EFFECT OF AUTOCLAVE CURE CYCLES ON THE INTERFACIAL PROPERTIES OF COMPOSITE LAMINATES

Lihua Zhan[1,2 *],Wei Tan[1,2,3], Tengfei Chang[1,4], Xingxing Ding[1,2]

[1] State Key Laboratory of High Performance Complex Manufacturing, Central South University, Changsha, 410083, P.R.China
[2] School of Mechanical and Electrical Engineering, Central South University, Changsha, 410083, P.R.China
[3] School of Mechanical and Aerospace Engineering, Queen's University Belfast, Belfast, BT9 5AH, United Kingdom
[4] Light alloys research institute, Central South University, Changsha410083, P.R.China

* Corresponding Author: L. Zhan (yjs-cast@csu.edu.cn)

Keywords: Composite manufacturing, autoclave curing, interfacial properties, characterization

Abstract
Autoclave curing of reproducible and high quality composite aerostructures with carbon fibre prepregs is often compromised by high costs, long development time, and poor quality due to multiple defects, particularly in massive complex parts. The consolidation cycle of the prepregs, accompanied by curing reaction and rheological change in the thermoset resin, significantly influences the fibre/matrix interfacial properties which determine the mechanical behaviour of composite structures. In order to tailor composites to achieve desired performance, the effect of manufacture process on the interfacial properties and structural performance is required to be investigated. In this work, single fibre push-in tests of unidirectional composite specimens consolidated under different pressure was conducted to characterize the interface bonding strength at the micro-scale level. Interlaminar shear strength of these samples which describes the bonding quality at the meso-scale level was evaluated by a short-beam three point bending test. An empirical model to describe the influence of different consolidation parameters on both micro-scale and meso-scale level interfacial properties of composite laminates was presented. This will greatly enhance the connections between manufacture process and design of composite structures.

1. Introduction

Carbon fibre reinforced composites material are nowadays widely used in aero-structural components due to their high specific stiffness and strength. For high-performance elements, manufacturing is carried out using prepreg laminates, which are stacked (either manually or with an automatic lay-up machine) and consolidated under the simultaneous application of pressure and temperature in an autoclave. Autoclave pressure impedes the growth of voids or leads to the collapse of air bubbles, giving rise to materials with excellent mechanical properties and very low porosity, as required by aerospace and automobile industries. However, they are often compromised by high costs, long development time, and poor quality due to multiple defects, particularly in massive complex parts.

The consolidation cycle of the prepregs, accompanied by curing reaction and rheological change in the thermoset resin, significantly influences the interlaminar and fibre/matrix interfacial properties which determine the mechanical behaviour of composite structures. There is still a lack of research regarding the multi-level influence of curing conditions on the fibre/matrix interface, interlaminar and structural performance of composite laminates.

Excerpt from ISBN 978-3-00-053387-7

One of the most significant phenomena in composite materials for applications to load bearing primary structures is the stress transfer between the fibre and matrix across the interface when the composites are subjected to various loading conditions. Various forms of microcomposite tests as a means of evaluating the bond quality has been conducted to understand the stress transfer at the fibre-matrix interface region. Since the in-situ condition of fibre/matrix interface is affected by different fibre packing density and polymer morphology cross-link density, single fibre fragmentation test [1] and microbond test [2] are not representative of the mechanical and physical-chemical condition of actual composites. Medina M et al. [3] demonstrated that the interfacial shear strength (IFSS) measured by the push-in and the push-out tests were very close for the materials analysed and the differences were always within the experimental scatter. These results indicate that both methodologies are valid to measure the interface properties. Compared to fibre push-out test that requires labour intensive preparation of a very thin membrane (40 μm ~ 60 μm), push-in test to push individual fibres on the cross-section of a bulk specimen is much easier to implement and can offer more stable and reproducible results. Therefore, push-in test was used to investigate the influence of cure pressure on the interfacial properties in this work.

In this work, single fibre push-in tests of unidirectional composite specimens consolidated under different pressure was conducted to characterize the interface bonding strength at the micro-scale level. Interlaminar shear strength of these samples which describes the bonding quality at the meso-scale level was evaluated by a short-beam three point bending test. Optical microscopy and scanning electron microscopy was used to assessment the manufacture quality and fracture surface of failed short-beam respectively. An empirical model to describe the influence of different consolidation parameters on both micro-scale and meso-scale level interfacial properties of composite laminates was presented. This will greatly enhance the connections between manufacture process and design of composite structures.

2. Material and methods

2.1. Sample preparation

The fibre reinforced composite used in this study was manufactured from unidirectional carbon fibre (T800) reinforced/epoxy (X850) prepregs. Pre-impregnated sheets were supplied by Commercial Aircraft Co., Ltd., China with fibre volume fraction of 65% and areal density of 190 g/m^2. Cytec's CYCOM X850 resin system, as the next generation epoxy resin for aerospace industry, is formulated with the most advanced epoxy chemistry available to provide good heat resistance, high specific strength and adhesion properties.

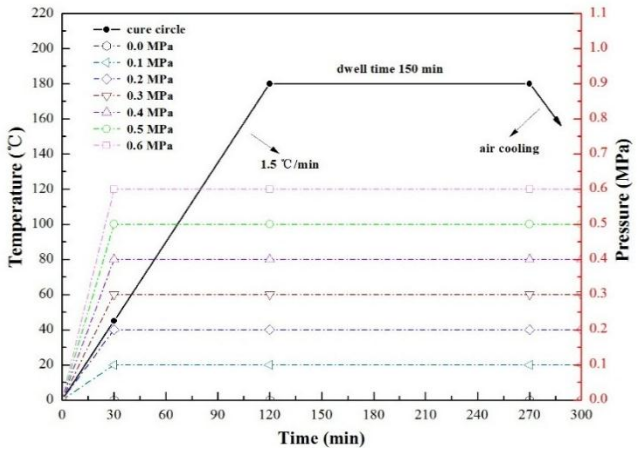


Figure 1. Consolidation cycles to process T800/X850 composite prepregs

Excerpt from ISBN 978-3-00-053387-7

Eight unidirectional $[0]_{10}$ composite laminates were fabricated using autoclave process by applying different kinds of external pressures. Pressures were set as 0.0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6 and 0.8 MPa respectively. For the consolidation heating profile, laminate plates were heated at a constant rate of 1.5 ℃/min until 180 ℃ and was held for 150 min, after which the samples were cooled down to room temperature under the same pressure, as shown in Figure 1. The designed laminates dimension was 200 mm (length) ×200 mm (width) ×2 mm (thickness), and the final cured thickness of composite laminates were 2.0 ± 0.2 mm.

2.2. Test setup

2.2.1 Characterization of fibre/matrix interfacial properties

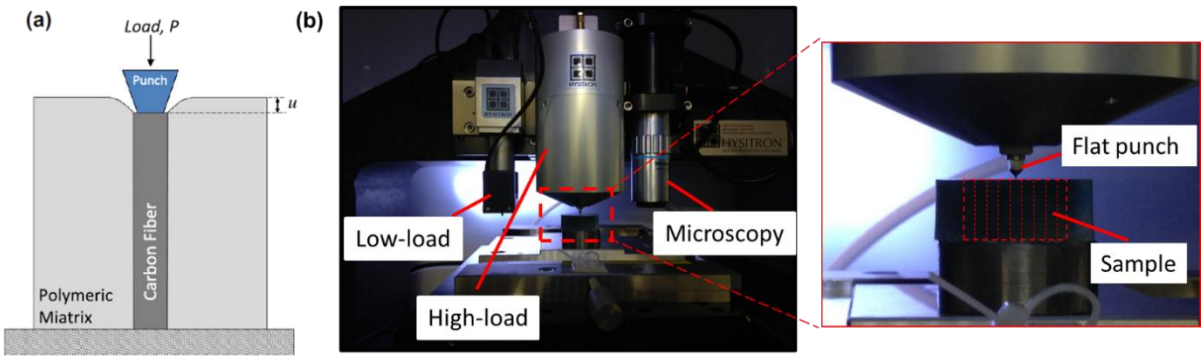


Figure 2. (a) Schematic of the fibre push-in test [4] (b) Experimental setup with the Hysitron TI950 Triboindenter

In order to understand influence and mechanisms of cure pressure on the material properties. Push-in tests were conducted by the Hysitron TI 950 triboindenter instrument with a diamond flat punch of 5 μm in diameter. Micromechanical tests were performed under displacement control at 40 nm/s with the maximum displacement of 1200 nm and then unloading was conducted to the original position. The tests were performed on over 15 fibres per sample groups. The fibres were tested with a typical hexagonal packings formed by one fibre surrounded by its six nearest neighbors, to obtain an average interfacial shear strength (IFSS) value τ_{SL}.

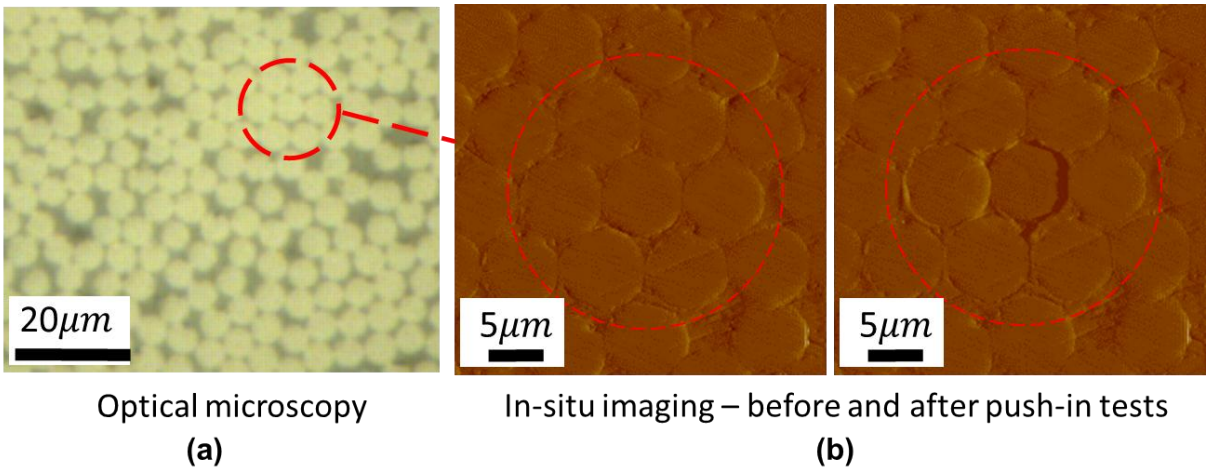


Figure 3 . (a) Optical micrograph of composite cross-section (b) In-situ imaging showing the detail of one hexagonal packing before and after fibre push-in tests

The overall tests were conducted following these procedures: first of all, the sample was inspected under

Excerpt from ISBN 978-3-00-053387-7

the optical microscopy to find the position of typical hexagonal fibre packing patterns. Afterwards, the sample was moved beneath the low-load with maximum load of 120 mN indenter probe. The in-situ images are obtained by raster scanning the low-load indenter probe over the sample surface to allow for precise test placement and microstructure identification. Afterwards, the sample was tested under the high load indenter probe with maximum load of 950 mN. As shown in Figure 2b, post-test imaging also provides the ability to verify that the test was performed in the desired location. This maximizes the reliability of data and aids in the explanation of unexpected test results.

The push-in test is performed by loading an individual fibre within the composite until interface fracture occurs. The load–displacement curve ($P-u$) presents an S shape, and the initial region corresponds to an imperfect contact between the indenter and the fibre. This is followed by a linear region (with slope S_0) due to the elastic deformation of the fibre and the matrix, which is followed by a non-linear region due to the onset of interface failure. The IFSS can be determined from the critical load P_c at the onset of interface failure through the shear-lag model [5], which is determined from two parallel lines with the initial stiffness S_0 drawn with offset displacement of 2% and 10%. This definition provides a good indication of the crictical load at the initiation of debonding as follow,

$$\tau_{SL} = \frac{nP_c}{2\pi r^2} \tag{1}$$

Where n is a parameter that depends on the elastic properties of the fibres and the matrix and local fibre patterns and volume fraction. n can be determined from the slope of the $P-u$ curve in the linear region, S_0, according to:

$$n = \frac{S_0}{\pi r E_f} \tag{2}$$

Where E_f is the longitudinal elastic modulus of the fibre modulus.

2.2.2 Characterization of interlaminar shear strength

The interlaminar shear strength were determined according to ASTM standard D2344/D2344M [6] using three point bending method. The sample was taken of The mechanics performance test was conducted on CMT5105 tensile testing apparatus (produced by Sansi Taijie Co., Ltd., China) and three tests for each group of samples were performed under displacement control at a constant crosshead rate of 1 mm/min. The specimens were cut by diamond saw to nominal dimensional of 20 mm×10 mm (length×width). The interlaminar shear strength of the composite samples was determined according to equation:

$$\tau_{ILSS} = \frac{3}{4}\frac{P_{max}}{bh} \tag{3}$$

Where P_{max} is the maximum load (N), b is the sample width (mm) and h is the sample thickness (mm).

2.3. Optical microscopy and SEM

For the optical microscopy inspection and push-in tests, samples consolidated at different cure pressures were cut by a precision diamond saw to nominal specimen dimensions of 10 mm×10 mm. Samples were embedded in epoxy resin to facilitate handling during polishing. Surfaces perpendicular to the fibres were polished with a sequence of silicon carbide papers of 600, 1000, 2000, and 4000, and finished with polishing diamond pastes of 0.3 μm and 0.1 μm, followed by the ultrasonic cleaning. The optical digital microscope and metallurgical microstructure image analysis systems were employed to study the voids distribution of composite laminates. The fracture surfaces of the tested samples were examined by scanning electron microscopy (SEM) to investigate the interfacial failure mechanisms.

Excerpt from ISBN 978-3-00-053387-7

3. Result and discussion

3.1. Manufacture quality assessment

The final plastograhpic of each specimen under different cure pressure is illustrated in Figure 4, clearly indicating that the volume fraction of voids or porosity decreases as the cure pressure increases. Voids may be created as the results of different parameters, such as entrapped air during resin impregnation or layup, and volatiles arising from the resin system during cure. The distribution of voids is mainly found in the interface between plies, while a small amount of voids locate at the intraply area. It is also evident that the porosity is almost negligible when cure pressure is higher than 0.4MPa. This is because high-pressure processing during the cure cycle encourages voids to dissolve and disperse within the matrix, resulting in low-void content and high-fibre volume fraction. The specimen consolidated without cure pressure also shows an extensive wrinkle deformation of individual ply.

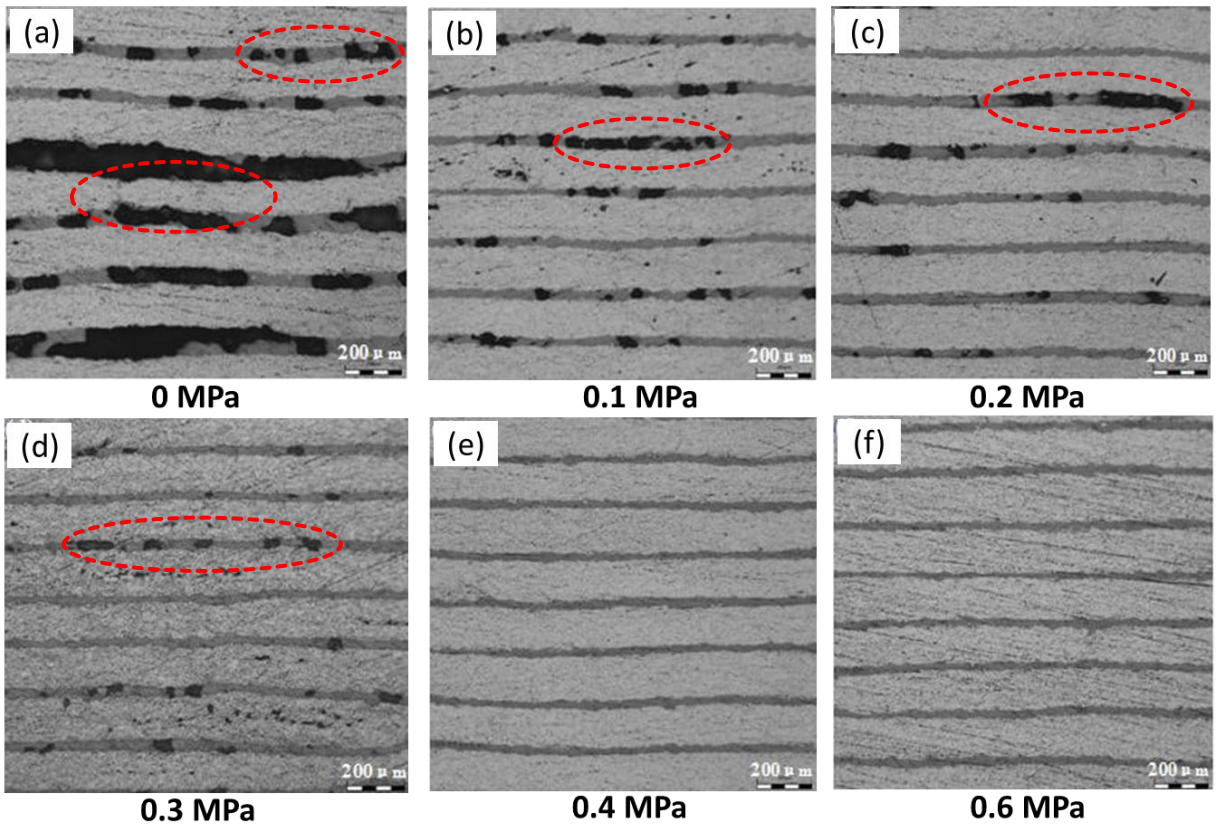


Figure 4. Void distribution under different cure pressure

3.2. Fibre/matrix interfacial shear strength

The representative load–depth curves obtained during the push-in tests for each sample under different cure pressures are shown in Figure 5a. All of them present the same initial elastic stiffness, followed by a non-linear region which indicates the propagation of the interface decohesion from the upper surface into the bulk matrix. The difference between the non-linear regimes of these curves demonstrated that critical load increased with increasing cure pressures. For this hexagonal fibre patterns, the interfacial strength can be considered approximately proportional to the critical loads. Hence, these curves may indicate the same trends of interfacial strength.

The interfacial shear strength (IFSS) was calculated from the experimental results according to Equation (1) and is plotted in Figure 5b as a function of cure pressure. It is shown that a high cure pressure leads

Excerpt from ISBN 978-3-00-053387-7

to a substantial growth of IFSS. Interfacial strength has increased by 43% after reaching cure pressure of 0.8 MPa. The relationship between IFSS and cure pressure could be rationalized with a very simple model using polynomial fitting, which is given by equation as follows,

$$\tau_{IFSS} = 43.55\,Y^3 - 66.23Y^2 + 55.75Y + 56.685 \quad (4)$$

Where Y is the applied cure pressure, τ_{IFSS} is the interfacial strength calculated by shear-lag model.

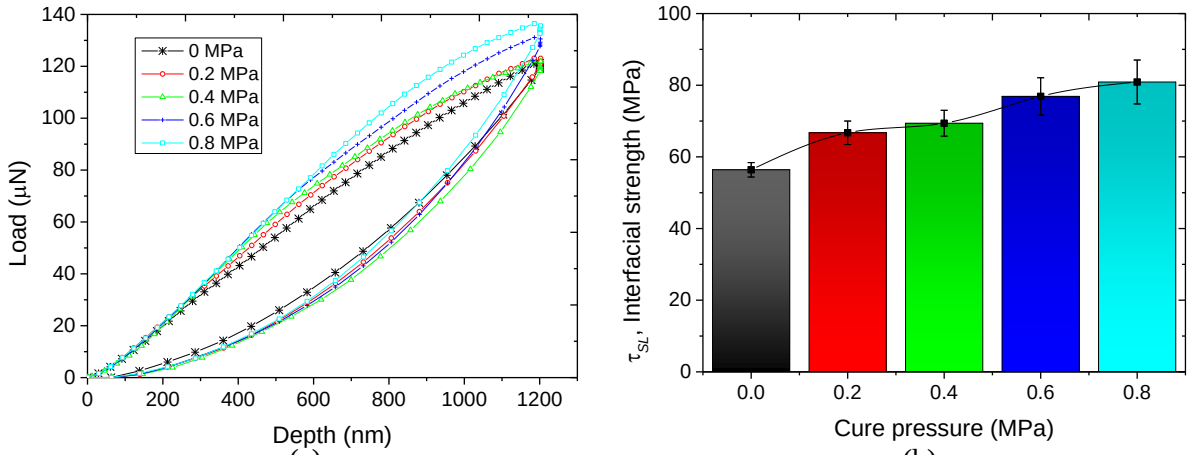
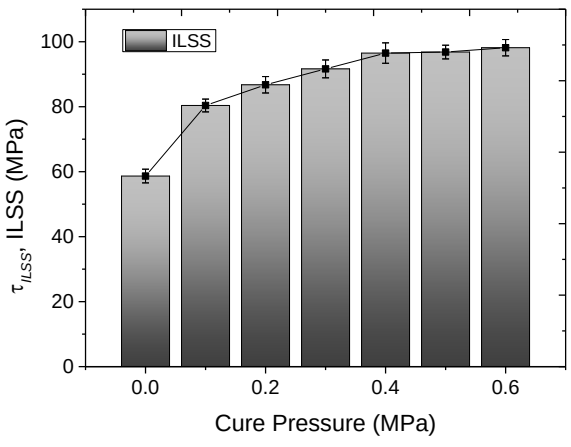


Figure 5. (a) Load-indentation depth curves and (b) fibre/matrix interfacial strength vs. pressure

3.3. Interlaminar shear strength

3.3.1 Characterization of interlaminar shear strength

As shown in Figure 4 and Figure 6, porosity leads to a marked reduction in the composite mechanical properties, particularly those dominated by the matrix behavior like the interlaminar shear strength. Laminates with the highest porosity (0 MPa) showed the lowest ILSS, while those with the minimum volume fraction of voids (0.4-0.6 MPa) presented the highest ILSS. It is evident that the porosity of laminates cured with pressure higher than 0.4 MPa is almost negligible. Once the porosity is almost zero (0.4 MPa and 0.6 MPa), the ILSS reaches a plateau. This confirms the ILSS is mainly controlled by the void volume fraction. Since voids are one of the most influential defects in the composite material as they weaken the matrix-dominated mechanical properties, such as shear and interface bonding strength, by inducing localized stress concentration. Therefore, high-pressure cure cycles which lead to the minimum porosity results in the best mechanical behavior. After ILSS reaches a plateau, the ILSS is controlled by both interlaminar and fibre/matrix interface properties. It should be noted that the increasing IFSS with cure pressure from 0.4 MPa to 0.6 MPa is not affecting the ILSS.



Excerpt from ISBN 978-3-00-053387-7

Figure 6. Interlaminar shear strength vs. cure pressures

The relationship between ILSS and cure pressure could be rationalized with a very simple model using polynomial fitting, which is given by equation as follows,

$$\tau_{ILSS} = 369.26\,Y^3 - 489.83Y^2 + 225.91Y + 59.63 \quad (5)$$

Where Y is the applied cure pressure, τ_{ILSS} is the interlaminar strength calculated by Equation (3).

3.3.2 Failure mechanisms

SEM images in Figure 7 show the typical resin cusp structures (platelets inclined on the surface), indicative of fracture by shear along the fiber direction. Cusps are formed as successive, parallel microcracks initiated by shear in the epoxy matrix along the main crack propagation direction.

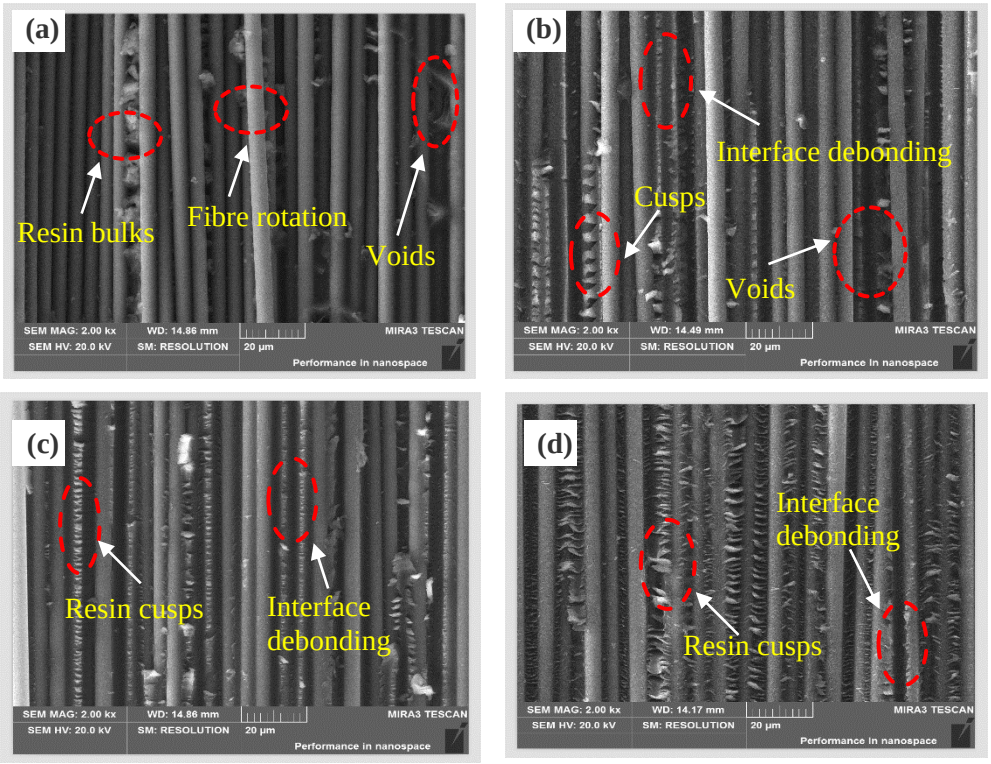


Figure 7. Scanning electron micrograph detail of fracture surfaces of specimens for interlaminar shear strength tests

Figure 7a shows the surface cured under autoclave pressure of 0 MPa, where the surface of fibres is smooth and gap between fibres is evident, indicating that the adhesion between fibre and matrix is very weak. Some fibres are even inclined and rotated due to the lack of support from matrix. The resin rich pocket and voids is frequently observed, which means impregnation of resin with the laminar is poor and inhomogeneous, leading to weak fibre/matrix interfacial and interlaminar strength. SEM image in Figure 8b illustrated the typical interface debonding under shear loading, the fibre from the upper layer is debonded from the matrix in the adjacent lower layer by mode II shear loading along the fibre direction. There is some resin remained on the surface of fibres after debonding, while voids are still presented in this sample due to inadequate cure pressure.

The fracture surfaces in Figure 8c and Figure 8d is showing no distinguish difference with resin cusps and interface debonding. Increasing pressure leads to more resin cusps areas. Upon the application of pressure, most of the load was transferred through a continuous skeleton of fiber-rich regions. The higher

Excerpt from ISBN 978-3-00-053387-7

pressure in these regions led to the migration of resin as well as voids. These SEM images confirms the cure pressures drive the resin flow to impregnate the fibre. The higher cure pressure will lead to better impregnation performance and hence facilitated the coalescence and elimination of voids. At the same time, it will improve the adhesion between fibre and matrix and corresponding interfacial strength (IFSS) and interlaminar strength (ILSS). Nevertheless, pressure should not be too high to avoid the excessive resin loss/bleed out and fibre wrinkles. For manufacturing high-quality composite components, cure pressure between 0.6 MPa and 0.8 MPa is recommended.

4. Conclusions

Unidirectional laminates of a T800/X850 polymer matrix composite were manufactured by autoclave consolidation under the same temperature cycles with different cure pressure ranging from 0 MPa to 0.8 MPa. A single fibre push-in tests of unidirectional composite specimens consolidated under different pressure was conducted to characterize the interface bonding strength at the micro-scale level. Interlaminar shear strength of these samples which describes the bonding quality at the meso-scale level was evaluated by a short-beam three point bending test. Both interfacial strength and interlaminar strength shows a substantial growth with increasing cure pressure. Optical microscopy and scanning electron microscopy confirms the consistency in terms of the relationship between porosity and mechanical behaviour, indicating high-pressure cure cycles which lead to the minimum porosity results in the best mechanical behaviour. An empirical model to describe the influence of different consolidation parameters on both micro-scale and meso-scale level interfacial properties of composite laminates was presented. This work greatly contribute the understanding of cure effect on the interfacial properties from different length scale, providing the basis for multi-scale modelling of composite processing. It also enhances the connection between manufacture process and design of composite structures.

Acknowledgments

The authors would like to gratefully acknowledges the funding from the Major State Basic Research Development Program of China (973 Program) under Grant No. 2014CB046502. The authors would also like to gratefully acknowledge Prof. Javier Llora, Dr. Jon Molina, Mr. Lingwei Yang of IMDEA Material Institute for their support to conduct nano-indentation tests and useful discussions in this research.

References

[1] A.C. Johnson, S.A. Hayes, F.R. Jones, The role of matrix cracks and fibre/matrix debonding on the stress transfer between fibre and matrix in a single fibre fragmentation test, Compos. Part A 43(1) (2012) 65-72.
[2] U. Gaur, B. Miller, Microbond method for determination of the shear strength of a fiber/resin interface: evaluation of experimental parameters, Compos Sci. Technol. 34(1) (1989) 35-51.
[3] C. Medina M, J.M. Molina-Aldareguia, C. Gonzalez, M.F. Melendrez, P. Flores, J. Llorca, Comparison of push-in and push-out tests for measuring interfacial shear strength in nano-reinforced composite materials, J. Compos. Mater. (2015).
[4] M. Rodr ́guez, J.M. Molina-Aldaregu á, C. Gonz ́alez, J. Llorca, A methodology to measure the interface shear strength by means of the fiber push-in test, Compos Sci. Technol. 72(15) (2012) 1924-1932.
[5] M. Zidi, L. Carpentier, A. Chateauminois, F. Sidoroff, Quantitative analysis of the micro-indentation behaviour of fibre-reinforced composites: development and validation of an analytical model, Compos Sci. Technol. 60(3) (2000) 429-437.
[6] A.T. Standard, D2344/D2344M-00e1,“, Standard Test Method for Short-Beam Shear Strength of Polymer Matrix Composite Materials and Their Laminates,” ASTM International, West Conshohocken, PA (2000).

Excerpt from ISBN 978-3-00-053387-7