A Viscoelastic-Viscoplastic Time-Temperature Equivalence for Thermoplastics

Vincent Dorléans [1], Rémi Delille [2], Franck Lauro [2], Delphine Notta-Cuvier [2], Benjamin Bourel [2], Gregory Haugou [2], Bruno Bennani[2], Hervé Morvan [2], Eric Michau[1]

[1] FAURECIA Interior system, Méru, France
[2] University Polytechnique Hauts de France, LAMIH, UMR CNRS 8201 Valenciennes, France

1 Abstract

For automotive suppliers, it is essential to model the behavior of thermoplastics under crash loading and for a large range of temperature typically from -30° until 85℃. Thermoplastics are very sensitive to both strain rate and temperature with an inverse relation: hardening with strain rate and softening with temperature. Generally, a large experimental campaign has to be carried out to identify different behavior laws of the material, each of them for a specific range of strain rate and temperature. Then, according to the characteristics of the loading case, e.g. impact, corresponding behavior laws are chosen in the database to run the numerical simulations. This results in an important experimental cost and a large database to manage. It is then interesting to explore the time-temperature equivalence of thermoplastics to act on both aspects. Relations between strain rate and temperature sensitivities are identified through dynamic mechanical analysis (DMA) in the viscoelastic domain and described through the Williams, Landel and Ferry model or the Arrhenius model for example. For that, a shift factor is experimentally determined and introduced to modify the time step in the behavior model for the finite element simulation, thus simulating an adapted strain rate. As a novelty, the time-temperature equivalence is here extended to the viscoplastic domain by keeping the same shift factor. It therefore becomes possible to cover all the scope of temperature and strain rate of automotive applications from only DMA and tensile tests at room temperature and different strain rates.
This approach is implemented in association with viscoelastic, viscoplastic with non-associative plasticity constitutive laws and non-local damage model [1][2][3] and applied to the case of a polypropylene. The time temperature equivalence is validated for the viscoelastic as well as for the viscoplastic parts of the behavior with good experimental/numerical correlation. As a result, the number of material cards required in Ls Dyna is reduced to only one to cover all the simulations. This approach is also under investigation to be applied to the failure model.

2 Introduction

The studied material is a copolymer propylene ethylene, which is 15 wt% mineral (talc) filled and impact modified (P/E-MD15 impact modified). This type of material is used in the design of injection-molded parts such as door panels, center consoles and dashboards. During a car crash, all the plastic parts are submitted to severe loads. It is the case for the top cover of the dashboard. For example, the deployment of the frontal airbag will generate significant level of deformation on the associated parts up to failure. That is why it is very important to be able to well characterize the behavior of the material in a wide strain-rate range (from 10^{-5} to $300s^{-1}$) and at different temperatures (from -30℃ to +85℃). The main objective is to predict the risk o f failure of the material in case of ductile as well as brittle behavior by combining static and dynamic loadings at different temperature. Therefore, a wide test campaign of characterization is necessary. In this paper, we will assess whether the time temperature superposition principle is applicable for our material in the viscoelastic and viscoplastic domain, so as to drastically reduce the number of tests.

As shown in the literature, the material is very sensitive to different parameters such as strain rate, hydrostatic pressure and temperature. A model that covers the wide range of behavior encountered in automotive applications has been developed by Balieu et al [1][2][3]. This model takes viscoelasticity and viscoplasticity into account with a non-associative approach and a non-local damage model at room temperature. All the parameters of the model are identified thanks to a direct identification method proposed by Epee et al [4][5].

This paper will focus on studying the coupling between the strain rate and the temperature sensitivities by using the theoretical approach called “time temperature superposition principle”. For that, an experimental test campaign has been performed in the framework of small and large deformations. A relationship between the strain rate and the temperature sensitivities is bring out and is mathematically expressed. That equation which allows linking strain rate and temperature effect is identified and then implemented in Balieu's model. An experimental and numerical validation is proposed to validate that time temperature equivalence for viscoelasticity and viscoplasticity. The results of this study and the new implementation in the model will allow reducing significantly the number of physical tests with a new experimental procedure only based on a dynamic mechanical analysis and few tensile tests.

3 Time Temperature Superposition Principle

The time temperature superposition principle is based on the dependence between sensitivity to strain rate and temperature of polymer behavior. Usually the principle is applicable in linear viscoelasticity domain. It is the case for PP [6], PA [7] [8] or PMMA [9] eg. To validate the dependence, it is necessary to characterize the viscoelastic properties of the material by dynamic mechanical analysis. A measurement of storage and loss moduli is done in a frequency range of loading/unloading cycles, at different temperatures. A shifted factor is applied to join all the segments, each of them being representative of behavior under a given temperature, in order to build a master curve (Figure 1). From this master curve, a mathematical model can be identified to link the strain rate and the temperature dependencies. Currently some mathematical models are proposed by Williams-Landel-Ferry, Vogel or Arrhenius [10][11][12][13].

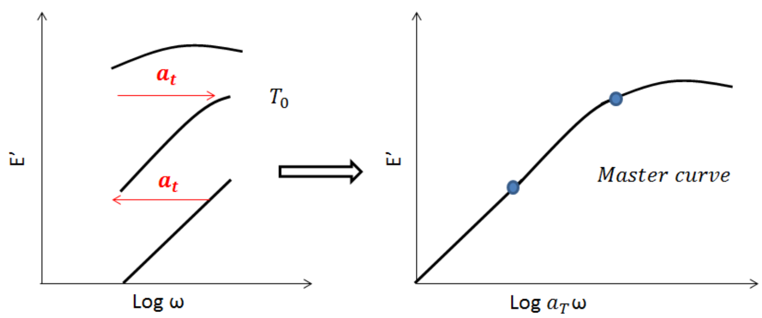

Fig. 1. Building of a master curve

4 Experiments

Viscoelastic properties of the P/E-MD15 impact modified are obtained by performing DMA with an Instron Electropulse E3000 with pneumatic grips. A sinusoidal signal with amplitude of 0.1mm is applied in tension on a sample of rectangular shape (80*10mm). The characterization is achieved in a frequency range between 0.05 and 30Hz at different temperatures from -30℃ to +80℃. For testing performed at cold and high temperatures, a thermo-regulated oven (-100 to 350℃) is used.

The results exposed below describes the evolution of the storage modulus in function of the frequency (or strain rate) for temperature step of 10℃.

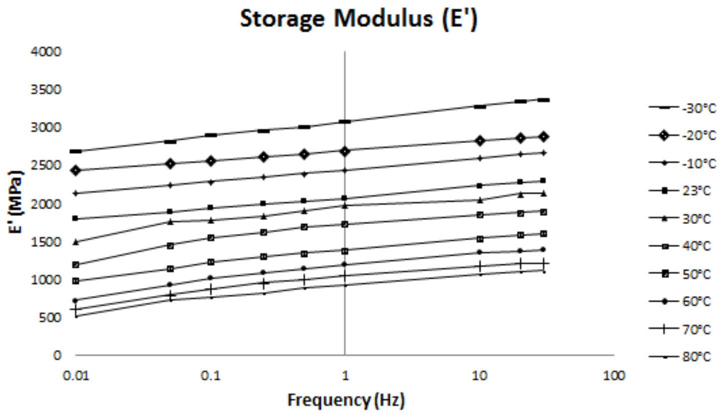

Fig. 2. Storage modulus versus frequency at different temperatures.

Several observations can be drawn from the results. We can observe a linear increase of the storage modulus when increasing frequency (300MPa), with the same slope whatever the temperature. It shows that the influence of the strain rate on material viscoelastic properties is not depending on the temperature. Then, it can be checked whether time temperature superposition principle is applicable for our material.

To build the master curve, a reference temperature has first to be defined. For the study, the reference temperature chosen is 23℃. Then, a shift factor, $a_T\,(T)$, is applied on each E' vs f curves obtained at other temperature, *T*, in order to build the master curve (Figure 3). The relationship between the shift factors and the temperature is given by:

$$a_T\,(T) = exp(-C\,(T - T_{ref})) \qquad (1)$$

Where *T* and T_{ref} are the current temperature and the reference temperature, respectively, and *C* is a constant.

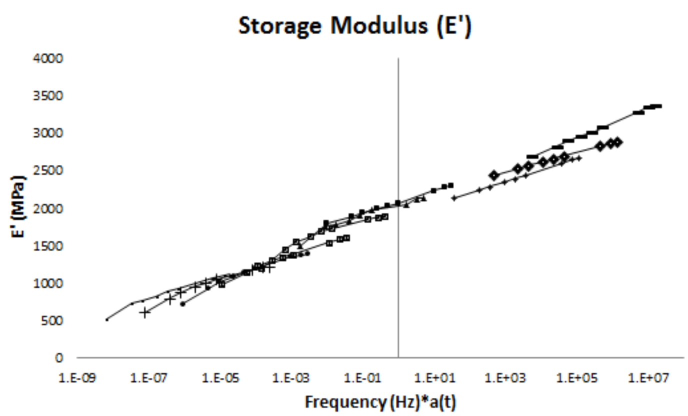

Fig. 3. Storage modulus of the master curve versus frequency range*shift factors

As expected, the obtained master curve has a linear shape and the value of parameter C can be identified at 0.25.
To complete the study, some uniaxial tensile tests (described in details hereafter) have been performed at different temperature and strain rate. The aim was to measure the elastic modulus to be compared to DMA measurements. As the loss modulus is low, the elastic modulus is directly compared to the storage modulus. The figure 4 shows that the two test methods give similar results. Time temperature equivalence principle is therefore validated within the viscoelastic domain.

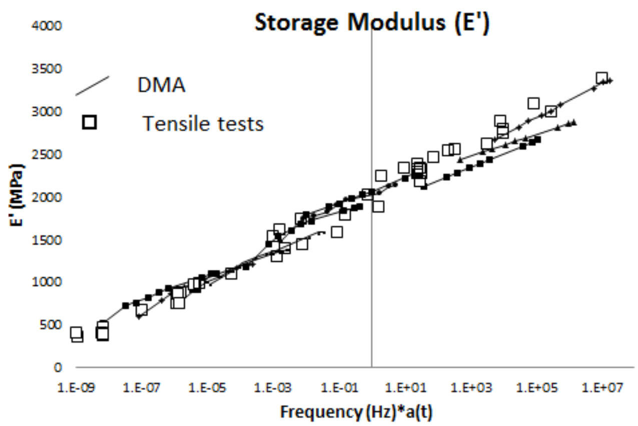

Fig. 4. Determination of the elastic modulus from two test methods: DMA and tensile tests

Next step is to assess the validity of that principle in the viscoplastic domain by using the same shift factor as determined in viscoelasticity. For that it is essential to characterize the viscoplastic behavior of the material. Some uniaxial tensile tests are performed at different strain rate (from 5 $10^{-3}s^{-1}$ to $30s^{-1}$) in a temperature range between -30℃ and +85℃. T he quasi-static tensile tests are carried out on the same test device used for the DMA tests, namely electromagnetic device Instron E3000. For the dynamic loadings, a hydraulic high speed device Instron VHS with a 5kN cell force is used.

2D digital image correlation technique combined with a high speed camera allow measuring the fields of displacements on specimen's surface of interest, thanks to the painting of a random pattern. Some specific technics for pattern creation are employed (black and white colour painting for quasi-static/dynamic loadings at room and high temperature and grease with alumina balls for low temperature).

The different results of the tensile tests between -30℃ and +85℃ at various strain rate ~~s~~ are shown below.

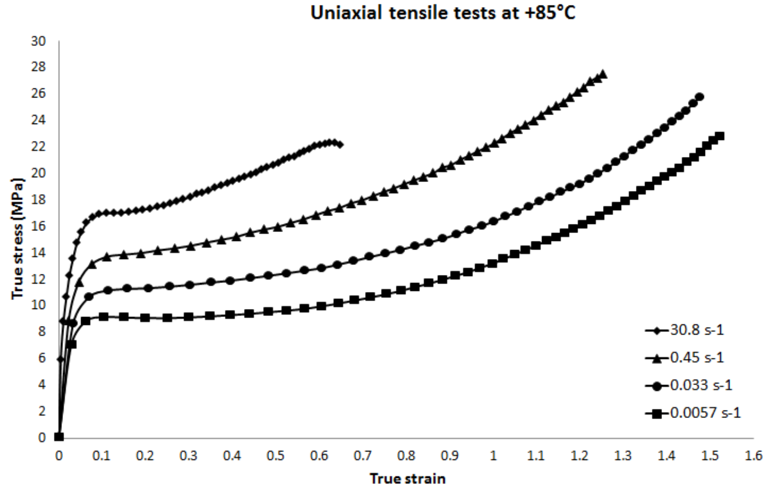

Fig. 5. Uniaxial tensile tests at +85℃

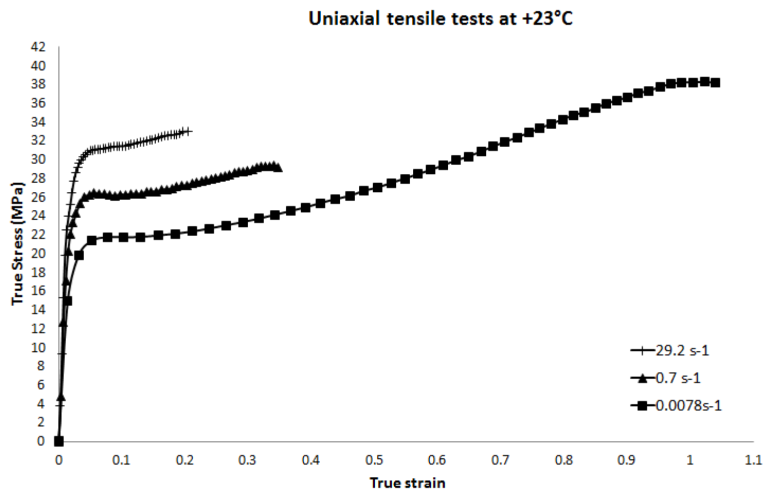

Fig. 6. Uniaxial tensile tests at +23℃

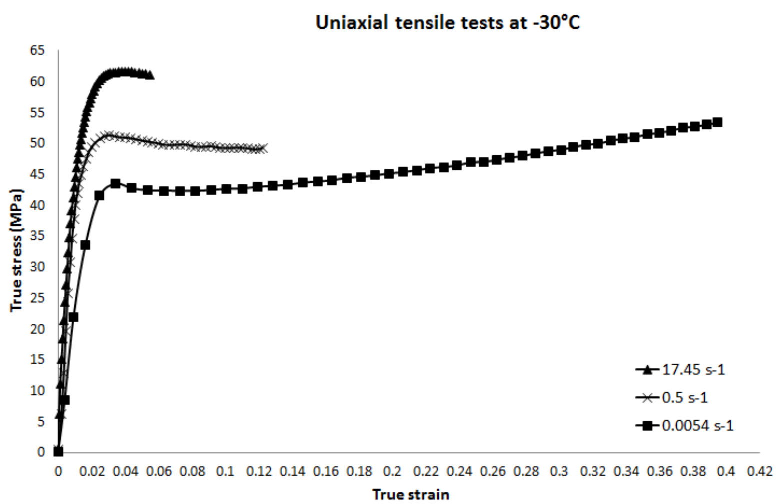

Fig. 7. Uniaxial tensile tests at -30℃

At high temperature (+85℃), the material has a duc tile behavior with a plastic strain level which exceeds 100%. This high deformation level is due to the presence of elastomer domains in the compound of the material. At the contrary, at low temperature (-30℃), the material shows a brittle behavior with a plastic strain level lower than 7%.

The SEE method developed by Epee et al. [4] [5] is used to characterize the viscoplastic behavior of the P/E-MD15 impact modified. From the uniaxial tensile tests, a 3D behavior surface is build. This surface describes the evolution of true stress in function of true plastic strain for a large ~~strain~~ strain-rate range. It is then possible to obtain the response of the material law by “cutting” the surface at the desired strain rate. By using the shift factor defined in viscoelasticity, the time temperature superposition principle is applied on the results of tensile tests which provide a unified SEE surface, as can be seen in Figure 7.

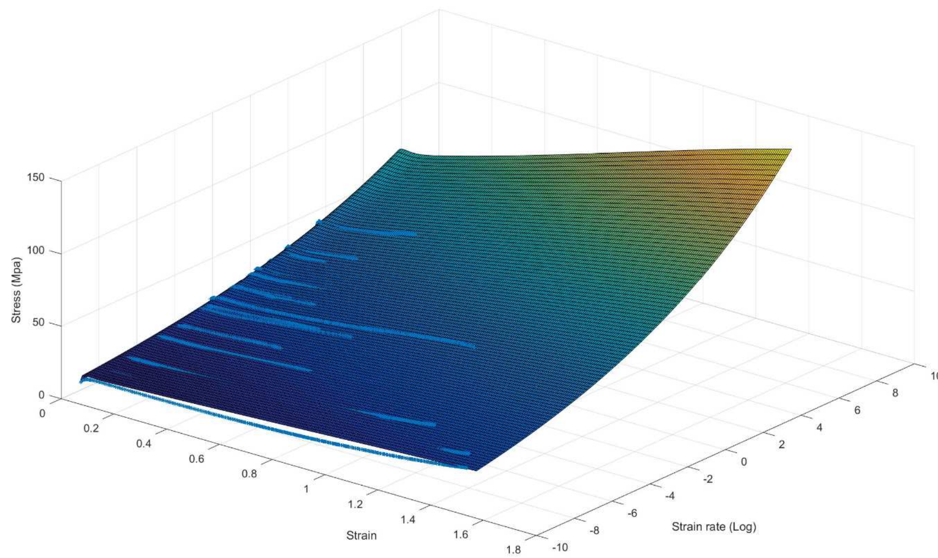

Fig. 7. 3D behavior surface of the P/E-MD15 impact modified

The constitutive laws which best describe behavior of P/E-MD15 impact modified is the model developed by G'Sell-Jonas [14], integrated in developments of Balieu et al [15], with main constitutive equations described in section 5. The identification of the parameters is carried out by optimization process thanks to Matlab tool.

Once all the parameters of the mathematical model have been identified, additional tensile tests were performed under some selected temperature/strain rate couples in order to assess the validity of the principle of time temperature superposition in viscoplasticity domain. In the graphs below, behavior curves obtained at $T \neq T_{ref}$ are shifted using the same shift factor as identified by DMA and superposition of curves is examined.

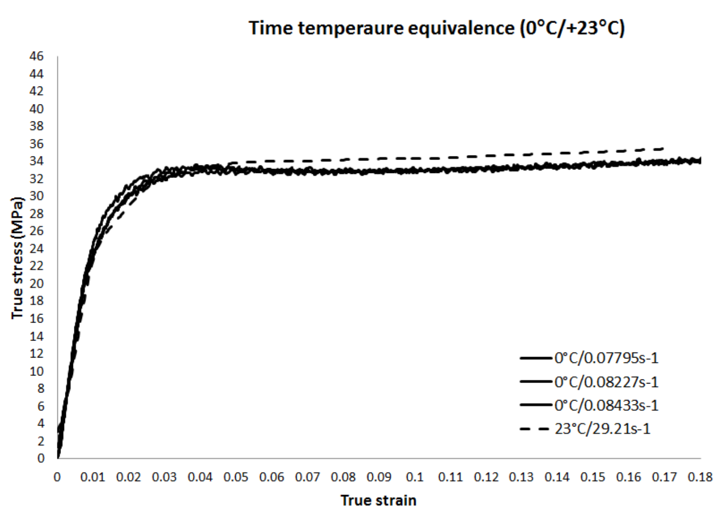

Fig. 8. Couples tested at 0℃ and +23℃

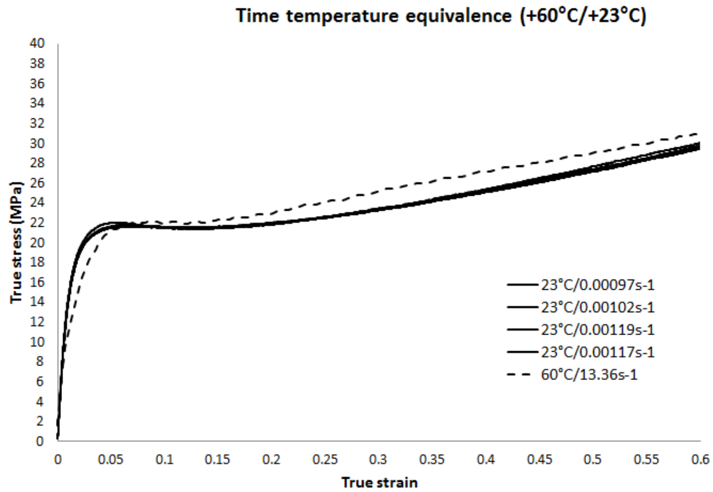

Fig. 9. Couples tested at +23℃ and +60℃

The results show that it is possible to link both sensitivity to strain rate and temperature also into the viscoplasticity domain. Indeed, reference curves (T=23℃) and shifted curves ($T \neq T_{ref}$) are well overlapped. For example, a tensile test carried out at high strain rate at room temperature gives results equivalent to a tensile test at low strain rate at low temperature. We can do the same observation between room and high temperature. Therefore, the time temperature equivalence principle is demonstrated and validated for viscoplasticity domain concerning our P/E-MD15 impact modified.

Thanks to this study a new test procedure for material characterization can be defined. The identification of the shift factor by DMA combined to tensile tests at room temperature allows determining the whole viscoelastic and viscoplastic behavior law for such polymer and reducing significantly the experimental campaign of behavior characterization.

5 Numerical approach: Constitutive model

5.1 Presentation of the constitutive model

As explained previously, for modeling the complex behavior of the polymers, a lot of parameters have to be taken into account. For that, Balieu [1][2][16] developed a viscoelastic viscoplastic model enhanced by a non-associative approach (non-isochoric deformation) and a non local damage formulation to simulate the behavior of a mineral filled semi-crystalline polymer.

To represent the viscoelastic effect, a linear Wiechert model is implemented into the model. It is composed of n Maxwell elements in parallel with a Hooke element which allows describing the long term effect. The damage is coupled with the viscoelastic law. In the constitutive model, the corresponding time-dependent stress is given by:

$$\underline{\sigma}(t) = (1 - D) \int_0^t R^{ve}(t - \zeta) \frac{d\underline{\varepsilon}^{ve}}{d\zeta} d\zeta \qquad (2)$$

Where D is the isotropic damage variable, R^{ve} a relaxation tensor and $\underline{\varepsilon}^{ve}$ the viscoelastic part of the strain tensor.

To capture the tension-compression dissymmetry, a non symmetric yield surface developed by Raghava et al [17] is introduced into the model:

$$f(\underline{\sigma}, R, D) = \frac{(\eta - 1) I_1(\underline{\sigma}) + \sqrt{(\eta - 1)^2 I_1^2(\underline{\sigma}) + 12\eta J_2(\underline{S})}}{2\eta(1 - D)} - \sigma_t - R(\kappa) \qquad (3)$$

Where $J_2(\underline{S})$ and $I_1(\underline{\sigma})$ are respectively the second invariant of the deviatoric stress tensor and the first invariant of the stress tensor. The dependence to the hydrostatic pressure is defined by the parameter η which is the ratio between the compression and tension initial yield stresses σ_c and σ_t, such that:

$$\eta = \frac{\sigma_c}{\sigma_t} \qquad (4)$$

$\boldsymbol{R(\kappa)}$ is the nonlinear isotropic hardening function defined by:

$$R(\kappa) = Q_1 \kappa\, exp(-b_1\kappa) + \ Q_2\big(1 - exp(-b_2\kappa)\big) + b_3\kappa^3 + b_4\kappa^2 + b_5\kappa, \qquad (5)$$

with Qi and bi material parameters,$\forall_i$. κ is the equivalent viscoplastic strain.

During the deformation of a polymer material, we can observe a variation of the volume. To represent this phenomenon, a viscoplastic potential F is used as follows:

$$F(\underline{\sigma}, \mathrm{D}) = \frac{\sqrt{3J_2(\underline{S}) + \alpha^+ < p >^2 + \alpha^- < -p >^2}}{1 - \mathrm{D}} = \frac{\mathrm{g}(\underline{\sigma})}{1 - \mathrm{D}} \qquad (6)$$

The parameters α^+ and α^- define the volume variation for positive and negative hydrostatic pressure. Thanks to the formulation using the Macauley bracket “< . >”, the viscoplastic flow can evolve independently for dilatation and compression.

To take viscoplasticity into account, a Perzyna-type [18] viscoplastic model is chosen, which considers a viscous overstress.

$$\sigma^v = \big(\sigma_t + R(\kappa)\big)\left(\frac{\dot{\kappa}}{\dot{\kappa_0}}\right)^n \qquad (7)$$

Where $\dot{\kappa}$ is the equivalent viscoplastic strain rate and n and $\dot{\kappa_0}$ are material parameters.

Considering the viscous stress, the dynamic yield surface, f^{d}, can be expressed from the static one, f, so that:

$$\mathrm{f}^d = \frac{(\eta - 1)I_1(\underline{\sigma}) + \sqrt{(\eta - 1)^2 I_1^2(\underline{\sigma}) + 12\eta J_2(\underline{S})}}{2\eta(1 - \mathrm{D})} - (\sigma_t + R(\kappa))\left\{1 + \left(\frac{\dot{\kappa}}{\dot{\kappa_0}}\right)^n\right\} = 0 \qquad (8)$$

For modeling the damage, a phenomenological model based on stress differences between the isochoric and non-isochoric deformation process is proposed. The evolution of the damage variable is represented by the scalar variable D and defined by the ratio between the true stresses calculated with the incompressibility and compressibility assumptions. In the constitutive model, the damage variable is expressed as:

$$D = 1 - \ exp\left(-\frac{\kappa}{\kappa_c}\right) \qquad (9)$$

Where κ_c is a material parameter.

In the context of the present study, the principle of time temperature equivalence has been added into the constitutive model, with shift factors reminded as follows:

$$a_T\,(T) = exp(-C\,(T - T_{ref})) \qquad (10)$$

More precisely, the principle of time temperature superposition is introduced in numerical simulations by re-computing a specific time step, $\Delta t'$, thanks to the coefficient $a_T\,(T)$ in order to take the link between strain rate and temperature sensitivity into account.

$$\Delta t' = \frac{\Delta t}{a_T\,(T)} \qquad (11)$$

5.2 Results

In this subsection, we propose at first to validate definitively all the parameters of the constitutive law identified for the studied material. Thus a correlation study is achieved on the different geometry of samples tested in tension at various strain rates and temperatures.

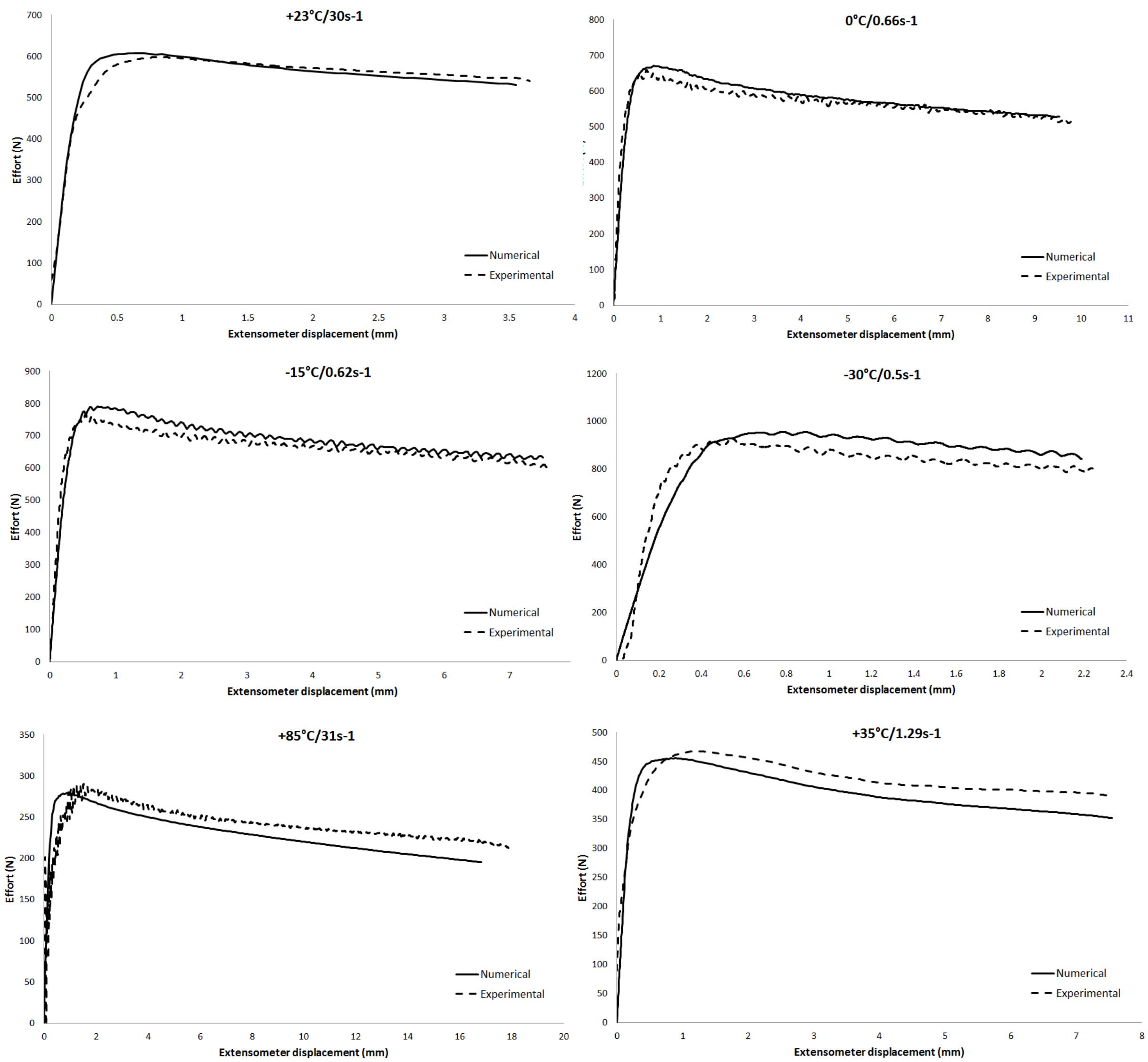

Fig. 10. Correlation study at different strain rates and temperatures

By comparing the reaction forces of the numerical model with the experimental tests, we can observe a good level of correlation. The non associated viscoplastic formulation of the constitutive model is able to predict very well the complete behavior of the material between -30℃ and +85℃ for various strain rates.

As shown in Fig.8. and Fig.9., the principle of time temperature superposition is validated experimentally. It is now essential to check it by numerical simulation. For that, we propose to test the principle for two temperature/strain-rate couples, as shown in Fig. 11 and 12.

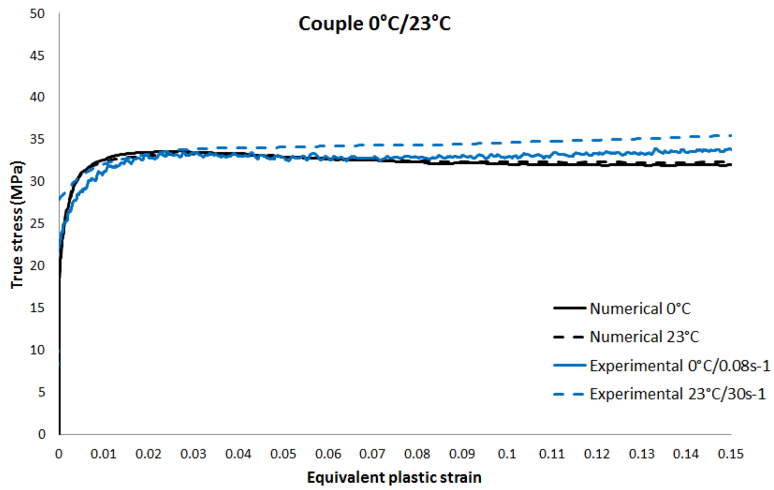

Fig. 11. Correlation study: Couple tested at 0℃ and +23℃

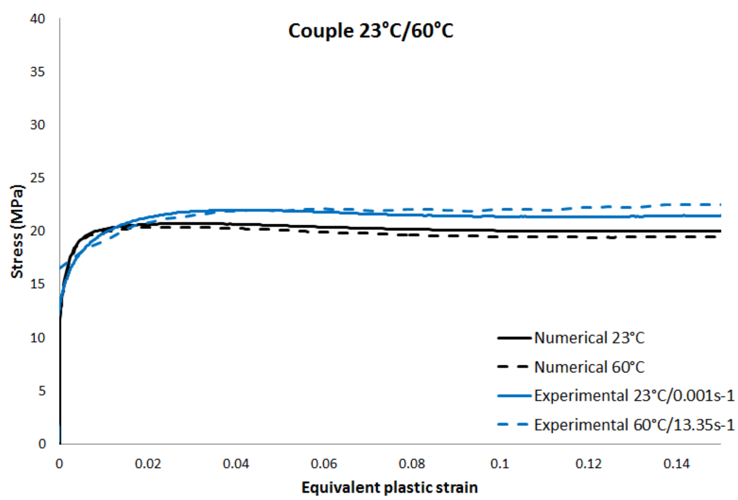

Fig. 12. Correlation study: Couple tested at 23℃ and +60℃

The results show a good level of correlation between the physical tests and the numerical simulations. Concerning the two tested strain rate/temperature couples, the results are the same. The numerical model describes completely the behavior of the studied material. The principle of time temperature superposition is well taken into account thanks to its introduction in the constitutive laws. In simulation, we reproduce very well the physical phenomena observed in physical test since curves are perfectly similar.

6 Summary

In this paper, a complete characterization of a P/E-MD15 impact modified at various strain rates and temperature has been proposed. First the principle of time temperature superposition has been validated in the viscoelastic domain thanks to DMA tests. From these results, a mathematical equation which allows linking the shift factors to the temperature has been defined. Furthermore, the coefficient C also identified by DMA tests has been used for viscoplasticity characterization. It has been shown that the principle of time temperature superposition is also applicable in the viscoplasticity domain. Then the model has been introduced in the constitutive laws developed by Balieu et al. and tested for two strain rate/temperature couples using numerical simulation. The numerical model is able to reproduce very well the principle of time temperature equivalence and the level of correlation between physical tests and simulations is remarkable. Thanks to this study, a new method of polymer characterization in wide temperature and strain-rate range that allows a significant reduction of required tests can be proposed. In addition, use of superposition principle can now be a tool to predict behavior under strain rate and temperature values which cannot be achieved by the test devices. In a future work, it will be investigated whether the time temperature superposition can be applied for failure prediction.

7 Acknowledgements

The present work has been supported by the LAMIH UMR CNRS 8201, the research department of the University Polytechnic Hauts-de-France and the Faurecia Interior System Company. I would like to express my sincere gratitude to all of them as well as Toyota Motor for the work carried out on the constitutive law.

8 Literature

[1] Balieu, R., Lauro, F., Bennani, B., Delille, R., Matsumoto, T., Mottola, E. (2013). A fully coupled elastoviscoplastic damage model at finite strains for mineral filled semi-crystalline polymer, International Journal of Plasticity, 51, 241-270

[2] Balieu, R., Lauro, F., Bennani, B., Matsumoto, T., Mottola, E. (2014). Non-associated viscoplasticity coupled with an integral-type nonlocal damage model for mineral filled semi-crystalline polymers, Computers and Structures, 134, 18-31

[3] Balieu, R., Lauro, F., Bennani, B., Haugou, G., Chaari, F., Matsumoto, T., Mottola E. (2015). Damage at high strain rates in semi-crystalline polymers. International Journal of Impact Engineering, 76, 1-8.

[4] Lauro, F., Bennani, B., Morin, D., and Epee, A. (2010). The SEE method for determination of behaviour laws for strain rate dependant material: Application to polymer material. International Journal of Impact Engineering, 37,715-722, impact Loading of Lightweight Structures.

[5] Epee, A., Lauro, F., Bennani, B., and Bourel, B. (2011). Constitutive model for a semi-crystalline polymer under dynamic loading. International Journal of Solids and Structures, 48, 1590-1599.

[6] Roncin Armelle, Ph.D. thesis. Étude de la modification des propriétés rhéologiques linéaires et non linéaires par ingénierie moléculaire. Vers le contrôle des propriétés adhésives de matériaux autocollants.www.theses.fr. Submitted on 12 Dec 2011.

[7] Maurel-Pantel Aurelien, Baquet Erwan, Bikard Jerome, Bouvard Jean-Luc, and Billon Noëlle. A thermo-mechanical large deformation constitutive model for polymers based on material network description: Application to a semi-crystalline polyamide 66. International Journal of Plasticity, 67:102-126, 2015.

[8] Billon Noëlle. New constitutive modeling for time-dependent mechanical behavior of polymers close to glass transition: Fundamentals and experimental validation. Journal of Applied Polymer Science, 125(6):4390-4401, 2012.

[9] 6. F.-J. Wortmann, K.V. Schulz. Stress relaxation and time/temperature superposition of polypropylene fibres. Polymer Vol. 36 Ko. 2, PP. 315-321, 1995.

[10] Rongzhi Li. Time-temperature superposition method for glass transition temperature of plastic materials. Materials Science and Engineering A278 (2000) 36–45.

[11] Malcolm L. Williams, Robert F.Landel and John D.Ferry. (1955). The Temperature Dependence of Relaxation Mechanisms in Amorphous Polymers and Other Glass-forming Liquids. Temperature dependence of relaxation mechanisms 3701 [Contribution from the department of chemistry, University of Wisconsin].

[12] Darko Ljubic, Marina Stamenovic, Chad Smithson, Maja Nujkic, Bojan Medo, Slavisa Putic. Time - temperature superposition principle - Application of WLF equation in polymer analysis and composites. Scientific paper. UDC:629.179.13:678.06

[13] Jean-Jacques Pesce. Influence de l’orientation moléculaire d’un polymère fondu en écoulement sur l’anisotropie induite des propriétés à l’état solide.

[14] G’Sell, C. and Jonas,J.(1979). Determination of the plastic behaviour of solid polymers at constant true strain rate. Journal of Materials Science,14,583-591.

[15] Romain Balieu. Modèle viscoélastique-viscoplastique couplé avec endommagement pour les matériaux polymères semi-cristallins. Other. Université de Valenciennes et du Hainaut-Cambresis, 2012. English. <NNT : 2012VALE0031>. <tel-00819902>

[16] Romain Balieu, Franck Lauro, Bruno Bennani, Benjamin Bourel. Modèles de comportement pour matériaux polymères soumis au crash. 10e colloque national en calcul des structures, May2011, Giens, France. pp.Cl_e USB, 2011. <hal-00592851>

[17] Raghava R, Caddell RM, Yeh GSY. The macroscopic yield behaviour of polymers. J Mater Sci 1973;8(2):225–32.

[18] Perzyna P. Fundamental problems in viscoplasticity. Adv Appl Mech 1966;9:243–377.