows time and cost effective solution and keeps constitutive models much more useful [43–53]. In the present work, we intend to develop and evaluate a numerical tool which allows to identify the overall constitutive model variable sets, directly from stress–strain curves, to contrast with the analytical deterministic approach. Moreover, such identification procedures should be reliable, useful and convenient for a large number of users. The numerical tool proposed in this work is dedicated to non-specialists of mathematical optimization heuristics, thus to exhibiting a practical "engineering-like" tool design. A great deal of researches have been conducted on the model parameters identification for metals, but few researches have been carried out to determine the model parameters for polymers, especially at large strains.

The present work is focused on the constitutive modeling of semi-crystalline polymers but also on the problem of model parameters identification. A genetic algorithm (GA) based optimization procedure is designed to determine the parameters of large-strain viscoelastic–viscoplastic constitutive models, and the results determined by GA compared to results of an analytical deterministic scheme. The application is performed on polyethylene with a wide range of crystal fractions including thermoplastic elastomer and stiff thermoplastic polymer mechanical responses. To illustrate the interest, reliability and usefulness of the proposed numerical identification tool, two constitutive models are retained. One [7] supposes the semi-crystalline material as homogeneous and the other [42] considers it as heterogeneous. The robustness of both constitutive models is examined. A secondary objective is to revise the Ayoub et al. [42] constitutive model in order to reduce the required model parameters.

The present paper is organized as follows. In Section 2, we review large-strain viscoelastic–viscoplastic constitutive modeling approaches. In Section 3, we present the analytical deterministic scheme. In Section 4, we present the computational formulation of the problem and the designed GA-based identification tool. In addition the robustness, reproducibility and uniqueness of the solutions are discussed. In Section 5, the identification results are presented and discussed. The concluding remarks are detailed in Section 6.

2. Large-strain viscoelastic–viscoplastic constitutive models

In this section, the main elements of two recently developed constitutive models for thermoplastic polymers are summarized. The first one (referred to the BSL model for "Boyce–Socrate–Llana model") considers the material as homogeneous while the second one (referred to the MBSL model for "modified BSL model") treats the material as heterogeneous by distinguishing amorphous and crystalline phases. These constitutive models satisfy the continuum mechanics rules within the context of the large-strain viscoelastic–viscoplastic framework. A common point of these constitutive models is that the resistance to deformation in the semi-crystalline polymers is the sum of a resistance A describing the intermolecular interactions and a resistance B describing the molecular network stretching and orientation process. The intermolecular resistance is composed of a linear elastic spring in series with a viscoplastic damper and the molecular network resistance is composed of a non-linear spring in series with a viscous damper.

As a point of departure, a summary of the finite strain kinematic framework is given. The key quantity is the deformation gradient defined by: $\mathbf{F} = \partial \mathbf{x}/\partial \mathbf{X}$ where $\mathbf{x}$ is the position of a material point in the current configuration and $\mathbf{X}$ is the position of this material point in the initial configuration. Note that all tensors are denoted by bold-face symbols.

Due to the model structure, the Taylor assumption prevails, i.e. the intermolecular deformation gradient $\mathbf{F}_A$ and the network deformation gradient $\mathbf{F}_B$ are equal to the total deformation gradient $\mathbf{F}$:

$$\mathbf{F} = \mathbf{F}_A = \mathbf{F}_B \qquad (1)$$

Note that for the MBSL constitutive model the effective contribution of the crystalline and amorphous phases to the overall intermolecular resistance are also integrated with the Taylor assumption:

$$\mathbf{F}_A = \mathbf{F}_A^a = \mathbf{F}_A^c \qquad (2)$$

where the superscripts a and c denote the amorphous and crystalline phases, respectively.

Following the Lee [54] decomposition, schematically illustrated in Fig. 1, the deformation gradient tensors can be further decomposed multiplicatively into elastic (network) and viscoplastic (flow) parts as:

$$\mathbf{F}_A = \mathbf{F}_A^e \mathbf{F}_A^p \qquad (3)$$

$$\mathbf{F}_B = \mathbf{F}_B^N \mathbf{F}_B^F \qquad (4)$$

where the superscripts e, p, N and F denote the elastic, viscoplastic, network and flow parts, respectively. Note that the decomposition given in Eq. (3) is also applicable to crystalline and amorphous phases in the case of the MBSL constitutive model.

According to the polar decomposition, the deformation gradient tensors can be further decomposed into stretch and rotation movements:

$$\mathbf{F}_A = \mathbf{V}_A^e \mathbf{R}_A^e \mathbf{V}_A^p \mathbf{R}_A^p \qquad (5)$$
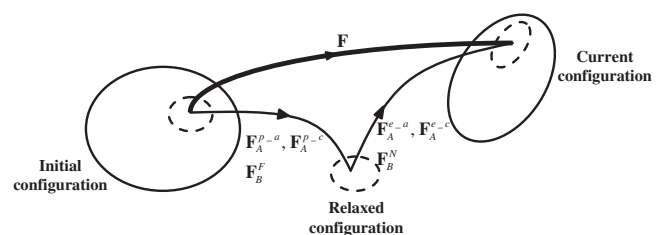

Fig. 1. Schematic illustration of the strain multiplicative decomposition.