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A thermodynamically consistent phase-field framework for structural damage in Polytetrafluoroethylene (PTFE) polymer

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Considering the significant increasing of polymeric products and components in many applications, with different safety and quality requirements, the appropriate description of the mechanical behaviour of polymers is necessary for design purposes. Usually, polymers have quite different structural and thermal responses when compared to metals, which can impact their manufacturing process and usage conditions. Deformation in polymers is highly dependent on molecular structures and motion and relaxation mechanisms, which are highly influenced by temperature and mechanical load history. These features imply that some models can fit for specific classes of polymers and not for others, and also include several non-linearities, which turns out to be challenging for computational simulation. This work develops and simulates a phase field model for the polytetrafluorethylene (PTFE) polymer. Using finite element techniques in a one-dimensional domain, the stress-strain and fracture response of this model is compared to experimental result. The constitutive multimechanism model used in this work considers a non-linear elastoviscoplastic condition, originally used for amorphous polymers. Conceptually, the simulation is able to represent elastic molecular interactions and viscoplastic flow from polymer segments, like sliding and unkinking. Hardening at higher strains, due to polymer chain alignment, is also included. The fitting of material parameters from the rheological model is obtained by a genetic algorithm, in order to adjust curves from simulated models to stress-strain experiments available in literature. The result of stress-strain curve, followed by rupture is plotted in comparison with experimental results, presenting a reasonable fit.
Keywords: Phase-field model, Finite element method, Constitutive models

INTRODUCTION

Nowadays, polymeric materials have been used largely in engineering applications. Due to advances in reinforcement fibres and combined molecular structures, polymeric materials can offer many advantages when compared to others materials even in most precise or under extremely severe usage conditions. For example, the importance of polymers in the automotive industry is asserted by the average global plastic usage of about 120 kg per vehicle according to Patil et al. (2017).

Usually, polymers have quite different structural and thermal responses when compared to metals, which can impact their manufacturing process and also their usage conditions. For this reason, many material constitutive models were developed in last decades and still represent a growing field.

Deformation in polymers is highly dependent on their molecular structures and motion and relaxation mechanisms, which are highly influenced by temperature and mechanical load history. These features imply that some models can fit well for specific classes of polymers and not for others and include several non-linearities, which turns out to be challenging for computational simulation.

In this context, the polymers containing fluorine present advantages related to improved mechanical, electrical, chemical and friction wear resistance. According to Brown et al. (2004), the most used fluorocarbon polymer in applications is the Polytetrafluorethylene (PTFE) commonly known under the trade name Teflon, which presents a complex microstructure with different crystalline phases in terms of temperature at ambient pressure. These phases influence the mechanical behavior, including the resistance to fracture.

For describing properly the behavior of polymers, a large number of works, started decades ago, aimed to implement constitutive models in order to reproduce different aspects of mechanical response. When properly calibrated, this effort created an important background whose models can, with reasonable accuracy, reproduce the macroscopic response of a polymer under multi-axial loading conditions over a wide range of strain rates and temperatures.

The starting point for many others authors describing amorphous polymers response is Haward and Thackray (1968), who presented an one-dimensional rheological model composed by two mechanisms (Eyring dashpot and a Langevin spring). The stress response is built from different contributions, from intermolecular interactions and also from preferred

orientation of the entangled molecular network. Subsequently, and counting on development of rubber elasticity by Treloar (1975), many others authors extended constitutive models for including more dimensions and complex behaviors, as Parks et al. (1984), Boyce et al. (1988), Wu and Van Der Giessen (1993), Tervoort et al. (1997), Govaert et al. (2000), Anand and Gurtin (2003), Buckley et al. (2004) Anand et al. (2009), Srivastava et al. (2010) and Bergstrom (2015).

Many methods for evaluating damage mechanisms are based on Griffith (1921) theory, which introduced the concept of energy release rate G as the responsible for the onset of a crack. An important variant of Griffith method was proposed by Francfort and Marigo (1998), in which the crack initiation, propagation and ramification process is driven by a minimization of an energy functional, in terms of displacement field and crack surface. Considering that crack surface is unknown in the beginning of process, it is replaced by a continuous scalar field, named phase-field, which interpolates smoothly between undamaged and broken material.

Due to these new techniques, more attention has been given to the phase-field methodology as a promising way to deal with modelling of material damage. Originally applied to problems related to separation of fluids by Cahn and Hilliard (1958), these models have been also used in a great variety of multiphase problems such as in the study of tumor growth by Silva (2009), vesicle-fluid iteration by Du et al. (2007), solidification by Allen and Cahn (1972) and fluid–structure interation problems by Sun et al. (2014). In addition to these categories, many fracture and crack initiation problems are solved using Ginsburg–Landau free-energy functional in order to predict the phase field evolution in continuum mechanics and solid materials, as described by Fabrizio et al. (2006), Amendola et al. (2016), Boldrini et al. (2016), Schänzel (2015), Miehe et al. (2015), and Narayan and Anand (2021).

This work develops a thermodynamically consistent model for PTFE polymer, in order to describe its behaviour under large displacement. The viscoelastoplastic constitutive model described by Marin et al. (2007) is used and new parameters are found in order to describe the response of this polymer. Moreover, a phase-field formulation is added to the model in order to include failure response.

In the next sections, the general equations are shown, followed by the specialization of the phase-field model and, finally, the results are compared to experiments.

GENERAL BALANCE EQUATIONS

The one-dimensional phase-field model for polymers is adapted mainly from arguments used by Boldrini et al. (2016), based on continuum mechanics and thermodynamics conservation laws. In this context, the basic governing equations (mass conservation, linear momentum balance, energy conservation, entropy inequality and Kroner-Lee decomposition) in material reference frame are

$$
\begin{cases}
\dot{\rho}_0 = 0, \\
\dot{u} = v, \\
\rho_0 \ddot{u} = \displaystyle\sum_{\alpha=1}^{M} \frac{d\left(JF_e^{(\alpha)}\tilde{T}_e^{(\alpha)}\left(F_e^{(\alpha)}\right)^T\left(F^{(\alpha)}\right)^{-T}\right)}{dX} + \rho_0 b_0, \\
0 = \dfrac{dh_0}{dX} - b_{\varphi 0} + \rho_0 a_0, \\
\rho_0 \dot{e}_0 = -\dfrac{dq_0}{dX} + \rho_0 g_0 + \displaystyle\sum_{\alpha=1}^{M} (J\tilde{T}_p^{(\alpha)}\dot{E}_p^{(\alpha)} + \frac{1}{2}J\tilde{T}_e^{(\alpha)}\dot{C}_e^{(\alpha)}) + b_{\varphi 0}\dot{\varphi} + h_0\dfrac{d\dot{\varphi}}{dX}, \\
-\rho_0\dot{\psi}_0 - \rho_0\eta_0\dot{\theta} + \displaystyle\sum_{\alpha=1}^{M} (J\tilde{T}_p^{(\alpha)}\dot{E}_p^{(\alpha)} + \frac{1}{2}J\tilde{T}_e^{(\alpha)}\dot{C}_e^{(\alpha)}) + b_{\varphi 0}\dot{\varphi} + h_0\dfrac{d\dot{\varphi}}{dX} - \dfrac{d\theta}{dX}\dfrac{q_0}{\theta} \geq 0, \\
F^{(\alpha)} = F_e^{(\alpha)}F_p^{(\alpha)}, \\
F_p^{(\alpha)}\tilde{T}_p^{(\alpha)}\left(F_p^{(\alpha)}\right)^T = C_e^{(\alpha)}\tilde{T}_e^{(\alpha)},
\end{cases}
\tag{1}
$$

where ρ_0 is the density, u is the displacement field, v is the velocity, J is the determinant of deformation gradient $F^{(\alpha)}$, α is a given mechanism of the rheological model, $F_e^{(\alpha)}$ and $F_p^{(\alpha)}$ are the elastic and plastic parts of $F^{(\alpha)}$, $T_e^{(\alpha)}$ and $T_p^{(\alpha)}$ are the elastic and plastic stresses and b_0 is the external body force along the bar length, $\bar{C}_e^{(\alpha)}$ is the elastic right Cauchy-Green, $E_p^{(\alpha)}$ the plastic Green-Lagrange deformations, $\tilde{T}_e^{(\alpha)} = \left(F_e^{(\alpha)}\right)^{-1} T_e^{(\alpha)}\left(F_e^{e(\alpha)}\right)^{-T}$ and $\tilde{T}_p^{(\alpha)} = \left(F_p^{(\alpha)}\right)^{-1} T_p^{(\alpha)}\left(F_p^{(\alpha)}\right)^{-T}$.

Moreover, φ is the phase-field variable, to describe the failure of the material and a_0 is the density of energy supplied by exterior to evolving structures (such as phase-field describing damages in material). Microscopic terms $b_{\varphi 0}$ and h_0 are, respectively, the volumetric density of energy exchanged by variation of a unit of φ and the flux of energy associated to

the spatial variation of a unit of φ in a unit of time. θ is the absolute temperature, η_0 is the specific density of entropy, q_0 is the heat flux and g_0 is the specific density of heat sources and sinks.

All these balance laws are not enough to fully characterize the physical response of a given body and must be completed with the material constitutive equations.

Similarly to Frémond and Shitikova (2002), $\tilde{T}_e$, $\tilde{T}_p$, $b_{\varphi 0}$, h_0 and q_0 are split in their reversible (non-dissipative) and irreversible (dissipative) parts, which are indicated, respectively, by the superscripts $(.)^{(r)}$ and $(.)^{(ir)}$ below

$$\begin{aligned}
\tilde{T}_e &= \tilde{T}_e^{(r)} + \tilde{T}_e^{(ir)},\\
\tilde{T}_p &= \tilde{T}_p^{(r)} + \tilde{T}_p^{(ir)},\\
b_{\varphi 0} &= b_{\varphi 0}^{(r)} + b_{\varphi 0}^{(ir)},\\
h_0 &= h_0^{(r)} + h_0^{(ir)},\\
q_0 &= q_0^{(r)} + q_0^{(ir)}.
\end{aligned} \tag{2}$$

For simplicity, as Frémond and Shitikova (2002), we consider

$$h_0^{(ir)} \equiv 0, \tag{3}$$

$$q_0^{(r)} \equiv 0. \tag{4}$$

Considering the chain rule for $\psi^{(\alpha)}$, the entropy inequality is written in terms of the free-energy as

$$\begin{aligned}
&-\rho_0\left(\eta_0 + \frac{\partial \psi^{(\alpha)}}{\partial \theta^{(\alpha)}}\right)\dot{\theta} + \left(-\rho_0\frac{\partial \psi^{(\alpha)}}{\partial \varphi^{(\alpha)}} + b_{\varphi 0}^{(r)} + b_{\varphi 0}^{(ir)}\right)\dot{\varphi} - \left(\rho_0\frac{\partial \psi^{(\alpha)}}{\partial \nabla\varphi^{(\alpha)}} - h_0^{(r)}\right)\nabla\dot{\varphi}\\
&\quad + \left(\frac{1}{2}J\tilde{T}_e^{(r)} + \frac{1}{2}J\tilde{T}_e^{(ir)} - \rho_0\frac{\partial \psi^{(\alpha)}}{C_e^{(\alpha)}}\right)\dot{C}_e^{(\alpha)} + \left(JT_p^{(r)} - 2\rho_0\frac{\partial \psi^{(\alpha)}}{\partial A^{(\alpha)}}A\right)D_p^{(\alpha)}\\
&\quad -\rho_0\frac{\partial \psi^{(\alpha)}}{\partial \nabla\theta^{(\alpha)}}\nabla\dot{\theta} - \rho_0\left(\frac{\partial \psi^{(\alpha)}}{\partial A^{(\alpha)}}G^{(\alpha)}\right)d_p^{(\alpha)} + JT_p^{(ir)}D_p^{(\alpha)} - \frac{1}{\theta}q^{(ir)}\frac{1}{\theta}\frac{d\theta}{dX} \geq 0,
\end{aligned} \tag{5}$$

with $A^{(\alpha)}$ being a dimensionless squared stretch-like quantity, whose evolution depends on the dynamic ($G^{(\alpha)}$) recovery term.

Given this inequality, the constitutive equations are found to be

$$\eta_0 = -\frac{\partial \psi^{(\alpha)}}{\partial \theta^{(\alpha)}}, \tag{6}$$

$$\frac{\partial \psi^{(\alpha)}}{\partial \nabla\theta^{(\alpha)}} = 0, \tag{7}$$

$$b_{\varphi 0}^{(r)} = \rho_0\frac{\partial \psi^{(\alpha)}}{\partial \varphi^{(\alpha)}}, \tag{8}$$

$$\tilde{T}_e^{(r)} = 2J^{-1}\rho_0\frac{\partial \psi^{(\alpha)}}{\partial C_e^{(\alpha)}}, \tag{9}$$

$$\tilde{T}_p^{(r)} = 2J^{-1}\rho_0\frac{\partial \psi^{(\alpha)}}{\partial A^{(\alpha)}}A, \tag{10}$$

$$h_0 = h_0^{(r)} = \rho_0\frac{\partial \psi^{(\alpha)}}{\partial \nabla\varphi^{(\alpha)}}, \tag{11}$$

$$q^{(ir)} = -\tilde{c}\frac{1}{\theta}\frac{d\theta}{dX}, \tag{12}$$

$$b_{\varphi 0}^{(ir)} = \tilde{\gamma}\dot{\varphi}, \tag{13}$$

$$\tilde{T}_e^{(ir)} = 2\tilde{b}_d\dot{C}_e^{(\alpha)}, \tag{14}$$

$$\tilde{T}_p^{(ir)} = JC_e^{(\alpha)}\tilde{T}_e^{(r)} - 2J^{-1}\rho_0\left(\frac{\partial \psi^{(\alpha)}(C_e^{(\alpha)}, A^{(\alpha)}, \theta)}{\partial A^{(\alpha)}}A^{(\alpha)}\right), \tag{15}$$

where $\tilde{\gamma}$ and $\tilde{b}_d$ are non-negative coefficients.

CONSTITUTIVE MODELS AND PARAMETER FITTING

A large number of constitutive models for polymers, specially for amorphous type, can be found in literature. Most of these models, for simplicity reasons, make assumptions in order to reduce material parameters, many of them being difficult to be predicted or collected through experimental tests.

This work uses a constitutive model proposed by Marin et al. (2007), which will not be described here for brevity reasons, and for more details the reader is suggested to check the original paper. It uses the multimechanism concept, where each one has a specific deformation nature, aiming to represent different sources of resistance in polymers (intramolecular and intermolecular), as suggested earlier by Haward and Thackray (1968).

Defining the material internal parameters for these complex rheological models requires an extensive effort, since most of these terms are not obtained directly from experiments. For this reason, it is necessary to apply optimization techniques, in order to adjust the material model response to the results of stress-strain experiments available in literature. The optimization technique used here for parameter fitting was the genetic algorithm, which is used for searching optimal solutions based on the principle of natural selection.

For this isothermal constitutive model, considering its lower number of parameters, the convergence is not difficult using the genetic algortihm called *eaSimple*, from the Python library DEAP (*Distributed evolutionary algorithms in Python)*.

SPECIALIZATION OF PHASE-FIELD EQUATIONS

The free energy functional including a phase-field damage variable is assumed to be

$$\psi(\varphi,\nabla\varphi,C_e,A,C) = \psi^{e(1)}(\varphi,C_e^{(1)}) + \psi_p^{(1)}(A) + \psi_e^{(\alpha)}(C) + \mathcal{J}(\varphi,\nabla_p\varphi), \tag{16}$$

with parameter α being dependent on the constitutive model.

The free-energy related to elasticity is given by the first term on the right hand side of Eq. (16), comprised by degradation ($D(\phi)$) and energetic terms (ψ_0^e), such that

$$\psi_e^{(1)}(\varphi,C_e) = D(\varphi)\psi_0^{e(1)}(\theta,C_e^{(1)}). \tag{17}$$

The energetic part is given by the constitutive models cited earlier and the degradation function is based on a two-well potential, shown in Fig.1, and defined as

$$D(\varphi) = \frac{1}{4\eta^2}\left((\varphi^2-1)^2 - a\varphi(\varphi-1)^2\right), \tag{18}$$

where $0 \leq \varphi \leq 1$ is the phse-field variable and $a > 0$ is a small term used to ensure the damage evolution from an initial condition when the damage is zero.

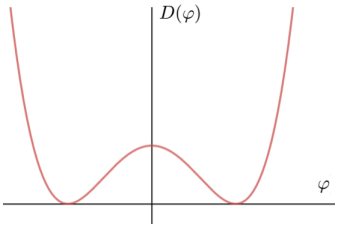

Figure 1 – Two well potential function.

The free energy related to damage is

$$\mathcal{J}(\varphi,\nabla\varphi) = g_c\left(\frac{\gamma}{2}\left|\frac{d\varphi}{dX}\right|^2 + \frac{1}{\gamma}\mathcal{H}(\varphi)\right), \tag{19}$$

with the material parameter g_c representing the Griffith's critical energy release rate and γ a positive constant related to

the fracture width. The expression for the potential $\mathcal{H}$ is given by

$$\mathcal{H}(\varphi) = \begin{cases} \frac{1}{4}(4\varphi^2 - 4\varphi)^2 & \text{for } 0 \leq \varphi \leq 1, \\ \frac{1}{2} & \text{for } \varphi > 1, \\ 0 & \text{for } \varphi < 0. \end{cases} \tag{20}$$

The degradation function and the damage free energy are able to correctly describe the abrupt rupture of the material under tension, as noticed, for instance, in experimental results at -10°C shown in Gamboni et al. (2016).

RESULTS

Consider a bar of material with the constitutive model described by Marin et al. (2007), discretized in 20 linear elements, with constant cross section, under tension and isothermal conditions (ambient temperature). An initial zero damage is considered all over the domain except in its center, where a small damage (0.01), representing a stress concentration, is imposed. The central element response is shown in Fig. 2, where it can be seen an abrupt (in terms of time) reduction of resistance, which indicates a rupture in material.

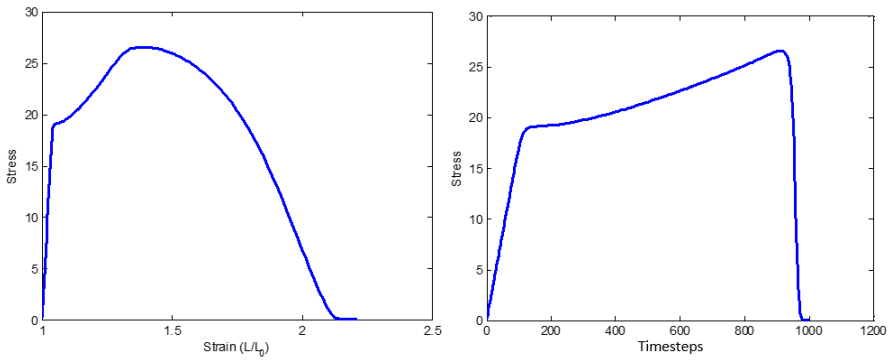

Figure 2 – Stress on the central element as a function of strain and timesteps, with influence of damage leading to fracture.

As observed in experiments, fracture in PTFE occurs in an abrupt manner. This behaviour is detected in this simulation. The evolution of damage variable for the center element is shown in Fig. 3.

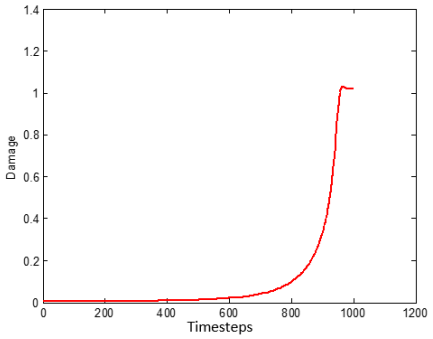

Figure 3 – Damage evolution on the central element.

In order to obtain fracture at the same strain as verified in the experiments, damage parameters can also be defined. With all material constants, it is possible to compare model with experimental results, as shown in Fig. 4. In the experiment, right after the last point in blue curve, there is fracture, in accordance with the adjusted model.

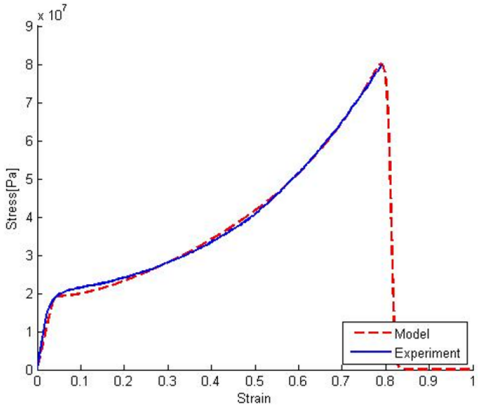

Figure 4 – Comparison between model and experimental stress-strain test.

CONCLUSION

The stress-strain values from the adjusted model, along with the failure response from phase-field model, present good agreement to PTFE experimental results, despite its complex structure. This simulation, however, does not take into consideration different temperature and strain-rates, which would bring additional requirements to constitutive model and, consequently, parameter fitting.

Considering the high number of applications of PTFE in engineering, the results presented here and the possibilities of an improved model considering a broader range of conditions (such as temperature and strain-rates) emerge as a interesting field for improving product development with this material.

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