Experimental study of fatigue behaviour of polypropylene-based micro and nanocomposites

M. Basso
Università di Padova, Dipartimento di Processi Chimici dell'Ingegneria

A. Crevatin
Università di Trieste, Dipartimento dei Materiali e delle Risorse Naturali

M. Celotto, M. Sanità
Electrolux Home Products Italy, CTI's Materials and Chemistry Department

RIASSUNTO. Oggigiorno molte realtà industriali stanno effettuando grandi sforzi nella direzione della riduzione costi, incremento della produttività e miglioramento della qualità dei prodotti. In modo particolare abbiamo focalizzato la nostra attenzione nel ramo del design di componenti strutturali termoplastici, includendo sia l'ottimizzazione delle strutture esistenti, sia il design di nuove strutture. Dal punto di vista dei materiali, questo si traduce nell' esigenza di avere determinate proprietà meccaniche, di porre attenzione ai margini di sicurezza e nella necessità di un controllo dei parametri chiave del design. Per ottenere questi risultati è necessario un nuovo approccio nel design di materiali plastici e componenti che incontrino le richieste di durata e performance.

L'obiettivo del presente progetto è quello di trovare strumenti utili alla previsione della durata in vita e dell'evoluzione del danneggiamento di materiali plastici e componenti sottoposti a carichi meccanici/termici in condizioni di servizio, in modo da supportare lo sviluppo di nuovi materiali con diverse formulazioni ed il design e l'ottimizzazione di componenti strutturali.

In modo particolare abbiamo incentrato il nostro lavoro sulla caratterizzazione e modellazione della durata in vita e dei meccanismi di danneggiamento dei materiali.

Uno dei principali problemi legati alla durata dei materiali è dovuto al cedimento per fatica. Il processo di fatica è un progressivo indebolimento di un componente nel tempo dovuto ad applicazioni ripetute del carico. Il cedimento per fatica non deve essere visto solo come una rottura del provino in due pezzi separati, ma piuttosto come un progressivo accumulo del danneggiamento. Il danneggiamento di un materiale sollecitato a fatica si manifesta come una progressiva riduzione di stiffness e sotto forma di creep.

Dal momento che i test di fatica standard sono dispendiosi in termini di tempo e denaro, è essenziale sviluppare nuovi approcci che siano rapidi e di facile implementazione. Uno dei risultati più importanti raggiunti nel presente lavoro, è il setting di un metodo di analisi, (test di fatica accelerato) estremamente semplice da implementare, che è in grado di differenziare le performance di diversi materiali e formulazioni in termini di accumulo del danneggiamento e di durata, in tempi brevi.

ABSTRACT. Nowadays most industrial realities undergo a strong push to improve cost-effectiveness, productivity and quality of manufactured products. In particular we focussed our attention in the area of design of plastic structural components, including both optimization of existing structures and design of new ones. In this case, but the following considerations have a more general value, these needs could be translated into demanding requirements of cost-effectiveness, weight reduction, reduced time-to-market with guarantee reliability. From a material perspective this means demanding mechanical performances, attention to safety margins and need of a better control of key design parameters. To obtain these results, we need to develop a new approach and effective tools in the design of plastic materials and components aimed at tailoring part behaviour to endurance and performance requirements.

The target of the project is to find effective tools for predicting life endurance and damage evolution of plastic materials and components under mechanical/thermal service loading, in order to support the development of new material formulations and the design and optimization of structural components.

In a particular way, we focussed our work in the characterization and modellization of materials durability and damage mechanisms.

One of the main problems related to materials durability is due to fatigue failure. Fatigue process is a progressive weakening of a component with increasing time under load such that loads to be supported satisfactorily for short duration produce failure after long durations [1, 2, 3]. Fatigue failure should not be thought only as the breaking of the specimen into two separated pieces, but as a progressive material damage accumulation [2]. Material damage during fatigue loading manifests as progressive reduction of stiffness and as creep [5].

As standard fatigue testing are expensive in terms of money and time, it is essential to develop new approaches less time consuming and simpler to be implemented. One of the most important goals of the present work is the setting of an investigation method (accelerated fatigue test) very simple to be implemented that is able to differentiate damage accumulation and durability performances of various material formulations in reduced time.

KEYWORDS: fatigue, thermoplastic polymers, composite materials, mechanical properties, fracture

1 INTRODUCTION

Standard fatigue testing requires long time to test a single material. The aim of this study is to develop a faster procedure to differentiate materials damage accumulation with a reduced number of cycles in shorter times than those required by conventional life tests.

The accelerated fatigue test developed is a new faster method to obtain information about material damage with the application of a tensile oligocyclic load with different max load levels (in terms of % tensile yield stress) by using a standard universal testing machine. In order to calculate the characteristics parameters for comparing the different materials, a dedicated Matlab® program was developed. In particular we chose as indicators of the cumulative damage for the tested sample: the area of the stress-strain cycle, the evolution of the elastic modulus and the residual strain trend [4, 5, 6].

The accelerated fatigue test was applied to different PP composites in order to understand and compare their fatigue properties and the induced cumulative damage and then compared with data obtained from conventional fatigue test in order to validate the new method.

2 MATERIALS AND MATERIALS PREPARATION

Two different formulations were considered in this work: a $CaCO_3$-PP microcomposite (MAT1) and a $CaCO_3$-Glass-fibre-PP composite (MAT2). Regarding MAT1, three different versions were prepared, varying the PP matrix (S1, S2, S3).

The PP matrices used in the materials are general purpose isotactic homopolymer suited for both technical and packaging injection moulded components.

MAT1 contains about 40wt% of micrometric ground calcium carbonate and others additives (mainly antioxidants).

MAT2 is made of 21wt % of $CaCO_3$, 15 wt% of glass fibres and others additives (0.5 wt %).

The fillers used are all commercial products from different suppliers.

The matrix, the fillers and the additives have been premixed in a dry blender for approximately 15 minutes and then extrusion compounded using a *Maris TM35V* co-rotating twin screw extruder (screw external diameter: 35 mm, L/D=32). Different melt temperatures and screw speeds have been used, in the range of 180-240 °C and 170- 220 rpm, respectively.

Tensile and flexural test samples according to ASTM D 638 and to ASTM D 790 have been injection moulded from the pelletised extrudate using a *Negri Bossi NB 125* injection moulding machine. The melt and mould temperatures have been set at 200 °C and 30 °C, respectively. Specific injection pressure was nominally 600 bar [8].

In Tab 1 and 2, the tensile and flexural properties of the two PP-composites are compared.

3 TEST CONDITIONS

The materials used in this study were tested by uniaxial tensile tests conducted at room temperature (23°C) in an Instron 5583 universal testing machine on injection moulded ASTM standard “dog-bone” shaped tests specimens.

The accelerated fatigue tests were run at a constant amplitude stress level applied at a constant rate. The fatigue stress was applied with a triangular waveform (Fig. 1), meaning that the frequency was not constant. In fact the testing machine used only allowed to control the crosshead speed and to set the maximum stress level (σ_{max}). The maximum stress level was selected on the basis of the tensile properties (yield stress) of each mate-

rial tested, while the minimum stress level was zero.
A maximum of 4000 cycles was chosen for each test. The crosshead speed was 50 mm/min. The axial load and crosshead displacement were stored digitally for each cycle throughout the duration of the test.

4 TESTS DATA ELABORATION

The first problem encountered to describe the fatigue behaviour of the tested materials was to identify which parameters to use to follow the progressive material damage.
G. Tao and Z. Xi [9], for example, studied the uniaxial fatigue behaviour of an epoxy resin with non-contact real-time strain measurement and control system. A relation of strain amplitude vs. fatigue life for fully-reversed strainrange-controlled uniaxial fatigue tests was obtained. Quantitative analyses of evolutions of various mechanical properties (including stress range, elastic modulus, nonlinear stress-strain relation, dissipated strain energy density, etc.) during the entire fatigue life period were carried out based on recorded stress-strain data. From the evolution of the stress-strain hysteresis loops, a gradual degradation of modulus and a decrease of nonlinear effect in stress-strain response were observed. The authors also found that these two phenomena were independent of the loading control mode (stress-control or strain-range-control) and of the mean stress/strain values in the cyclic loading.
A similar approach was applied in this work for the eva-

	MAT1 (S1) (PP-$CaCO_3$)	**MAT2 (PP-$CaCO_3$-GF)**
Tensile yield stress [MPa]	22	-
Tensile yield strain [%]	3	-
Tensile stress at break [MPa]	22	51.5
Tensile strain at break [%]	60	3
Flexural Modulus [GPa]	3200	4200

Table 1: Tensile and Flexural Properties of PP-composites.

	S1	**S2**	**S3**
PP MFI [g/10min] - matrix	7	7	5
% filler	40	40	40
Tensile yield stress [MPa]	22	23	24
Tensile yield strain [%]	3	4.0	3.0
Tensile stress at break [MPa]	22	16.8	18.3
Tensile strain at break [%]	60	60	65
Flexural Modulus [MPa]	3200	3200	3400
80% of yield stress [MPa]	17,5	18,2	19,5

Table 2: Characteristics of the three MAT1 versions PP composites compared.

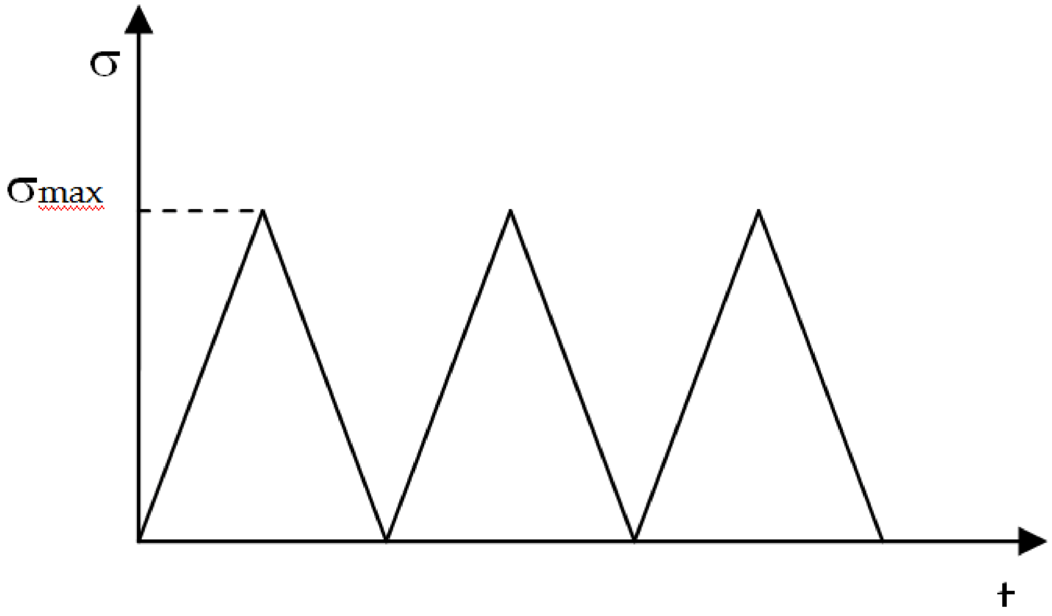

Figure 1: Triangular waveform.

luation of the accelerated fatigue test data. To elaborate the great amount of data, a dedicated Matlab® program was developed for each type of test (conventional and accelerated fatigue tests) since the way the data stored in each test was different: in the conventional tests the data were stored digitally for predefined cycles throughout the duration of the test, while in the accelerated tests the data have been stored digitally for each cycle. In both cases, three channels have been considered: the axial load, the crosshead displacement and the cycle counter. From these channels it was possible to evaluate the stress-strain plot with the hysteresis loops for each stress level tested for a material. A progressive modification of the shape of the hysteresis loops could be observed during the fatigue test, resulting from the progressive changes of various material mechanical properties (progressive damage accumulation). In particular for each cycle the following parameters were considered [4, 5, 6]:

- *Secant Modulus, E [MPa];*
- *Relative Secant Modulus,* E_i/E_0*;*
- *Hysteresis Loop Area [J* 10^{-3}*];*
- *Dissipated Strain Energy [J/*cm^3*];*
- *Maximum Strain,* ε_{max} *[%];*
- *Minimum Strain or Residual Deformation,* ε_{min} *[%];*
- *Strain Range,* $\Delta\varepsilon$ *[%];*

Supposing that only the gauge length of the injected molded ASTM standard "dog-bone" shaped tests specimen participates to sample deformation, the stress and strain have been calculated as:

- *Stress [MPa] = Crosshead Load [N]/ Gauge section of specimen [*mm^2*]*
- *Strain [MPa] =Crosshead Displacement [mm]/Gauge Length [mm]*100*

The secant modulus E, was computed from the stress-strain curve as the slope of hysteresis loop (Fig. 2).
The drop of the secant modulus has been computed as the ratio of the secant modulus of the first cycle (at which the material is supposed not damaged), E_0, and its value at the i-th cycle, E_i.
The hysteresis loop area was calculated integrating with the trapezoidal integration method the axial load versus crosshead displacement plot for each cycle. Consequently the dissipated strain energy was computed as the ratio of the hysteresis loop area and the volume of the gauge length of the tested sample.
The difference between the maximum and minimum strain gives the strain range, Δε.

$$\Delta\varepsilon = \varepsilon_{MAX} - \varepsilon_{MIN}$$

From the comparison of these parameters we could emphasize the differences in the fatigue behaviour of the different polypropylene-based composites.

5 RESULTS AND DISCUSSION

It is possible to divide the experimental study into two phases. In the former two materials completely different for mechanical properties (MAT1 and MAT2) were tested in order to evaluate their progressive fatigue damage, and a conventional fatigue test has been carried out to validate the data obtained. In the latter three materials quite similar for composition and mechanical properties

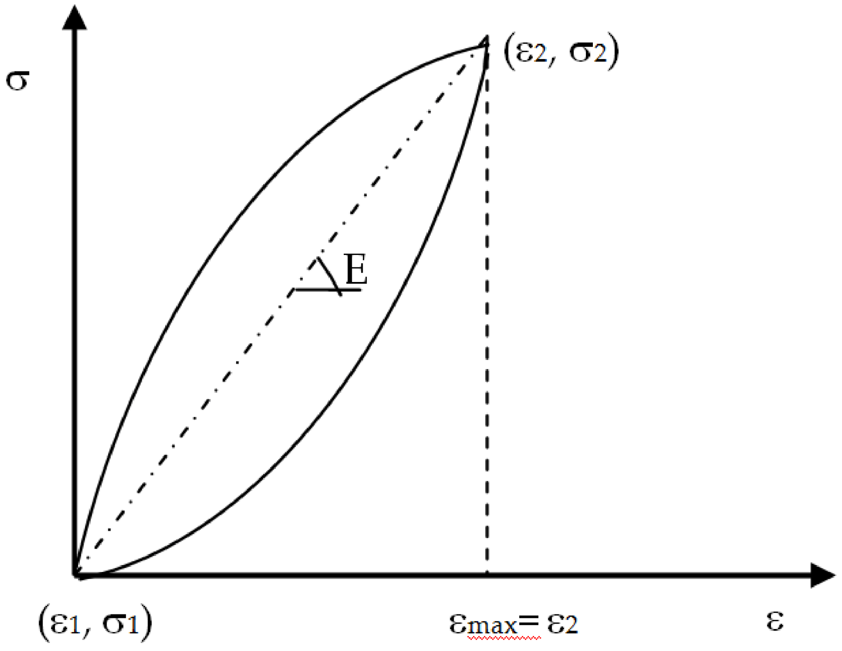

Figure 2: Secant Modulus E as slope of the hysteresis loop.

(S1, S2, S3) have been considered. The purpose of this phase is to demonstrate the usefulness of the accelerated fatigue method for industrial applications. In fact in these cases a fast method able to compare, for example, the fatigue behaviour of two or more matrices furnished by different suppliers or the consequence of a little variation in the formulation of a composite prepared with the aim of cost reduction, could be fundamental.

Tab. 3 resumes the maximum stress levels and the total number of cycles tested with the accelerated fatigue tests for MAT1 and MAT2, while the samples S1, S2 and S3 have been all tested at 19.5 MPa for 4000 cycles.

From Fig. 3 to Fig. 5 the damage parameters obtained for MAT1 from the tests carried out at 17,5MPa, 18,2MPa, 19,5MPa and 23MPa for MAT1 are reported. For the two lower stress levels the material doesn't show important damage accumulation, while the tests at higher stress levels emphasize the evolution of the parameters towards breaking conditions.

It is important to notice that for MAT1 we have reached the break of the sample only for a stress level higher than the yield stress.

For the PP-$CaCO_3$-GF composite (MAT2) the most significant results were the ones at 31MPa, 36MPa and 41MPa that correspond to the 60, 70 and 80% of the tensile stress at break respectively. Like MAT1, the first two tests don't show an appreciable fatigue damage, in fact the relative storage modulus remains substantially constant during the 4000 cycles. The test at 41MPa shows an evident evolution of the damage parameters and in this case the breaking of the sample is reached. Plots in Fig. 6-8, the three damage parameters versus the number of cycles for MAT2 are represented.

The conventional fatigue tests were carried out with the purpose of validating the fatigue data obtained from the accelerated fatigue tests.

Before comparing these two methods it is important to take into consideration the differences of the two cases. First of all the stress waveforms are different: in the conventional fatigue test, the load is applied with a sinusoidal waveform, while in the accelerate one it has a triangular waveform. Moreover a conventional fatigue test is carried out at constant frequency (3Hz for MAT1 and MAT2) [2, 7], while for the accelerated fatigue method it was not. Notwithstanding it, the conventional tests have substantially confirmed the results obtained with the accelerate fatigue tests. In Fig. 9 and 10 a comparison between the relative secant modulus and the dissipated strain energy obtained from the two different fatigue tests carried out at 41MPa on MAT2 samples are reported.

The accelerated fatigue test is above all useful to compare similar materials at the same maximum stress level. The main requirement is that the materials have similar matrix, and similar filler type and content.

S1, S2 and S3 are three similar composites as results from the standard mechanical tests (Tab. 2), but if they are tested with the accelerated test some important differences are emphasized.

The test at 18,2MPa has shown that there are some differences in the fatigue properties of these materials. The differences increase as the stress level increases. At 19,5MPa S1 and S3 have similar damage parameters path, while S2 denotes a higher damaging rate.

In Fig. 11-13, the three damage parameters versus the number of cycles for the three composites tested at 19,5MPa are represented.

6 CONCLUSIONS

In the present work a new method for the analysis of fatigue damaging of thermoplastic composite materials has been developed. The accelerated fatigue test is useful to obtain in short time information about the tendency of a material to damage (two-three hours instead of one week or more requested by conventional fatigue tests) by using a standard universal testing machine. We have demonstrated that the data obtained with this method are in agreement with those of the conventional fatigue one, although some fundamental differences in the set up for the two methods exist. The principal limitation of the method is given by the fact that high stress levels are needed in order to highlight in

	% of tensile yield stress	**σ_{max} [MPa]**	**N_{TOT}**
MAT1 (PP-$CaCO_3$)	80	17,5	4000
	83	18,2	4000
	89	19,5	4000
	105	23	Break @ 695
MAT2 (PP-$CaCO_3$-GF)	60$^{(a)}$	31	4000
	70$^{(a)}$	36	4000
	80$^{(a)}$	41	Break @ 1070

Table 3: Summary of the Accelerated Fatigue Test. (a) % of tensile stress at break (max. stress).

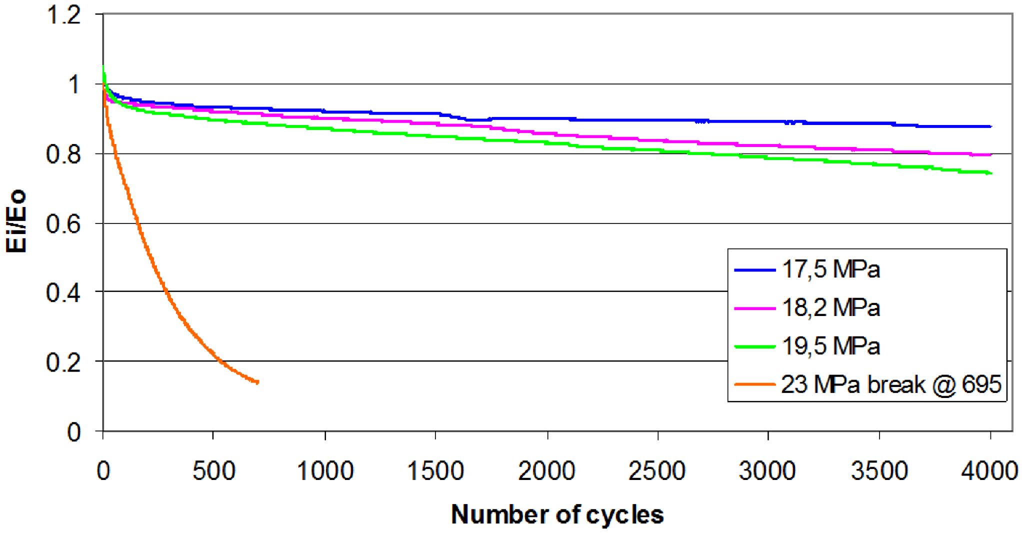

Figure 3: Relative secant modulus for MAT1.

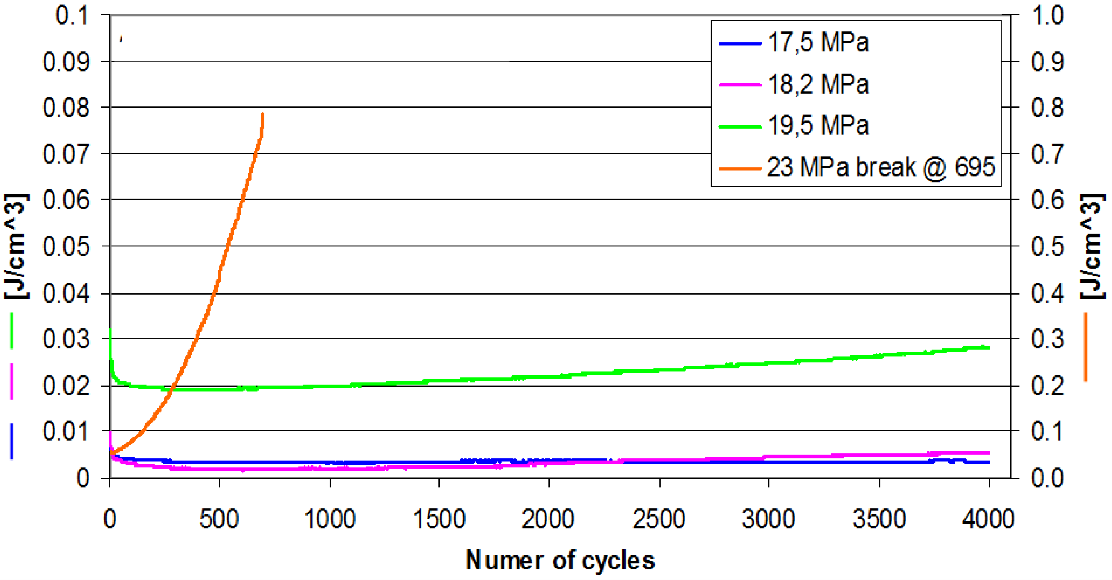

Figure 4: Dissipated strain energy for MAT1.

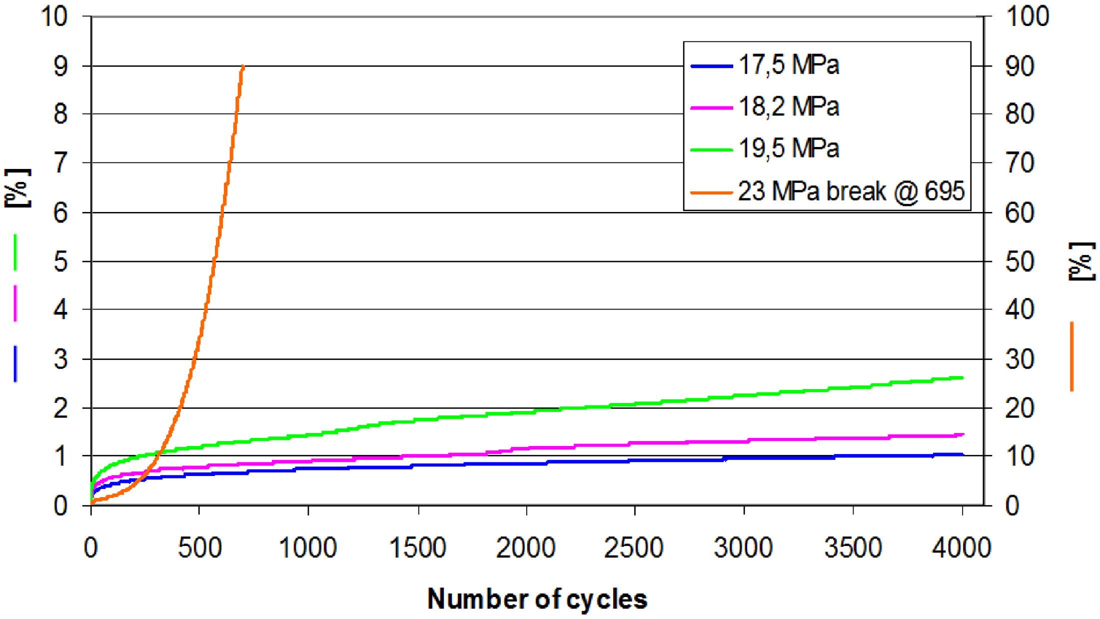

Figure 5: Residual strain for MAT1.

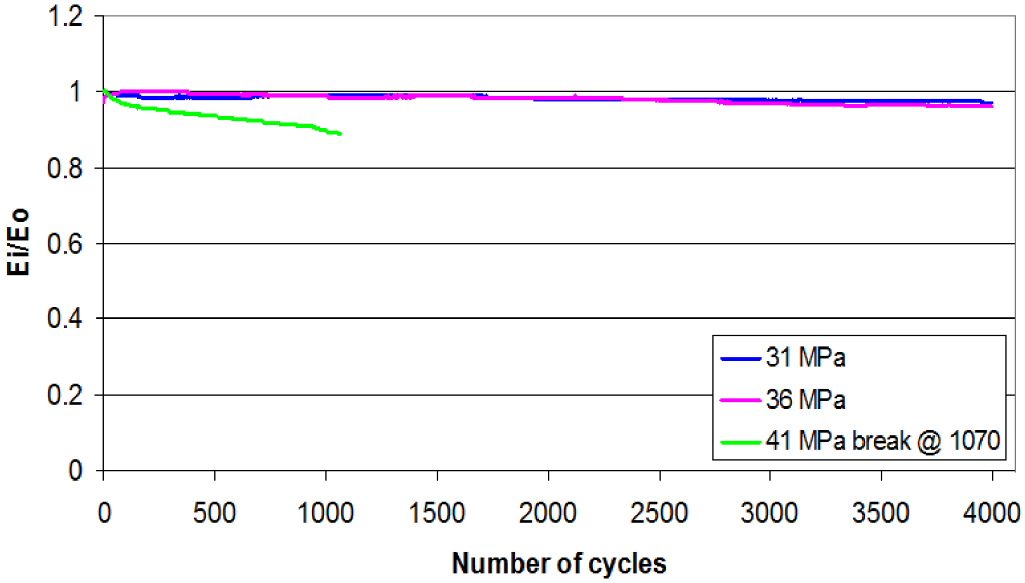

Figure 6: Relative secant modulus for MAT2.

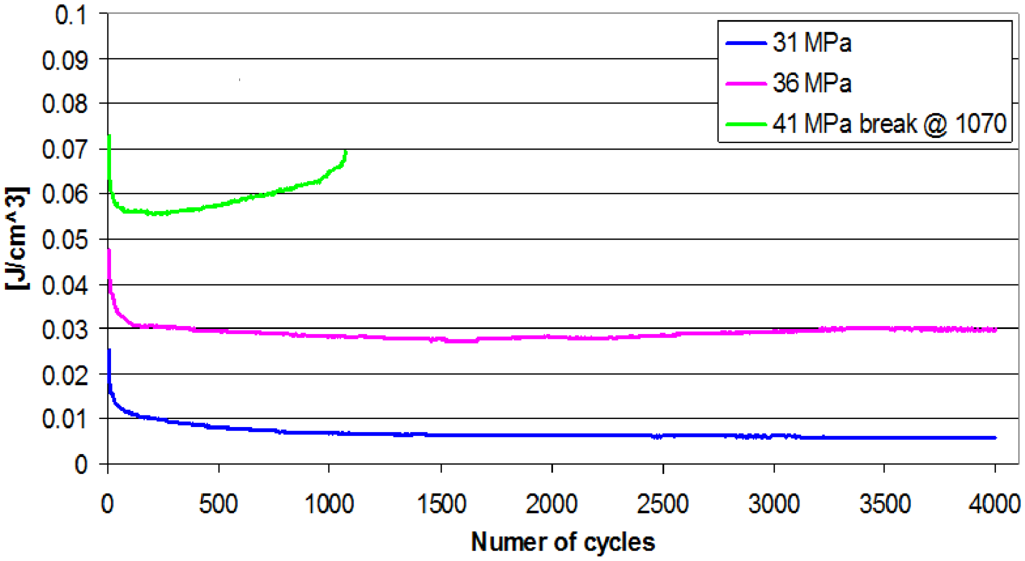

Figure 7: Dissipated strain energy for MAT2.

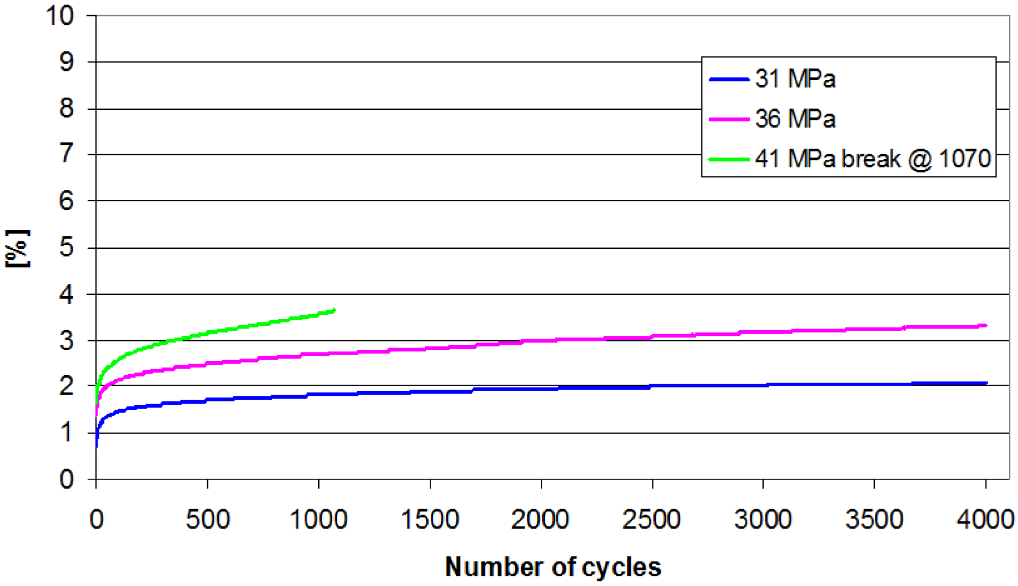

Figure 8: Residual deformation for MAT2.

short time different trends for the damage parameters considered and compare endurance performances of various materials accordingly.

7 REFERENCES

[1] N. W. M. Bishop, F. Sherratt, Finite Element Based Fatigue Calculations, NAFEMS Ltd (2000).

[2] Fatigue of Materials, 2nd ed., S. Suresh, Cambridge Univ. Press. (1998).

[3] Y.L. Lee, J. Pan, R. Hathaway, M. Barkey, Fatigue Testing and Anslysis – Theory and Practise, Elsevier (2005).

[4] A.J. Jones, A.J. Lesser, Journal of Polymer Science. Part B: Polymer Physics, 36 (1998) 2751-2760.

[5] S. Rangaray, Durability Assessment and Modelling (18) of Wood-Thermoplastic Composites,Master Thesis ,Washington State University (1999).

[6] A.E. Slaugther, Design and Fatigue of a Structural Wood-Plastic Composite, Master Thesis, Washington State University (2004).

[7] Y. Zhou, P.K. Mallick, Polymer Engineering and Science, 45-4 (2005) 510-516.

[8] Yuanxin Zhou, P.K. Mallick , Polymer Engineering and Science, 45-6 (2005) 755 – 763.

[9] G. Tao, Z. Xia, Polymer Engineering and Science, 47 (2007) 780-788.

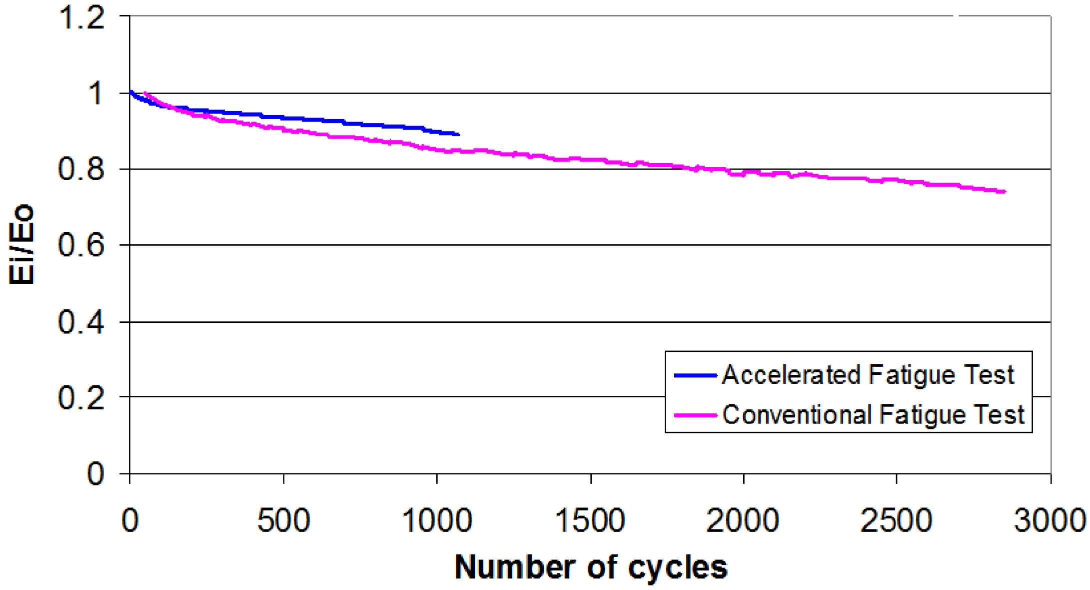

Figure 9: Comparison between conventional and accelerated fatigue test on MAT2.

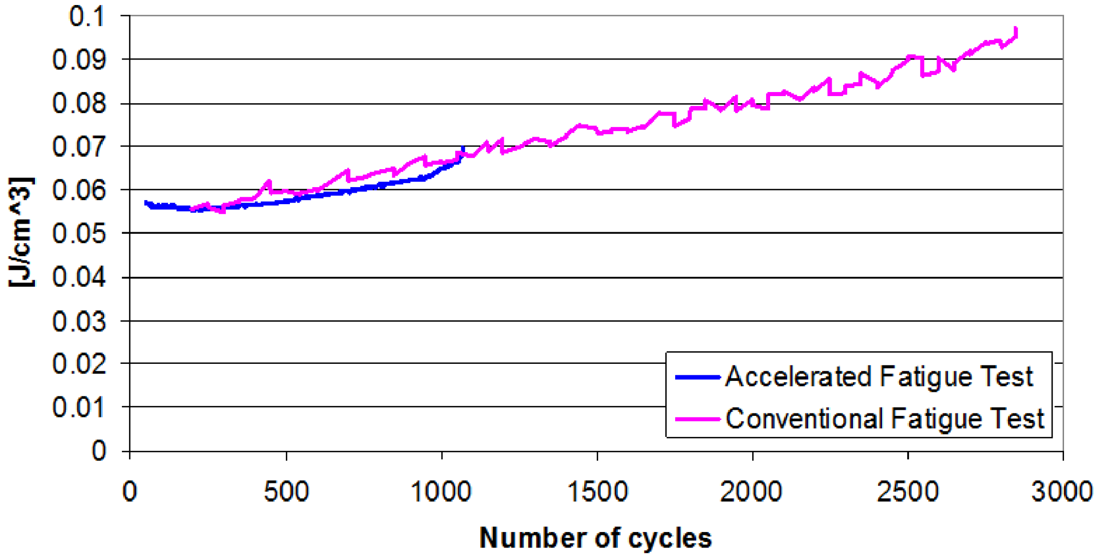

Figure 10: comparison between conventional and accelerated fatigue test on MAT2.

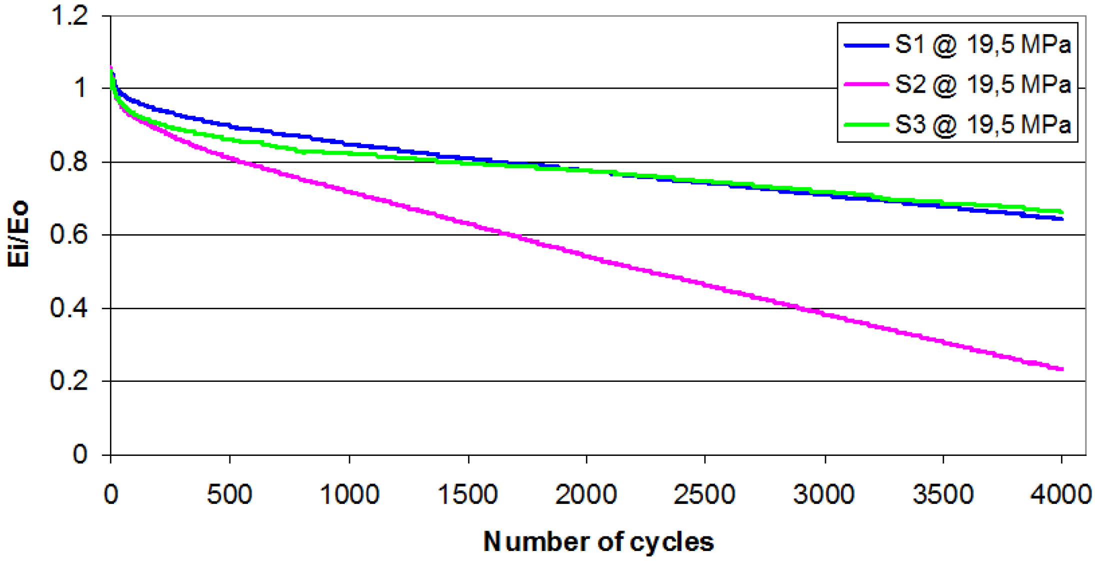

Figure 11: Comparison between S1, S2 and S3 at 19.5 MPa.

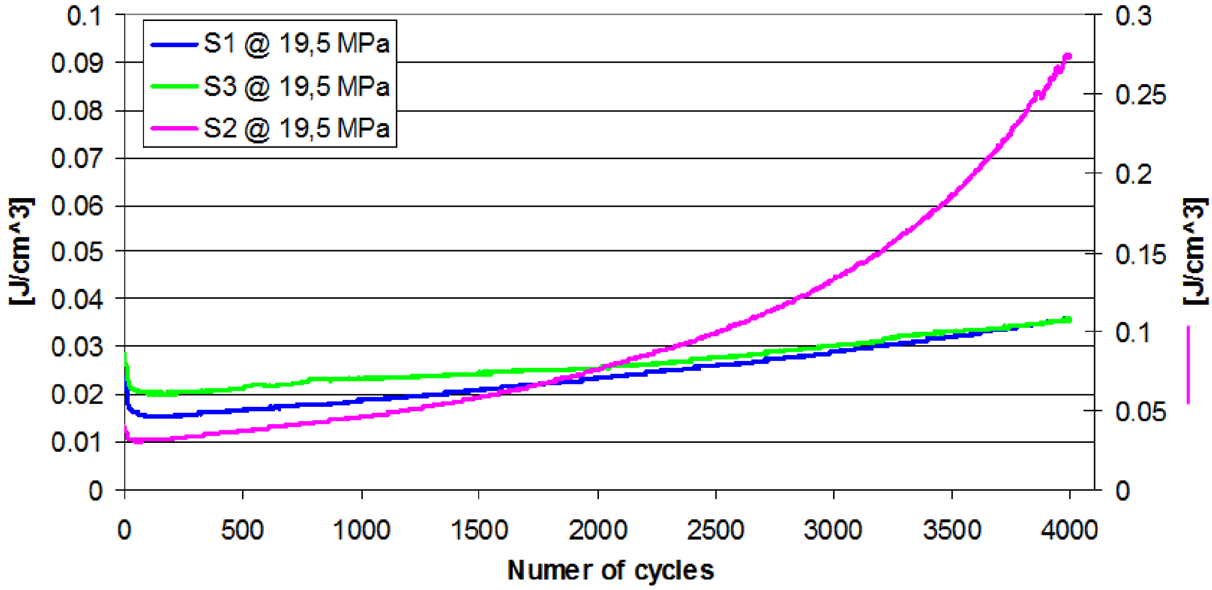

Figure 12: Comparison between S1, S2 and S3 at 19.5 MPa.

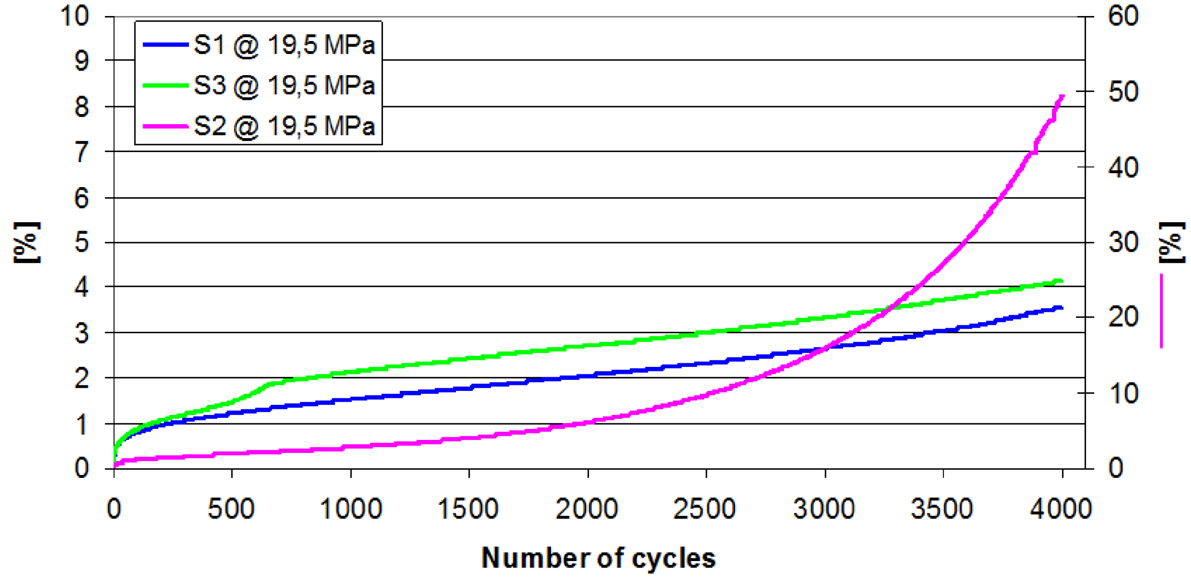

Figure 13: Comparison between S1, S2 and S3 at 19.5 MPa.