UDC 539.4

Experimental Analysis of the Influence of Structural Parameters on the Behavior of Glass-Fiber Reinforced Polypropylene Composites

L. Bounouas[a,b], **S. Benmedakhene**[a], **A. Laksimi**[a], **F. Neumann**[a], **A. Imad**[c], **and Z. Azari**[d]

[a] Université de Technologie de Compiègne (UTC), Compiègne, France

[b] Université Hassan II, Faculté des Sciences Ben M'sick-sidi othman, Casablanca, Maroc

[c] HEI Laboratoire d'Études des Structures, Lille, France

[d] Faculté de Mathématique et d'Informatique, Université de Picardie, Amiens, France

УДК 539.4

Экспериментальное исследование влияния параметров структуры на свойства стеклопластика на основе полипропилена

Л. Бунуа[а,б], **С. Бенмедахен**[а], **А. Лаксими**[а], **Ф. Ньюман**[а], **А. Имад**[в], **З. Азари**[г]

[а] Технологический университет Компьеня, Компьень, Франция

[б] Университет Хасана II, Факультет естественных наук, Касабланка, Марокко

[в] Лаборатория изучения структуры, Лилль, Франция

[г] Университет Пикарди, Факультет математики и информатики, Амьен, Франция

В композитном материале, армированном случайно расположенными короткими волокнами, повреждения при механическом нагружении возникают в результате действия разных элементарных механизмов, таких как микрорастрескивание матрицы, выдергивание волокон, разрушение межфазной границы волокно–матрица, разрушение волокон. Эти повреждения оказывают большое влияние на макроскопические свойства композитного материала. Для получения приемлемых механических характеристик композитного материала важно оптимизировать концентрацию волокон, а также качество межфазной границы волокно–матрица, которые оказывают непосредственное влияние на перечисленные выше повреждения. Главной целью статьи является определение влияния структурных параметров на эволюцию повреждаемости двух типов стеклопластиков на основе полипропилена. Одновременно с классическим подходом к механической теории повреждаемости, основанной на испытаниях на растяжение с периодической разгрузкой, использование метода акустической эмиссии дает возможность изучать в реальном времени природу и важность механизмов повреждений в процессе нагружения. В дополнение к этому фрактографический анализ позволяет подтвердить различные предпосылки и заключения настоящего исследования.

Ключевые слова: композит, полипропилен, стеклопластик, акустическая эмиссия, повреждение.

Introduction. Composite materials offer several advantages over the conventional ones. In fact, they have higher rigidity and specific strength, higher oxidation resistance and improved flexibility in application. However, they often involve long manufacturing cycles that result in high fabrication costs. Moreover, their high sensitivity to damage is often inconvenient for their structural applications. A solution to overcome this sensitivity to damage is the use of thermoplastic-matrix composites. One of the thermoplastics most often used as a matrix in this new type of composites is polypropylene (PP), which is prepared by a continuous process of injection moulding. In fact, glass-fiber reinforced polypropylene offers very interesting mechanical and environmental performances. The use of these materials in automobile industry is in expanding continuously. However, their application is still limited due to the lack of data concerning their behavior and damage resistance. Thus, much research work has to be done in this field in order to optimize and extend their application.

Damage initiation in composites may often be initiated at low-energy interactions during service life, such as drop of tools or parts, impacts during transport and/or due to environmental agents (such as hail–stones). This has been considered a serious problem encountered during the use of composite materials and consequently it has attracted increasing attention in recent years, assuming that damage results in a more or less appreciable reduction in mechanical properties depending on the loading conditions. As for conventional composites, a complete study is required in order to understand damage resistance and behavior of thermoplastic composite materials.

The literature presents several studies conducted in this area. In fact, many studies are concerned mainly with the influence of structural parameters such as fibers, volume ratio and the quality of fiber/matrix interface. However, the majority of the authors limit their investigations to the theory of mechanical damage. In this study, an effort has been made to implement the classical approach of the theory of mechanical damage through the load–unload tensile tests combined with a nondestructive method to follow the material behavior in real time.

The latter is an acoustic emission method, which allows one to obtain precise information on the chronology and damage intensity during loading. In addition, fractographic analysis is used to confirm different hypotheses and conclusions.

Thus, the aim of this work is the identification of damages in glass-fiber/polypropylene composites and determination of the influence of different structural parameters on damage behavior of composites. Both acoustic emission and microscopic observations have been used for the identification of damage mechanisms.

Experimental Procedure.

Materials. The material of the study is a polypropylene composite matrix reinforced with short glass fibers approximately 4 mm in length and 10 μm in diameter. The injection-moulded samples used in this study were prepared using a computer-controlled Fast-Inject type injection machine. The operation conditions such as injection speed, injection pressure, injection temperature and mould temperature regulation are set by the operator and are chosen according to the ISO R527 standard. Figure 1 summarizes the materials and the most essential parameters considered in this study.

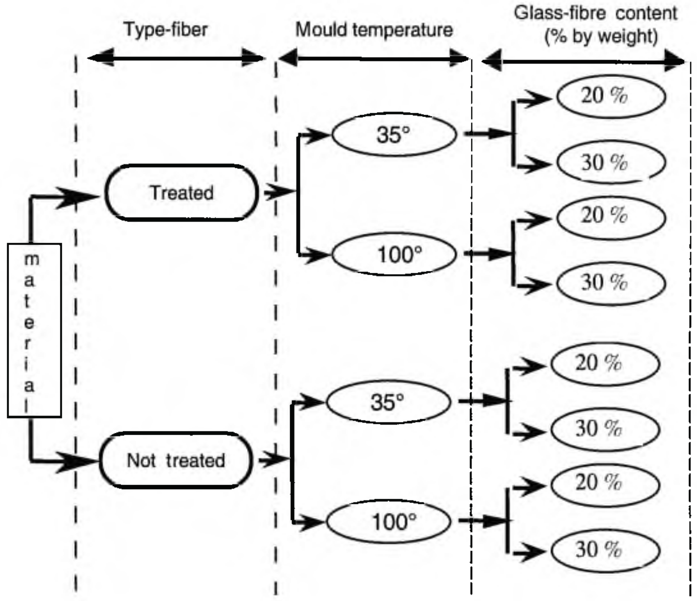
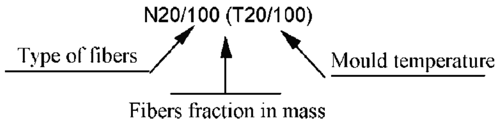


Fig. 1. Presentation of various materials studied.

In this study, the materials used will be designated as follows:



T and *N* stand for treated and untreated glass fibers, respectively. For example, T20/100 indicates a PP composite reinforced by 20% of treated glass fibers injected at 100°C and N30/35 indicates a PP composite reinforced by 30% of untreated glass fibers injected at 35°C.

Tensile Tests. Tensile tests are carried out on INSTRON 1186 static type machine. In addition to the acoustic emission signals, the acquisition system collects the data on the load applied to the specimen and the information from the strain gauges.

The sample is instrumented with bidirectional gauges that allows the measurement of longitudinal and transverse strains in the material (Fig. 2). After testing, scanning electron microscope (SEM) observations are carried out on fractured samples.

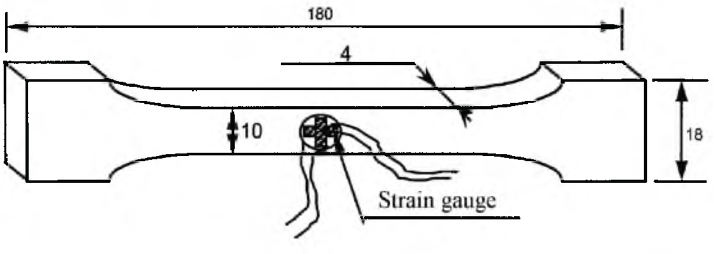


Fig. 2. Tensile specimen (dimensions in mm).

Tensile Load-Unload Test. The load-unload tensile tests are carried out on the same type of samples as used under simple tension. In this test, two levels of unloading are chosen:

– partial unloading: in each sequence of loading, unloading is applied up to 30% of the previously reached load;

– complete unloading: in each loading sequence unloading is complete, i.e., the load is equal to zero.

Damage is quantified by the measurements of the elastic modulus using the following relationship: $D = 1 - (\tilde{E} / E)$, where D is the damage parameter, E is Young's modulus of an undamaged material, and $\tilde{E}$ is Young's modulus of a damaged material.

The elastic parameters are measured within the linear part of the loading phase. This phase generally shows a total absence of any reaction of acoustic emission.

Acoustic Emission. Different studies have been accomplished using acoustic emission as nondestructive method to follow damage in composite materials. They involve different composites with different types of matrix and reinforcements [1–7]. They all suggest that various damage mechanisms encountered in the composites may be classified according to different ranges of acoustic emission amplitudes (Fig. 3).

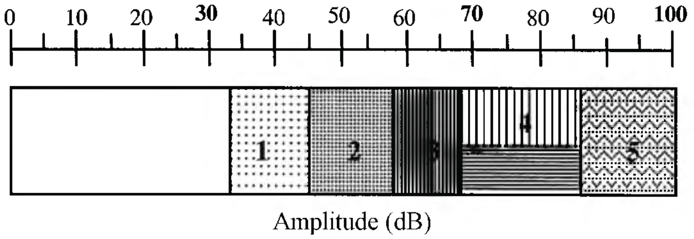


Fig. 3. Typical amplitude distribution and areas definition (*1* – matrix microcracking; *2* – matrix/matrix friction; *3* – interface decohesion; *4* – fiber/matrix friction and fibre debonding; *5* – fiber breakage and bundle shattering).

Thus, analysis of the amplitudes of acoustic emission signals generated during the occurrence of different damage mechanisms facilitates the damage growth detection during testing.

Processing of acoustic activity and data acquisition have been carried out by means of an acoustic emission system (DUNEGANE/ENDEVCO 3000) with the following physical characteristics:

preamplifier gain: 39 dB;

preamplifier filters: 450 B and 190 B;

AE threshold: selectable, fixed at 30 dB for the present study;

the range of amplitude channels is from 0 to 100 dB;

channel width (resolution): 1 dB;

dead time: 1 ms (emission by bursts).

Acoustic emission signals are detected using a piezoelectric transducer (PAC-MICROPHONE 80) and have a large range of frequencies from 200 kHz to 1 MHz.

The AE investigation method is combined with microscopic observations using a scanning electron microscope. This contributes to a better understanding of the occurrence and evolution of the basic damage mechanisms.

Results and Discussion.

Tensile Test. The aim of tensile tests is to determine the influence of structural and other parameters on the elastic behavior, damage and failure of the materials.

Influence of the Interface. The following figures illustrate the difference in the mechanical behavior between two glass-fiber reinforced polypropylene materials with different interface qualities.

The stress-strain curve belonging to T20/100 material shows a better mechanical performance. This is the result of improved interface quality due to the addition of oiling. The stress-strain curve of N20/100 shows low mechanical characteristics resulting from untreated fibers. In the case of T20/100 material (Fig. 4), three significant phases are observed on the curve:

– The first linear elastic phase (phase *A*) without acoustic emission.

– The second non-linear phase (phase *B*) related to the initiation of damage mechanisms. During this phase, the first events of acoustic emission generated by the material are observed.

– The third phase (phase *C*) characterized by a change of slope of the stress-strain curve. This change is accompanied by a significant acoustic emission. An exponential evolution is observed on the last portion of the acoustic emission cumulative curve indicating the presence of large damage leading to final failure of the material.

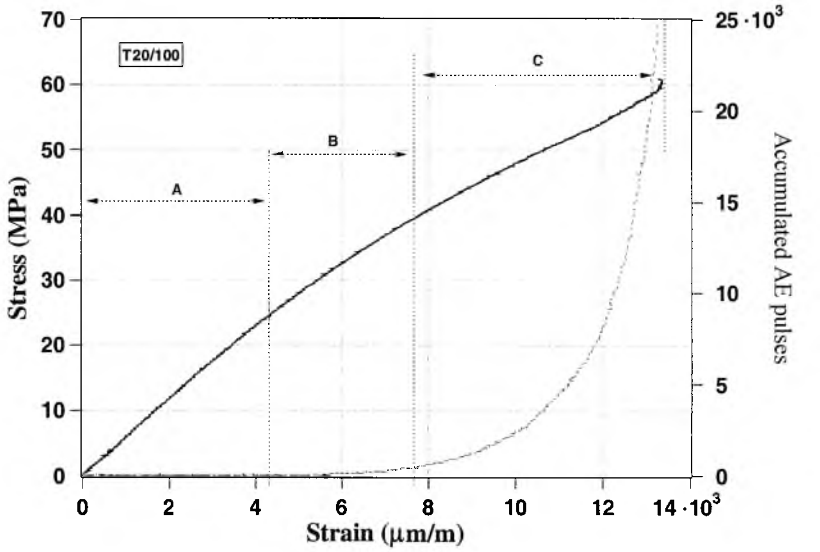


Fig. 4. Stress and acoustic emission versus strain (T20/100).

In the case of N20/100 material (Fig. 5), the behavior is characterized by three phases different from those of T20/100:

– The first linear elastic phase (phase *A*), which allows the measurement of the elastic properties of the material without acoustic emission.

Table 1

Results of Tensile Testing of Composite Materials with Poor and Good Quality Interfaces

	Mould $T = 100^\circ$C				Mould $T = 35^\circ$C	
Content of glass fibers	20%		30%		20%	
Fiber type	treated	untreated	treated	untreated	treated	untreated
E (GPa)	5.15 (0.30)	5.43 (0.49)	6.81 (0.41)	6.88 (0.64)	5.58 (0.15)	5.00 (0.23)
σ_r (MPa)	62.87 (6.73)	33.09 (1.90)	70.46 (3.67)	32.70 (1.07)	73.35 (4.23)	32.83 (1.01)
ε_r (%)	1.56 (0.36)	1.33 (0.25)	1.53 (0.25)	1.20 (0.60)	1.67 (0.63)	2.23 (0.25)

Note. Here E is Young's modulus, σ_r is the failure stress, and ε_r is the failure strain (standard deviations are given in parentheses).

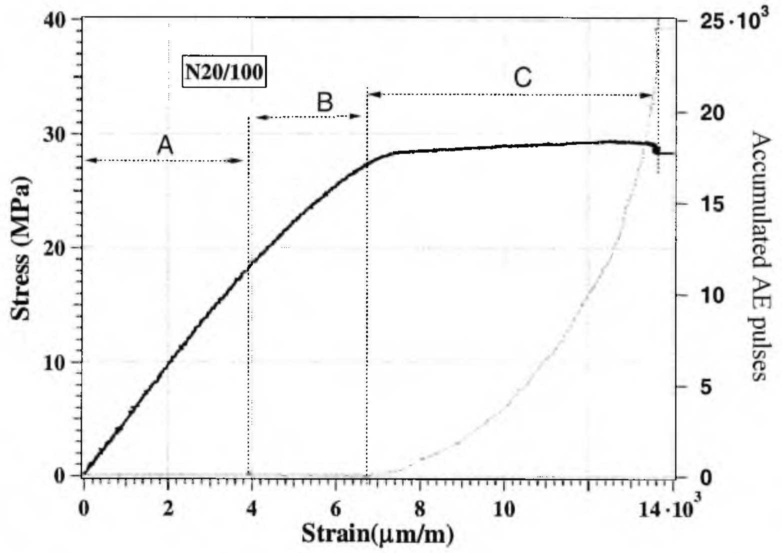


Fig. 5. Stress and acoustic emission versus strain (N20/100).

– The second non-linear phase (phase *B*), wherein no acoustic emission is detected. This phase indicates plastic deformation without damages.

– The third phase (phase *C*), wherein the stress-strain curve reaches a creep plateau and deformation increases under constant stress. This phase is characterized by a high acoustic emission activity. The creep plateau indicates the absence of load transfer between the matrix and glass fibers beyond the elastic field. Table 1 presents comparison between the elastic constant values and failure characteristics of the two materials.

These results show that oiling of fibers allows us to double the failure stress of the composite materials studied. However, it has no effect on the elastic modulus. It is well established in both cases (with oiled and not oiled fibers) that acoustic emission allows the initiation and evolution of damages taking place after the elastic phase to be detected.

Influence of Fiber Content Increase. The fraction of fibers has an appreciable influence on the mechanical properties of the composite and particularly on the elastic modulus. In fact, the increase of fibers ratio from 20% (in weight) to 30% results in an increase in elastic modulus of 25% in the case of treated fibers and 21% in the case of untreated fibers.

Damage Analysis. Through the study of the damage mechanisms, a continuous correlation is established between the acoustic emission and microscopic observations.

The distribution of events according to their amplitudes [8–13] has an advantage. It allows the classification of damage mechanisms according to a given range of amplitudes. This method also allows a chronological detection of damages during loading.

Figures 6 and 7 present amplitude distributions during tensile loading in the case of both oiled and not oiled fiber/matrix interfaces.

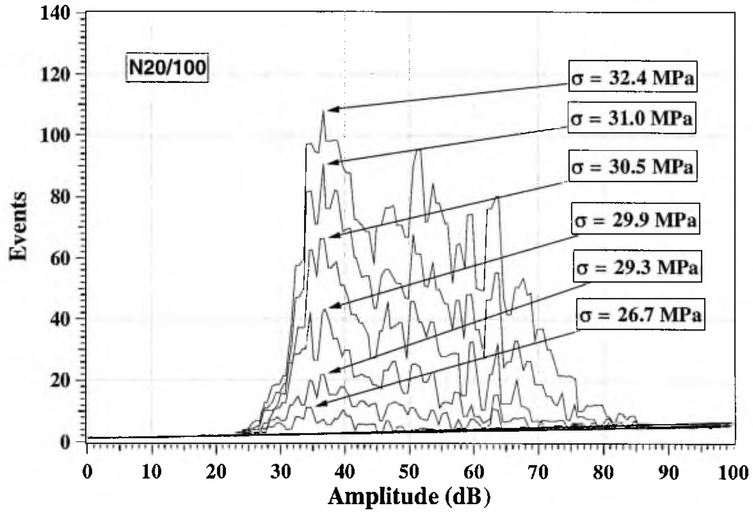


Fig. 6. Amplitude distribution for N20/100 for various stress levels.

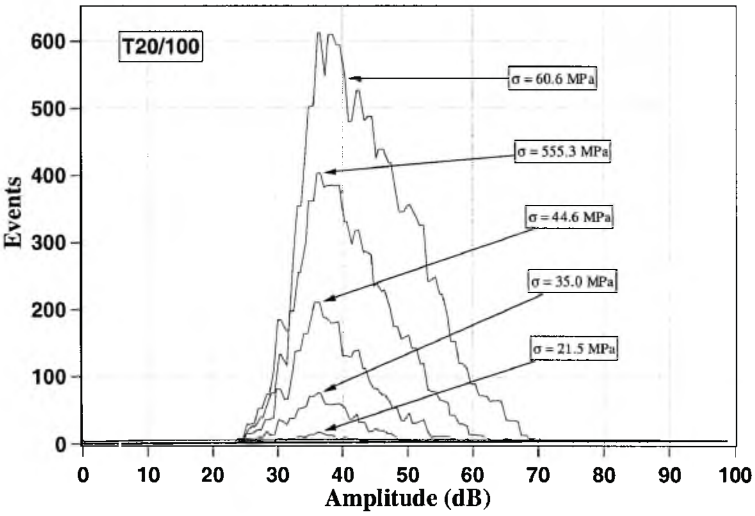


Fig. 7. Amplitude distribution for T20/100 for various stress levels.

The evolution of amplitude distributions shows that the total number of recorded events obtained in the case of treated fibers is more significant than that obtained in the case of untreated ones. The results also show that for both oiled and not oiled fibers the number of events decreases when the fiber content increases. Figure 6 shows that for untreated fibers, the amplitude distribution is very wide. This is an evidence of the presence of several types of damages such as matrix cracking, microscopic crack coalescence, interface failures, matrix/fiber friction and fiber failure. This is especially the case where the fiber content is low.

In the case of treated fibers, the amplitude distribution is narrow (Fig. 7). The evolution of damages is less important between 60 and 80 dB, which indicates that failure of the fiber/matrix interface and fiber pullout are not important. This is the result of good matrix/fiber adherence that, consequently, ensures a better mechanical behavior.

Microscopic observations (Figs. 8–11) show the presence of matrix microcracking in the T20/100 and N20/100 material. Figure 9 confirms the good fiber/matrix adhesion for the same material.

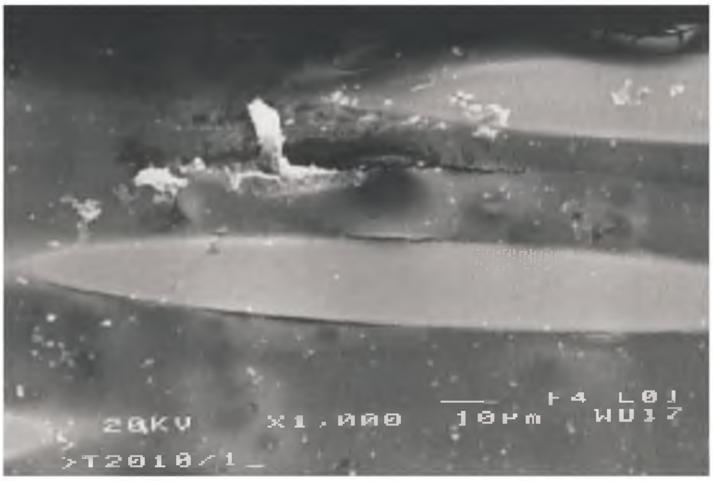


Fig. 8. Matrix microcracking and failure of fiber/matrix interface (T20/100).

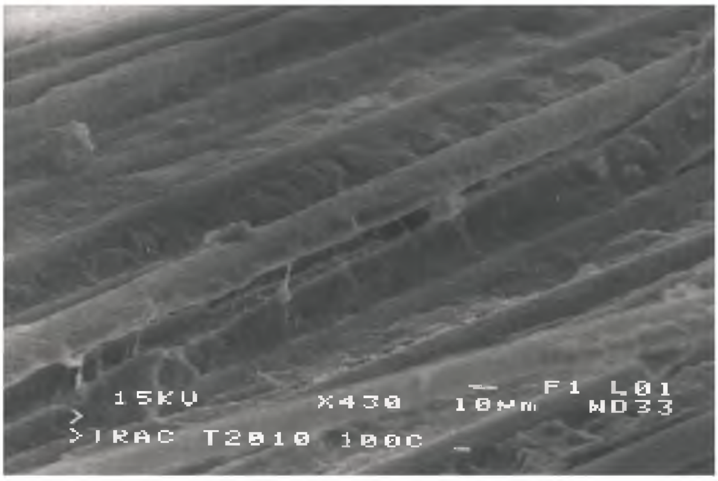


Fig. 9. SEM fractograph of the T20/100 material.

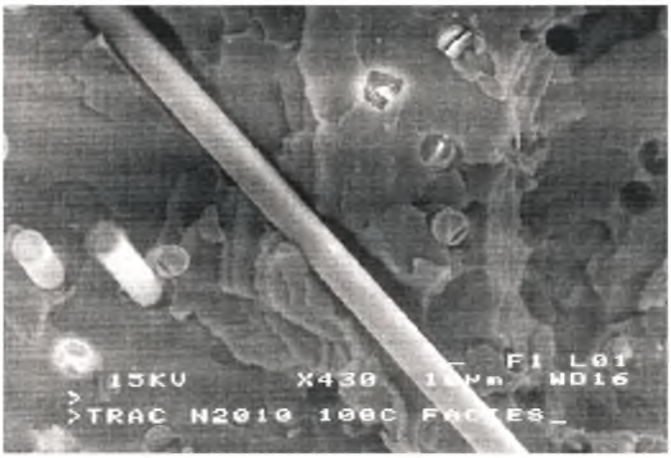


Fig. 10. SEM fractograph of the N20/100 material.

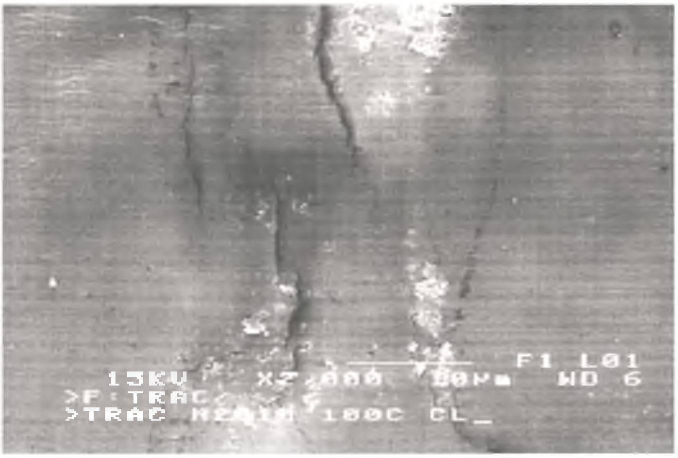


Fig. 11. Matrix microcracking (N20/100 material).

Tensile Load–Unload. Quantitative estimation of the damage resulting from load–unload tests is based on a comparison with the initial elastic characteristics of an undamaged material. This type of test allows the determination and quantification of the damage that cannot be measured in a simple tensile test.

The acoustic emission technique is used in order to show that damages are not caused by unloading to which the material is subjected.

Isotropic Damage. Damage is defined as a gradual deterioration phenomenon leading to failure of the loaded material. Physically it results in the appearance of microscopic defects within the material. These microdefects influence the behavior of a specimen, particularly, in terms of the degradation of macroscopic properties such as Young's modulus.

The fact that the rigidity of the material is affected during the load–unload tests makes the measurement of the elastic modulus essential in order to be able to use a scalar D, which will permit us to quantify the damage mechanics. This assumption of isotropy is confirmed in our case by the constancy of Poisson's ratio (as shown by the graphs in Fig. 12).

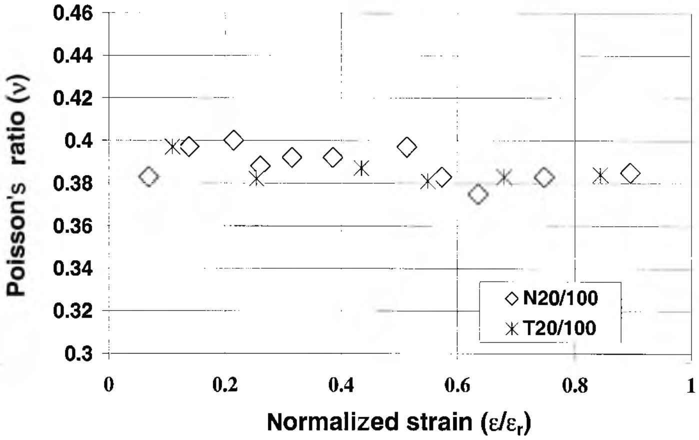


Fig. 12. Evolution of Poisson's ratio ν (oiled and not oiled samples).

The introduction of a variable damage parameter representing the surface density of discontinuities is made using the concept of effective strain, i.e., the strain, which takes into account the load-sustaining part of the specimen section area [14, 15]. In the presence of isotropic damage of magnitude D, the load-sustaining part of the specimen section area then becomes: in the case of simple tension, the ratio between the applied force and the section of the representative volume $\sigma = F / S$ gives the usual strain that satisfies the equilibrium conditions, therefore

$$\tilde{S} = S - S_D = S(1 - D).$$

Here S_D is the damaged section.

The effective strain is expressed by

$$\tilde{\sigma} = \sigma \frac{S}{\tilde{S}} \Rightarrow \tilde{\sigma} = \frac{\sigma}{1 - D}.$$

In this formula, D is the damage parameter defined by

$$D = 1 - \frac{\tilde{E}}{E},$$

where $\tilde{E}$ is Young's modulus of the damaged material, E is Young's modulus of undamaged material.

The limits of this scalar can be defined as follows: when $D = 0$, the material is undamaged and when $D = 1$, the material is fractured.

Evolution of the Damage Parameter D. While following the load–unload tensile mechanical tests, the damage phenomenon is characterized by means of an indirect technique. The latter consists in the measurement of the Young's modulus of the damaged material from the stress-strain curves. The ratio between the Young's moduli of the damaged and undamaged materials quantifies the extent of damage in the material at different stages of loading.

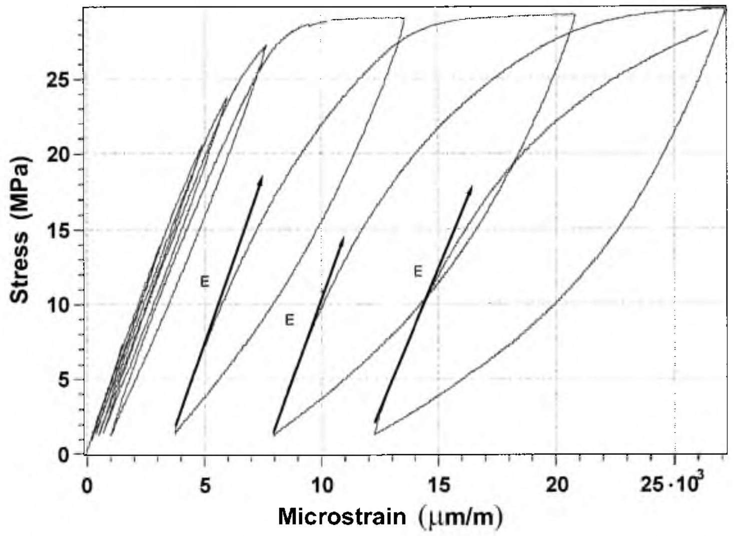


Fig. 13. Measurement of Young's modulus (E) in the damaged material in the case of complete unloading.

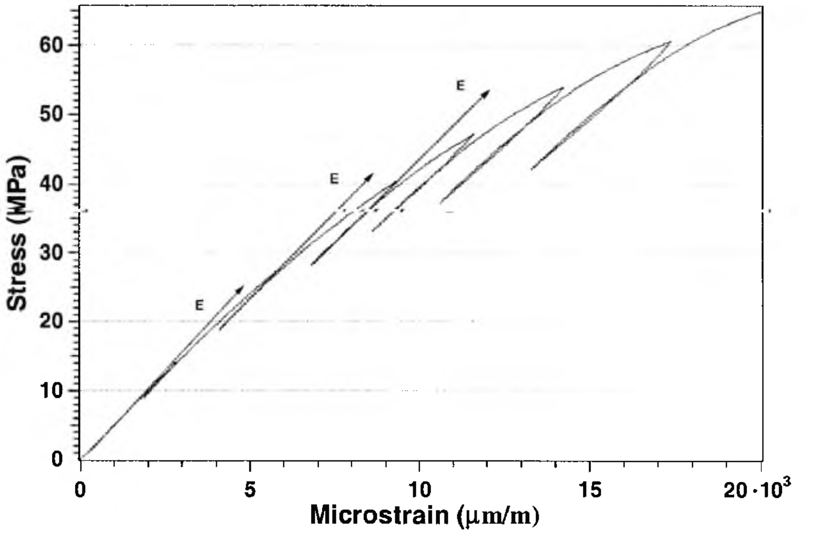


Fig. 14. Measurement of Young's modulus (E) in the damaged material in the case of unloading to 30% of the maximum load.

Figures 13 and 14 show the way of measuring the Young's modulus in the damaged material during the steps of reloading.

Different situations are obtained depending on the mode of unloading and on the interface quality. In the case of complete unloading (Fig. 15), the loss in rigidity is rather low for all types of materials. This case is equivalent to the situation, where the test was restarted from the beginning. This is related to the total relaxation of the material. In this case, when reloading, the measure of the initial elastic modulus is related to a coupling of elasticity with damage. This can be explained by the state of the majority of microcracks, which are rather closed, and the material behaves almost as an undamaged one.

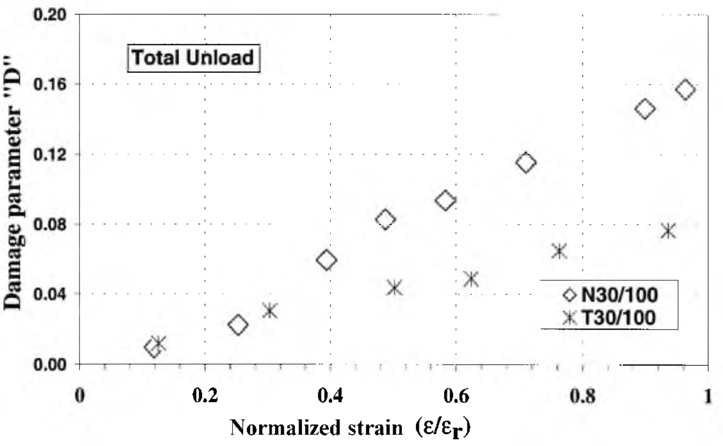


Fig. 15. Damage parameter versus normalized strain for materials N30/100 and T30/100 in the case of complete unloading.

Figure 16 shows that when unloading goes 30% below the maximum load, the stress field during unloading remains high and maintains the material in the state of significant damage. The graph in Fig. 16 illustrates this phenomenon.

According to the quality of the fiber/matrix interface, we can observe two types of evolution of the damage parameter D. Thus, as shown in Fig. 16, in the case of a good-quality interface, the damage grows in a regular way.

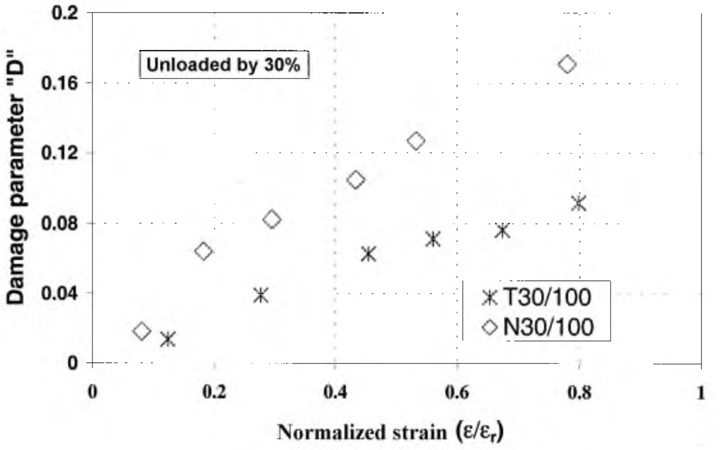


Fig. 16. Damage parameter versus normalized strain for materials N30/100 and T30/100 in the case of partial unloading.

In the case of an untreated material, an irregular evolution of damage is observed: first, a linear evolution of the behavior can be noted coupled with the damage where the elastic modulus does not change significantly. In the second stage, when the creep plateau is reached, the deformation grows at a constant stress followed by a rapid increase in damage. The Kaiser effect is valid for metallic materials, but for the composite structures it is replaced by the Felicity effect. It consists in that the acoustic emission will start again before the preceding maximum load has been reached. The Felicity ratio is equal to the ratio between the stress corresponding to the acoustic emission and the stress reached before unloading (Felicity ratio "*R*" is equal to P1/P2) (Fig. 17).

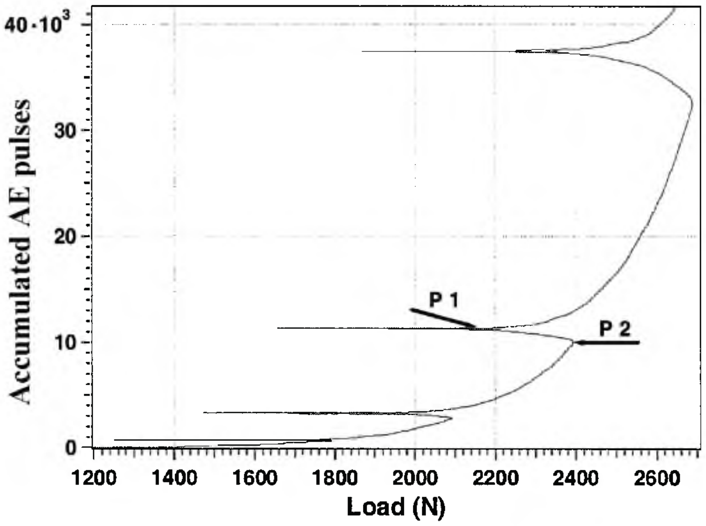


Fig 17. Measure of the Felicity ratio.

In general, the Felicity ratio decreases and tends to zero. Indeed, when this ratio is equal to 1, the material is undamaged. When the Felicity ratio lies between zero and 1, the material is damaged and when the ratio is equal to zero, the material is fractured.

The graph below shows the evolution of the Felicity ratio according to the quality of the fiber-matrix interface (Fig. 18).

Note that in the case of an untreated sample, the ratio decreases sharply and its values approach zero showing the weakness of the interface.

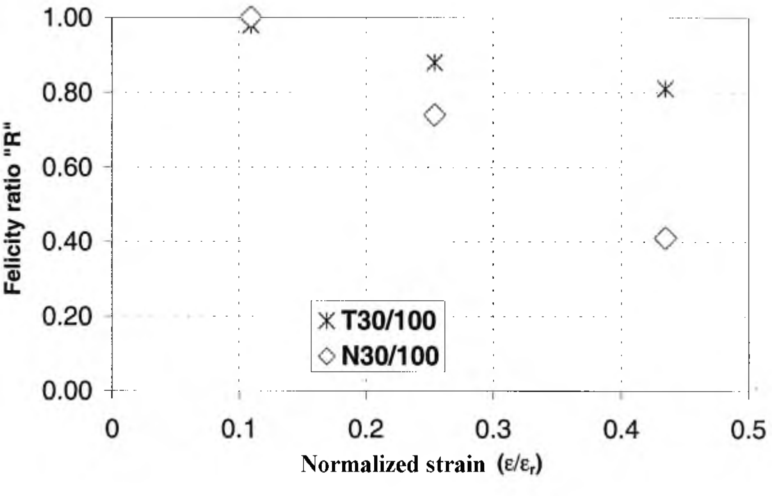


Fig. 18. Evolution of the Felicity ratio with the damage parameter.

On the other hand, in the case of a treated sample, the Felicity ratio decreases slowly and is much higher than in the case of the weak interface. This indicates that the material undergoes weak damage owing to a good fiber/matrix interface.

Conclusions. An experimental study of damage of thermoplastic composites was carried out according to two different approaches.

The first is a qualitative approach based on the acoustic emission technique and scanning electron microscopy. It allowed the study of the global behavior of the material. The use of the cumulated acoustic emission and the amplitude distribution method allowed the identification and classification of various damages according to the chronology of their occurrence.

Microscopic observations enabled the localization of damages and the characterization of the fiber/matrix interface quality. In fact, the fiber pullout is sudden and the surface of the fibers is smooth in the case of a weak interface.

The second is a quantitative approach, which involves a damage theory based on the concept of the effective stress. This method uses the measure of the level of damage reached in the material under cyclic loading such as load–unload tension. The evolution of the material rigidity obtained through the measurements of Young's modulus at each step of successive loading makes it possible to determine the damage parameter D and its evolution during loading.

The approach presented allowed a better understanding of the mechanical behavior of the materials studied, their damage and failure in respect to structural and moulding parameters.

Резюме

У композитному матеріалі, що армований випадково розташованими короткими волокнами, пошкодження при механічному навантаженні виникають внаслідок дії різних елементарних механізмів, таких як мікророзтріскування матриці, висмикування волокон, руйнування міжфазової границі волокно–матриця, руйнування волокон. Ці пошкодження зумовлюють значний вплив на макроскопічні властивості композитного матеріалу. Щоб отримати прийнятні механічні характеристики композитного матеріалу, важливо оптимізувати концентрацію волокон, а також якість міжфазової границі волокно–матриця, які безпосередньо впливають на ці пошкодження. Головна мета роботи – визначення впливу структурних параметрів на еволюцію пошкодження двох типів склопластиків на основі поліпропілену. Одночасно з класичним підходом до механічної теорії пошкодження, що заснована на випробуваннях на розтяг із періодичним розвантаженням, використання методу акустичної емісії дає змогу вивчати в реальному часі природу і важливість механізмів пошкоджень у процесі навантаження. Додатково до цього фрактографічний аналіз дозволяє підтвердити різні передумови і висновки даного дослідження.

1. D. Laroche, *Accumulation d'Endommagement Dans le Carbone Époxyde Détecté par Emission Acoustique*, Thèse de docteur-Ingénieur de l'école de mines de Paris, (1980).

2. D. Valentin, *Etude par Émission Acoustique de l'Endommagement Soussollicitations Mécaniques des Composites Carbone–Résine*, Thèse de Docteur–Ingénieur de l'école des mines de Paris (1983).
3. M. Fuwa, A. R. Bunsell, and B. Harris, "Tensile failure mechanisms in carbon-fiber-reinforced plastics," *J. Mater. Sci.*, **10**, 353–364 (1975).
4. B. Ponsot and D. Valentin, "Analyse qualitative et quantitative de l'endommagement de structures composites par émission acoustique," *JNC5*, Paris (1986), pp. 123–129.
5. Xl. Gong, A. Laksimi, and M. L. Benzeggagh, "Nouvelle approche de l'émission acoustique et son application à l'identification des mécanismes d'endommagement dans les matériaux composites," in: *Revue des Composites et des Matériaux Avancés*, **8**, No. 1, 7–23 (1998).
6. A. Laksimi, M. Cherfaoui, and C. Bathias, "Etude par émission acoustique de l'amorçage et de la propagation dans un composite verre-époxy," *JNC5*, Paris (1986), pp. 625–637.
7. A. Laksimi, S. Benmedakhene, and L. Bounouas, "Monitoring of acoustic emission during tensile loading of thermoplastic composite materials," 12th International Conference on Composite Materials, July 5–9, 1999, Paris.
8. J. M. Berthelot and J. Rhazi, "Caractérisation des mécanismes de rupture dans les matériaux composites à l'aide de l'amplitude des signaux d'émission acoustique," *JNC5*, Paris (1986), pp. 230–238.
9. J. R. Wadim, *Acoustic Emission Applications*, Dunegan Endevco, San Juan Capistrano CA (1978).
10. S. Barre, "Etude de l'endommagement des matériaux composites à fibres courtes et à matrice thermoplastique sous chargement statique et cyclique," DEA UTC, Compiègne (1991).
11. L. Bounouas, S. Benmedakhene, A. Laksimi, G. Villoutreix, and A. Ilidrissi, "Etude de l'influence des paramètres structuraux et de mise en oeuvre sur le comportement mécanique des matériaux composites," in: *Revue des Composites et des Matériaux Avancés*, **8**, No. 2, 179–205 (1998).
12. F. Meraghni, S. Benmedakhene, and M. L. Benzeggagh, "Identification and modeling of damage mechanisms in short glass-fiber-reinforced polypropylene composites," in: Proc. ICCM 10, Vancouver, Aug. 1995, pp. 366–369.
13. S. Benmedekhene, *Etude Phénoménologique du Comportement à l'Impact des Matériaux Composites*, Thèse de doctorat UTC, Compiègne (1996).
14. L. M. Kachanov, "Time of the rupture process under creep conditions," *Izv. Akad. Nauk SSSR*, **8**, 26–31 (1958).
15. J. L. Chaboche, "Sur l'utilisation des variables d'état internes pour la description du comportement viscoplastique et de la rupture par endommagement," in: Symp. Franco-Polomais de Rhéologie et Mécanique, Cracovie (1977).

Received 28. 04. 2000