EVALUATION OF GLASS FIBER/POLYPROPYLENE COMPOSITES BY QUANTITATIVE LASER ULTRASONIC AND ACOUSTIC EMISSION TECHNIQUES

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Abstract: The objective of this work is to study the influence of the moulding conditions of manufacture on the mechanical behaviour of the composites, when which they are submitted to mechanical loading. In order to evaluate the quality of interfacial adhesion, two non-destructive testing procedures were combined. Laser ultrasonic was employed to evaluate the state of the composite microstructure depending on the moulding conditions. Measurements of ultrasonic velocity and attenuation were applied to characterize microstructure and to assess the mechanical properties of the composite. Acoustic Emission techniques coupled with mechanical testing in mode II were then used to study the progressive nature of interface damage due to mechanical loading in mode II.

Introduction: The use of thermoplastic composite in the manufacturing applications has become well recognized. The major obstacle to the efficient application of composite materials, in general, is their tendency to develop fiber/matrix interface damage and delamination. Interface damage is the most predominant failure mechanism in composite structures and may due to non-optimum curing conditions. The mechanical and environmental performances of these materials depend largely on fibers, the matrix and the quality of adhesion to the interface fibre/matrix. In general, these materials are moulded by simultaneous application of heat and pressure. During moulding, the crystallization of polymer in a molten state rests on processes of germination and crystalline growth. Consequently, the degree of crystallinity of polymer, function of the temperature, has a direct influence on the tensile strength, on the toughness, and on the long-term performance of the composite. Laser-Ultrasonic technique has been used as non-contact evaluation technique to evaluate microstructure variation due to different moulding cycles of the thermoplastic materials. Ultrasonic high-energy pulses were generated on a variety of composite samples, using a Q-switched Nd: YAG laser. The detection of ultrasound waves was performed by a confocal Fabry-Pérot interferometer. The fracture toughness has been analyzed by fracture mechanics analysis using the end notched flexure test (ENF) in pure mode II [1] and the strain energy release rate was correlated to Acoustic Emission energy dissipated by the material in three-point bending test. Ultrasonic wave measurements were used in conjunction with mechanical tests to distinguish between toughness behaviour due to different composite moulding conditions.

Laser-Ultrasonic Measurements: The purpose of the present investigation was to apply a non-destructive evaluation method based on laser-ultrasonic technology to characterize microstructure variations in polypropylene composites by means of measurements of high frequency ultrasonic attenuation and ultrasonic velocity. Laser ultrasonic technique (Figure 1) is a novel inspection tool using lasers for the generation and detection of ultrasound waves in materials. The technology is now well established [2,3,4]. When a low power light pulse from a laser beam strikes a solid surface of materials, the material absorbs one part of electromagnetic energy; its produces localized heating. Just below the surface, the fast rising of temperature produces a thermal expansion of the material. In the thermoelastic regime, the induced thermoelastic stresses generate elastic waves propagating in the material in shear and bulk mode. The detection of ultrasonic waves is based on optical interferometry [3]. For detection of ultrasonic waves, a confocal Fabry-Pérot interferometer uses light reflected from surface of material, and detects

changes in the frequency of reflected light. The interferometer output signal depends on the velocity of waves arriving at the material surface using the Doppler effect.
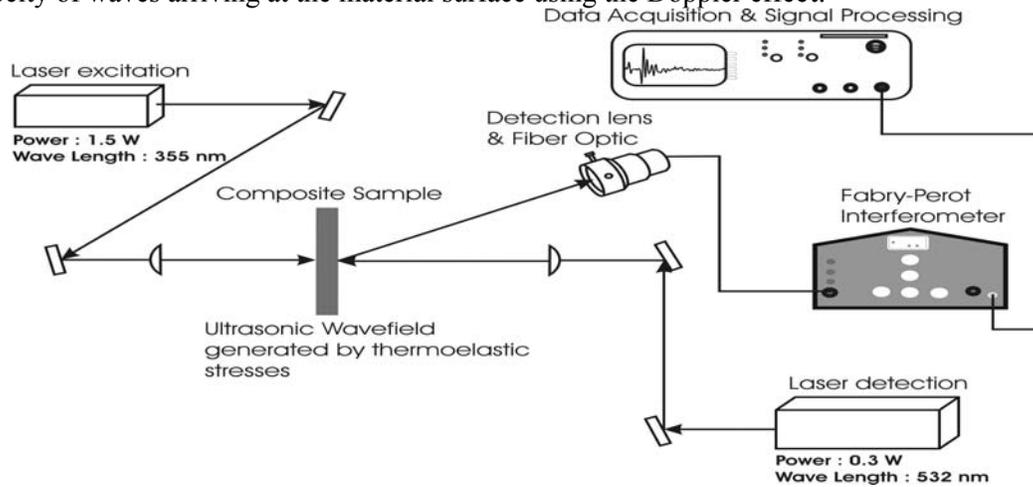

Figure 1. Laser ultrasonic experimental setup

Composition and microstructure influence ultrasonic wave propagation, then ultrasonic characterization of molding conditions effects on composite materials is possible with laser ultrasonic. Attenuation measurements will permit to establish correlations between composite microstructures and variations of mechanical properties related to effects of composite molding conditions. Ultrasonic attenuation measurements, although strongly frequency dependant, have been used successfully to determine grain size and grain-size distribution. Wave scattering and absorption is the energy loss mechanisms that govern ultrasonic attenuation in the frequency ranges of interest. Ultrasonic wave attenuation is relatively high in polypropylene composites, primarily because of scattering by the fibers and isothermal absorption of the polypropylene resin. High attenuation can be reduced by using the through-transmission ultrasonic mode, passing through the test object only once before arriving to detector [5]. The ultrasonic velocity **v** is given by: $v=2d/t$, where, **d** is the sample thickness and t is the time for ultrasonic wave propagation to travel through the sample in round-trip. By using Fast Fourier algorithm applied to laser ultrasonic signals, amplitude spectra **A(f)** is used to calculate the echoes attenuation by using [5]:
$\alpha(f) = (ln\ A_1(f)-lnA_2(f))/2d$, where **f** is the frequency, **α(f)** is the attenuation of ultrasonic wave echoes, $A_1(f)$ and $A_2(f)$ represent the amplitudes spectra of arriving wave and its echo respectively.

Toughness in pure shear mode II: Under load, Interlaminar stresses may develop between layers and these can lead to interlaminar fractures or delamination. These failures constitute a major damage in thermoplastic composites since it's reduce the stiffness, strength and the fatigue life of the structure. The interlaminar toughness is commonly characterized by so-called mode I tension test and mode II shear test. Energy release rates G_I and G_{II} obtained from these tests provide a valuable measure of delamination fracture toughness. In the present study, the emphasis is placed on analysis of crack propagating in shear mode II. End notched flexure test (ENF) is mechanical test were used to evaluate the fibre/matrix interface properties of the composite, depending in moulding condition of materials. Many investigations, concerned with measurements of interlaminar fracture [6], show that fracture toughness can be determined using the end notched flexure test (ENF) in pure shear mode II, as shown in figure 2, where the crack of length a_0 is parallel to the plane of plies.
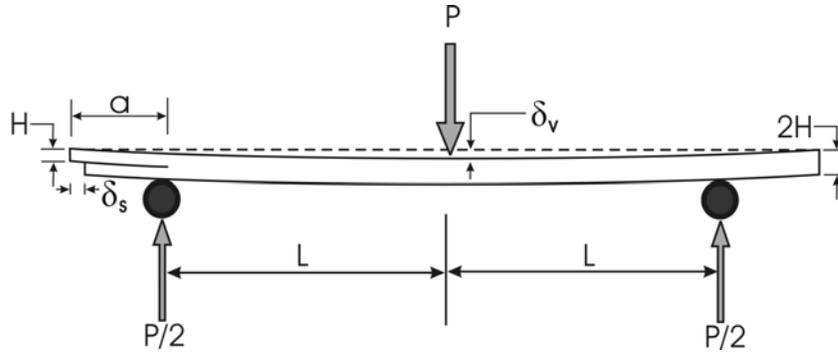
Figure 2. End notch flexure test (ENF)

The ENF specimens are containing an artificial delamination, therefore generating a fracture energy value for stable and unstable crack growth conditions. Using elementary beam theory for applied load to ENF specimen is possible to determine the energy release rate G_{II}. Many model are used to obtain the energy release rate, such area method, compliance calibration, beam theory and other methods [1]. Let us consider the load-displacement response of cracked plate of thickness 2H subjected to a concentrate force *P* (figure 2). As long as there is no crack growth and the elastic behaviour is linear, the load-displacement relation is *u= CP*, where **C** is the compliance of the plate, and **P** is the load on the R-curve. Compliance *C*, reciprocal of stiffness, is evaluated from the load and displacement curve. The crack shear displacement compliance, λ, is defined as the ratio of the crack shear displacement, δ_s, to the applied load, **P**: $\lambda = \delta_s/P$, as shown in figure 2. The energy release rate is defined as; $G_{II}=(P^2/2B).(dC/da)$, where **B** is the width of the ENF specimen, **a** is the crack length. The energy release rate G_{II} can then be evaluated as function of crack length using equation obtained from the approximate analysis based on elementary beam theory [1], $G_{II}=(9CP^2a^2)/2B(2L^3+3a^3)$, where **2L** is span length.

Acoustic Emission technique (AE): Acoustic Emission testing (figure 4) of fibre composite materials is gaining a broad industrial acceptance as a monitoring method of defects growth in composite structure under load. Abrupt internal stresses caused by crack growth, liberate stress waves that travel through the material and eventually are detected by piezoelectric or electro-optic sensors. Acoustic emission appears [7,8], to be one of few techniques that form the basis of monitoring damage in composites in real time. The detected acoustic emission signals are ultrasonic waves generated in composites by failure modes involving delamination, debonding, fibre failure, matrix failure and fibre pull out. Generally, the emissions are broadband transients at their source but the waveform detected by the sensor will be distorted by frequency-dependant attenuation, dispersion, multipath propagation and mode conversion depending on the wavelength and the specimen geometries. The relation between the energy released by the fracture process and the energy detected by AE sensor, have been discussed by *Rotem and Altus* [9]. It has been shown that, the acoustic energy stored in the propagating stress wave would be proportional to the square of voltage produced by broadband transducer, by: $E_{EA}=\alpha \int V(t)^2 dt$, where α is the gain, *V(t)* is the amplitude of acoustic emission signal, and E_{EA} is the energy of the signal. Assuming that this acoustic energy is caused by release of strain energy as the crack advances suggests that is might be proportional to G_{IIc}, calculated experimentally by: $G_{II}=(9CP^2a^2)/2B(2L^3+3a^3)$.

Experimental Procedures: Two types of samples are used. All of them are made from prepreg unidirectional composite with glass fibre volume fraction of 60% and polypropylene matrix. Sample dimensions respect the ASTM standards D5528. All samples have eight unidirectional layers. For the moulding cycles (figure 3), the first category of samples (B samples) was heated up to 220^0C for five minutes while the others (W samples) only reached 165°C for five minutes. The constant pressure was held at 100 Psi during the moulding process.
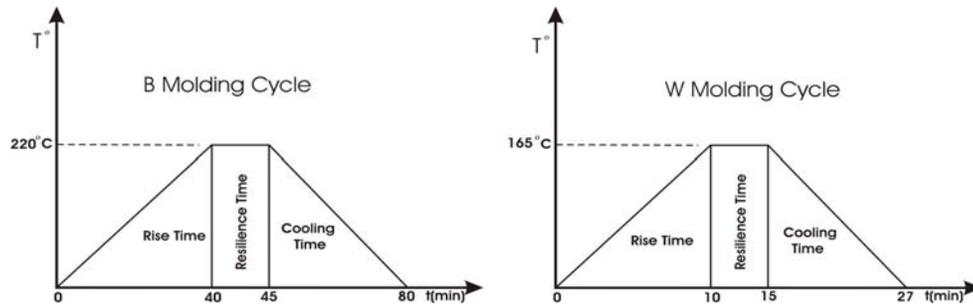
Figure 3. Moulding conditions of polypropylene/glass samples

For Laser-ultrasonic measurements, the system is based on the use of laser beams for the generation and detection of ultrasonic waves (figure 1). The waves are generated using a Q-switched Nd:YAG laser Brilliant from Quantel with variable wavelength (1064 nm and 355 nm) and corresponding pulse energies 350 mJ and 60 mJ. The detection laser is a continuous wave Compass laser (Adlass) with 532 nm wavelength and 150 mW output power. The detection is based on Doppler effect and wave velocity measurements. The sensing of ultrasound is operated by LISOR (Light-In-Signal-Out-Receiver) an optical sensor based on the technology of confocal Fabry-Pérot interferometry. For fracture toughness measurements, the ENF specimens were prepared by inserting a Teflon tape into one end at the desired interface to help initiate delamination under loading. During delamination growth, Acoustic Emission stress waves was detected and recorded using a large broadband *B&K* sensor attached to specimen (figure 4). For each sample, crack propagation was monitored by acoustic emission and by using a visual graduation on the sample. The broadband transducer was connected to a wide band conditioning amplifier that has the purpose of filtering and amplifying. The signal amplification was by 66dB. The filter is set at a 50kHz-2MHz range so that interference between AE signals and low frequency noise is avoided. A DATA 6100B system recorded the AE signals emitted during loading. While the sampling period was set at 300ns, the threshold was fixed at 480mV. Each signal was formed of 1024 points.
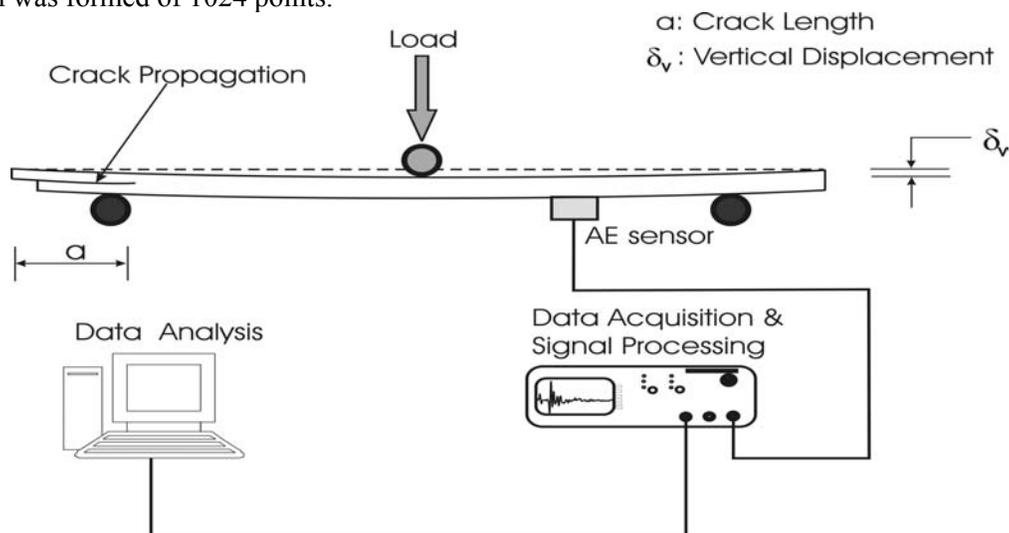

Figure 4. Acoustic emission experimental setup

Results and discussion: Typical waveforms obtained from the sample B, heated up to 220^0C for five minutes, and sample W, heated up to 160^0C for five minutes, are shown in figure 5a. and

figure 6a. respectively. Figures 5b. and 6b. correspond to envelope of received laser ultrasonic signal calculated by using Hilbert transform in time domain. The components of signals are very short in time and correspond to multiple ultrasonic reflections in composite. In all laser ultrasonic tests, a longitudinal beam is incident normal to the polypropylene composite surface. Under these conditions, the effect of material anisotropy can be neglected and the approach is similar to tests of polypropylene isotropic media [5]. Even though the materials behave as if it were isotropic, its layered nature contributes to reflections and increased attenuation. Only a small portion of the initial pulse energy is returned to optic probe at the end of its first round trip through the test material. As each echo returns to the front surface, only a small fraction of its energy passes through the material surface and interacts with detection laser beam. The ultrasonic pulse energy remaining within the test composite material continues to bounce back and forth between the parallel front and back surfaces until it has dissipated within the test material. A comparison of these multiple back reflections for two different samples are shown in figure 6 a) and figure 6 b) representing envelope of laser ultrasonic signals. Comparing to Sample B, the ultrasonic signal detected in sample W it is significantly reduced in amplitude. These amplitude variations are caused certainly by differences in microstructures due to different crystallization levels and molding conditions. This result gives a clear indication of the interaction of microstructure and high frequency ultrasonic waves. Figure 7 shows the variation of attenuation with frequency for various samples. The attenuation increase with frequency, it can be seen that attenuation is lower for the sample B and higher for sample W. The only difference between materials B and W is the degradation of the polypropylene matrix affected by inappropriate molding cycles. One knows the differences in temperature that there was between the samples B and W during their manufacture. It is easy to deduce from it that the temperature of molding of the composite intervenes on the quality of the microstructure of the polypropylene composite. To quantify the resistance of polypropylene to crack propagation, toughness of interfaces are measured by means of ENF test. Fracture toughness properties of different molding condition samples are evaluated by interlaminar fracture toughness test in mode II. The curves of bending normal stress are presented at figure 8. It is seen that initially for the deflection going until 8mm, the two sample groups B and W behave same manner. Figure 9 represents the variation of bending modulus Ef calculated from the expression; $E_f = (L^3P/4Bh^3w)$, where **L** outdistances between supports, **h** the thickness and **w** the fleche of the neutral axis. As one sees it on figures, the resistance and the module of bending are higher for the samples B than for the samples W before rupture zone that is visible of share the brutal fall of the curves. As of the beginning of the loading, the samples B have more raised rigidity and more resistance. The rupture of the samples B occurs with a bending stress much higher than in the case of the samples W (figure 8). The samples B broke without there being cracking observed visually as a preliminary. For this reason, it was difficult to calculate the critical release rate energy in mode II, G_{IIc}, for the samples B. Figure 10 plots, G_{IIc}, against the crack length for a group of W samples. Being given that the samples B did not fissure, we conclude that the samples B offers a better opposition to the propagation of the crack. By consequent, the samples B offer a better tenacity than the samples W. It is clear that the interface has a role paramount in the propagation of the crack in the direction of fibers. It is the quality of the interphase that will determine propensity with delamination between fibers. This performance is surely connected to a better behavior of the material B at the time of shearing to the interface fibre/matrice. As that had already been shown with other means, by *J.Denault* [10], the molding of material at high temperatures improves quality of the interface in the case of the composite polypropylène/fibre of glass, these results agree with those obtained by this work. The AE results obtained for ENF tests and emitted by samples for materials B and W are shown in figure 11 and figure 12 respectively. Figures show the histogram of the cumulative energy values of AE signal versus number of events emitted during pure mode II end-notched flexure testing. The rate of acoustic emission activity, as indicated in figure 11 and 12, tends to increase with increasing load as the interlaminar damage becomes more important. First of all, the sample B (figure12) is much

less emissive than the sample W, from a point of view number of signals. In the case of the B sample, it seems that the energy of the signal increases rapidly with the bending stress applied. The initiation of the damage appears for a stress from approximately 240 MPa corresponding to 10 mm deflection. On the other hand for the sample W, the initiation of cracking
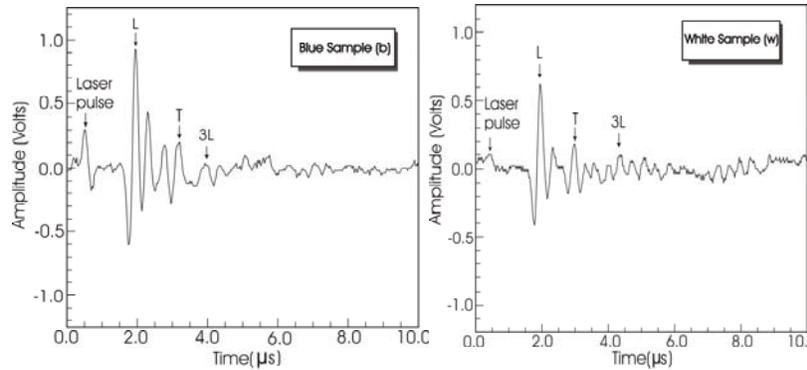

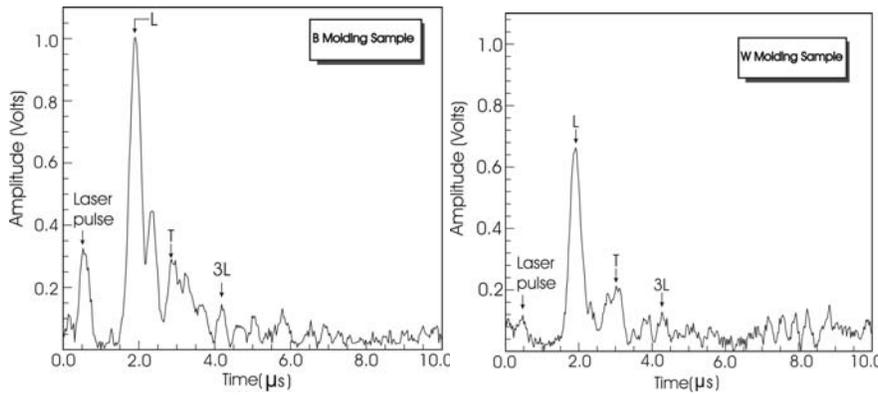Figure 5a. Waveform detected from sample B   Figure 5b. Waveform detected from sample W

Figure 6a. Waveform envelope of fig.5a.   Figure 6b. Waveform envelope of fig.5b.
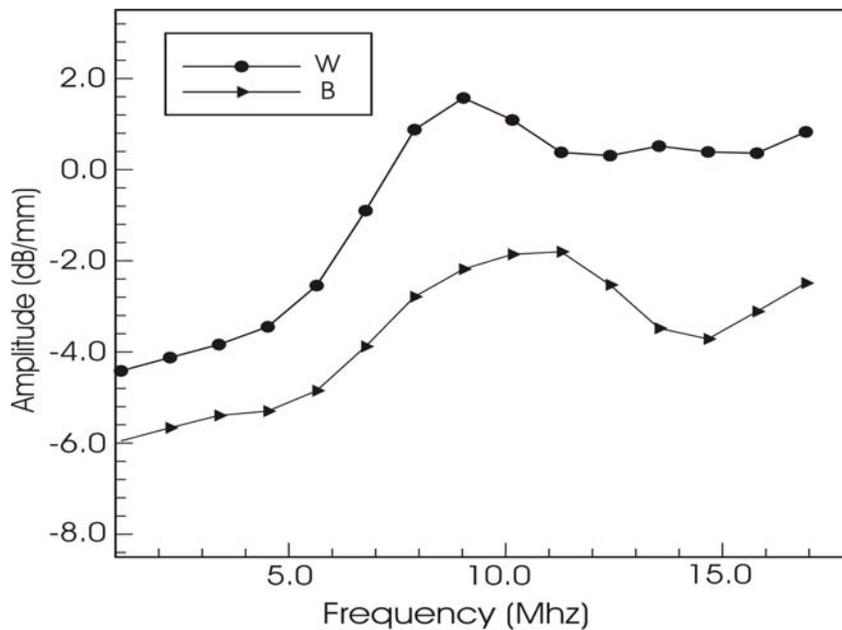
Figure 7. The attenuation spectra of samples measured by laser ultrasonic
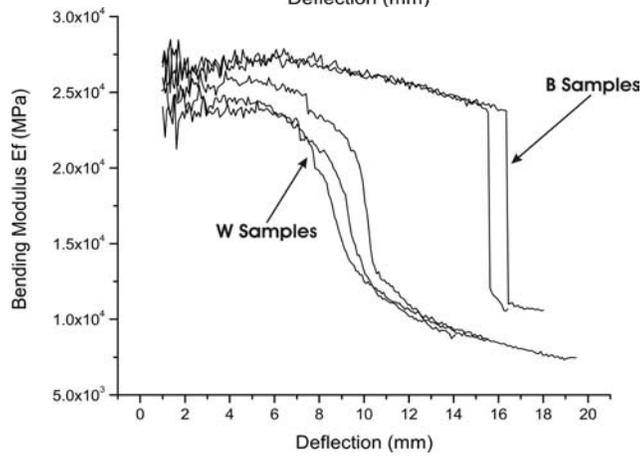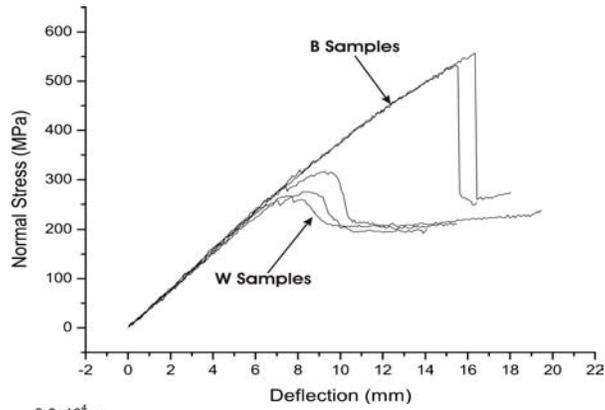

Figure 8. Normal bending stress
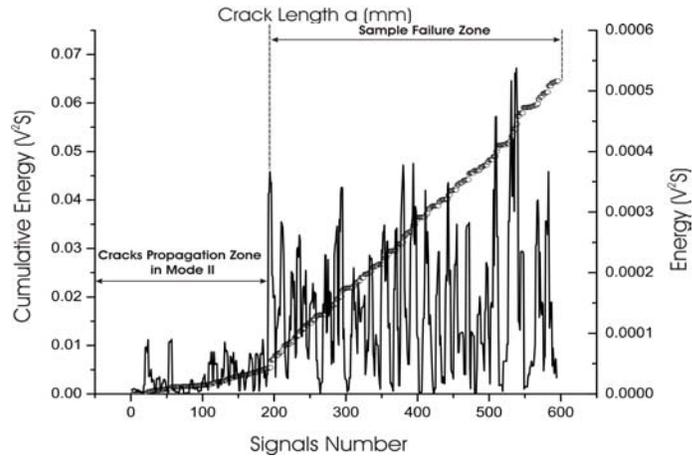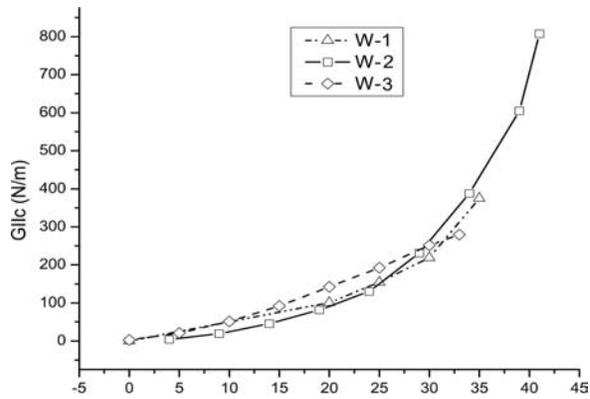
Figure 9. Bending modulus versus deflection

Figure 10. Critical energy release rate G_{IIc} by sample W

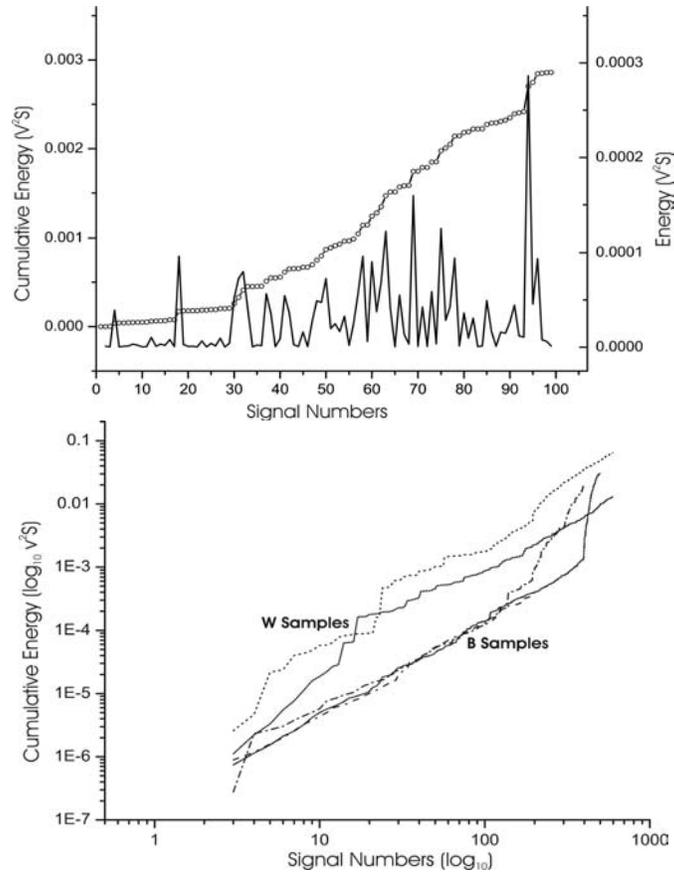Figure11. AE energy distribution emitted

Figure 12. AE energy distribution emitted by sample B samples

Figure 13. AE activities of

is done with stresses much lower than 50 MPa and for deflections about a few millimetres. The moulded composite with 165°C goes deteriorated much more quickly under a loading in bending than the moulded composite to 220°C. The samples W seem "to crack" much more easily than the sample B. Figure 13 presents, in logarithmic scale, the cumulative energy distribution for all the samples tested, according to the number of detected signals. We notice once again whom the samples W have an acoustic activity of stronger energy than the samples B. With regard to the samples W, we see appearing jumps in the curves. These jumps were assigned to advance of the interlaminar cracks and delaminations in the sample, and thus with the appearance of acoustic events of greater amplitudes.

Conclusion: In the present study, we compared the behaviour of two polypropylene/glass cross-ply laminates submitted to pure shear loading in three-point bending test. The toughness of materials subjected to different moulding condition was analysed. Before interlaminar mechanical testing, a non-destructive evaluation method based on laser-ultrasonic technology was applied to characterize microstructure variations in polypropylene composites by means of measurements of ultrasonic attenuation. It has been demonstrated how quantitative information concerning the microstructure state can be discriminate from the ultrasonic signals and attenuation versus frequency. The laser ultrasonic technique allowed separation in a very clear way the groups of samples having different conditions of moulding. The temperature of crystallization has an influence on the global microstructure of material and especially on the interphase since it

influences resistance to the delamination of composite material. The interlaminar damage progression in materials was mainly followed using end notch flexure tests, ENF, coupled with acoustic emission technique. The results show clearly, that the temperature of moulding has an effect on the mechanical properties and on the tenacity of material. A temperature of moulding of 220°C confers on material a better resistance to cracking than a temperature of moulding of 165°C. The results of this study, make it possible to conclude that the acoustic emission is a reliable technique which offers the potential to follow the development of the interface damage during the test, and to quantify the capacity of material to resist the propagation of cracks under shearing in mode II.

References:

[1] K. Kageyama, M. Kikuchi, and N. Yanagisawa, "Stabilized End Notched Flexure Test: Characterization of Mode II Interlaminar Crack Growth", *Composites Materials: Fatigue and Fracture (Third Volume), ASTM STP 1110, T.K. O'Brien Ed. American Society for Testing and Materials*, Philadelphia, pp.210-225, 1991.

[2] J. P. Monchalin, C. Néron, J. F. Bussiere, et al., Laser-Ultrasonics: From the Laboratory to the Shop Floor, *Physics in Canada*, pp. 122-130, March/April, 1995.

[3] C.B. Scruby, and L. Drain, *Laser Ultrasonics-Techniques and Applications*, Adam Hilger-IOP Publishing, Bristol, 1990.

[4] A. Maslouhi, R. Leblanc " Application of Laser-Ultrasonic Technique to Evaluate Microstructure Transformation In Aluminium Alloy'' ASME *Proceeding of 7th NDE Topical Conference, 7th Nondestructive Evaluation Topical Conference,* April 23-25, San Antonio, Texas (USA), 2001.

[5] Non Destructive Testing Handbook, Volume Seven*, Ultrasonic Testing*, ASNT, 1991.

[6] N. Sela, and O. Ishai, "Interlaminar fracture toughness and toughening of laminated composite materials: a review", *Composites,* 20(5), pp. 423-435, 1989.

[7] H. Kioua, A. Maslouhi, and C. Roy " AE monitoring of Free-Edge Delamination in Graphite/Epoxy Laminates Under Fatigue Loading*", Canadian Aeronautics and Space* J., Vol. 41, No. 1, pp 21-27, 1995.

[8] A. Maslouhi., H. Saadaoui, S. Beland and C. Roy, Acousto-Ultrasonic Signal Classification to Evaluate High Temperature Degradation in Composites, *Journal of Acoustic Emission*, 12, Number 1/2, pp. 45-54, 1994.

[9] A. Rotem and E. Altus, " Fracture Modes and Acoustic Emission of Composite Materials", *1979, Journal of Testing and Evaluation*, JTEVA, Vol.7, No.1, Jan., pp.33-40, 1979.

[10]   J. Denault, Interfacial interaction in carbon fiber/thermoplastic composites, Composite Interfaces, Vol. 2, No. 4, pp. 275-289 (1994).