Effect of in situ treatment on the quality of flat thermoplastic composite plates made by automated fiber placement (AFP)

F. Shadmehri\textsuperscript{a,b}, S. V. Hoa\textsuperscript{a,b}, J. Fortin-Simpson\textsuperscript{a,b} and H. Ghayoor\textsuperscript{a,b}

\textsuperscript{a}Mechanical, Industrial & Aerospace Engineering, Concordia University, Montreal, Canada; \textsuperscript{b}Center for Research in Polymers and Composites (CREPEC), Montreal, Canada

ABSTRACT
Composite structures used as aerosurfaces in aerodynamic applications are required to have a certain surface finish quality. In manufacturing of thermoplastic composites using automated fiber placement (AFP) for aerodynamic applications, it is not only desirable to achieve good consolidation by using AFP alone and avoiding secondary treatment in an autoclave, but also to achieve acceptable surface smoothness required for aerosurfaces. In this study, an \textit{in situ} treatment called “repass” was implemented to achieve surface finish quality required for aerodynamic applications. Moreover, the effect of this \textit{in situ} treatment on the quality of the thermoplastic laminates, including void content and crystallinity was investigated. Autoclave-treated samples were used as references for comparing surface quality and other quality indicators.

1. Introduction
Technical advances in the automated manufacturing of thermoplastic composites have increasingly attracted the interests of the aerospace industry due to flexibility of the process, allowing fabrication of complex composite parts. Specifically, automated fiber placement (AFP) has provided a new look in the manufacturing of large-scale thermoset composite aero-structures in comparison with the traditional manufacturing processes. However, AFP manufacturing of large-scale thermoplastic composites is lagging behind due to technical challenges and difficulties of processing at high temperature.

In manufacturing of thermoplastic composite structure, it is highly desirable to reach good quality by using AFP process alone and to avoid additional autoclave process to reduce manufacturing cost. However, due to various parameters involved in the AFP process, achieving a desirable quality of the AFP-made part is challenging. Previous studies point out that the quality of fiber placed thermoplastic composites strongly depends on the manufacturing parameters such as temperature, compaction force (consolidation pressure) and layup speed.\textsuperscript{1–21} One of the important requirements for structures categorized as aerosurfaces is the surface roughness and surface waviness. The surface finish has a direct impact on the amount of drag and lift generated as airstream flows over the structure. Traditional \textit{in situ} manufacturing of thermoplastic composites using AFP, generally leads to a very rough surface if layup is performed on a male tool (e.g. mandrel). Thus, it is highly desirable to
achieve a good surface finish using an \textit{in situ} treatment technique in applications where aerodynamics requires a smooth surface finish.

In this study, an \textit{in situ} treatment called “repass” has been implemented to primarily improve the surface finish quality of the carbon fiber/PEEK composites for aerodynamic applications. However, since this \textit{in situ} treatment could potentially have an effect on other quality indicators of the parts, the effect of applying a repass treatment one or two times on the void content and crystallinity of the samples is studied. One set of AFP-made samples was treated in the autoclave to obtain reference values for all quality indicators, namely, surface roughness, void content, and crystallinity.

2. Manufacturing and treatment of samples

To quantify the effect of including repasses between layers, flat coupon samples were made using the AFP workcell and flat paddle tool at the Concordia Centre for Composites (CONCOM), shown in Figure 1 below. The AFP system employs a nitrogen hot gas torch to melt the incoming tape and a steel roller for compaction.

The samples were made using ¼-inch wide AS4/ APC-2 carbon fiber/PEEK tape from CYTEC Solvay Group. The unidirectional tape consists of a 68:32 weight percentage mixture of carbon fiber (AS4) and Polyetheretherketone (PEEK) matrix (APC-2). The nominal value of fiber volume fraction is 61%. The first step required to make the laminate was to wrap the paddle tool with a substrate layer that doubles as the first layer in the laminate. The substrate layer is processed with the following parameters: 910 °C hot gas temperature, 80 lbf compaction force, 2 in/s layup speed. Subsequently, a panel was made on top of the substrate layer, including the substrate as the initial ply. The total layup sequence was [0]_{24}. The panel was made with the same process parameters as the substrate layer, except that the gas temperature was lowered to 875 °C.

2.1. Repass treatment

The term “repass” refers to the application of heat and pressure via the AFP head to a substrate layer, without the addition of new material.

To observe the effect of including repasses between layers on the mechanical properties of the finished laminate, the panels were manufactured in four sections, as illustrated in Figure 2. First, a layer of material was placed on the entire laminate, then, one repass was performed on half of the laminate, followed by a second repass on one quarter of the laminate, leaving half of the laminate without repasses before moving on to the next layer and repeating the process. The repasses were performed by applying heat and pressure using the same processing parameters as during material deposition, and the torch position remained unchanged. After placement, three quarters of the laminate with zero, one and two repasses were sectioned into samples for surface roughness measurement, microscopic inspection, and differential scanning calorimeter (DSC) tests.

2.2. Autoclave treatment

Following the AFP layup of the laminate, one half of the section of the laminate with zero repasses (one-quarter of the full laminate) was bagged and consolidated in the autoclave to be used as reference. The bagged section of the laminate can be seen being placed into the autoclave in Figure 3 with the autoclave cycle in Figure 4. A schematic cross-section of the bagged laminate can be seen in Figure 5. The autoclave air temperature was used to control the cycle. Once consolidated in the autoclave, the final quarter of the laminate was sectioned for inspection and testing to be used as reference to compare to the non-autoclave-treated sections.

3. Surface roughness measurements

To demonstrate the effect of the repass treatment on the surface quality, the surface roughness of the last placed ply of the samples manufactured using different number of repass treatments and autoclave-treated ones were measured using a Mitutoyo SJ-400 surface roughness tester. The measurements were performed on at least three samples with the same treatment along the fiber direction and perpendicular to the fiber direction. The measurement setup is shown in Figure 6. Arithmetic mean deviation from the mean ($R_a$) is reported in Table 1 and plotted in Figures 7 and 8 perpendicular and along the fiber direction, respectively.

As it can be seen from Table 1, the repass treatment improves significantly the surface roughness both along and perpendicular to the fiber direction compared to non-treated samples. With only one repass, the surface roughness improves in the direction perpendicular to the fibers by 6.5 times (from 40.7 to 6.2 μm) and along
These results are very promising and show that applying repasses as *in situ* treatment significantly improves surface finish quality of thermoplastic composites made by AFP. However, as can be seen from Table 1, comparing to the autoclave-treated samples, even samples treated with two repasses have a much rougher surface.

### 4. Crystallinity measurements

The degree of crystallinity of carbon fiber/PEEK specimens were measured using a differential scanning calorimeter (DSC) from TA Instruments. Specimens of approximately 10 mg were prepared and analyzed in a heat-cool-heat cycle with a heating rate of 10 °C/min in nitrogen and cooling rate of 5 °C/min. The degree of crystallinity, $X$, is calculated by:

$$X = \frac{\Delta H_m - \Delta H_c}{(1 - \alpha)\Delta H_f}$$

where $\Delta H_m$ and $\Delta H_c$ are the enthalpies of fusion at melting point and crystallization measured by the area under the endothermic melting peak and the area under the exothermic crystallization peak, respectively. $\Delta H_f$ is the enthalpy of fusion for fully crystalline PEEK which is considered 130 J/g in the analysis. The presence of carbon fibers can be accounted for by $\alpha$ in Equation (1) which is the fiber weight volume of the material. Table 2 summarizes the average value of the degree of crystallinity of at least three specimens tested per section with different treatments.

As it can be observed from Table 2, repass treatments decrease the degree of crystallinity. This could be attributed to the fact that when the AFP head applies heat and pressure without deposing any new material during repass treatment, PEEK material on the surface of the composite layer is re-melted and re-cooled in a very short period of time due to the direct exposure to the environment, which leads to more amorphous phase in the structure of PEEK and lower degree of crystallinity. As expected, autoclave-treated samples
have a higher degree of crystallinity since the cooling rate during autoclave treatment is controlled and much slower than during the AFP process. Melting points of samples with and without repass treatment are the same, however, autoclave consolidated samples have slightly higher melting points.

5. Micrographic study

The effect of repass and autoclave treatments on the microstructure of AFP-made carbon fiber/PEEK composites were studied using microscopy. Samples were cut, embedded in resin, and polished to prepare them for microscopic imaging. Typical micrographs of samples with different treatments are shown in Figure 9. It is evident from these micrographs that repass treatments noticeably change the fiber distribution and makes

![Figure 5. Cross-section of vacuum-bagged laminate: 1. Steel tool plate (9.5 mm) 2. Stainless steel plate (0.744 mm) 3. Bagging film (Thermalimide, Airtech) coated with release agent (Frekote® 770-NC™, Henkel) 4. AFP processed laminate 5. High-temperature sealant tape (SM-5160 TACKY-TAPE®, Schnee-Morehead) 6. 6 oz. plain woven fiberglass cloth 7. Breather (Airweave® UHT 800, Airtech), 8. Vacuum valve (VAC VALVE SSHTR and AHTC 1000 QTD, Airtech).](image)

Figure 6. Roughness measurement setup.

**Table 1. Roughness measurement.**

| Sample condition | \( R_a (\mu m) \) | SD* | \( R_a (\mu m) \) | SD* |
|------------------|------------------|-----|------------------|-----|
| Without repass   | 40.7             | 3.57| 19.3             | 0.44|
| 1 repass treatment | 6.2             | 0.41| 3.8              | 0.27|
| 2 repass treatment | 5.9             | 0.39| 2.6              | 0.32|
| Autoclave treatment | 1.0             | 0.05| 0.5              | 0.04|

*SD = Standard Deviation.

![Figure 7. Roughness measurement perpendicular to fiber direction.](image)

**Table 2. Degree of crystallinity of carbon fiber/PEEK.**

| Sample condition | Crystallinity (%) | \( T_m (°C) \) | SD* |
|------------------|-------------------|----------------|-----|
| Without repass   | 30.4              | 1.3            | 342.6| 0.2|
| 1 repass treatment | 26.1             | 1.3            | 342.8| 0.2|
| 2 repass treatment | 23.7             | 0.2            | 342.6| 0.3|
| Autoclave treatment | 35.9             | 0.3            | 344.0| 0.2|

Note: The weight ratio of PEEK was taken to be 32%.

*SD = Standard Deviation.

![Figure 8. Roughness measurement along the fiber direction.](image)
adjacent layers more distinguishable by separating each layer from its adjacent one. The repass makes the separation lines more visible (yellow splines in the images). However, autoclave treatment allows the fibers to move at each layer interface in a way that adjacent layers are not distinguishable anymore and no discernable layer separation or resin rich area between layers can be seen compared to samples which were not treated in the autoclave.

The effects of repass and autoclave treatments on void content of different composite samples were also measured using micrograph analysis. Several micrographs with 20X magnification were taken along the thickness of each sample, and then image stitching technique through a plugin in ImageJ software was used to stitch the micrographs together to obtain one image with relatively high magnification covering the whole thickness of the sample. The void contents of samples were calculated by color threshold of the images to distinguish the voids from the resin and fibers (see Figure 10(b)). The limitation of this method is that it can capture only voids that are visible at 20X magnification. The values for void content are reported in the Table 3.

Table 3 shows that the void content decreases with repass treatment. While applying one repass treatment after deposition of each layer reduces the void content by 33% (from 0.42 to 0.28%), applying a second repass treatment reduces the void content by a total of 45%.

| Sample condition         | Void content (%) | CV*  |
|--------------------------|------------------|------|
| Without repass           | 0.42             | 0.56 |
| 1 repass treatment       | 0.28             | 0.46 |
| 2 repass treatment       | 0.23             | 0.51 |
| Autoclave treatment      | 0.03             | 0.83 |

CV = Coefficient of Variation.
compared to not applying any repass treatment at all. Autoclave consolidated samples have by far less void content (only 0.03%) in comparison with in situ-treated samples.

6. Discussion and conclusion

The effect of in situ repass treatment on the quality of AFP-made thermoplastic laminate with [0]24 layup sequence was investigated. It should be mentioned that during repass treatments, all process parameters (e.g. torch temperature, compaction force, layup speed etc.) were kept the same as manufacturing process parameters. AFP-made laminate with autoclave treatment was also analyzed to show the effect of autoclave consolidation on the quality of the thermoplastic laminate and to serve as a reference in quality indicators comparison.

The in situ repass treatment significantly improves the surface finish of the laminate, making it more possible to meet aerodynamic smoothness requirements for thermoplastic composites used as aero-surface in aerodynamic application (around $R_a < 5 \mu m$ in the flow direction). The repass treatment effect could be considered analogous to the ironing effect on the garments. While the repass treatment significantly improves the surface smoothness of the thermoplastic laminate both in the fiber and perpendicular to the fiber directions, autoclave-treated samples had the smoothest surface finish among all samples.

Micrograph study showed that repass treatment makes layers more distinguishable from one another. However, autoclave treatment makes no discernable layer separation and reduces resin rich areas between layers. Another contributing factor is crystallinity; autoclave-treated samples have a higher degree of crystallinity in comparison with non-autoclave-treated ones. This is mainly due to the fact that the cooling rate is much slower in the autoclave compared with the AFP in situ process. It was also observed that repass treatments slightly reduce the degree of crystallinity due to re-melting and re-consolidating the matrix in a short period while applying the repass. In order to avoid reduction in the degree of crystallinity due to repass, repass treatment is recommended to be applied only to the last placed ply. This way, while the significant surface smoothness can be achieved, the degree of crystallinity will not be affected. Another observation from the micrograph study was that repass treatments reduce the void content significantly in comparison with non-treated samples. The dominant consolidation-related void dynamics mechanism is void migration (void transportation) along with the resin. During repass treatment heat diffuses into the already placed layers and performs repetitive consolidation.\textsuperscript{8,27,28} However, autoclave-treated samples still have the lowest void content among all samples.

In summary, it can be concluded that for an AFP-made thermoplastic laminate with many layers in one direction, such as the [0]24 lamination in this study, applying one repass treatment significantly improves the surface smoothness and void content. If the primary goal is only to improve the surface smoothness of the AFP-made thermoplastic laminate, it is suggested that repass treatments be applied only to the last layer.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was supported by the Natural Sciences and Engineering Research Council of Canada [grant number N00004].

ORCID

J. Fortin-Simpson \textsuperscript{4} http://orcid.org/0000-0003-2190-4351

References

1. M. A. Khan, P. Mitschang and R. Schledjewski: ‘Identification of some optimal parameters to achieve higher laminate quality through tape placement process,’ \textit{Adv. Polym. Technol.}, 2010, 29, (2), 98–111.
2. M. A. Khan, P. Mitschang and R. Schledjewski: ‘Parametric study on processing parameters and resulting part quality through thermoplastic tape placement process,’ \textit{J. Compos. Mater.}, 2012, 47, (4), 485–499.
3. R. Schledjewski: ‘Thermoplastic tape placement process – In-situ consolidation is reachable,’ \textit{Macromol. Eng.}, 2009, 38, (9–10), 379–386.
4. F. Shadmehri, X. Cai, M. Hojjati, et al.: ‘Determination of optimal process parameters for manufacturing thermoplastic composite rings by automated fiber placement,’ American Society for Composites (ASC), 26th Technical Conference (the second joint US-Canada conference on composites), Montreal, Canada, September 2011.
5. X. Cai, F. Shadmehri, M. Hojjati, et al.: ‘Determination of optimum process conditions for processing AS4/APC-2 thermoplastic composites by automated fiber placement,’ The Society for the Advancement of Material and Process Engineering (SAMPE) conference, Baltimore, MD, May 2012.
6. J. Fortin-Simpson, M. I. El-Geuchy, F. Shadmehri, et al.: ‘Effect of processing parameters on the bending behavior of thermoplastic composite tubes made by automated fibre placement process, American Society for Composites (ASC), 28th Technical Conference, 2013, State College, PA.
7. S. V. Hoa: ‘Principles of the manufacturing of composite materials,’ 2nd ed., Lancaster, PA, DEStech Publications, Inc, 2017.
8. M. A. Khan, P. Mitschang and R. Schledjewski: ‘Tracing the void content development and identification of its effecting parameters during in-situ consolidation of thermoplastic tape material,’ \textit{Polym. Polym. Compos.}, 2010, 18, (1), 1–15.
9. A. Yousefpour and M. N. Ghasemi Nejhad: ‘Experimental and computational study of APC-2/AS4 thermoplastic
10. W. Grouve: 'Weld strength of laser-assisted tape-placed thermoplastic composites,' PhD thesis, University of Twente, Enschede, the Netherlands, August 2012.

11. J. T. John and W. Gillespie Jr.: 'Modeling of heat transfer and void dynamics for the thermoplastic composite tow-placement process,' J. Compos. Mater., 2003, 37, (19), 1745–1768.

12. J. T. John and W. Gillespie Jr.: 'Modeling of in situ strength development for the thermoplastic composite tow placement process,' J. Compos. Mater., 2006, 40, (16), 1487–1506.

13. C. Nicodeau: 'Continuous welding modeling of thermoplastic matrix composites,' Engineering Sciences [physics], Arts et Métiers ParisTech, 2005.

14. J. J. Tierney, R. F. Eduljee and J. W. Gillespie Jr.: 'Material response during robotic tow placement of thermoplastic composites,' Proceedings of the 11th Annual Advanced Composites Conference, 315–329.

15. M. N. Ghasemi Nejhad, R. D. Cope and S. I. Güceri: 'Thermal analysis of in-situ thermoplastic-matrix composite tape laying,' J. Compos. Mater., 1990, 4, (1), 29–45.

16. G. Regnier, C. Nicodeau, J. Cinquin, et al.: 'A Multiphysic and Multiscale Approach to Model the Continuous Welding of Thermoplastic Matrix Composites,' 16th International Conference on Composite Materials (ICCM-16), Kyoto, Japan, 2007.

17. M. N. Ghasemi Nejhad: 'Issues related to processibility during the manufacture of thermoplastic composite using on-line consolidation technique,' J. Composite Mater., 1993, 6, 130–145.

18. M. Hojjat, G. Chouinard, A. Yousafpour: 'Crystallization behavior of PEKK thermoplastic polymer,' The Society for the Advancement of Material and Process Engineering (SAMPE) conference, Long Beach, California, 30 April–4 May 2006.

19. M. J. El-Geuchy: 'Bending behavior of thick-walled composite tubes,' PhD thesis, Concordia University, April 2013.

20. M. Duc Hoang: 'Procedure for making flat thermoplastic composite plates by Automated Fiber Placement and their mechanical properties,' MSc thesis, Concordia University, April 2015.

21. S. V. Hoa, M. Duc Hoang, and J. Simpson: 'Manufacturing procedure to make flat thermoplastic composite laminates by automated fibre placement and their mechanical properties,' J. Thermoplast. Compos. Mater., 2017, 30, (12), 1693–1712.

22. Thermoplastic composite material handbook, Cytec Engineered Materials, 1999.

23. S.-L. Gao and J.-K. Kim: 'Cooling rate influences in carbon fibre/PEEK composites. Part 1. Crystallinity and interface adhesion,' Composites: Part A, 2000, 31, 517–530.

24. D. J. Blundell and B. N. Osborn: 'The morphology of poly (aryl-ether-ether-ketone),' Polymer, 1983, 24, (8), 953–958.

25. C. A. Schneider, W. S. Rasband and K. W. Eliceiri: 'NIH Image to ImageJ: 25 years of image analysis,' Nat. Methods, 2012, 9, (7), 671–675.

26. S. Preibisch, S. Saalfeld and P. Tomancak: 'Globally optimal stitching of tiled 3D microscopic image acquisitions,' Bioinformatics, 2009, 25, (11), 1463–1465.

27. S. Ranganathan, S. G. Advani and M. A. Lamontia: 'A non-isothermal process model for consolidation and void reduction during in-situ tow placement of thermoplastic composites,' J. Compos. Mater., 1995, 29, (8), 1040–1062.

28. R. Pitchumani, S. Ranganathan, T. R. C. Don, et al.: 'Analysis of transport phenomena governing interfacial bonding and void dynamics during thermoplastic tow-placement,' Int. J. Heat Mass Transf., 1996, 39, (9), 1883–1897.