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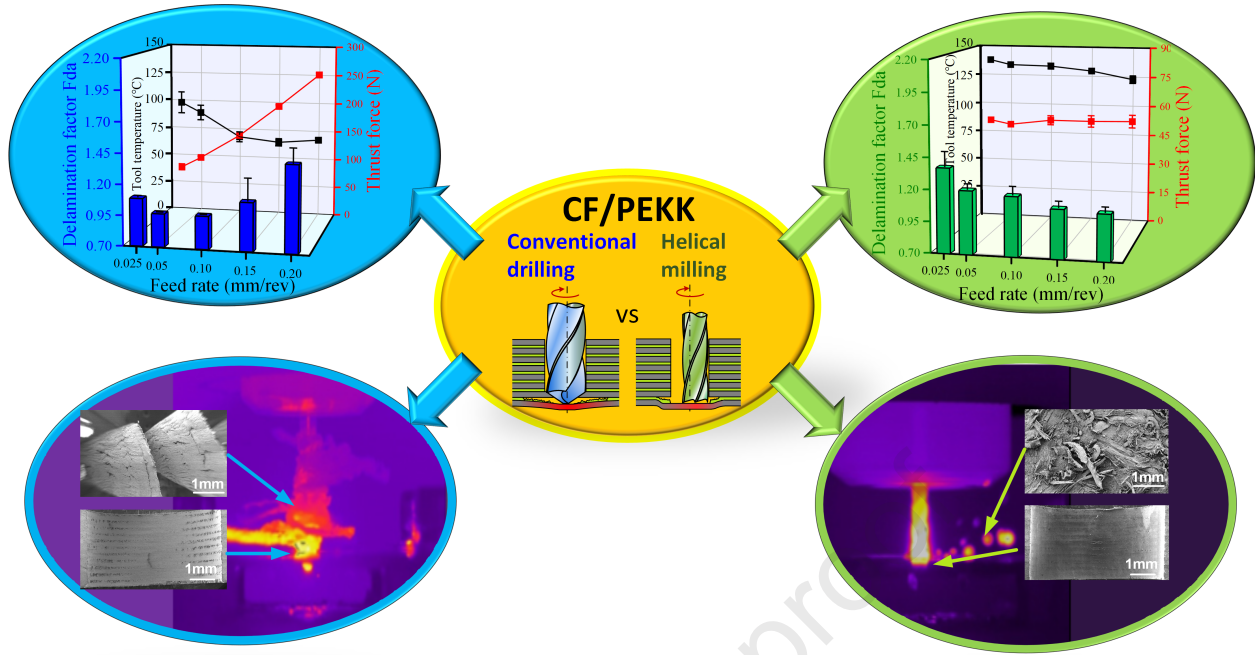
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**CRedit authorship contribution statement**

**Jia Ge:** Conceptualization, Methodology, Investigation, Writing - Original Draft **Giuseppe Catalanotti:** Supervision, Writing - Review & Editing **Brian Falzon:** Writing - Review & Editing **John McClelland:** Software, Resources, Writing - Review & Editing **Colm Higgins:** Resources, Writing - Review & Editing **Yan Jin:** Supervision, Writing - Review & Editing, Funding acquisition, Project administration **Dan Sun:** Conceptualization, Methodology, Investigation, Writing - Review & Editing, Funding acquisition, Project administration.

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# Towards understanding the hole making performance and chip formation mechanism of thermoplastic carbon fibre/polyetherketoneketone (CF/PEKK) composite

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## Abstract

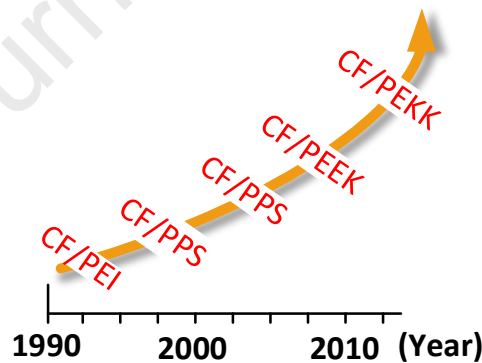
Here, we report the first study on the hole making performance of thermoplastic carbon fibre/polyetherketoneketone (CF/PEKK). Different hole making methods (conventional drilling vs. helical milling) have been compared and the effect of different feed rates has been investigated. The effect of thermal-mechanical interaction on the resulting hole damage has been elucidated for the first time for carbon fibre reinforced thermoplastics (CFRTPs) hole making. In the material science dimension, advanced material characterization techniques have been deployed to reveal the material removal mechanisms at microscopic scale and unveil the underlying material structural change at a molecular level. Results show that the delamination damage of CF/PEKK is a result of the thermal-mechanical interaction. For conventional drilling, the high machining temperature (at low feed rate < 0.1 mm/rev ) has a stronger influence on the delamination damage and the delamination starts to show stronger dependence on the thrust force at high feed rate > 0.1 mm/rev. In contrast, helical milling generates a much higher machining temperature which plays a more predominant role in the associated delamination damage. Microstructural analysis shows that all the hole surfaces feature matrix smearing, as a result of combined in-plane shear stress and high machining temperature. Conventional drilling leads to more severe hole wall microstructural damage (matrix loss and surface cavity) as compared to helical milling. Finally, thermal analysis reveals that the hole making process has led to significantly increased crystallinity in the PEKK matrix as a result of the strain-induced crystallization under the combined effect of shear stress and high temperature.

**Keywords:** CFRTP, CF/PEKK composite, hole making, delamination damage, polymer crystallinity

## 1 1. Introduction

2 Carbon fibre reinforced plastics (CFRPs) have been intensively deployed in aerospace and automotive  
 3 industries nowadays, as their high specific strength favours the lightweight design and energy saving  
 4 requirements of the modern industry. In recent years, there is a surge of interest in the application of  
 5 novel carbon fibre reinforced thermoplastics (CFRTPs) in the manufacturing sector, as these materials  
 6 demonstrate several prominent advantages. Compared to the conventional thermosetting CFRPs,  
 7 CFRTPs require less stringent storage condition and have infinite shelf-life under ambient conditions.  
 8 Their out-of-autoclave processing can achieve much shorter manufacturing cycle and requires less  
 9 energy consumption [1]. The thermoplastic nature of the material also endows CFRTP with better  
 10 recyclability and greater reparability [2], which can contribute greatly to the carbon emission reduction  
 11 in sustainable manufacturing.

12 Amongst the available CFRTPs, carbon fibre reinforced polyetherketoneketone composite (CF/PEKK)  
 13 stands out for its exceptional mechanical properties (tensile strength  $\sim 2.4$  GPa), strong chemical  
 14 resistance, high thermal stability (glass transition temperature  $T_g \sim 160$  °C) and wide processing  
 15 window (330 – 380 °C) [3]. The roadmap for development of thermoplastic composites in Europe,  
 16 supported by Airbus and a variety of national aerospace consortia (see Fig. 1), suggests that CF/PEKK  
 17 will be top choice of composite in high end applications such as primary and secondary structural  
 18 components such as leading edges, floor panels, wing spars and engine pylons in future aircrafts [4–  
 19 7].



20  
 21 Fig. 1 EU development road map of thermoplastic composites [8]

22 Like many other CFRTPs, CF/PEKK parts can be manufactured into net shape through compression  
 23 moulding or assembled by means of welding [9]. However, secondary processing such as mechanical  
 24 riveting through fastener holes still remains an imperative manufacturing process, particularly in  
 25 joining of dissimilar materials (e.g. metal/CFRTP joining) and load bearing components [10]. In  
 26 composite part assembly, conventional drilling (CD) is the most commonly deployed hole making  
 27 technique due to its great efficiency. Other emerging hole making technology, such as helical milling  
 28 (HM), has also attracted increasing attention [11]. In HM, the cutter proceeds in both the tangential

1 and the axial directions thus providing combined frontal and peripheral cutting. Compared with CD,  
2 reduced burr formation, improved hole geometrical accuracy and smaller cutting force have been  
3 reported in HM of aeronautical alloys [12] and thermoset CFRPs [13].

4 To date, a large body of literature has been dedicated to the study of hole making performance of  
5 conventional thermosetting CFRP, covering wide ranging research topics including process  
6 optimization [14–16], cutting tool design [17,18], cutting force modelling [19–21] and delamination  
7 damage modelling [22–24]. The readers are referred to the review papers [18,25,26] in this field for  
8 more information. In comparison, the number of studies concerning hole making performance of  
9 CFRTP is very limited. The first study on hole making of CFRTP was conducted by Hocheng and Puw  
10 [27] on drilling machinability of CF/acrylonitrile butadiene styrene (CF/ABS). They found that the  
11 thrust force in drilling of CF/ABS was proportional to feed rate and highly finished surface ( $R_a < 1\mu\text{m}$ )  
12 can be achieved for a wide range of drilling parameters (cutting speed = 1.96 – 50 m/min and feed rate  
13 = 30 – 3000 mm/min). In a later study by Hocheng et al. [28], the drilling performance of CF/polyether  
14 ether ketone (CF/PEEK) and CF/Polyphenylene sulfide (CF/PPS) was reported. The results showed  
15 that thrust force, hole-exit burr and delamination all showed significant increase with increasing feed  
16 rate. CF/PEKK composite showed less severe delamination damage due to its high interlaminar  
17 fracture toughness. Xu et al. [29] compared the hole making performance of thermoplastic  
18 CF/polyimide (PI) and thermosetting CF/epoxy composites under different drilling parameters. The  
19 results suggested that CF/PI demonstrated better machinability in terms of lower thrust  
20 force/machining temperature and better hole geometrical accuracy. For CF/PI, increasing the cutting  
21 speed helped to reduce cylindricity errors, but at the price of higher specific energy consumption. Xu  
22 et al. [30] also conducted a full-factorial experimental study to investigate the effect of cutting speed  
23 and feed rate on the drilling delamination damage in CF/PEEK and CF/PI. The results showed that  
24 delamination damage in both composites can be reduced by increasing the cutting speed or decreasing  
25 the feed rate. Lopez-Arraiza et al. [31] compared the performance of three different cutters (reamer,  
26 twist drill and drill-end cutter) in hole making of carbon fibre reinforced poly-cyclic butylene  
27 terephthalate (CF/pCBT). Results showed that the tool geometry played a crucial role in the resulting  
28 hole delamination damage and that the twist drill caused the least delamination damage due to the  
29 clearer cutting of the carbon fibre and the pCBT matrix. According to Hocheng et al. [28], the hole  
30 making performance of CFRTPs depend on machining parameters, the cutting tools used, as well as  
31 the properties of the thermoplastic matrix and the fibre content. To date, no work has been conducted  
32 to investigate the hole making performance of high performance CF/PEKK composite and  
33 comparative studies on different hole making methods are in scarcity in CFRTP research.

34 It is known that manufacturing of the thermosetting CFRPs involves a curing process. The resin  
35 undergoes a non-reversible chemical reaction and form rigid, covalently bonded crosslinks which  
36 maintain their rigidity under elevated temperature [32]. In contrast, CFRTPs show typical temperature-

1 dependent property. They soften (lose their mechanical properties) as the temperature approaches its  
2  $T_g$  [33] due to the relatively weak Van der Waals' force between their molecular chains. As such, the  
3 machining temperature can significantly influence the hole making performance of CFRTPs and  
4 should be strictly monitored and controlled. With this in consideration, Xu et al. [29] measured the  
5 CF/PI drilling temperature under different feed rates and cutting speeds. Their results suggested that  
6 increasing the feed rate can lead to reduced drilling temperature, whereas cutting speed did not have  
7 any significant effect. In another study by Xu et al. [30], the CF/PEEK drilling temperature was  
8 measured and results showed that the high machining temperature can soften the matrix and the highly  
9 ductile PEKK matrix was recast on to the hole wall, generating the distinct matrix smearing. Similar  
10 matrix smearing phenomena were also reported by Hocheng and Puw [27], Ferreira et al. [34] and Kim  
11 et al. [35] in drilling of carbon fibre reinforced acrylonitrile butadiene styrene (CF/ABS) and CF/PPS.  
12 Although above researchers have studied the machining temperature in hole making of several  
13 CFRTPs, they failed to elucidate the impact of high machining temperature on the resulting hole  
14 damage formation (especially delamination and hole microstructural damage) and the associated  
15 material removal mechanisms.

16 Recently, several researchers studied the chip formation in drilling of CFRTPs with the aim of  
17 establishing the associated material removal mechanisms. Hocheng and Puw [27] compared chips  
18 produced from drilling of thermoplastic CF/ABS and thermoset CF/epoxy composites. It was found  
19 that CF/ABS featured long continuous chips, which was associated with the better ductility of the  
20 thermoplastic matrix and the considerable plastic deformation it experienced during the machining  
21 process. In contrast, fragmented chips were produced in drilling of CF/epoxy, indicating the material  
22 removal was by means of brittle fracture. In their later work, Hocheng et al. [28] reported that the  
23 extent of plasticity in chip formation and the chip length were dependent on the level of fibre content  
24 and the deformation behaviour of the matrix. Ahmad et al. [36] and Xu et al. [30] found that the  
25 elongated CFRTP chips adhered to the drill bit main cutting edge, which led to severe tool clogging,  
26 deteriorated tool cutting performance and accelerated tool wear [36]. So far, no work has been done to  
27 investigate the chip microstructure and the associated material removal mechanism of CF/PEKK. The  
28 polymer chains within CFRTP matrix will be highly mobile when subjected strong mechanical  
29 shearing and high temperature during the hole making process, there has been no study dedicated to  
30 understanding the microstructural evolution of machined CFRTP at a molecular level.

31 In this paper, the hole making performance of advanced thermoplastic CF/PEKK composite is reported  
32 for the first time. The effect of different hole making methods (CD and HM) and feed rates on the  
33 resulting delamination and hole wall microstructural damage will be discussed, considering the  
34 thermal-mechanical interaction arise from the hole making process. The microstructure of the formed  
35 chips will be analysed in detail to reveal the fundamental material removal mechanism. In addition,  
36 thermal analysis will be carried out for the chips to elucidate the material molecular structural evolution

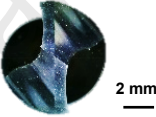
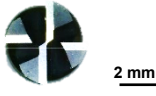
1 during the machining process. This study will not only reveal the fundamental material science  
 2 involved in the hole making process, but also provide a parametric and methodological guidance for  
 3 assembly of load-bearing aircraft parts involving CFRTP components (such as composite-metal  
 4 stacked structures in frames and wings) [37,38].

## 5 2. Experiment setup

### 6 2.1. Materials

7 CF/PEKK (3 mm thick, fibre content 66 wt%) laminates consisting of 22 plies of unidirectional  
 8 prepregs with a stacking sequence of  $[0/90]_{11}$  were manufactured by consolidation method [39]  
 9 following recommended protocol. The thermoplastic PEKK matrix has a  $T_g$  of 160 °C and a melting  
 10 temperature ( $T_m$ ) of 337 °C. The coupons for hole making experiment measures 120 mm × 120 mm ×  
 11 3 mm. Twist drill bit for CD (namely AT Drill) and end milling cutter for HM (namely AT Mill), were  
 12 supplied by Changzhou Aitefasi Tools Co., Ltd. Changzhou, China. More details of the cutting tools  
 13 can be found in Table 1.

14 Table 1 Details of the cutting tools used in this study

Tool abbreviation	Tool name	Image	Diameter (mm)	Flute number	Point angle (°)	Helix Angle (°)
AT Drill	Aitefasi 3D drill bit (TiAlN coated)		6	2	104	30
AT Mill	Aitefasi standard square end mill (TiAlN coated)		4	4	-	30

15

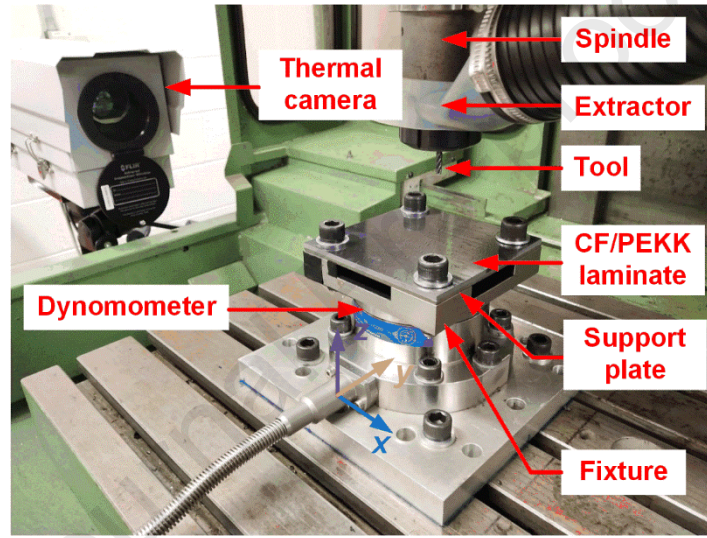
### 16 2.2. Hole making experiments

17 Both CD and HM experiments were conducted under dry condition at room temperature. Since axial  
 18 feed rate is a key parameter in relation to hole delamination formation due to Mode-I opening fracture,  
 19 five different levels of feed rates ( $F = 0.025, 0.05, 0.10, 0.15, 0.2$  mm/rev) were selected for CD and  
 20 HM. A cutting speed of  $V_c = 50$  m/min was selected to avoid generation of excessive heat and thermal  
 21 damage of the matrix [40,41]. A tangential feed 0.04 mm/tooth was selected for HM based on previous  
 22 research [40]. All machining parameters were selected based on the published literature [10,30,41] and  
 23 per tool manufacturers' recommendation. Both CD and HM were carried out on a Deckle FP3A 3 axis  
 24 Computer Numerical Control (CNC) machine, which is equipped with an in-house chip extraction  
 25 system to comply with health and safety regulations. Fig. 2 illustrates the details of the experiment  
 26 setup. A 3 mm thick steel plate with 10 mm diameter pre-drilled holes was placed on the CFRTP drill-



1 exit side, to ensure consistent supporting stiffness for each hole made [42]. During the hole making  
 2 process, the thrust force was recorded by a Kistler 9272 4 component dynamometer with a 5070A  
 3 charge amplifier and a 5697 data acquisition Unit. A thermal camera (FLIR A6751, temperature range:  
 4 -20 to 350 °C, frame frequency: 125 Hz) was used for real time temperature measurement. The camera  
 5 calibration and temperature measurement procedures were in accordance with ASTM E1933-14. The  
 6 image of the cutting tool tip emerging from the hole-exit was captured by the thermal camera and a  
 7 region of interest (ROI) was selected on the software to calculate the average tool tip temperature, see  
 8 Appendix Fig. A.1. Three holes were produced and inspected under each set of parameters for  
 9 repeatability. The cutting tools were replaced after making every three holes to eliminate the effect of  
 10 tool wear.

11



12  
 13 Fig. 2 Image showing the setup of the hole making platform

14

### 15 2.3. Hole quality evaluation

16 The hole-exit delamination was imaged by Alicona infinite focus G5 microscope (Bruker, UK, 2.5X  
 17 magnification). The exit delamination (Fig. 3 grey area) can be quantified by the delamination factor  
 18  $F_{da}$  [43], which can be calculated following Eq. 1-3:

$$F_{da} = \alpha \frac{D_{max}}{D_{nom}} + \beta \frac{A_{max}}{A_{nom}} \quad (1)$$

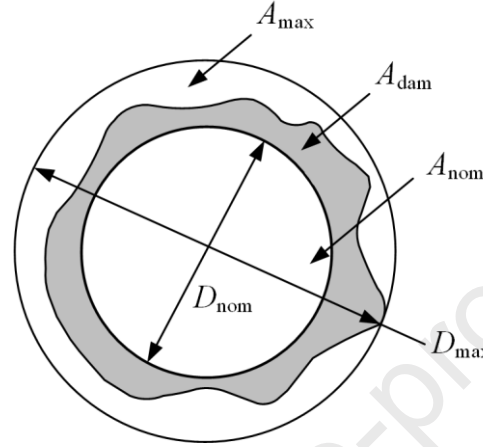
$$\beta = \frac{A_{dam}}{A_{max} - A_{nom}} \quad (2)$$

$$\alpha = 1 - \beta \quad (3)$$

19 where  $D_{max}$  is the maximum diameter of damaged area,  $D_{nom}$  is the nominal diameter of the hole,  
 20  $A_{max}$  is the area related to the maximum diameter of the delaminated zone ( $D_{max}$ ),  $A_{nom}$  is the

1 nominal hole area and  $A_{dam}$  is the damaged area. The delamination factor was calculated through  
 2 image analysis using MATLAB R2019b software.

3 The hole wall microstructures and chip morphology were inspected by Scanning Electron Microscopes  
 4 (SEMs) FlexSEM 1000 (Hitachi Ltd., Japan) under an acceleration voltage of 5 kV. The samples were  
 5 sputter-coated with gold before SEM observation.



6  
 7 Fig. 3 Schematic of hole-exit delamination

#### 8 2.4. Thermal analysis

9 The thermal properties of the CF/PEKK samples were measured using a thermogravimetric analysis  
 10 (TGA) and differential scanning calorimetry (DSC) thermal analysis system (TGA/DSC2, Switzerland)  
 11 under a N2 atmosphere in a temperature range 30 to 550 °C and a heating rate of 10 °C/min.

12 The crystallinity  $\chi_c$  of the samples is calculated following Eq. 4 [44]:

$$\chi_c = \frac{\Delta H_m - \Delta H_{cc}}{\Delta H_{100\%}(1 - W_f)} \times 100\% \quad (4)$$

13 Where  $\Delta H_m$  is the melting enthalpy,  $\Delta H_{cc}$  is the cold crystallization enthalpy,  $\Delta H_{100\%} = 130$  J/g is  
 14 the melting enthalpy of the theoretical 100% crystalline PEKK [45] and  $W_f = 66\%$  is the fibre  
 15 weight fraction in the CF/PEKK composite.

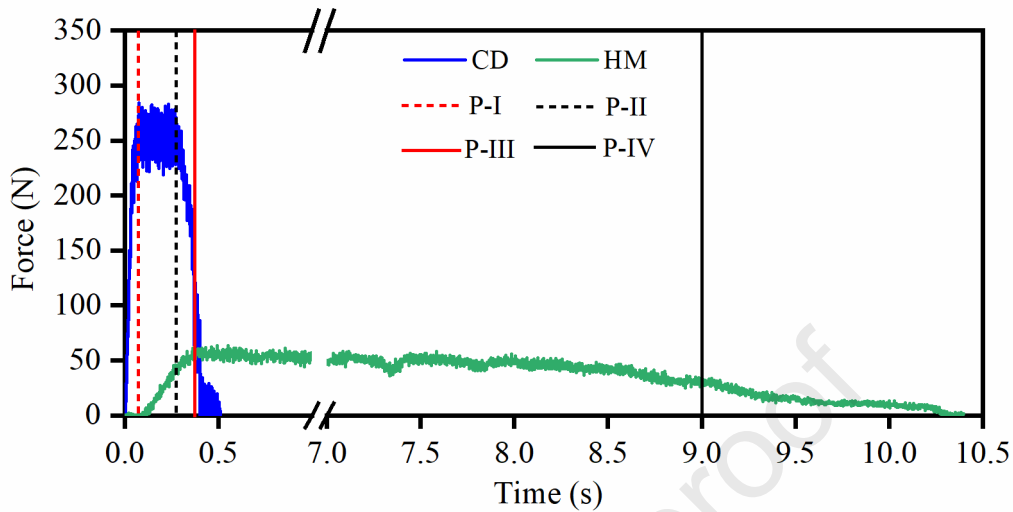
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### 17 3. Results and discussion

#### 18 3.1. Thrust force

19 In hole making of CF/PEKK, thrust force (also known as axial force) corresponds to the level of load  
 20 exerted on the workpiece by the cutting tool. The thrust force is thought to be directly related to the  
 21 hole-exit delamination damage caused by Model-I opening fracture, particularly in thermosetting  
 22 CFRP [46]. The typical force signals in hole making of CF/PEKK are shown in Fig. 4. The thrust force  
 23 signals for both CD and HM feature three phases, namely, cutting in, stable cutting and cutting out  
 24 phases. The thrust force measured during the stable cutting phase is representative of the force exerted

1 on the workpiece, therefore the average thrust force of the stable cutting phase was used for further  
 2 analysis.



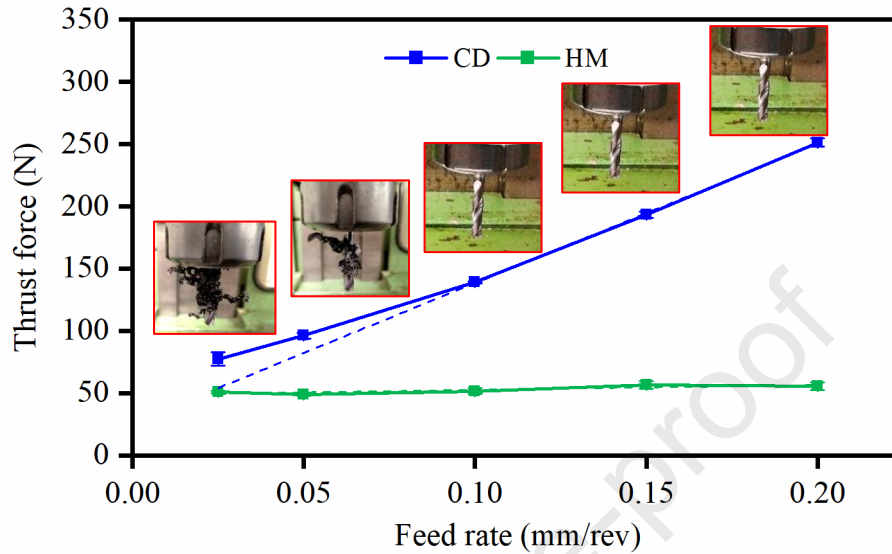
3  
 4 Fig. 4 Representative thrust force signal in CD and HM of CF/PEKK with feed rate  $F=0.2$  mm/rev.  
 5 (P-I and P-II depict the starting and ending of the CD stable cutting phase; P-III and P-IV depict the  
 6 starting and ending of the HM stable cutting phase)

7 The average stable cutting phase thrust force generated under different feed rates is shown in Fig. 5.  
 8 HM generated a consistently low thrust force ( $\sim 50$  N), which is independent of the feed rate used.  
 9 Under CD however, the measured thrust forces are significantly higher than that of HM and shows a  
 10 clear increasing trend with the feed rate.

11 The difference in thrust force seen in CD and HM can be attributed to the distinctly different material  
 12 removal mechanisms involved in the two processes. For CD, the cutting speed near the borehole centre  
 13 approaches zero. The material removal is achieved by extrusion in the axial direction rather than  
 14 cutting [13], which induces a much higher axial thrust force. For HM, a large portion of material  
 15 removal is achieved by the peripheral cutting edge as the tool travels along its helical tool path. As a  
 16 result, less force is exerted in the axial direction.

17 It can be seen from Fig. 5 that increasing feed rate from 0.025 mm/rev to 0.2 mm/rev has led to  $\sim 224\%$   
 18 increase in thrust force for CD. The drastic increase can be attributed the thicker material removed per  
 19 tool revolution under the higher feed rate, as shown in Appendix C Fig.C.1. This creates more  
 20 resistance for tool progression, as greater work is required to achieve deformation/removal of thicker  
 21 material [19]. For CD, the thrust force showed a quasi-linear correlation with the feed rate, such linear  
 22 correlation is similar to that reported by Guo et al. [19] in their thrust force models for CD of CF/epoxy  
 23 composite. The slight deviation from the linearity at lower feed rates ( $F \leq 0.05$  mm/rev) may be  
 24 attributed to the tool clogging (see Fig. 5 insets). When tool clogging occurs, extra force is required to  
 25 evacuate the stuck chips, and this is added onto the thrust force measured in the axial direction. Tool

1 clogging has also been reported by Ahmad et al. [36] and Xu et al. [30] in their study on drilling of  
 2 CF/PEEK. Such phenomenon is unique to drilling of CFRTP (not reported in thermosetting CFRP  
 3 drilling) and should be considered for thrust force modelling in future studies.



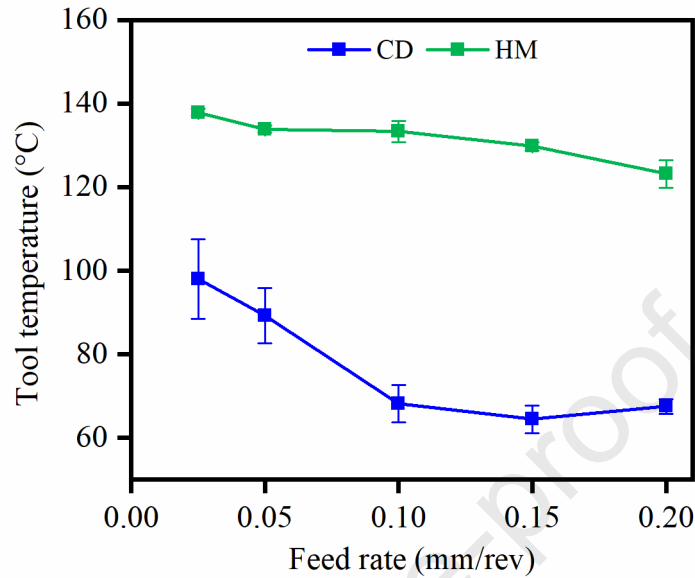
4 Fig. 5 Effect of feed rate on the thrust force under different hole making conditions (insets: evidence  
 5 of tool clogging by chips on drill bits)  
 6

### 7 8 3.2. Tool temperature

9 The heat generated during the hole making process is primarily due to the shear deformation of  
 10 CF/PEKK and the friction between the cutting tool and the workpiece [47]. The accumulated heat is  
 11 then distributed to the chips, the workpiece and the cutting tool. According to previous experimental  
 12 and numerical simulation studies by Thirukkumaran et al. [48], the workpiece temperature follows a  
 13 quasi-linear increasing trend with cutting tool temperature. Given the workpiece hole-exit temperature  
 14 cannot be measured directly with our existing experimental setup, hole-exit tool temperature was used  
 15 to inform the machining temperature. The tool temperature under different hole making methods and  
 16 feed rates is shown in Fig. 6. In general, the tool temperature decreases with increasing feed rates for  
 17 both CD and HM. For CD, the tool temperature decreased from 98.03°C to 68.20°C (a 30.43%  
 18 reduction) as the feed rate increased from 0.025mm/rev to 0.1 mm/rev. The tool temperature stabilized  
 19 at ~ 70 °C for feed rate > 0.1 mm/rev. In contrast, the tool temperature recorded for HM is much  
 20 higher for all the feed rates used. The highest HM tool temperature (137.9°C) was recorded at F =  
 21 0.025mm/rev, which gradually decreased to 123.15°C at F = 0.2 mm/rev, marking a 10.70% overall  
 22 reduction.

23 For both CD and HM, the lower tool temperature recorded at a higher feed rate can be attributed to the  
 24 reduced tool-workpiece contact time (i.e., tool engagement time, see Appendix Table B.1). It is clear  
 25 that the tool engagement time decreases with increasing feed rate and shorter tool engagement time

1 creates less thermal energy accumulation during the machining process. The considerably longer tool  
 2 engagement time in HM (see Appendix B Table B.1) is accountable for its much higher tool  
 3 temperature.



4  
 5 Fig. 6 Tool temperature of CD and HM against feed rate  
 6

### 7 3.3. Assessment of delamination damage

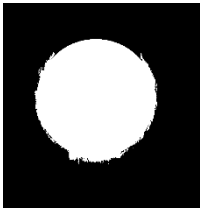
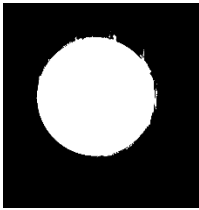
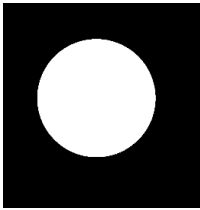
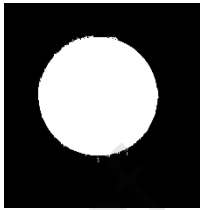
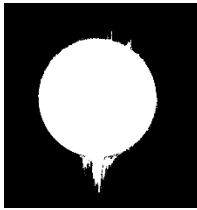
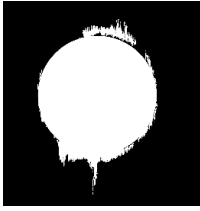
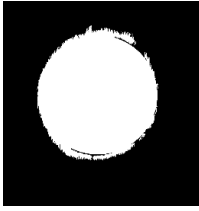

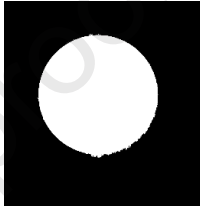
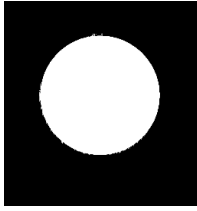
8 Delamination, a commonly seen defect at the CFRP hole entry and hole exit, accounts for ~ 60% of  
 9 composite laminates rejection during assembly [49]. Hole delamination reduces the assembly tolerance  
 10 and compromises the component's reliability under fatigue loads [46], and therefore should be strictly  
 11 monitored/controlled during the hole making process. Table 2 shows the binary images of delamination  
 12 damage produced under different hole making conditions. Although the literature suggests HM induces  
 13 less hole delamination in hole making of thermosetting CFRP [50], our results show that the extent of  
 14 hole delamination under different hole making condition is actually feed rate dependent. Under low  
 15 feed rate, the delamination seen for HM is more severe than CD.

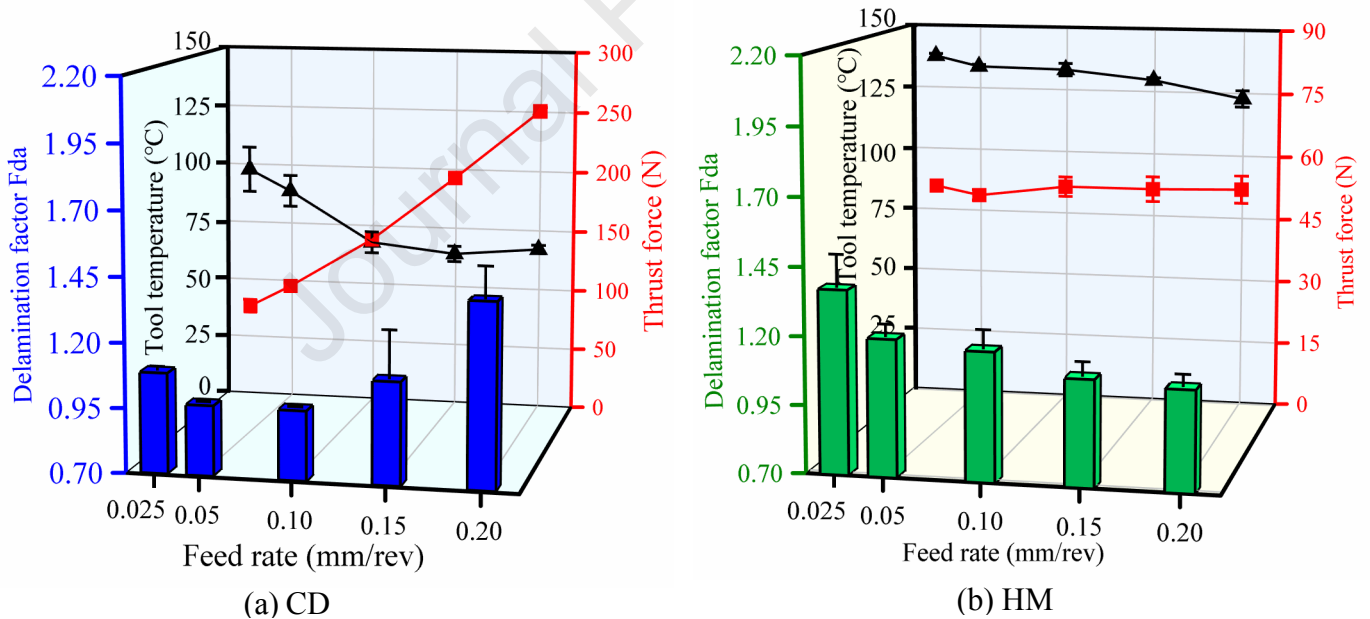
16 The variation of delamination factor against the feed rate is depicted in Fig. 7. The corresponding tool  
 17 temperature and the thrust force was also included in the plots. For CD, the initial declining  $F_{da}$  ( $F <$   
 18  $0.1$  mm/rev) correlates well with the declining tool temperature. Under a lower feed rate ( $F < 0.1$   
 19 mm/rev), the machining temperature plays a more predominant role in the delamination damage, as  
 20 the much higher machining temperature under such condition can lead to significant loss of stiffness  
 21 of the CF/PEKK ply which makes it more prone to delaminate. At higher feed rate, the machining  
 22 temperature decreased significantly and the drastically increased thrust force would take over its  
 23 influence on the delamination damage. This is reflected by the increased  $F_{da}$  with increasing thrust  
 24 force for  $F > 0.1$  mm/rev.

1

2

3 Table 2 Binary images of hole exit delamination under different hole making conditions

Feed rate (mm/rev)	0.025	0.05	0.1	0.15	0.2
CD					
HM					



4

5 Fig. 7 Delamination factor  $F_{da}$  against feed rate (front bar chart) and variation of tool temperature  
 6 and thrust force against feed rate (back line chart) for (a) CD and (b) HM

7 For HM, the thrust force is independent of feed rate and remained at a constant low level (50 N). At  
 8 low feed rate (0.025 mm/rev), the tool temperature approaches the  $T_g$  of PEKK. The gradual declining  
 9 trend of tool temperature also correlates well with the decreasing  $F_{da}$ , implying the influence of tool  
 10 temperature on the HM delamination

1 The finding of this study is in contrast to the reports [25,46,51] on thermosetting CFRP, where thrust  
2 force is considered the sole factor determining the delamination damage. In thermosetting CFRP, hole-  
3 exit delamination is mainly caused by mode I opening fracture as the cutting tool approaches the hole  
4 exit and the resulting delamination factor increases with thrust force [18,25,26]. For CF/PEKK  
5 however, the elevated temperature caused by the machining process also plays a significant role in the  
6 resulting delamination. Given the thermoplastic nature of the PEKK matrix, high machining  
7 temperature (approaching  $T_g$ ) can result in softened matrix [52] and weakened matrix  
8 support/encapsulation around the fibre bundles, which contribute to the more severe delamination  
9 damage.

#### 10 3.4. Hole wall microstructure

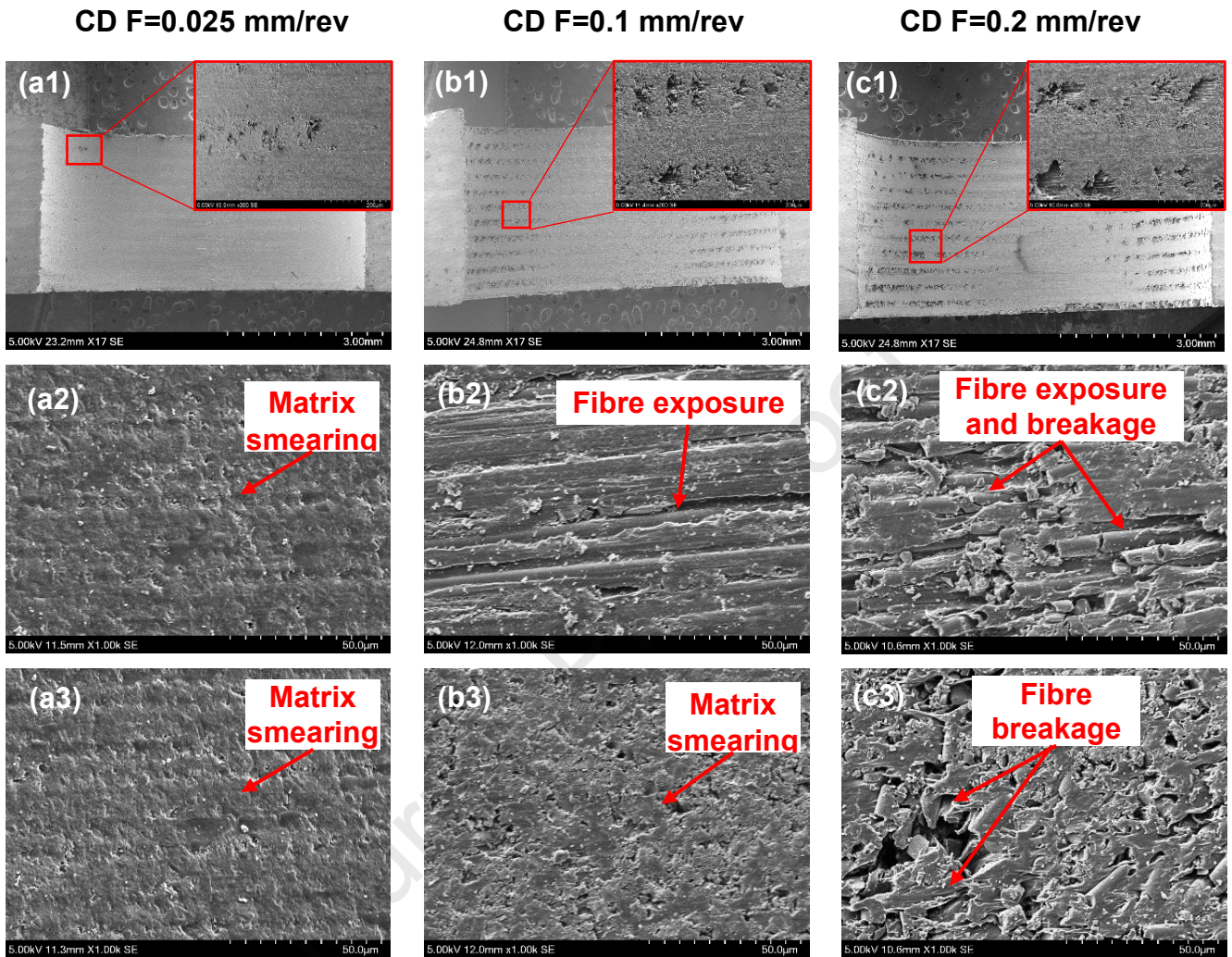
11 Microstructural damage such as fibre breakage, matrix loss and debonding can occur during hole  
12 making of CFRP. The typical hole wall surface SEM images produced under different hole making  
13 conditions is shown in Fig. 8 and Fig. 9.

14 For CD, low feed rate ( $F = 0.025$  mm/rev) resulted in relatively smooth hole wall surface finish, with  
15 negligible matrix loss, see Fig. 8 (a1). Increasing feed rate has led to more severe matrix loss and  
16 surface cavity as shown in Fig. 8 (b1) and (c1). The matrix loss and surface cavity typically take place  
17 at the obtuse fibre orientation where the material was removed by bending fracture [53]. The cracks  
18 and tears caused by the bending stress can be easily transmitted to the hole wall subsurface, resulting  
19 in distorted pits when the chip was separated from the workpiece [53]. As can be seen in Fig. 8 (a2-c2)  
20 (a3-c3), for both  $0^\circ$  and  $90^\circ$  fibre orientation, the extent of hole wall damage increases with increasing  
21 feed rate. The hole wall produced under low feed rate ( $F=0.025$  mm/rev) mainly features matrix  
22 smearing, where the fibre is well covered by the thermoplastic PEKK matrix. When the feed rate is  
23 increased from 0.025 mm/rev to 0.2 mm/rev, more severe fibre debonding and breakage are evident  
24 on the hole surface.

25 The hole wall microstructure under HM condition is significantly different from that of CD, see Fig.  
26 9. No significant surface matrix loss / cavity has been observed. This may be because the much lower  
27 cutting force of HM minimized the fracture and tears produced by the bending stress. The feed rate did  
28 not have significant impact on the resulting hole microstructure. For all the feed rates under  
29 investigation,  $0^\circ$  ply features long lip-like matrix smearing, whereas  $90^\circ$  ply shows blotchy matrix  
30 smearing. This is in contrast to the more even matrix smearing seen on the CD hole wall. The difference  
31 can be attributed to the disparate cutting behaviour of HM and CD: the cutting edges of drill bit is in  
32 constant contact with the hole wall surface and this can facilitate the evenly smeared PEKK matrix  
33 against the hole wall. For HM, the peripheral cutting edges of milling cutter is in intermittent contact  
34 with hole wall, which causes more localized matrix smearing. The much higher machining temperature  
35 (approaching PEKK  $T_g$ ) under HM may also lead to reduced matrix viscosity, and hence different

1 matrix smearing behaviour.

2



3

4 Fig. 8 (a1-c1) Low and high magnification SEM images showing hole wall microstructures produced

5 by CD under different feed rates; (a2-c2) high magnification SEM images showing damage on 0°

6 ply; (a3-c3) high magnification SEM images showing damage on 90° ply.



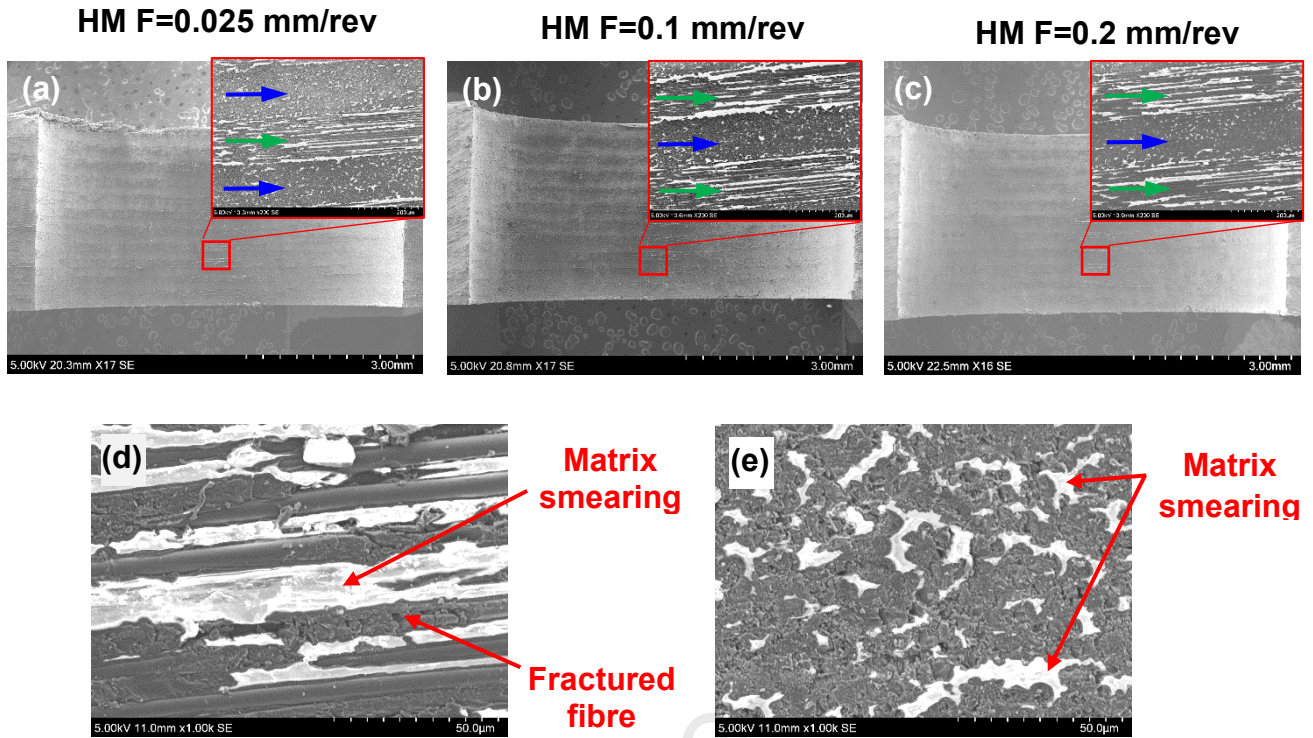


Fig. 9 (a-c) Low and high magnification SEM images showing hole wall microstructure produced by HM under different feed rates (Green arrow: 0° ply, blue arrow: 90° ply); Representative SEM images showing HM hole wall microstructure of (d) 0° ply and (e) 90° ply.

### 3.5. Chip analysis

#### 3.5.1 Chip morphology






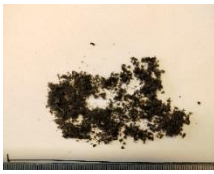


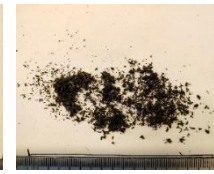
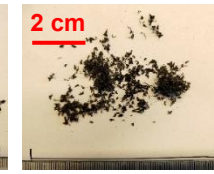
Table 3 shows the optical images of the chips collected from different hole making conditions. In general, chips produced by CD are continuous and ribbon-like, similar to chips produced from drilling of CF/ABS [27] and CF/PEEK [30]. In contrast, chips produced from HM are finer and powder-like. This distinct difference in chip morphology can be attributed to the different material removal mechanisms: CD is a continuous cutting process where the main cutting edge of the drill bit is in continuous contact with the workpiece and the chip flows continuously along the flute after leaving the cutting edge [54]. HM on the other hand, is an intermittent cutting process where the material removal is achieved by the periodic contact between peripheral cutting edge and the workpiece [55].

For CD, extremely long continuous chips were produced at low feed rates ( $F = 0.025$  mm/rev and  $F = 0.05$  mm/rev), which has led to severe tool clogging, (also see Fig. 5). The length of the chips generally decreased with increasing feed rate, as the chips are prone to fracture under greater thrust force.

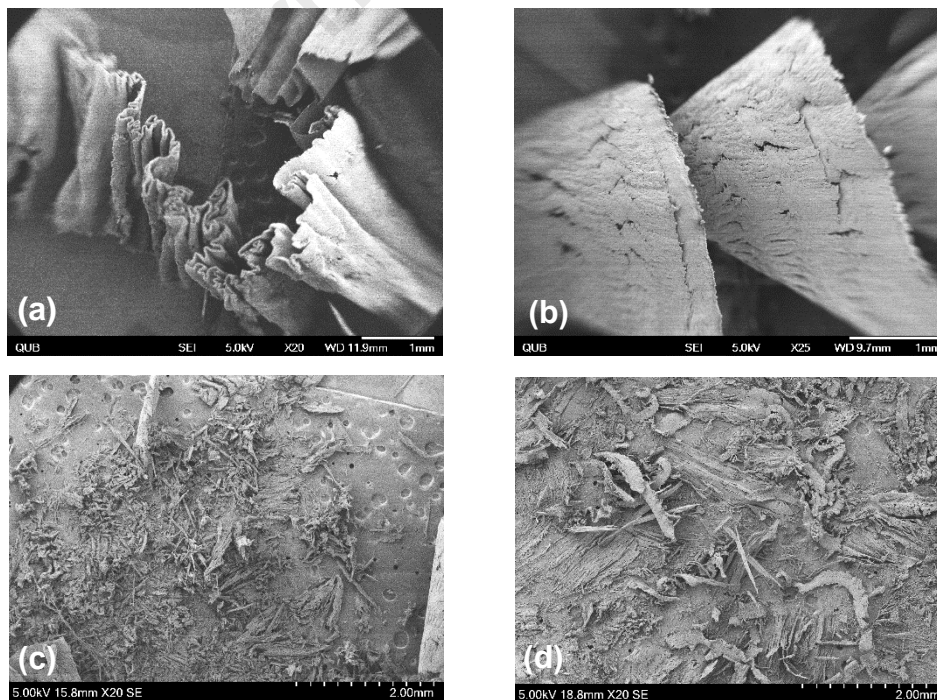
The microstructure of the chips was further analysed by SEM and the results are presented in Fig. 10. It can be seen that for CD under low feed rate ( $F = 0.025$  mm/rev), the chips show a folded morphology. This is because the extremely long chips tend to congest in the flute of the drill bit, impeding their evacuation. In addition, the chips produced under low feed rate are thinner (See Appendix C Fig. C.1)

1 and easily deformed (compressed) when being pushed out of the flute. The spiral CD chip morphology  
 2 produced under high feed rate ( $F = 0.2$  mm/rev) resembles metal chips produced by CD [56,57]. Such  
 3 morphology is in contrast to the powdery chips produced from brittle CF/epoxy composite under the  
 4 similar CD process [30,58,59]. This is mainly due to the excellent ductility of the PEKK matrix (~  
 5 50.0% failure strain) [60], where the thermoset epoxy matrix has a much lower ductility (1.5 - 8.0%  
 6 failure strain) [60]. For HM, finer and more fragmented chips were evident, see Fig. 10 (c) and (d).  
 7 Higher feed rates tend to produce slightly thicker chips, as the volume of material removed per tooth  
 8 increases with the feed rate [13].

9 Table 3 Morphology of chips produced in hole making of thermoplastic CF/PEKK composite

Feed rate (mm/rev)	0.025	0.05	0.1	0.15	0.2
CD					
HM					

10



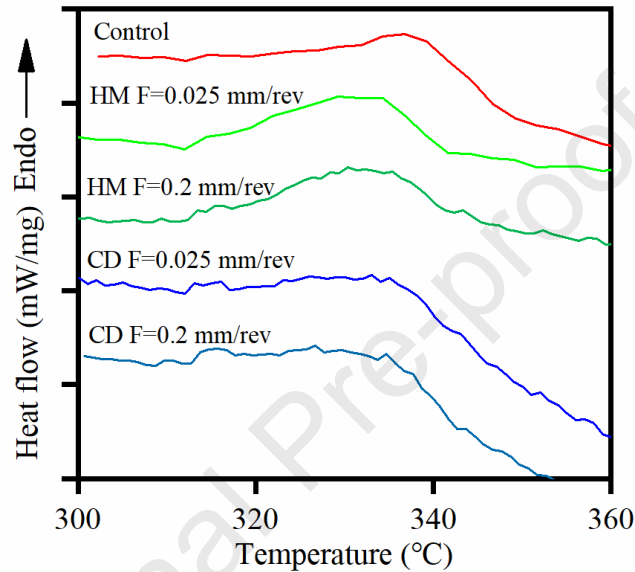
11

12 Fig. 10 SEM images of chips produced by (a) CD at  $F=0.025$  mm/rev, (b) CD at  $F=0.2$  mm/rev (c)

13 HM at  $F=0.025$  mm/rev (d) HM at  $F=0.2$  mm/rev

### 1 3.5.2 Chip thermal properties

2 To evaluate the potential impact of the hole making process on the machined composite molecular  
 3 structure, thermal analysis was carried out for the machined chips using DSC technique, see Fig. 11.  
 4 The corresponding material thermal parameters obtained from DSC analysis were summarized in Table  
 5 4. From Table 4, it can be seen that the crystallinity data  $\chi_c$  follows the trend  $CD > HM > \text{control}$ ,  
 6 with CD under  $F = 0.2\text{mm/rev}$  giving the highest  $\chi_c$  (28.35%), representing  $> 300\%$  increase against  
 7 the control (unmachined CF/PEKK).



8

9 Fig. 11 DSC results of raw material and chips produced under different hole making conditions

10

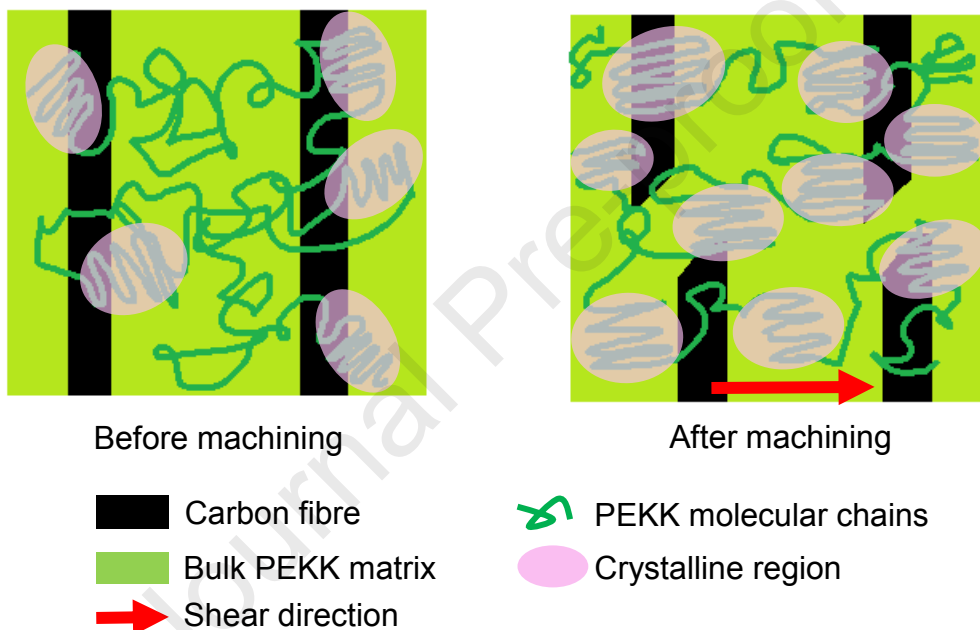
11 Table 4 Crystallinity values of unmachined CF/PEKK and chips produced under different hole making  
 12 conditions

Hole making method	Feed rate (mm/rev)	$\Delta H_m$ (J/g)	$\Delta H_{cc}$ (J/g)	$\chi_c$ (%)	% increase
Control	-	3.183	-	7.20	-
CD	0.025	9.739	-	22.03	205.96
CD	0.1	12.53	-	23.76	293.65
CD	0.2	12.82	-	28.35	302.76
HM	0.025	6.496	-	14.70	104.08
HM	0.1	6.759	-	15.29	112.34
HM	0.2	6.624	-	14.98	108.10

13

14 The increase in crystallinity seen in the machined chips can be attributed to the shear induced

1 crystallization in the thermoplastic PEKK matrix [61]. Shear induced crystallization has been  
 2 previously reported for thermoplastic polymers such as uniaxially stretched PEEK [61] and powder  
 3 reinforced thermoplastic nanocomposites (such as boron nitride reinforced polyethylene [62] and  
 4 graphene reinforced polypropylene [63]) subjected to tension. However, this is the first time such  
 5 phenomenon was discovered for machined CFRTP. During machining, the tool cutting edge exerts a  
 6 significant in-plane shear stress on both the hole wall and the adjacent chip. PEKK is a semi-crystalline  
 7 polymer consisting of both crystalline and amorphous regions. Under the shear action, randomly  
 8 arranged molecular chains within the PEKK amorphous region will be stretched and re-aligned along  
 9 the shear direction, forming more orderly arranged crystalline region, as illustrated by  
 10 Fig. 12.



11  
 12 Fig. 12 Schematic diagram of strain induced crystallization in machined CF/PEKK chips

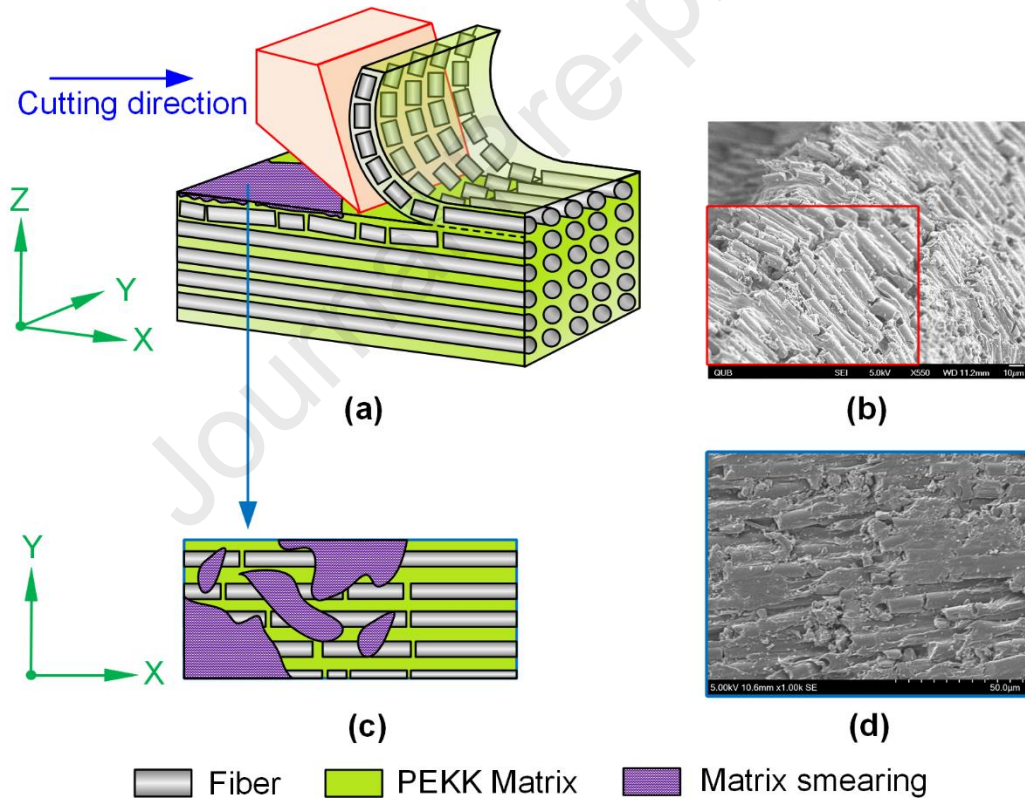
13 Compared to HM, CD exhibited greater crystallinity increase. This can be attributed to the greater  
 14 cutting force and temperature experienced by the chip under CD. As the temperature approaches  $T_g$ ,  
 15 the PEKK matrix will experience the transition from glass to rubber-like state, the better flow and  
 16 sliding of the molecular chains can further facilitate the re-alignment of the polymer molecules and  
 17 increase crystallinity.

### 18 3.6. Material removal mechanism

19 For easier microstructural inspection and interpretation of the underlying material removal mechanism,  
 20 the analysis was focused on CD chips and hole wall with  $0^\circ$  and  $90^\circ$  fibre orientation. As depicted in  
 21 Fig. 13 and Fig. 14, for both fibre orientations, the material removal involves combined brittle fibre  
 22 fracture and plastic deformation of the PEKK matrix. The cutting process can be explained by the

1 Ernst and Merchant's shear cutting model [64], where the shear plane assumption is applied to interpret  
 2 the formation of continuous chips formed by the CF/PEKK composite.

3 As shown in Fig. 13 (a), for  $0^\circ$  fibre orientation, the cutting edge exerts a compressive stress to the  
 4 workpiece along the fibre direction and an in-plane shear stress is generated along the fibre-matrix  
 5 interface [53]. With the advancement of the tool, the removed carbon fibre will be subjected to  
 6 extensive bending against the tool rake face. Brittle fibre fracture occurs when the external stress  
 7 exceeds the bending strength limit of carbon fibre. Despite the segmented fibres within, the chip  
 8 remains an overall intact morphology, as the ductile PEKK matrix can sustain a significant amount of  
 9 plastic deformation, holding the fractured fibres in place, see Fig. 13 (b). On the machined hole surface,  
 10 the high machining temperature can soften the matrix, and the highly viscous matrix can be re-casted  
 11 and smeared at certain regions of hole surface (see Fig. 13 (c, d)). The evident fibre breakage on the  
 12 machined hole surface is a result of the compressive action exerted by the tool cutting edge and rake  
 13 face in the workpiece thickness direction.

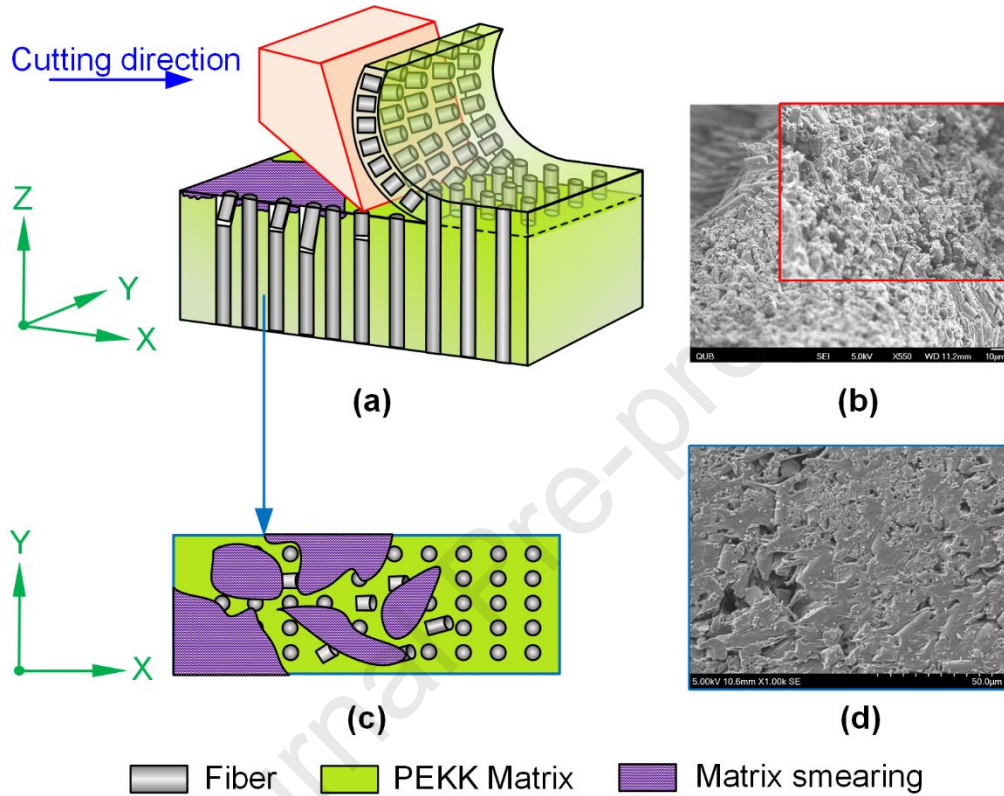


14

15 Fig. 13 (a) Schematic diagram of chip formation of CF/PEKK ( $0^\circ$  fibre orientation), (b) Close-up  
 16 SEM image of chip surface ( $0^\circ$  fibre orientation) showing fractured fibres, (c) Top view schematic of  
 17 the machined hole wall and (d) Close-up SEM image showing matrix smearing and fractured fibres  
 18 on machined hole wall.

19 For  $90^\circ$  fibre orientation as illustrated by Fig. 14 (a), the cutting speed is perpendicular to the fibre  
 20 direction. The carbon fibres undergo brittle fracture as a result of the shearing action of the cutting

1 edge and the fracture plane is perpendicular to the fibre direction. Again, despite its plastic deformation,  
 2 the highly ductile PEKK matrix has held the broken fibres in place, Fig. 14 (b). Similar matrix smearing  
 3 was found on the hole surface. Bending-induced fibre debonding and fibre breakage have been found  
 4 on the machined hole surface. Some of the broken fibres were found to be embedded in the smeared  
 5 matrix on the machined hole surface, Fig. 14 (c, d).



6  
 7 Fig. 14 (a) Schematic diagram of chip formation of CF/PEKK (90° fibre orientation), (b) Close-up  
 8 SEM image of chip surface (90° fibre orientation) showing fractured fibres, (c) Top view schematic  
 9 of the machined hole wall and (d) Close-up SEM image showing matrix smearing and fractured  
 10 fibres on machined hole wall.

11 It is expected the unique material removal mechanism and the associated material structural change  
 12 discovered in this study can be generalized to a wide of range of CFRTP materials, such as CF/PEEK,  
 13 CF/PPS, CF/ABS, etc. Our preliminary work also inspires the research into orthogonal cutting of  
 14 CFRTPs, through which more comprehensive understanding on the effect of cutting depth, cutting  
 15 speed and temperature on the material removal mechanism can be elucidated. Although in-depth  
 16 microstructural and thermal analysis has been carried out for chips in this study, measuring thermal  
 17 property of the machined hole wall surface layer is not feasible with the current instrumentation. It is  
 18 expected that the hole wall surface, particularly the smeared matrix, would have experienced similar  
 19 shear induced crystallization. According to the literature, strain induced crystallization can potentially  
 20 alter the stiffness, strength and fracture toughness of the material [62,65]. In the future, it will be

1 valuable to investigate how machining induced structural/property variation would impact the  
2 mechanical performance of CFRTP components.

3

#### 4 **4. Conclusions**

5 This paper reports the first investigation on hole making performance of thermoplastic CF/PEKK  
6 composite. Different hole making methods (CD and HM) and the effect of different feed rates on the  
7 resulting thrust force, machining temperature, delamination damage, hole microstructure and chip  
8 morphology/crystallinity have been investigated in detail. The main conclusions and contributions are  
9 as follows:

- 10 • CD generates higher thrust force than HM. The CD thrust force and feed rate follow a quasi-linear  
11 increasing relationship, whereas the thrust force of HM is independent on its feed rate.
- 12 • The tool temperature for both CD and HM decreases with the feed rate. HM shows a much higher  
13 tool temperature than CD, and this can be attributed to its prolonged tool engagement time and the  
14 associated heat accumulation.
- 15 • The CF/PEKK delamination damage is a result of combined thermal-mechanical interaction. For  
16 CD under low feed rate, the high machining temperature plays a more predominant role whereas  
17 under higher feed rate (lower temperature), the high thrust force would be more dominating. For  
18 HM, the delamination damage formation is mainly influenced by the high machining temperature,  
19 as the thrust force remains constant within the range of feed rate being investigated.
- 20 • Matrix smearing has been observed on both CD and HM hole walls. This is due to the softening  
21 and recasting of the highly ductile PEKK matrix onto the hole surface. Hole wall microstructural  
22 damages such as matrix loss and surface cavity are more severe for CD.
- 23 • CD leads to formation of continuous chips, which can be attributed to the continuous material  
24 removal process during drilling and the excellent ductility of the PEKK matrix. Under lower feed  
25 rate ( $< 0.05$  mm/rev), the long and folded chips tend to clog the tools. Under higher feed rate ( $>$   
26  $0.1$  mm/rev), shorter, spiral shaped chips can be effectively evacuated. In contrast, HM produced  
27 short fragmented powdery chips as a result of the intermittent material removal process.
- 28 • Chips produced by both CD and HM show increased crystallinity as a result of shear induced  
29 crystallization, with CD ( $0.2$  mm/rev) giving the greatest (300%) crystallinity enhancement.
- 30 • Through advanced material characterization, a greater insight has been developed into the  
31 machined CFRTP deformation characteristics, material removal mechanisms, and the associated  
32 material structural evolution at microscopic and molecular levels. These findings would inspire  
33 future researchers to better deploy the material process - structure - property relationship for  
34 optimized CFRTP manufacturing.

1 **CRedit authorship contribution statement**

2 **Jia Ge:** Conceptualization, Methodology, Investigation, Writing - Original Draft **Giuseppe**  
3 **Catalanotti:** Supervision, Writing - Review & Editing **Brian Falzon:** Writing - Review & Editing  
4 **John McClelland:** Software, Resources, Writing - Review & Editing **Colm Higgins:** Resources,  
5 Writing - Review & Editing **Yan Jin:** Supervision, Writing - Review & Editing, Funding acquisition,  
6 Project administration **Dan Sun:** Conceptualization, Methodology, Investigation, Writing - Review &  
7 Editing, Funding acquisition, Project administration.

8  
9 **Declaration of competing interest**

10 The authors declare that they have no known competing financial interests or personal relationships  
11 that could have appeared to influence the work reported in this paper.

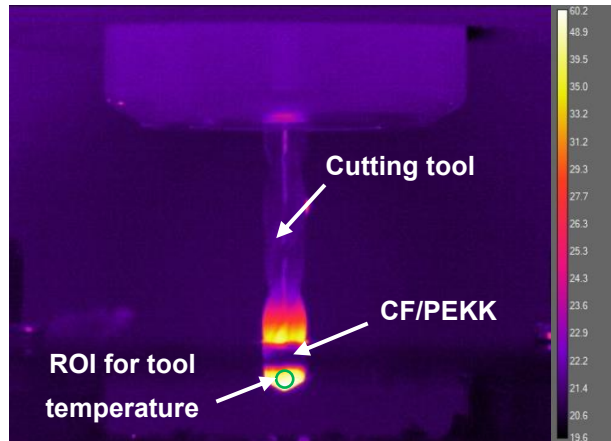
12  
13 **Acknowledgements**

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15 project has also received funding from the European Union's Horizon 2020 research and innovation  
16 programme under grant agreement No 734272.

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## 1 Appendix A



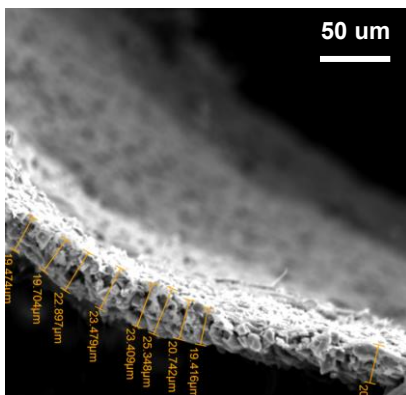
2  
3 Fig. A.1 Tool temperature measurement at the hole exit (CD with feed rate  $F=0.2\text{mm/rev}$ )

## 4 Appendix B

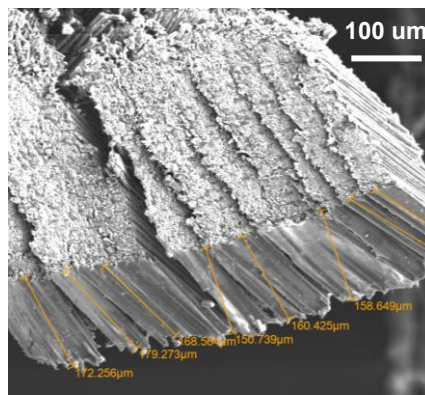
5 Table B.1 Tool engagement time of CD and HM under different feed rates

Feed rate (mm/rev)	Method	
	CD	HM
0.025	4.4 s	77.7 s
0.05	2.2 s	37.8 s
0.1	1.1 s	18.7 s
0.15	0.7 s	12.4 s
0.2	0.5 s	9.3 s

## 7 Appendix C



8 (a) CD  $F=0.025\text{ mm/rev}$



9 (b) CD  $F=0.2\text{ mm/rev}$

10 Fig. C. 1 Thickness measurement of chips produced by CD under different feed rates

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**Highlights:**

- Hole making performance of CF/PEKK is investigated for the first time.
- A comparative study on conventional drilling and helical milling is reported.
- The role of thermo-mechanical interaction is elucidated for CF/PEKK hole making.
- Chip morphology and the associated material removal mechanism are revealed.
- Shear-induced crystallization caused by hole making is reported for the first time.

Journal Pre-proof

### **Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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