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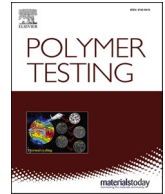
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Time-temperature superposition in viscoelasticity and viscoplasticity for thermoplastics

Vincent Dorléans^{a,b}, Rémi Delille^b, Delphine Notta-Cuvier^{b,c,*}, Franck Lauro^b, Eric Michau^a

^a FAURECIA Interior System, Méru, France

^b Univ. Polytechnique Hauts-de-France, LAMIH, CNRS, UMR 8201, F-59313, Valenciennes, France

^c INSA Hauts-de-France, F-59313, Valenciennes, France

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ABSTRACT

The use of thermoplastics in the automotive industry is continuously increasing. Design of parts require the characterization and numerical modelling of thermo-mechanical behaviour of those materials under wide strain rate ($0\text{--}300\text{ s}^{-1}$) and temperature ranges (-30 °C to $+85\text{ °C}$). Large and costly experimental campaigns are therefore necessary to identify all behaviour's sensitivities. Yet, the time-temperature-superposition (TTS) principle may be a powerful tool to strongly reduce the number of required tests.

This paper demonstrates the possibility to extend this principle to the viscoplastic domain while keeping the same shift factors identified for viscoelasticity. So, a limited number of tests (DMA and tensile tests at room temperature only) now enable the characterization of whole thermoplastic behaviour. This new approach is implemented in a viscoelastic, viscoplastic behaviour model. A good correlation between numerical simulations and experiments validate the approach.

1. Introduction

Nowadays, in vehicles interiors, dashboards, door panels or center consoles, e.g., are made of thermoplastic polymers. These parts may be submitted to severe loading cases such as head impact or airbag deployment. Then, with the aim to prevent or at least reduce vehicle occupants injuries, those parts must comply with high specifications, in terms of thermo-mechanical behaviour under severe loadings, as imposed by international safety regulations for the homologation of the vehicle. In that context, it is crucial to well understand polymers thermo-mechanical behaviour up to failure and to dispose of an accurate behaviour model that takes all behaviour specificities into account for numerical simulations.

Focusing for instance on airbag deployment during a car crash, the inflation of the airbag generates a significant level of deformation on the module, the chute channel and the top cover of the dashboard. Numerical simulation is thus a key tool to anticipate the risk of failure on those parts and to optimize the design before parts' production. This kind of simulation require a complete characterization of polymer behaviour under wide strain rate (from 10^{-5} to 300 s^{-1}) and temperature ranges (from -30 °C to $+85\text{ °C}$). Generally, this large amount of data is introduced into the finite element solver by considering several sets of

behaviour curves (several curves at different strain rate, at fixed temperature, e.g.).

This study focusses on the behaviour of a copolymer propylene-ethylene, which is 15 wt% mineral (talc) filled and impact modified by elastomeric nodules (P/E-MD15 impact modified). This kind of material is commonly used for injection-molded parts such as the top cover of a dashboard. As mentioned previously, it is necessary to characterize all specificities of its thermo-mechanical behaviour.

As shown in the literature, thermoplastic behaviour is sensitive to strain rate [1,2], hydrostatic pressure and temperature [3,4]. It appears also that the plastic (or viscoplastic) flow is not isochoric. Therefore, behaviour characterization requires a large test campaign, including dynamic mechanical analysis (DMA) for characterization of viscoelasticity and tensile tests at various strain rate and temperature for characterization of viscoplasticity and temperature sensitivity. Aiming at reducing the number of required experiments, the time-temperature-superposition (TTS) principle is investigated. This principle is based on a correlation between behaviour sensitivity to strain rate on one hand and to temperature on the other hand. As exposed in Appendix, "shift factors" can be identified in order to define a couple of shifted strain rate and given temperature that leads to a similar behaviour than under initial (i.e non-shifted) strain rate and reference temperature.

* Corresponding author. Univ. Polytechnique Hauts-de-France, LAMIH, CNRS, UMR 8201, F-59313, Valenciennes, France.

E-mail address: delphine.notta@uphf.fr (D. Notta-Cuvier).

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In the viscoelastic domain, the coupling between strain rate and temperature sensitivities is generally assessed for the evolution of storage modulus. This one can be identified by DMA at different temperatures and can be described by models of William, Landel and Ferry [5,6] or Arrhenius [7]. If the validity of TTS principle has been demonstrated for different thermoplastics for viscoelasticity [8–11], its application in viscoplasticity has never been investigated before for a large strain rate range, to the authors' best knowledge. In this paper, it is therefore proposed to assess the validity of TTS scheme in viscoplasticity and to investigate whether the shift factors identified in viscoelasticity could be suitable for TTS principle in viscoplasticity.

In recent past, some models have been developed to predict thermoplastics mechanical and/or thermal behaviour, aiming at being input in numerical simulations, but listing them is outside the scope of this study. The present study will focus on the model developed by Balieu et al. [12–14], dedicated to the prediction of mechanical behaviour of semi-crystalline thermoplastic polymers, at constant temperature. Please note that main constitutive equations of model of Balieu et al. are given in Appendix. The viscoelasticity is taken into account by considering a rheological model, namely Wiechert model [15]. To consider the hydrostatic pressure dependency, a Raghava yield surface [16] is considered while viscoplasticity is described by Perzyna's constitutive law [17]. Finally, the non-isochoric plastic flow is taken into account through the expression of the thermodynamic potential.

The present paper is dedicated to the investigation of time-temperature-superposition principle in the framework of viscoelasticity (section 3.2) and, as a novelty, of viscoplasticity (section 4.2), focusing on P/E-MD15 impact modified polymer. In that framework, whole characterization of the polymer behaviour is first performed for various strain rates and temperatures (sections 3.1-4.1). Then, the time-temperature-superposition principle is investigated in viscoelasticity. An evolution law for the shift factors with respect to temperature is identified (section 3.2). It is then proposed to extend the TTS principle to viscoplasticity by using the same shift factors determined in viscoelasticity (section 4.2). The TTS principle is coupled to viscoelastic-viscoplastic constitutive laws originally defined for behaviour prediction at ambient temperature and implemented in a user defined material model for numerical simulations (section 5.1). The approach is finally validated by comparison of experimental and numerical material response, under different strain rate/temperature couples (section 5). Based on those results, a new characterization procedure is proposed to significantly reduce the number of required tests for whole material thermo-mechanical behaviour characterization but also to predict behaviour under strain rate and temperature ranges that are not reachable with classic test devices (section 6).

2. Viscoelastic behaviour

2.1. Characterization using dynamic mechanical analysis

The viscoelastic properties of the material, namely the complex modulus, E^* , and the loss angle, $\tan\delta$, are characterized using Dynamic Mechanical Analysis (DMA). An Instron Electropulse E3000 with pneumatic grips (Fig. 1) is used. A sinusoidal displacement with amplitude varying from 0 to 0.1 mm is applied in tension to a rectangular specimen ($80 \times 10 \text{ mm}^2$) in a frequency range between 0.01 and 30 Hz and at different temperatures from -30°C to $+85^\circ\text{C}$. At cold and high temperature, a thermoregulated oven is used (Fig. 1). The temperature at specimen surface was verified just before testing, after a period of acclimatization. Note that thermal fields on the specimen surface, and therefore possible self-heating during tests, were not measured.

Directly arising from imposed displacement, the nominal strain is given by:

$$\varepsilon(t) = \varepsilon_0 \cos(\omega t) = \varepsilon_0 R_e \{ \exp(i\omega t) \} \quad (3.1)$$



Fig. 1. Test device chamber used for DMA.

where ε_0 is the strain amplitude and ω is the angular frequency. R_e represents the real part of a complex quantity. For low strain amplitude, the measured stress is also of sinusoidal shape but with a phase shift, δ , also called loss angle, compared to the strain, so that:

$$\sigma(t) = \sigma_0 \cos(\omega t + \delta) = \sigma_0 R_e \{ \exp(i(\omega t + \delta)) \} \quad (3.2)$$

The ratio of the stress over strain, gives the complex modulus E^* :

$$E^*(i\omega) = \frac{\sigma_0}{\varepsilon_0} \exp(i\delta) \quad (3.3)$$

The complex modulus can be split into a real part, the storage modulus, E' , and an imaginary part, the loss modulus, E'' (Eq (3.4)). The storage modulus is related to the elastic strain energy of the material while the loss modulus is related to dissipated energy through viscous effects:

$$E^* = E' + iE'' \quad (3.4)$$

with

$$E' = \frac{\sigma_0}{\varepsilon_0} \cos(\delta) \quad (3.5)$$

$$E'' = \frac{\sigma_0}{\varepsilon_0} \sin(\delta) \quad (3.6)$$

The loss angle is given by:

$$\tan\delta = \frac{E''}{E'} \quad (3.7)$$

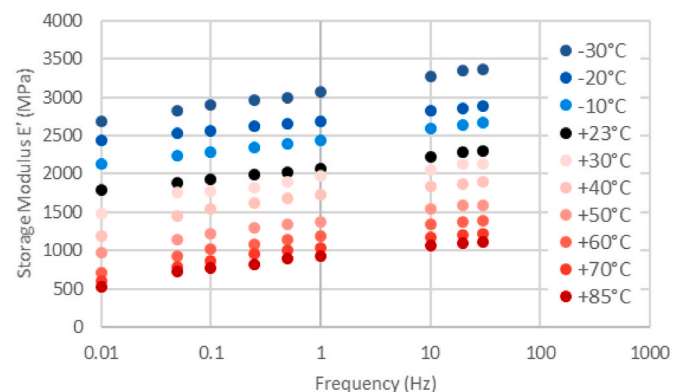


Fig. 2. Storage modulus versus frequency at different temperatures.

Fig. 2 below shows the evolution of the storage modulus, E' , versus the frequency for different temperatures, with a step of 10 °C. 3 specimens were tested by frequency and by temperature, showing a good repeatability of results (standard deviation of about 4%).

As expected, DMA shows an increase of the storage modulus, E' , when increasing frequency (increase of about 300 MPa in the covered frequency range, at given temperature) and when decreasing temperature (decrease of about 2200 MPa in the covered temperature range, at given frequency). Yet, it is interesting to note that the evolution of E' with the frequency is very similar for all tests. It indicates that the influences of the strain rate and temperature on storage modulus are uncoupled. On that basis, it is possible to investigate the validity of time temperature superposition principle in the viscoelastic domain.

The studied material is a semi-crystalline copolymer propylene-ethylene, which is 15 wt% mineral (talc) filled and impact modified by elastomeric nodules (P/E-MD15 impact modified). This compound has a block structure with combination of a polypropylene homopolymer and a propylene/ethylene copolymer rich in ethylene (50–60% by weight). Usually, semi-crystalline polymers show 2 types of temperature transitions: T_g and T_f . The transition of the amorphous phase from vitrous to rubbery state occurs at, T_g , while T_f is the melting temperature of crystalline phase. Due to the presence of different polymeric phases, P/E-MD15 impact modified shows multiple glass-transition temperatures at -87.7 °C (ethylene phase), at -52.4 °C (elastomeric phase) and at $+7.8$ °C (propylene phase) as highlighted by DSC analysis (Fig. 3).

It is noticed that the glass transitions actually have a little impact on the evolution of the moduli due to the low proportion of amorphous phase in our material. This observation allows to attempt to find a time-temperature-superposition model on the whole studied temperature range.

2.2. Time temperature superposition principle (TTS)

Time temperature superposition principle is a scheme based on some equivalence between material behaviour sensitivity to strain rate and temperature. In other words, TTS principle allows to find a couple of strain rate/given temperature that leads to a similar behaviour than another strain rate/reference temperature couple. It is based on the identification of frequency shift factors, which depend on temperature, to build a master curve, at a reference temperature. Please note that more details are given in Appendix.

Focusing on our material, the DMA (section 3.1) have highlighted that similar values of storage modulus are measured for different couples

temperature/frequency. As an example, the storage moduli measured at -30 °C and 0.01 Hz and at -10 °C and 30 Hz are very close to each other (Fig. 4a). Aiming at verifying the applicability of the TTS principle, a master curve is built, following the procedure described in Appendix. The reference temperature chosen is $+23$ °C. Shift factors $a_T(T)$ are identified for each temperature curve (Fig. 4b) in order to align all these curves on the master curve, by horizontal translation (along the frequency axis) (Fig. 4c).

For our material, it is therefore possible to build a master curve (Fig. 4d) for the evolution of the storage modulus versus shifted frequency f_{TTS} defined by:

$$f_{TTS} = f \times a_T(T) \quad (3.8)$$

with $a_T(T)$ the shift factor which is equal to 1 for the reference temperature.

As a first step, it was assessed whether classic models of Williams-Landel-Ferry or Arrhenius (cf Appendix) could be used to predict the evolution of the shift factors $a_T(T)$ with temperature. It appeared that for our material, none of these models were suitable to predict all the shift factors of the master curve (Fig. 5).

Therefore, a new model which is an adaptation of Williams-Landel-Ferry and Arrhenius models is proposed here, as follows (Fig. 4d):

$$a_T(T) = \exp(C(T - T_{ref})) \quad (3.9)$$

where T and T_{ref} are the current temperature and the reference temperature, respectively, and C is a material parameter to be identified.

This model reproduces very well all the shift factors ($R^2 = 0.9998$) of the master curve, with a value of C of 0.25. That allows to validate definitively the time temperature superposition principle in the viscoelastic domain for our material and to extrapolate the data of the master curve to other temperatures.

From now on, it is possible to know the value of the storage modulus on extended frequency ranges by extrapolating experimental data. Up to now, the measurements were made in a frequency range between 0.01 and 30 Hz due to the limitation of the test device. Thanks to the time temperature superposition principle, the storage modulus at $+23$ °C can be estimated on a wider frequency range between 1.10^{-9} and $+1.10^7$ Hz. Therefore, provided its validity is checked, this principle allows to determinate the viscoelastic properties of a material in a wider frequency and temperature ranges with a reduced number of tests and to overcome limitations of conventional test devices.

The next challenge of this study is to check if the time temperature

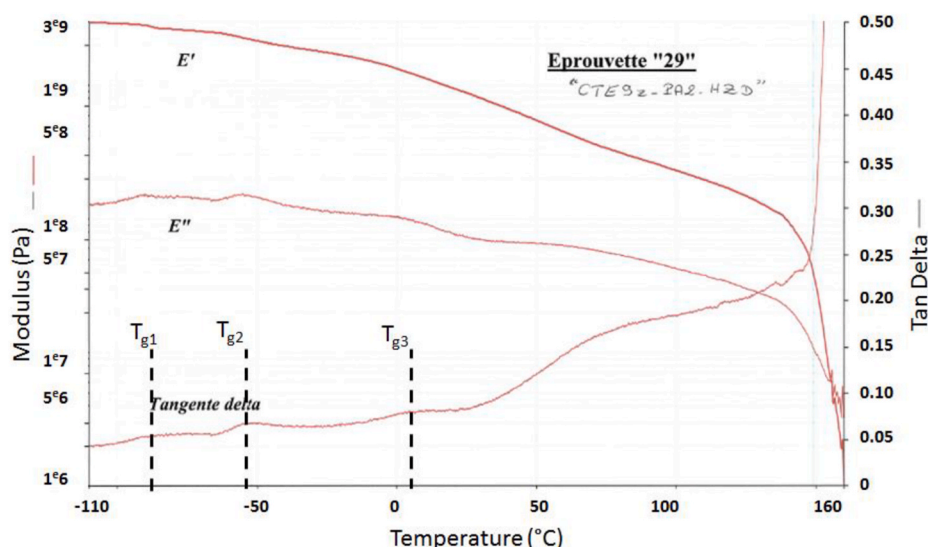


Fig. 3. Storage and loss moduli and loss factor versus temperature (Heat from -110 °C to $+180$ °C at 3 °/min).

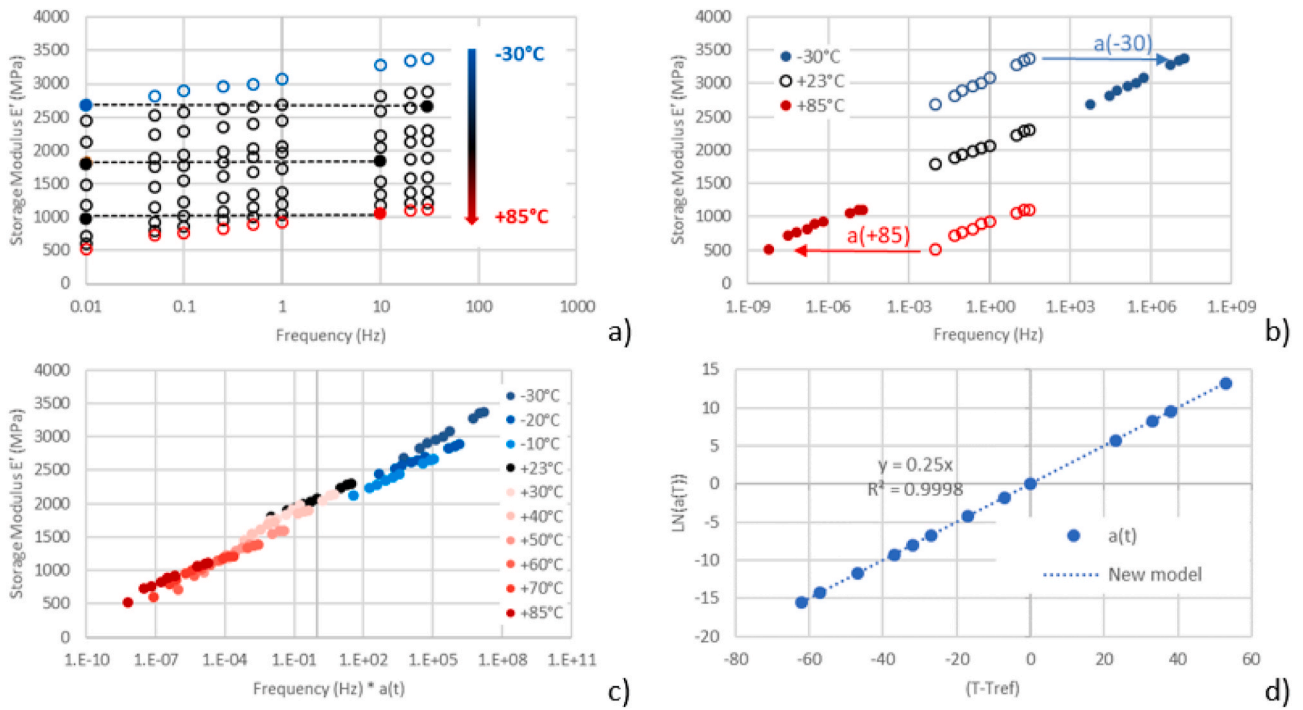


Fig. 4. Building of the master curve and characterization of time temperature superposition principle for present material.

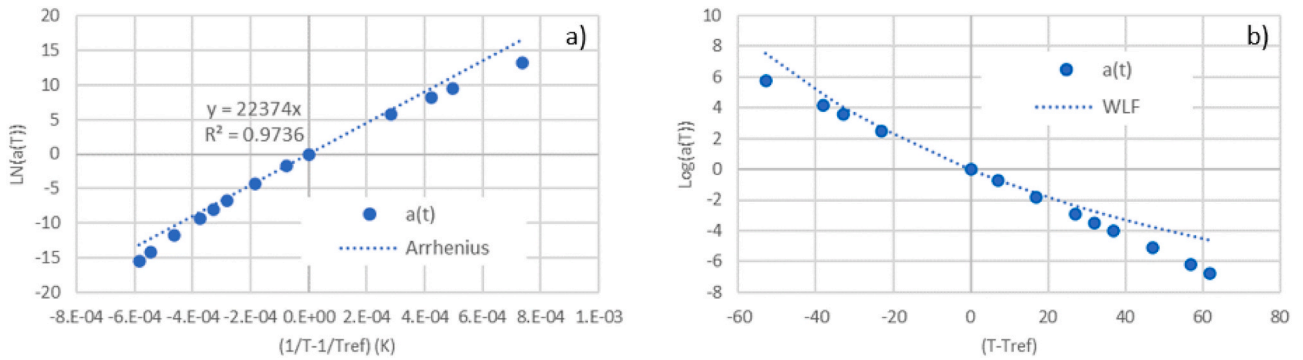


Fig. 5. Evolution of $a_r(T)$ a) Arrhenius, b) WLF.

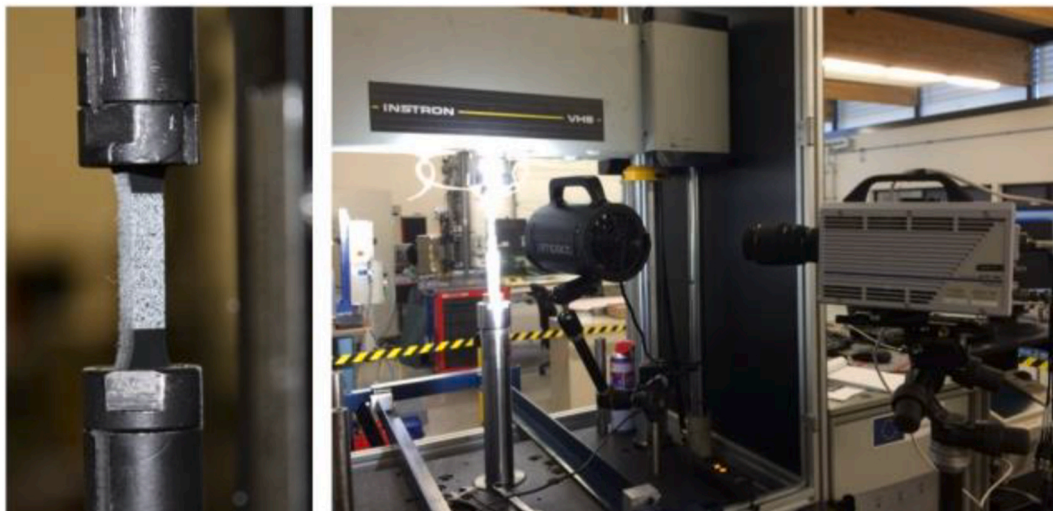


Fig. 6. Hydraulic high-speed device.

superposition can be applied in viscoplasticity by using the same shift factors identified in viscoelasticity, $a_T(T)$. Before that, a complete characterization of the viscoplastic properties at different strain rate and temperature is necessary. The following section is dedicated to this topic.

3. Viscoplastic behaviour

3.1. Characterization using tensile tests

To characterize the viscoplastic behaviour of the polymer, uniaxial tensile tests are performed at different strain rate (from 5.10^{-3} s^{-1} to 30 s^{-1}) in a temperature range between $-30 \text{ }^\circ\text{C}$ and $+85 \text{ }^\circ\text{C}$. The quasi-static tensile tests are carried out on an electromagnetic device (Instron E3000) with a 3 kN cell force. A hydraulic high-speed device Instron VHS with a 5 kN cell force is used for the dynamic loadings (Fig. 6).

For specimens, two geometries are proposed:

- One quasi-static specimen which dimensions are adapted from standards NF EN ISO527 2B. Indeed, the gauge length is reduced to 10 mm in order to reach high deformation levels;
- One dynamic specimen with a gauge length of 20 mm with a geometry specially design for the high-speed tensile device.

Specimens were cut by water jet from injection-molded rectangular plates of 2.5 mm of thickness (Fig. 7).

2D digital image correlation technique combined with a high-speed camera is used to measure the displacement fields and so on extract the strain fields on specimen's surface of interest, thanks to the painting of a random pattern. Different techniques for pattern creation are employed. At room and hot temperature, a black and white paint speckle is applied on specimen surface for all tests. However, at cold temperature, black and white painting was replaced by grease covered by alumina balls (Fig. 8). Indeed, insufficient adhesion between the painting and the specimen resulted in a peeling of the pattern at $-30 \text{ }^\circ\text{C}$.

More information about DIC analysis is given in Table 1.

Again, note that thermal fields on the specimen surface, and therefore possible self-heating during tests, were not measured.

Fig. 9 below shows a summary of all tensile tests results performed at different strain rate (from 5.10^{-3} s^{-1} to 30 s^{-1}) in a temperature range between $-30 \text{ }^\circ\text{C}$ and $+85 \text{ }^\circ\text{C}$.

As expected, the polymer tensile behaviour is highly sensitive to strain rate and temperature. Behaviour is highly ductile at $+85 \text{ }^\circ\text{C}$ with a

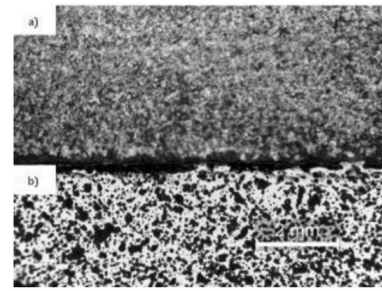


Fig. 8. Two techniques for random pattern deposit: a) Grease with alumina balls (low temperature) b) black and white color painting (ambient and high temperature).

Table 1
Parameters of DIC analysis.

Camera	Quasi-static tests: Manta G: 12.4 Million pixels, full resolution: $4112 \times 3008 \text{ pix}^2$ Dynamic tests: Photron APX RS, full resolution: $1024 \times 1024 \text{ pix}^2$
Pixel size	From 30 to 90 μm
Frame rate	From 5 to 10,000 images per second
Facet size	Quasi-static tests: 59 pix^2 Dynamic tests: 13 pix^2
Step size	Quasi-static tests: 30 pix Dynamic tests: 6 pix

plastic strain level higher than 120%. As commonly observed, material ductility dramatically decreases at low temperature ($-30 \text{ }^\circ\text{C}$), with a plastic strain level lower than 10%. As expected, tensile strength and rigidity increase when increasing strain rate and decreasing temperature.

3.2. TTS principle in viscoplasticity

Experimental results show that similar tensile behaviour can be observed for different strain rate/temperature couples (Fig. 9). For example, behaviour at $+23 \text{ }^\circ\text{C}$ and 0.7 s^{-1} , is similar to that at $0 \text{ }^\circ\text{C}$ and 6.10^{-3} s^{-1} . The same phenomenon is observed for other couples such as at $+23 \text{ }^\circ\text{C}$ at 4.10^{-2} s^{-1} and $+40 \text{ }^\circ\text{C}$ at 5.9 s^{-1} . Based on these observations, it seems that the TTS principle could be applied also in the viscoplastic domain.

To apply the TTS principle in viscoplasticity the previous behaviour

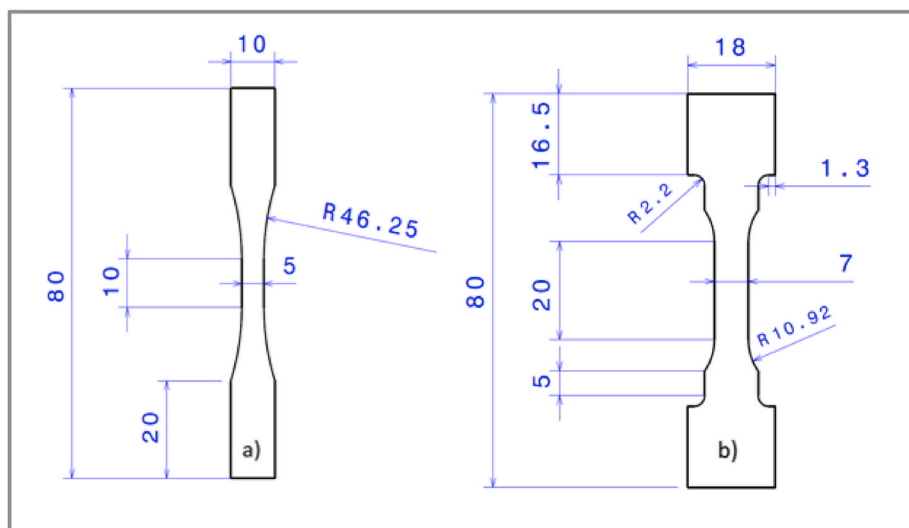


Fig. 7. a) Quasi-static specimen, b) Dynamic specimen.

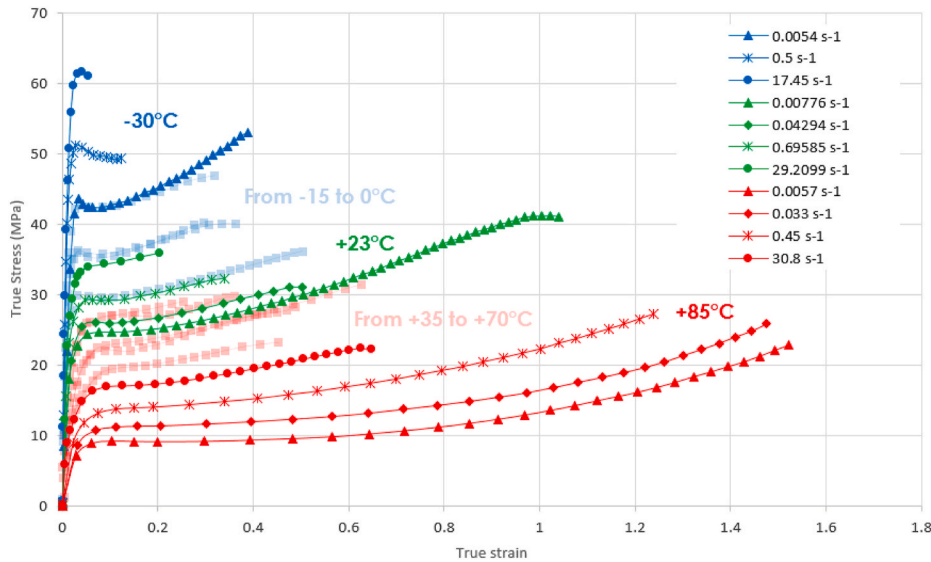


Fig. 9. Tensile tests at different strain rate (from 5.10 to 3 s^{-1} to 30 s^{-1}) and temperature (from $-30 \text{ }^\circ\text{C}$ to $+85 \text{ }^\circ\text{C}$).

curves obtained at a given temperature are plotted in a 3D space (true strain; true stress; strain rate) by using the *SEE* (for stress-strain-strain rate) method [18,19]. This method allows to take benefits of the measurement of heterogeneous mechanical fields by DIC since data extracted from all the facets are plotted in the 3D space. A continuous surface is then extrapolated between experimental data (Fig. 10). Finally, any viscoplastic behaviour, at a given strain rate included in the covered range, is extracted by “cutting” the surface in the corresponding stress-strain plane.

As in viscoelasticity, a master surface is built by applying the time temperature superposition principle. Input data are now the evolution of true stress vs true strain, at various strain rate and temperature. The same reference temperature as in viscoelasticity is obviously chosen ($+23 \text{ }^\circ\text{C}$) and the same shift factors are used. Then, considering a test at a given strain rate and temperature, T , the strain rate is multiplied by the shift factor $a_T(T)$, for each viscoplastic stress vs strain law, which allows to define an equivalent viscoplastic behaviour at T_{ref} (Fig. 11).

As a result, behaviour surface can be plotted for the reference temperature, for a wide range of shifted strain rate (Fig. 12). For instance,

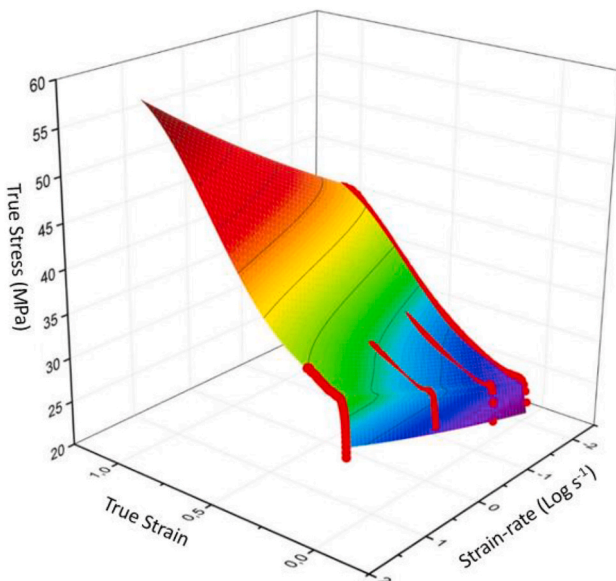


Fig. 10. Behaviour surface obtained by the SEE Method at $+23 \text{ }^\circ\text{C}$.

data measured at $+85 \text{ }^\circ\text{C}$ for strain rate varying from 0.006 s^{-1} to 31 s^{-1} can be used to extend the *SEE* surface at T_{ref} for strain rate varying from $1.6 \cdot 10^{-9}$ to $5.7 \cdot 10^{-6} \text{ s}^{-1}$.

The validity of TTS principle in viscoplasticity, with the same shift factors than in viscoelasticity, is checked by comparing experiments and curves extracted from the *SEE* surface. To this aim, two different couples strain rate/temperature among performed experiments are considered. Then, the corresponding strain rate which should lead to a similar behaviour at $+23 \text{ }^\circ\text{C}$ by applying the shift factors $a_T(T)$ is computed. Fig. 13 shows the results of the tensile tests for two couples.

The reference curves ($T = +23 \text{ }^\circ\text{C}$) and shifted curves ($T \neq T_{\text{ref}}$) are well overlapped, demonstrating that TTS principle is valid into the viscoplastic domain. Moreover, the shift factors $a_T(T)$ identified for viscoelasticity are still valid in viscoplasticity for the studied polymer.

4. Numerical validation

4.1. Implementation of TTS principle

The time temperature superposition principle has been validated experimentally in viscoelasticity and viscoplasticity. The identified model (Eq (3.9)) is now implemented into the constitutive laws of Balieu et al. behaviour model [12–14] in order to validate the time temperature superposition model thanks to a correlation study.

Please note that main constitutive equations of Balieu et al. model are presented in Appendix. Briefly, this model considers strain partition, so that the total strain, ϵ , is splitted into a reversible viscoelastic part, ϵ^{ve} , and an irreversible viscoplastic part, ϵ^{vp} . Beyond the limits of viscoelasticity, G' self hardening law [20,21] is considered for describing the plastic flow (Eq (5.1)).

$$R(\kappa) = Q_1 \kappa \exp(-b_1 \kappa) + Q_2 (1 - \exp(-b_2 \kappa)) + b_3 \kappa^3 + b_4 \kappa^2 + b_5 \kappa \quad (5.1)$$

where κ is the cumulated viscoplastic strain, defined by $\kappa = \sqrt{2/3} \epsilon^{\text{vp}} : \epsilon^{\text{vp}}$ and the Q_i and b_i are material parameters.

Viscous effects are then introduced based on overstress theory and Perzyna law (Eq (5.2)).

$$\sigma^v = (\sigma_t + R(\kappa)) \left(\frac{\dot{\kappa}}{\kappa_0} \right)^n \quad (5.2)$$

with σ^v the viscous overstress, $\dot{\kappa}$ the equivalent viscoplastic strain rate, σ_t the initial yield stress in quasi-static tension and κ_0 a threshold of

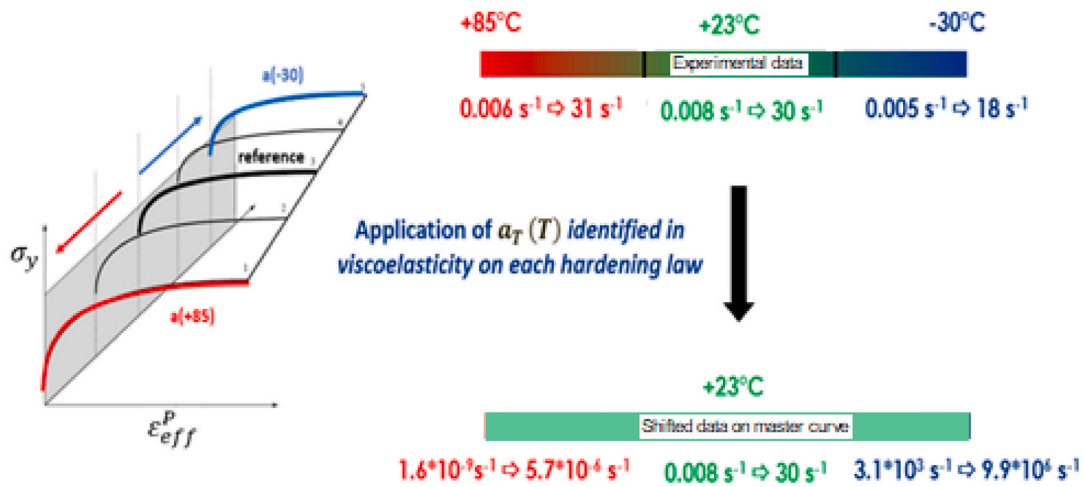


Fig. 11. Mechanism of the time temperature superposition application in viscoplasticity.

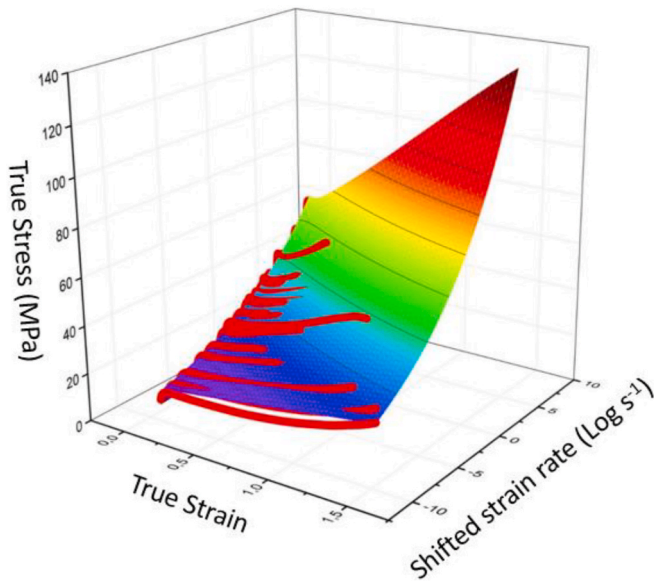


Fig. 12. Behaviour surface obtained by the SEE Method at temperatures between -30 and + 85 °C.

equivalent viscoplastic strain. The identification of the parameters is carried out with the “SEE Method” [18,19], using optimization process (Origin tool).

To take the TTS principle into account, a new specific time step,

$\Delta t'(T)$, is introduced in the discrete domain and formulated as follow:

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

$$\Delta t'(T) = \frac{\Delta t}{\exp(C(T - T_{ref}))} \tag{5.3}$$

where Δt is the time step of the finite element calculation.

The time temperature superposition acts on the viscoelastic and viscoplastic properties without differentiation between the two mechanisms as proposed by the use of same shift factors $a_T(T)$. In practice, constitutive equations are solved using the material parameters identified at reference temperature, but the strain rate is adapted in the viscoelastic and viscoplastic constitutive laws using the modified time step. This way, behaviour can be numerically predicted over a wide range of temperature, in addition to a wide range of strain rate. It should be noted that the self-heating by plastic work deformation is not taking into account in the model but it is implicitly introduced as acting on experimental data used for the characterization of behaviour laws.

4.2. Correlation study

To validate the robustness of the behaviour model the tensile tests used for the identification of the parameters of the model are numerically simulated and compared to experiments. Ls Dyna explicit finite element code is used together with a user-defined subroutine made of viscoelastic-viscoplastic constitutive laws coupled with TTS principle, as described in section 5.1.

For this study, full integration 2D shell elements with 5 integration

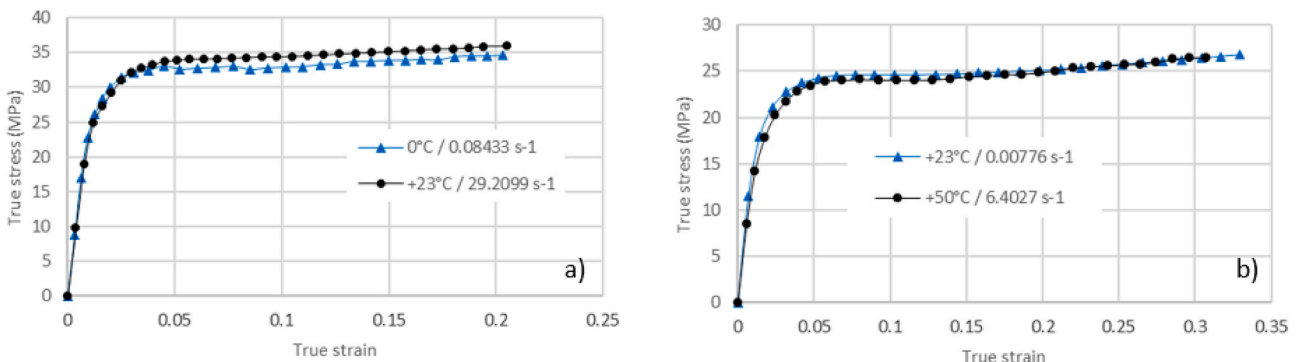


Fig. 13. 2 couples tested a) 0 °C/+23 °C b) +23 °C/+50 °C.

points in the thickness are selected. The size of the mesh is 0.5 mm. The thickness is adjusted for each specimen according to the actual thickness variability measured on injected plates. The average value is around 2.5 mm. Experiments and numerical simulations are compared for tensile tests at various strain rate and temperature (Fig. 14).

The comparative study shows a good level of correlation, ie. experiments and numerical tensile behaviour are very close to each other. The extended behaviour model therefore reproduces accurately the whole behaviour of the polymer between $-30\text{ }^{\circ}\text{C}$ and $+85\text{ }^{\circ}\text{C}$ for a wide range of strain rate. From only one behaviour law characterized at a reference temperature, it is now possible to describe the behaviour for a wide range of test conditions by coupling the effect of strain rate and temperature. Correlation study is enriched by simulating material behaviour under the two equivalent strain rate/temperature couples identified in section 4.2 (Fig. 15).

For the two tested couples, the time temperature superposition principle, already experimentally checked, is now proven to be well reproduced numerically. The correlation level is satisfying even if some gaps are observed, mainly due to harsh characterization of hardening law parameters.

5. Towards a streamlined procedure for behaviour characterization of a semi-crystalline polymer in a wide range of strain rate and temperature

After establishing that the time temperature superposition principle can be applied in viscoelasticity and viscoplasticity using the same shift factors identified in viscoelasticity, a new characterization procedure of polymer behaviour, in a wide strain rate and temperature ranges, can now be proposed, aiming at significantly reducing the number of required experiments (Fig. 16). Note that the same number of DMA, under different temperature, is required for both classic and optimized procedures. Currently, with a classical method of characterization (here based on first author's own professional experience in automotive sector), 15 tests are necessary per temperature (5 different strain rates and 3 tests to assess repeatability) to characterize viscoplasticity and temperature sensitivity. In case of 3 different temperatures (as usually considered in automotive sector), 45 tests are therefore needed. New characterization procedure allows a dramatic reduction up to 17 tests (15 tests at a reference temperature + 2 tests at other temperatures to validate the numerical response), as explained hereafter. In addition, these 17 tests allow behaviour characterization over a wide range of temperature and not for only 3 discrete values.

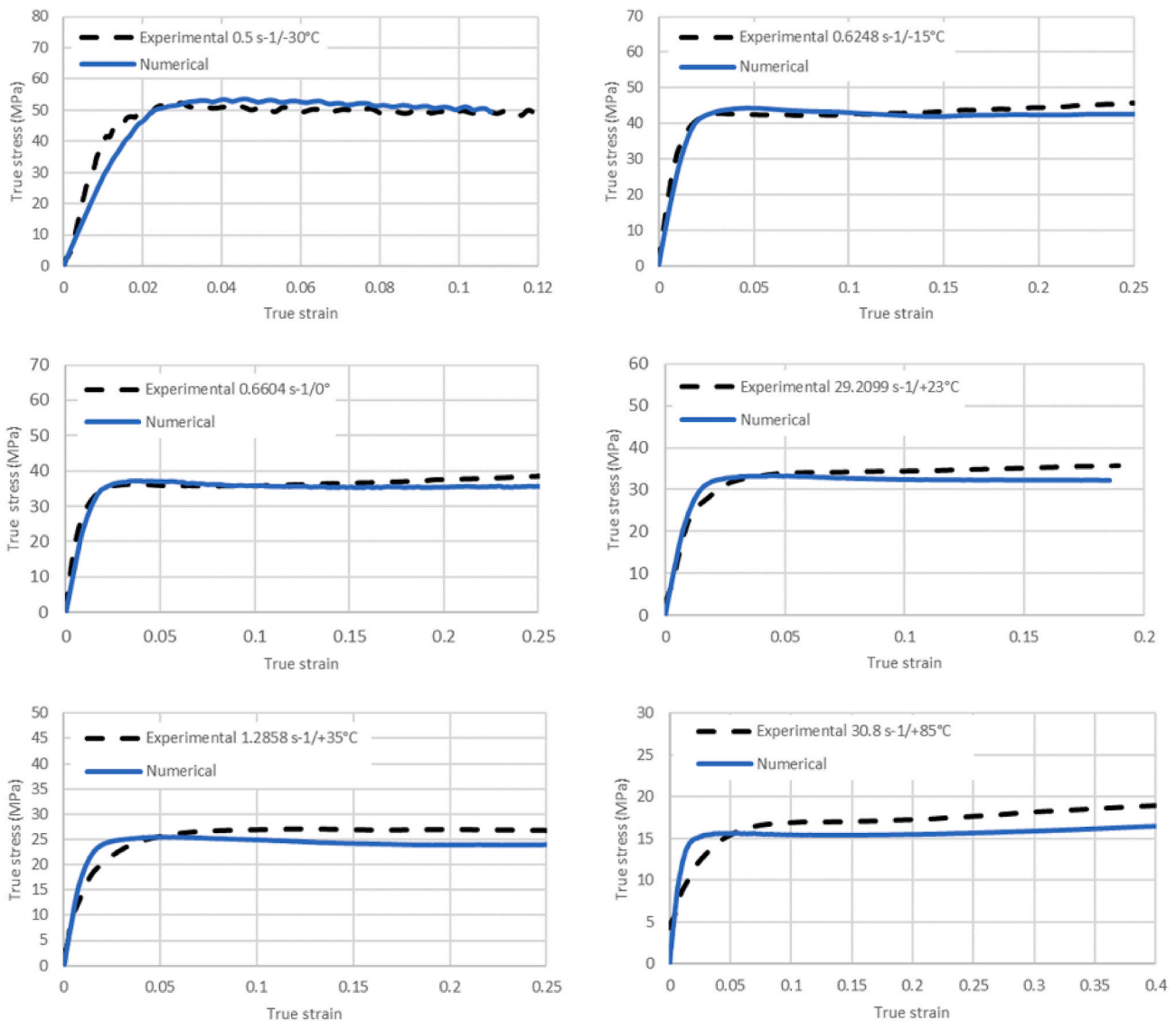


Fig. 14. Comparative study between experiments and behaviour law for various strain rate and temperature.

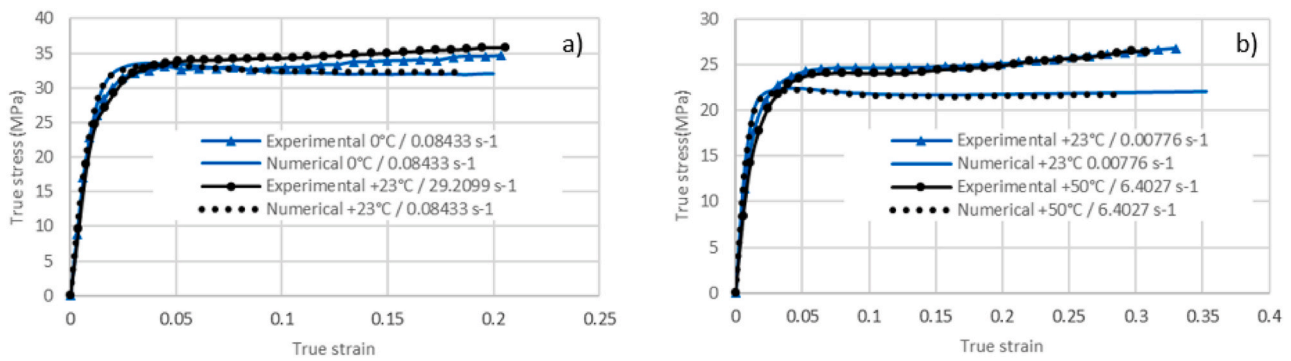


Fig. 15. Comparative study between experiments and numerical simulation for two couples a) 0 °C/+23 °C and +23 °C/+50 °C.

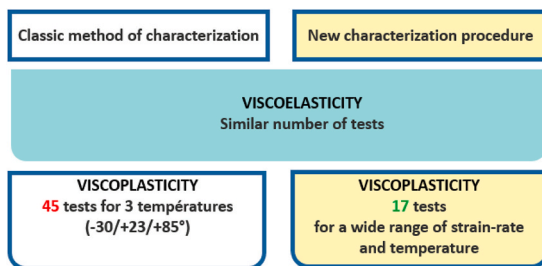


Fig. 16. Estimation of the number of tests for a whole characterization of a semi-crystalline polymer by introducing the time temperature superposition principle.

Indeed, the implementation of TTS principle into viscoelastic/viscoplastic behaviour model allows to predict material response under strain rate and temperature which are not tested and/or not reachable with classic tensile test devices, thanks to numerical simulation. The new approach is detailed in Fig. 17:

As an example, it is proposed to characterize the viscoelastic/viscoplastic behaviour of a polymer from DMA results between -30 °C and +85 °C and tensile tests at +23 °C at different strain rates (Fig. 18). By applying TTS principle, tensile tests at temperature different from ambient temperature are not required anymore (Table 2).

Fig. 19 shows the experiments and the numerical simulation based on classic or optimized procedure of characterization.

For all the tests, the two simulations give similar results. In particular, the optimized procedure (viscoplastic model identified at +23 °C coupled with TTS principle) allows a good correlation level at other temperature. These results validate the new streamlined characterization procedure. Indeed, tensile tests at a reference temperature and some different strain rate are enough to characterize the viscoplastic properties of the material in a wide range of temperature.

6. Conclusions

This paper deals with the viscoelastic and viscoplastic behaviour characterization for a semi-crystalline polymer, under wide strain rate and temperature ranges.

First of all, the viscoelastic properties have been characterized by DMA for temperature ranging from -30 to +85 °C. The time temperature superposition (TTS) principle has been introduced and checked. A mathematical model which allows to link the shift factors to the temperature has been identified for this polymer. Some tensile tests have been carried out at different strain rates and temperature to characterize viscoplastic properties. The results show a superposition of tensile behaviour for different strain rate/temperature couples, thus suggesting that TTS can be considered also in the viscoplastic domain for the present material. For that, the same shift factors as in viscoelasticity are

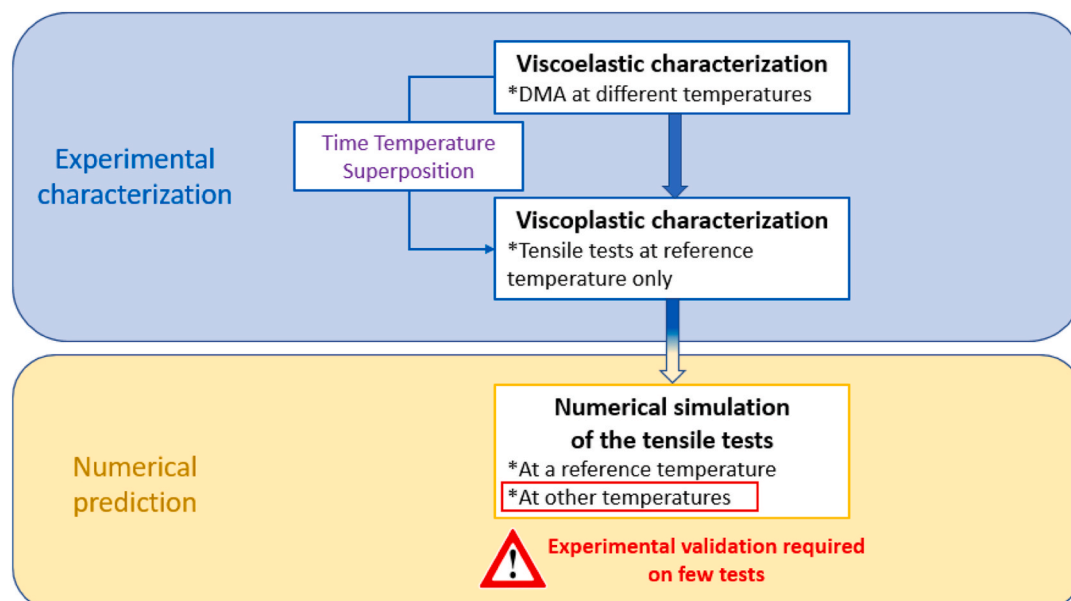


Fig. 17. New characterization procedure for a semi-crystalline polymer for a wide strain rate range and temperature.

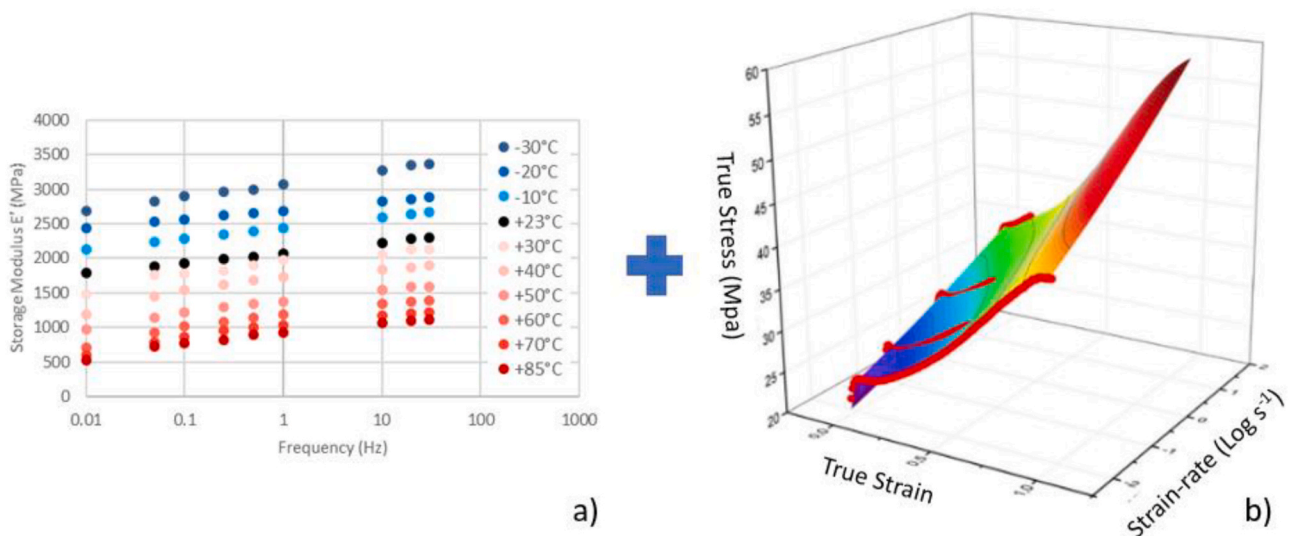


Fig. 18. Data's input for the behaviour law with the new characterization procedure: a) DMA results between -30 and $+85$ °C, b) Behaviour surface at $+23$ °C.

Table 2
Nature of experiments required for whole behaviour characterization.

		Classical method of characterization	Optimized method
DMA	+23 °C	Required	Required
	Other temperature	Required	Required
Tensile tests	+23 °C	Required	Required
	Other temperature	Required	Useless

applied to shift the strain rate in viscoplastic constitutive law (Perzyna model). Then, TTS principle has been validated experimentally. For the studied polymer, it has therefore been established that the TTS principle can be applied in the viscoplastic domain by keeping the same shift factors identified in viscoelasticity.

One section has been dedicated to the implementation of the time temperature superposition principle in the model developed by Balieu et al. A numerical validation has been proposed by simulating several tensile tests for different temperatures and strain rates. The results highlight a good correlation level.

The modification of the behaviour law has therefore been validated and a new characterization procedure has been established. For our material, it is possible to reduce the viscoplastic characterization to a unique reference temperature, knowing the shift factors identified in

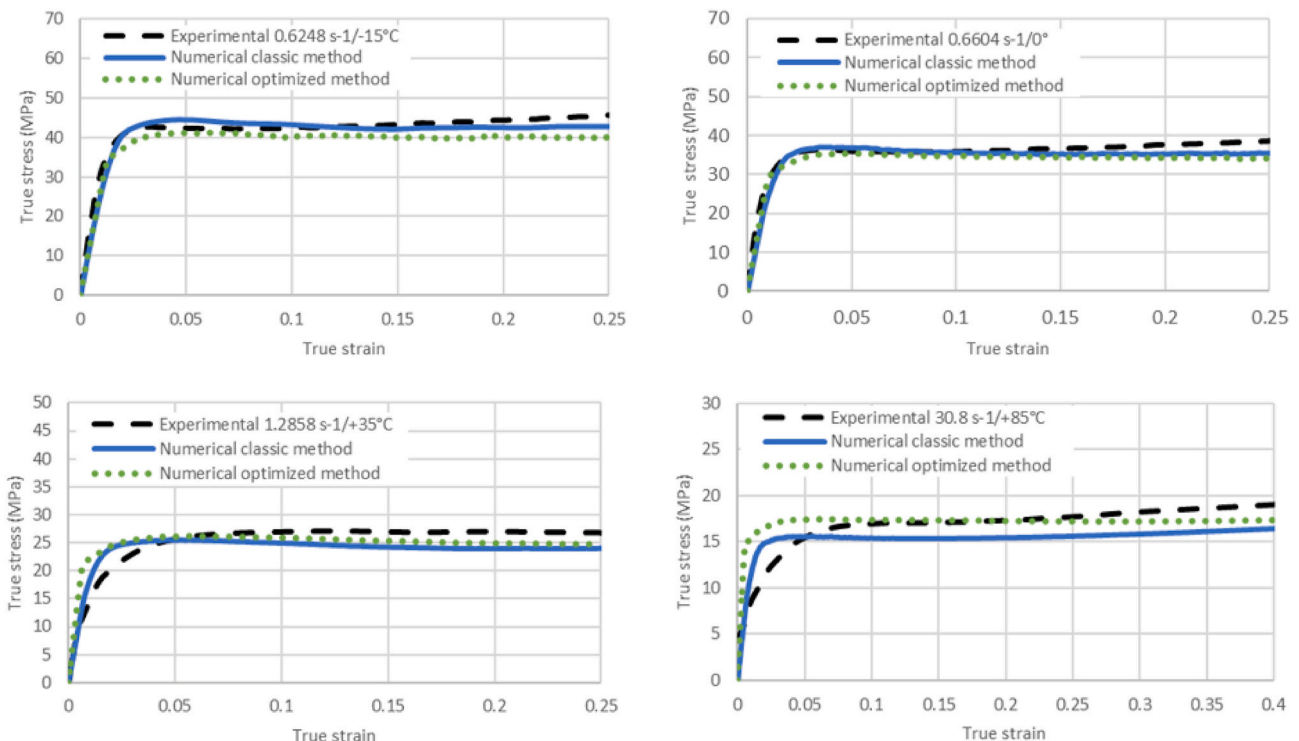


Fig. 19. Validation of the new characterization procedure.

DMA. It is now possible to describe the polymer behaviour in a wide strain rate range and temperatures from the characterization of a viscoplastic behaviour law to a unique temperature, by coupling the sensitivity time/temperature. Therefore, the TTS principle offers the possibility to significantly reduce the number of tensile tests for the viscoplastic characterization and to reach some strain rates not achievable with classical test devices. In a future work, it will be investigated whether the time temperature superposition can be applied for failure prediction.

Author statement

Vincent Dorléans: Conceptualization, Formal analysis, Investigation, Methodology, Validation, Writing – original draft, Writing – review & editing, Rémi Delille: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Writing – original draft, Writing – review & editing, Delphine Notta-Cuvier: Conceptualization, Formal analysis, Investigation, Methodology, Validation, Writing – original draft, Writing – review & editing, Franck Lauro: Conceptualization,

Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Writing – original draft, Writing – review & editing, Eric Michau: Funding acquisition, Project administration, Resources, Software, Supervision, Validation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability statement: no availability for confidentiality purpose.

Appendix. Principle of Time Temperature Superposition – building of the master curve in viscoelasticity

In viscoelasticity, TTS principle is based on DMA. Isotherm evolution of complex or storage modulus with frequency is first plotted and a single master curve, at a reference temperature (T_0), is search (Fig. A1). To that aim, a shift factor, $a_T(T)$, dependent on temperature, is applied to each storage modulus measured at another temperature in order to align all the results on the master curve (at T_0). Obviously, shift factors are applied to translate a given curve segment to higher frequency, in the case of $T > T_0$, or to lower frequency, in the case of $T < T_0$ (Fig. 4).

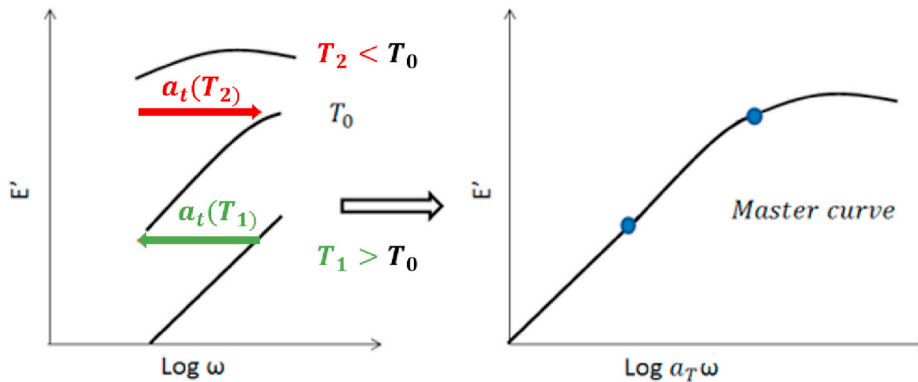


Figure A1. Building principle of a master curve.

From the master curve, it is possible to define a couple (shifted strain rate, given temperature) which gives an equivalent behaviour at another strain rate at reference temperature, provided that shift factors can be identified.

In the literature, several authors have developed models which allow to predict the evolution of the shift factors with temperature. For example, Williams-Landel-Ferry [5,6] proposed the following model:

$$\log a_T(T) = \frac{-C_1 (T - T_{ref})}{C_2 + (T - T_{ref})} \quad (\text{A1.1})$$

where T and T_{ref} are respectively the current temperature and the reference temperature. C_1 and C_2 are two material parameters to be identified.

Another model describing the dependence of the shift factors to temperature is proposed by Arrhenius [7]:

$$a_T(T) = \exp\left(\frac{E_a}{R} \left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right) \quad (\text{A1.2})$$

With E_a the activation energy and R the gas constant.

As already stated, TTS principle has been checked for thermoplastics such as PP [8], PA [9,10] or PMMA [11], in the viscoelastic domain.

Appendix. Main constitutive equations of model of Balieu et al.

General statements

The framework of small deformation is considered and total strain, ϵ , is split into a reversible viscoelastic part, ϵ^{ve} , and an irreversible viscoplastic

part, ε^{vp} . Coupling with damage model is not presented here, as it is not considered in the present article.

Main constitutive equations in viscoelasticity.

Linear model of Wiechert (or generalised Maxwell model) is considered. This rheological model is composed of n Maxwell elements (linear spring and damper in series), disposed in parallel, and of an additional branch with a spring, to model elastic recovery. The expression of the stress tensor, $\sigma(t)$, in viscoelasticity is given by:

$$\sigma(t) = \int_0^t R^{ve}(t-\zeta) \frac{d\varepsilon^{ve}}{d\zeta} d\zeta \quad (\text{A2.1})$$

where R^{ve} is the relaxation tensor.

Yield surface and plasticity criterion

Raghava's plasticity criterion [16] is selected to take the behaviour sensitivity to hydrostatic pressure into account. Then, the expression of the yield surface, f , is given by:

$$f(\sigma, \kappa) = \frac{(\eta - 1)I_1(\sigma) + \sqrt{(\eta - 1)^2 I_1^2(\sigma) + 12\eta J_2(\sigma)}}{2\eta} - \sigma_t - R(\kappa) \quad (\text{A2.2})$$

where $I_1(\sigma)$ et $J_2(\sigma)$ are respectively the first and second invariant of the stress tensor. κ is the cumulated viscoplastic strain, defined by $\kappa = \sqrt{2/3\varepsilon^{vp} : \varepsilon^{vp}}$. η is a material parameter that governs flow dependency to hydrostatic pressure. It is defined by the ratio between the initial yield stress in compression, σ_c , and the initial yield stress in tension, σ_t , so that $\eta = \sigma_c/\sigma_t$. R is the hardening law. In present work, a non-linear hardening law, adapted from G'Sell model [18] is considered (Eq. (A2.3)).

$$R(\kappa) = Q_1 \kappa \exp(-b_1 \kappa) + Q_2 (1 - \exp(-b_2 \kappa)) + b_3 \kappa^3 + b_4 \kappa^2 + b_5 \kappa \quad (\text{A2.3})$$

where Q_i and b_i are material parameters.

Non-isochoric viscoplastic flow

The thermodynamic potential, ϕ , is defined to take a non-isochoric plastic flow into account, as stated by Eq. (A2.4).

$$\phi = \sqrt{3J_2(\sigma) + \alpha^+ \langle p \rangle^2 + \alpha^- \langle -p \rangle^2} \quad (\text{A2.4})$$

where $\langle . \rangle$ is the Macaulay operator, stating that $\langle p \rangle = \left(\frac{p+|p|}{2} \right)$. α^+ and α^- are material parameters which model the positive or negative volume variation during viscoplastic flow, depending on the sign of hydrostatic pressure, p . The thermodynamic potential is used in normality laws to compute the viscoplastic strain increment, in particular.

Viscoplasticity is modelled by the computation of a viscous overstress, σ^v , following Perzyna's model (Eq. (A2.5)) [17].

$$\sigma^v = (\sigma_t + R(\kappa)) \left(\frac{\dot{\kappa}}{\dot{\kappa}_0} \right)^n \quad (\text{A2.5})$$

where $\dot{\kappa}$ is the time derivative of the cumulative viscoplastic strain, κ (i.e. equivalent viscoplastic strain rate). n and $\dot{\kappa}_0$ are material parameters.

Finally, a viscoplastic yield surface can be expressed, taking into account viscous overstress:

$$f^{vp} = \frac{(\eta - 1)I_1(\sigma) + \sqrt{(\eta - 1)^2 I_1^2(\sigma) + 12\eta J_2(\sigma)}}{2\eta} - (\sigma_t + R(\kappa)) \left\{ 1 + \left(\frac{\dot{\kappa}}{\dot{\kappa}_0} \right)^n \right\} = 0 \quad (\text{A2.6})$$

All those constitutive equations are implemented in user defined material card of Ls-Dyna FE software. An explicit formulation is considered to solve the classical (visco)elastic trial-(visco)plastic correction scheme.

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