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- Effect of resin-rich bond line thickness and fibre migration on the
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Effect of resin-rich bond line thickness and fibre migration on the toughness of unidirectional Carbon/PEEK joints

15 Abstract

It is a common practice in fusion bonding of thermoplastic composites to add a matrix 16 layer between the two substrates to be joined. The aim is to ensure proper wetting of 17 18 the two parts. The effect of this additional matrix layer on the mechanical performance was studied by mode I fracture toughness measurements. The additional matrix was 19 inserted at the interface in the form of films of various thicknesses. Three different 20 21 manufacturing techniques, namely autoclave consolidation, press consolidation and stamp forming, were used to prepare different sets of specimens with varying resin-rich 22 bond line thickness. The occurrence of fibre migration towards the matrix rich 23 interface was induced by the manufacturing techniques used due to their different 24 processing times. The interlaminar fracture toughness was observed to increase with 25 26 increasing amount of extra-matrix at the interface, while no effects of the fibre migration on the fracture toughness were observed. 27

28 Keywords: thermoplastic composites, fusion bonding, matrix interleaving, fracture mechanics,

29 fractography

31 **1. Introduction**

Fusion bonding can be considered as an affordable way to assemble thermoplastic composite 32 parts [1]. From a practical viewpoint, the process involves heating of the interface between 33 the parts, followed by the application of pressure and cooling down. There are many fusion 34 bonding techniques available, all differing in the way heat and pressure are applied to the 35 interface [2, 3]. Two groups of fusion bonding techniques can be distinguished by the size of 36 the area heated, namely bulk heating and welding, which is characterised by local heating. 37 38 The first group consists of bringing the entire part to melt and using the tooling to maintain pressure throughout the process. Consequently, this technique is characterised by a relatively 39 long processing time (1-2 hours) [4]. The second group is characterised by local heating, and 40 41 sometimes by local application of pressure, which means that a short processing time can be 42 achieved (minutes or seconds).

From a physical viewpoint, the fusion bonding process involves intimate contact 43 development between the two surfaces (also known as wetting), followed by interdiffusion of 44 polymer chains across the interface (also known as healing) [5]. Proper wetting may be a 45 challenge for thermoplastic composites with a high fibre volume fraction due to the lack of 46 matrix material at the interface; this may result in poor bond performance [6]. To solve this 47 problem, an additional layer of neat polymer can be inserted (interleave) at the interface in 48 49 order to promote wetting [7, 8, 9]. Moreover, some welding techniques may, in any case, require such an additional resin layer at the interface. For example, a resin layer is added as 50 an energy director in the case of ultrasonic welding, while resistance welding requires a metal 51 52 mesh embedded in a matrix layer at the interface [5, 10]. This additional layer of pure polymer may lead to a matrix rich bond line which in turn may affect the joint performance. 53 A proper understanding of the interrelation between the matrix rich bond line thickness and 54 the joint performance is required to enable optimisation of the joint design. 55

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Earlier research showed that the interlaminar fracture toughness of Carbon/PEEK increases 58 with interleaving thickness (i.e. with increasing thickness of the matrix rich bond line) [11, 59 9]. This is in line with the work on other material systems [12, 13, 14, 15] and adhesives 60 joints [16, 17] and is generally related to the size of the plastic yielding zone in front of the 61 62 crack tip. An increase in matrix interface thickness allows for a larger plastic yielding zone, resulting in a higher interlaminar fracture toughness. It is proposed that the interlaminar 63 toughness eventually reaches a plateau value equal to the matrix toughness for larger matrix 64 interleaved thicknesses [18, 16]. To the best of the authors' knowledge, the aforementioned 65 studies were all performed on samples manufactured using a typical (bulk heating) 66 consolidation technique, i.e. either autoclave or press consolidation. These techniques are 67 characterised by a long processing time, which allows fibres to migrate into the matrix rich 68 area at the interface. The long processing times are not representative of what happens during 69 welding. In this case, the short processing times are expected to significantly reduce fibre 70 migration. It is not clear how this fibre migration affected the measured toughness values 71 reported in the literature. Two effects may play a role, on one hand, fibre migration leads to 72 more fibre-fibre contact, which effectively reduces the plastic zone size and, hence, the 73 fracture toughness [19, 13]. On the other hand, fibre migration may also lead to fibre nesting, 74 resulting in so-called fibre bridging which causes an increase in toughness [20, 21]. 75

In this research, the effect of interleaving thickness and fibre migration on the interlaminar fracture toughness of unidirectional carbon fibre reinforced poly-ether-ether-ketone (Carbon/PEEK) fusion bonded samples was studied. The interleave thickness was varied by adding unreinforced PEEK films of varying thickness at the interface between the laminates, while the extent of fibre migration was varied by using different processes. Two slow processes, autoclave consolidation and press consolidation, and one fast process, stamp forming, were used to prepare interleaved fusion bonded samples. The slow processes are expected to yield samples with a high degree of fibre migration, while the fast process should prevent significant fibre migration. A mode I double cantilever beam (DCB) test was used to evaluate the fracture toughness of the joint under mode I failure. Fractographic analysis of the samples was performed after mechanical testing to investigate the failure behaviour of the different samples.

88 2. Experimental methods

Sample preparation consisted of two steps. First, laminates were press consolidated following the procedure described below. Second, these laminates were used as substrates for a fusion bonding step in which two substrates were joined to form a sample. The substrates were fusion bonded using three processing technologies as described in this section. Subsequently, the physical state and the interlaminar fracture toughness of the samples was characterised by cross-sectional microscopy and DCB test respectively. The procedures followed to perform these measurements are described in the following sub-sections.

96 2.1. Materials and substrate manufacturing

Press consolidation was used to prepare unidirectional Carbon/PEEK laminates with a 97 stacking sequence of [0]₁₂. The material was provided by TenCate and is known as Cetex® 98 TC1200. The fibres used in the prepreg is a T300 JB 3K while the polymer is a Victrex 99 PEEK 150. The prepreg was stacked in a picture frame mould of 300 by 300 mm² and 100 101 subsequently consolidated using a static Pinette Emidacau Industries press following the 102 consolidation cycle suggested by TenCate, which is shown in Figure 1. To ensure deboning of the laminates from the mould, Marbocote® 227CE, a silicon based semi-permanent mould 103 104 release agent was used as a release media. These laminates were then used as the substrates for the fusion bonding processes. 105



106

Figure 1: Press cycle used to manufacture the laminates.



108 Three different processing techniques were used to prepare the fusion bonded samples, i.e. autoclave consolidation, press consolidation and stamp forming. Regardless of the processing 109 method, a sample was prepared by stacking two substrates on top of each other with 110 optionally additional PEEK film inserted at the interface. The film was manufactured by 111 Victrex and is known under the tradename APTIV®. It was available in two different 112 thicknesses, namely 38 µm and 100 µm. Moreover, a 13 µm thick polyimide film was also 113 inserted between the substrates prior to fusion bonding in order to introduce the pre-crack 114 required for DCB testing. It is worth to notice that in the area where the polyimide film was 115 inserted the additional PEEK films were not inserted. The remainder of this section describes 116 each of the aforementioned processing techniques. 117

118

2.2.1. Autoclave consolidation

An autoclave consolidation process was used to fusion bond the first sample set. Seven samples were prepared. The first sample was prepared without an additional film at the interface, while for the other six samples, one to six layers of film with a thickness of 38µm were inserted at the interface prior to consolidation.

123 A schematic illustration of the autoclave table preparation can be found in Figure 2. The press

consolidated substrates were cut into square sections of 150 by 150 mm² and subsequently stacked together with the required film material. Brass picture frames with different thicknesses were used as a shim at the interface to maintain the distance between substrates and thereby to prevent the added matrix from being squeezed out. A 10 mm thick aluminium caul sheet was used to ensure the flatness of the laminate. After wrapping the table in a vacuum bag, the substrates were fusion bonded in an autoclave at 6 bar pressure and a temperature of 380 °C based on the process cycle recommended by TenCate, which is shown in Figure 3.



Figure 2: Sketch of the preparation of the autoclave table. In the top view, the upper substrate is not shown for clarity.



Figure 3. Autoclave consolidation cycle used to fusion bond the substrates.

141 2.2.2. Press consolidation

A second sample set was prepared by press consolidation of two substrates in a press using a 300 by 300 mm² picture frame mould. A total of three samples were prepared: one without an additional polymer film, one with a 38 μ m PEEK film and one with a 100 μ m PEEK film. Contrary to the autoclave consolidation process, no shims or spacers were added as any squeeze flow was restricted by the picture frame mould. The consolidation cycle was the same as the one used to manufacture the substrates i.e. the cycle as shown in Figure 1.

148 *2.2.3. Stamp forming*

149 The last sample set was prepared by using a Pinette Emidacau Industries stamp forming set up to fusion bond two substrates. Two substrate laminates were stacked and placed on a 150 polyimide film of 50 µm thickness, meant for carrying the laminates from the material 151 loading position to the infrared oven and from the oven to the pressing/stamping position. 152 The infrared oven was set at a temperature of 450 °C. The substrates were heated up to 153 154 complete melting (the temperature at the interface between the two laminates was measured to be 400 °C, taking around 4 minutes of heating time). Then, the substrates were transferred 155 to the stamping station where they were fusion bonded and consolidated between two flat 156 aluminium moulds with a dimension of 250 by 250 mm². The mould temperature was set to 157 220 °C. The mould halves were quickly closed, and a pressure of 10 bar was applied for 1 158 minute. The measured temperature and pressure during stamp forming are illustrated in 159 Figure 4. Three samples were prepared: one without extra polymer, one with a 38 µm PEEK 160 polymer film and one with a 100 µm PEEK polymer film at the interface between the two 161 laminates. Table 1 summarises all the samples that were prepared. 162





Figure 4: Measured temperature and pressure during stamp forming process.

	Number and thickness (um)	Fusion bonding technique/ Sample name				
of PEEK film plies	Autoclave	Press	Stamp-			
				forming		
	None	A-None	P-None	S-None		
	1 x 38 μm	A-1x38	P-1x38	S-1x38		
	2 x 38 μm	A-2x38	-	-		
	1 x 100 μm	-	P-1x100	S-1x100		
	3 x 38 μm	A-3x38				
	4 x 38 μm	A-4x38				
	5 x 38 µm	A-5x38	-	-		
	6 x 38 um	A-6x38				

165

Table 1: Sample description, the thickness of interleaving, and fusion bonding technology used.

166 2.3. Characterization

After fusion bonding, cross-sectional micrographs of the samples were prepared.
Subsequently, double cantilever beam (DCB) tests were performed followed by a
fractography analysis.

170

2.3.1. Cross-sectional microscopy

The consolidation quality of the fusion bonded samples was characterised using thickness measurements and cross-sectional microscopy. The micrographs were taken close to the edge of the fusion bonded laminates, while the centre was kept for mechanical testing, as it is shown in Figure 5. The microscopy images were also used to evaluate, in a qualitative manner, the thickness of the matrix rich bond line and the degree of fibre migration at the



Figure 5: Sketch of the location of the cross-sectional sample preparation and the position of the DCB
 samples



2.3.2. Double cantilever beam experiments

The interlaminar fracture toughness of the bond line was evaluated using the double 180 cantilever beam (DCB) test method. DCB specimens were cut from the fusion bonded 181 samples in the longitudinal direction of the fibres and then tested according to ISO 15024 182 [22]. The ISO Standard 15024 is based on the linear elastic fracture mechanics (LEFM). As 183 such, the conformance of the linear elastic behaviour of the specimens during testing was 184 evaluated. Figure 5 shows schematically the location of the test specimens cut to a width of 185 20 mm from the fusion bonded laminates. The specimens were loaded in a servohydraulic 186 Instron 8500 universal testing machine equipped with a 1 kN force cell. A mode I pre-187 cracking procedure was performed for all the specimens according to the standard. The 188 189 specimens were loaded at a constant speed of 1.2 mm/min until a delamination crack growth of about 5 mm has occurred, followed by the specimens unloading until zero force. Next, the 190 specimens were re-loaded at the same constant speed of 1.2 mm/min until the final 191 192 delamination length of about 100 mm has been reached. A travelling recording camera was used to measure the delamination crack length during testing. The corrected beam theory 193 (CBT) was used to analyse the data. The interlaminar fracture toughness was calculated as: 194

$$G_{\rm IC} = \frac{3P\delta}{2w(a+\Delta)} \left(\frac{F}{N}\right),\tag{1}$$

where P is the force, δ is the displacement, a is the crack length, w is the width of the 195 specimen, F is a correction factor for large displacement, N is a correction factor for the 196 197 loading blocks and Δ is a correction factor for the rotation of the beam at the crack tip. Since the delamination length was measured using the horizontal position of the travelling camera 198 system, there is no need for a large-displacement correction factor (F) to be applied to the 199 measurements [22] (i.e. F can be considered equal to one). The interlaminar fracture 200 toughness was calculated both for initiation and propagation. The initiation values were 201 calculated following the procedure called 5 % / MAX point in the ISO 15024 standard. From 202 that point on the values measured were considered as propagation values. 203

204

2.3.3. Fractography analysis

Two cross-sectional optical micrographs were prepared after testing. One with a sectioning 205 plane perpendicular to the crack propagation direction and the other with a sectioning plane at 206 20° with respect to the crack propagation direction. A schematic view of how these cross-207 sectional cuts were taken is shown in Figure 6. All the micrographic specimens were 208 209 embedded in epoxy and then polished. A Leica DMRX and a Keyence VHX optical microscope were used to obtain the optical micrographs. Moreover, SEM micrographs of the 210 fracture surface were made with a Jeol Neoscope JCM-5000. The cross-sectional and 211 fractography images were analysed in order to determine the crack propagation path and to 212 identify the main failure modes. 213



Figure 6: Sketch of the location of cross section micrograph cuts for the fractography analysis

215 **3. Experimental results**

The experimental results are elaborated in the present section. First, the physical state of the samples and bond line microstructure will be evaluated. Subsequently, the fracture toughness data is provided, followed by the fractographic analysis.

3.1. Physical state of the samples

The fusion bonded samples prepared using the autoclave consolidation process showed non-220 uniform thickness, with the centre of the laminates being thicker than the edges. Despite their 221 thickness (10 mm), the aluminium caul sheets were bent and permanently deformed during 222 the autoclave cycle as a result of the high pressure applied. In some case, the difference in 223 thickness between the edge and the centre was up to 0.15 mm. The quality of the samples 224 manufactured using press consolidation and stamp forming process, in terms of variation in 225 sample thickness, was superior to the autoclaved samples with the variation in thickness 226 being always less than 0.05 mm. 227

Typical cross-sectional micrographs for the three fusion bonding techniques and with 228 229 different interleave thicknesses are presented in Figure 7. All micrographs showed good consolidation quality with no voids in the substrates or the interface. For the cases in which a 230 PEEK film was inserted between the laminates prior to fusion bonding, two different regions 231 can be distinguished in all the micrographs shown in Figure 7, i.e. a matrix poor region 232 mainly in the substrates, and a matrix rich region at the bond line. Besides, two different 233 morphologies can be identified in the matrix rich region. The first is characterised by matrix 234 material in which many fibres are randomly distributed as shown in the first and second 235 columns in Figure 7. This morphology arises when fibres migrate, during processing, from 236 the substrates into the interleaved film at the interface. This happened during the slower 237 processes, i.e. during autoclave and press consolidation. The second morphology is 238

characterised by matrix material with very few or no fibres. This is evident in the stamp
formed samples (last column in Figure 7), for which there is not enough time for the fibres to
migrate during processing.



Figure 7: Cross-sectional micrographs of 6 different specimens close to the interface. Left to right: autoclave
consolidated specimen, press consolidated specimen, and stamp formed specimen. Top row: specimen
interleaved with a 38 μ m thick film. Bottom row: specimen interleaved with 100 μ m film in the case of press
consolidation and stamp forming, specimen interleaved with 3 layers of 38 μ m thick films in the case of
autoclave consolidation. The white bar on the left of the micrograph indicates the thickness of the interleaved
films before processing.

The thickness of the matrix rich region was not uniform along the cross-sectional plane for the autoclaved specimens, which was associated with significant matrix flow during processing. The effect of this non-uniformity on toughness will be further elaborated in the next section. On the contrary, the press consolidated, and the stamp formed samples showed a more uniform thickness of the matrix rich region.

3.2. Double cantilever beam experiments

254 This section presents the results of the DCB experiments. First, the issues encountered during

testing are described and examples of force vs. displacement curves are shown. At the end of

this section, the results from all the samples tested are combined to generate a plot of fracturetoughness as a function of interleaving thickness.

3.2.1. General observations during DCB testing.

Five DCB specimens were tested for each sample. Nevertheless, several issues were 259 encountered during DCB testing which made the analysis difficult and reduced the number of 260 specimens kept for the analysis. The main problems encountered were instability of crack 261 propagation and the presence of a non-flat resistance curve (toughness vs. crack length). The 262 former leads to a small number of propagation values, making the specimen less statistically 263 relevant, while the latter indicates possible variations in crack propagation mechanisms, such 264 as for example fibre bridging. As both complicate data reduction, two criteria were 265 implemented to obtain a set of specimen data for analysis. A specimen was kept for analysis 266 in case it showed i. at least 10 mm of stable crack propagation, and ii. less than 20% variation 267 268 in interlaminar toughness along the 10 mm of crack propagation. An exception to the second criteria was made for the stamp formed specimens. The threshold was changed to 50% in 269 order to have enough specimens for analysis. It is worth to notice that only few stamp formed 270 specimens were kept for the analysis which were close to the second criterion. These criteria 271 led to only three to four consistent specimens from an initial lot of five specimens per sample. 272 An exception was the sample from the autoclave which was interleaved with three 38 µm 273 films. Out of the five specimens tested, only two were kept for the analysis. Table 2 274 summarises the number of specimens discarded and the reason for not using the data. The last 275 column shows the number of specimens kept for the analysis. From the table, it can be noted 276 that the standard samples, i.e. the autoclave and press consolidation samples without 277 interleaving, did not present any problem during testing and all the specimens were kept for 278 the analysis, while all the samples that were manufactured by a nonconventional procedure, 279

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281 specimen.

	Number of specimens						
Sample Name	Presented at least one point of unstable crack propagation	Did not show at least 10 mm of stable crack propagation	More than 20% or 50% derivation in R-curve	Used for the analysis			
Autoclave							
A-None	0	0		5			
A-1x38	1	1		4			
A-2x38	5	2	0	3			
A-3x38	4	3	0	2			
A-4x38	4	1		4			
A-5x38	3	1		4			
A-6x38	3	2		3			
Press							
P-None	0	0	0	5			
P-1x38	3	1	0	4			
P-1x100	2	1		4			
Stamp							
S-None	2	0	1	4			
S-1x38	4	0	1	4			
S-1x100	4	0	2	3			

282 283

 Table 2: Overview of the number of specimens discarded and the reason for not using the data. The last column shows the number of specimens used for analysis.

Two characteristic force - displacement and crack length - displacement curves are shown in 284 the upper graphs of Figure 8. The left graph corresponds to a specimen which showed stable 285 crack propagation, while the right graph belongs to a specimen which showed a combination 286 of stable and unstable crack propagation. During the evaluation of the initiation point, the 287 maximum force point occurs almost always before the 5% point. Furthermore, almost no 288 289 residual displacement was observed after the specimens were unloaded. The previous two 290 observations means that the material can be analysed according to LEFM by following the 291 ISO15024 standard. Fibre bridging was not observed during testing.

The R-curves corresponding to the four specimens are shown in the bottom row of Figure 8. As shown, only the stable part was used to calculate the interlaminar fracture toughness. The first point of the R-curve corresponds to the initiation value for interlaminar fracture toughness. It can be noted that stable crack propagation is correlated with a continuous Rcurve, whereas in the presence of an unstable crack propagation the R-curve is interrupted
and therefore shows separate segments.







Many of the autoclave consolidated specimens suffered from unstable crack propagation as 305 was illustrated in Table 2. Moreover, some of the specimens showed non-uniform toughness 306 along the crack length. In those cases, the trend of the R-curve was mostly decreasing. 307 Although the press consolidated specimens also suffered from unstable crack propagation, 308 they showed longer paths of stable crack propagation compared to the autoclave consolidated 309 samples. Moreover, the R-curves observed in press consolidated specimens were flatter than 310 the ones observed for the autoclave consolidated samples. Finally, the stamp formed samples 311 despite several cases of unstable crack propagation showed a long path of stable crack 312 313 propagation. Some of these specimens showed a rising R-curve, which in some cases was too large (more than 50%), leading to the rejection of these specimens for the analysis. 314

The origin of the unevenness in the R-curves observed in the autoclave and stamp formed 315 specimens were attributed to two different phenomena. For the case of the Autoclave samples 316 the decreasing R-curve could be caused by a decreasing interleave thickness towards the end 317 of the specimen, which is the result of resin outflow during processing. The non-flat R-curves 318 of stamp-formed specimens may be related to variations in consolidation quality. Although 319 no voids were observed in the specimens, the degree of healing may vary from place to place. 320 As the process is highly non-isothermal, it is difficult to control temperature during the 321 process. However, a complete picture requires an in-depth investigation, which is deemed to 322 be out of scope of this paper. 323

324

3.2.2. Fracture toughness vs. interleaved thickness

The results of all the samples tested are summarised in Table 3. An average initiation and propagation fracture toughness values were calculated for all the samples. The average initiation value of each sample was calculated by averaging the initiation values of all the specimens within one sample. The average propagation value per sample was determined by 329 averaging the mean propagation value of each specimen within that sample.

The last column of Table 3 shows the overall trend of the R-curve for each sample, i.e. whether the R-curve was observed to be flat (-), ascending (/) or descending (\). It can be seen that the trend of the R-curve is closely related to the relation between initiation and propagation. In the cases of a flat R-curve the initiation and propagation values are similar, whereas with an ascending R-curve initiation values are lower than propagation, and the opposite occurs with a descending R-curve.

Sample	Fracture To	R-curve	
type, name	Initiation (kJ/m^2)	Propagation (kJ/m ²)	trend
Autoclave			
A-None	1.28 ± 0.10	1.30 ± 0.10	-
A-1x38	1.59 ± 0.10	1.55 ± 0.10	-
A-2x38	1.87 ± 0.16	1.89 ± 0.12	-
A-3x38	1.93 ±0.20	2.05±0.16	-
A-4x38	2.46 ± 0.30	2.22±0.18	\
A-5x38	2.74 ± 0.15	2.61±0.15	\
A-6x38	2.85 ± 0.28	2.66 ± 0.20	\
Press			
P-None	1.17±0.10	1.19±0.10	-
P-1x38	1.54 ± 0.10	1.51 ± 0.10	-
P-1x100	2.06±0.17	2.12 ± 0.18	-
Stamp			
S-None	1.10±0.37	1.25±0.25	/
S-1x38	1.57±0.15	1.60 ± 0.10	/
S-1x100	1.80 ± 0.20	1.84 ± 0.25	/

Initiation and propagation fracture toughness as a function of the interleaved PEEK film thickness is shown in Figure 9 for the three different process technologies used. It is worth noticing that the x-axis is the nominal thickness of the added films and not the actual matrix rich bond line thickness after processing, which in some cases may be smaller due to outflow of matrix. Measurements of the actual matrix rich bond line thickness were difficult to perform from the micrograph and therefore not used. The trend is similar for all three processes, where the fracture toughness increases with increasing interleave thickness. No

³³⁶ 337

Table 3: Fracture toughness values for initiation and propagation for all the sample tested. The error was calculated as one standard deviation of the set of values within one sample.

significant differences can be observed between the three processes and between initiation and propagation. Despite the similar average toughness values, the stamp forming process resulted in a higher scatter within the sample. This may be due to a non-uniform pressure and temperature distribution during fusion bonding, which may locally have resulted in incomplete wetting or healing.



Figure 9: Interlaminar fracture toughness as a function of interleaved thickness for the three processes,
 autoclave consolidation, press consolidation, and stamp forming.

352 3.3. Fractography

The fracture behaviour of the different samples is compared in this section using cross-353 354 sectional micrographs and fractography analysis. First, a comparison between samples without film interleaving and with film interleaving is shown. Later, the comparison between 355 samples with fibre migration and without fibre migration is presented. Three types of images 356 were used for the analyses. Figure 10 shows cross-sectional micrographs perpendicular to the 357 crack propagation direction. These micrographs show the position of the crack at a single 358 instant, though they do not give information about how the crack propagates along the length 359 of the specimen. Figure 11 shows pictures of the optical cross-sectional micrographs with the 360 cross-sectional plane oriented at 20° with respect to the crack propagation direction. These 361 362 pictures show how the crack propagates through the specimen. Finally, Figure 12 shows the SEM micrographs of the fracture surfaces where the interaction between fibre and matrix and 363 the deformation of the matrix after testing can be observed. 364

The comparison between samples with and without PEEK film interleaving is presented here. 365 Due to the similarity among the images within each test group, only one representative 366 micrograph per group is shown. The left micrograph in Figure 10 shows a specimen without 367 interleaving. It can be seen that the crack is located at the centre plane of the specimen. The 368 right micrograph shows a specimen with interleaving. In this case, the crack is located close 369 to the interface between the fibre rich and the matrix rich region, slightly out of the centre of 370 371 the specimen. Other images in the same cross-sectional plane, thus to the left or right of the presented image, showed the same crack at the interface between matrix rich region and the 372 373 matrix poor region of the upper substrate.





The straightness of the crack along the propagation direction was analysed using the 20° cross-sectional micrographs. The top micrograph in Figure 11 shows a non-interleaved specimen, while the bottom shows an interleaved specimen. It can be noted that the crack path remains flat when the specimens are not interleaved as is shown in the top. However, the crack propagates with some waviness, seemingly avoiding the matrix rich region in the centre, and this is the case for interleaved specimens shown in the bottom image.

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385





393 A comparison between the fracture surface of an interleaved and a non-interleaved specimen is shown in Figure 12, while Figure 13 shows a schematic illustration of the accompanying 394 cross-section. The SEM image on the left shows that the fracture surface of a non-interleaved 395 specimen is characterised by fibre imprints in the matrix and bare fibres. Also, microscale out 396 of fracture plane plastic deformation of the matrix can be observed, which is a typical feature 397 of the fracture surface of carbon/PEEK laminates tested in mode I [23]. This deformation is 398 present at the edges of the fibres in the schematic view. The SEM micrograph on the right 399 shows that the fracture surface of an interleaved specimen is characterised by two distinct 400 regions. The first region shows a combination of fibre imprints in the matrix and bare fibres, 401 similar to the case of the non-interleaved sample. The second region is characterised by a 402 matrix rich area where large microscale plastic deformation of the matrix can be observed as 403 evidenced by the white polymer regions. 404





Figure 12: Scanning electron micrograph of the fracture surfaces. Left: Autoclave consolidated specimen with no interleaving. Right: interleave press consolidated specimen.



Figure 13: Schematic view of a cross-section of a fracture surface. Left: Sample without matrix interleaving.
 Right: Sample with matrix interleaving [Figure 13 near here]

The interleaved samples can be subdivided into two groups. The first comprises the samples 410 prepared using a slow process (autoclave and press consolidation), while the second group 411 consists of samples manufactured using the fast process (stamp forming). Figure 14 and 412 Figure 15 show the cross-sectional micrographs and their schematic illustration for both 413 groups, respectively. The crack shape and location look similar for both cases, irrespectively 414 of whether fibre migration occurred or not. The crack seems to remain at the interface 415 between the matrix rich and matrix poor region. The crack path was observed to alternate 416 between the top and the bottom substrate trying to minimise the crack path length through the 417 matrix rich region, similar to what is observed in Figure 11. 418



Figure 14: Cross-sectional micrographs perpendicular to the crack propagation direction. Left) autoclave
 consolidated specimen with interleaving. Right) stamp formed specimen with interleaving.

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Figure 15: Schematic view of a cross-section micrograph of an interleaved specimen. Left: Specimen with
 fibre migration as obtained using autoclave or press fusion bonding. Right: Specimen without fibre migration as
 obtained using stamp fusion bonding.

425 **4. Discussion**

In this section, the results obtained are combined and discussed with the purpose of getting a deeper understanding of the mechanisms that govern the interlaminar fracture toughness of fusion bonded joints that present a matrix rich bond line.

The interlaminar fracture toughness improves by increasing the matrix rich bond line thickness, as was expected. This is true even if the crack does not propagate through the matrix rich area but through the matrix poor area or the interface between the matrix-poor (one of the two substrates) and matrix-rich (the interleave) regions. This phenomenon was explained by Hojo et al. [13] for interleaved laminates, who reasoned that by increasing the interleave thickness, even if the crack does not propagate fully through the matrix rich area, the plastic yield zone in front the crack tip is still less constrained by the fibres and is

therefore allowed to increase in size. Moreover, it was proposed that when the matrix rich 436 region is smaller than the maximum plastic yield zone size, the crack path migrates towards 437 the weakest region, i.e. the boundary between matrix poor and matrix rich regions, resulting 438 in adhesive failure [13]. However, when the thickness of the matrix rich region increases 439 further than the plastic yield zone, the crack will remain within this region resulting in a 440 cohesive failure of the interleave [13]. The change in plastic zone size and the position of the 441 442 crack propagation path is schematically represented in Figure 16. A larger plastic yield zone area means that more energy will be dissipated, which is reflected by a higher interlaminar 443 444 fracture toughness. The SEM fractography, as presented in Figure 12, confirmed that more plastic deformation is observed in the interleaved samples compared to the samples without 445 additional matrix at the interface. Besides, the tortuosity of the crack path, as shown in the 446 lower micrograph in Figure 11, may also contribute to an increased fracture toughness 447



448Figure 16: A schematic explanation of crack growth behaviour and plastic zone development having a radius449 r_y . Left) Base material, no interleaved. Centre) Material with an interleaving thickness below maximum plastic450yield zone $(2r_y)$. Right) Material interleaved with a thickness above the maximum plastic yield zone. Figure451adapted from [13].

Plastic deformation of the matrix was found to be the main mechanism to increase the interlaminar fracture toughness of the interleaved specimens. Nevertheless, as the plasticity is localised only at the fracture surface, the global linear elastic behaviour of the specimen during testing was retained. As such, the tests still comply with the LEFM assumption, which makes the comparison of the values obtained for the different samples acceptable.

457 It was suggested that the maximum theoretical toughness of an interleaved system is the 458 toughness of the pure polymer, which is reached when the interleave thickness is equal or larger than two times the plastic yield radius (Figure 16 right) [16, 18]. A first approximation of the plastic zone radius (r_y) of a polymer can be calculated following Irwin's plastic zone model for plane strain reported by Ozdil and Carlsson [19] (Equation (2)).

$$r_{y} = \frac{1}{4\pi} \left(\frac{K_{IC}}{\sigma_{y}} \right)^{2} \left(\frac{3}{2} \left(1 - 2\nu^{2} \right) \right), \tag{2}$$

where K_{IC} is the stress intensity factor which relates to the fracture toughness of the polymer, σ_y is the tensile yield stress of the polymer, and v is the Poisson's ratio. The following expression can be used to relate the stress intensity factor K_{IC} to the energy release rate G_{IC} in case of a plane strain situation:

$$G_{IC} = \frac{(1 - v^2)K_{IC}^2}{E},$$
(3)

where E is the elastic modulus of the polymer. Material data from the literature is required to 466 calculate the maximum theoretical fracture toughness of this system. The following values 467 were reported in the data sheet of Victrex PEEK 150, which is used as matrix in the prepregs; 468 tensile yield point (σ_v) of 105 MPa, an elastic modulus (E) of 3.5 GPa and a poisson's ratio 469 (v) of 0.4. The stress intensity factor K_{IC} for Victrex PEEK 450G, a similar grade of the 470 polymer use for interleaving, is reported in literature to lie between 3 to 6 MPa \cdot m^{1/2} [24]. An 471 average value of 4.5 MPa \cdot m^{1/2} will be used for the following analysis. Using Equation (2) 472 and Equation (3) a plastic radius of 0.225 mm and an energy release rate of 4.8 kJ/m^2 can be 473 calculated for this polymer. The result shows that the pure polymer has almost two times 474 higher toughness than the interlaminar fracture toughness measured in the experiments in this 475 study. Nevertheless, the theoretical matrix rich bond line thickness required to develop the 476 fracture toughness (0.45 mm) was not tested in the experiments reported in this work, where 477 a maximum matrix rich bond line thickness of 0.2 mm was tested. Thus, the fracture 478 toughness is expected to keep increasing by increasing matrix rich bond line thickness. 479

480 Similar observations were made for thermoset composites [18]. For these material systems,
481 smaller interleave thicknesses are required to achieve the maximum (i.e. polymer) toughness,
482 which is caused by the more brittle nature of thermosets compared to thermoplastics.

The matrix rich bond line thickness after processing was observed to be not uniform, this is 483 particularly true for the autoclaved samples where material flow occurs during processing. 484 This non uniformity and the difficulty to distinguish between the matrix rich and matrix poor 485 486 region makes it difficult to evaluate the actual matrix rich bond line thickness after processing. Besides, this non uniformity may, moreover, also be one of the causes for the 487 488 unstable crack propagation observed as it most probably resulted in a non-uniform interlaminar fracture toughness along the crack path. It is known that the unstable crack 489 propagation may occur when the crack propagates from a region of higher toughness to a 490 region of lower toughness, as the elastic energy stored in the specimen is more than required 491 for making the crack to propagate in a stable manner. Or more precisely formulated unstable 492 crack propagation may occur at the locations where dG/da exceeds dR/da [25]. 493

The high cooling rates observed during stamp forming may have induced a different level of 494 crystallinity compared to the other two (slower) processing techniques, possibly affecting the 495 measured toughness values. DSC experiments showed, however, that a non-interleaved press 496 consolidated specimens and non-interleaved stamp formed specimens have the same level of 497 crystallinity of approximately 35% using an enthalpy of crystallisation value of 130 (J/g) [26] 498 499 with a matrix weight fraction of 34%. Although the difference in thermal history may have resulted in different crystal morphologies, this seemed to have no effect on the measured 500 toughness. 501

In conclusion, it seems that the interlaminar fracture toughness is independent of the three processes used in this work. It solely depends on the interleave thickness and is not affected by fibre migration. The amount of fibres, in the fibre migration region, is too small to

505 constrain the plastic zone, nor does it result in excessive fibre bridging.

506 **5. Conclusions**

The effect of a matrix rich interface and fibre migration on the fracture toughness of fusion 507 bonded samples was studied. For this purpose, samples were prepared using manufacturing 508 having different characteristic processing times, technologies namelv: autoclave 509 consolidation, press consolidation, and stamp forming. Autoclave and press consolidation 510 were considered as slow processes, while stamp forming was considered as a fast process 511 with conditions similar to those in many welding techniques. Matrix rich bond lines with 512 different thicknesses were obtained by interleaving matrix films at the interface between two 513 adherents prior to fusion bonding. 514

Microscopy showed that two regions can be identified in the interleaved samples, namely the 515 matrix poor adherent(s) and a matrix rich bond line. The processing time, moreover, affected 516 the matrix rich bond line morphology. On the one hand, fibre migration from the adherents 517 into the matrix rich bond lines was observed during (the slower) press and autoclave 518 consolidation, resulting in a matrix rich zone with many loose fibres. On the other hand, fibre 519 migration was prevented during (the faster) press forming, resulting in a bond line with very 520 few or no fibres. Double cantilever beam experiments were performed and showed that the 521 increase in the matrix rich bond line improves the fracture toughness. This increase is 522 attributed to the development of microscale matrix plastic deformation. Moreover, it was 523 shown that fibre migration has a negligible effect on the interlaminar fracture toughness, i.e. 524 the toughness only depends on the matrix interleave thickness. 525

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531 7. Reference

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