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Ultrasonic consolidation (UC) debulking of thermosetting prepreg for autoclave curing of composite laminates

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Abstract

Debulking of prepreg (pre-impregnated resin system) layers during hand lay-up manufacturing of carbon fibre reinforced polymers (CFRP) is a key-step to reduce air content and maximise the mechanical properties of the final product. Debulking is usually performed using vacuum-bag cycles of 10-15 minutes applied after the lay-up of every three or five prepreg layers, leading to a considerable time-consuming process. In this work, the use of ultrasonic stimulation during vacuum is studied to improve the efficiency of the debulking process and reduce the number of operations in order to decrease the overall manufacturing time. Three CFRP laminates were laid-up using the proposed ultrasonic consolidation (UC) with three different exposition times (5, 10 and 15 minutes) and cured in autoclave. The UC debulking process consists in a vacuum cycle with ultrasonic waves sent to the uncured material through an ultrasonic transducer. In order to evaluate the efficiency of this process interlaminar shear strength (ILSS) and in-plane compressive properties were tested. Experimental results show for 15 minutes compressive properties comparable with the ones obtained from reference samples manufactured using the traditional debulking technique, and high improvements in terms of ILSS (>20%). Therefore, UC debulking process can be used during hand lay-up of prepreg in order to improve the interlaminar properties of the final part and reduce the debulking time by over 85%.

Keywords: ultrasound; ultrasonic consolidation; prepreg debulking; thermosetting prepreg curing; CFRP.

1. Introduction

The use of composite materials for both primary and secondary structures is increased in the last decades, particularly for high-performance applications such as aerospace, racing automotive and wind blades due to its high mechanical properties (such as strength, elastic modulus and fatigue strength) and low weight. An example of advanced composite materials commonly used is Carbon Fibre Reinforced Polymer (CFRP), a polymeric matrix (thermosetting or thermoplastic) reinforced with carbon fibres. Several manufacturing processes are available for CFRPs. Breuer published [1] a detailed state of the art of composite materials manufacturing technologies, reporting that the majority of airframe composite components are manufactured by lay-up of pre-impregnated layers known as 'prepreg'. Prepreg material is a layer of carbon fibres arranged in a Unique Direction (UD) or woven together to form a texture impregnated with a thermosetting resin and stored at low temperatures (i.e. -18ºC) in order to delay the cure of the matrix. The influence of the fibre orientation of each prepreg ply along the stacking sequence over the mechanical properties of the final part is well known [2–5], providing high flexibility in the design of the final properties of the part.

Despite the increasing interest of industry in automated processes of advanced composite manufacturing, such as Automated Tape Laying (ATL) [6] and Automated Fibre Placement (AFP) [7], hand layup of prepreg layers remains the main manufacturing method in many cases, especially for research and development of new components and several high performance manufacturing facilities due to its flexibility and adaptability [8]. The process consists in cutting prepreg layers to the desired dimensions and shapes, and manually laminating each one to a mould or to the previous layer. Very high-quality and flexible structures can be designed and produced by hand lay-up with a low start-up cost.

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In order to guarantee the maximum quality and mechanical properties of the laminate, the removal of the air entrapped between the layers (consolidation) is a key step in the prepreg lay-up. Porosity directly affects the mechanical properties of the laminate, especially interlaminar shear strength (ILSS) and compressive strength [9–13]. Indeed, Jeong [9] presented an experimental study on laminated composites with a wide range of void contents, showing that the mechanical properties (in particular the ILSS) are strongly dependent not only on void content but also on voids geometry. A statistical approach on a similar study was also published by Yoshida et al. [11] while Hancox [13] carried out a theoretical study on the influence of the contents and geometry flaws and voids in shear properties of CFRP rods and tubes.

In order to remove air and reduce (final) void content, different procedures are used during the hand lay-up process. Firstly, pressure is manually applied during the lamination step of every ply using a specific tool (‘Dibber’ tool) to remove big air packages (Figure 1.a) [8]. Then, as suggested by Hexcel® in the HexPly® Prepreg Technology guidelines [14], prepreg sheets need to be consolidated using a vacuum bag assisted procedure (Figure 1.b) for 10-15 minutes every 3 or 5 layers, depending on the geometry and shape of the part. After the stacking has been completed, the consolidation is ensured by vacuum and pressure applied during the autoclave cure. Given that the autoclave pressure is applied during the resin cure, it has no influence on the total duration of the manufacturing, while the hand pressure and the numerous vacuum cycles during the lamination are the main time consuming steps for prepreg hand lay-up process, particularly for components with large thickness.

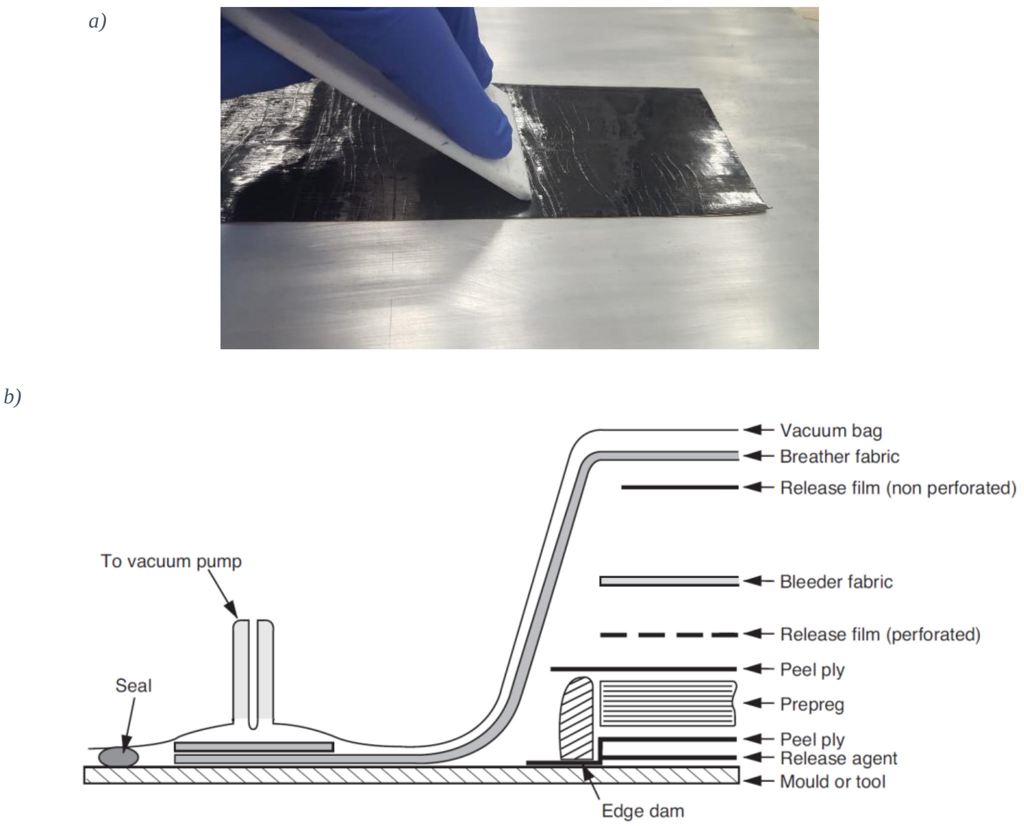


Figure 1 – a) Dibber tool for air removal and b) vacuum bag set-up (figure from [14])

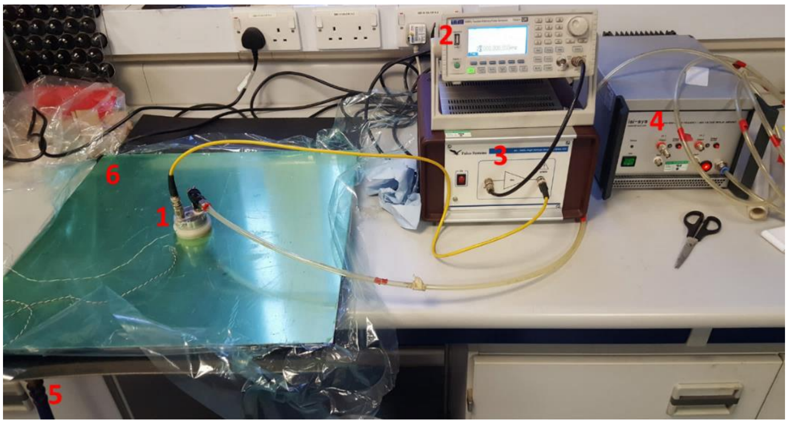
Based on these premises, in order to reduce the process time and ensure high level of consolidation, the use of ultrasound waves is considered. This technique, called ultrasonic consolidation (UC), takes advantage of vibrations generated by the waves in to the material in order to facilitate the removal of the air trapped between layers. The benefits in the use of UC are well known for automated lay-up of both thermosetting and thermoplastic composites [15–18], where ultrasound waves are used for both matrix debulking and curing (heat generation), with a frequency generally in the range of 20-120 kHz [15]. Lionetto et al. [16] presented both experimental and numerical analyses of an automated lay-up process that uses ultrasonic propagation in order to provide pressure and heat during filament winding of thermoplastic matrix composite, reporting a void content within the typical range for composites processed by filament winding and other traditional methods. Rizzolo et al. [17] experimentally studied the UC process during AFP of PET/carbon composite samples resulting in significantly higher mechanical properties in comparison with those obtained by hot-press manufacturing process. In their work, Chu et al. [18] analysed the influence of ultrasonic AFP (UAFP) on the mechanical properties and microstructure crystallization of thermoplastic composites showing a good match with properties of the same specimens produced by hot-press.

However, there are cases in which debulking and heating are undesired to take place at the same moment. When UC is applied to a thermosetting prepreg, the transmission of high levels of energy to the material through ultrasonic waves may activate chemical reactions, generating cross-links between polymer's molecules via frictional heat and consequently cure the matrix. This is particularly undesirable when maximum mechanical properties in the part can be reached only with autoclave cure, or when it is necessary to maintain the stickiness of the material. Foster-Miller company, now part of QinetiQ, patented several manufacturing methods and devices based on the use of ultrasound waves for prepreg compaction: they designed a method for the manufacturing composites, called ultrasonic tape lamination, where an ultrasonic horn induces shear waves with a small angle and low frequency [19] to the surface of thermosetting prepreg plies for consolidation [20]; the same process was also applied on thermoplastics [21]. The device was proved to generate enough energy to remove the air between the plies without activating the matrix cure and allowing the stuck of the following layer.

In this work, the UC is proposed as debulking procedure of manual lay-up before the autoclave cure for CFRP prepregs in order to decrease the time of the manufacturing process. Also, the process is used to reduce the porosity and therefore improve the interlaminar properties of the material. Using a low level of frequency (and thus of generated heat) and constantly monitoring the material temperature, the layers were consolidated without generating the amount of heat required to activate the cure. A manufacturing set-up is then presented and used to fabricate three different sample using different UC exposition times. In order to prove the efficiency of the process, the mechanical properties (compressive and interlaminar) of the samples were experimentally evaluated and compared with reference samples, obtained using a standard consolidation technique.

2. Samples manufacturing

In this section, the manufacturing of three different samples using ultrasonic consolidation (UC) method is illustrated. Three different laminates of dimensions 400x100 mm were manufactured via manual stratification of prepreg layers with a very limited pressure applied manually with a Dibber tool and without any vacuum-bag assisted process during the lay-up. The chosen lamination sequence was $[(-45/0/45/90)_2/-45/0/45/\overline{90}]_S$ and the used material is a unidirectional carbon fibre prepreg with 977-2 epoxy resin system and Tenax$^{®}$ - E IMS65 fibres produced by Cycom$^{®}$. Each laminate was attached to an aluminium plate (previously covered with release agent to facilitate the removing) and enclosed in a vacuum bag. Afterwards, the UC process was carried out: an ultrasonic transducer was connected to the centre of the opposite side of the aluminium plate through a vacuum pump, as shown in Figure 2, in order to transmit the ultrasonic waves to the plate and thus to the material. Simultaneously, a second pump attached to the bag applied the vacuum over the uncured laminated to apply pressure over the prepreg. In order to limit the frictional heat generated inside the material and avoid the matrix to start the cure, the used signal was a sine wave with a frequency of 28.0 kHz (central frequency of the transducer) and the material temperature constantly monitored using thermocouples. Figure 3 shows a schematic representation of a section of the consolidation plate. The ultrasonic consolidation procedure was applied for 5, 10 and 15 minutes respectively to each of the three uncured laminates (UC1, UC2, and UC3).



a)

Figure 2 - Ultrasonic consolidation setup: 1 vacuum ultrasonic transducer; 2 waves generator; 3 amplifier; 4 transducer vacuum pump; 5 vacuum bag pump; 6 aluminium plate bottom part.

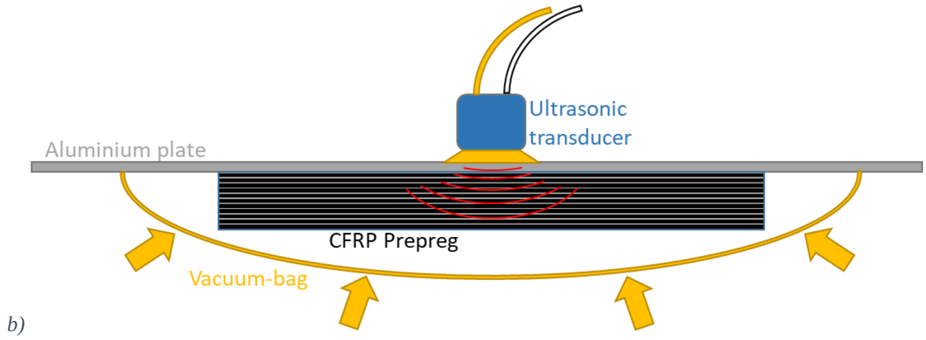


Figure 3 – Ultrasonic consolidation schematic representation

In order to evaluate the effective advantages and possible limitations of the UC process for the manual stratification, a reference laminate sample was manufactured in similar conditions following the standard debulking process: a vacuum cycle of 15 minutes every 3 plies, for a total of 120 minutes. Afterwards, the three UC samples and the reference sample (PC) were cured via autoclave at 180ºC and a pressure of 100 psi for 3 hours (Figure 4.a). A heating and cooling rate of 2ºC/min was used. Table 1 summarises the debulking techniques, times and average thicknesses of the four cured laminates. In the calculus of process times, side procedures were not considered, such as the time to prepare the vacuum bag and enclose the laminate in it, because strongly depending on the operator ability. Anyway, these side procedures are significantly less in the UC debulking process (only one vacuum bag) in comparison with traditional one (one vacuum bag for every three layers). After the cure, the laminates were cut using a diamond blade obtaining the appropriate specimen dimensions (Table 2) for the experimental campaign (Figure 4.b).

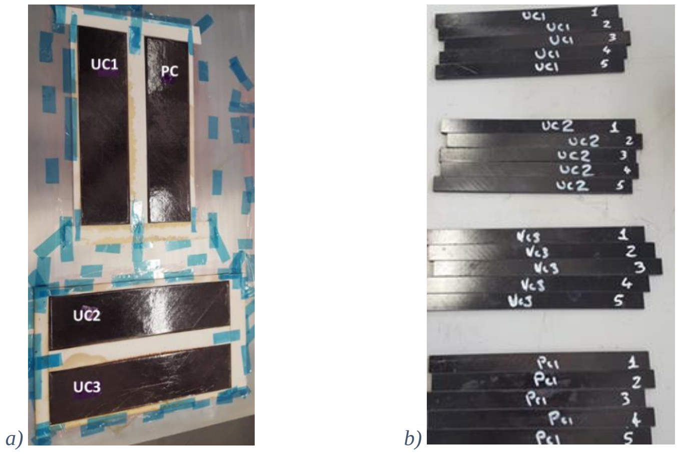


Figure 4 - a) Laminates after autoclave cure and b) samples after diamond blade cut

Table 1- Manufacturing debulking times and thicknesses of samples

	UC1	UC2	UC3	PC
Debulking method	Ultrasonic Consolidation	Ultrasonic Consolidation	Ultrasonic Consolidation	Classic
Debulking time (*min*)	5	10	15	120
Thickness (*mm*)	3.78	3.74	3.76	3.77

Table 2 - Tests sample dimensions

Test	L (*mm*)	W (*mm*)	Number of specimens
ILSS	38	19	5
Compression	110	10	5

Based on the number of layers used in this experimental study (23), the reductions in terms of debulking process time are 95.8%, 91.7% and 87.5% (UC1, UC2 and UC3 respectively). The total debulking time of the classic procedure can be illustrated with equation Equation 1:

$$T = t * (\frac{n}{3})$$

Equation 1

where t is the debulking time of each cycle and n is the number of layers. Considering the manufacturing conditions of this work, the equation of UC debulking time is:

$$T_{UC} = t_{UC} * (\frac{n}{23})$$

Equation 2

where t_{UC} is the optimal exposure time (5, 10 or 15 minutes) of the process to consolidate 23 plies, that will be identified via experimental campaign. It is possible to calculate the time reduction as follow:

$$T_{UC} = \frac{t_{UC}}{t} * \left(\frac{3}{23}\right) * T$$

Equation 3

3. Experimental Setup

In order to investigate the mechanical properties of the different samples and evaluate the influence of inner porosity on mechanical properties in function of the UC time of process, two experimental tests were performed.

3.1. Compression

In order to evaluate in-plane compressive properties, compression tests were carried out according to the standard BS-EN-ISO-14126:1999. The samples were placed into a dedicated fixture (Figure 5) in order to avoid bending and buckling and guarantee a uniform uniaxial compressive load. The sample was inserted into the fixture and symmetrically clamped on both edges using two steel blocks and eight bolts (four for each edge) to guarantee a good grip during the test. Two axial rods, parallel to the sample axis, were used as guiderail to ensure the correct positioning of the blocks. Afterwards, the assembly was placed on a Universal testing machine Instron 5585 and tested with a cross-head speed of 1mm/min.

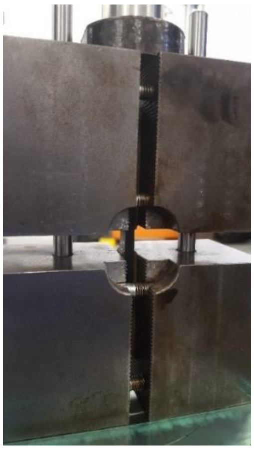


Figure 5 - Compression-dedicated fixture used during experimental campaign tests

The compressive strength σ_c was then obtained using the formula:

$$\sigma_c = \frac{F_c}{A}$$

Equation 4

where F is the maximum compressive load (N) applied during the test and A is the cross-section of the sample (mm^2).

3.2. Interlaminar shear strength (ILSS)

In order to evaluate the interlaminar mechanical properties of CFRP parts obtained with different time of exposition to UC, ILSS tests were carried out using a Universal testing machine Instron 3369, according to BS EN ISO 14130:1998. Two steel rollers (4 mm in diameter) were used as supports for the CFRP samples with a span of 19mm (span-to-ratio 5:1) and a steel roller of 10mm was used as loading nose to apply the transverse load. A sketch of the test setup is reported in Figure 6.

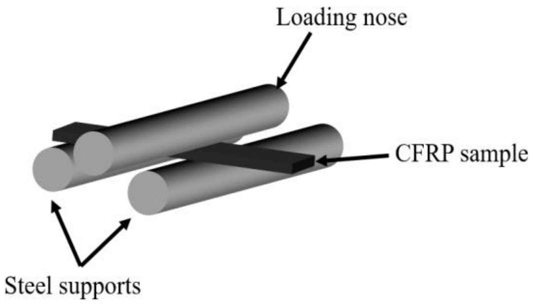


Figure 6 – Sketch of ILSS test setup

In order to calculate the interlaminar shear stress τ (MPa) for ILSS test, the following equation is used:

$$\tau = 0.75\frac{F}{bh}$$

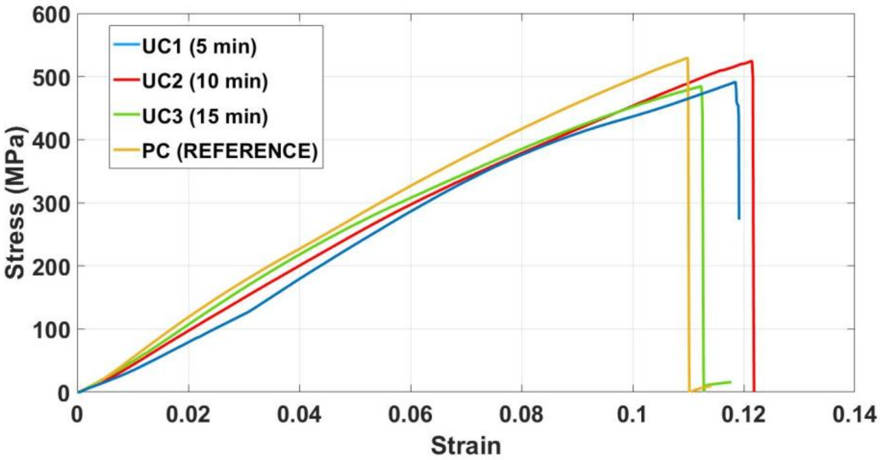
Equation 5

Where b is the width (mm) of the sample, h is the thickness (mm) and F is the applied load (N). To obtain the interlaminar shear strength τ_s (MPa), the maximum force value recorded during the test is used.

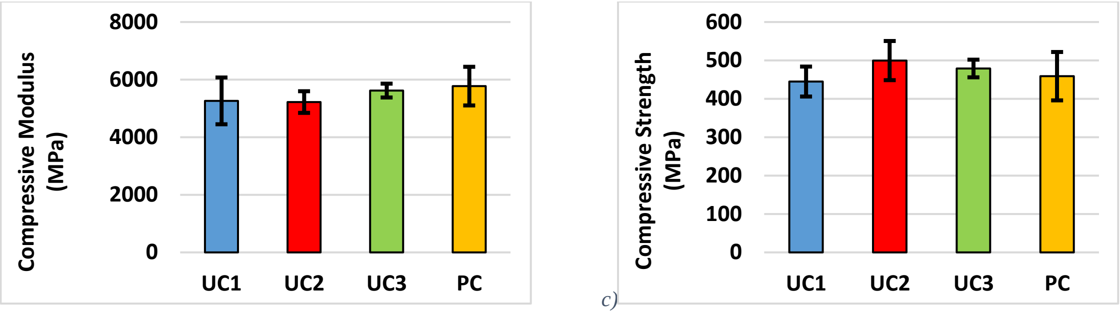
4. Results and discussion

Results of compression tests are reported in Figure 7 where stress-strain curves are reported with relative mean and standard deviation on compressive strength and compressive modulus.

Analysing the experimental data from Figure 7, it is possible to notice that the UC samples presented a variation of -9%, -10% and -3% for the compressive modulus and -3%, +9% and +4% for compressive strength (UC1, UC2 and UC3, respectively) in comparison with the reference samples.



a)



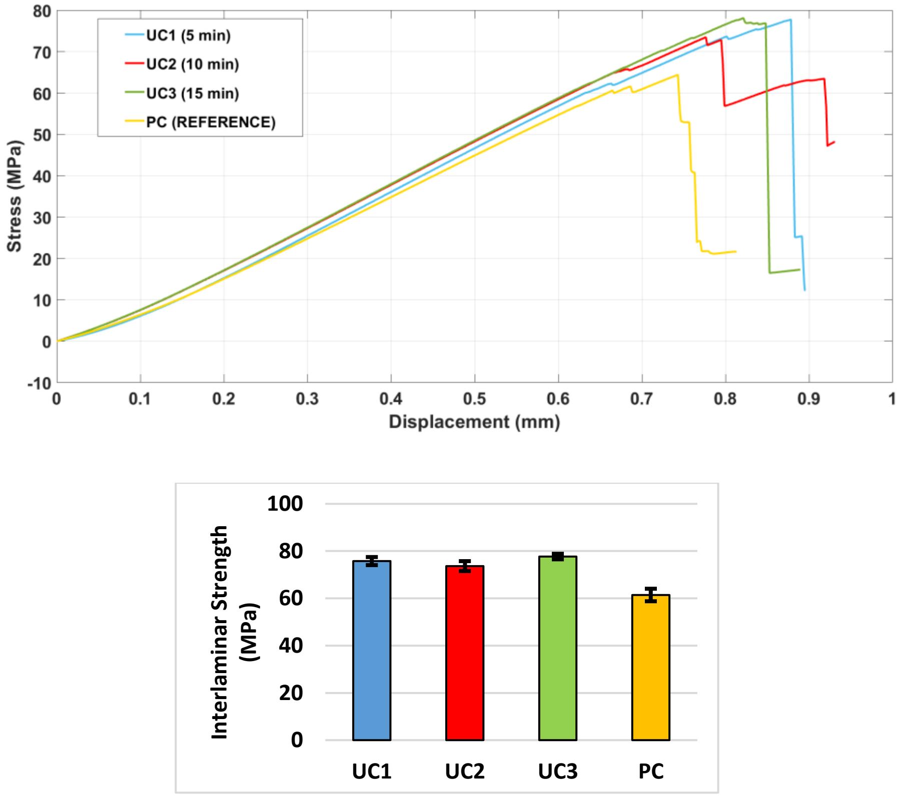
b) *c)*

Compression	Compressive modulus (*MPa*)			Compressive strength (*MPa*)		
Sample	Mean	Standard Deviation	% variation	Mean	Standard Deviation	% variation
UC1	5262	812	-9%	445	39	-3%
UC2	5221	376	-10%	499.6	51	9%
UC3	5620	240	-3%	479	23	4%
PC	5775	671	0%	459	63	0%

d)

Figure 7 - Plots of compression tests results for 3 UC configurations and reference: a) stress-strain curves, b) compressive strength, c) compressive modulus and d) relative statistical data and percentage variation in comparison with reference (PC)

Results on ILSS are reported in Figure 8 where stress-displacement curves are displayed reporting mean and standard deviation of interlaminar strength in bar plot.



a)

b)

ILSS	Interlaminar strength (*MPa*)		
Sample	Mean	Standard deviation	% variation
UC1	75.78	1.71	23%
UC2	73.65	2.1	20%
UC3	77.69	1.18	26%
PC	61.42	2.65	0%

c)

Figure 8- Plots ILSS results: a) stress-displacement curves, b) interlaminar strength graph with standard deviation bars and c) relative statistical data and percentage variation in comparison with reference (PC).

The results from the ILSS tests showed excellent interlaminar properties for all the UC samples, with an increase of 23%, 20% and 26% (UC1, UC2 and UC3 respectively) in ILSS in comparison with the reference and small values of standard deviation.

Considering all the experimental results, it is possible to analyse the effect of UC on the mechanical properties of the samples obtained with the three different exposition times. UC3 showed similar compressive properties when compared to reference ones with a very low percentage variation (under 5%). Similarly, UC1 and UC2 showed good results, but with higher percentage variations (up to 10%). Anyway, analysing the standard deviation and error bar plot of these results, it is possible to state that these variations fall in the statistical errors. On the other hand, the increment in terms of ILSS of all the UC samples compared to reference ones is clear (above the 20%), confirming the effective reduction of voids between layers due to the UC process [9]–[13]. It is important to highlight that this test presented relative low values of standard deviation. This is probably due to the strong dependency of the ILSS properties from laminate matrix characteristics and inner porosity. On the other hand the compressive test have a higher dependency on different experimental variables, including position of the laminates during the autoclave cure, position of the sample cut along the laminate, samples thickness, and others. Although this dependency leads to higher standard deviation values, the UC3 results showed a very good match with reference ones, with both positive and negative variations under 5%. Based on this, it is possible to consider 15 minutes (UC3) as enough UC process time to reach a considerable increment in terms of ILSS and thus of consolidation without affecting the compressive properties. The reduction in terms of debulking time can be estimated using equation Equation 3 substituting $t_{uc} = 15\ min$ and $t = 15\ min$:

$$T_{UC} = 0.13 * T \qquad \text{Equation 6}$$

5. Conclusions

In this work, ultrasonic consolidation was studied and used to improve the debulking step for hand lay-up of autoclave cure prepregs and decrease the manufacturing process time. Debulking is a considerable time-consuming process usually performed using vacuum-bag cycles of 15 minutes applied after the lay-up of every three prepreg layers. An ultrasonic vacuum bag system was used on three uncured CFRP laminates, laid without the traditional vacuum debulking steps. UC was applied for three different times for the three laminates: 5, 10 and 15 minutes (UC1, UC2 and UC3 samples respectively). The resonant frequency of the ultrasonic transducer was set to low levels (in the range of the common UC frequencies) in order to avoid the generation of high levels of frictional heat between the polymer molecules, and thus the cure of the matrix. The procedure led to a consolidation time reduction from the 120 minutes of the traditional process down to 5 minutes for the laminate analysed.

In order to evaluate the reliability of the debulking process, the mechanical properties of the samples were experimentally studied and compared with a reference laminate, manufactured with traditional debulking steps. The ILSS property is particularly effective for the study of voids content between layers in composite laminates. The results from this experimental test showed an increment of ILSS for all the three UC samples of at least 20% in comparison with the reference ones. Moreover, compressive properties were experimentally studied. Results showed a very good match with reference for UC3 sample, with percentage variations under ±5%.

In conclusion, the results confirmed the reliability of the proposed process for the consolidation and debulking of prepregs, with a substantial reduction in terms of time of hand lay-up manufacturing process (over 85%) and large improvements in terms of interlaminar properties.

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