Modeling and Investigation of Volume Strain at Large Deformation under Uniaxial Cyclic Loading in Semi Crystalline Polymer

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Abstract—This study deals with the experimental investigation and theoretical modeling of Semi crystalline polymeric materials with a rubbery amorphous phase (HDPE) subjected to a uniaxial cyclic tests with various maximum strain levels, even at large deformation. Each cycle is loaded in tension up to certain maximum strain and then unloaded down to zero stress with N number of cycles. This work is focuses on the measure of the volume strain due to the phenomena of damage during this kind of tests. On the basis of thermodynamics of relaxation processes, a constitutive model for large strain deformation has been developed, taking into account the damage effect, to predict the complex elasto-viscoelastic-viscoplastic behavior of material. A direct comparison between the model predictions and the experimental data show that the model accurately captures the material response. The model is also capable of predicting the influence damage causing volume variation.

Keywords—Cyclic test, large strain, polymers semi-crystalline, Volume strain, Thermodynamics of Irreversible Processes.

I. INTRODUCTION

Modeling of nonlinear elasto-viscoplastic behavior of semi crystalline polymers including the effects of variation of volume, mainly associated with the damage phenomena, has received a considerable attention in many researches. This physical phenomenon has been studied in numerous publications under uniaxial monotonic tensile test [1]-[3].

Although there are many experimental and mechanical models that have been developed on polymers to characterize the mechanical response at uniaxial tensile test with strain-controlled program [4],[5], very limited studies focus on the evaluation of true axial stress – strain and volume strain during uniaxial cyclic tensile tests in large deformation at different maximum strain levels. This paper is aimed to modeling the effect of cyclic deformation on the elasto-viscoplastic behavior of high density polyethylene (HDPE). The work consists of three parts: 1) the experimental investigation, some uniaxial cyclic tests were performed in various maximum strain levels; 2) a constitutive model based on the tests data was developed using the thermodynamic approach of nonlinear relaxation phenomena; 3) predict the whole of experimental results.

II. EXPERIMENTAL PROCEDURE

A. Material – Preparation Procedure

The high density polyethylene investigated in this study was supplied by Roehling Company. The glass transition temperature and the melting point are –125°C and 135°C respectively. Its molecular weight distribution is M_w=50000g/mol. Differential Scanning Calorimetry (DSC) and the X-ray experiments indicate a crystallinity ratio of about 66%. The density of the material, measured by the hydrostatic weighting is ρ=0.964g/cm^3.

The tensile specimens were machined by milling from 6mm extruded sheet, relative to the extrusion direction Fig. 1. This specific geometry of specimen has been designed, in order to control the appearance of necking and to localize the deformation (damage) in the central part where the mechanical variables are determined. Before any mechanical tests, samples were submitted to annealing heat treatment for 24 hours at 120°C in a vacuum, and then they were cooled slowly to obtain a microstructural state without residual stresses.

Fig. 1 Geometry of tensile specimen (dimension in mm)

B. Experimental Results

The mechanical behavior of HDPE was characterized at large deformation under series of uniaxial cyclic tests with maximum strains; ε_1 max =0.2, 0.5, 0.8, 1.1 and 1.4. In each cyclic, a specimen was loaded up to a maximum strain ε_1 max with the strain rate of ė =0.001s⁻¹, unloaded down to zero stress with the strain rate of - ė, reloaded up to the maximum strain with the strain rate ė, etc. Each test consisted at least 14 cycle of deformation. To avoid bulking of sample during unloading stage, an anti-buckling system was used.

All tests were conducted at controlled room temperature on a universal hydraulic tensile machine (MTS 810) equipped with a system for measuring the true local strain without any
contact with the specimen, called Videotraction [7], [8]. This optical extensometer gives access to the elasto-viscoplastic response and volume change of polymer under test by determining in real time the true strain in the 3 orthotropic directions of a representative volume element situated at the center of the neck. This technique is based on follow up, using CCD camera interfaced with a computer, of 7 markers printed on the center of specimen surface, 5 makers are aligned along tensile axis and the two others according to transverse direction.

The evaluation of true stress – volume strain – true strain curves for cyclic deformation with various maximum strains are presented in the Fig. 2. Similar observations were reported by [4]-[6] for HDPE, except that we are more interested to the finite deformation and volume strain during the cycles.

Contrarily to the stress responses, this effect will disappear and the volume strain tends to stabilize around specific values at greater numbers of cycles.

Generally the following conclusions can be drawn from the results; The plastic strain, measured at zero stress, increase with number of cycles The most pronounced growth of the plastic strain occurs during the first three cycles of deformation. The maximum true stress per cycle of the HDPE also changes with number of cycles. Another interesting find of this test was that, the cycle damage leading to increase of the average slop of the hysteresis loops with increasing number of cycles. For comparison with the results of continuous tensile test, the interruption by the cyclic tests does not have any change on the stress and volume strain responses memory.

III. THERMODYNAMIC MODELING OF MECHANICAL BEHAVIOR

A. Background of DNLR Approach

The non linear behavior of high density polyethylene is modeled using a thermodynamic approach called Distribution of Nonlinear Relaxation, DNLR, [9]-[11]. It is based on the study of the internal rearrangements such as relaxation processes from thermodynamic point of view; thermodynamic of irreversible processes. It allows describing, even outside of equilibrium, the evolution of a RVE of materials submitted to the controlled perturbation. Based on the chemical reaction of DE DONDER [12] and the generalization of the fundamental Gibbs equation, one can assume the existence of a thermodynamic potential \( \psi_2 = \psi_1 \) which completely describe the state of material. This potential depends on the state variables \( \gamma \) (controlled variables necessary to characterize the non dissipation evolution of RVE and on the internal microstructural variables \( \beta \) (dissipative variables which represent the different internal reorganization) associated to their dual forces \( \beta \) and \( \gamma \), respectively.

The global response \( \beta \) (observable variables) and the thermodynamic forces \( \gamma \) (Affinities of DE DONDER) are defined like the first derivatives of the potential \( \psi = \psi_1 \) which directly leads to the following nonlinear incremental law:

\[
\frac{\beta}{\gamma} = \frac{\dot{\gamma}^2}{\gamma^2} + \frac{\dot{\beta}}{\gamma} + \frac{\dot{\gamma}}{\gamma^2}
\]

\[
A = -b \dot{\gamma} - g \dot{\gamma}
\]

where \( a^u = \frac{\partial^2 \psi_1}{\partial \gamma^2} \) is Tisza’s stability matrix which couples the state variables \( \gamma \) and \( \beta \) to each others. \( g = \frac{\partial^2 \psi_1}{\partial \gamma^2} \) and \( b = \frac{\partial^2 \psi_1}{\partial \gamma^2} \) are the coupling matrices. The DNLR approach is characterized by the following two original features:
i. The distribution of the dissipative modes including nonlinear effects. The spectrum of relaxation time is statistically obtained by using the Prigogine's equipartition of created entropy theorem [13]. In [9], Cunat showed that, this distribution is characterized by the following equations:

\[ P^0_j = B \sqrt{\tau^*_j} \quad \text{and} \quad \sum_{j=1}^{N} P^0_j = 1, \]

\[ B = \frac{1}{\sum_{j=1}^{N} \sqrt{\tau^*_j}} \]  

where, \( \tau^*_j \) represent the relaxation time of the process \( j \), \( P^0_j \) the relative weight of the process \( j \), \( N \) number of dissipative modes, \( B \) a normalization constant and the index \( r \) means relaxed.

ii. The modeling of the relaxation time based on activation state theory. This theory rests on the existence of a transient state in a complete thermodynamic nonequilibrium state. The relaxation time of process \( j \) can then write:

\[ \tau^*_j = \frac{h}{k_b T} \exp \left( \frac{\Delta F^\tau_j}{RT} \right) \]  

where, \( \Delta F^\tau_j \) represents the activation Helmholtz free energy of the spontaneous fluctuation around the relaxed state for the dissipation mode \( j \). \( h, k_b \) and \( T \) are constants respectively of Plank, Boltzmann and perfect gases. \( T \) represents Kelvin temperature. To take into account the nonlinearity for the situations far from the equilibrium, the relaxation times become dependent on the actual state of the continuous media as:

\[ \tau_j = \frac{h}{k_b T} \exp \left( \frac{\Delta F^\tau_j}{RT} \right) \exp \left( \frac{\Delta(\Delta F^\tau_j(t))}{RT} \right) \]  

\[ \tau^j = \tau^*_j a_j(t) \]  

with \( a_j(t) = \exp \left( -V^\tau_j (\beta - \beta^\tau_j) \right) \) and \( \tau^j = \frac{h}{k_b T} \exp \left( \frac{\Delta F^\tau_j}{RT} \right) \),

where \( a_j \) represent the nonlinearity shift factor, which is the same for all processes \( a_j(t) = a_j(t) \), associated to the activation volume average \( V^\tau_j \). The general constitutive equation of DNLR is given by

\[ \dot{\sigma} = \sum_{j=1}^{N} \dot{\sigma}^j = \sum_{j=1}^{N} \left( P^0_j E^\tau_j \dot{\varepsilon} - \frac{\sigma^j - P^0_j \sigma^\tau_j}{a_j(t) \tau^*_j} \right) \]

where, \( u \) and \( r \) denote the instantaneous and relaxed values, respectively.

In the field of mechanical engineering, considering the case of tensile test, \( \dot{\varepsilon} \mapsto \dot{\varepsilon} \cdot \beta \mapsto \dot{\sigma}^j \), at imposed strain \( \dot{\varepsilon} \), the constitutive equation of DNLR approach can be written in the following incremental equation:

\[ \dot{\sigma} = \sum_{j=1}^{N} \dot{\sigma}^j = \sum_{j=1}^{N} \left( P^0_j E^\tau_j \dot{\varepsilon} - \frac{\sigma^j - P^0_j \sigma^\tau_j}{a_j(t) \tau^*_j} \right) \]

where, \( \{ P^0_j E^\tau_j \} \) represents the instantaneous response for a mode \( j \). \( E^\tau_j \) is a Young's modulus, and \( \{ \sigma^j - P^0_j \sigma^\tau_j \} / a_j(t) \tau^*_j \) represent the dissipation part of response for a mode \( j \), which is governing the evolution of relaxed state. To complete the modeling, it is necessary to describe the definition and the role of the relaxed state \( \sigma^\tau \) and the relaxation times \( \tau^j \) (in particular the shift factor \( a_j(t) \)).

B. Modeling of Non Linear Behavior of HDPE

The mechanical behavior of semi crystalline polymers as characterized by elasto-viscoplastic deformation is intimately related to their two phase microstructures, constituted by amorphous and crystalline phases. The elasto-viscoplastic behavior strongly depends on the degree of crystallinity, the initial crystallographic texture and molecular orientation, as well as the evolution of this microstructure during the deformation [1], [14]-[16]. Thus, in our case of HDPE, one must take into account the existence of two phases which is the key in determining the overall macroscopic mechanical behaviour of HDPE.

Based on the deformation mechanisms of HDPE [15], [1], [17], [18], the constitutive model can be obtained after considering three distinct internal reorganization families. The first family is denoted \( \mathcal{Z} \) and it is related to the relaxation mechanisms that depend on the imposed strain rate (viscous process), mainly due to the amorphous phase. The other one is denoted \( (\mathcal{P}) \) for the mechanisms, non viscous, that are independent of this strain rate due to the fragmentation of the crystallites (plastic process). A third family \( \mathcal{F} \) is corresponding to the phenomena of damage by cavitation.

In the case of uniaxial tensile test, \( (\dot{\varepsilon}_t, \varepsilon_t) \) is applied and \( (\sigma_t, \epsilon_t, \varepsilon_t, \ldots) \) is measured knowing that \( \sigma_2 = \sigma_3 = 0 \), which can be treated as controlled variables. One can chose, thus, a new thermodynamic potential as \( \phi = \phi(\varepsilon, \sigma, \gamma; \mathcal{Z}, \mathcal{P}, \mathcal{F}) \). The system of constitutive equations that take into account these three types of internal reorganizations can be written in the DNLR form as:

\[ \dot{\sigma} = \sum_{j=1}^{N} \dot{\sigma}^j = \sum_{j=1}^{N} P^0_j E^\tau_j \dot{\varepsilon} - \frac{\sigma^j - P^0_j \sigma^\tau_j}{a_j(t) \tau^*_j} \]
\[
\begin{pmatrix}
\sigma_i \\
\varepsilon_i \\
-\frac{A}{\bar{\lambda}}
\end{pmatrix}
= \begin{pmatrix}
a_{i,j} & a_{i,j} & a_{i,j} \\
a_{i,j} & a_{i,j} & a_{i,j} \\
a_{i,j} & a_{i,j} & a_{i,j}
\end{pmatrix}
\begin{pmatrix}
\sigma_j \\
\varepsilon_j \\
\sigma_{a,i}
\end{pmatrix}
+ \begin{pmatrix}
b_{i,j} \\
b_{i,j} \\
b_{i,j}
\end{pmatrix}
\begin{pmatrix}
\dot{\varepsilon}_i \\
0 \\
0
\end{pmatrix}
\tag{8}
\]

The experimental observations of ROGUE ET al [19] and MRABET ET al [20] show that, for semi crystalline polymer, the relaxed state differs from the true thermodynamic equilibrium state. The relaxation state, which is accessible via relaxation test, tends to the true equilibrium state after a very long relaxation time. Note that, the shapes of relaxed stress – strain curve and the tensile response curves are similar, with the fact that the relaxed stress is independent of the applied strain rate.

Thus to complete the modeling, one admit that the spectrum of the relaxation time associated to the dissipation of viscoplastic modes exhibits the same spectrum of damage modes, and the fragmentation of crystals which occurs much later on the time scale depending on another spectrum. These observations and hypotheses, with the analysis steps of DNLR approach (presented in details in [11], [21], lead to the following differential equation:

\[
\dot{\varepsilon}_i = \sum_{j=1}^{N} \sigma_{i}^{\text{j}} \dot{\varepsilon}_j = \sum_{j=1}^{N} \left( \sigma_{i}^{\text{j}} - P_{i}^{\text{j}} \sigma_{i}^{\text{\alpha}} \right) / \tau_{i}^{\text{\alpha}} \tag{9}
\]

\[
\varepsilon_i = \sum_{j=1}^{N} \varepsilon_{i}^{\text{j}} = -V_{i}^{\text{\alpha}} \dot{\varepsilon}_i - \sum_{j=1}^{N} \left( \frac{\varepsilon_{i}^{\text{j}}}{E_{i}^{\text{\alpha}}} \right) \left( \sigma_{i}^{\text{j}} - P_{i}^{\text{j}} \sigma_{i}^{\text{\alpha}} \right) / \tau_{i}^{\text{\alpha}} \tag{10}
\]

with \(\varepsilon_i = \dot{\varepsilon}_i \) for transversally isotropic strain case, which is experimentally proved for this HDPE in the extrusion direction [22]. The volume strain can be determined as

\[
\varepsilon_i = \varepsilon_{\text{i}} + 2\varepsilon_{\text{a}} \tag{11}
\]

The relaxed stress evolution is governed by the following relation in DNLR form as:

\[
\sigma_{i}^{\text{\alpha}} = \sum_{j=1}^{N} \sigma_{i}^{\text{j} \text{\alpha}} = E_{i}^{\text{\alpha}} \dot{\varepsilon}_i - \sum_{j=1}^{N} \left( \frac{\varepsilon_{i}^{\text{j} \text{\alpha}}}{E_{i}^{\text{\alpha}}} \right) \left( \sigma_{i}^{\text{j}} - P_{i}^{\text{j}} \sigma_{i}^{\text{\alpha}} \right) / \tau_{i}^{\text{\alpha}} \tag{12}
\]

where \(E_{\text{\alpha}}\) is the instantaneous modulus of elasticity, \(E_{i}^{\text{\alpha}}\) the damaged modulus of elasticity at relaxed state. \(\sigma_{i}^{\text{\alpha}}\) and \(\sigma_{i}^{\text{\alpha}}\) are respectively the relaxed and equilibrium stress, corresponding to their relaxation time \(\tau_{i}^{\text{\alpha}}\) and \(\tau_{i}^{\text{\alpha}}\) of the \text{j}\text{\alpha} mode. The global response of the material is the sum of each modal stress represented by \(\sigma_{i}^{\text{j}}\). The weight of each mode is obtained by \(p_{i}^{\text{j}} = p_{i}^{\text{j}} + (p_{i}^{\text{j}} - p_{i}^{\text{j} \text{\alpha}}) / 2\), with \(p_{i}^{\text{j}} = 1 / N [20]\).

One of the objectives in this modeling is to take into account the effect of strain rate, with respect the independency of the relaxed state of the strain rate which agrees with the experimental results. We propose to introduce this effect in the nonlinearity shift factor, \(a_{i} (\varepsilon)\) which will take place in both \(\tau_{i}^{\text{\alpha}}\) and \(\tau_{i}^{\text{\alpha}}\). The relaxation spectrums which control the phenomenon of relaxation from the instantaneous state to the relaxation state and from the relaxation state to the equilibrium state are:

\[
\tau_{i}^{\text{\alpha}} = h / k_{i} T \exp \left( \frac{\Delta E_{i}^{\text{\alpha}}}{RT} \right) \exp \left( -V_{i}^{\text{\alpha}} \sigma_{i}^{\text{\alpha}} / RT \right) \tag{13}
\]

\[
\tau_{i}^{\text{\alpha}} = h / k_{i} T \exp \left( \frac{\Delta E_{i}^{\text{\alpha}}}{RT} \right) \exp \left( -V_{i}^{\text{\alpha}} \sigma_{i}^{\text{\alpha}} / RT \right) \tag{14}
\]

with

\[
\tau_{i}^{\text{\alpha}_{\text{max}}} = h / k_{i} T \exp \left( \frac{\Delta E_{i}^{\text{\alpha}}}{RT} \right)
\]

and

\[
\tau_{i}^{\text{\alpha}_{\text{max}}} = h / k_{i} T \exp \left( \frac{\Delta E_{i}^{\text{\alpha}}}{RT} \right)
\]

where \(\exp [-V_{i}^{\text{\alpha}} \sigma_{i}^{\text{\alpha}} / RT] \) and \(\exp [-V_{i}^{\text{\alpha}} \sigma_{i}^{\text{\alpha}} / RT] \), the nonlinearity shift factor \(a_{i} (\varepsilon)\).

The term \(\varepsilon_{\text{i}} / \varepsilon^{\text{\alpha}}\) represents the effect of strain rate in \(a_{i} (\varepsilon)\), \(m\) is the strain rate sensitivity factor. Indeed \(m=1\) when the material behavior is independent of strain rate as in the relaxed state. \(\dot{\varepsilon}_{\text{i}}\) is the critical strain rate below which the tensile behavior is no more dependent on the strain rate [19]. \(V_{i}^{\text{\alpha}} = V_{0} \exp (k_{i} e^{b_{i}})\), where \(V_{0}\) is the activation volume which increases with deformation (damage effect), \(k_{i}\) and \(b_{i}\) are the parameters. \(V_{i}^{\text{\alpha}}\), the activation volume which is related to the fragmentation/orientation mechanisms of crystallites lamellae.

The modeling of equilibrium stress, \(\sigma_{i}^{\text{\alpha}}\), is based on a statistical model with eight-chain as proposed by Arruda and Boyce [23], which is related to hyperelastic hardening at large strain of rubber and other polymers, and due to changes in molecular chains configuration.

\[
\sigma_{i}^{\text{\alpha}} = \frac{N k_{i} T}{3 \lambda_{i}} \eta^{\alpha} \left( \lambda_{i} - 2 \lambda_{2} \right) \varepsilon^{\text{\alpha}} \left( \frac{\lambda_{2}}{n^{\alpha}} \right) \tag{15}
\]

with \(\dot{\lambda}_{2} = \exp (\varepsilon)\), \(\lambda_{2} = \exp (\varepsilon / 2) \sqrt[3]{\lambda_{i}}\), \(\lambda_{i} = \sqrt{\lambda_{i}^{2} + 2 \lambda_{2}^{2} / 3}\), and \(\varepsilon^{\text{\alpha}} (\lambda_{2} / n^{\alpha})\) is the inverse of the Langenvin function. The
term $Nk_B T$ represents the initial shear modulus, and $n$ the number of rigid segments per macromolecular chain [20].

The damage may be represented by an elastic properties change. In empirical way, the evolution of $E^{\text{off}}$ and $\nu^{\text{off}}$ can be modeled by considering two physical phenomena, the first contribution (microcavities) leading to fall of the elastic modulus and the second representing their increase (reorientation of chains) [22].

$$E^{\text{off}}_{i} = E_{i}' \left[ 1 - [\alpha_i(1 - \exp(-\beta_i \varepsilon_i)) - X \ell^{-1} (\lambda_i^n)] \right]$$ (16)

$$\nu^{\text{off}}_{ij} = \nu_{ij}' \left[ 1 - [\alpha_{ij}(1 - \exp(-\beta_i \varepsilon_{ij})) + \varepsilon_i^n] \right]$$ (17)

The above equation can be written in the following form:

$$E^{\text{off}}_{i} = E_{i}' \left[ 1 - D_{1i} \right], \quad \nu^{\text{off}}_{ij} = \nu_{ij}' \left[ 1 - D_{ij} \right]$$ (18)

where $D_{1i}$ and $D_{ij}$ are the damage variables resulting from the two contributions

$$D = D_{\text{cavitation}} + D_{\text{orientation}}$$

with

$$X = \frac{1}{n^{0.5}} \frac{3n - 1}{n - 1}$$

$\alpha_i$ represents the level of maximum damage, $\beta_i, \alpha_{ij}$ and $\beta_{ij}$ (characterizing the damage evaluation) are the parameters to be adjusted.

C. Validation of Modified Model

Our aim now is to demonstrate the capability of the model to produce the elasto-viscoplastic behavior of HDPE during cyclic tests, and particularly to predict the volume strain. Seventeen parameters must be identified to run the model, the values of the required material parameters for HDPE were obtained using the Sido software. $E' = 1200$MPa, $V_0 = 1305$ cm$^3$/Mol at, $k_1 = 0.56$, $k_2 = 1.99$, $\tau_{\max} = 39800$sec, $E^{\text{off}} = 330$MPa, $\alpha_1 = 0.899$, $\beta_1 = 18$, $\tau_{\text{max}}^{\text{off}} = 19\times10^7$sec, $\nu^{\text{eq}} = -490$ cm$^3$/Mol, $\dot{\varepsilon}_0 = 9\times10^2$sec$^{-1}$, $m = 0.18$, $Nk_B T = 0.53$MPa, $n = 90$, $\nu_{ij}' = 0.448$, $\alpha_{ij} = 0.229$, $\beta_{ij} = -1.58$, $\delta_{ij} = 0.05$. Note that, all simulations were obtained by using the same set of parameters.

Figs. 3 and 4 show the simulated stress response of uniaxial cyclic tensile tests at different maximum strain levels. Most features of stress–strain behavior during the tests, initial stiffness, and non linear response during loading and unloading and hardening zone were well predicted over the complete range of the test program. For volume strain during cyclic deformation, a quite nice agreement is observed between the predicted behavior and experimental results, even at large deformation Fig. 4.

IV. CONCLUSIONS

The non linear behavior of high density polyethylene was studied experimentally under series of uniaxial cyclic tests over a range of different maximum strain levels. Volume changes, measured during cyclic test of HDPE, have revealed that damage process are significant and have to be taken into account.

Fig. 3 Simulation results for true stress–strain curve during uniaxial cyclic tensile tests at different maximum strain levels and strain rate of $0.001/s$.

Fig. 4 Simulation results for volume strain during uniaxial cyclic tensile tests at different maximum strain levels and strain rate of $0.001/s$.

An elasto-viscoplastic model for the constitutive behavior of semi-crystalline polymers has been developed in the
framework of thermodynamic of irreversible process. The
DNLR approach was used and modified. To take into account
the morphology and deformation mechanism (including damage), a new thermodynamic potential was proposed and
coupled with DNLR approach. The introduction of two
spectrum of relaxation time, allowed describing the relaxation and
equilibrium states. This approach leads to very general
formalism which well adapted to the complex loading path. It
allows to describe behavior which highly non linear. By
comparing the predictions from the proposed constitutive
model with experimental data for cyclic tensile tests, one
concludes that the modified model correctly describes
observations on HDPE.

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