Effect of Cure Pressure on Void Content and Interlaminar Shear Strength of Interlayer Toughened Composite Laminates

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ABSTRACT

There is increasing interest in cure cycle optimization for composite materials to increase production rates and lower the cost in the aerospace industry. However, modified cure cycles may have a detrimental effect on physical and mechanical properties and there is a need to understand the effect of cure cycle parameters on microstructures and properties of composites. It is known that cure pressure has a significant effect on void content and mechanical properties of untoughened composites. This paper evaluates the effect of cure pressure and the role of interlayer particles on the void content and interlaminar shear strength of an interlayer toughened prepreg system, 3900-2B/T800SC, at different heating rates.

KEYWORDS: toughened composites, porosity, interlaminar shear strength

1 INTRODUCTION

Cure cycle optimization for composite materials aims to reduce the residual stress and manufacturing defects (Web-1) [1-2]. Additionally, the increasing interest in improving the production rate and lowering the manufacturing cost for the aerospace industry also requires faster cure cycles and broader processing windows of composite materials [3]. However, the material manufacturers usually provide narrow process windows for their material systems, and the Original Equipment Manufacturers (OEM) have strict specifications for thermal management of composites parts. This is because modified cure cycles may cause defects, induces residual stress and change laminate microstructures, and therefore may have a detrimental effect on physical and mechanical properties [4-6]. Composite materials used in the aerospace industry are mainly processed in the autoclave. The fundamental advantage of autoclave processing is that the high pressure facilitates gas removal from the laminate, collapses bubbles, and keeps volatiles in solution. However, autoclave processing is expensive in terms of equipment purchase and usage. To further lower the manufacturing cost, Out of Autoclave (OOA) composites processing is proposed as an ideal alternative method to traditional autoclave processing [7] (Web-2). The primary challenge of applying the OOA method for manufacturing aerospace composites is to meet the strict requirements of void content, which is usually required to be less than 1 or 2% for aerospace applications (Web-3). It is known that cure pressure has a significant effect on void content and mechanical properties of non-interlayer toughened composites [8-9]. Sources and sinks of porosity for non-interlayer toughened composite material have been identified and studied extensively [10-12]. Interlayer toughened composite materials such as 3900-2B/T800SC have been used in primary structures of the commercial aircraft for their high delamination and impact resistance. The aerospace industry is looking to OOA processing to replace autoclave processing for interlayer toughened composite materials. However, the effect of cure pressure and the role of interlayer particles on void content and mechanical properties of the interlayer toughened composites has not been studied. The work presented in this paper is focused on experimentally investigating the effect of cure pressure on void content and interlaminar shear strength (ILSS) of an interlayer toughened prepreg system (3900-2B/T800SC) at different heating rates. There are two critical questions answered in this paper. First, can 3900-2B/T800SC be cured with lower autoclave pressure or without pressure at the high heating rate? Second, what is the role of toughening particles and the toughened interlayer on void content?

2 EXPERIMENTAL

2.1 Material and manufacturing

The material used in this study is an interlayer toughened UD tape prepreg, Torayca® 3900-2B/T800SC, from which laminates with stacking sequence $[0]_{16}$ were made. Thermoplastic particles are dispersed in the interlayer to improve the delamination and impact resistance. The square laminates were prepared for present experimental investigation with a dimension of 150 mm× 150 mm× 3.1 mm. A rubber dam was placed around the laminates to prevent the resin bleed. A flat aluminum caul plate was placed on the top of the laminates to obtain similar surface quality at both sides of the laminates. The part temperature was recorded by the thermocouple inserted in the middle of the laminates. Full vacuum was applied under the vacuum bag throughout the cure cycle. Composites laminates were processed in an autoclave with different autoclave pressures and heating rates. In this paper, the cure pressure is equivalent to the autoclave pressure. After the curing process, laminate thicknesses at different locations of the cured laminates were measured by a caliper. Table.1 presents the curing parameters of eight cure cycles with similar final Degree of Cure (0.90-0.92). The final DoC was calculated by the cure kinetics model in the Raven software (Convergent Manufacturing Technologies). The manufacturer's recommended thermal cycle for 3900-2B/T800SC is to apply a temperature ramp to 177 ± 5 °C at a rate of 1.7 ± 1.1 °C/min, holding the cure temperature at 177 ± 5 °C for 120 -180 minutes and then cooling down to 60°C or lower at a maximum rate of 2.78 °C per/min. Cycle C-2 is the manufacturer's recommended cure cycle (MRCC) and Cycle C-1 is the same thermal cycle as C-2 but without autoclave pressure. The edge and center of the laminates processed by C-2 are denoted as P1 and P2. C-5 is also a MRCC although with the maximum heating rate allowed. Cycle C-3, C-4, C-5 were used to study the effect of pressure on void content with the maximum heating rate (2.78 °C/min) allowed by the MRCC. Cycles C-6, C-7, C-8 were used to evaluate the effect of pressure on void content with a faster heating rate (7.0 °C/min), above the maximum heating rate allowed.

Table 1 Cure cycles and void content

Cure cycle	Autoclave pressure (kPa)	Heating rate (°C/min)	Hold temperature (°C)	Hold time (min)	Average ply thickness (μm)	Average void content (%)	Standard Deviation (%)
C-1	0	1.0	180	120	194.1	0.51	0.30
C-2	586	1.0	180	120	--	0	0
C-3/P1	0	2.78	180	120	191.8	2.39	0.83
C-3/P2	0	2.78	180	120	193.6	1.56	1.33
C-4	100	2.78	180	120	194.1	0	0
C-5	586	2.78	180	120	192.9	0	0
C-6	0	7.0	180	135	216.6	10.82	1.47
C-7	100	7.0	180	135	193.3	0	0
C-8	586	7.0	180	135	--	0	0

2.2 Ultrasonic C-Scan Inspection

The porosity distribution was determined using ultrasonic C-scan using an Olympus ultrasonic equipment (OmniScan MX2) with phased array flaw detectors and GLIDER™ X-Y Scanner. All laminates were scanned in water. A TomoView software was used for the ultrasonic analysis.

2.3 Microscopic analysis

Optical microscopy (Keyence VHX-1000 digital Microscope) was used to observe the void size and void distribution. The scanning electron microscopy (FEI Quanta 650) is used to determine the failure mode of short beam shear specimens. To increase the conductivity of samples and improve the image quality, a sputter gold coating was applied on the fracture surfaces to form an ultra-thin coating of electrically-conductive metal. Moreover, an electrically-conducting tape was used to bridge the samples with SEM Mount. The morphology is imaged by the secondary electron (SE), at low vacuum condition, and with 14 kV.

2.4 Void content determination

The void content was determined by the density method in accordance with the Test Method A in ASTM D2734-16 [13]. The laminate density was measured by a Precisa EP-125SM semi-micro balance according to ASTM D792-13. Six samples (short beam shear specimen) for each cure cycle were tested. All samples have smooth surfaces and edges. The theoretical composite density is defined as the average density of reference laminates processed by 2.78 °C/min under 586kPa autoclave pressure. Results from optical microscopy were used to confirm that reference laminates were void-free. According to ASTM D 2734-16, void content is calculated by the following equation,

$$V = \frac{100(T_d - M_d)}{T_d} \tag{1}$$

where V is the void content (%), T_d is the theoretical composite density, and M_d is the measured composite density.

2.5 Short beam shear test

Short-Beam Shear (SBS) tests were performed with an Instron 5982 Floor Model Testing Systems (100 kN) using the ASTM D2344M-16 standard [14]. A water-lubricated precision diamond saw (Isomet 400) was used to cut the laminates into small SBS specimens with a dimension of 20 mm× 7.1 mm× 3.1 mm. Six rectangular specimens for each cure cycle were prepared. All specimens were stored in a standard laboratory atmosphere of 23±3 °C and 50±10% relative humidity for seven days and tested at the standard laboratory atmosphere. Specimens were loaded in three-point bending with a loading rate of 1.0 mm/min and a span length of 12.3 mm. The short-beam strength is calculated using the following equation:

$$F^{sbs} = 0.75 \times \frac{P_m}{b \times h} \tag{2}$$

where F^{sbs} is the short-beam strength (MPa), P_m is the maximum load observed during the test (N), b is the measured specimen width (mm), and h is the measured specimen thickness (mm).

3 RESULTS AND DISCUSSIONS

3.1 Cure cycles

Figure 1 presents the temperature profiles for 16-ply 3900-2B/T800SC laminates. Small thermal lags are observed at the end of the temperature ramp with high heating rates (2.78 °C/min and 7.0 °C/min). Thermal lags are smaller at the higher autoclave pressure due to higher convective heat transfer coefficient. Overall, the temperature profiles for the 16-ply laminates processed by the same thermal cycle are close despite the effect of autoclave pressure.

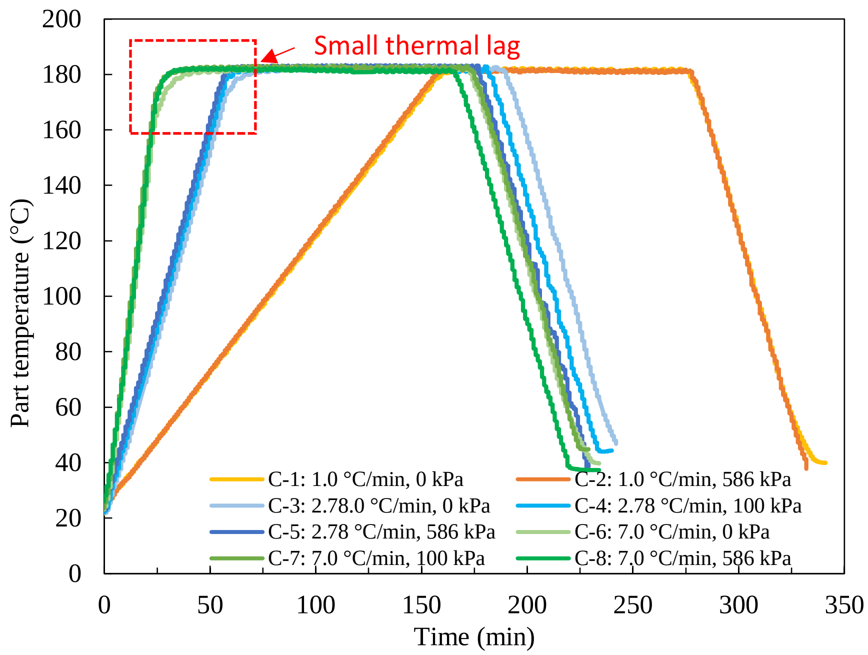

Figure 1: Temperature profiles for 16-ply 3900-2B/T800SC laminates cured with different cure pressures and heating rates

3.2 Void content

Figure 2 presents the ultrasonic C-Scan mapping of voids in the laminates cured with different heating rates and cure pressures. The red zones correspond to low echo from the bottom surface and suggest voids beneath the surface. Figure 2 (a) shows that voids distribute along the fiber direction in the laminates cured with 1.0 °C/min and without pressure. Increasing the heating rate to 2.78 °C/min, more voids are observed at the laminate edge than at the laminate center, as shown in Figure 2 (b). Table 1 shows that the average void content at the laminate edge is 2.39% which is higher than the 1.56% at the laminate center. For the faster heating rate (7.0 °C/min), the void content increases to 10.47% with voids uniformly distributed within the laminates. This is because the resin gels quickly, and thus there is not enough time for void migration. Figure 2 (d-f) shows that all laminates processed under 100 or 586 kPa autoclave pressure are void-free, which agrees with the results by the density method and the microscopic observation at polished cross-sections.

The thickness distributions of the laminates cured with different heating rates and cure pressures are shown in Figure 3. Generally, the laminate edge is thinner than the laminate center due to the resin flow and bleed when the resin is fluid. The average ply thickness of laminates is calculated from laminate thickness. Table 1 shows that the average ply thicknesses of laminates cured with 2.78 °C/min under different cure pressures are close despite that voids are found in the laminates cured without pressure. For the fast heating rate (7.0 °C/min), the average ply thickness of laminates increases from 193.3 μm to 216.6

µm as the pressure increases from 0 kPa to 100 kPa. The polished cross-sections in Figure 4 (g-h) shows that large voids are frozen within the laminates resulting in the ply thickening.

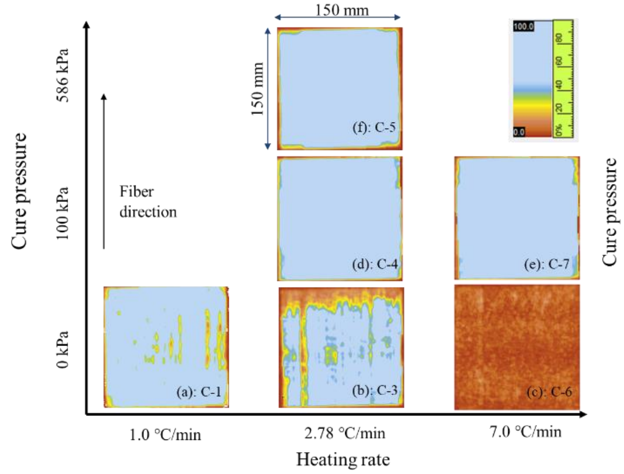

Figure 2: The ultrasonic C-Scan mapping of the void distribution of laminates cured with different heating

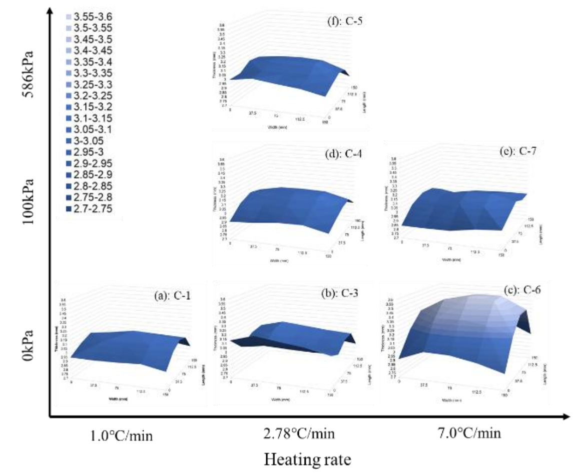

Figure 3: Thickness distribution in laminates cured with different heating rates and pressures

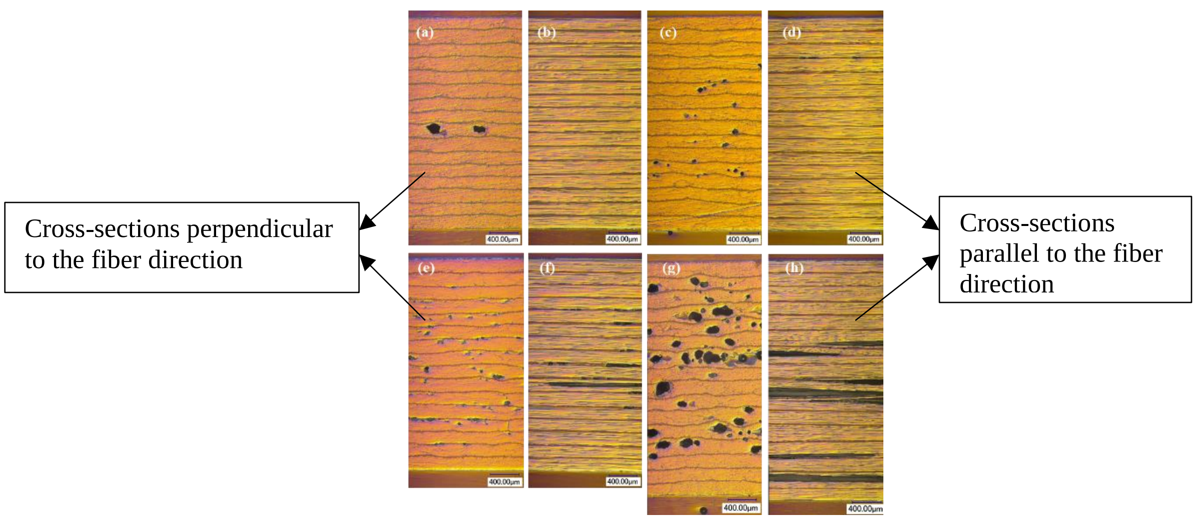

Figure 4: Two different views of void distribution within laminates cured without pressure at different heating rates, (a-b) C-1: 1.0 °C/min, (c-d) C-3/P1: 2.78 °C/min, sample cut from the laminate center, (e-f) C-3/P2: 2.78 °C/min, sample cut from the laminate edge, (g-h) C-6: 7.0 °C/min.

3.3 Void shape and distribution

Figure 4 presents the void shape and the through-thickness void distribution of laminates cured without pressure at different heating rates. In these cases, voids are oval in the cross-section perpendicular to the fiber direction and elongated along the fiber direction. This indicates that the voids mainly migrate along the fiber direction. As shown in Figure 4 (e-f), voids distribution in the intralayer region is higher than those in the interlayer region. These results suggest that interlaminar voids' removal is complicated by the toughening particles in this region, possibly due to their effect on resin infiltration at the interlaminar region. Further, through-thickness void removal also tends to be obstructed by the toughened interlayer, which increases ply waviness, Figure 5 (b). It is worth noting that a correlation can be understood between the ply waviness and void size. The ply waviness is lower at 2.78 °C/min with smaller intralaminar voids, as shown in Figure 5 (a).

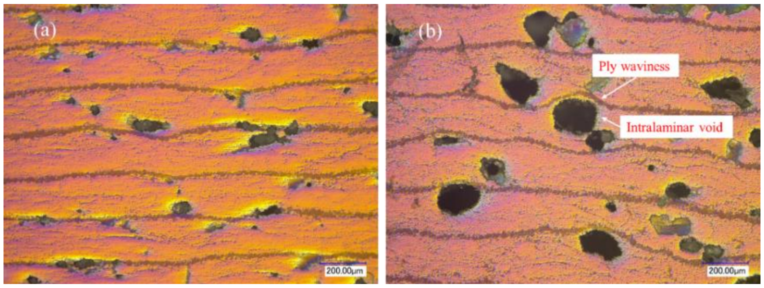

Figure 5: Polished cross-sections of laminates cured with (a) C-3: 2.78°C/min and (b) C-6: 7.0°C/min without autoclave pressure

3.4 Interlaminar shear strength (ILSS)

Figure 6 presents the interlaminar shear strength (ILSS) of laminates cured with different pressures and heating rates. The ILSS is not sensitive to cure parameters if 100-586 kPa autoclave pressure is applied and void-free parts are made. For cure cycle C2, C4, C5 and C8, the average ILSSs are similar, about 102 MPa. Figure 6 shows that porosity generally has a negative effect on the ILSS, and according to the least square fit, the ILSS decreases by 2.51 MPa for each 1% of voids. As shown in Figure 8, for all void-free specimens cured with cycle C-5 and C8, the load-displacement curves rise to the maximum values and then drop. For cycle C-3, the load-displacement curves follow a similar tendency but have lower peak force. For cycle C-6, there is no obvious load drop in the load-displacement curves due to the squeezing of porous laminates under compressive load. Figure 9 shows the SEM micrographs of the delamination initiated from intralaminar voids. The typical interlaminar shear failure is evidenced by 45° hackles in the delamination zones. Interestingly, as demonstrated in Figure 10, the delamination can only be observed on either side of void-free specimens (2.78 °C/min) and both sides of specimens with voids (7.0 °C/min).

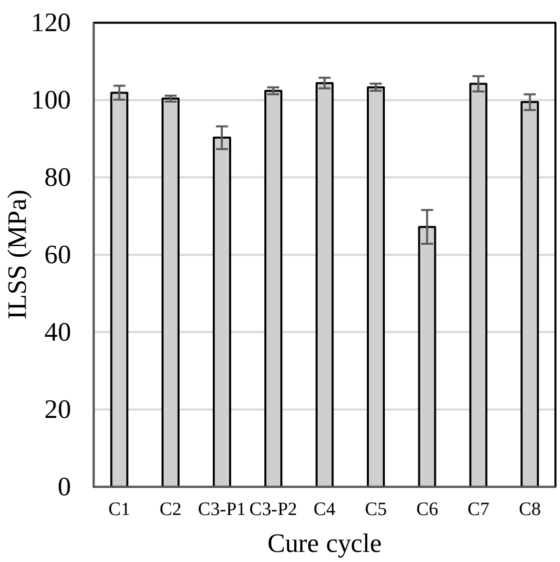

Figure 6: ILSS of laminates cured with different autoclave pressures and heating rates

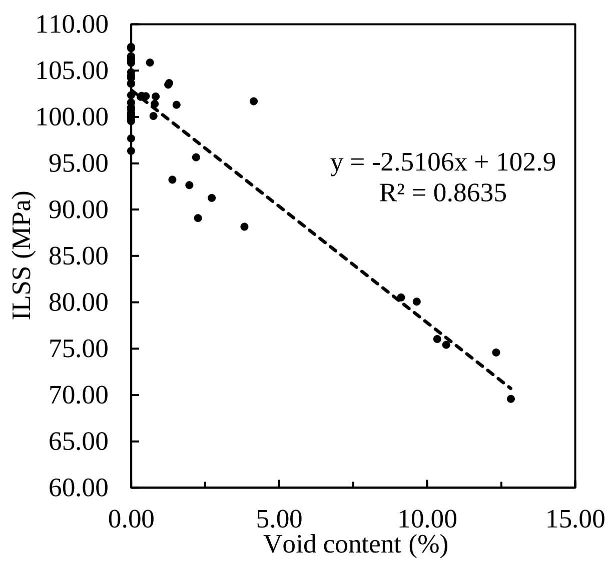

Figure 7: Correlation between void content and the ILSS of 3900-2B/T800SC laminates

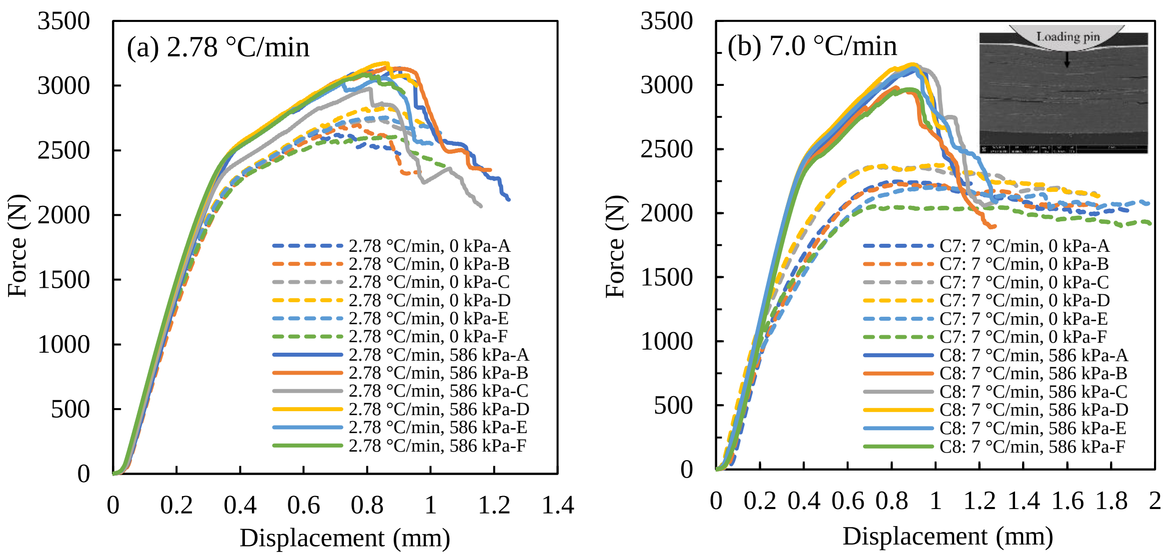

Figure 8: Comparison of load vs. deflection curves for short beam shear specimens cured with different heating rates and autoclave pressures, (a) 2.78 °C/min, (b) 7.0 °C/min

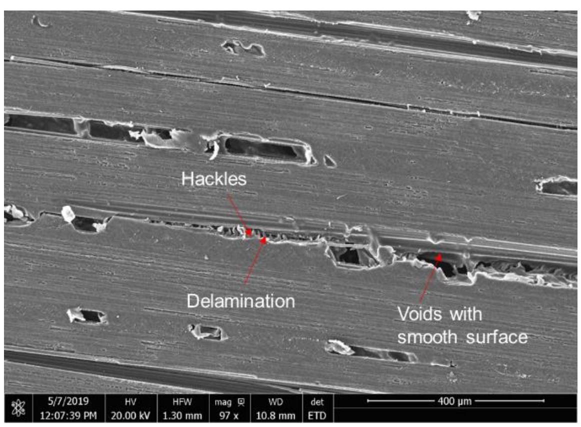

Figure 9: SEM micrograph of delamination initiated from interlaminar voids

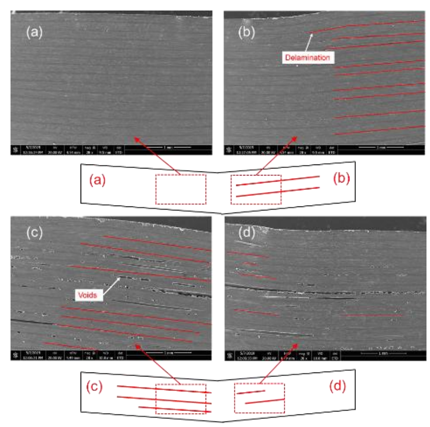

Figure 10: SEM micrographs of delamination distribution at left and right side of in SBS specimens, (a-b) C-3, (c-d) C-5

4 CONCLUSION

The present study investigated the effect of cure pressure and the role of interlayer particles on the void content and interlaminar shear strength of interlayer toughened composite laminates at different heating rates. The following conclusions can be drawn:

There might be a wider acceptable processing window for 3900-2B/T800SC than the MRCC. Void-free laminates can be made with low autoclave pressures. Lower heating rates allow more time for void migration and result in lower void content in laminates cured without autoclave pressure. The interlayer dispersed with particles inhibits in-plane and through-thickness resin transport and void migration. Therefore, voids mainly migrate along the fiber direction in the intralaminar region with the flowing resin. Porosity generally has a negative effect on the ILSS of 3900-2B/T800SC. The ILSS decreases by 2.51 MPa for every 1% of voids. Like untoughened composites, there is no noticeable reduction of the ILSS when void content is below 1%.

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