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A RHEOLOGICAL ANALYSIS OF SOLID POLYMERS USING AN INVERSE METHOD APPLIED TO A FINITE ELEMENT MODEL OF THE TORSION AND TENSILE TESTS

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The torsion and traction mechanical tests are generally used in order to analyses the rheology of solid polymers. This paper deals with an identification of the rheological constitutive equation for a polycarbonate and a polyethylene material. To take into account strain localization, caused by increasing values of the plastic strain and of the temperature gradients, a finite element simulation of the experimental tests is used. Then, the values of the constitutive parameters are computed from an Inverse Analysis method. Accuracy of the material behavior identification results obtained by the Inverse Finite Element Method will be demonstrated.

Key words: Rheology, Solid Polymers, Torsion, Tensile, Inverse Analysis.

1. INTRODUCTION

Recent developments of the industrial forming processes of solid polymers, such as the thermoforming, the stamping or the fiber stretching [1, 2] show that these materials are subject to important thermoplastic phenomena. At the macroscopic scale is observed several instabilities, as for example the diffuse necking or shear band formation [3]. This last one corresponds generally to the strain or temperature localization mechanisms [4]. Moreover it is necessary to reflect the structural modifications undergone by the material during its plastic deformation Today, the use of a numerical modeling of forming processes, such as the Finite Element Method, requires a well known of the intrinsic constitutive equation of the material. This equation must express the influence of strain, strain rate and temperature on the plastic flow stress defined by the Von-Mises yield criterion [5, 6]. Identification of all the constitutive parameters is today very important for numerical simulations. For this purpose classical mechanical tests are used, and, after the computation of the true stress, true strain and strain rate, a non-linear regression method gives the values of the constitutive parameters. According to the possibility to simulate numerically the experimental tests, new methods, based on the Inverse Analysis principle, were developed [7, 8]. The principal goal of this paper is to develop a more rigorous analysis of the formulation and the identification of the rheological behavior of two different solid polymers: an amorphous one and a semi-crystalline one. The importance of the Inverse Analysis will be shown starting from comparison of results with those obtained from classical analytical method.

2. CONSTITUTIVE EQUATION

To describe the viscosity of polymers and its thermoplasticity, Eyring proposes to use a theory of molecular dynamics starting from thermal activates motions and local jumps around existing physical "barriers" [9]. Taking into account the frequency of probable forward and back lock jumps, the equivalent strain rate can be written from the Eyring equation by the following relationships:

$$\dot{\bar{\varepsilon}}_f = \dot{\bar{\varepsilon}}_0 \exp\left(-\frac{G_0 - \bar{\sigma} V_{\exp}}{k_B T}\right) \text{ respectively,} \tag{1}$$

$$\dot{\bar{\varepsilon}}_b = \dot{\bar{\varepsilon}}_0 \exp\left(-\frac{G_0 + \bar{\sigma} V_{\exp}}{k_B T}\right).$$

Here $\dot{\bar{\varepsilon}}_0$ is a reference strain rate value, G_0 is the free energy, k_B represents the Boltzman constant, T is the temperature and $V_{\exp}$ is the experimental local activation volume.

Starting from a phenomenological description and from an experimental analysis made by Sellars and Tegart, the total strain rate $\dot{\bar{\varepsilon}}$ is obtained from the difference between the forward strain rate $\dot{\bar{\varepsilon}}_f$ and the back lock one $\dot{\bar{\varepsilon}}_b$, and can be written in the general form [10]:

$$\dot{\bar{\varepsilon}} = \dot{\bar{\varepsilon}}_0 \underbrace{\left[\exp\left(\frac{\bar{\sigma} V_{\exp}}{k_B T}\right) - \exp\left(-\frac{\bar{\sigma} V_{\exp}}{k_B T}\right)\right]}_{2\sinh(\alpha\bar{\sigma})} \exp\left(-\frac{G_0}{k_B T}\right) \approx A\dot{\bar{\varepsilon}}_0 \left[\sinh(\alpha\bar{\sigma})\right]^p \exp\left(-\frac{G_0}{k_B T}\right), \tag{2}$$

where A, α, p are material specific parameters.

This formula permits to write the Von-Mises stress expression:

$$\bar{\sigma} = \frac{1}{\alpha} \operatorname{Arcsinh}\left[\frac{1}{A}\frac{\dot{\bar{\varepsilon}}}{\dot{\bar{\varepsilon}}_0} \exp\left(\frac{Q}{RT}\right)\right]^{1/p}, \tag{3}$$

where $Q = RG_0 / k_B$ represents the activation energy and R the perfect gas constant.

For small values of the plastic strain rate (available for quasi-static and static loads), the Arcsinh function ($\operatorname{Arcsinh}(x) = \sinh^{-1}(x) = \ln\left(x + \sqrt{1+x^2}\right)$) degenerate mathematically in a linear function (Arcsinh(x)=x) and consequently it is possible to write the law (3) in the Norton-Hoff form [11]:

$$\bar{\sigma}\left(\bar{\varepsilon}, \dot{\bar{\varepsilon}}, T\right) = K\left(\bar{\varepsilon}\right)\sqrt{3}(\sqrt{3}\dot{\bar{\varepsilon}})^m \exp\left(\frac{\beta}{T}\right). \tag{4}$$

Here $\bar{\varepsilon}$ represents the cumulated plastic strain, K is the material hardness function, m=1/p is the strain rate sensitivity parameter and $\beta = mQ/R$. For solid polymers undergoing large plastic deformations, G'Sell [5, 12] proposes to express the material hardness function variation by:

$$K\left(\bar{\varepsilon}\right) = K_0\left[1 - \exp(-w\bar{\varepsilon}\right] \quad \left[1 + c\exp\left(-d\bar{\varepsilon}\right)\right]\exp(h\bar{\varepsilon}^n). \tag{5}$$

The first strain function corresponds to the visco-elastic phenomenon, the second one describes the plastic "hook" of the stress and the third one is linked to the structural material hardening. Relationships (4) and (5) are generally used to describe the experimental stress-strain curves of a lot of solid polymers in amorphous or semi-crystalline state. In the case of high values of generalized strain rates (more great that 100 s^{-1}), many authors proposes to use a logarithmic law, which can be easy obtained from the relationship (3), if an asymptotical approximation of Arcsinh function is used, i.e. $\operatorname{Arcsinh}(x) \approx \ln(2x)$. In this case the constitutive equation can be written in the following form:

$$\bar{\sigma}\left(\bar{\varepsilon}, \dot{\bar{\varepsilon}}, T\right) = K\left(\bar{\varepsilon}\right)\left[D' + D\ln\left(\frac{\dot{\bar{\varepsilon}}}{\dot{\bar{\varepsilon}}_0}\right) + \frac{\beta}{T}\right]. \tag{6}$$

Consequently, in order to cover all the strain rates range it is recommended to use the Arcsinh law (3). For quasi-static conditions (generalized strain rates smallest that 10 s^{-1}), the material behavior description can use a Norton-Hoff constitutive equation defined by the relationships (4) and (5).

3. PARAMETER IDENTIFICATION BY INVERSE ANALYSIS

In a classical way, to identify the constitutive parameters values, firstly, is necessary to compute the Von-Mises stress and to determine its variation with the cumulated plastic strain, generalized strain rate and

temperature. This method requires building an analytical model able to express all the above intrinsic thermo-mechanical variables in function of the global experimental measurements corresponding to the mechanical tests: operating conditions, torques, loads, geometry, etc. After the computation of all thermo-mechanical variables, a non-linear regression model, generally based on the least squares method, can be used. According to the actual development of the finite element model it is now possible to simulate numerically the experimental mechanical test. It is then possible to take into account the all complexity of the specimen deformation: strain localization, self-heating phenomenon and variation of specimen shape. Because the experimental space is now defined by the direct measured quantities (loads, torques, and specimen shape variables), the corresponding constitutive parameters can be identified only from an Inverse Analysis Method [7], based on the diagram pictured in Fig. 1.

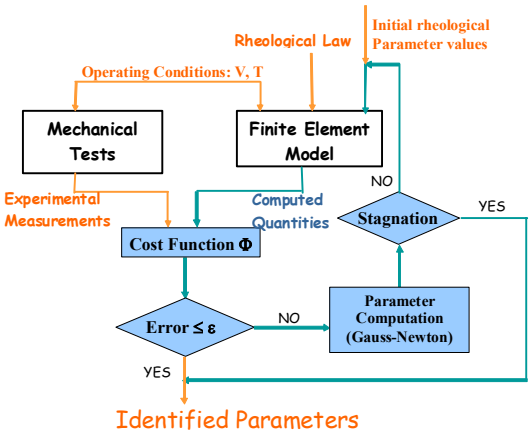


Fig. 1 – Principle of the Inverse Analysis Method.

The principle is to find the parameters values, which minimizes a cost function Φ, expressed in a least squares sense, in terms of the global experimental measurements and of the corresponding numerical values. The iterative parameter computation uses a Gauss-Newton method, which requires evaluation of parameter derivatives of computed finite element quantities.

4. ANALYSIS OF THE TORSION TEST OF A POLYCARBONATE

For a solid amorphous polymer, as a polycarbonate one, to analyze its rheology properties corresponding to large plastic deformations, a torsion test can be used (Fig. 2).

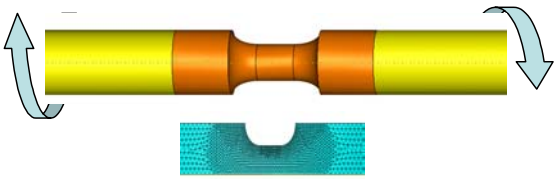


Fig.2 – The mechanical torsion test and the corresponding finite element model.

The useful cylindrical part of the specimen has a length L of 15 mm and a diameter d of 10 mm. A lot of torsion experiments show that a cylindrical specimen keeps constant its axisymmetry. Fields and Backofen have developed an analytical model of the torsion test. It uses some assumptions as the isotropy of the material and the axial homogeneity of mechanical and thermal variables, taking into account only the radial variation of the mechanical variables. In this case the authors demonstrate that in the outside surface of the specimen the Von-Mises stress can be written from Fields and Backofen formulas:

$$\bar{\sigma}\big|_{r=R} = \frac{\sqrt{3}C(R)}{2\pi R^3}\left(3+\tilde{n}+\tilde{m}\right), \tag{7}$$

where:

$$\tilde{n} = \left(\frac{\partial \ln C}{\partial \ln N} \right)\Big|_{\dot{N}=\text{cte}} \text{ and } \tilde{m} = \left(\frac{\partial \ln C}{\partial \ln \dot{N}} \right)\Big|_{N=\text{cte}} . \tag{8}$$

Here C is the torque, R is the radius of the cylindrical specimen corresponding to the useful part with the length L, N is the rotation number and $\dot{N}$ is the rotation speed (corresponding to the number of the rotations per second). The corresponding cumulated plastic strain and the generalized strain rate can be computed with the formula:

$$\dot{\bar{\varepsilon}}\big|_{r=R} = \frac{2}{\sqrt{3}} \pi \dot{N} \frac{R}{L} \text{ and } \bar{\varepsilon}\big|_{r=R} = \int_0^t \dot{\bar{\varepsilon}} \mathrm{d}\tau = \frac{2}{\sqrt{3}} \pi N \frac{R}{L}. \tag{9}$$

Three different torsion tests have been made at the room temperature for three different rotation speeds: 0.48 rpm (a), 0.048 rpm (b) and 0.0048 rpm (c). These values correspond respectively to a maximum generalized strain rate of 10^{-2} s^{-1}, 10^{-3} s^{-1} and 10^{-4} s^{-1}. A classical non-linear regression of the analytical method, in the stress-strain-strain rate space, gives the parameters values presented in Table 1. The corresponding cost function or error function is written by:

$$\Phi(P) = \left\{ \sum_{i=1}^{N\exp} \left[\sigma_i^{\exp} - \sigma_i^{c} \right]^2 \right\} / \left\{ \sum_{i=1}^{N\exp} \left[\sigma_i^{\exp} \right]^2 \right\}, \tag{10}$$

where $N_{\exp}$ represents the total number of the experimental points, $\sigma^{\exp}$ are the "experimental" stress values estimated from (7) and σ^{c} are the stress values computed from (4) and (5).

To take into account the all complexity of the torsion test phenomena, especially the thermal changes between the specimen and the tool device and the possible localization of the strain, strain rate and temperature, a complete finite element simulation it is proposed. All the parameters are now identified by an Inverse Analysis, starting directly form the measured torques (Table 1).

Table 1

Parameter Identification of a polycarbonate behavior from torsion tests (β = 774) using formulas (4), (5), (10) and (11)

Parameters	Identification		
	Initial Values	Analytical Model	Inverse Analysis
K_0[MPa s^m]	5.8	8.07	4.502
w	1.	0.84	3.505
c	10.	9.28	7.395
d	10.	9.60	13.470
h	0.	-0.075	0.328
n	2.	2.	2.
m	0.001	0.002	0.033
R^2	-	0.884	0.972

The cost function, which can be minimized with respect to all the parameters, is defined now from the global quantities represented here by the torques values:

$$\Phi(P) = \left\{ \sum_{i=1}^{N\exp} \left[C_i^{\exp} - C_i^{FE} \right]^2 \right\} \cdot \left\{ \sum_{i=1}^{N\exp} \left[C_i^{\exp} \right]^2 \right\}^{-1} . \tag{11}$$

Figure 3 shows that the estimated stress-strain curves are very different between those obtained from the classical model and those obtained from the Inverse Analysis. The more important differences are caused by the strain rate sensitivity parameter. Moreover the true stress has a scale factor which changes from 1.25 to 2 between the Inverse Analysis method and the analytical one. This error can be caused by the wrong evaluation of the analytical model of the useful specimen length L and of the torques derivatives with respect to the rotation number and rotation speed (formulas 8).

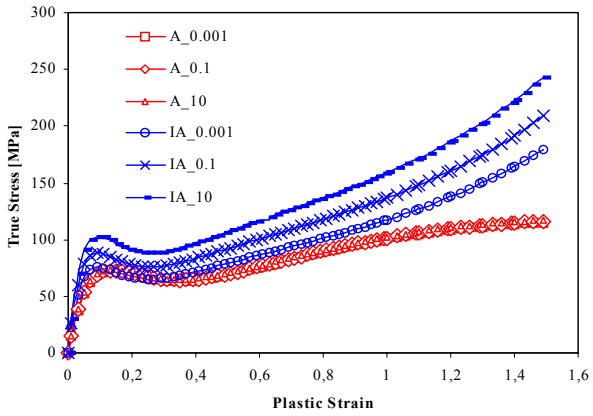
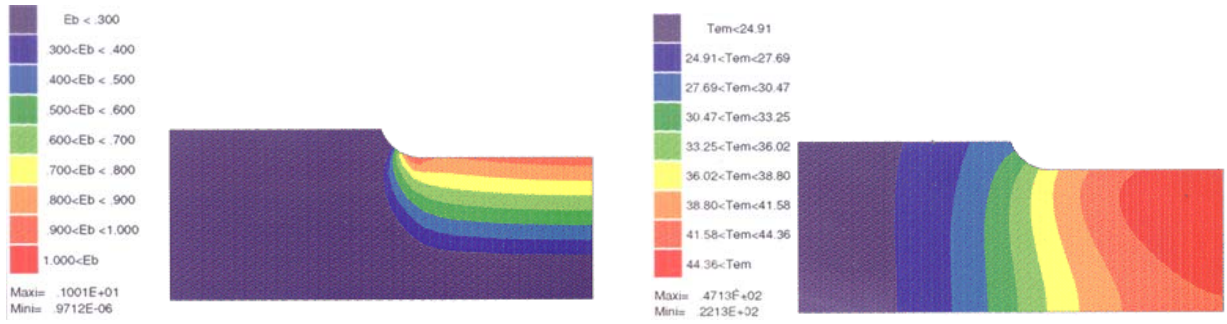


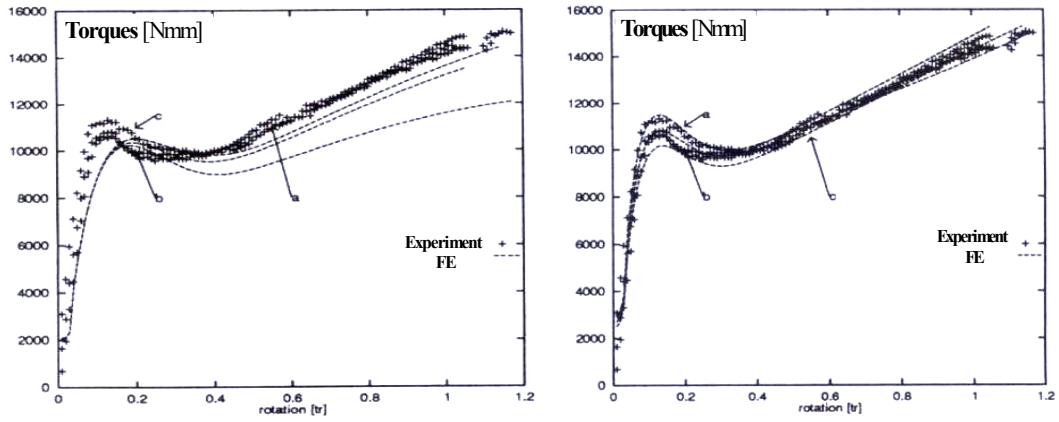
Fig. 3 – True stress – true strain curves of the solid Polycarbonate behavior obtained from analytical identification (A) and from the Inverse Analysis one (IA), for different strain rate values (0.001 s^{-1}, 0.1 s^{-1} and 10 s^{-1}).

Numerical simulations show that softening of the material after the "hook" of the stress and the self-heating of the specimen, due to the plastic work, leads to a localization of the plastic strain, which modify the spatial distribution of the all kinematics and thermo-mechanical variables (Fig. 4).



a) b)

Fig. 4 – Finite element simulation of the torsion test for the Polycarbonate at 0.48 rpm: a) distribution of the plastic strain; b) distribution of the temperature.



a) b)

Fig. 5 – Comparison of the experimental torques with the numerical ones (FE): a) for parameters identified from analytical model; b) for parameters identified with Inverse Analysis.

Moreover, Fig. 5a illustrates that, if a classical analytical model is used for the parameter identification, the prediction of the torques variation is far from the experiment (global error of cost function around 12%). Regarding the torques variation obtained from parameters identified by Inverse Analysis (Fig. 5b), the

agreement with the experimental values is more precisely (global error of 2.8%). It can be then concluded that the true stress values are, in this case, more correctly estimated.

5. ANALYSIS OF THE TENSILE TEST OF A POLYETHYLENE

For a semi-crystalline solid polymer, as the polyethylene one, the torsion test shows that the cylindrical sample has important change of the shape. In this case it is necessary to use a 3D finite element simulation, taking into account physical phenomena coupled to the observed anisotropy of the material in the radial-axial space. A more simple choice is to use the tensile test. In order to have a better control of the necking phenomenon, a dumbbell cylindrical specimen is used. In this case it is necessary to add experimental measurements of the central shape of the specimen, using video metric equipment (Fig. 6). Then the global experimental measurements are represented by the variation of the loads, of the diameter on the central area of the specimen and of the principal curvature radius.

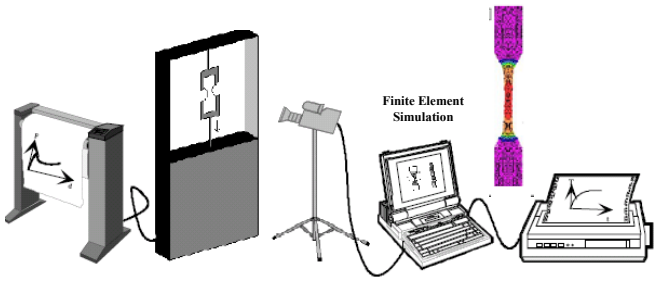


Fig. 6 – Video-Metric measurements for the Tensile Test.

Starting from a Bridgman description of the tensile test, the analytical model, leads to the following relationships:

$$\bar{\sigma} = \frac{4F}{\pi D^2}\left[\left(1+\frac{4\rho}{D}\right)\ln\left(1+\frac{D}{4\rho}\right)\right]^{-1}, \tag{12}$$

with $\dot{\bar{\varepsilon}} = -\frac{2}{D}\left(\frac{\mathrm{d}D}{\mathrm{d}t}\right)$ and $\bar{\varepsilon} = 2\ln\frac{D_0}{D}$.

In these formulas D_0 is the initial diameter of the central area of the specimen (here 6 mm), D is the diameter corresponding to the time t, F is the load and ρ is the curvature radius on the central area of the specimen. The length of the dumbbell specimen is chosen to be equal to 60 mm and the head diameter is equal to 12 mm. Three different tensile tests have been made at room temperature for three different traction speeds: 0.00834 mm/s (a), 0.0417 mm/s (b) and 0.162 mm/s (c), which correspond approximately to an initial generalized strain rate of $0.01s^{-1}$, $0.05s^{-1}$ and $0.2s^{-1}$. The application of a classical non-linear regression method (named Analytical Method) leads to the parameter values indicated in the Table 2.

Table 2

Parameter identification of a polyethylene behavior from tensile tests ($c = d = 0$, $\beta = 466$) using formulas (4), (5), (12) and (13)

Parameters	Identification			
	Initial Values	Analytical Method	Inverse Analysis I ($\lambda = 0$)	Inverse Analysis II ($\lambda = 1$)
K_0[MPa s^m]	10.	11.014	6.031	6.039
w	30.	30.786	286.53	322.06
h	0.5	0.515	0.896	1.001
n	2.	2.	1.255	1.215
m	0.1	0.156	0.103	0.121
R^2 (F, R_{min})	–	0.99 (0.864, 0.907)	0.945 (0.971,0.908)	0.954 (0.953, 0.956)

If an Inverse Analysis method, based on a numerical finite element simulation, is used, the parameters have different values. The first Inverse Analysis (I) uses only the tensile loads (F) on the formulation of the cost function and ($\lambda = 0$) and the second one (II), uses simultaneously the load values (F) and the minimal radius ones ($R_{\min}$), according to the following formula:

$$\Phi(P) = \frac{\sum_{i=1}^{N\exp}\left[F_i^{\exp} - F_i^{FE}\right]^2}{\sum_{i=1}^{N\exp}\left[F_i^{\exp}\right]^2} + \lambda \frac{\sum_{i=1}^{N\exp}\left[R_{\min i}^{\exp} - R_{\min i}^{FE}\right]^2}{\sum_{i=1}^{N\exp}\left[R_{\min i}^{\exp}\right]^2}. \tag{13}$$

To explain the difference between the analytical model and the Inverse Analysis methods it is necessary to build the stress-strain curves (Fig. 7) and to compare the prediction of the loads and of the minimal radius of the specimen (Fig. 8 and Fig. 9).

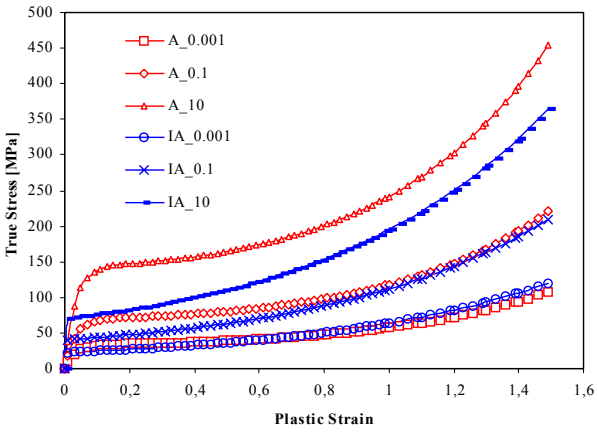
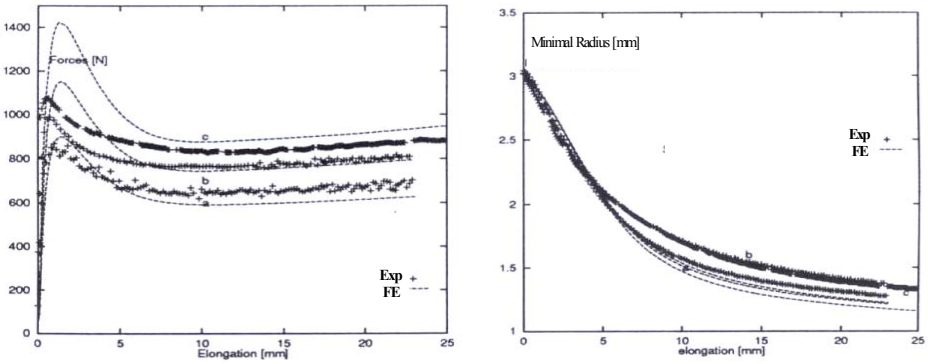


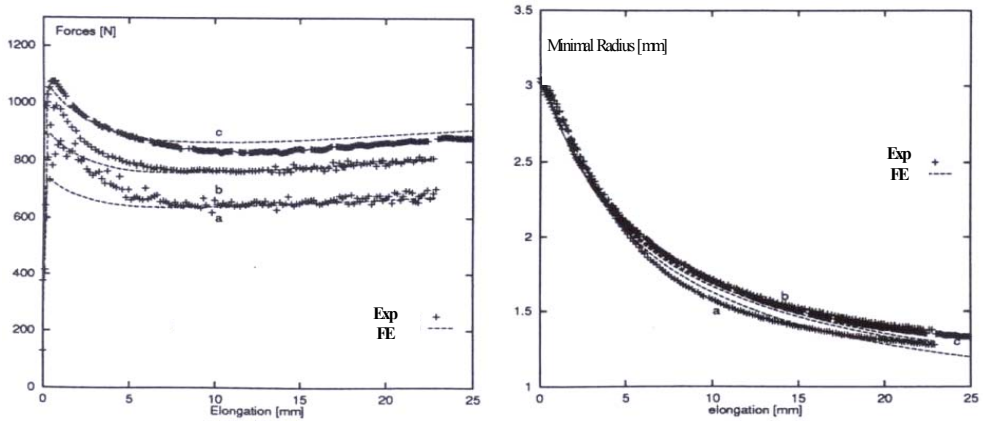
Fig. 7 – True stress – true strain curves of the solid Polyethylene behavior obtained from analytical identification (A) and from the Inverse Analysis II one (IA) for different strain rate values (0.001, 0.1 and 10 s^{-1}).



a) b)

Fig. 8 – Comparison of the experimental data with the numerical ones (FE) obtained from the analytical identification: a) for the loads; b) for the minimal radius.

Regarding the stress-strain curves estimated from the both methods, closed values are obtained for the smallest value of the strain rate. The differences start to be more pronounced for strain rate up to 0.1s^{-1} and with an evident change of the curve shape for the strain rate of 10s^{-1}. It is possible to explain these differences by the specimen temperature increasing linked to the self-heating phenomenon caused by the dissipated plastic energy. In this case, the temperature gradient change the Von-Mises stress distribution on the central area of the specimen and the Bridgman approximation used by the analytical model is no more valid. Moreover finite element prediction of the loads and of the minimal radius variation (Fig. 8) shows that the results are in this case far from the experimental recordings.



a) b)

Fig. 9 – Comparison of the experimental data with the numerical ones (FE) obtained from the Inverse Analysis II: a) for the loads; b) for the minimal radius.

Using the parameter values identified by the Inverse Analysis, a best prediction of the experimental curves was obtained (Fig. 9).

6. CONCLUSIONS

It has been demonstrated that, for solid polymers undergone large plastic strain, the classical hypothesis of the analytical analysis of the specimen deformation during the mechanical tests are generally no valid. Heterogeneity of plastic strain, strain rate and temperature lead to strain localization phenomena and the computed true stress – true strain curve can be wrong. The use of the proposed Inverse Analysis via a numerical simulation of the mechanical test permits to take into account the all complexity of the specimen deformation. This research paper shows that using this Inverse Analysis method it is possible to identify more precisely the intrinsic constitutive equation defining the solid polymer behavior.

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