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Coupling rheometry and computed tomography to study the evolution of voids during the consolidation of CF/PEKK composites

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Abstract: One of the manners to optimize the consolidation quality of composites is to understand how the voids present within or between the tapes are reduced. This work proposes to study tomography scanned specimens (unidirectional and cross-ply), obtained from interrupted squeeze flow experiments on rheometer. Results show that inter-ply porosities are rapidly reduced and that the ply-ply interface is more rapidly indistinguishable for UD laminates than cross-ply ones suggesting a good {fiber + matrix} homogenization at the interface. It is also shown that intra-ply voids, surrounded by densely packed fibers are hardly reduced. These porosities limit the consolidation quality, suggesting that an effort in better separating the fibers during the pre-impregnation step is essential for the quality of composites.

Keywords : CF/PEKK composites, Synchrotron radiation, micro-tomography, porosity, consolidation

1. Introduction

Carbon fiber reinforced thermoplastic composites have gained interest over thermoset ones because of their recyclability and weldability, as well as quicker manufacturing processes [1,2]. The Polyaryletherketone (PAEK) family is commonly chosen as the matrix of such composites because of its high service temperature, high oxidation and chemical resistance and good impact properties [3,4]. Specifically, polyetherketoneketone (PEKK) has been developed for its versatility in terms of service and processing temperatures, which can be tuned by varying the terephthaloyl (T) and isophthaloyl (I) isomers ratio, [5,6] as shown in Figure 1.

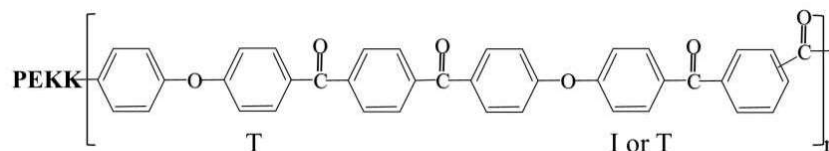


Figure 1: Chemical structure of PEKK

Nevertheless, optimizing the manufacturing of CF/PEKK composites, involving a pre-impregnation step, a placement step and a consolidation one, remains an open area of research. To do so, the rheology of the system responsible for the reduction of voids present within the ply (intra-laminar voids) and between the plies (inter-laminar voids) needs to be better understood. It is then proposed in this study to conduct interrupted squeeze flow experiments on UD (unidirectional) and cross-ply [0/90] CF/PEKK

tapes. The obtained samples are then characterized by high-resolution X-ray micro-computed tomography in order to visualize the microstructural evolution of the composite during consolidation, especially the size, location and shapes of the voids.

2. Materials and methods

2.1. Materials

Continuous (UD) tapes (plies) consist in PEKK with a T/I ratio equal to 70/30 (Arkema) impregnating Hexcel High Strength carbon fibers (Tensile strength ~5 GPa and young modulus ~240 GPa), with a fiber volume content about 60 v% and a thickness close to 200 μm (see a typical optical image of the tape in Figure 2).

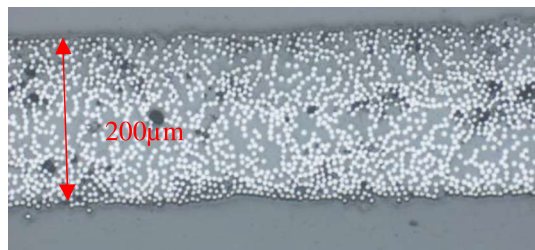


Figure 2: CF/PEKK tapes typical microstructure as observed with an optical microscope (x20). White circles represent the fibers, the grey area is the matrix and the black spots are the initial voids.

2.2. Equipment for squeeze flow experiments

Squeeze flow experiments are conducted on an Anton Paar MCR 502 rheometer with a plan-plan geometry (25 mm) in a constant surface configuration. This means that the samples initially fill all the surface of the plates such that, during all the experiment, the contact surface between the plates and the sample remains constant. The samples are cut into 25 mm diameter disks and then hand-laid up to stack manually UD 8-ply $[0]_8$ and cross-ply $[0/90]_4$ laminates. The laminate is carefully placed on the rheometer at T_1 (above T_g). Then heating occurs until T_2 ($T_2 > T_m$) at a constant force $F=1$ N to maintain contact between the plates and the sample. After a few minutes at T_2 , a controlled ramp of force is applied till a value F_0 is reached (leading to a pressure around 1 bar, such as in VBO process). The force is then kept constant for the duration of the test. The whole procedure is schematized in Figure 3. Interrupted test are conducted where the sample is removed from the rheometer at certain times shown by the blue dots in Figure 3.

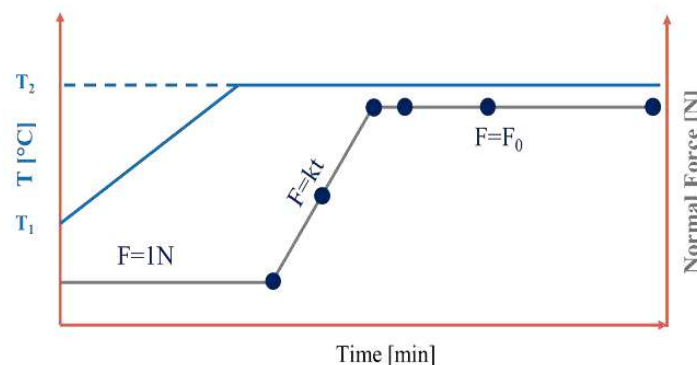


Figure 3: Temperature and normal force profiles of the squeeze flow experiments

2.3. Micro-computed tomography

The samples are cut into bar shape (with a cross section around 1.5 mm x 1.5 mm) using a diamond wire saw and fixed on a sample carrier for tomography measurements. The tomography measurements were made at the Anatomix beamline of Synchrotron SOLEIL [7]. A polychromatic (“white”) X-ray beam with a central energy around 10 keV was used. The detector had 2048x2048 pixels; the effective pixel size was 0.65 μm . For each scan, 2000 projections were taken over an angular range of 180 degrees. The reconstructed volumes have a diameter and height of 1.3 mm. After tomographic reconstruction using the standard pipeline at the beamline and the PyHST2 reconstruction software [8], all volumes were post-treated with the ImageJ software [9] to quantitatively characterize the voids size, location and shape.

3. Results and discussion

Through the squeeze flow experiments on the rheometer, one can plot the variation of the laminate thickness as a function of time (Figure 4). The gap rapidly decreases when the force increases and stabilizes after about 15 min.

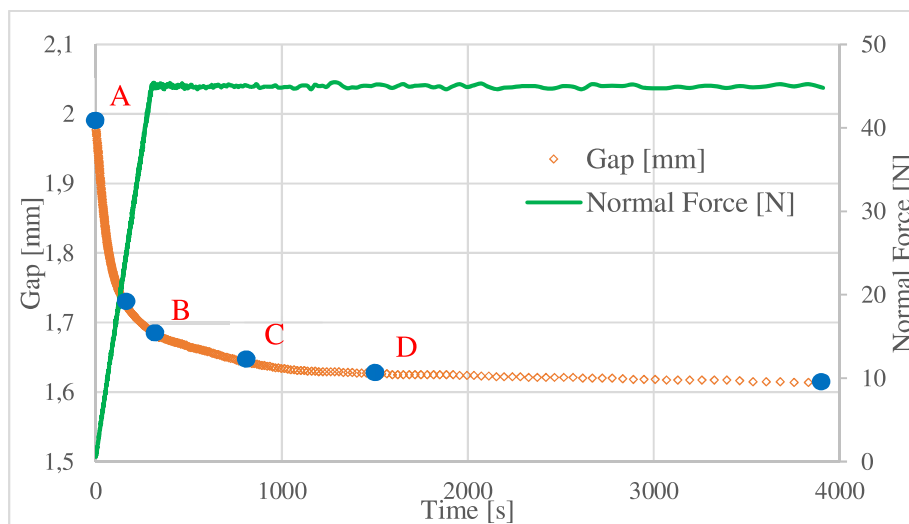


Figure 4: Evolution of the $[0/90]_4$ CF/PEKK laminate during the squeeze flow experiment. The blue dots represent the points where the test is interrupted. Letters A, B, C and D are for the four images shown in Figures 5 and 6

The observed porosities are strongly anisotropic, oriented along the fibers. They decrease in terms of size and quantity when the time of squeeze increases (Figure 5). The large thickness variation observed at the beginning of the experiment is mainly due to the tapes' initial surface roughness, which flatten under compression. Very rapidly, this rough interface between plies disappear and the laminate starts to form a homogeneous composite.

However, the voids present within the tapes remain, even for longer squeeze times (see Figure 5. D). Such porosities appear surrounded by very dense areas of fibers. During the impregnation step, some fibers have not been sufficiently separated leading to a poor matrix impregnation in these areas and the presence of trapped voids. The pressure applied (close to 1 bar) combined with high matrix viscosity cannot enable the filling of these voids in a reasonable squeeze time. Hence, it is suggested here, that, for VBO consolidation, an effort should be done on the homogenization of the {fiber + matrix} repartition.

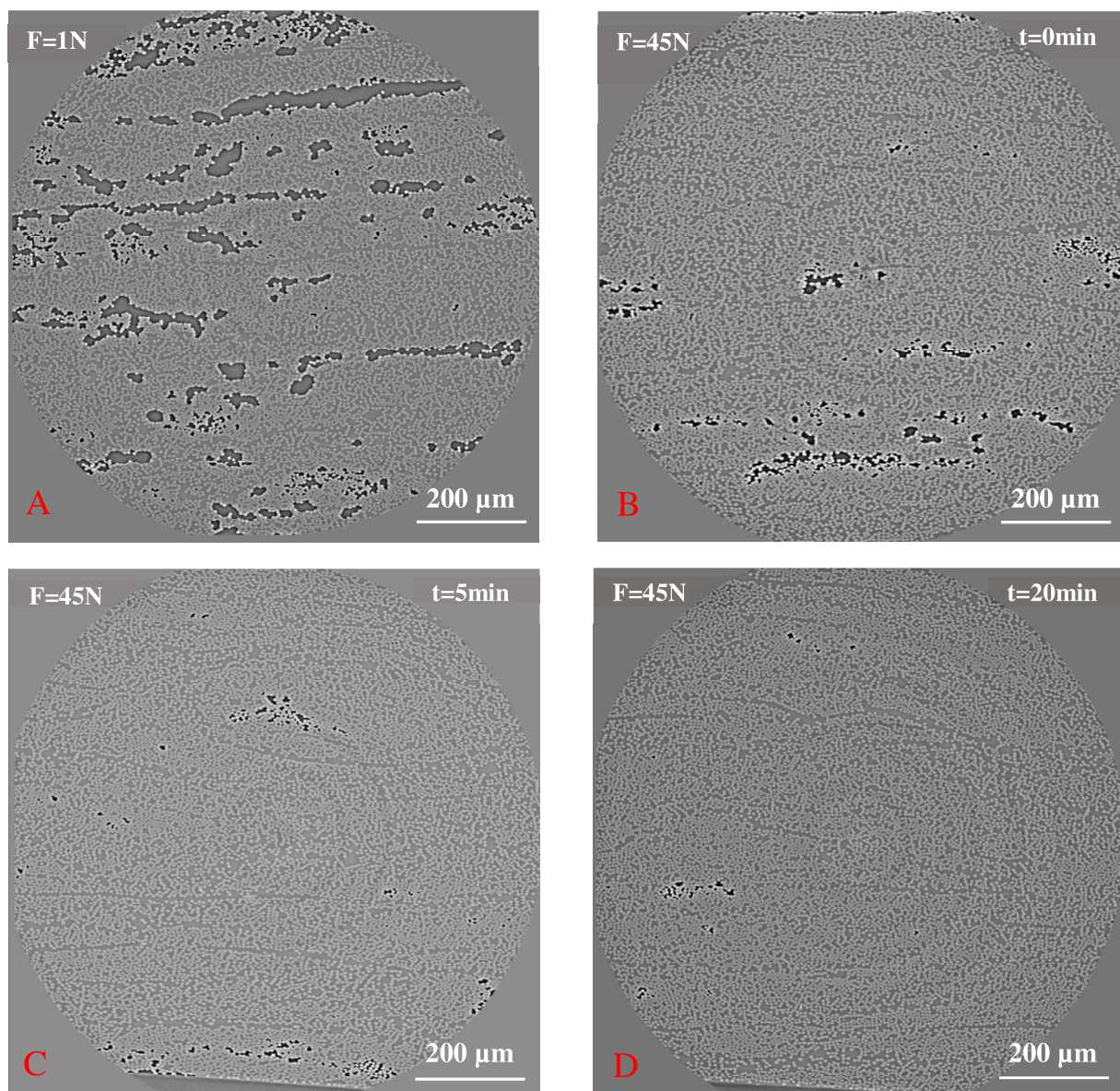


Figure 5: Tomography images, orthogonal to the fibers direction, of the squeezed UD [0]₈ laminates for four different moments: A, B, C and D shown in Figure 4.

Quite similar results can be extracted from the tomography measurements on cross-ply [0/90] laminates (Figure 6). As for UD laminates, the voids are progressively reduced; inter-laminar voids seem to disappear faster than intra-laminar ones; porosities trapped are not able filled by the matrix during the time of the experiment.

Nevertheless, contrary to UD laminates, the individual plies can be distinguished late in the process, as shown by the green arrows. This is explained by the fact that cross-ply stacking leads rapidly to fibers blocking, while fibers in the same orientation favor the interpenetration of one ply towards the other.

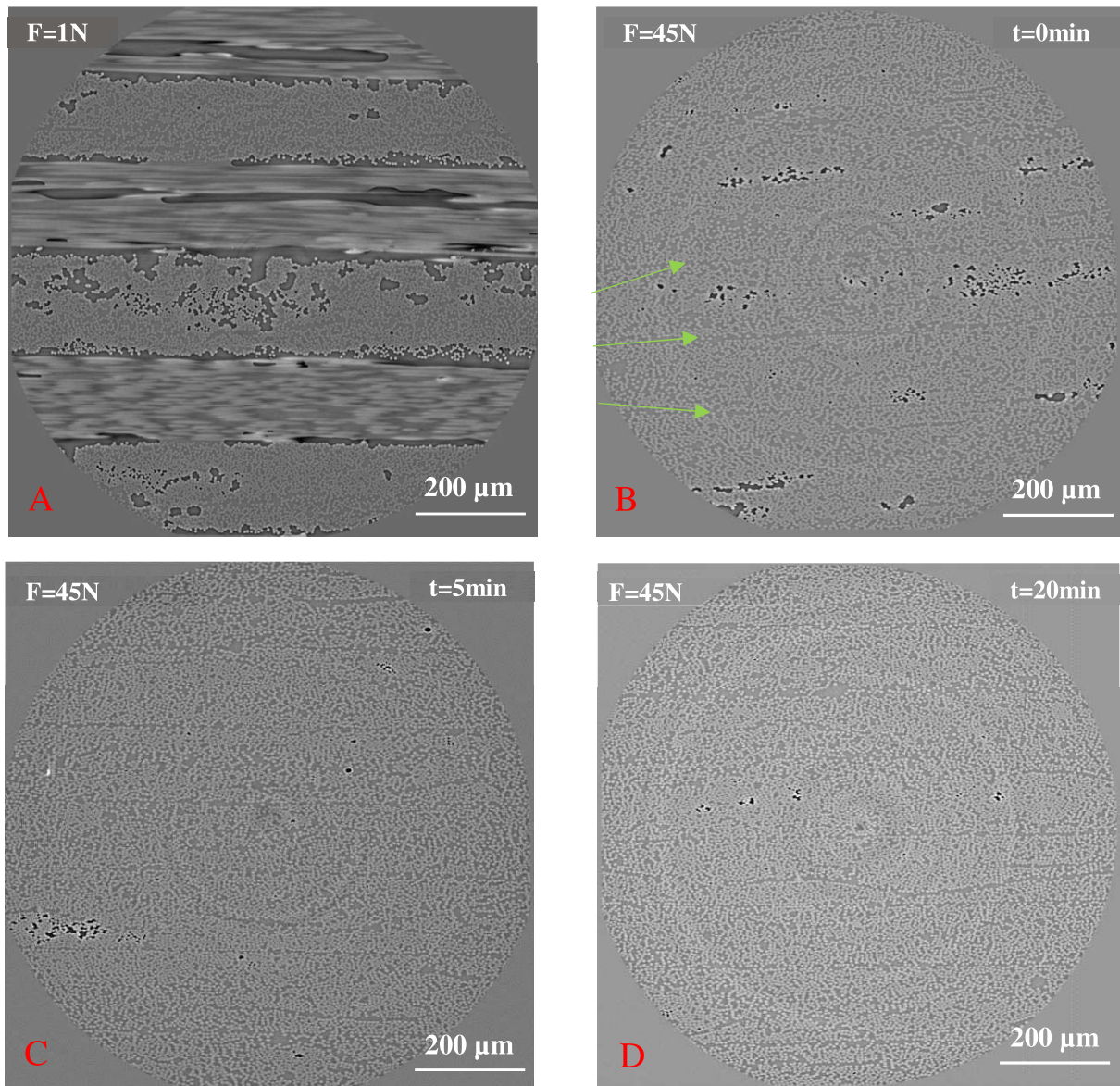


Figure 6: Tomography images of the squeezed cross-ply $[0/90]_4$ laminate for four different moments: A, B, C and D shown in Figure 4. In A, the slice is either parallel to the fibers (plies with lines of fibers) or transverse to the fibers (plies with circular fibers). The view of the three other images (B, C, and D) is either at 45° or -45° towards the fibers, hence the elliptical shape of the fibers in the images.

At the beginning of consolidation, the matrix flows first and fills the inter-laminar voids leading to resin-rich regions at the interface but, with time, local micro movement of fibers lead to a homogenization at the interface in terms of {fibers + matrix} repartition (see Figure 6.C and D).

4. Conclusion

This work studies the evolution of voids during consolidation of CF/PEKK by visualizing tomography scans of UD and $[0/90]$ laminates through interrupted squeeze flow tests. It is observed that inter-ply porosities are rapidly reduced whereas those within the plies, surrounded by high concentrated fibers areas, are much more hardly filled. A progressive homogenization in terms of {fibers + matrix} repartition at the ply-ply interface is also highlighted for the cross-ply $[0/90]$ laminates.

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6. References

- [1] Clyne TW, Hull D. An Introduction to Composite Materials. Second Edi. In: Cambridge Solid State Science Series; 2019.
- [2] Vaidya UK, Chawla KK. Processing of fibre reinforced thermoplastic composites. *Int Mater Rev* 2008;53:185–218. <https://doi.org/10.1179/174328008X325223>.
- [3] Choupin T, Fayolle B, Régnier G, Paris C, Cinquin J, Brulé B. Isothermal crystallization kinetic modeling of poly(etherketoneketone) (PEKK) copolymer. *Polymer (Guildf)* 2017;111:73–82. <https://doi.org/10.1016/j.polymer.2017.01.033>.
- [4] Choupin T, Fayolle B, Régnier G, Paris C, Cinquin J. A more reliable DSC-based methodology to study crystallization kinetics : Application to poly (ether ketone ketone) (PEKK) copolymers. *Polymer (Guildf)* 2019;155:109–15. <https://doi.org/10.1016/j.polymer.2018.08.060>.
- [5] Choupin T, Debertrand L, Fayolle B, Régnier G, Paris C, Cinquin J, et al. Influence of thermal history on the mechanical properties of poly(ether ketone ketone) copolymers. *Polym Cryst* 2019;2:1–8. <https://doi.org/10.1002/pcr2.10086>.
- [6] Veazey D, Hsu T, Gomez ED. Next generation high-performance carbon fiber thermoplastic composites based on polyaryletherketones. *J Appl Polym Sci* 2017;134:19–21. <https://doi.org/10.1002/app.44441>.
- [7] Weitkamp T, Scheel M, Giorgetta J, Joyet V, Le Roux V, Cauchon G, et al. The tomography beamline ANATOMIX at Synchrotron SOLEIL. *J Phys Conf Ser* 2017;849:012037. <https://doi.org/10.1088/1742-6596/849/1/012037>.
- [8] Mirone A, Brun E, Gouillart E, Tafforeau P, Kieffer J. The PyHST2 hybrid distributed code for high speed tomographic reconstruction with iterative reconstruction and a priori knowledge capabilities. *Nucl Instruments Methods Phys Res Sect B Beam Interact with Mater Atoms* 2014;324:41–8. <https://doi.org/10.1016/j.nimb.2013.09.030>.
- [9] Schneider CA, Rasband WS, Eliceiri KW. NIH Image to ImageJ: 25 years of image analysis. *Nat Methods* 2012;9:671–5. <https://doi.org/10.1038/nmeth.2089>.