EFFECT OF THERMAL AGEING ON THE BEHAVIOUR OF GLASS POLYETHYLENE COMPOSITE : EXPERIMENTS AND MODELLING

L. Meylheuc(*), Y. Remond(*) and Ch. Wagner(**)

(*)LPMP EP647 CNRS
Université Louis Pasteur – ECPM
25, rue Becquerel
F-67087 Strasbourg Cedex
(**)LPMT
Université de Haute Alsace – ENSITM
11, rue Alfred Werner
F-68093 Mulhouse Cedex

SUMMARY : After defining different ways to implement accelerated ageing of a reinforced short glass fibre thermoplastic, we try to study the changes of its mechanical behaviour in function of time. A model is then build up with an ageing internal variable taking into account the viscoelasticity and the damage evolution of the material.

KEYWORDS : ageing, polyethylene, short glass fibre, creep, tension test

INTRODUCTION

The performance of composite materials is conditioned by their mechanical properties. For thermoplastics, these properties are often too bad for mechanical parts. However, composite materials are in common use since they are easy to manufacture and very cheap. The use of these materials at industrial ends is conditioned by its long-term behaviour.
The study of the ageing of PE has been known for a long time, the short fibres addition results in modifying deeply theories used initially. In the case of composites ageing, observed phenomena are more varied and thus, no predictive method of lifespan calculations currently seems to give whole satisfaction. Most current methods are founded on the time-temperature equivalence, extensively used for polymers. But those methods raise difficulties for the semicrystalline in function of some temperature ranges. Other methods take account approaches in time-stress equivalence.
On the macroscopical level, it is necessary to handle carefully these two classes of methods insofar, since various ageing mechanisms could be activated depending on each material constituent. Each of them has specific thresholds and ageing kinetics. Indeed, the matrix

ageing, the fibre-matrix interface ageing, and the fibre ageing do not have the same evolutions. That is why it is very difficult to obtain a global ageing modelling.
On the other hand, microscopic approaches try to modelise the ageing behaviour according to the microstructure of each component. In spite of many results, these approaches are difficult to implement because of the complex behaviours and the complex geometries. All of these approaches are reviewed in [Cardon 96], [Choqueuse 98], [Bobet 98], [Halary 86] and [Van Ger Griten 96].
Our modelling of GF-PE long time behaviour, linking the time-temperature to corresponding ageing, is here presented with a specific analysis of quasi-static mechanical tests of the composite after ageing. This approach seems more reliable than others exclusively based on creep-rupture measurements, whose results are scattered.

EXPERIMENTAL METHODOLOGY

The composite investigated was a medium density polyethylene reinforced with 20% of short glass fibres. The fibres length are measured after calcination, according to the standard NF-T-57.518,and are given fig. 1. Samples were obtained by an extrusion molding process. This process favours a fibre orientation in the sample axis as show the X-Ray micro-tomography views (fig. 3). The left and right was parts of the figure represent respectively paralled and orthogonal views of the sample axis. The right view is perpendicular to the sample axis. The average fibres length is 0.4 mm. Although it is smaller than the 2 mm critical length [Simon 84], it has a favourable effect, especially in the elasticity modulus and strength.
Accelerated ageing consists here in creep tension tests in 3 steel boxes with regulated water. Such experiments are often used for pure PE. The immersion in water is moreover associated with the operating conditions of the materials.
These accelerated ageing method can be evaluated by analysing evolutions of time-temperature shift factors. These shift factors were calculated first with creep curves analysis and then with RDA (Rheometrical Dynamical Analysis) measurements (fig. 2). The obtained curve shift factors affine of temperature [Davant 98], because they represent the lowest boundary of the modelling.
.

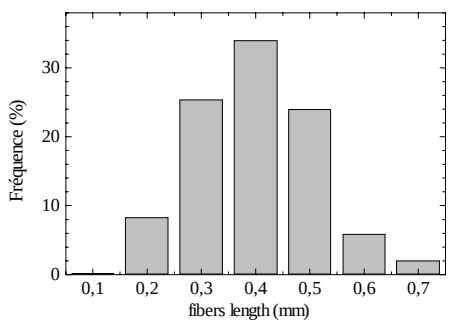

Fig. 1: Fibres length repartition frequency

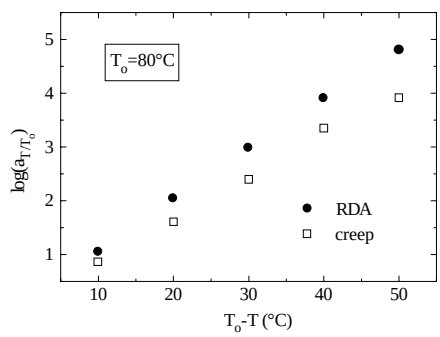

Fig. 2 : Time-temperature shift factor

180 samples were tested under various tension-creep loads in boxes filled with temperature regulated water at 30°C, 60°C or 80°C. After 30 ageing days, samples are removed from their ageing test stand and are tested in a tension machine after their viscoelastic recovering during

several days. The mechanical behaviour is then identified for a real ageing of 30 days to 8000 days (with the temperature reference T_{ref}= 30°C). The tensile tests are carried out at ambient temperature, at a constant strain velocity with return to null stress in a INSTRON tension machine. The apparent young's modulus related to the successive discharges, the maximal stresses and the strains are then measured.

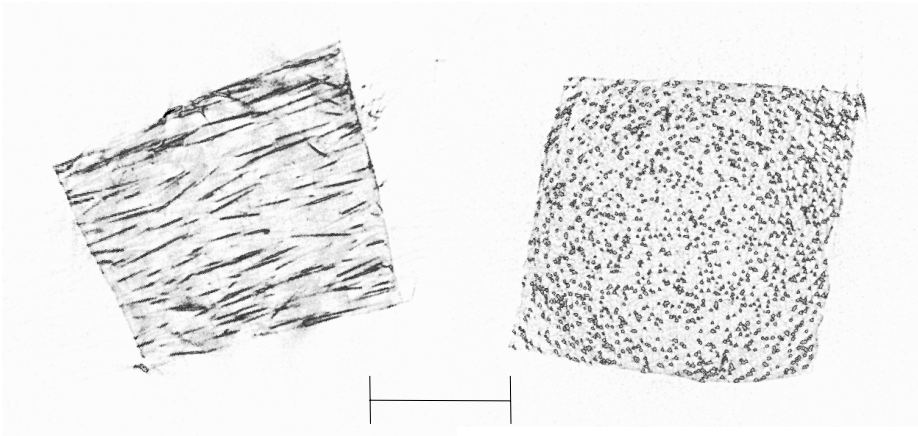

Fig.3 : X-ray microtomographic views of GF-PE

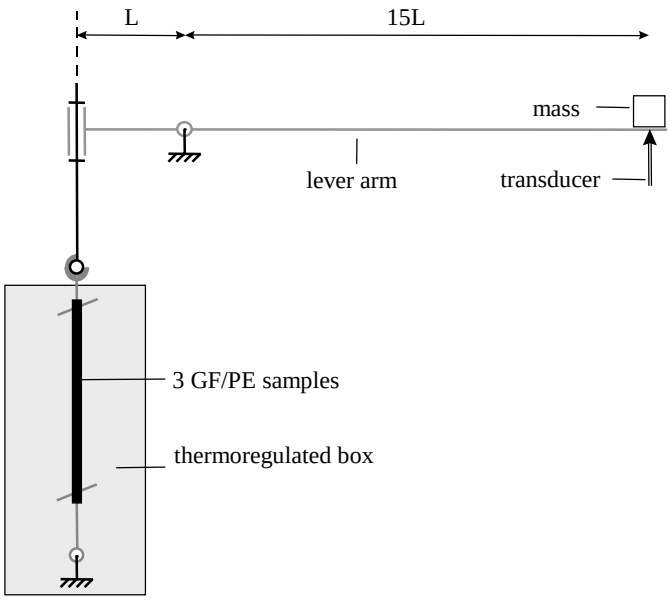

Fig. 4 : Experimental creep setup

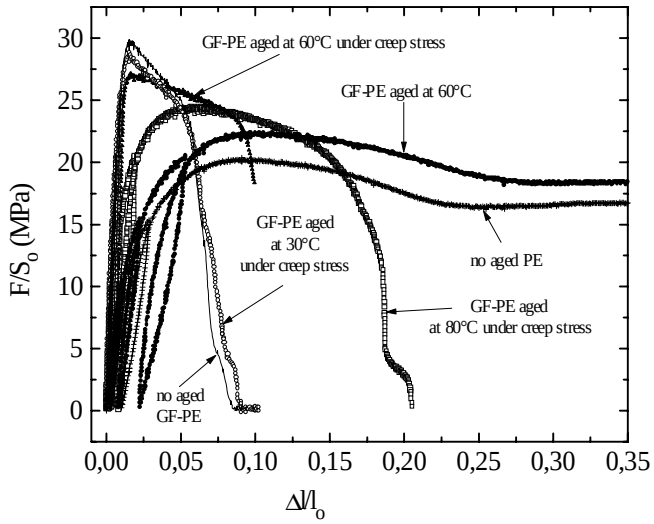

Fig. 5 : Tension curves after ageing of PE and GF/PE

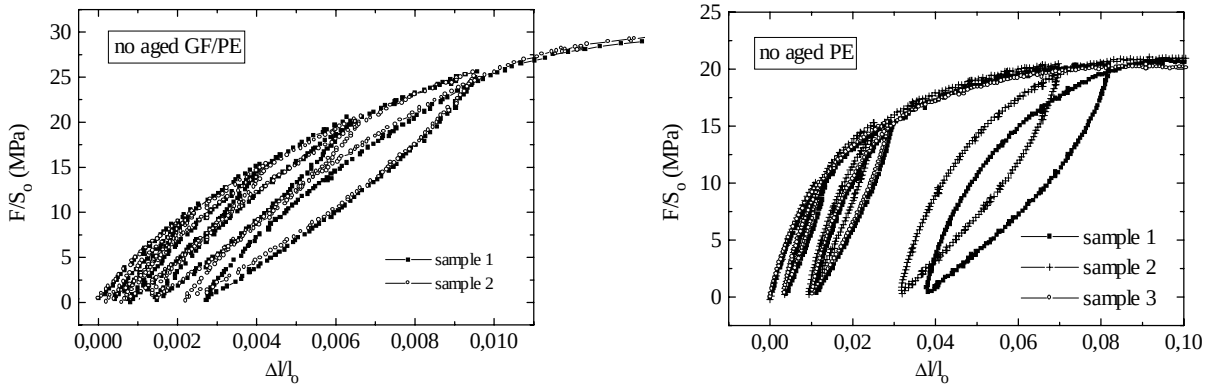

Fig. 6 : Tension behaviour of non aged PE and GF-PE

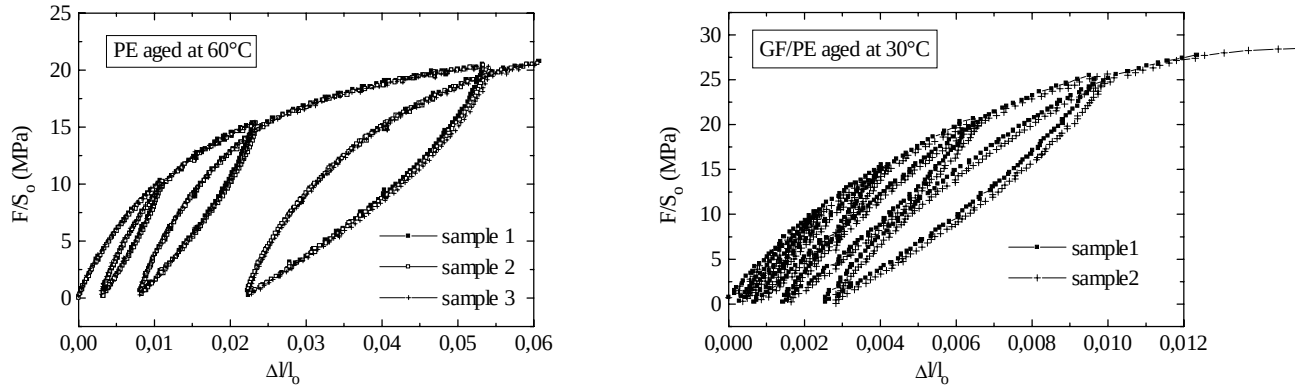

Fig. 7 : tension behaviour of PE and GFR-PE after ageing at 30°C and 60°C

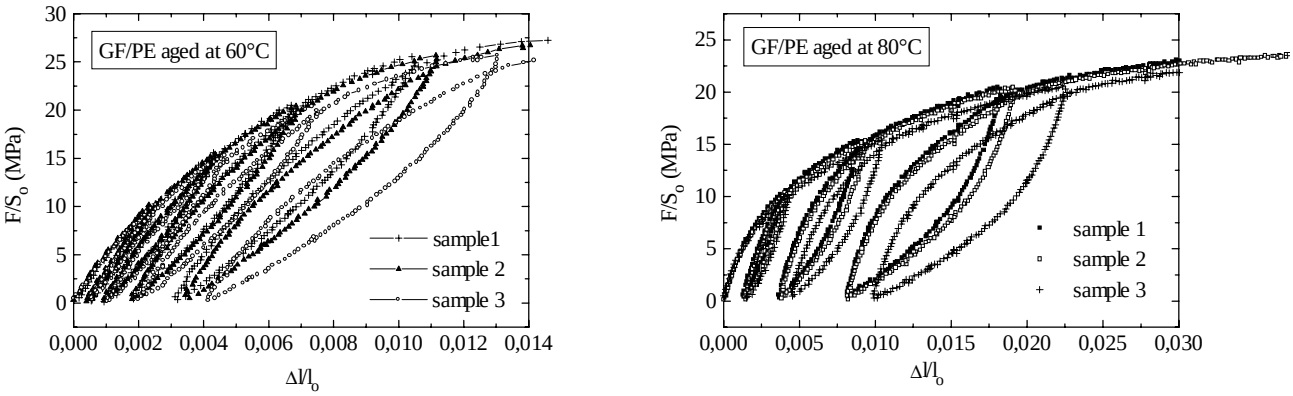

Fig. 8 : Tension behaviour of GF-PE after ageing at 60°C and 80°C

RESULTS AND MODELLING

Mechanical behaviours in traction of the healthy and aged GF-PE are given fig. 5, 6, 7 and 8 like those of the healthy and aged PE. The very small deformations remainders, the results are presented in conventional stresses and deformations. The measurement of the Young's moduli of return to null constraint of the GF-PE composite and the PE constitutes a good indicator of the mechanical behaviour evolution of these materials. Indeed, they integrate the matrix and interfaces viscoelastic behaviour evolution as well as the appearance of a specific damage of the interface starting from a threshold in unknown stress level.

The damage of the composite is non-existent below 10 MPa or for deformations lower than 2 to 3%. Therefore the decreasing of the apparent modulus is mainly due to the viscoelasticity, then a coupling is done with a progressive damage of the interfaces. It is interesting to compare these modules defined in a given stress level and for two different states of ageing. We obtain then a measurement of the effect of ageing on material. We propose to build an internal variable of ageing V_σ defined by the report of module of an aged material for one given period Δt on the module of the none aged material request by the same constraint.

$$V_\sigma = \frac{E_{\Delta t}}{E_{\Delta t=0}} \qquad (1)$$

This state variable leads then to a thermodynamical model as the classical damage variable D. D. We define

$V_\sigma=1 \leftrightarrow$ Material without ageing
$V_\sigma>1 \leftrightarrow$ Material displaying hardening with ageing
$V_\sigma<1 \leftrightarrow$ Material displaying softening with ageing

The identification of the variable V_σ is given fig. 9, and show that the GF-PE Young's modulus decrease with ageing. The fig. 10 shows the evolution of the ageing amplitude versus time. The ageing amplitude ΔV after a given ageing time t, is measured by the variation of V_σ between $\sigma = 0$ and σ_{max}. The prolongation of this curve up to $\Delta V = 0$ permits to identify the time threshold t_s from which the ageing is beginning. We obtain $t_s = 10^6$s. Note also that the ageing effect on the PE without fibres induces hardening ($V_\sigma>1$).

The ageing kinetics is quite linear versus log(t) (fig. 10) ; a linear model is then built up as follows:

$$t < t_s \quad \leftrightarrow \quad V_\sigma = 0$$
$$t \geq t_s \quad \leftrightarrow \quad V_\sigma = 1 - f(\sigma)\log\frac{t}{t_s}$$

Where $f(\sigma)$ is an linear function given by $f(\sigma)=a\sigma+b$ with $a=5{,}3.10^{-3}$ and $b=0{,}068$. For each strain rate, a critical value of V_σ can be identify ; this model becoming then an useful tool for simulation (fig.11).

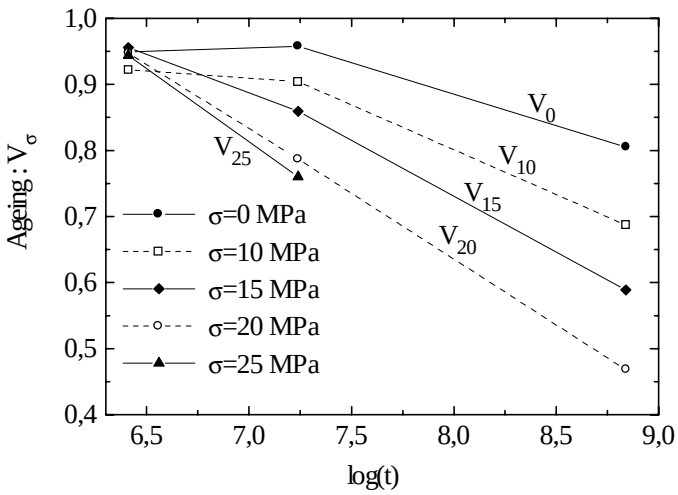

Fig. 9 : Ageing variable evolution for different stress level

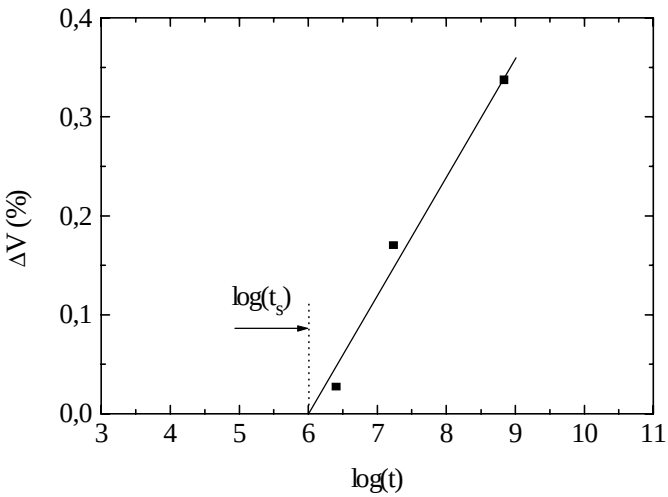

Fig. 10 : Ageing threshold time identification

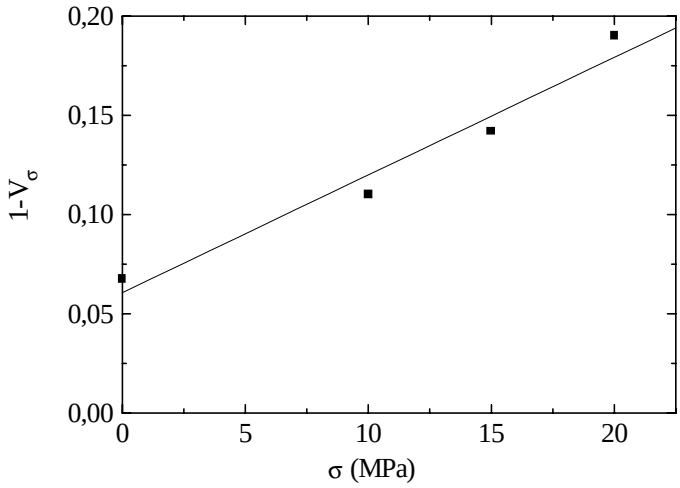

Fig. 11 : Identification of the ageing model

(MPa)	GF/PE T=20°C	GF/PE 30 d. at 30°C	GF/PE 30 d. at 60°C	GF/PE 30 d. at 80°C	PE T=20°C	PE 30 d.at 60°C
E_0	5900	5600	5650	4750	1650	2000
$E_{\sigma=10MPa}$	5530	5100	5000	3800	1750	1370
$E_{\sigma=15MPa}$	5180	4950	4450	3050	850	1050
$E_{\sigma=20MPa}$	4700	4450	3700	2200	570	675
$E_{\sigma=25MPa}$	4080	3850	3100			
σ_{max}	30	29	27.5	24	20	22
$\varepsilon_{\sigma max}$(%)	1.5	1.5	2	5		

Table 1 : Mechanical characteristics of PE and PEFV after ageing

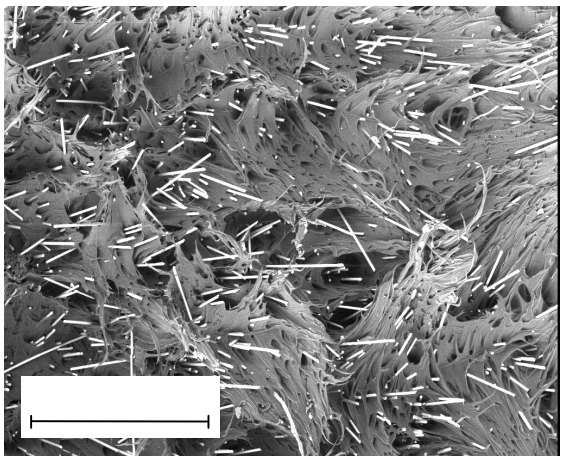

Fig. 12 : Typical tensile fracture surface of GF-PE

CONCLUSION

The analysis of the ageing effect on a composite with thermoplastic matrix was identified by the time-temperature equivalence method. But this method, raises nevertheless some difficulties because they include a mixture of phenomena. An ageing indicator is built in this study. It allows by identification, to model and to consider the ageing evolution up to the critical values.

REFERENCES

[Bodet 98] :Bobet V., Lory P. "Prévision de la durée de vie en fatigue des pièces mécaniques en matériaux composites" JST AMAC – Commission durabilité, Arcueil 1998, pp.14-20.

[Cardon 96] : Cardon A.H. "Comportement à long terme des systèmes composites à composante polymèrique : objectifs, développement récents et orientations de recherche, Durabilité des matériaux composites, JST AMAC 1996, pp. 5-13.

[Choqueuse 98] : Choqueuse D., Davies P., Roy A. "Comportement à long terme – application au milieu marin", JST AMAC – Commission durabilité, Arcueil 1998, pp. 24-26.

[Davant 98] : Davant M., Rémond Y ., Wagner C. "Etude du vieillissement de polymères thermoplastiques chargés de fibres de verre ", C.R. JNC11, Arcachon, 1998, Vol.1, pp. 295-301.

[Halary] : Halary J.L. "Traitement pratique des résultats des essais viscoélastiques" Introduction à la mécanique des polymères, INPL-MECAMAT-GFP-APOLLOR-FIRCHTEC, pp. 169-189.

[Simon 84] : Simon H. "Propriétés interfaciales dans les composites : énergie de surface et adhésion fibre de verre/matrice macromoléculaire" Thèse de 3ème cycle, Mulhouse 1984.

[Van der Grinten 96] : Van der Grinten F. , Wickers Scheur P.W.M. "Use of fatigue testing to evaluate long term performance of polyethylene", Plastics, Rubber and Composites Processing and Applications, 1996, Vol.25 n°6 pp. 294-298.