INFLUENCE OF SURFACE PREPARATION ON FUSION BONDING OF THERMOPLASTIC COMPOSITES

Francisco Sacchetti[1,2], Wouter J.B. Grouve[1], Laurent L. Warnet[2], Irene F. Villegas[3]

[1]ThermoPlastic composites Research Center (TPRC)
Palatijn 15, P.O. Box 770, 7500AE Enschede, The Netherlands
Email: francisco.sacchetti@tprc.nl, web page: http://www.tprc.nl

[2]Faculty of Engineering Technology, Chair of Production Technology, University of Twente
Drienerlolaan 5, P.O. Box 217, 7500AE Enschede, The Netherlands
Email: l.warnet@utwente.nl, web page: http://www.utwente.nl

[3] Structural Integrity and Composites, Delft University of Technology
Kluyverweg 1, 2629 HS Delft, The Netherlands
Email: I.FernandezVillegas@tudelft.nl, web page: http://www.tudelft.nl

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ABSTRACT

Carbon fibre-reinforced thermoplastic composites laminates (CFRP) meant for fusion bonding have been moulded using different release media. The potential contamination of the laminate surface by the release media and its effect on the mechanical performance of fusion bonded joints was studied. The physical and chemical state of the bonding surface were analysed with surface energy and roughness measurements. The chemical composition of the laminate surfaces has been investigated by X-ray photoelectron spectroscopy (XPS). The laminates were then fusion bonded through co-consolidation. The mechanical performance of the bonds was studied by a mode I fracture toughness measurement using the double cantilever beam (DCB) test. The results show significant effect of contaminations on mechanical performance of the joints.

1 INTRODUCTION

Continuous fibre reinforced thermoplastic composites are gaining more and more interest from the aerospace and automotive industry. Thermoplastic matrix based composite materials have shown similar, if not better, basic mechanical properties (e.g. strength and stiffness) than thermoset matrix based composite materials [1]. Moreover, thermoplastic based composite materials have some advantages over thermoset based composites materials. These include a higher toughness, better environmental resistance and improved flammability properties[2,3]. From a manufacturing point of view the main advantage of thermoplastic composites lies in their potential to achieve high production rates [4,5]. The ability to be thermally formed and fusion bonded give these materials a great potential for process automation [6–9]. Nevertheless, further research on these topics is required before these processes achieves their maturity.

Fusion bonding of thermoplastic composites is based on their ability to soften when heated and to recover their properties after cooling down under pressure. The performance of the joint develops after molecular interdiffusion at the joint interface. Proper interdiffusion requires intimate contact, which involves polymer flow to flatten the surface asperities. Fusion bonding can be performed with different techniques, which are usually classified by the mechanism of heat generation at the welding interface. Three techniques are well suited for long fibre thermoplastic composites: resistance, ultrasonic and induction welding [10,11].

Much research has been done in order to gain deeper insight into the effect of different variables on the quality of these processes [11–13]. The majority focusses on the effect of process parameters on the mechanical performance of the bond. Similar to adhesives, the state of the adherent prior to the welding process may have an influence on the mechanical performance of the bond [10]. However there is not much research done in this direction for fusion bonding. Variables such as surface contamination, degree of crystallinity of the matrix, residual stresses, or any other defects present in the adherent prior to fusion bonding may influence the properties of the joints.

Some authors have stated that the state of the surface of the composites part before fusion bonding is not relevant, since the matrix melts during the process [14,15]. However, contamination on the surface of the composite can be trapped in the bond line during the fusion bonding process which might affect the mechanical performance of the bond. It is known that for adhesives contamination trapped in the bond line was shown to have a strong influence on the mechanical performance of adhesively bonded joints [16]. Research on this topic demonstrated that the surface must be carefully treated before bonding with an adhesive. Good bond quality is only achieved when a minimum amount of contamination is guaranteed on the surface.

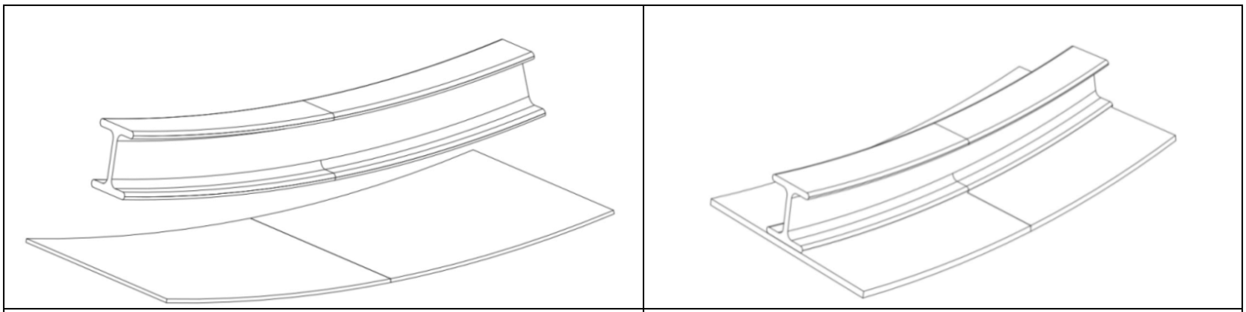
There are different sources for surface contamination, such as during handling and machining of the composites parts. During these processes, dust and grease can get deposited on the surface before the bonding process takes place. Therefore, the bonding areas of the parts are often cleaned before fusion bonding in order to remove these visible contaminations. Yet another, difficult-to-detect source of contamination is the release media used during consolidation and forming of the parts. Fibre reinforced thermoplastic composites are manufactured and shaped by applying pressure and heat with metal moulds or metal dies. This implies that the composites are formed against surfaces from which they must be released after consolidation. A release media is usually applied to the metal tooling surface to ensure the release of the composite part.

This research focusses on the influence of surface contamination coming from release media employed during the consolidation of the adherent on the mechanical performance of fusion bonded joints. In this study two separate consolidation steps were used as fusion bonding technique. The process is called co-consolidation and the specimen manufacturing comprises of two steps:

1. Two laminates were consolidated individually in a press using various release agents.
2. The parts were fusion bonded in a co-consolidation step in an autoclave.

Usually in co-consolidation, none of the parts that are being joined together are previously consolidated. However, in our case the two parts to be bonded are previously consolidated. A schematic representation of this process is shown in figure 1.

Pressure and temperature are the two main variables of the fusion bonding process. Nevertheless a few differences between welding and co-consolidation can be noticed. Firstly, the co-consolidation process takes more time (1-2 hours) than welding techniques (few seconds up to a minute). Secondly, the whole part is melted during co-consolidation while during welding polymer only melt near the interface. This makes co-consolidation a simplified fusion bonding technique compared to the welding process, allowing to isolate and study the influence of surface contamination on the quality of fusion bond.



Step 1: The parts are press consolidated

Step 2: Parts are fusion bonded by co-consolidation

Figure 1. Schematic of the two-step consolidation process (Co-consolidation)

Mechanical testing is required to evaluate the performance of fusion bonded components. The mechanical performance of the fusion bonded samples is compared with the parent material as a baseline for this study. Typically the mechanical performance of fusion bonded samples is characterised by lap shear tests [10-16]. The manufacturing of samples with the proper geometry for this test is difficult, particularly for the parent material [17]. This makes the comparison between bonded samples and the parent material problematic. Moreover, the geometry of the specimens has a great influence on the stress distribution and the results of the single lap shear test [18]. To overcome these difficulties, The double cantilever beam (DCB) test was chosen as mechanical characterisation technique in this research. This technique is suitable for testing the bonded samples as well as the parent material.

Surface contamination was measured using different techniques. A qualitative state of the surface was measured by surface energy. Surface topology changes were analysed by roughness measurements. Chemical surface changes were characterized by X-ray photoelectron spectroscopy (XPS). The state of the surface was then correlated with the mechanical performance of the fusion bonded joints.

2 EXPERIMENTAL METHODS

2.1 Material

A 200 kN Pinette Press and a 25.4 x 25.4 cm^2 picture frame mould were used to press consolidate flat laminates. Uni-directional carbon/polyetheretherketone (PEEK) pre-preg material (Cetex TC1200 produced by TenCate) was used in this research. Laminates of 12 layers were press consolidated, the stacking sequence used was $[0]_{6s}$. The nominal laminate thickness after press consolidation was 1.5 mm. In total 6 laminates were press consolidated, each two laminates were press consolidated using the same release media. Three different release media were used namely, Polyimide A, which has a T_g (285 °C) below the processing temperature of PEEK; Polyimide B, which has a T_g above the processing temperature of PEEK, and finally a semi-permanent silicon based release agent. The laminates were press consolidated simultaneously in batches of three laminates, separated by steel caul sheets of 1 mm thickness. A typical press consolidation cycle provided by TenCate was used and is shown in figure 2.

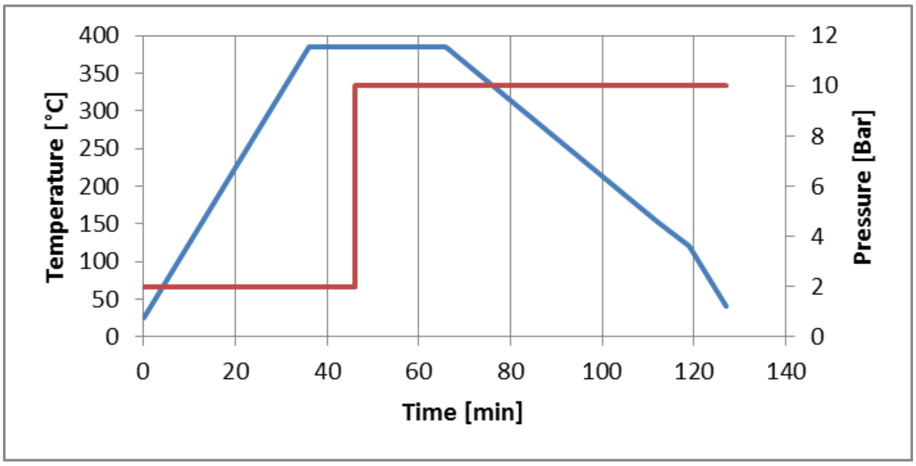


Figure 2. Press cycle. Blue line indicates temperature and red line indicates pressure.

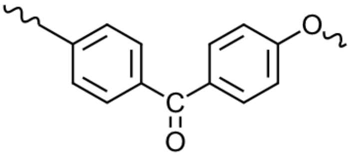
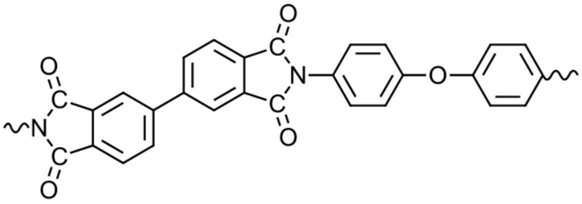
After the press consolidation step, the laminates were subjected to cleaning with the aim to reduce the amount of contamination on the surface. The surfaces of the laminates were wiped for 2 minutes with cloth wetted in isopropyl alcohol, applying a normal force of approximately 10 N.

2.2 Surface analysis

The surface tension of the laminates was determined from contact angle measurements using different liquids at room temperature. This technique is known to be sensitive to contamination of the surface [19]. The three different liquids used were demineralized water, n-Hexane, and ethylene glycol. Contact angle measurements provided combined information about roughness and chemical composition of the surface. Roughness measurements were carried out using a confocal microscope to isolate the effect of the surface topology.

Chemical composition of the composite surface was determined by X-ray photoelectron spectroscopy (XPS). Peaks in the XPS spectra can be assigned to chemical elements and the amount of each element can be calculated from the area under the peaks.

Reference values are required to improve the interpretation of XPS analysis/results.. For this purpose, the XPS spectra of the two pure polyimide (PI) films and the Carbon/PEEK pre-preg were obtained. The chemical structure of the components are shown in figure 3. The spectra of the PI film and Carbon/PEEK samples were then compared with the spectra of the laminate surfaces.

	PEEK
	Polyimide A

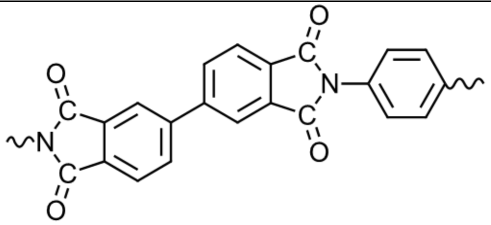
	Polyimide B

Figure 3. Chemical structure of PEEK, Polyimide A, and Polyimide B.

2.3 Fusion bonding

A second consolidation step was performed in an autoclave, to create the fusion bonded laminates. The autoclave cycle was performed following the specifications of TenCate, the cycle is depicted in figure 4. Two laminates which were press consolidated with the same release media were fusion bonded together. The nominal thickness of the laminate after consolidation was 3 mm.

In the same autoclave cycle one laminate of 24 layers was consolidated from the pre-preg tape (parent material). A 60 mm-wide and 15 μm thick polyimide film strip was placed between each two pre-consolidated laminates and in the mid plane of the reference laminate to form a pre-crack for the DCB testing.

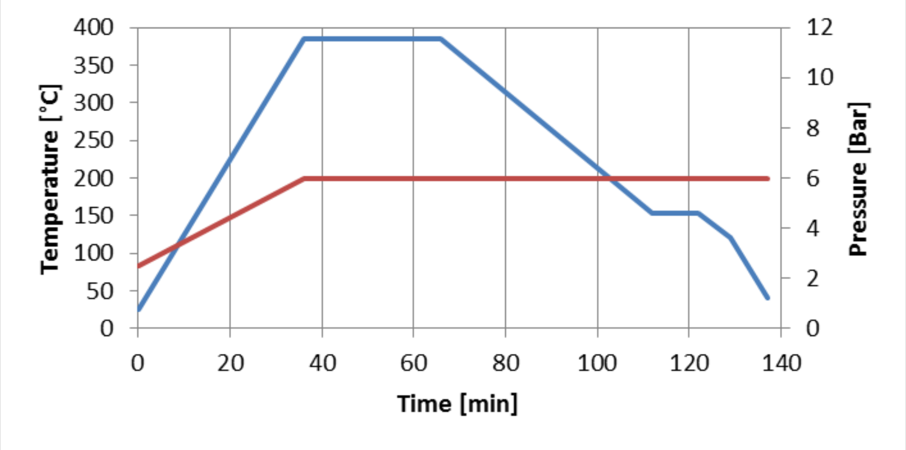


Figure 4. Autoclave cycle. Blue line indicate temperature and red line pressure.

2.4 Mechanical testing

DCB samples were cut from the laminates in the longitudinal direction of the fibres. The samples were cut and then tested according to ISO 15024. A traveling camera was used to measure the crack length during testing. The corrected beam theory (CBT) data reduction analysis was followed to analyse the data.

Cross-section micrographs of in the midplane of the samples were used to assess the state of the fusion bonded interface. Fractography was used to evaluate the main failure modes of the samples with different welding conditions.

3 RESULTS AND DISCUSSION

The influence of different release media on the surface of the laminate in terms of chemical and physical change was studied after press consolidation. Surface energy measurements, roughness measurements and XPS spectroscopy were carried out. The mechanical performance of the interface was tested using DCB test method after fusion bonding of the laminates by co-consolidation.

3.1 Surface analysis

3.1.1 Surface energy and roughness

The contact angle measurements were used to calculate the surface energy of the different laminates after press consolidation and cleaning. Surface energy results with standard deviations are given in Table 1. Three specimens were tested for each sample and 5 measurements were performed for each specimen. It is important to note that the measurements were performed parallel to the fibre direction. The contact angle in the direction perpendicular to the fibres would be different due to influence of the surface topography. The Owens, Wendt, Rabel and Kaelble (OWRK) method [20] was used to calculate the surface energy. In all the cases a contact angle of 0 degree was observed when n-hexane was used.

Release agent	Surface Energy [mJ/m2]
Si based	29±1
Polyimide A	30±1
Polyimide B	48±1

Table 1: Surface energy of the different samples. The liquid used were demineralized water, n-Hexane, and ethylene glycol.

It can be noted that surface energy of the laminates consolidated with polyimide A and Si based semi-permanent release media showed similar values. While laminates which where press consolidated using Polyimide B showed a different surface energy.

A confocal microscope was used to measure the roughness of the three specimens. Three measurements were performed for each specimen over an area of 0.25 cm^2. An average value R_q of 0.25 ± 0.05 µm was obtained for all the samples. The variance of the results of the different samples was within the error of the measurement technique.

The different surface energy values indicate either a different surface chemistry or different surface topology. However the surface roughness was observed to be the same in all the specimens among the samples. Hence, the difference in surface energy has to be due to differences in the chemistry of the surface.

3.1.2 XPS

The XPS results are summarized in Table 2. which shows the amount of each element detected on the surfaces laminates. The first 4 rows of table 3 show reference values for the neat material. The last three rows show the results for the laminates consolidated using Si based release agent, Polyimide A and Polyimide B.

Element	C	O	Si	N	Others
Binding Energies [e.V.]	285	532	102	689	-
Theoretical PEEK [REF?]	86.4	13.6	-	-	-
Carbon/PEEK Pre-preg	85.2	13.3	0.1	-	1.4
Polyimide A	81.5	13.9	-	4.5	0.1
Polyimide B	79.6	14.3	-	6.1	-
Laminate – Si base	81.4	15.9	2.4	-	0.3
Laminate - Polyimide A	82.7	14.7	-	2.0	0.6
Laminate Polyimide B	73.5	17.9	-	4.9	3.7

Table 2: XPS results, amount of each element.

The XPS results for Polyimide and PEEK are quite similar. This makes it difficult to distinguish these two components only by comparison of their constitutive elements. Nevertheless, a clear difference between them is the presence of Nitrogen in the Polyimide.

Differences between the theoretical PEEK value and the pre-preg are observed, however the values are similar. This gives an idea of the sensitivity of this technique. It can be noted that all the samples have some contamination elements namely, Na, S, Si and N among others. The elements that are not a constituent part of the release agent or PEEK were grouped in “other” column in table 3. Even the reference samples such as the pure films and pre-preg have showed extra elements.

The laminates consolidated with the semi-permanent Si based release agent and Polyimide A are the ones with least contamination. The main element detected besides the constitutive elements of PEEK are Si and N which are the constitutive elements of the semi-permanent release agent and polyimide A respectively. The laminates in which Polyimide B was used as a release media shows higher concentration of N than the previous sample. Moreover, the combination of Nitrogen and other elements add up to 8.6 %, three times more contamination than what is observed in the other samples.

3.2 Mechanical testing

3.2.1 Fracture toughness

The fracture toughness in Mode I of the different samples was tested following ISO 15024. The crack propagation was observed to be stable in all cases. Force vs. displacement and crack length vs. displacement curves are shown in figure 5. The calculated fracture toughness is shown in figure 6.

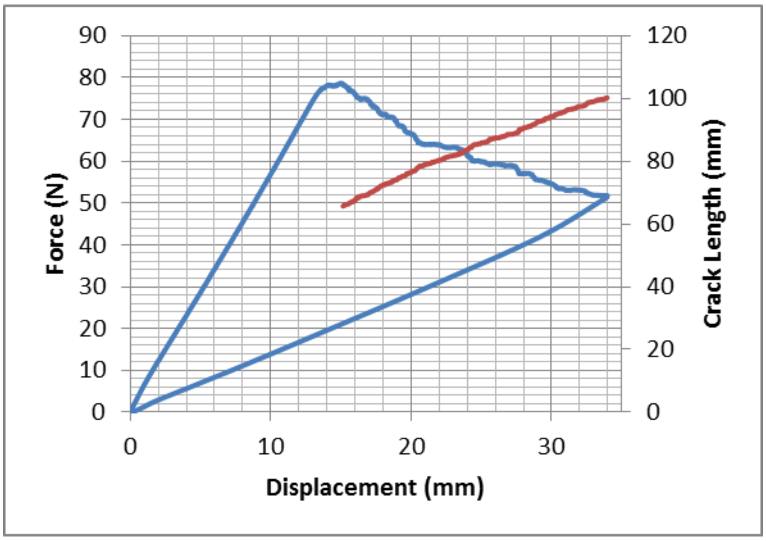


Figure 5. Blue line indicates force vs. displacement curve, and red line indicates crack length vs. displacement.

The co-consolidated samples which were first press consolidated against the Si based release media and against Polyimide A show similar toughness as the parent material. This suggests that the amount of surface contamination in these samples was not enough to affect the mechanical performance of the fusion bonds. However, co-consolidated sample in which adherents were press consolidated using Polyimide B shows a significant drop in toughness compared to the other samples. This correlates with higher contamination of the laminate surface observed in the XPS measurements.

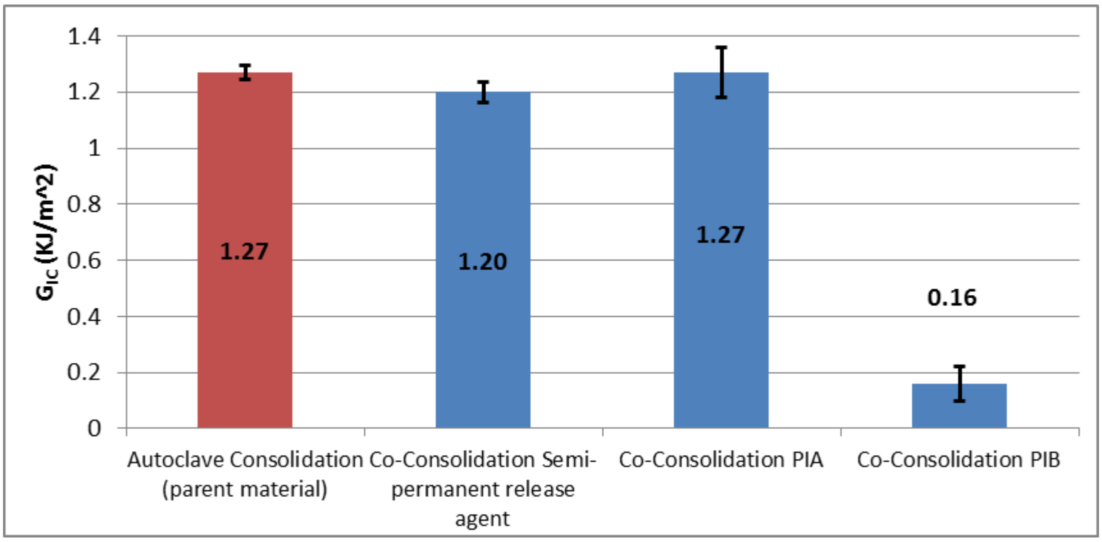


Figure 6. Delamination fracture toughness of different samples.

3.2.2 Cross section micrographs and fractography

Cross sectional micrographs of all the samples were analysed. The bonded area between two tapes in one step consolidation technique looks similar to the bonded area of two laminates in the two step consolidation (fusion bonding) process (figure 7, first column). No significant differences were observed among the samples.

The cross section of the samples and the fracture surface were analysed using optical and scanning electron microscopy (SEM) after performing the tests. All samples which showed higher toughness look similar under the microscope, characterized by a tortuous crack path, with a fibre rich surface, some broken fibres and fibre imprints in the matrix. A completely different fracture surface was found in the samples with low toughness, where only a matrix rich fracture surface was observed and a smooth crack path (figure 7). Proper intimate contact in all the cases can be observed on the micrographs. Nevertheless, the samples with the highest contamination concentration, have not developed higher toughness. This may be due to poor interdiffusion obstructed by the contamination on the interface.

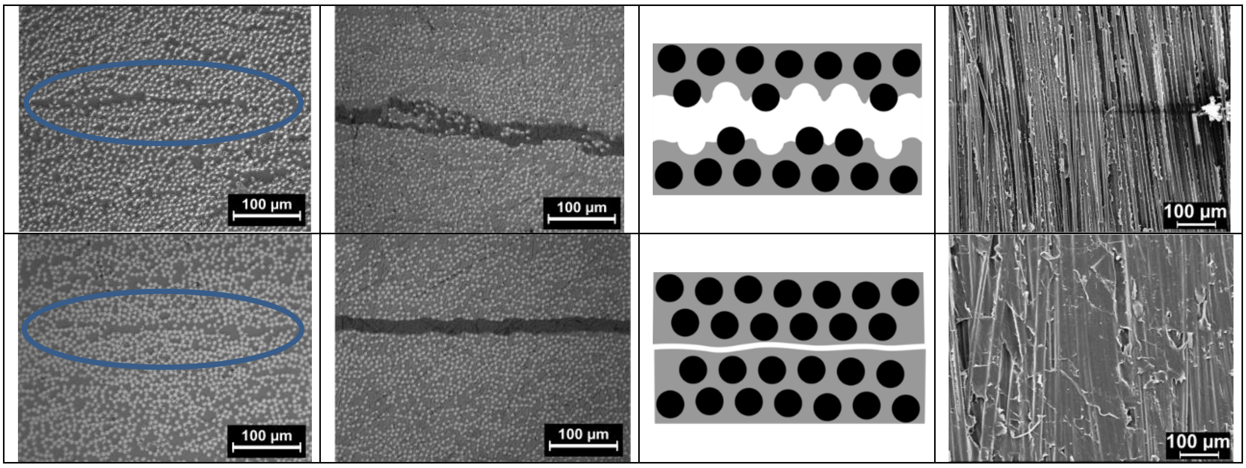


Figure 7. From left to right, cross section micrographs before testing (the blue circle indicates the interfaces), cross section after testing, scheme of the failure, and SEM micrograph of the fracture surface. From top to bottom, one step press consolidation of 24 layers, and co-consolidation with Polyimide B release media.

4 CONCLUSION

The contamination of the surface of laminate due to the release media and its effect on the mechanical performance of fusion bonded joints was studied.

Surface energy was used as a quick indicator of the cleanliness of the surface. The surface of the laminates which were press consolidated in contact with silicon based release agent and Polyimide A have shown the same value, whereas use of Polyimide B has shown higher surface energy.

XPS analysis has shown that main source of composite surface contamination in these samples were from the release media and it always occurs irrespective of the type of release media used. Samples which show a lower amount of contamination in the XPS results have identical surface energy. Higher value of surface energy was measured which correlates to the higher amount of contamination observed in XPS results.

The fracture toughness of the co-consolidation samples were comparable with the performance of the parent material, when lower levels of contamination were observed. High surface contamination resulted in joints with an extremely low toughness.

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