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# Experimental Investigation and Constitutive Modeling of Volume Strain under Uniaxial Strain Rate Jump Test in HDPE

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***Abstract*—**In this work, tensile tests on high density polyethylene have been carried out under various constant strain rate and strain rate jump tests. The dependency of the true stress and specially the variation of volume strain have been investigated, the volume strain due to the phenomena of damage was determined in real time during the tests by an optical extensometer called Videotraction. A modified constitutive equations, including strain rate and damage effects, are proposed, such a model is based on a non-equilibrium thermodynamic approach called (DNLR). The ability of the model to predict the complex nonlinear response of this polymer is examined by comparing the model simulation with the available experimental data, which demonstrate that this model can represent the deformation behavior of the polymer reasonably well.

***Keywords*—**Strain rate jump tests, Volume Strain, High Density Polyethylene, Large strain, Thermodynamics approach.

## I. Introduction

HIGH density polyethylene (HDPE) is one of an important technological material, because it is considered as of the most best candidates for many industrial applications. Many researchers have focused on understanding their nonlinear elasto-viscoplastic behavior and modeling their mechanical behavior under different loading conditions [1]-[4].

The analysis studies of the nonlinear behavior of semi crystalline polymers at different strain rates have received a considerable interest because the mechanical properties and deformation mechanisms are strongly dependent on the applied strain. There are several experimental studies and mechanical models that have been developed on polymers to characterize the strain rate history effect [2], [5], [6], but limited researches were carried out to investigate this effect during strain rate jump test, especially for the variation of volume strain response which not seen, to our knowledge, in the literature.

In this work, examples will be given of the observable mechanical behavior of HDPE in uniaxial strain rate jump tests and monotone tensile tests at various strain rates, in order to analysis the influence of strain rate on the nonlinear behavior of semi crystalline polymers and to highlight the ability of HDPE to resume its initial behavior. In next section, a constitutive model based on the thermodynamic approach of nonlinear relaxation phenomena, DNLR is developed to predict the whole of experimental results.

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## II. Material and Experimental Procedure

In this research, the test material was HDPE (high density polyethylene, grade 500 natural) produced by Roechling Engineering Plastics in form of 6mm thick extruded sheets. The supplier indicates that the molecular weight distribution and density are respectively $M_w$= 500000 g/mol and $\rho=0.964\ g/cm^3$, the glass transition temperature and the melting point are – 125°C and 135°C. The crystallinity ratio, obtained from differential scanning calorimetry and from the x-ray is about 66%.

Test samples were cut from the polymer sheet by milling along the extrusion direction following the shape represented in Fig. 1. This specific geometry has been designed, with the median region of cube of 6mm, in order to control the appearance of necking and to localize the deformation in this predetermined zone where the mechanical variables are determined.

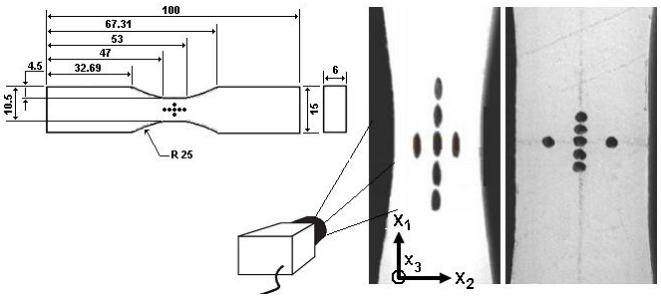



Fig. 1 Tensile specimen (dimension in mm)

The influence of strain rate and strain rate jump on the elasto-viscoplastic behavior, particularly on volume strain, of HDPE was characterized at large deformation under series of uniaxial tensile tests at several constant strain rates. The experiments were carried out at controlled temperature of 23°C on a universal hydraulic tensile machine (MTS 810) equipped with a Video extensometer, called Videotraction, which gives access to the elasto-viscoplastic response and volume change of polymer under test by determining in real time the true strain in the three orthotropic directions of a representative volume element situated at the center of the neck. An important feature of the Videotraction is its ability to

impose in real time any desired input path of local strain rate and keep it constant during the test [7], [8].

Based on Videotraction technique, seven dot markers printed on the center of specimen surface Fig.1, five makers are aligned along tensile axis and the two others according to transverse direction. The measurement of local deformation were done by follow up the displacement of the center of gravity of seven dot markers, using CCD camera interfaced with a computer.

## III. Experimental Results

The influence of strain rate on stress and volume are shown in the Fig. 2. As indicate in previous studies [1], [2], [5], [7], [9], [8], there is a pronounced effect, as increasing the strain rate remarkably, the true stress increases at all strains. The initial young's modulus appears also to be strain rate dependant. For the volume strain evaluation, the results show a highly strain rate dependent. For all strain rates, before the yield strain, there is a monotone increase of volume strain observed at very beginning of deformation, which may be attributed to the contributions from elastic expansion and cavitations [1], [10]. The maximum of 0.05 is reached when the strain is around 0.1. After the yield point the volume strain decrease approximately to its initial value, this may be attributed to the volume compaction of the amorphous due to rearrangement of lamellae orientation accompanied by the changing of voids shape and/or to the closure of small cavities initially presented in the HDPE [11], [12]. When the strain is larger than 0.5, again the volume strain increases gradually which seems to be connected the formation and growth of cavities [1], [10], [13]. It is clear from the results that, before the yield point there is no strain rate dependant, once the plastic stage begins, the phenomenon of volume compaction decrease as increasing the strain rate, and for the fastest strain rate, the onset of the volume dilatation took place much earlier.

Fig. 3 shows the results obtained for the strain rate jump test. The strain rate, initially equal to $0.001s^{-1}$, is increased to $0.005s^{-1}$ then to $0.01s^{-1}$ for a selected time intervals. We reverse the transient regime, Fig. 4; the initial strain rate is increased to $0.01s^{-1}$ and then decreased to $0.005s^{-1}$. For comparison, also the results of continuous tensile test at various strain rates were included. An instantaneous stress increase is immediately observed after the strain rate is increased; the stress attaints the level it would have held if the new strain rate had been used from the beginning of the test. Hence, the interruption by the strain rate jump does not change the memory of the previous strain rate. This test indicates that the stress response of HDPE does not have strain rate history dependency. Contrarily for the evaluation of volume strain, the memory effect during the transient regime of strain rate change is very important, the volume strain increase with the increasing of the strain rate but the HDPE keeps this change in volume although the strain rate return to its initial value, one can attributed this phenomenon to the microcavities, which are already created during the deformation in the material, does not change its size even after decreasing strain rate. Therefore the volume strain evaluation of HDPE have a highly strain rate history dependency.

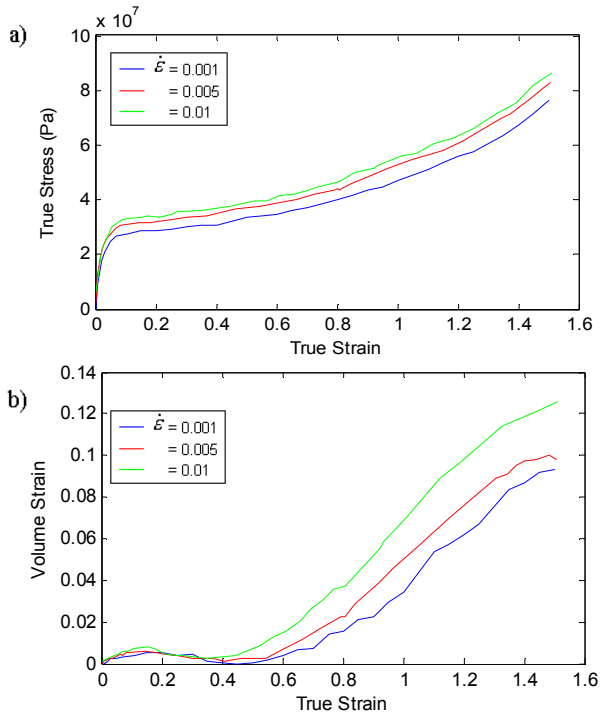



Fig. 2 Influence of strain rate on mechanical behavior of HDPE; a) True stress vs. true strain and b) Volume strain vs. true strain

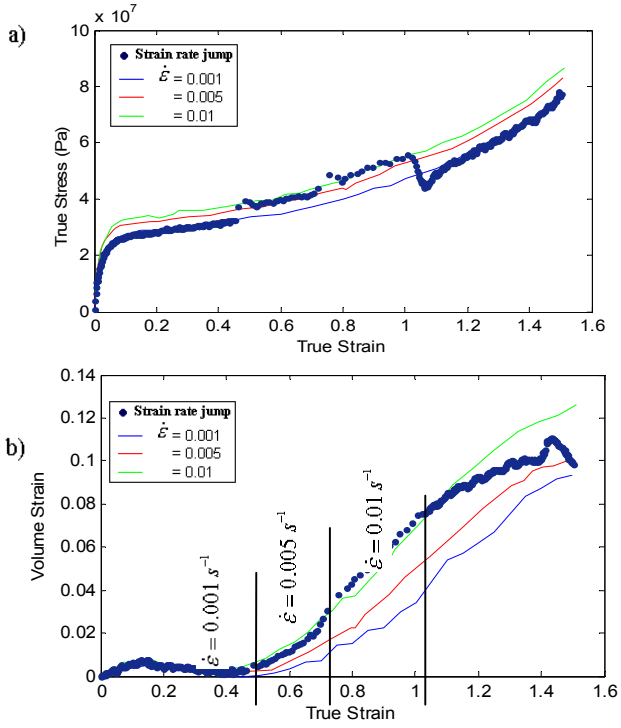



Fig. 3 Experimental results for a strain rate jump tests; a) True stress vs. true strain, and b) Volume strain vs. True strain

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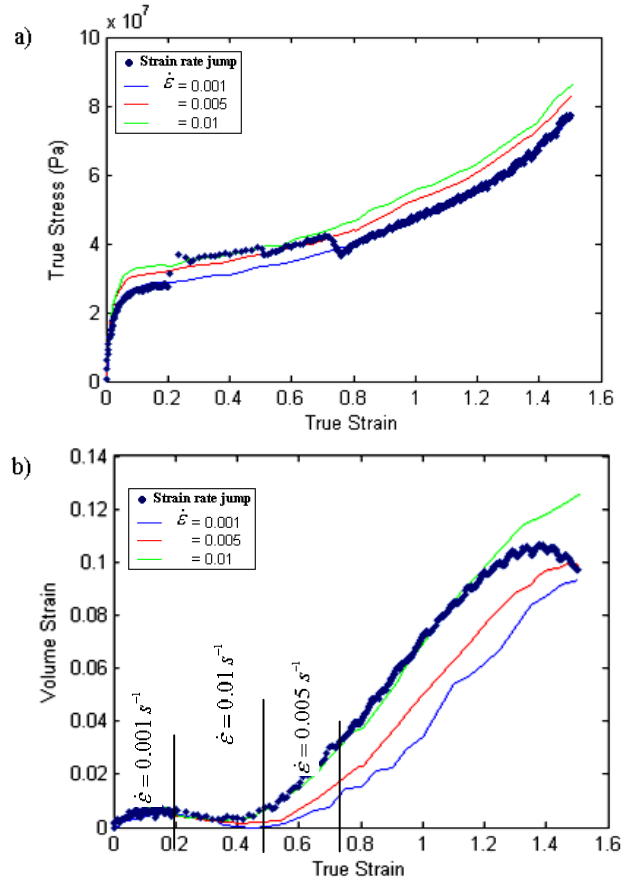



Fig. 4 Experimental results for a strain rate jump tests; a) True stress vs. true strain, and b) Volume strain vs. True strain

## IV. Thermodynamic Modeling of Mechanical Behavior

### *A. Thermodynamic Bases of DNLR Modeling*

On the bases of thermodynamic of irreversible processes, the mechanical behavior of HDPE is modeled. The model used in this study is referred as DNLR, Distribution of Nonlinear Relaxation [14], [15]. This thermodynamic approach is closely related to the relaxed state concept, which is defined as the results of microstructural evolution after loading followed by complete stress relaxation. The internal reorganization processes is considered in this formalism as a chemical reaction. Its originality comes from its expression of the relaxation spectrum, the introduced of non-linearity and the specificity of the relaxed state.

Based on the analysis of the chemical reaction of DE DONDER [16] and the generalization of the fundamental Gibbs equation for the system even outside equilibrium, the existence of a thermodynamic potential $\psi = \psi(\underline{\gamma}, \underline{\bar{z}})$ can be taken, which completely describe the state of material. This potential depends on the state variables $\underline{\gamma}$ (controlled variables necessary to characterize the non dissipation evolution) and on the internal microstructural variables $\overline{Z}$ (dissipative variables which represent the different internal reorganization) associated to their dual forces $\underline{\beta}$ and $\overline{A}$, respectively. The response $\underline{\beta}$ (observable variables) and the forces $\overline{A}$ (Affinities of DE DONDER) are defined like the first derivatives of the potential $\psi$ which directly leads to the following general constitutive equation in rate form:

$$\begin{aligned} \dot{\underline{\beta}} &= \underline{\underline{a}}^{u}.\dot{\underline{\gamma}} + \underline{\overline{b}}.\dot{\overline{z}} \\ \dot{\overline{A}} &= -\underline{\overline{b}}^{t}.\dot{\underline{\gamma}} - \overline{\overline{g}}.\dot{\overline{z}} \end{aligned} \tag{1}$$

where $\underline{\underline{a}}^{u} = \partial^2 \psi_k(\underline{\gamma}, \overline{z}) / \partial \underline{\gamma}^2$ is Tisza's stability matrix which couples the state variables $\underline{\gamma}$ and $\underline{\beta}$ to each others. $\underline{\overline{b}} = \partial^2 \psi_k(\underline{\gamma}, \overline{z}) / \partial \underline{\gamma} \partial \overline{z}$ and $\overline{\overline{g}} = \partial^2 \psi_k(\underline{\gamma}, \overline{z}) / \partial \overline{z}^2$ are the coupling matrixes.

The originality of DNLR approach is characterized by two essential keys: 1)- The distribution of the dissipative modes including nonlinear effects, and 2)- The modeling of the relaxation time based on activation state theory.

The spectrum of relaxation time is statistically obtained by using the Prigogine's equipartition of created entropy theorem [17]. In [14], Cunat proposed a universal spectrum which is characterized by the following equations:

$$P_0^j = B\sqrt{\tau_j^r} \text{ and } \sum_{j=1}^{N} P_0^j = 1, \text{ with } B = \frac{1}{\sum_{j=1}^{N} \sqrt{\tau_j^r}} \tag{2}$$

$\tau_j^r$ represents 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.

According to Eyring's theory, the existence of a transient state in a complete thermodynamic non-equilibrium state is postulated. The relaxation time of process $j$ can then write:

$$\tau_j^r = \frac{h}{k_B T} \exp\left(\frac{\Delta F_j^{+,r}}{RT}\right) \tag{3}$$

where, $\Delta F_j^{+,r}$ represents the activation Helmholtz free energy of the spontaneous fluctuation around the relaxed state for the dissipation mode $j$. $h$, $k_B$ and $R$ 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:

$$\tau_j = \frac{h}{k_B T} \exp\left(\frac{\Delta F_j^{+,r}}{RT}\right) \exp\left(\frac{\Delta\left(\Delta F_j^{+}(t)\right)}{RT}\right) \tag{4}$$

$$\tau^j = \tau_j^r a_f(t) \tag{5}$$

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with $a_f^j(t)=\exp\left(\frac{-V^+(\beta-\beta^r}{RT}\right)$ and $\tau_j^r=\frac{h}{k_B T}\exp\left(\frac{\Delta F_j^{+,r}}{RT}\right)$, where $a_f$ represent the nonlinearity shift factor, which is the same for all processes $a_f^j(t)=a_f(t)$, associated to the activation volume average $V^+$.

The general constitutive equation of DNLR is given by

$$\underline{\dot{\beta}}=\sum_{j=1}^{N}\underline{\beta}^j=\sum_{j=1}^{N}\left(P_0^j\,\underline{\underline{a}}^u.\underline{\dot{\gamma}}-\frac{\underline{\beta}^j-\underline{\beta}^{j,r}}{a(t)\tau_j^r}\right) \tag{6}$$

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

In the field of mechanical engineering, considering the case of tensile test, at imposed strain $\dot{\varepsilon}$, the overall true stress $\sigma^r$ is the sum of all modal components $\sigma^j$ and the general relation of DNLR approach can be written in the following incremental equation:

$$\dot{\sigma}=\sum_{j=1}^{N}\dot{\sigma}^j=\sum_{j=1}^{N}\left(P_0^j\,E^u\,\dot{\varepsilon}-\frac{\sigma^j-P_0^j\sigma^{j,r}}{a_f(t)\tau_j^r}\right) \tag{7}$$

where, $\left(P_0^j\,E^u\,\dot{\varepsilon}\right)$ represents the instantaneous response for a mode $j$, $E^u$ *is a* Young's modulus, and $\left(\sigma^j-P_0^j\sigma^{j,r}/a_f(t)\tau_j^r\right)$ 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 $\sigma^r$ and the relaxation times $\tau^j$ (in particular the shift factor $a_f(t)$).

*B. Application to HDPE*

The plastic damage deformation of most of semi crystalline polymers under uniaxial tension induced an important large strain deformation accompanied with volume change. This behavior is strongly depending on their two phases microstructures, constituted by crystalline lamellae and macromolecules in an amorphous state. The elasto-viscoplastic behavior strongly related to its degree of cristalinety, initial crystallographic texture and molecular orientation, as well as the evolution of this microstructure during the deformation [1], [18], [19]. Thus, in the case of HDPE, the existence of two phases must take into account which is the key in determining the overall macroscopic mechanical behaviour of HDPE.

Based on the deformation mechanisms of HDPE [20], [1], [10], [13], the constitutive model can be obtained after considering three distinct internal reorganization families. The first family is denoted $\bar{z}$ 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 ( $\bar{\rho}$ ) for the mechanisms, non viscous, that are independent of this strain rate due to the fragmentation of the crystallites (plastic process). A third family ( $\bar{e}$ ) is corresponding to the phenomena of damage by cavitation.

For uniaxial mechanical test with imposed strain, $\sigma_1,\varepsilon_2$ *and* $\varepsilon_3$ are measured knowing that $\sigma_2=\sigma_3=0$, which can be treated as controlled variables. One can chose, thus, a new thermodynamic potential as $\phi=\phi\left(\varepsilon_1,\sigma_2,\sigma_3,T;\bar{z};\bar{\rho};\bar{e}\right)$.

The experimental observations of Roguet et al. [21] and Mrabet et al. [22] 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, for modeling the relaxations spectrum, 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 crystallites 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 [15], [23], lead to the following differential equations:

$$\dot{\sigma}_1=\sum_{j=1}^{N}\dot{\sigma}_1^j=E_1^u\,\dot{\varepsilon}_1-\sum_{j=1}^{N}\left(\frac{(\sigma_1^j-P_j^r\sigma_1^{j,r})}{\tau_j^r}\right) \tag{8}$$

$$\dot{\varepsilon}_2=\sum_{j=1}^{N}\dot{\varepsilon}_2^j=-\nu_{12}^{r,eff}\dot{\varepsilon}_1-\sum_{j=1}^{N}\left(\frac{\delta_{12}}{E_1^{r,eff}}\frac{(\sigma_1^{j,r}-P_j^r\sigma_1^{j,eq})}{\tau_j^{eq}}\right) \tag{9}$$

with $\dot{\varepsilon}_3=\dot{\varepsilon}_2$ for transversally isotropic strain case, which is experimentally proved for this HDPE in the extrusion direction [25]. The volume strain can be determined as $\varepsilon_v=\varepsilon_1+2\varepsilon_2$.

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

$$\dot{\sigma}_1^r=\sum_{j=1}^{N}\dot{\sigma}_1^{j,r}=E_1^{r,eff}\,\dot{\varepsilon}_1-\sum_{j=1}^{N}\left(\frac{(\sigma_1^{j,r}-P_j^r\sigma_1^{j,eq})}{\tau_j^{eq}}\right) \tag{10}$$

where $E^u$ is the instantaneous modulus of elasticity, $E^{r,eff}$ the damaged modulus of elasticity at relaxed state. $\sigma^r$ and $\sigma^{eq}$ are respectively the relaxed and equilibrium stress, corresponding to their relaxation time $\tau_j^r$ and $\tau_j^{eq}$ of the j$^{th}$ mode. The global response of the material is the sum of each modal stress represented by $\sigma_1^j$. The weight of each mode is obtained by $p_j^r=p_j^0+\left(p_j^u-p_j^0\right)/2$, with $p_j^u=1/N$ [22].

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, $a_v(\dot{\varepsilon})$ which will take place in both

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$\tau_j^r$ and $\tau_j^{eq}$. 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:

$$\tau_j^r = \frac{h}{k_B T}\exp\left(\frac{\Delta F^+}{RT}\right)\exp\left(\frac{-V\left|\sigma - \sigma^r\right|}{RT}\right)\left(\frac{\dot{\varepsilon}_0}{\left|\dot{\varepsilon}\right|}\right)^{1/m} \quad (11)$$

$$\tau_j^{eq} = \frac{h}{k_B T}\exp\left(\frac{\Delta F^{+,eq}}{RT}\right)\exp\left(\frac{-V^{eq}\left|\sigma^r - \sigma^{eq}\right|}{RT}\right)\left(\frac{\dot{\varepsilon}_0}{\left|\dot{\varepsilon}\right|}\right) \quad (12)$$

with $\tau_{\max}^r = \frac{h}{k_B T}\exp\left(\frac{\Delta F^+}{RT}\right)$ and $\tau_{\max}^{eq} = \frac{h}{k_B T}\exp\left(\frac{\Delta F^{+,eq}}{RT}\right)$. Where $\exp\left(-V\left|\sigma - \sigma^r\right|/RT\right)$ and $\exp\left(-V^{eq}\left|\sigma^r - \sigma^{eq}\right|/RT\right)$, the nonlinearity shift factor $a_f(t)$.

The term $\left(\dot{\varepsilon}_0/\left|\dot{\varepsilon}\right|\right)^{1/m}$ represents the effect of strain rate in $a_v(\dot{\varepsilon})$, m is the strain rate sensitivity factor. Indeed m=1 when the material behavior is independent of strain rate as in the relaxed state. $\dot{\varepsilon}_0$ is the critical strain rate below which the tensile behavior is no more dependent on the strain rate [21]. $V = V_0\exp(k_1\varepsilon^{k_2})$, where $V_0$ is the activation volume which increases with deformation (damage effect), $k_1$ and $k_2$ are the parameters. $V^{eq}$, the activation volume which is related to the fragmentation /orientation mechanisms of crystallites lamellae.

The modeling of equilibrium stress, $\sigma^{eq}$, is based on a statistical model with eight-chain as proposed by Arruda and Boyce [24], which is related to hyperelastic hardening at large strain of rubber and other polymers, and due to changes in molecular chains configuration.

$$\sigma_1^{eq} = \frac{Nk_B T}{3\lambda_c} n^{0.5}\left(\lambda_1^2 - \lambda_2^2\right)\ell^{-1}\left(\frac{\lambda_c}{n^{0.5}}\right) \quad (13)$$

with $\lambda_1 = \exp(\varepsilon)$, $\lambda_2 = \exp(-\varepsilon/2) = 1/\sqrt{\lambda_1}$, $\lambda_c = \sqrt{\lambda_1^2 + 2\lambda_1^2/3}$, and $\ell^{-1}\left(\lambda_c/n^{0.5}\right)$ is the inverse of the Langevin function. The term $Nk_BT$ represents the initial shear modulus, and n the number of rigid segments per macromolecular chain.

The damage may be represented by an elastic properties change. In empirical way, the evolution of $E^{r,eff}$ and $\nu_1^{r,eff}$ 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) [25].

$$E_1^{r,eff} = E_1^r\left[1 - \left[\alpha_1\left(1 - \exp(-\beta_1\varepsilon_1)\right) - \mathrm{X}\ \ell^{-1}\left(\frac{\lambda_c}{n^{0.5}}\right)\right]\right] \quad (14)$$

$$\nu_1^{r,eff} = \nu_{12}^r\left[1 - \left[\alpha_{12}\left(1 - \exp(-\beta_{12}\varepsilon_1)\right) + \varepsilon_1^2\right]\right] \quad (15)$$

The above equation can be written in the following form:

$$E_1^{r,eff} = E_1^r\left[1 - D_{11}\right], \quad \nu_1^{r,eff} = \nu_{12}^r\left[1 - D_{12}\right] \quad (16)$$

where $D_{11}$ and $D_{12}$ are the damage variables resulting from the two contributions $D=D_{cavitation}+D_{orientation}$, with $\mathrm{X} = \sqrt{\frac{1}{n^{0.5}}}\frac{3n-1}{n-1}$, $\alpha_1$ represents the level of maximum damage, $\beta_1$, $\alpha_{12}$ and $\beta_{12}$ (characterizing the damage evaluation) are the parameters to be adjusted.

### C. Validation of Modified Model

The numerical simulation of the model are generated and compared with the experimental data, to produce the elasto-viscoplastic behavior of HDPE during strain rate jump test, and particularly to predict the volume strain. An explicit integration schema in Matlab was used to numerically integrate our model. All simulations were obtained by using the same set of parameters, the values of materials parameter that need to determine, obtained using the SiDolo software, are,

$E^u$ = 1180 MPa, $V_0$ = -1444 cm$^3$/Mol at, $k_1$=0.525, $k_2$= 2, $\tau_{\max}^r$ = 50300 sec, $E^{r,eff}$ = 295 MPa, $\alpha_1$= 0.899, $\beta_1$ = 18, $\tau_{\max}^{eq}$ = 10*10$^7$ sec, $V^{eq}$ = -480 cm$^3$/Mol, $\dot{\varepsilon}_0$ = 12*10e$^{-7}$ sec, m= 0.18, $Nk_BT$= 0.60 MPa, n= 90, $\nu_{12}^r$=0.465, $\alpha_{12}$=0.2105, $\beta_{12}$=-1.638, $\delta_{12}$=0.029.

As shown in Figs. 5 & 6, the results clearly show the ability of the model to correctly predict the behavior and the strain rate history effect for HDPE. The highly nonlinear and strain rate dependent behavior of stress – strain curve were in an excellent agreement with experimental observations during this kind of tests. For comparison with the evolution of volume strain during strain rate jump test, also a quite nice agreement is observed between the predicted behavior and experimental results, even at large deformation.

## V. Conclusions

A series of uniaxiale tensile tests at various strain rates and strain rate jump tests were performed in order to study the influence of strain rate dependency on the deformation behavior of HDPE, practically on the volume strain. The mechanical response of HDPE is significant affected by the strain rate. No strain rate history dependency for the stress response was found during strain rate jump test, but it was have a highly history dependency for the volume changes.

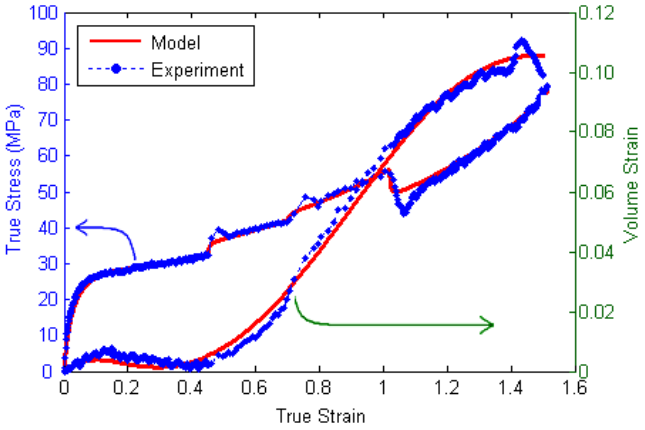



Fig. 5 Simulation results for a strain rate jump tests. The strain rate of $0.001s^{-1}$ was suddenly changed to $0.005s^{-1}$, then to $0.01s^{-1}$ for a time interval, then back to $0.001s^{-1}$

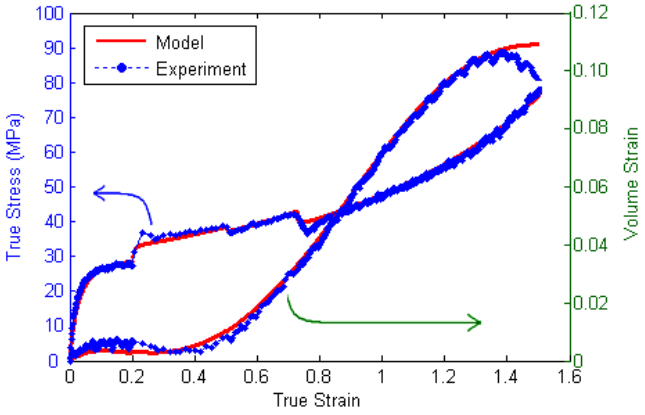



Fig. 6 Simulation results for a strain rate jump tests. The strain rate of $0.001s^{-1}$ was suddenly increased to $0.01s^{-1}$, and then decreased to $0.005s^{-1}$ for a time interval, then back to $0.001s^{-1}$

Based on the distribution of non linear relaxation approach, a model of mechanical behavior was developed taking into account the two phases microstructures and deformation mechanism (including damage) of HDPE. A new thermodynamic potential was proposed and coupled with DNLR approach which allowed describing the relaxation and equilibrium states. The theoretical simulations have shown how the modified model cans correctly describe the experimental observation and the deformation histories even in large strain during strain rate jump tests on HDPE.

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