

**POLYTECHNIQUE MONTRÉAL**

affiliée à l'Université de Montréal

**Développement d'un processus de pultrusion de tresses à géométrie creuses de  
composites thermoplastiques pour une application sportive de hockey**

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## **Développement d'un processus de pultrusion de tresses à géométrie creuses de composites thermoplastiques pour une application sportive de hockey**

présentée par **Maissaloun El-Jakl**

en vue de l'obtention du diplôme de *Philosophiæ Doctor*

a été dûment acceptée par le jury d'examen constitué de :

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## DÉDICACE

*À mon père.*

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## RÉSUMÉ

Les bâtons de hockey d'aujourd'hui sont principalement fabriqués à partir de matériaux composites thermodurcissable. En fin de vie, ils sont dirigés vers les sites de décharges. La fabrication des bâtons de hockey est basée principalement sur des procédés manuels, situés dans des pays à faibles coûts de main d'œuvre. Cela a encouragé à explorer des solutions alternatives de fabrication locale. D'où la solution de production de bâtons de hockey en composite thermoplastique par *Braid-trusion*. C'est la combinaison du procédé de tressage textile et de la pultrusion thermoplastique. Ce processus de fabrication est automatisé et peu coûteux qui produit des poutres à sections constantes ayant des fibres orientées en angle qui peuvent être remodelées, réparées et recyclées. Le procédé de *Braid-trusion* thermoplastique reste en cours de développement en raison de certains enjeux de performances. Dans le cadre d'une collaboration industrielle, des éléments de cette technologie sont étudiés pour développer le procédé de production de pièces adaptées à l'industrie.

Le premier objectif consiste à caractériser un nouveau matériau appelé DREF pour limiter l'ondulation des fibres en pultrusion thermoplastique. Les études sur la pultrusion thermoplastique se sont concentrées sur des problèmes d'imprégnation avec des matériaux assemblant de longues fibres polymères aux fibres de renforcement, tels que des fils mélangés et co-enroulés. Cependant, une attention limitée a été accordée à la réduction de l'ondulation des fibres. Une étude expérimentale, basée sur l'application de tensions 3N et 0N sur des fils co-enroulés (CW) et DREF, visait à valoriser la contribution des fibres polymères discontinues à la réduction de l'ondulation des fibres. La réaction de ces matériaux au chauffage a montré un retrait des fibres PET longues, induisant l'ondulation des fibres de verre (GF) dans les fils CW, à l'inverse, les fils DREF présentaient des GF droit. Pour les tensions 3N et 0N, les expériences ont montré une fraction de porosité plus élevée dans les composites avec des fils CW (4,21 % vs 2,19 %) et (2,87 % vs 1,11 %) pour les fils DREF. Les images par tomographie ont révélé une ondulation évidente des fibres dans CW-0N alors que dans DREF-0N, l'ondulation des fibres était difficilement détectable. La pultrusion thermoplastique utilisant des fils DREF a abouti à des composites avec une ondulation des fibres restreinte et des performances mécaniques supérieures malgré l'absence de tension.

Le deuxième objectif consiste à proposer une approche expérimentale démontrant les avantages d'utilisation de plusieurs filières en pultrusion thermoplastique. Les études qui ont adopté la pultrusion thermoplastique multi-filières montrent des résultats prometteurs. Les hypothèses n'ont

pas été démontrées pour l'imprégnation améliorée. Une approche expérimentale imitant les cycles de consolidation de la pultrusion multi-filières par moulage par compression, visait à déterminer l'effet de l'utilisation de plusieurs filières. Le processus multi-filières a effectivement augmenté le temps de consolidation de 260 % et réduit la force de compression de 38 %. De plus, le procédé multi-filières présentait le plus haut degré de micro-imprégnation avec un  $R_{imp}$  de  $96 \pm 4$  %. La filière unique répliquée 5° présentait la charge de compression la plus élevée, ce qui conduisait à une perméabilité réduite des fibres et à un  $R_{imp}$  de  $73 \pm 12$  %. Ce qui indique que l'imprégnation améliorée résulte d'un temps de consolidation prolongé et d'une pression réduite. La principale découverte concerne le potentiel d'optimisation du nombre et de la longueur des matrices pour obtenir la qualité composite souhaitée.

Le troisième objectif consiste à concevoir une tresse permettant d'obtenir un remplissage correct d'une structure creuse rectangulaire avec une approche de consolidation interne. Dans le cadre limité des recherches sur la *Braid-trusion* de composite thermoplastiques creux, l'attention s'est principalement portée sur les structures tubulaires. Il est à souligner que ces études n'ont pas utilisé un modèle de tresses et que toutes les études expérimentales ont été réalisées à l'aide d'un mandrin droit. L'étude présente une méthodologie de conception de tresses pour poutres pultrudées creuses pour des tresses triaxiales. Deux modèles de tresses sont proposés. Le premier présente une circonférence de mandrin de tressage et de pultrusion constante. Le second a une circonférence du mandrin de tressage correspondant à 0,9 fois celle du mandrin de pultrusion, en référence à l'approche du mandrin expansible. La conception a démontré l'effet d'un mandrin expansible sur la réduction des ondulations des fibres en obtenant des longueurs de fil de tressage similaires. Des composites fibre de verre/polyéthylène téréphtalate ont été pultrudés avec des mandrins constants et à expansion. Il a été démontré que l'approche du mandrin expansible peut être utilisée pour la pultrusion de tresses tubulaires. Ces résultats conduisent à la conclusion que le modèle de conception de tresse présente un potentiel prometteur pour la production continue de structures de tresses composite thermoplastique creuses pultrudées avec une ondulation réduite des fibres.

L'étude présentée établit les fondations de Braid-trusion locale de bâtons de hockey. Des travaux futurs sont nécessaires pour fournir un modèle de tresses présentant les caractéristiques mécaniques nécessaires pour des bâtons de hockey de qualité spécifique.

## ABSTRACT

Today's hockey sticks are primarily constructed of thermosetting polymer materials. At the end of their life, they go to landfills, but do not decompose. The manufacturing of hockey sticks is mainly based on manual processes, therefore located in countries with low labour costs. This prompted the exploration of local manufacturing alternatives solutions. Hence, the solution of production thermoplastic composite hockey sticks by Braid-trusion. Braid-trusion is the combination of the braiding textile process and thermoplastic pultrusion. This process is an automated, low-cost manufacturing process that produces beams with constant cross-sections having angle-oriented fibres that can be reshaped, repaired, and recycled. Thermoplastic Braid-trusion remains under development due to some performance issues. As part of industrial collaboration, elements of this technology are being investigated to develop the process for producing suitable parts for the industry.

The first objective consists of characterizing a new input material called DREF to limit fibre waviness in thermoplastic pultrusion. Findings revealed that long thermoplastic fibres induce waviness in the reinforcement fibres. Studies in thermoplastic pultrusion concentrated on mitigating impregnation issues with materials blending long polymer fibres to the reinforcement fibres, such as commingled and co-wound yarns. However, limited attention was given to reducing fibre waviness. DREF yarns are composed of a continuous fibre core onto which discontinuous polymer fibres are applied using the friction spinning process. An experimental study, based on the application of 3N and 0N tension on Co-wound (CW) and DREF yarns, aimed to showcase the contribution of discontinuous polymer fibres on reducing fibres waviness. Input materials' reaction to heating showed long PET fibres shrinkage resulting in wavy glass fibres (GF) in CW yarns, conversely DREF yarns presented straight GF. For 3N and 0N tension, experiments showed higher porosity fraction in composites with CW yarns (4.21% vs 2.19%) and (2.87% vs 1.11%) for DREF yarns. CT scan images revealed a greater amount of fibre waviness in CW-0N whereas in DREF-0N fibre waviness was hardly detectable. The average in-plane shear strength was 55.17 MPa for CW-0N and 119.35 MPa for DREF-0N. Thermoplastic pultrusion using DREF yarns resulted in composites with restricted fibre waviness and superior mechanical performances despite the absence of tension.

The second objective involves on proposing an experimental approach that demonstrates the benefits of using multiple dies in thermoplastic pultrusion. Studies have adopted multi-die systems for thermoplastic pultrusion showing promising results. Some assumptions for the improved impregnation were discussed; however, these hypotheses were not demonstrated. An experimental approach that imitates the consolidation cycles of the multi-die pultrusion by compression moulding, aimed to determine the effect of using multiple dies. The multi-die process effectively increased the consolidation time by 260% and reduced the compressive tool force by 38%. Moreover, multi-die process significantly decreased void content to 2%. Voids in the consolidated plates with single die process were detected in the fibre bundles. Whereas voids with multi-die process were displayed in the resin. Suggesting that the sudden elevated force in the single die reduced fibre permeability. The multi-die consolidated plates had inter-laminar shear strength of 46.5 MPa compared to 28.0 MPa for the single die process. Indicating that enhanced impregnation resulted from prolonged consolidation time and reduced pressure. The primary finding relates to the potential of optimizing dies number and length for desired composite quality.

The third objective consists on designing a braid model for pultrusion of hollow thermoplastic structures. During the braid-trusion process, braids are subjected to tension and compression, which leads to braid deformation and fibre orientation variations. Successful braid-trusion of tubular structures need a braid design methodology that accounts for braid deformations during its passage in the pultrusion die. The study presents a design methodology for hollow pultruded beams reinforced by triaxial braids. Two braid design are proposed. The first has a constant braiding and pultrusion mandrel circumference. The second has a braiding mandrel circumference being 0.9 times of the pultrusion mandrel's, referred to the expanding mandrel approach. The design demonstrated the effect of an expanding mandrel in reducing fibre waviness by achieving similar braiding yarn lengths. Glass fibre/polyethylene terephthalate composites were pultruded with the constant and expanding mandrels. It was demonstrated that the expanding mandrel approach can be used for tubular braid-trusion. Finally, results of these three studies were used to manufacture hockey sticks. These hockey sticks were tested at Bauer Hockey Ltd and successfully met the requirements.

The study presented lays the groundwork for local braid-trusion of hockey sticks. Future work is required to develop a braid model specifying the necessary conditions for achieving specific mechanical properties of hockey sticks.

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## LISTE DES SIGLES ET ABRÉVIATIONS

|               |                                               |
|---------------|-----------------------------------------------|
| <b>DREF</b>   | DR. Ernest Fehrer                             |
| <b>CW</b>     | Co-enroulé                                    |
| <b>PET</b>    | Polyéthylène téréphtalate                     |
| <b>GF</b>     | Fibres de verre                               |
| <b>C/PEEK</b> | Carbon / Polyéther-éther-cétone               |
| <b>PLA</b>    | Acide polylactique                            |
| <b>PP</b>     | Polypropylène                                 |
| <b>PEI</b>    | Polyéthérimide                                |
| <b>PA66</b>   | Nylon 66                                      |
| $T_g$         | Température de transition vitreuse            |
| $T_m$         | Température de fusion                         |
| <b>BY</b>     | Fil de tressage                               |
| <b>AY</b>     | Fil axial                                     |
| <b>DSC</b>    | Calorimétrie Dynamique à Balayage             |
| <b>PVT</b>    | Pression-Volume-Température                   |
| $R_f$         | Taux de remplissage                           |
| $v_f$         | Taux volumique de fibres de renfort           |
| $v_{f,th}$    | Taux volumique de fibres de renfort théorique |

|             |                                                          |
|-------------|----------------------------------------------------------|
| $v_{n,f}$   | Taux volumique de fibres de renfort nominale             |
| $F_f$       | Densité linéaire de fibres de renfort(tex)               |
| $F_m$       | Densité linéaire de fibres de polymère (tex)             |
| $v_v$       | Taux de vides                                            |
| $R_{imp}$   | Ratio de micro-imprégnation                              |
| $L_{eff}$   | Longueur effective de la filière de pultrusion           |
| <b>CAD</b>  | Conception Assistée par Ordinateur                       |
| <b>CBY</b>  | Fil de tressage dans le sens horaire                     |
| <b>CCBY</b> | Fil de tressage dans le sens anti-horaire                |
| <b>UD</b>   | Fil unidirectionnel                                      |
| $R_c$       | Taux d'ondulation de fil                                 |
| $R_m$       | Taux de réduction de la section du mandrin de pultrusion |

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## CHAPITRE 1 INTRODUCTION

Historiquement, le bois dur était le matériau de choix des bâtons de hockey, cependant l'arrivée des matériaux composites a amené à la fabrication de bâtons à haute performance. Ces bâtons sont principalement construits de matériaux à base de polymère thermodurcissable. En fin de vie, ils sont dirigés vers les sites de décharges; cela en raison de la difficulté à recycler les polymères thermodurcissables [1]. Cette difficulté découle du fait que les polymères thermodurcissables se solidifient par une réaction chimique irréversible [2]. En plus, la fabrication des bâtons est principalement basée sur des procédés manuels, donc dépend fortement de la capacité du travailleur [3]. La majorité de ces procédés sont localisés dans les pays à faible coût de main-d'œuvre impliquant des frais de transport intercontinentaux. D'où l'importance de trouver un procédé de fabrication automatisé permettant la production locale, à grande échelle, de bâtons à faible coût, avec une empreinte environnementale réduite tout en garantissant des propriétés mécaniques de haute qualité.

La pultrusion est une méthode de fabrication qui permet une production rapide et peu coûteuse de pièces composites [4]. L'une des principales limitations de ce procédé, est que les fibres sont principalement unidirectionnelles. Une autre méthode de fabrication, qui peut être utilisée pour produire des poutres composites avec des propriétés mécaniques de haute qualité, est le tressage-pultrusion, appelé *Braid-trusion* en anglais. Le procédé *Braid-trusion* est réalisé à partir d'une machine à tresser reliée à une filière de pultrusion pour produire des structures de manière continue avec des sections constantes ayant des fibres orientées [5]. Cela en fait un choix idéal pour la production de bâtons.

Les composites thermoplastiques offrent un potentiel avantageux pour réduire l'impact environnemental par rapport aux matériaux composites thermodurcissables. Cependant, leurs productions demeurent en cours de développement en raison de divers défis, principalement liés à la viscosité élevée des polymères thermoplastiques, ce qui engendre des problèmes d'imprégnation, particulièrement dans les applications où les coûts sont un enjeu majeur [6]–[8]. Pour compenser la viscosité élevée des polymères thermoplastiques, une large variété de fils hybrides est disponible. Les fils hybrides sont composés de fils de polymères et de fils de fibres de renforcement. Ainsi, ils garantissent une faible distance d'imprégnation, ce qui permet de maintenir des temps de procédé réduits [9]. Certains fils hybrides sont coûteux, que ce soit en production ou en stockage. Un

nouveau fil hybride à faible coût est le fil DREF. Celui-ci n'a pas encore été utilisé dans la pultrusion thermoplastique alors qu'il présente un fort potentiel pour être utilisé en industrie de production de composite thermoplastique à faible coût. Une autre voie envisagée par les chercheurs pour réduire les problèmes d'imprégnation des composites thermoplastiques est l'utilisation de multiples filières en pultrusion.

Dans le cadre d'un projet alliant trois partenaires industriels, une initiative de recherche a été entreprise afin de concevoir et promouvoir de nouvelles technologies de mise en forme des composites thermoplastiques. Bauer Hockey Ltd. est une compagnie mondiale reconnue pour ses bâtons de hockey et s'intéresse à la fabrication de bâtons de hockey à faible coût. Filspec, une compagnie productrice de fils textiles, s'intéresse aux applications des fils hybrides dans les composites thermoplastiques. Pultrusion Technique Inc. est un fabricant de pultrusion qui montre un intérêt pour le développement de la pultrusion thermoplastique. L'objectif de ce projet de recherche est de développer un processus de pultrusion de bâtons de hockey en composite thermoplastique. Afin de réaliser ce projet de recherche, trois sous-objectifs sont mis en œuvre. Tout d'abord, il faut évaluer le fil hybride DREF en pultrusion thermoplastique, en examinant son effet sur la réduction de l'ondulation des fibres. Ensuite, il faut démontrer l'impact de la pultrusion multi-filières sur la micro-imprégnation des composites thermoplastiques par réplique par moulage, par compression. Enfin, il faut développer un modèle simplifié de pultrusion de tresse pour obtenir un remplissage correct d'une structure creuse rectangulaire en utilisant une approche de consolidation interne. Dans cette thèse, une revue de littérature est tout d'abord introduite pour situer le travail et définir les objectifs spécifiques du projet. Une méthodologie de travail sera par la suite présentée pour terminer avec les études publiées et les résultats générés avant de conclure.

## CHAPITRE 2 REVUE DE LITTÉRATURE

Les matériaux composites sont définis comme étant un assemblage de deux ou plusieurs matériaux, avec un assemblage final ayant des propriétés supérieures aux propriétés de chacun des matériaux qui le constitue [10]. Les matériaux composites à matrices polymérique sont divisés en deux principales catégories : les composites à matrice thermodurcissable et les composites à matrice thermoplastique. Dans un polymère thermodurcissable, les molécules sont liées ensemble par une réaction chimique irréversible durant le processus de durcissement, pour former des réseaux polymériques à structure tridimensionnelle rigide [11]. Cependant, dans un polymère thermoplastique, les molécules ne sont pas liées par des liaisons covalentes. Elles sont liées par des liaisons faibles d'hydrogène ou de van der Waals qui peuvent être facilement brisées par application de chaleur et puis restaurées lors du refroidissement. Ainsi, un polymère thermoplastique peut être fondu et remodelé [11]. La caractéristique qui sépare les polymères thermoplastiques est l'arrangement de ces molécules après refroidissement. Un polymère thermoplastique amorphe ne contient aucun arrangement des molécules, mais un polymère semi-cristallin contient une phase bien ordonnée, la phase cristalline, dispersée dans une phase amorphe. Le pourcentage volumique de cristallinité influence les propriétés mécaniques du composite et varie selon l'histoire thermique du polymère, la vitesse de refroidissement et aussi d'un polymère semi-cristallin à l'autre [12]. Normalement, un thermoplastique qui subit un refroidissement lent, va cristalliser alors que si le refroidissement est rapide, il subira une solidification dans un état vitreux [13]. Les fibres sont les principaux constituants d'un matériau composite renforcé. Ils partagent la partie majoritaire de la charge qui agit sur une structure composite. Ces fibres renforcent le composite pour avoir de bonnes propriétés mécaniques [11]. Plusieurs fibres sont disponibles en commerce comme les fibres de verre, de kevlar et de carbone.

Les données statistiques révèlent qu'environ 80 % des plastiques consommés sont des thermoplastiques, dont la moitié est utilisée dans des applications à usage unique [14]. Parmi ces plastiques, le polyéthylène téréphtalate (PET) se distingue comme le polyester le plus fabriqué au niveau mondial, largement utilisé dans la fabrication de bouteilles de boissons, d'emballages et de vêtements [15]. Malgré la collecte annuelle de quantités importantes de thermoplastiques recyclables tels que le PET, trouver des marchés viables pour ces matériaux demeure un obstacle majeur [16]. Les thermoplastiques pultrudés présentent un potentiel de réutilisation. Les composites pultrudés pourraient être coupés en granulés pour être utilisés comme thermoplastiques

renforcés de fibres courtes. Les granulés peuvent servir à la fabrication de panneaux de carrosserie et de portes pour l'industrie automobile, ou encore de panneaux et de boîtiers pour l'industrie des appareils électroménagers [16]. Une étude exhaustive a évalué divers thermoplastiques recyclables sur le marché de la pultrusion [16]. L'étude a indiqué que le PET est le choix privilégié en raison de ses propriétés mécaniques et thermiques, de son coût abordable et de sa recyclabilité. Peu de publications ont été trouvées utilisant le PET comme matrice polymérique.

## **2.1 Pultrusion thermoplastique**

Le processus de pultrusion permet la production de produits avec une section transversale constante, qui peuvent atteindre une fraction volumique de fibres élevée [3]. Cette méthode de fabrication est hautement automatisée en raison du faible besoin d'intervention de l'opérateur [3]. Les produits pultrudés présentent d'excellentes propriétés mécaniques, chimiques et structurales, et trouvent des applications dans diverses industries [17]. La pultrusion de composites renforcés de fibres continues avec matrice thermodurcissable, est une méthode de fabrication bien développée et largement utilisée, comparée à la pultrusion avec matrice thermoplastique [2]. Pourtant les composites thermoplastiques offrent de nombreux avantages, notamment une ténacité et une tolérance aux dommages plus élevées, ainsi que la possibilité d'être reformés, recyclés et soudés [2], [18]. Cependant, la viscosité élevée des thermoplastiques fondus par rapport aux thermodurcissables rend le traitement de ces polymères plus difficile. Une mauvaise dispersion des fibres et une mauvaise qualité d'imprégnation peuvent être anticipées [19]. Pour essayer de compenser la viscosité élevée du thermoplastique, plusieurs techniques de pultrusion ainsi que des matériaux d'entrées ont été développés. Ces techniques de pultrusion sont similaires à celles utilisées dans la pultrusion thermodurcissable, telles que la pultrusion par injection de thermoplastiques [20] et la pultrusion thermoplastique réactive [21]. Pour ces techniques, les fibres de renfort secs sont introduites dans la filière de pultrusion séparément du polymère. Des granulés thermoplastiques fondus ou des monomères thermoplastiques sont injectés dans la filière où a lieu l'imprégnation. La pultrusion réactive est utilisée pour profiter de la faible viscosité des monomères et catalyseurs en entrée, afin d'imprégner les fibres avant de compléter la réaction de polymérisation in-situ. Cependant, il existe peu de polymères qui conviennent au traitement réactif et surtout pour des polymères à haute performance [22]. De même, la pultrusion par injection directe nécessite des résines modifiées à faible viscosité pour obtenir une imprégnation complète [20].

### 2.1.1 Pultrusion thermoplastique à fils hybrides

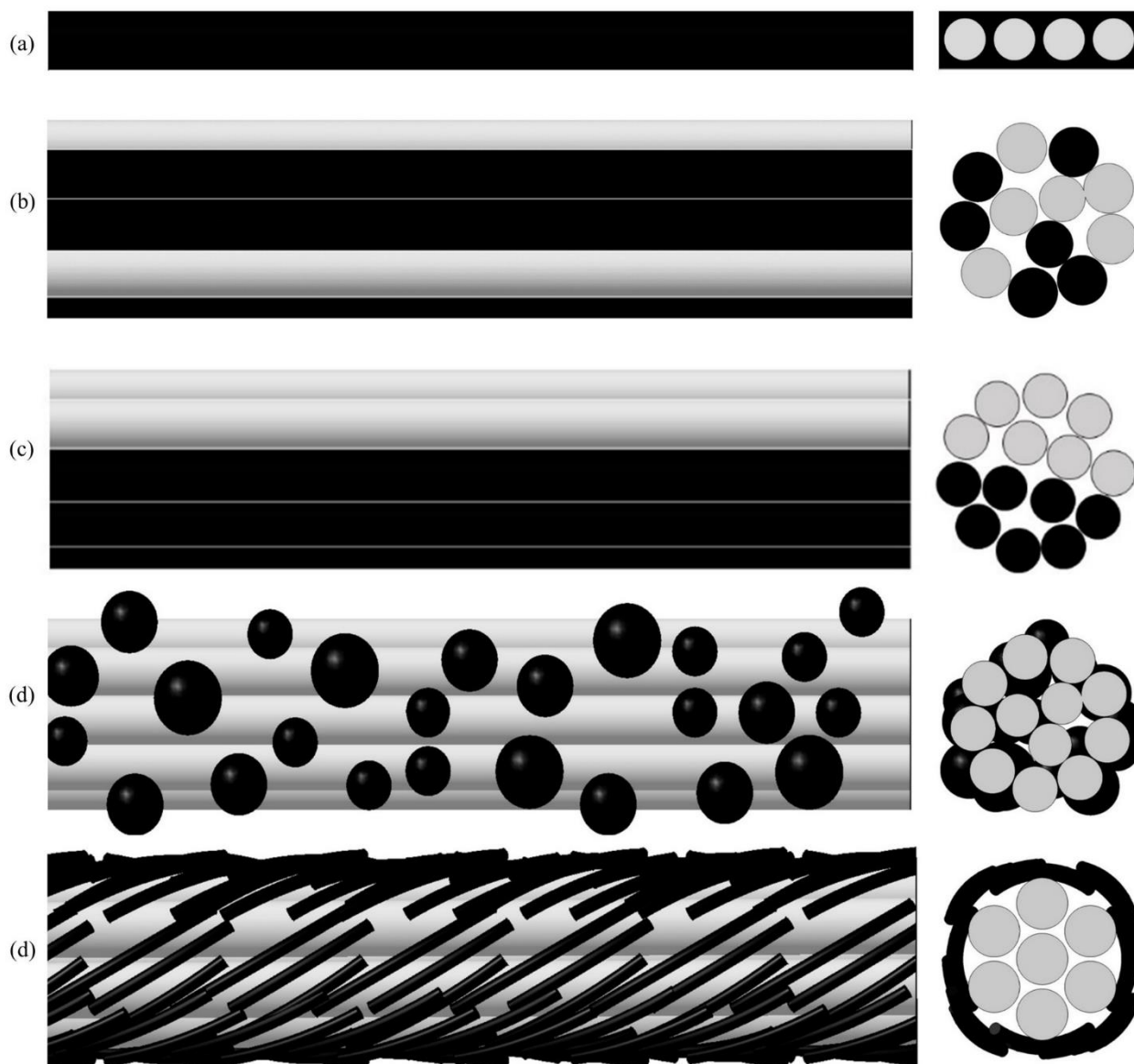


Figure 2-1 Illustrations de fils hybrides (a) préimprégné (*prepregs*), (b) fil co-mélangé (*commingled*), (c) fil co-enroulé (*co-wound*), (d) fil enrobe de poudre (*towpregs*), (e) fil DREF.

Une autre approche pour compenser la viscosité élevée des thermoplastiques, consiste à utiliser des fils hybrides comme matériaux d'entrée, où le polymère est intégré au fil de renfort. Le but d'avoir des fils hybrides comme matériaux d'entrée, est de réduire la distance que le polymère doit parcourir pour imprégner les fibres de renfort. Figure 2-1 présente des illustrations d'exemples de fils hybrides, tels que : ruban préimprégné (*prepregs*) (Figure 2-1 (a)), fil co-mélangé (*commingled*)

(Figure 2-1 (b)), fil co-enroulé (CW) (*co-wound*) (Figure 2-1 (c)), fil enrobe de poudre (*towpregs*) (Figure 2-1 (d)), fil DREF (Figure 2-1 (e)). Les fibres de renfort sont représentées en gris et les fibres de polymère sont représentées en noire. Un ruban préimprégné est constitué de fibres de renforts complètement imprégnées d'un polymère thermoplastique et présente donc une forme assez rigide et une épaisseur très faible; ces rubans sont généralement coûteux [23]. En raison de la rigidité des rubans préimprégnés, le tressage et la fabrication de géométries complexes deviennent des défis supplémentaires [24], [25]. Un fil co-mélangé est composé de fibres de renfort mélangées mécaniquement avec des fibres de polymère thermoplastique continues. Les techniques de mélange impliquent généralement le passage des fibres à travers des canaux à jet d'air [26], [27]. Ces techniques peuvent conduire à des dommages aux fibres [27]. Un fil co-enroulé consiste à enrouler des fibres de renfort et des fibres de polymère thermoplastique continues en parallèle. L'assemblage de ce fil ne nécessite aucune technique de mélange, mais les fils peuvent être tordus [28], [29]. Ce fil est simple à produire, mais la dispersion ou mélange de fibres de renfort et polymère est faible. Un fil enrobé de poudre est constitué de polymère en poudre, mélangé aux fibres de renfort [30]. Ce fil démontre une bonne dispersion de fibres de renfort et polymère, cependant la friction élevée inter fils engendrent une manipulation complexe des fils [31]. Ernst Fehrer a développé et commercialisé un système de fil textiles de filage par friction sous le nom de DREF (DR Ernest Fehrer) [32]. Un fil DREF est constitué de fibres de renfort central continu couverts par une couche de fibres discontinues de polymère. Cette technologie DREF assemble les fibres de polymère discontinus autour de fibres de renfort continu à l'aide d'un procédé de filage par friction [32], [33]. À la connaissance des auteurs, les fils DREF n'ont jamais été utilisés comme matériaux d'entrée dans la pultrusion thermoplastique. Tableau 2-1 résume les fils hybrides utilisés en pultrusion thermoplastique ainsi que leurs avantages et inconvénients.

Tableau 2-1 Fils hybrides utilisé en pultrusion thermoplastique ainsi que leurs avantages et inconvénients

| Fils Hybrides        | Avantages                                                                                                                                    | Inconvénients                                                                                                                           |
|----------------------|----------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------|
| Pré-imprégné         | <ul style="list-style-type: none"> <li>• Vitesse de pultrusion élevée</li> <li>• Ne nécessite pas de processus d'imprégnation</li> </ul>     | <ul style="list-style-type: none"> <li>• Matériel très dispendieux</li> <li>• Ne peut être utilisé dans les procédés textile</li> </ul> |
| Fil co-mélangé       | <ul style="list-style-type: none"> <li>• Flexible</li> <li>• Moyenne dispersion renfort-polymère</li> </ul>                                  | <ul style="list-style-type: none"> <li>• Matériel dispendieux</li> <li>• Endommagement de fibres</li> <li>• Vitesse moyenne</li> </ul>  |
| Fil co-enroulé       | <ul style="list-style-type: none"> <li>• Matériel moins dispendieux</li> <li>• Flexible</li> <li>• Endommagement de fibres minime</li> </ul> | <ul style="list-style-type: none"> <li>• Moyenne dispersion renfort-polymère</li> <li>• Vitesse faible</li> </ul>                       |
| Fil enrobé de poudre | <ul style="list-style-type: none"> <li>• Bonne dispersion renfort-polymère</li> </ul>                                                        | <ul style="list-style-type: none"> <li>• Friction de fibres élevée</li> <li>• Difficile à manipuler</li> </ul>                          |

La Figure 2-2 présente une illustration de la pultrusion thermoplastique avec des fils hybrides comme matériaux d'entrée. Le premier composant de la ligne de pultrusion est le cantre, celui-ci supporte puis oriente les bobines contenant les matériaux d'entrée. Dans certains cas, le cantre applique une tension sur les fils [28]. Le deuxième composant est le préformage ayant pour fonction d'aligner les fils en vue de leur introduction dans la filière, permettant ainsi d'obtenir une disposition contrôlée. Le troisième composant constitue la partie du traitement thermique et du formage du produit final. Celui-ci est divisé en chambre de préchauffe, filière(s) de chauffage et une filière de refroidissement. Lors du passage des fils dans la filière de pultrusion, le matériau est alors appelé "pultrudat". C'est là où le polymère thermoplastique fond et imprègne les fibres de renfort. Le composite passe ensuite dans une filière de refroidissement où la solidification se produit, entraînant la formation du produit composite pultrudé. Finalement, un tracteur situé en aval de la ligne de pultrusion contrôle la vitesse d'avancement et tire les fils.

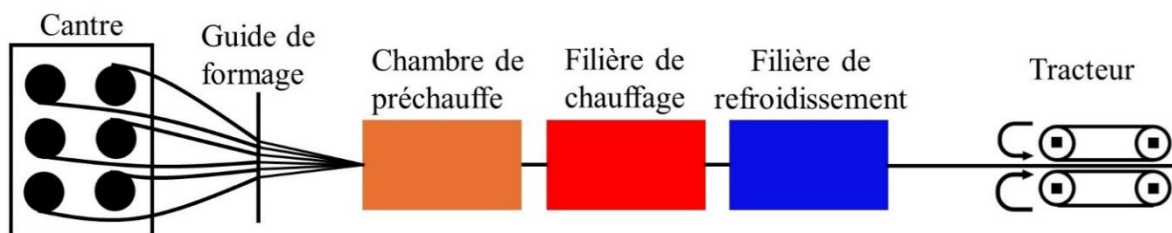


Figure 2-2 illustration de la pultrusion thermoplastique avec des fils hybrides comme matériaux d'entrée

### 2.1.2 Ondulations des fibres

Certaines études de pultrusion thermoplastique avec des fils hybrides comme matériaux d'entrée, soulignent l'importance de maintenir une tension appropriée lors de la pultrusion [9], [28]. Cette tension est maintenue afin d'éviter les ondulations des fibres, susceptibles d'entraîner des dommages aux fibres dans la filière [28]. L'endommagement des fibres peut éventuellement provoquer un arrêt de la pultrusion dans certains cas [28]. Dû à leur procédé de fabrication, les fibres thermoplastiques ont une faible cristallinité et ainsi des contraintes résiduelles élevées [34]. Alors, lors du chauffage les contraintes résiduelles sont libérées, entraînant le retrait des fibres thermoplastiques [28]. Dans les matériaux d'entrée, les fibres thermoplastiques continues sont en contact direct avec les fibres de renfort. En conséquence, le retrait des fibres thermoplastiques entraîne également les fibres de renfort, induisant ainsi des ondulations si la tension n'est pas adéquate. Il est important de noter qu'une tension excessive pourrait également entraîner des dommages aux fibres [35] et réduire leur perméabilité transversale [24].

Les ondulations et le désalignement des fibres contribuent à la réduction des performances mécaniques des composites [36]–[40]. Potter *et al.* [40] a révélé que l'ondulation des fibres non seulement impactait directement les propriétés des composites, mais aussi influençait la variabilité de la fraction volumique en fibres de ces derniers. Lapointe *et al.* [28] a noté une observation de dommages aux fibres de renfort dans le cas où la tension était à 0 N, entraînant ainsi l'interruption de la pultrusion thermoplastique de tiges C/PEEK. Une tension de 3 N était nécessaire pour éviter les dommages aux fils co-mélangés lors de la pultrusion. Cependant, Volk *et al.* [41] a démontré une pultrusion thermoplastique réussie de GF/PET utilisant des fils co-mélangés où aucune tension n'était appliquée aux fibres. Ces expériences de pultrusion n'étaient pas en continu. À l'aide de la

filière transparente développée dans l'étude, il a été observé que, dans la section conique de la filière, les fils semblaient légèrement entremêlés; également, à travers la section droite de la filière, les fibres de renforts semblaient se redresser. La littérature ne montre aucune caractérisation claire sur l'impact de l'ondulation des fibres sur les tiges pultrudées et leurs qualités. De plus, étant donné que la tension peut également endommager les fibres en provoquant la compaction des fibres, aucune étude n'a proposé de solution alternative à l'application de tension sur les fils.

## **2.2 Filière de pultrusion thermoplastique**

La conception de la filière est un aspect critique de la pultrusion thermoplastique. Une filière typique comprend une section décroissante suivi d'une section droite. Cette filière doit être remplie avec le matériel d'entrée. Afin d'assurer l'imprégnation des fibres de renfort, le taux de remplissage doit être supérieur à 100%. Le taux de remplissage de la filière est déterminé par le rapport entre la section transversale du matériau d'entrée et la section de sortie de la filière [12], [42]. Comme la section de la filière de pultrusion se réduit, un excès de résine est repoussé vers l'arrière, générant ainsi un écoulement inverse dans la filière [43]. L'écoulement de la résine entraîne la création d'une pression d'imprégnation à l'intérieur de la filière de pultrusion [26], [43]. Lorsque le pultrudat atteint la section à profil constant, la pression d'imprégnation est supposée se relâcher [43], [44]. Des études ont montré que le degré d'imprégnation dans le pultrudat dépend non seulement de la taille des agglomérations de fibres mais de la géométrie de la filière [20], [43].

### **2.2.1 Micro-imprégnation**

Dans la littérature, plusieurs approches ont été utilisées pour évaluer la qualité de l'imprégnation des composites thermoplastiques lors du processus de pultrusion. Kim *et al.* [43] a proposé une approche analytique liant deux sous-modèles pour représenter distinctement, l'écoulement du polymère entre les agglomérations ainsi que l'imprégnation de ces derniers. Cela a permis la catégorisation de l'écoulement de polymère en deux types : l'écoulement de macro-imprégnation et micro-imprégnation. L'écoulement de macro-imprégnation est identifié comme l'écoulement le long de la direction des fibres de renfort. Généralement, la perméabilité axiale des fibres de renfort est élevée. L'écoulement de micro-imprégnation est identifié comme l'écoulement à l'intérieur des agglomérations des fibres où la perméabilité est faible. Dans ce cas, le polymère s'écoule de l'extérieur des agglomérations de fibres vers le centre et se diffuse entre les fibres de renfort. La

micro-imprégnation est généralement modélisé par la loi de Darcy [26], [43]. L'écoulement inverse créer lors de la pultrusion constitue l'écoulement de macro-imprégnation, celui-ci génère une pression qui entraîne l'écoulement de micro-imprégnation. Plusieurs études ont évalué le degré d'imprégnation dans la pultrusion thermoplastique en évaluant l'écoulement de micro-imprégnation [26], [43], [45]. Ces études caractérisent cette micro-imprégnation par l'avancement du polymère. Afin d'identifier expérimentalement la qualité d'imprégnation générale des composites pultrudés, des chercheurs ont procédé à des analyses de porosité en examinant les microscopies des sections [12], [28], [42]. Récemment, Kazano *et al.* [46] a caractérisé la micro-imprégnation en calculant un ratio de micro-imprégnation, identifié en divisant la zone imprégnée par la surface totale de la section transversale d'un toron de fibres. Cette méthode de caractérisation semble être une approche expérimentale efficace pour identifier la micro-imprégnation des composites.

## 2.2.2 Pultrusion multi-filière

Dans les dernières années, un système de pultrusion à filières multiples (multi-filières) a été développé au labSFCA de Polytechnique Montréal [47], [48]. Ce processus induit un profil de pression cyclique lors de la fabrication du pultrudat. Oswald *et al.* [29] ont rapporté la fabrication de tiges cylindriques en lin/PLA en utilisant la pultrusion multi-filières. Lapointe *et al.* [28] ont aussi utilisé le processus de pultrusion thermoplastique multi-filières pour la fabrication de tiges en C/PEEK. Les résultats de ces deux études ont montré que le processus multi-filières a amélioré l'imprégnation sans induire une force de traction élevée. Eichenhofer *et al.* [49] ont également développé un système de pultrusion multi-filières pour une utilisation dans la fabrication additive. L'étude a indiqué une imprégnation adéquate, ainsi qu'une réduction du taux de déconsolidation. Plusieurs hypothèses ont été discutées concernant l'amélioration de l'imprégnation. Une proposition a indiqué que le processus multi-filières aidait à atteindre la température de fusion du polymère ainsi que la division de l'écoulement de polymère [29]. Une autre perspective a suggéré que la durée sous pression était prolongée avec le processus multi-filières [49]. Lessard *et al.* [45] a proposé une approche de modélisation pour explorer le mécanisme de déconsolidation et son effet sur la microstructure du composite. Les résultats ont indiqué que les cycles de déconsolidation ont induit une amélioration de la perméabilité des fibres. La littérature propose plusieurs théories concernant les avantages du processus multi-filières. Cependant, il serait nécessaire de mener

davantage d'investigations pour comprendre l'amélioration de l'imprégnation lors de l'utilisation d'une séquence de filières courtes.

## **2.3 Pultrusion de tresse (tresse-trusion ou Braid-trusion)**

### **2.3.1 Les tresses et leurs avantages**

Le tressage est défini comme étant un processus textile qui consiste à entrelacer au moins trois fils pour fabriquer une structure continue [50]. Les formes de tressage conventionnelles sont les tresses planes (2D) et les tresses tridimensionnelles (3D). Parmi les tresses 2D, on retrouve les tresses plates, les tresses tubulaires biaxiales et les tresses tubulaires triaxiales [51]. Les tresses triaxiales sont différenciées des biaxiales par la présence de fils dans l'axe de la tresse, appelé fils axial (AY). Principalement, la distinction entre les tresses à 2D et 3D est par rapport à l'épaisseur et l'entrelacement de leurs fils [50]. Pour une tresse tubulaire 2D avec une épaisseur, la forme totale est formée de plusieurs couches, donc plusieurs tresses, une sur l'autre. Pour une tresse tubulaire 3D, il y aura des fils orientés dans la direction de l'épaisseur liant les couches entre elles. La Figure 2-3 présente un schéma qui montre les principales caractéristiques d'une tresse : le diamètre ( $D$ ), l'angle ( $\alpha$ ) et le pas ( $p$ ), ainsi que leur relation géométrique. L'angle de la tresse est l'angle entre les fils de renforcement et l'axe central de la tresse. Le pas de la tresse est la distance de la section répétable de la tresse c'est-à-dire la distance que prend le fils de tressage (BY) pour retourner à sa position sur l'axe. L'utilisation de composites obtenus par tressage offre la possibilité de réaliser des structures à hautes performances [52]. La raison de cet intérêt est le fait de pouvoir maximiser les propriétés mécaniques dans plusieurs directions de la pièce fabriquée.

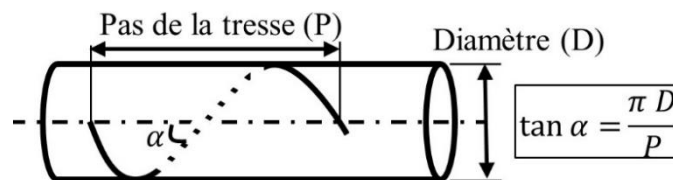


Figure 2-3 Les paramètres de la tresse, le diamètre, le pas de la tresse et l'angle, ainsi que la formule qui les relie

### 2.3.2 Pultrusion de tresse

Le processus de braid-trusion combine le tressage et la pultrusion où la machine de tressage est jointe à la ligne de pultrusion [53]. Ce processus produit des profils à sections constantes ayant des fibres orientées selon des angles précis. Michaeli *et al.* [25] a été un des premiers à introduire le tressage à la pultrusion thermoplastique et a aussi tenté la production de tubes. L'étude a comparé deux types de fils et matériels, des fils hybrides parallèles et des fils mélanger utilisant PP/verre et PEEK/Carbone respectivement. À l'aide de leur modèle de préchauffage, Bechtold *et al.* [54], [55] a démontré l'efficacité d'utiliser un système de préchauffage par contact pour la pultrusion de la tresse de verre/PP, en montrant une amélioration des propriétés mécaniques. Ces études n'ont pas proposé de méthodologie de conception pour la tresse à pultruder, mais ont utilisé un système d'essai-erreur. Une des premières publications à introduire la conception de tresse pour le processus de Braid-trusion est celle de Laberge Lebel et Nakai [5]. Ils ont développé un modèle géométrique simple pour les tresses triaxiales pleines. Celui-ci est basé sur la déformation circonférentielle d'une tresse triaxiale lors de la compression dans la filière. Leur modèle a été utilisé pour concevoir la tresse permettant un remplissage correct de la filière et limiter la déformation de la tresse durant la pultrusion. Par l'application du modèle, une poutre thermoplastique en forme de "L" a été fabriquée avec succès. Ils ont assumé que, seule la déformation circonférentielle latérale des tresses triaxiales se produit au cours du processus puisque les fils axiaux empêchent la déformation longitudinale; et ce, contrairement aux tresses biaxiales où la déformation pendant la pultrusion se produit à la fois dans le sens longitudinal et latéral [56]. Ghaedesharaf *et al.* [56] a réalisé une pultrusion de tresse biaxiale. Dans un premier article, un simple modèle géométrique qui prédit la géométrie finale de la tresse après pultrusion a été proposé. Le résultat présenté montre des erreurs significatives dans la prédiction du diamètre initial de la tresse, avant pultrusion. Cela pourrait être dû à une mauvaise prédiction du ratio de compaction. Par la suite, Ghaedesharaf *et al.* [57] a

proposé un modèle par éléments finis pour prédire la géométrie 3D des tresses biaxiales en modélisant les fibres comme poutre en flexion. Le même auteur [53] a réussi à développer ce modèle pour prédire la déformation de la tresse durant la pultrusion. Cette étude néglige la présence du polymère et ses effets et considère un coefficient de frottement (fibre-fibres) indépendant de la position et de la température.

### 2.3.3 Pultrusion de tresse creuse

Tableau 2-2 résume les tresses thermoplastiques creuses pultrudées trouvées dans la littérature.

**Conception de la tresse** - Les tresses utilisées sont des tresses plates (2D), biaxiales et triaxiales, composées de plusieurs couches. Le nombre des couches ainsi que leur angle varient entre 2 et 4 et 30° et 45° respectivement. Cependant, la conception de la géométrie de la tresse, le choix de tresse biaxiale ou triaxiale, de diamètre, de l'angle et du nombre de couches n'a pas été montré. Memon *et al.* [58] a noté l'importance de la conception de la tresse, mais ne la décrit pas. Schafer *et al.* [59] pultrude une section carrée, mais n'indique aucun problème de remplissage de coins.

**Mandrin** - Memon *et al.* [58] a utilisé un mandrin chauffé à une température qui est légèrement inférieure à la Tg du polymère PLA. Cependant, Saito *et al.* [60] a utilisé un mandrin chauffé à la Tg du PA66. Les auteurs n'ont pas donné d'explication concernant le choix de température. L'étude a montré que lorsque la matière commence à être tirée du mandrin dans la filière de refroidissement, la force augmente. Ce qui indique une friction élevée de la matière sur le mandrin à cette zone. Schafer *et al.* [59] a utilisé un préchauffage par contact pour fondre le polymère en surface, et pour pouvoir atteindre la température de traitement dans la première couche de la tresse. Tous les articles ont utilisé une filière longue à différentes zones de chauffage.

**Qualité du pultrudat** - Memon *et al.* [58] a montré que la qualité d'imprégnation dans la tresse augmente avec le taux de remplissage et la température de traitement, mais le taux de vide est toujours élevé. Saito *et al.* [60] a montré que l'entrelacement des filaments de verre et de PA66 à basse viscosité a amélioré l'imprégnation, mais le fini de surface reste irrégulier. Schafer *et al.* [61] a indiqué que le mandrin reste dans la structure. La microscopie indique la présence de vide entre les couches et des ondulations de fibres.

Aucune étude ne semble avoir utilisé une conception de tresse. De plus, toutes les études ont utilisé un mandrin à section constante. Finalement, vu la qualité des pultrudats creux tressés disponibles dans la littérature, la pultrusion réussie de tresses creuses thermoplastiques n'a pas été démontrée.

Tableau 2-2 Tresses thermoplastiques creuses pultrudées trouvées dans la littérature

| Tresse tubulaire et conception                                    | Mandrin                                  | Qualité de pultrudat                                            | Ref           |
|-------------------------------------------------------------------|------------------------------------------|-----------------------------------------------------------------|---------------|
| Tresse triaxiale<br>Angle = 30- 38°<br>48 BY – 24 AY<br>2 couches | Mandrin à section<br>constante chauffé   | Finis de surface: Très<br>irrégulier<br><br>Taux de vide : >10% | [58],<br>[62] |
| Tresse biaxiale<br>Angle = 30°<br>16 BY<br>4 couches              | Mandrin à section<br>constante chauffé   | Finis de surface :<br>Irrégulier<br><br>Taux de vide : 0.5-1.7% | [60]          |
| Tresse biaxiale<br>Angle = 45°<br>36 BY<br>4 couches              | Mandrin à section<br>constante en mousse | Finis de surface : N/A<br><br>Taux de vide : N/A                | [61]          |

## CHAPITRE 3 OBJECTIFS DE RECHERCHE

### 3.1 Identification du problème

Premièrement, la revue de littérature rapporte des exemples de fils hybrides utilisés dans la pultrusion de composite thermoplastique en tant que matériaux d'entrée. Ceci met en évidence diverses considérations concernant l'utilisation des fils hybrides. Parmi celles-ci, le coût élevé de certains fils hybrides constitue un obstacle majeur. De plus, la répartition des fibres de polymères et renforts dépend fortement des techniques de mélange, ces techniques peuvent également créer des risques d'endommagement des fibres. De plus, la manipulation difficile et la rigidité de certains fils hybrides présentent des défis pour leur incorporation dans des procédés de tressage. Par ailleurs, des chercheurs ont remarqué la création de dommages aux fibres de renfort causés par un manque de tension sur celles-ci. Il était supposé que le retrait des fibres de polymère continu entraîne et replie les fibres de renfort, créant ainsi des dommages et éventuellement un arrêt de la pultrusion. Cependant, ce retrait n'a pas été évalué ni mis en évidence. Par conséquent, il est important de développer un nouveau matériel d'entrée pour la pultrusion de composite thermoplastique pour remédier aux problèmes mentionnés précédemment.

Deuxièmement, la revue de littérature fait état des hypothèses avancées concernant l'amélioration de l'imprégnation dans un système multi-filières de pultrusion de composite thermoplastique. Ces hypothèses se focalisent sur la réduction de la pression de consolidation, la prolongation de la durée de consolidation, tandis que d'autres voient dans la déconsolidation, la principale cause des bénéfices du processus multi-filières. Néanmoins, des investigations supplémentaires sont nécessaires pour une compréhension approfondie de l'amélioration de l'imprégnation lors de l'utilisation de séquence de filières courtes dans le processus de pultrusion de composites thermoplastiques.

Finalement, les recherches sur la pultrusion des composites thermoplastiques de tresses creuses ont principalement reposé sur une approche d'essais-erreurs, sans qu'une méthodologie correspondante pour la conception de la tresse soit mise en place. Les modèles présentés dans la revue de littérature suggèrent que les fils utilisés dans la préforme tressée doivent avoir la capacité d'adapter leur forme pendant la consolidation. Cependant, ces modèles sont complexes, nécessitant des ressources importantes en termes de temps et de moyens, et n'ont pas été validés pour les tresses tubulaires. De plus, toutes les méthodes de consolidation de pultrusion des composites thermoplastiques de

tresses creux ont utilisé un mandrin à section constante. La revue de littérature met en évidence le besoin de concevoir un modèle simple et abordable pour la braid-trusion à section creuse.

### **3.2 Objectifs de recherche**

L'objectif général du projet est de développer un processus de pultrusion de tresse à géométries creuses de composites thermoplastiques pour une application dans le domaine du hockey. Ce processus doit combiner le tressage de fils hybrides et la mise en forme par pultrusion.

**Objectif 1 : Caractériser le fil hybride 'DREF' pour limiter les ondulations des fibres en pultrusion thermoplastique.**

Une étude expérimentale est réalisée pour mettre en évidence la contribution des fibres de polymères discontinues à la réduction des ondulations des fibres de renfort dans les tiges pultrudées.

**Objectif 2 : Démontrer l'effet de la pultrusion multi-filières sur la micro-imprégnation des composites thermoplastiques.**

Une approche expérimentale a été développée pour reproduire le processus de consolidation de la pultrusion de composites thermoplastiques par moulage par compression. La micro-imprégnation des composites a été étudiée pour mettre en évidence l'effet de multi-filières.

**Objectif 3 : Développer un modèle pour la pultrusion de tresses creuses en composite thermoplastique en utilisant une approche de mandrin à section ascendante.**

Un modèle géométrique de tresse tubulaire a été développé pour permettre un remplissage correct d'une structure creuse rectangulaire avec un mandrin à section ascendante.

### **3.3 Cohérence des articles**

Les trois objectifs de cette thèse sont présentés sous forme d'articles.

L'article 1, couvrant la réalisation de l'objectif 1, est soumis au journal Composites Part B: Engineering et présenté au chapitre 4. Cet article est en processus de révision par les paires. L'étude introduit la faisabilité de la pultrusion thermoplastique avec des fils hybrides 'DREF'. L'analyse de la structure du fil a révélé une uniformité remarquable dans la répartition des fibres de polymère et de renfort. Des expériences de pultrusion sont réalisées avec des fils co-enroulés aussi. Les expériences ont confirmé le rétrécissement des fibres polymères continues dans les fils co-enroulés,

induisant l'entrelacement des fibres de renforts. Le fils DREF a démontré que la tension n'est pas requise pour garantir des fibres droites et une bonne imprégnation dans un composite thermoplastique, validant ainsi son utilisation pour la pultrusion thermoplastique.

L'article 2, couvrant l'objectif 2, est soumis au journal *Composites Part A: Applied Science and Manufacturing* et présenté au chapitre 5. L'étude propose une méthodologie pour reproduire le processus de consolidation de la pultrusion de composites thermoplastiques par moulage par compression. Cette méthodologie est utilisée pour la conception de trois filières uniques ainsi qu'un système de quatre filières consécutives. La déconsolidation et la compaction cyclique lors de l'expérience multi-filières ont amélioré l'imprégnation du composite sans nécessiter de temps de consolidation excessif. Ces travaux suggèrent que l'utilisation de système multi-filières est recommandée.

L'article 3, couvrant l'objectif 3, est soumis au journal *Materials & Design* et présenté au chapitre 6. L'étude propose un modèle géométrique de tresse tubulaire avec un mandrin à section ascendante. Le modèle démontre que l'utilisation d'un mandrin ascendant entraîne une adaptation des fils dans la tresse, favorisant ainsi une pultrusion correcte avec un remplissage adéquat de la filière. Indiquant ainsi la possibilité de réaliser une pultrusion continue de tresses creuses.

Finalement, les travaux expérimentaux rapportés dans le chapitre 6 démontre une pultrusion de bâton de hockey réussie. Cette pultrusion correspond à la pultrusion de tresses creuses à section rectangulaire en composite thermoplastique en utilisant des fils DREF à l'aide d'un processus de pultrusion multi-filières.

## **CHAPITRE 4     Article 1: Fibre waviness reduction in thermoplastic pultrusion by using DREF yarns.**

Maissaloun El-Jakl, Louis Laberge Lebel

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### **Abstract**

Non-reactive thermoplastic pultrusion impregnation issues are mitigated by using hybrid input materials. Co-wound (CW) and commingled yarns are an assembly of continuous polymer and reinforcement fibres. Continuous thermoplastic fibres have shown to induce waviness in the reinforcement fibres during pultrusion due to their shrinkage at high temperature. DREF yarns are composed of a core of continuous reinforcement fibres onto which discontinuous polymer fibres are applied using the friction spinning process. This study, based on the application of 3N and 0N tension on CW and DREF yarns, aimed to highlight the contribution of discontinuous polymer fibres on reducing reinforcement waviness in pultruded rods. CW yarns' reaction to heating showed continuous polyethylene terephthalate (PET) fibres shrinkage resulting in wavy glass fibres (GF). Conversely, the GF in DREF yarns remained straight. Pultrusion experiments with yarn tension of 3N were done to alleviate the GF waviness. However, the porosity was rather high at 4.2% for CW rods and 2.3% for DREF rods. Pultrusion experiments without tension showed lower porosity of level of 2.9% for CW yarns and as low as 1.1% for DREF yarns. However, CT-scan image indicated GF waviness in CW rods. GF in DREF rods remained straight. The in-plane shear strength reached 119 MPa. Thermoplastic pultrusion using DREF yarns resulted in composites without reinforcement fibre waviness, lower porosity level and superior shear strength.

### **4.1 Introduction**

The increasing adoption of thermoplastic composites in various industries can be attributed to their numerous advantages over thermoset composites [2]. These advantages include high impact resistance and enhanced fracture toughness [2], [11]. Additionally, their post-forming, re-shaping, and recycling ability makes them preferred replacements for metallic and traditional thermoset parts in sectors like automotive, aerospace, and marine industries [6], [63], [64]. Environmental concerns about waste management are increasing due to the recycling challenges posed by thermoset composites. These materials often end up in landfills, highlighting the need for better material alternatives, like thermoplastics [6], [64]. However, their production and usage remain

under development due to inherent challenges, primarily the high viscosity of thermoplastic polymers resulting in impregnation issues, especially in cost-sensitive applications [6]–[8].

The pultrusion process enables the production of products with a constant cross-section that can achieve a high fibre volume fraction [3]. This manufacturing process is highly efficient due to the little need for operator intervention [3]. Pultruded products exhibit excellent mechanical, chemical, and structural properties, and find applications across various industries [17]. Continuous pultrusion of fibre-reinforced composites with thermoset matrix is a well-established and widely used manufacturing method, compared to thermoplastic matrix pultrusion [2]. Due to the advantages offered by thermoplastic materials, thermoplastic pultrusion has gained popularity over the past 50 years through the usage of different technologies and input materials [2], [18].

The drawback in thermoplastic pultrusion is related to the impregnation challenges due to their molten viscosity being 2 to 3 orders of magnitude higher than thermosets polymer [19]. To try to compensate for the high viscosity of thermoplastics, several pultrusion techniques and input materials have been developed. The term ‘input material’ refers to a range of material formats used for pultrusion manufacturing. This includes reinforcement fibres, such as carbon or glass, polymer pellets, prepregs, and hybrid yarns. Similar techniques to those used in thermoset pultrusion, such as thermoplastic injection pultrusion [20], and reactive thermoplastic pultrusion [21] are employed. For these techniques, dry reinforcement yarns are fed into the pultrusion die separately from the polymer. Melted thermoplastic pellets or thermoplastic monomers are injected into the die where the impregnation takes place. Another approach is to use hybrid yarns as input materials, where the polymer is integrated to the reinforcement yarn. The idea is to reduce the distance that the polymer must travel to impregnate the reinforcement fibres. Such examples of input materials are called prepregs, towpregs, commingled, CW and DREF yarns. Prepregs are thin pre-impregnated tapes that are comprised of fully impregnated reinforcement fibres with a thermoplastic polymer, resulting in a relatively rigid form and are typically expensive [23]. Commingled yarns consist of reinforcement fibres that are mechanically mixed with continuous polymer fibres. Mixing techniques usually involve passing the fibres through air-jet channels [26], [27]. These yarns are typically more expensive than the ones that follow. Towpregs are made by mixing powdered polymer to the reinforcement tows [30]. CW yarns are composed of reinforcement tows and thermoplastic polymer tows arranged side by side. The assembly of these CW yarns does not require any mixing technique. To ensure their integrity, these yarns are sometimes twisted [28],

[29]. In the nineteen seventies, Ernst Fehrer developed and commercialised a friction spinning system for textiles under the name DREF [33]. DREF yarns are composed of continuous reinforcement fibre core onto which discontinuous polymer fibres are spun [33]. To the authors' knowledge, DREF yarns were never used as input materials in thermoplastic pultrusion.

During hybrid yarn input material thermoplastic pultrusion, input materials are wound into bobbins are positioned in a creel. The creel is used to hold the bobbins and to regulate their unwinding tension [28]. These yarns are then drawn through a preheater, a heated die, and a cooling system using a puller mechanism. At the end of the manufacturing line, a saw is employed to cut the finished products. The crucial stage of melting and impregnation occurs within the heated die. This die is typically configured with a tapered section to create impregnation pressure, followed by a short constant section for shaping the product [25]. The pultrusion process involves filling the die with the material. To ensure the impregnation of the reinforcement fibres, a filling ratio exceeding 100% is required. The die filling ratio is calculated as the ratio between the cross-sectional area of the input material and the exit area of the die [12], [42]. While input material yarns enter the heated dies, the excess melted polymer creates a backflow, which contributes to the impregnation pressure [65]. From that point on, the material is referred as a pultrudate. Subsequently, the pultrudate enters a cooling die. The cooling die maintains pressure while the composite temperature cools down. Its purpose includes preventing deconsolidation and facilitating shrinkage for releasing the pultrudate from the die surface [12]. In recent publications, a sequence of heated dies arranged with a decreasing final cross-section was employed to enhance impregnation in thermoplastic composites [12], [28], [29], [42], [66], [67]. This multi-die pultrusion system was used to produce Flax/PLA [29], Flax/PP [67], C/PEI [12], [42] and C/PEEK [12], [28] rods with low volume content. These studies have demonstrated the efficacy of the multi-die pultrusion system in enhancing impregnation.

Surface imperfections manifested as rough fibre finish on the composite surface are caused by a phenomenon known as sloughing in pultrusion terminology [65]. Sloughing is common to thermoset and thermoplastic pultrusion and is attributed to polymer adhesion to the metallic die surface. During pultrusion, as the composite is getting pulled through the cooling die, if polymer-metal adhesive forces are higher than the composite strength, fragments of the pultrudate can tear off [12]. Carlsson and Astrom [65] compared the imperfections caused by sloughing with different cooling die temperatures and pulling speeds. Alsinani *et al.* [12] proposed to minimize the apparent

sloughing critical distance which was considered to be the distance where the composite temperature is between the processing temperature and the glass transition temperature ( $T_g$ ). In order to mitigate the sloughing phenomenon, composite shrinkage must be exploited. Where in the case of semi-crystalline polymers shrinkage occurs during crystallization, which happens at temperature higher than  $T_g$  [12]. For both mentioned studies, surface finish was characterized using the roughness parameter. It is suspected that increasing composite strength would reduce the probability of sloughing [68] and that polymer-rich surface could lead to excess sloughing [69]. These propositions lead to the potential for a correlation between yarn tension and sloughing. Seeing that yarn tension induces yarn diameter compaction, which reduces transverse permeability of the fibres [19] and leads to polymer-rich areas around the compacted fibres.

The quality of a composite is typically assessed through reinforcement fibre tow impregnation, as it directly influences mechanical properties. Low fibre tow impregnation decreases the mechanical performance [70]. The fibre tow impregnation is most frequently analysed by measuring the void content [71]. Another route to characterize impregnation quality is by measuring the shear strength of the composite. Shear strength is affected by the composite constituents' individual properties, fibre-matrix interfacial strength, and void content [72]. For a given composite material, different manufacturing conditions have a greater influence on the latter than the first two. Studies have shown a decrease of more than 50% in in-plane shear strength when the void content increased from 1% to 4% [29], [73]. This suggests that conducting in-plane shear testing offers a reliable method for assessing composite quality.

Statistics reveal that approximately 80% of consumed plastics are thermoplastics, with half of them utilized in single-use applications [14]. Among these plastics, polyethylene terephthalate (PET) stands out as the most manufactured polyester globally, extensively employed in beverage bottles, packaging, clothing, and carpeting [15]. Despite the annual collection of significant amounts of recyclable thermoplastics such as PET, finding viable markets for these materials remains a major obstacle [16]. Notably, pultruded thermoplastics possess the potential to be repurposed by chopping them into chips for use as short fibre-reinforced thermoplastics. These can be used in moulding automotive body panels, doors, and fenders, or for panels and housings in the appliance industry [16]. A comprehensive study has evaluated various recyclable thermoplastics for their suitability in the pultrusion market [16]. Their findings indicate that PET emerges as the first choice

based on its mechanical and thermal properties, cost-effectiveness, and recyclability. Few publications were found using PET as the polymeric matrix. Asensio *et al.* [30] used recycled PET as matrix and E-glass fibres as reinforcements. Three types of recycled PET matrix were used in their study, one recycled from bottle waste, the second was the same recycled PET for bottle waste though it was modified to lower its viscosity and the third one was a multi-layered and coloured recycled PET. All three materials were ground into powders and transformed into towpregs before pultrusion with 45% matrix weight content. The pultruded profile was a 20 x 2 mm<sup>2</sup> bar. The pultrusion process speed was at 200mm/min, processing temperature was set at 280°C and cooling die temperature at 25°C. The pultruded profiles had an average of 64% fibre volume content and an average tensile modulus of 47 GPa. There were no quantitative measures for void content. The pultruded plate containing the modified PET with lower viscosity indicated the highest tensile modulus and lowest void content. The study did not analyse yarn tension or surface finish. This experimental study confirmed the feasibility of recycled PET pultrusion. Volk *et al.* [41] recently published a study on thermoplastic pultrusion using PET as their polymer matrix. A commingled yarn material containing PET and E-glass with a fibre volume content of 55% was used during this study. The pultrusion subsystem consisted of a batch pultrusion approach, which allowed the manufacturing of a pultruded rod with limited length of 1500mm. The batch pultrusion consisted of pulling a loop of input materials by the means of a steel wire. This approach was not continuous and did not necessitate a creel. This indicated that there was no tension applied on the yarn at the beginning of the pultrusion. Throughout their experiments, processing and cooling temperatures remained at 260°C and 30°C respectively. Multiple speeds and preheating temperatures were tested for different rod diameters. The study indicated that for speeds below 100 mm/min and for different rod diameters, the void content was below 2%. Micrographs showed an inhomogeneous fibre distribution over the cross-section, suggesting that the fibre arrangement within the yarns and prior to pultrusion, significantly affects the consolidation quality of the pultrudate. Assisted by the transparent die developed in their study, it was observed that the yarn bundles increased in diameter upon entering the die's tapered section and their configuration appeared somewhat entangled. Through the straight section of the die, the bundle of yarns appeared as if they were straightening. A crucial point to consider would be the absence of yarn tension and guidance in this study. No characterization was conducted on the effect of the entanglement or waviness of the commingled

yarns on the pultruded rods. Evidently, an investigation into the effect of the absence of tension on the quality of the pultrudate is necessary.

The absence of yarn tension during composite manufacturing may induce fibre waviness. The reason behind this is attributed to the response of thermoplastic fibres to heating. The thermoplastic fibres included in the input materials are produced through a melt spinning process, which results in thermoplastic fibres with low crystallinity and high residual stresses [34]. During heating, these residual stresses are released, resulting in the shrinkage of the thermoplastic fibres [28]. In the input materials, such as commingled or CW yarns, thermoplastic fibres are in close contact with the reinforcement fibres. Due to that contact, when the input materials are heated, the shrinkage of the thermoplastic fibres could induce entanglement or waviness in the reinforcing fibres. Several studies on non-pultruded composites have found that fibre waviness and misalignment contribute to the reduction of the mechanical performance of the composites. Yokozeki *et al.* [36] presented a numerical model and experimentally validated a reduction of 40-50% in flexural strength due to fibre waviness. Bogetti *et al.* [37] showcased a simulation revealing a 40% decrease in unit cell strength with increasing waviness of [90/0/90] composite laminate under uniaxial compression. In Karami *et al.* [38] micromechanics model, the results predicted that the Young's modulus in the fibre direction would decrease rapidly with the increase of fibre waviness. Chun *et al.* [39] included a simulation and an experimental validation demonstrating that the strength of the composite material decreases while the ultimate strain increases with the increase of fibre waviness. Additionally, Potter *et al.* [40] demonstrated that aside from directly affecting composite properties, fibre waviness influenced the variability in fibre or polymer content of composites. For pultruded thermoplastic composites, Lapointe *et al.* [28] identified that 3 N tension prevented yarn damage during thermoplastic pultrusion of C/PEEK rods of commingled yarns. By comparing experiments having 0 N tension and 3 N tension, fibre damage was apparent during their 0 N tension experiment and resulted in an interruption of the pultrusion. The yarn damage was attributed to the misalignment of the carbon fibres in the commingled yarns, with the author suspecting that this misalignment was induced by thermoplastic fibre shrinkage. Further investigation is required to explore the effect of thermoplastic fibre shrinkage on fibre waviness formation during thermoplastic pultrusion.

The objective of this study is to demonstrate an effective pultrusion of DREF yarns at different tension levels and highlight the effect of shrinkage of continuous polymer fibres on composite

properties and pultrusion outcome. Glass/polyethylene terephthalate (G/PET) hybrid yarns were used as input materials. First, a comparative discussion on the input materials' CW and DREF structure and their reaction to heating was conducted. Pultrusion experiments were conducted to produce cylindrical profiles, employing the CW and DREF yarns as input materials with in-creel tension levels of 0N and 3N. The impact of tension on void content, sloughing, mechanical properties and fibre waviness was analysed for both CW and DREF yarn rods.

## 4.2 Experimental

### 4.2.1 Materials

The pultrudates were made from glass fibres (GF) as the reinforcement and PET thermoplastic polymer matrix. Two types of input material were used. The first type was a CW yarn (Concordia Fibers LLC). The second type was a friction spun yarn, a.k.a DREF (Filspec Inc). The PET polymer properties for DREF and CW yarns are shown in Table 4-1. The melting point ( $T_m$ ), glass transition ( $T_g$ ) and degradation temperatures were obtained by Dynamic Scanning Calorimetry (DSC) (MDSC 2000, TA instruments). Solid density was measured by Pycnometer (Accupyc 1340, Micrometrics). PET's solid density of 1.43 g/cm<sup>3</sup> in DREF yarns, was higher than the PET's 1.38 g/cm<sup>3</sup> density in CW yarns. This is attributed to the additives added to the DREF's PET to modify its colour to black. The melt density was measured by Pressure-Volume-Temperature (PVT) (pvT 100, ThermoHaake).

Table 4-1 PET polymer properties in CW and DREF yarns.

|                                         | CW yarn's PET | DREF yarn's PET |
|-----------------------------------------|---------------|-----------------|
| <b>Melting point (°C)</b>               | 254           | 257             |
| <b>Glass transition (°C)</b>            | 80            | 80              |
| <b>Degradation temperature (°C)</b>     | 295           | 295             |
| <b>Solid density (g/cm<sup>3</sup>)</b> | 1.38          | 1.43            |
| <b>Melt density (g/cm<sup>3</sup>)</b>  | 1.06          | 1.06            |

The lower left side of Figure 4-1 shows a magnified image of the longitudinal and cross-sectional illustration of both CW and DREF tows. PET fibres and GF were represented in black and white respectively. The CW tow sketch is an assembly of one PET tow and one GF tow. The CW illustration reveals the parallel placement of the GF and PET fibres, as well as the continuous nature of the PET fibres. The DREF tow sketch shows GF enclosed by the spun PET discontinuous fibres.

The CW yarns were composed of PET fibres (Laser+ HS Ti818) and E-GF (ECDE75, AGY). One CW yarn is an assembly of seven PET tows of 100 tex each and 20 E-GF tows of 66 tex each. E-GF density was of 2.54 g/cm<sup>3</sup>. The total linear density of one CW yarn was 2020 tex. It included 700 tex of PET and 1320 tex of E-GF. DREF yarns were composed of PET fibres (staple fibre, Huvis Corporation) and S2-GF (SCG75, AGY). A DREF tow was comprised of discontinuous PET fibres, amounting to 38 tex, friction spun around one S-GF tow of 66 tex. S-GF density was of 2.46 g/cm<sup>3</sup>. Eight tows were assembled without a twist, to shape one DREF yarn. The total linear density of one DREF yarn consisted of 832 tex, with 304 tex of PET fibres and 528 tex of S-GF.

#### **4.2.2 Pultrusion apparatus**

Figure 4-1 illustrates the schematic of the pultrusion system used for this study. The apparatus consisted of a creel, a guiding plate, a preheater, three heating dies, one cooling die, cooling fans, a load-cell, an encoder and a pulling system. At the initial stage of the pultrusion system was a creel housing bobbin of yarn. The creel allowed yarn tension adjustment during unwinding. The guiding plate was used to position the yarns for entry into the preheater and obtain a controlled arrangement. The preheater featured a metallic tube measuring 300 mm in length and 16.2 mm in diameter. A heating tape was wrapped around the metallic tube to provide heat. Heating of the pultrusion dies was carried out using heating cartridges represented by the red circles. The temperature was monitored by type J thermocouples, visually indicated by the green triangles in Figure 4-1. The pultrusion dies serve the purpose of shaping the composite structure by combining and impregnating the reinforcement fibres with the polymer matrix. All four dies had a 10° tapered cavity with a length of 31.75 mm followed by a cylindrical cavity with a length of 6.67 mm. The dies' cylindrical cavities had exit diameters of 5.61, 5.00, 4.90 and 4.76 mm respectively. The dies were separated by a 5 mm gap. This multi-die configuration was similar to previously published study that showcased good pultrudate impregnation quality [12]. The last die had a thin-walled tube with a length of 27.6 mm for cooling purposes, as detailed in [12]. The tube length was subjected to the airflow generated by the fans. A puller controller the pultrusion speed with an encoder monitoring the real-time speed.

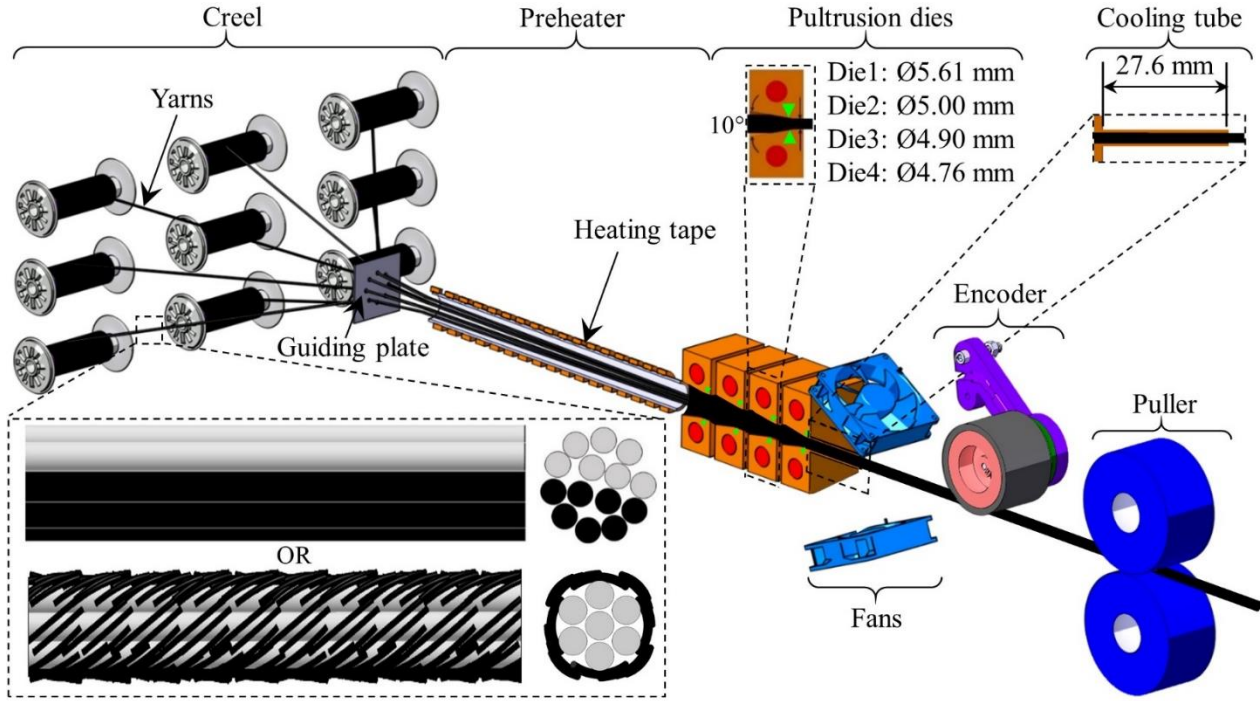


Figure 4-1 Schematic view of the thermoplastic pultrusion system used showing a transverse cross-section of the preheater and the pultrusion dies. Each component of the pultrusion system is labelled and identified accordingly. Red circles represent the heat cartridge location, and the green triangles represent the control thermocouple locations. On the lower left side is an illustration of the longitudinal and cross-section view of a both CW and DREF tows where PET and GF are represented in black and white respectively.

### 4.2.3 Pultrusion experiment

The pultrusion system shown in Figure 4-1 was used to manufacture five-meter-long GF/PET rods. Table 4-2 displays the sample name, material, tension, number of yarns, filling ratio ( $Rf_{die\ i}$ ) of each die and the theoretical fibre volume content ( $v_{f,th}$ ) for each experiment. The sample name consisted of the input material with the respective tension used. Two different tension adjustments were applied during the experiments. The first condition included the application of three N (3 N) tension to each bobbin, while the second involved no tension (0 N). The filling ratio was calculated for each pultrusion die and is expressed by the equation,

$$Rf_{die\ i} = \frac{(A_f + A_{m,i-1})}{A_{die\ i}} \quad i = 1, 2, 3, 4 \quad (4.1)$$

where  $A_f$  is the total GF cross-sectional area and  $A_{m,i-1}$  is the PET cross-sectional area located before the  $i$ th die.  $A_{die\ i}$  is the die's exit area calculated with the exit diameters of each die respectively.  $A_f$  was determined by,

$$A_f = \frac{N \times F_f}{1000 \cdot \rho_f} \quad (4.2)$$

where  $N$  is the number of input material yarns used in each pultrusion experiment.  $F_f$  is the linear density of the GF in tex, and  $\rho_f$  the solid density of the GF in g/cm<sup>3</sup>. Note that the cross-sectional area of the GF is constant throughout the pultrusion. The PET cross-sectional area at the first die entrance can be calculated with,

$$A_{m,0} = \frac{N \times F_m}{1000 \cdot \rho_m} \quad (4.3)$$

where  $F_m$  is the linear density of the PET in tex, and  $\rho_f$  the melt density of the PET in g/cm<sup>3</sup>. If the filling ratio  $Rf_{die\ i}$  is exceeding 100% and considering that the GF are impregnated at each die, the PET cross-sectional area entering the next die would be calculated with,

$$A_{m,i} = A_{die\ i-1} - A_f \quad i = 2, 3, 4 \quad (4.4)$$

The filling ratio should exceed 100% to insure consolidation during pultrusion. Table 4-2 indicates that the first die's filling ratio did not exceed 100%. Actually, the purpose of the first die was to ensure that all polymer fibres were reaching the processing temperature without significant polymer flow. To achieve comparable consolidation in all experiments, the number of yarns ( $N$ ) was adjusted to get similar filling ratios between CW and DREF experiments. The theoretical fibre volume content for the pultruded rod is calculated according to,

$$v_{f,th} = \frac{N \times A_f}{A_{die\ 4}} \quad (4.5)$$

For all pultrusion experiments, the speed was set at 35 mm/min. The preheater temperature was set to 240°C in order to bring the material closer to its processing temperature. The heating dies were set at 280°C. PET like other semi-crystalline polymer, crystallizes at temperature between  $T_g$  and  $T_m$  [74]. The cooling temperature was set between  $T_g$  and  $T_m$  at around 168°C to enhance the crystallization. Throughout the experiments, a data acquisition system (Labview) recorded the pultrusion parameters.

Table 4-2 Variable pultrusion parameters used for each experiment.

| Sample name    | Material | Tension | N  | $Rf_{die\ 1}$ | $Rf_{die\ 2}$ | $Rf_{die\ 3}$ | $Rf_{die\ 4}$ | $v_{f,th}$ |
|----------------|----------|---------|----|---------------|---------------|---------------|---------------|------------|
| <b>CW-3N</b>   | CW       | 3 N     | 18 | 84%           | 106%          | 104%          | 106%          | 58%        |
| <b>DREF-3N</b> | DREF     | 3N      | 42 | 81%           | 103%          | 104%          | 106%          | 56%        |
| <b>CW-0N</b>   | CW       | 0 N     | 18 | 84%           | 106%          | 104%          | 106%          | 58%        |
| <b>DREF-0N</b> | DREF     | 0 N     | 42 | 81%           | 103%          | 104%          | 106%          | 56%        |

#### 4.2.4 Characterisation

Microscopic images of the of the input materials cross-section were taken to reveal their structures. Input material yarns were enclosed in shrink tube (Polyolefin Plastic). The shrink tube was uniformly heated to induce tube shrinkage without PET polymer melting. The shrink tape covered yarn were embedded in epoxy resin (Epofix, Struers). Once the epoxy was cured, samples' cross-section was polished and analysed under a microscope (VHX 7000, Keyence). Input materials behaviour under heating was characterized by first securing one end of each yarn with polyimide tape (Kapton® Polyimide Plastic) in a metallic fixture. The yarns were then placed in an oven (F100HP, Pyradia, Brick Oven) at 240°C for one minute. A video of the yarns behaviour during heating was acquired.

During each pultrusion experiment, the pultrudate's temperature was acquired with a follow-on 36-gage type J wire thermocouple. The thermocouple was spliced to a yarn in the centre of the pultrudate's cross-section. Pultruded rods were inspected for surface imperfections. Samples were cut for microscopic observations at sloughing defects locations. The rods' diameter was measured using a micrometre (H-2780, Mitutoyo Digital). The defect-free diameter was measured at five locations where the rod appeared fully circular indicating that sloughing did not occur. To assess the sloughing defects, the actual diameter of the pultrudate was measured by performing 60 measurements along a 1-meter length of the pultruded rod. Twenty diameter measurements were acquired with 50 mm intervals. This measurement process was repeated three times, with each iteration involving a 45° rotation of the rod. These 60 measurements were acquired without paying attention to the surface finish, roundness, or presence of sloughing defects on the pultruded rods.

Impregnation was characterised by void content analysis and optical microscopy. Constituent content was measured per ASTM D3171-22 procedure G. The void content was calculated in accordance with the same standard where density measurements of samples were carried out following the ASTM D792. For microscopic observation, three samples of 30 mm were cut from

each experiment. The cylindrical samples were prepared according to the same procedure than the unconsolidated yarns, described above.

From each pultruded rod, a 50 mm sample was cut and scanned using an X-ray CT system (Zeiss, Xradia-520 versa). These samples were scanned and stitched at two positions overlapping each other, to create a 3D image of a composite rod measuring 10 mm in length. The CT scans were conducted with a voxel size of 6.5  $\mu\text{m}$  using a beam energy of 60 kV and 50 W in power. Subsequently, two sets of 1601 projections were obtained then stitched automatically with Scout-and-Scan Control System (Zeiss, v14.0), for the image reconstruction. The resulting stitched 3D image was comprised of 1850 slices perpendicular to the rod axis. Finally, for segmentation, 3D visualization, and quantitative analysis, reconstructed images were imported into an image processing software (Object Research Systems Inc. (ORS), Dragonfly software (version 2020.1). For manual segmentation of a specific yarn, 15 slices were chosen at an interval of 100 slices. Then using an interpolation operation tool within the software, the slices between the 15 selected ones were automatically filled to highlight the chosen yarn.

Two mechanical tests were conducted on the composite rods. To assess the elastic modulus of the composite manufactured by each experiment, three rods, each with a length of 150 mm, underwent tensile testing using grooved grips to prevent slipping. The composite rod was securely clamped within 20mm of the grips from both ends. Elongation was acquired using an extensometer (634.25F-25, MTS) with a gage length of 50mm. The tensile chord modulus of elasticity was determined by following the ASTM D3039. The strain range was measured at two points, one at 25% and another at 50% of the maximum strain. Shear strength was characterized following an adaptation of the ASTM D3914 [29]. Three coupons were tested per experimental conditions. Coupons had a length of 19 mm. Two grooves were cut on each side of the rod using a precision saw (Secotom-20, Struers). The distance between the grooves was 5mm. The coupons were loaded under compression (Insight, MTS). The maximum compression load was recorded and divided by the rod's longitudinal area between the grooves to obtain the in-plane shear strength.

## 4.3 Results and discussions

### 4.3.1 Input materials' structure

Figure 4-2(a) shows the microscopic image at 30-x magnification of a CW yarn with a black background. The image shows that the yarn had a flat structure with both GF and PET fibres being white. The CW input material was lacking consistent shape retention and tended to be deformed when touched. Figure 4-2(b) shows the microscopic image at 30-x magnification of a DREF yarn. The image shows an assembly of DREF tows with the GF in white and PET fibres in black. This input material was comprised of short PET fibres that had a soft texture. In some cases where friction levels were elevated during winding, the short fibres tended to be dislodged. However, the yarn maintained a consistent overall shape and exhibited resilience against deformation.

Figure 4-2(c) shows the microscopic image at 200-x magnification of a CW yarns cross-section contained in a shrink tube. The smaller circles represented the GF. The GF are of smaller size compared to those of PET fibres. It was observed that the PET fibres exhibited variations in size. The CW cross section showed unevenly blended bundles of PET fibres and agglomerations of GF. Figure 4-2(d) shows the microscopic image at 200-x magnification of a DREF yarns cross-section, also contained in a shrink tube. The GF are the densely packed particles forming eight agglomerations, while the PET fibres are dispersed around them. The DREF cross section exhibited uniform PET fibres distribution around the GF. The cross section of the CW represented 2020 tex of material, whereas the DREF represented 832 tex of material. However, Figure 4-2(c) and (d) show a yarn diameter of 1.30mm and 1.15mm for CW and DREF respectively, this suggested that DREF had a higher bulk factor than CW yarns. The bulk factor refers to the ratio of the volume occupied by the yarn in its uncompressed state to the volume it occupies when fully compacted. A high bulk factor in the yarns may pose challenges during the compression moulding of complexes structures with small corner radii [75]. However, during a linear process such as pultrusion, there is a limited impact of the high bulk ratio of the input material.

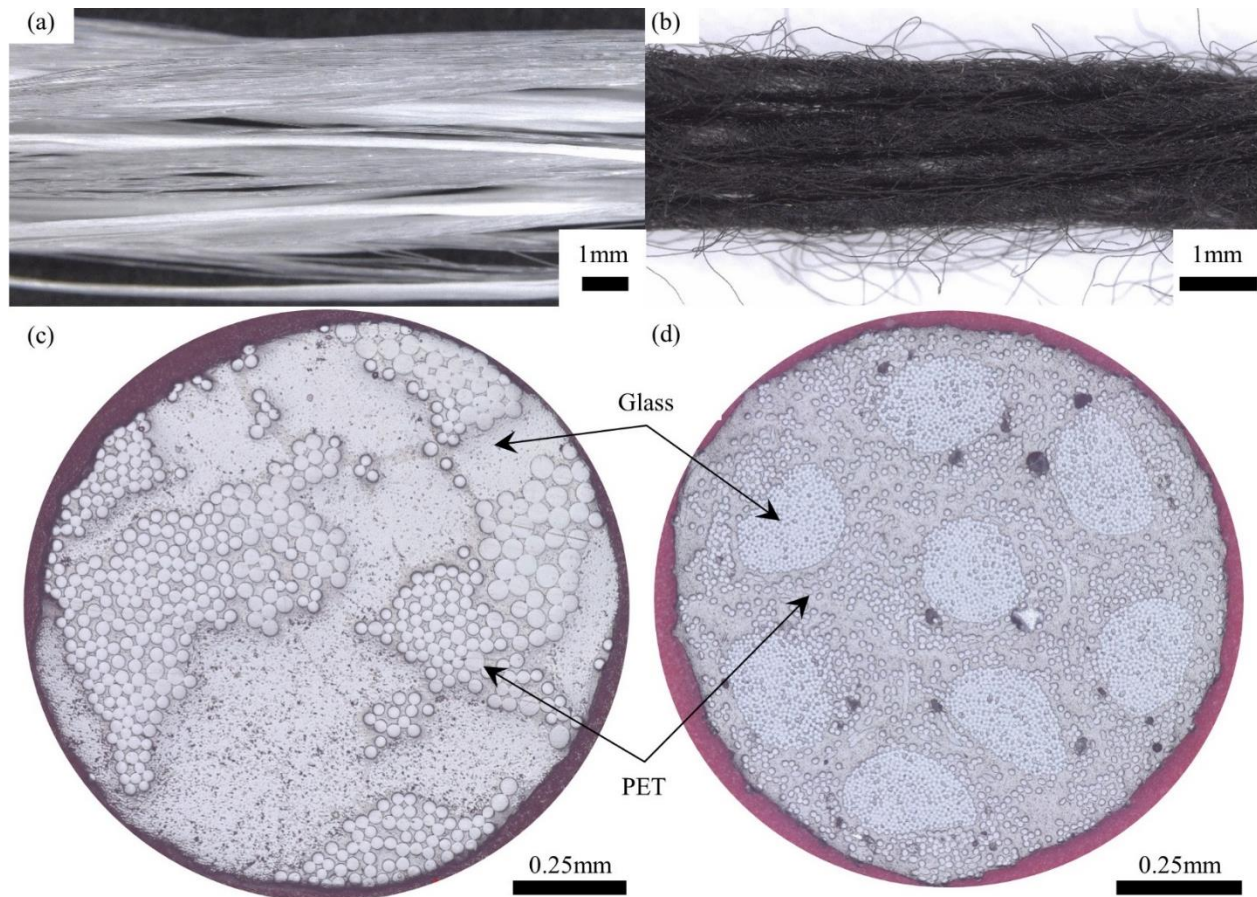


Figure 4-2 Input material yarns structures of (a) CW yarns (b) DREF yarns, and cross-sectional micrographs at a 200 magnification of (a) CW yarns (b) DREF yarns.

Figure 4-3 presents both input material yarns behaviour under heat and without tension. Figure 4-3(a) presents CW and DREF yarns placed side by side on a metallic fixture at room temperature. Figure 4-3(b) presents CW and DREF yarns placed side by side on a metallic fixture after being placed in an oven at 240°C for one minute. The temperature was set at 240°C to replicate the response of the yarns as they passed through the preheater without tension. A video of the yarns' behaviour under heat is available at <https://youtu.be/3UNLEjfZRUU>. As demonstrated in Figure 4-3(b), the heating the CW yarn resulted in an entanglement of the yarn structure. The long PET fibres shrunk before melting which resulted in dragging the reinforcement fibres and caused the waviness of the GF. On the other side, Figure 4-3(b) demonstrated that the DREF yarn kept its structure with GF staying straight after heating. This observation was also apparent during the experiments. In the preheater, when tension was not applied, CW yarns were entangled and entered

the dies all scattered. However, the DREF yarns retained their shape and behaved comparably in both experiments, with and without tension. Images of the CW and DREF yarn extracted from the preheater, illustrating the explanation above, are available in Annexe A.

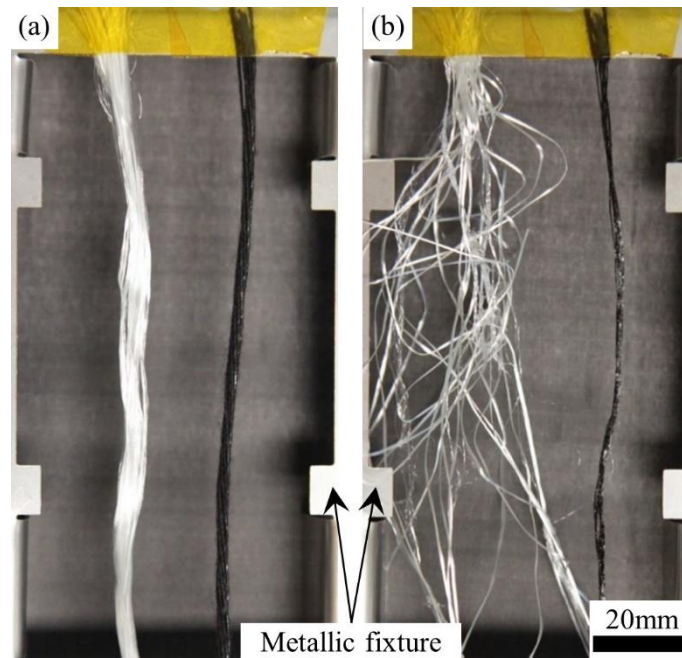


Figure 4-3 Un-tensioned CW and DREF yarns shape: (a) before heating, (b) after heating at 240°C for one minute.

### 4.3.2 Pultrusion experiment

Figure 4-4 displays a schematic of the transverse cross-section of the preheater and the pultrusion dies with the temperature profile recorded throughout the DREF-0N pultrusion experiment. This temperature profile was similar throughout all the pultrusion experiments. The green triangles represent the fixed thermocouples within the dies and on the cooling tube. The solid line represents the actual temperature measured. The dashed line corresponds to the set points of the preheater and dies and the desired cooling temperature of the cooling tube. The three black points on the graph represent the measured temperatures of the outside of the tube using glued-on thermocouples. The preheater was set to 240°C and the dies were set to 280°C. The cooling temperature at the exit of the thin-walled tube was 168°C. When entering the preheater, the recorded temperature gradually increased to reach the set point temperature. The temperature reached 240°C at the second half of

the preheater. At the end of the preheater, the temperature lowers due to the small gap between the dies and the preheater. When entering the first die, the temperature rises again. It reached the set point of 280°C at the middle of the first die. The temperature remained steady at 280°C in the first three dies. The small gaps did not affect the material temperature. At the middle of the last die the temperature increased to 290°C, then started dropping before reaching the start of the thin-walled tube. The fans directing airflow onto the die's surface could indirectly cause the temperature raise. This heat extraction by the airflow caused the heating cartridges to overheat the pultrudate at the Die 4 entrance. Then, due to the same airflow heat extraction, the pultrudate started to cool down before reaching the dies exit. The thermocouple placed on the surface of the thin-walled tube indicated temperatures comparable to those measured inside of the composite. The temperature decreased at the beginning of the tube from 234°C to the middle at 205°C and reached 168°C at the tubes exit. The measured temperature along the pultrusion experiments aligns with temperature expectations values.

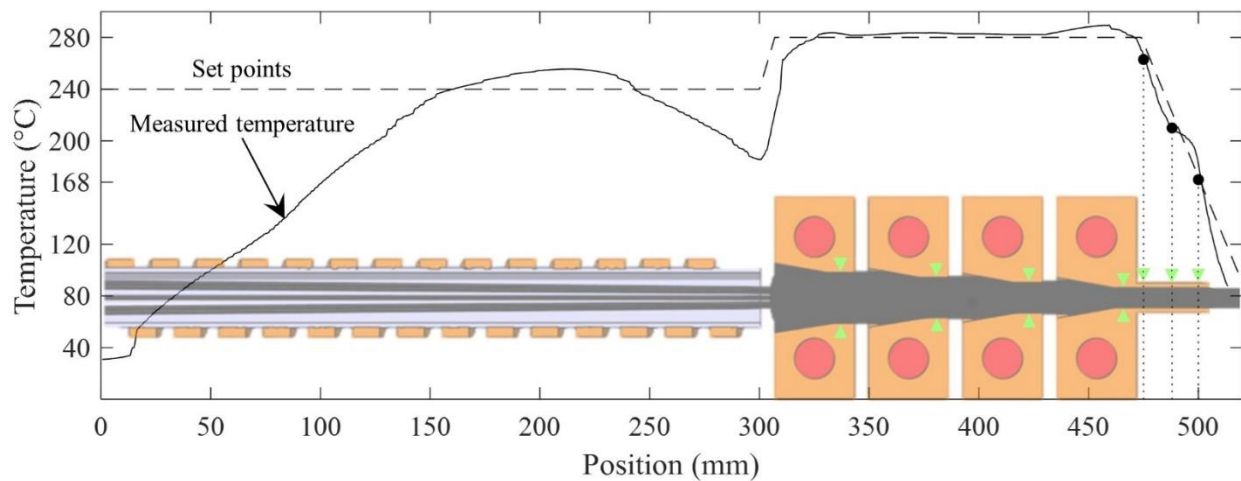


Figure 4-4 Temperature measurement and set points during DREF-0N pultrusion experiment. The solid line corresponds the actual reading temperature, while the dashed line corresponds to the set points.

### 4.3.3 Sloughing defects assessment

Figure 4-5 show photographs and cross-section microscopies of pultrudates where sloughing defects were apparent. Figure 4-5(a) and (b) displays the microscopic image at 20-x magnification of the CW-3N and DREF-3N rods. The pultrusion direction was displayed on the images. Both

images showed an aggregate of polymer-rich surface that was ruptured during the pultrusion with a diameter of 4.69 mm for the CW-3N rod and 4.57 mm for the DREF-3N rod. The aggregates had adhered to the cooling tube wall and subsequently de-bonded under the pressure from the incoming pultrudate. Before debonding, the aggregate ploughed into the pultrudate surface and caused a reduction in the pultrudate diameter and rough surface finish. The rough surface had visible GF with a smaller diameter of 4.18 mm for the CW-3N rod and 4.49 mm for the DREF-3N rod. Figure 4-5(c) and (d) presents the microscopic image of the cross-section of a CW-3N and DREF-3N sample that included a sloughing defect. The upper sections of the images in Figure 4-5(c) and (d) show the sloughing defect as a marginally detached fragment of the pultrudate. The sloughing defects reveal minimal GF presence as depicted in the enlarged view. Sloughing defects were observed in all experiments, but the exact extent or frequency of these events cannot be determined through this local observation alone. A diameter measurement distribution is provided below to quantify the intensity of the sloughing defects. In the literature, surface finish was characterized using the roughness parameter [12], [65]. However, in the current study, distributed diameter measurements were deemed to be more accurate for characterizing sloughing defects intensity.

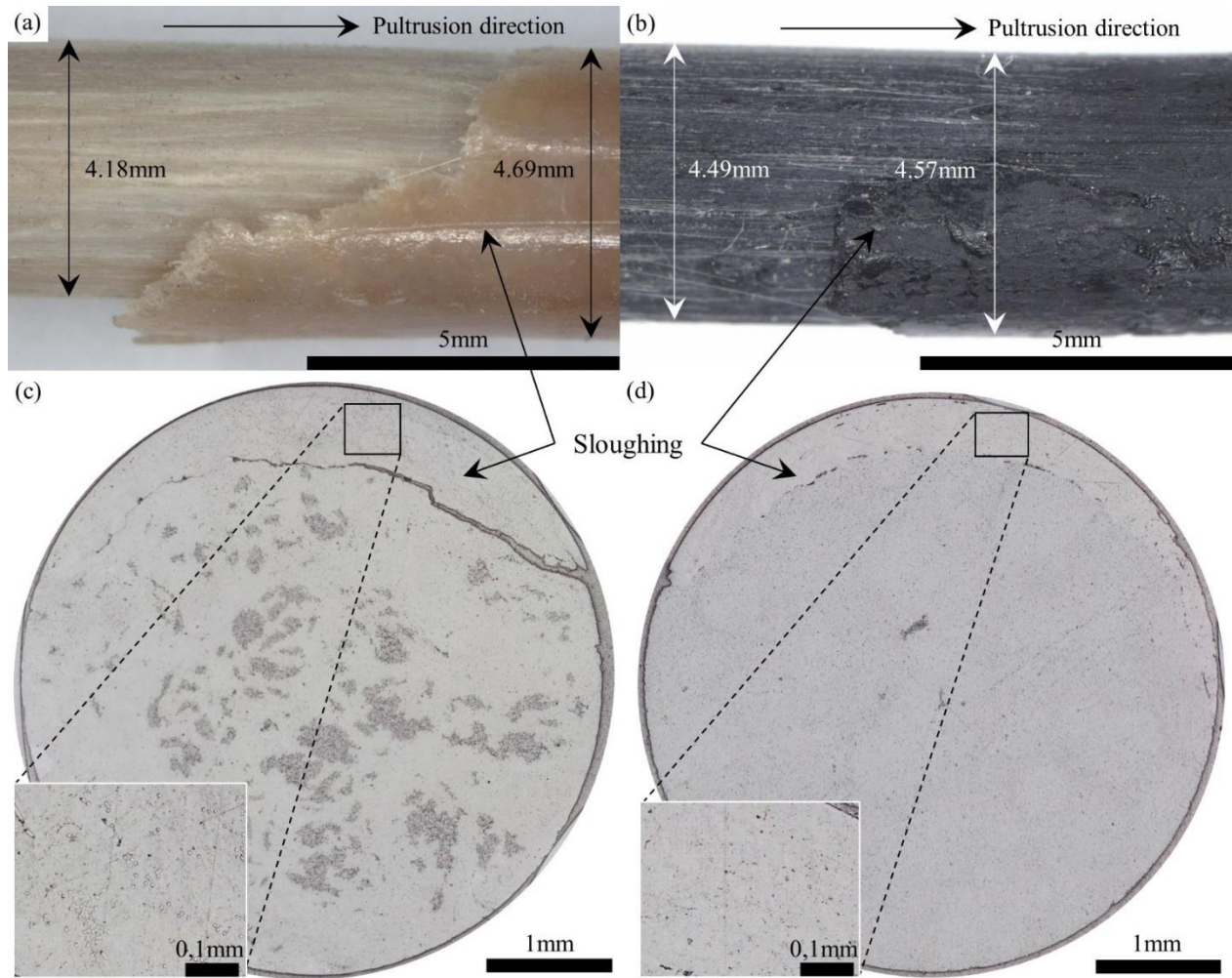


Figure 4-5 Top: Photographs at 20-x magnification of GF/PET rods at a section showing sloughing defects on the CW-3N (a) and DREF-3N (b) rods. Bottom: Cross-section image of the GF/PET rods at a section showing sloughing defects of CW-3N (c) of DREF-3N (d).

Figure 4-6 presents the corresponding diameter measurement the CW-3N, DREF-3N, CW-0N and DREF-0N. Displayed error bars for each experiment are the standard deviations on each measurement. The dashed line shown in the bar graph represented the 4.76 mm exit diameter of the Die 4. The average of the five diameter measures where sloughing defects did not occur are presented in grey. CW-3N and DREF-3N diameters without sloughing defects were  $4.59 \pm 0.03$  mm and  $4.64 \pm 0.01$  mm respectively. CW-0N and DREF-0N diameter without sloughing defects was  $4.66 \pm 0.02$  mm and  $4.64 \pm 0.01$  mm. These diameter measurements are relatively close to each other. The variation observed between the diameter without sloughing defects and the exit diameter of the Die 4 was caused by the pultrudate's shrinkage. The actual diameter along the 1-

m-long rod were presented in white. The rods made using yarn tension of 3N had the lowest average diameters with the largest standard deviation on the 60 equidistant measurements. CW-3N and DREF-3N actual diameters were  $4.53 \pm 0.19$  mm and  $4.54 \pm 0.17$  mm, respectively. The rods made using no yarn tension had average diameter closer to the 'Without Sloughing' diameters and with standard deviations almost half of the 3N tension experiments. CW-0N and DREF-0N actual diameters were  $4.60 \pm 0.09$  mm and  $4.63 \pm 0.09$  mm. The smaller standard deviations for CW-0N and DREF-0N indicates that when tension was not applied (0 N), the diameter was more consistent and exhibited less variation. Remarkably, for the DREF-0N, the actual diameter was the closest to its diameter without sloughing (with 0.02 mm difference). Indicating that DREF-0N had the least number of sloughing defects. This confirms visual observations during the experiment.

These observations revealed that tension influenced the diameter variation of the rods. Yarn tension induces yarn diameter compaction [28], thereby resulting in lowering transverse permeability of the fibres [19]. As a result, the polymer formed a layer around the bundle of the compacted fibres. Since a limited number of fibres are present in the polymer-rich layer, the adhesion force exceeded the strength of the composite, resulting in the detachment of the polymer-rich layer and the creation of sloughing defects. Additionally, the sloughing defects observed suggested that the cooling process might have been inadequate. While the focus of this study did not revolve around the pultrusion cooling process, improvement of the cooling should be investigated in future studies.

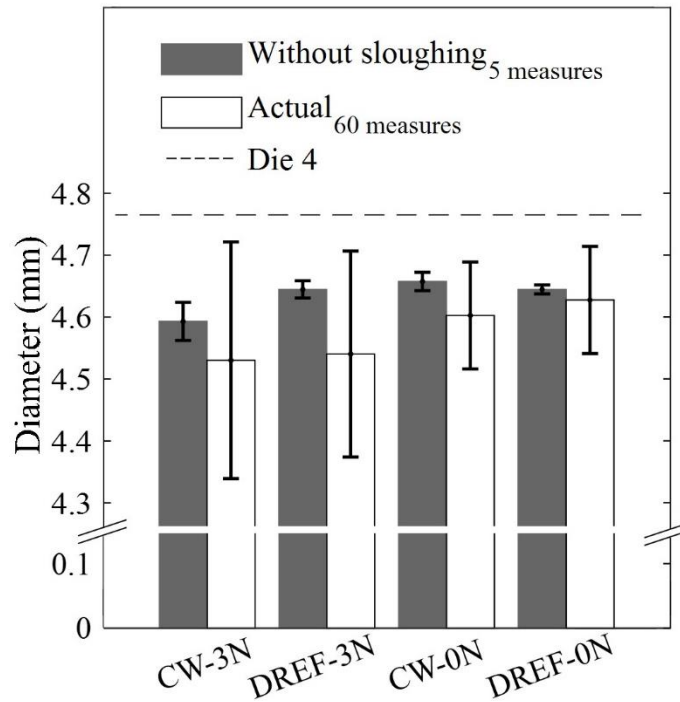


Figure 4-6 Average of the five diameter measurements on selected locations without sloughing and average of the 60 diameter measurements for CW-3N, DREF-3N, CW-0N and DREF-0N.

#### 4.3.4 Impregnation assessment

Figure 4-7(a) shows the fibre volume fraction of CW-3N, DREF-3N, CW-0N and DREF-0N. The theoretical fibre volume fraction  $v_{f,th}$  of 58% for CW-3N and CW-0N is shown with a dashed line. Whereas the theoretical fibre volume fraction  $v_{f,th}$  of 56% for DREF-3N and DREF-0N is shown with a dotted line. Displayed error bars for each experiment are the standard deviations on three measurements. Fibre volume fraction was measured at  $62.0 \pm 0.3 \%$ ,  $59.4 \pm 0.5 \%$ ,  $59.9 \pm 1.1 \%$  and  $56.0 \pm 0.7 \%$  for CW-3N, DREF-3N, CW-0N and DREF-0N respectively. For CW-3N and DREF-3N, the average measured  $v_f$  values were noticeably higher than the  $v_{f,th}$  with a 4.0 % and 3.4 % discrepancy respectively. The effect of sloughing is likely to have been the cause for this high discrepancy. For CW-0N and DREF-0N, the average measured  $v_f$  values exhibited a 1.9 % and 0.0 % discrepancy on the  $v_{f,th}$  respectively. In the previous section, CW-3N and DREF-3N demonstrated to have more sloughing defects than CW-0N and DREF-0N. It was explained that yarn tension induced fibre tow compaction, which lead to the presence of a polymer-rich area around the bundle of the compacted fibres and thereby resulted in polymer adhesion. The adhered

polymer's ploughing reduces the effective diameter of the Die 4. Consequently, for the same amount of fibres, less polymer was able to exit the Die 4. This resulted in a higher  $v_f$  for CW-3N and DREF-3N. It is important to note that the  $v_f$  was measured by pyrolysis and calculated using the actual density of the pultrudate. The actual density was measured through liquid immersion. For CW-0N, the standard deviation on the average measured  $v_f$  was the highest (1.1 %). Indicating that on samples that were characterized, there was a significant variation in the GF volume content. Fibre waviness may have contributed to the variation in the GF volume content of CW-0N. The evaluation of waviness is detailed in Section 3.5. For DREF-0N, average measured  $v_f$  showed low standard deviation and an insignificant discrepancy on the  $v_{f,th}$ . Demonstrating that for DREF-0N the average measured  $v_f$  was consistent with the theoretical predictions. This correlates with the previous section that indicated negligible number of sloughing defects. From the results above, it is likely that higher  $v_f$  is caused by yarn tension that increased the sloughing defects occurrence in CW-3N and DREF-3N. In the case of CW-0N, the higher  $v_f$  was attributed to fibre waviness.

Figure 4-7(b) shows the void content of CW-3N, DREF-3N, CW-0N and DREF-0N. The void content  $v_v$  was measured at  $4.2 \pm 0.4$  %,  $2.9 \pm 1.0$  %,  $2.2 \pm 1.0$  % and  $1.1 \pm 0.1$  % for CW-3N, DREF-3N, CW-0N and DREF-0N respectively. These measurements indicated that when 3 N tension was applied  $v_v$  was higher than when no tension was applied (0 N). This observation, also observed by Laponite *et al.* [28], is explained by the lower permeability of the compacted fibre bundle due to the tension. For the non-tensioned pultrusions, CW-0N  $v_v$  average was higher and had a larger standard deviation than DREF-0N. The greater  $v_v$  value of the CW-0N was attributed to the structure of the input material by itself. As explained in Section 4.3.1, CW yarns displayed unevenly blended bundles of PET fibres and greater agglomerations of GF. Large bundles of GF require more time for impregnation. DREF yarns have PET fibres well dispersed around the constant size GF bundles therefore requiring shorter time for impregnation as well as a similar impregnation time between GF bundles. In summary, both larger size of GF agglomerations in hybrid yarns, as well as higher yarn tension, increased the void content of the pultrudates.

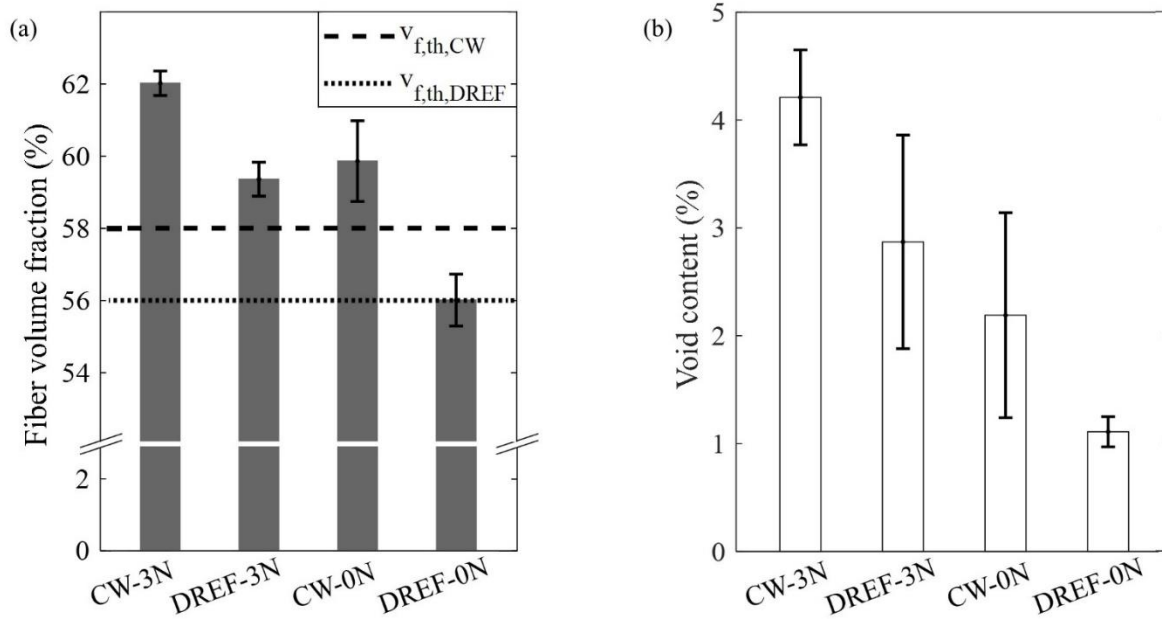


Figure 4-7(a) Fibre volume fraction of CW-3N, DREF-3N, CW-0N and DREF-0N. (b) Void content of CW-3N, DREF-3N, CW-0N and DREF-0N.

Figure 4-8(a) and (b) presents microscopic images of the cross-section of samples from CW-3N and DREF-3N. Black areas represent voids. The enlarged views show a bundle of un-impregnated fibres. Through the comparison Figure 4-8 (a) and (b), it was clear that the agglomerations of un-impregnated fibres were noticeably larger in CW-3N than of DREF-3N. This was directly linked to the better distribution of GF in the DREF yarns as shown in Figure 4-2. Figure 4-8(c) presents the microscopic image of the cross-section of a sample from CW-0N. This cross-section still presented black areas scattered around the image. Figure 4-8(d) presents the microscopic image of the cross-section of a sample from DREF-0N. This image shows an insignificant number of black areas. The cross-section appears to be very well impregnated. Indicating that DREF yarns showed notable improvement in impregnation without tension. These images directly align with the  $v_v$  measurement shown in Figure 4-7. For Figure 4-8(a) and (b), where a 3N tension was applied for both experiments, the porosity was remarkable. Therefore, once again validating that having extensive tension can lead to the compaction of the fibre bundles and reduces permeability and thus limits impregnation. However, in the case of the CW yarns, tension absence did not enhance impregnation. This was due to the CW yarn structure, precisely the size of GF agglomeration. Conversely, the structure of DREF yarns enabled achieving high impregnation without any application of tension during pultrusion.

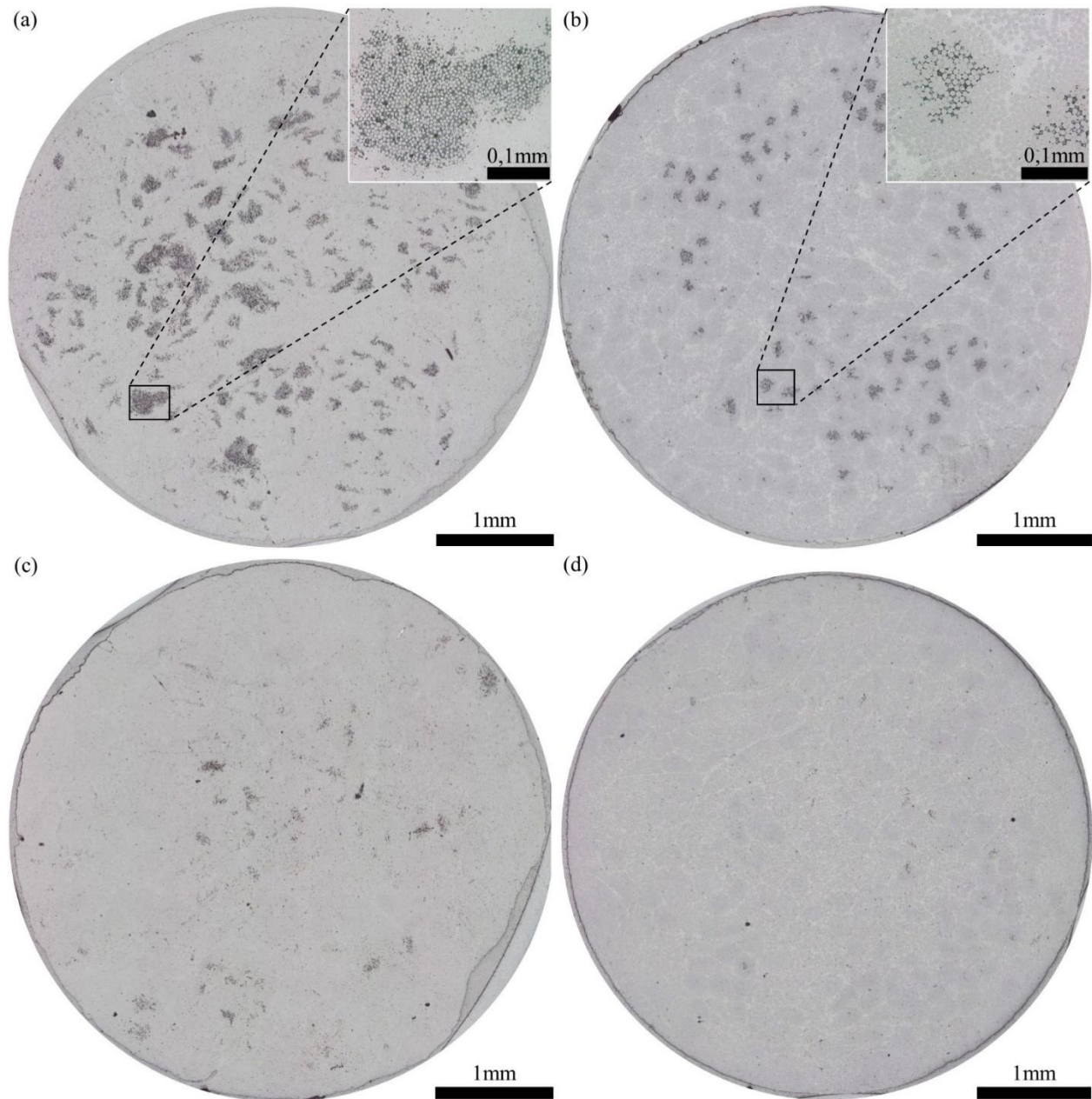


Figure 4-8 Cross-sectional images of the pultruded G/PET rods for (a) CW-3N (b) DREF-3N (c) CW-0N, and (d) DREF-0N.

#### 4.3.5 Waviness assessment

Figure 4-9 presents CT-scan images of 10 mm-long GF/PET pultruded rods with four-coloured glass fibre bundle (blue, purple, yellow and red). Figure 4-9(a) shows a CT scan image of a CW-0N pultrudate showing four wavy GF tows. Figure 4-9(b) shows a CT scan image of a DREF-0N

pultrudate showing four straight GF tows. CT scan images clearly illustrated that when no tension was applied during the pultrusion of CW yarns, fibre waviness occurred. Conversely, when no tension was applied to DREF yarns, fibre tows retained their form and remained straight throughout the pultrusion process. This contradicts the findings of Volk *et al.* [41], who observed apparent straightening of commingled in the straight section of the die. CT-scan image of CW-3N and DREF-3N pultrudates were not evaluated considering that yarn tension straightened the yarns in both cases. The findings presented here support that the absence of yarn tension during composite manufacturing induces fibre waviness in input materials consisting of continuous thermoplastic fibres, like CW yarns. Whereas, for DREF yarns consisting of discontinuous thermoplastic fibres, fibre waviness remains absent even without the application of tension.

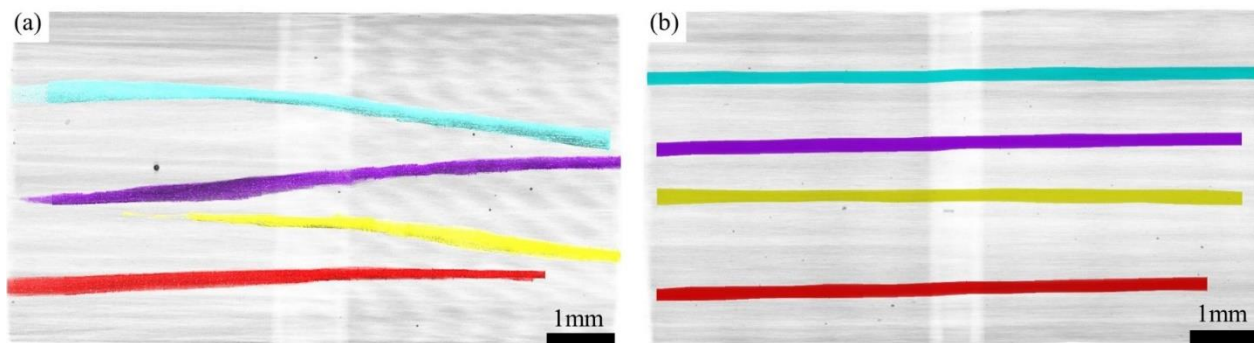


Figure 4-9 CT scan images of the pultruded GF/PET rods for (a) CW-0N showing four wavy fibre tows (b) DREF-0N showing four straight fibre tows.

### 4.3.6 Composites mechanical properties

Figure 4-10 presents the elastic modulus of the GF/PET rods. Theoretical elastic modulus for CW experiments ( $E_{CW}$ ) is presented in a dashed line, with a value of 43.27 GPa. Theoretical elastic modulus for DREF experiments ( $E_{DREF}$ ) is presented in a dotted line, with a value of 51.98 GPa. These theoretical values were calculated using  $v_{f,th}$ , E-GF (69 GPa) and S-GF (86 GPa) elastic modulus from material's technical datasheets. The elastic modulus of PET was (3.6 GPa) [76]. The bar graph displays the measured elastic modulus ( $E$ ) in GPa of samples from CW-3N, DREF-3N, CW-0N and DREF-0N experiments. Displayed error bars for each experiment are the standard deviations on three samples and are small compared to the modulus differences between experiments. Comparative analysis between CW and DREF experiments was challenging due to the difference in GF types. CW-3N and CW-0N presented close comparable values of  $48.87 \pm 0.19$

GPa for CW-3N, and a  $48.39 \pm 0.09$  GPa for CW-0N. The similar modulus values are explained by the similar volume fraction of the CW pultrudates (see Figure 4-7(b)). DREF-3N elastic modulus was  $57.26 \pm 0.17$  GPa. The DREF-0N modulus was  $53.88 \pm 0.09$  GPa. In this case, the 3.35 GPa lower value for DREF-0N is explained by the significantly lower fibre volume fraction of the DREF-0N pultrudates (see Figure 4-7(a)). It is also noticeable that the measured modulus value for DREF-0N was the closest to the theoretical value with a 1.9 GPa difference.

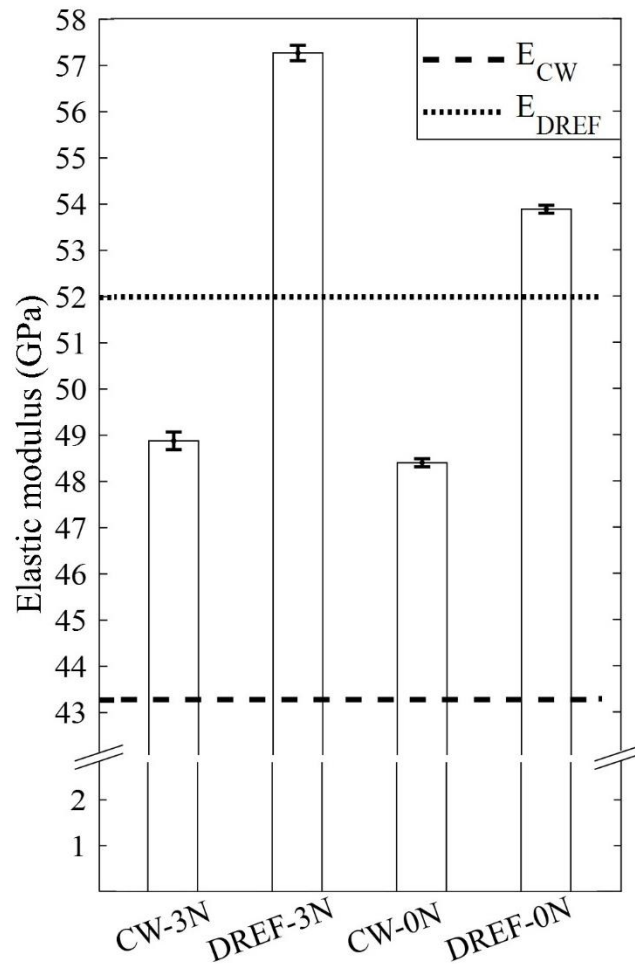


Figure 4-10 Elastic modulus of GF/PET rods for CW-3N, DREF-3N, CW-0N and DREF-0N. Dashed and dotted lines show the theoretical elastic modulus.

Figure 4-11 presents the in-plane shear strength of the GF/PET rods. Displayed error bars for each experiment are the standard deviations on three samples. The in-plane shear strength was  $26.93 \pm$

4.51 MPa,  $55.47 \pm 2.39$  MPa,  $55.17 \pm 15.17$  MPa and  $119.35 \pm 16.89$  MPa for CW-3N, DREF-3N, CW-0N and DREF-0N. The bar graph demonstrates a noticeable increase in the in-plane shear strength across the four experiments, progressing from CW-3N to DREF-0N. This was consistent with the decrease in void volume fraction shown in Figure 4-7. Supporting the literature's argument that in-plane shear strength decreases with increasing porosity [28], [73]. DREF-0N demonstrated a  $119.35 \pm 16.89$  MPa for in-plane shear strength, significantly exceeding those obtained in the other experiments. DREF yarns has the potential of producing superior performances in pultruded composite materials despite the absence of tension during pultrusion.

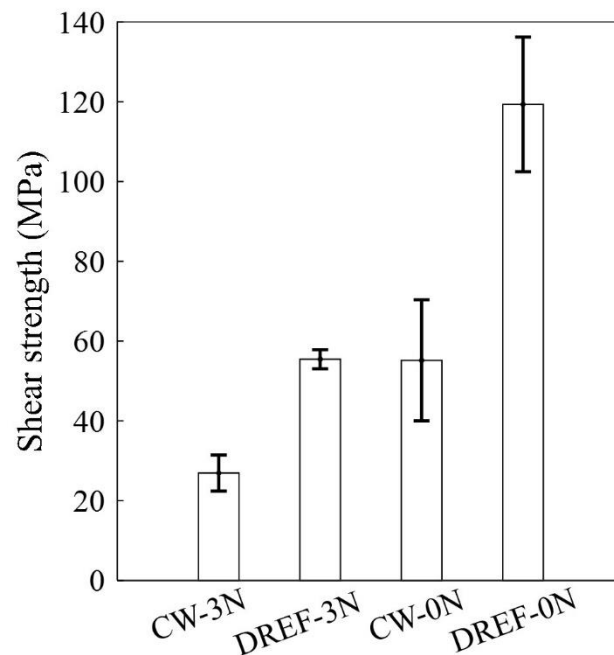


Figure 4-11 Average measured in-plane shear strength of GF/PET rods for CW-3N, DREF-3N, CW-0N and DREF-0N.

#### 4.4 Conclusion

This study introduced the feasibility of thermoplastic pultrusion with DREF yarns, and proved more effective results when tension was not applied to the input yarns. Composites rods were produced using a multi-die pultrusion system, where CW and DREF yarns were evaluated as input materials. DREF yarns' structure assessment highlighted a notable uniformity in the distribution of polymer and glass fibre. Additionally, heat response assessment confirmed that continuous

polymer fibres, similar to those present in CW yarns, have a tendency to shrink inducing the entanglement of GF. CW and DREF yarns were used to study the effect of yarn tension on sloughing, porosity, and mechanical properties of pultrudates. Yarn tension indicated increased the occurrence of sloughing defects. Due to the compacted GF and the reduced permeability, a weak layer of polymer was formed on the exterior of the pultrudate that tore away from the pultruded yarn after adhesion to the cooling die wall. DREF-0N had the least number of sloughing defects, with the actual diameter ( $4.63 \pm 0.09$  mm) being the closest to its diameter without sloughing ( $4.65 \pm 0.01$  mm). Additionally, yarn tension was shown to impact fibre volume fraction due to the presence of sloughing. In DREF-0N,  $v_f$  was measured at  $56.0 \pm 0.7$  % and precisely matched the theoretical value ( $v_{f,th}$ ) of 56%. While evaluating porosity, the size of GF agglomeration and yarn tension indicated to be directly linked to void content of the pultrudate. DREF-0N exhibited the lowest  $v_v$  at  $1.1 \pm 0.1$  %. Microscopic image of the cross-section of DREF-0N showed an insignificant amount of porosity and indicated that the structure of DREF yarns enabled achieving high impregnation without any application of tension during pultrusion. Moreover, CT scan images of pultrudates confirmed that the absence of yarn tension induces fibre waviness in input materials with continuous thermoplastic fibres like CW yarns, while fibre waviness was absent in DREF yarns with discontinuous thermoplastic fibres, even without tension. DREF-0N surpassed the other experiments with an in-plane-shear strength of  $119.35 \pm 16.89$  MPa. In conclusion, DREF yarns present promising potential for use in thermoplastic pultrusion of high-performance composites across various fields, particularly due to their ability to limit fibre waviness even without tension. Experiments will soon be conducted at higher pultrusion speed and with an improved cooling process to reduce sloughing defect occurrence. Nonetheless, this study paves the way for a broader utilization of DREF yarns as a pultrusion input material for commercial applications.

### **Credit authorship contribution statement**

M. El-Jakl: Conceptualization, Methodology, Investigation, Analysis, Writing – original draft.

L. Laberge Laberge: Conceptualization, Writing - Review & Editing, Supervision, Project administration, Funding acquisition.

### **Declaration of Competing Interest**

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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## CHAPITRE 5     **Article 2: Demonstration of the multi-die pultrusion's effect on the micro-impregnation of thermoplastic composites through compression moulding replication.**

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### **Abstract**

Thermoplastic pultrusion presents a major challenge in achieving a high degree of impregnation because of the high viscosity of thermoplastic polymers. Recently, studies have adopted multi-die process for thermoplastic pultrusion showing an improvement in the impregnation quality of the manufactured composites. Several assumptions to explain the improved impregnation quality were discussed without validation. This study presents an experimental approach that imitates the consolidation cycles of the multi-die pultrusion by compression moulding to determine the factors contributing to the enhanced impregnation of using multiple dies. Using a low-cost thermoplastic hybrid yarn, called DREF, this study explores the consolidation time and tool force of a MD process compared to three SD processes. The multi-die process effectively increased the consolidation time by 260% and reduced the compressive tool load by 38%, compared to a long single die with similar taper angle. Moreover, the MD process exhibited the highest degree of micro-impregnation with a  $R_{imp}$  of  $96 \pm 4$  %. The replicated SD-5° exhibited the highest compression load, which lead to a reduced fibre permeability and a  $R_{imp}$  of  $73 \pm 12$  %. The MD consolidated plates had the highest interlaminar shear strength of  $42.3 \pm 7.8$  MPa, suggesting that the multi-die process could result in manufacturing composites with higher mechanical properties.

### **5.1 Introduction**

Pultrusion is a well-established manufacturing method for continuous fibre-reinforced composites with thermoset matrices [2], [11]. This process enables the production of composite with a constant cross-section, capable of attaining high fibre content [3]. Pultruded profiles have demonstrated their capacity to meet cost, mechanical and physical standards with their production steadily rising and expanding into diverse market sectors [17]. However, the advancement of thermoplastic pultrusion remains limited [2], [18]. Thermoplastic composites offer numerous advantages, including higher toughness and damage tolerance [2] as well as the ability to be reshaped,

reprocessed and welded [11]. Above all, thermoplastic composite manufacturing enables cleaner processing [77]. In non-reactive thermoplastic pultrusion, input yarns containing polymer and reinforcement filaments are wound onto bobbins then placed in a creel. These yarns are then pulled through a hot die where the thermoplastic polymer undergoes melting and impregnates the reinforcement filaments. Upon entering the pultrusion die, the material is referred to as a pultrudate. The composite then transitions into a cooling die where solidification occurs, resulting in the formation of the pultruded composite product [12]. The process speed is controlled by a pulling system located downstream the pultrusion line, ensuring precise control over the production rate.

In non-reactive thermoplastic pultrusion, impregnation challenges arise from the considerably higher melt viscosity (10 orders of magnitude) compared to thermoset polymers [19]. To address this issue, various input materials have been developed. Input materials imply various material configurations, such as reinforcement fibres, polymer pellets, prepregs, and hybrid yarns. When the input materials are hybrid yarns, they are composed of polymer premixed with reinforcement fibres. The concept behind this premixing is to minimize the polymer flow distance, and therefore compensating for the high viscosity of thermoplastic [28]. Input materials include prepregs, towpregs, commingled, co-wound and DREF yarns. Prepregs, towpregs, commingled and CW yarns have been extensively used in thermoplastic pultrusion [2]. However, DREF yarns have few applications in thermoplastic pultrusion. DREF yarns consist of continuous reinforcement filaments core onto which discontinuous polymer filaments are spun [33]. DREF yarns provide advantages like consistent and even distribution of polymer filaments around the reinforcement filaments. In addition, DREF input materials offer improved handling capabilities when compared to other input material yarns.

In recent years, researchers have developed the multi-die process for thermoplastic pultrusion demonstrating promising outcomes [12], [28], [29], [56], [66], [78]. The multi-die thermoplastic pultrusion consist of utilizing a set of successive dies rather than a single one. This process induces a cyclic consolidation pressure applied to the pultrudate moving through the dies. Oswald *et al.* [29] reported flax/PLA cylindrical rod manufacturing using multi-die pultrusion. The void content decreased by 62% when using two successive dies instead of only one. The rods pultruded with the multi-die process had a 4.5% void content. It should be noted that the die length for the single die process was equal to each of the dies used in multi-die process. The results obtained showed that the multi-die process improved impregnation while decreasing the pulling force. Lapointe *et al.*

[28] reported carbon/PEEK rod manufacturing using multi-die thermoplastic pultrusion process. It was observed that the 4-die system contributed to a 4.7% reduction in void content compared to a single-die pultrusion. Additionally, multi-die experiments indicated a decrease of 240 N in pulling forces. Eichenhofer *et al.* [49] also developed a multi-die pultrusion system for use in additive manufacturing. By adapting the multi-die pultrusion process, a reduction of over 80% in the final void content of the pultrudate was achieved. Several assumptions were discussed regarding the enhanced impregnation. One proposal indicated that the multi-die process helped in attaining the polymer melt temperature and divided the polymer backflow into multiple streams, thereby reducing pressure in each die [29]. Another perspective suggested that the duration under pressure was prolonged with the multi-die process, while leaving other manufacturing parameters unchanged [28]. Other researchers considered the deconsolidation to be the underlying cause for the multi-die process advantages [45], [49]. Lessard *et al.* [45] proposed a modelling approach to explore the deconsolidation mechanism and its effect on the microstructure of the composite. The findings indicated that the benefits from deconsolidation cycles included enhancing the permeability of wetted fibres and facilitating polymer flow towards dry regions. In summary, several theories were explored regarding the advantages of the multi-die approach, with some attempts to model such phenomena. Nonetheless, to the authors' understanding, additional investigation is necessary to fully understand the enhanced impregnation when using a sequence of short dies.

A critical component of the thermoplastic pultrusion is the die design. A typical thermoplastic pultrusion die included a tapered segment and a straight segment having a constant exit area. The die filling ratio is determined by the ratio of the cross-sectional area of the input material to the exit area of the die [12], [42]. During pultrusion process, the material area needs be slightly greater than the die exit area to achieve a filling ratio exceeding 100%. The excess melted polymer creates a backflow in the tapered segment of the die [43]. The thermoplastic resin backflow towards the die's entrance generates an impregnation pressure [26], [43]. Therefore, the tapered section's length and angle are important factors to consider for the generation of impregnation pressure. Then, when the pultrudate reaches the final straight segment of the die, the impregnation pressure was supposed to be instantly released [43], [44]. It was found that the degree of impregnation in the pultrudate depended on the geometry of the die as well as fibre agglomeration size [20], [43]. Studies indicated that a longer taper length or increased time of residency, may improve impregnation for high speed

pultrusion [20], [43], [72]. However, it was suggested that this configuration could create problems at the entrance of the die and potentially harming the pultrudate due to the high shear rate flow in the opposite direction of pulling, leading to excessive pulling forces [72]. In summary, it is yet to be demonstrated if longer tapers in pultrusion dies can produce high quality composites.

Cyclic pressure can also be used in compression moulding; some refer to it as debulking [79]. The purpose of debulking is to remove trapped air from the part prior to the final high-pressure consolidation [80]. In some cases, the use of compaction cycles resulted in improved textile nesting due to the rearrangement of fibre bundles [81]. Betanzos *et al.* [82] reported the effects of pressure cycling on the mechanical properties of compression-moulded PLA/GF composites. The authors indicated that cyclic pressure effectively removed significant pockets of trapped air during compression. Their study showed that the flexural strength of the composite was 36% higher than that produced with the static pressure, suggesting a higher degree of impregnation. It was proposed that cyclic pressure has a potential to alter the fibre network permeability through the relaxation and pressure reapplication. In the case of a static pressure, the fibre network permeability was low due to the tightly packed fibre bundles. Thus, during the pressure release phase of the cyclic pressure, the permeability was restored within the fibre network, therefore enhancing the transverse micro-impregnation.

Polymer flow during thermoplastic composite manufacturing can be characterized in two types: the macro-impregnation-flow and the micro-impregnation flow. The macro-impregnation flow happens between tows which are zones with high permeability. The macro-impregnation flow can circulate along the tow direction. The micro-impregnation flow happens within the tows, i.e., within densely packed fibres which have lower permeability. The micro-impregnation flow mainly circulates between the fibres towards the centre of the tow. The impregnation quality is commonly evaluated through the characterization of macro-pores located between tows and micro-pores located intra-tows [83]. Macro-pores appear in mainly resin-rich areas and qualify the macro-impregnation. Alternatively, micro-pores appear in fibre-rich areas, defined by fibre agglomeration limits, indicating the micro-impregnation. During pultrusion, the backflow is a macro-flow between tows towards the die's entrance. As explained above, the backflow generates pressure that drives a micro-flow towards tow centres. Several studies evaluated the degree of impregnation in thermoplastic pultrusion through the assessment of micro-pores [26], [42], [43]. Recently, Kazano *et al.* [46] analysed cross-sectional microscopies using an image processing software (GIMP 2).

They characterized the micro-impregnation of elliptical fibre agglomerations. The micro-impregnation ratio was obtained by dividing the impregnated area by the total cross-section area of the fibre tow. Additionally, the degree of impregnation of composites directly impacts their mechanical properties. It has been demonstrated that low fibre tow impregnation decreases the mechanical performance of composites [70]. Properties such as inter-laminar shear strength (ILSS), exhibit a marked correlation with the void volume fraction ( $v_v$ ). In this relationship, ILSS typically decreases with an increase in  $v_v$  [70], [84]. These findings suggested that characterisation of micro-pores and conducting ILSS testing provides an effective approach for evaluating the impregnation quality of composites.

The objective of this study is to compare the micro-impregnation quality of composites using the multi-die and single-die processes. Compression moulding experiments were conducted to replicate the MD and SD pultrusion process. The impact of the multi-die process on processing parameters, micro-impregnation and interlaminar shear strength was assessed.

## 5.2 Experimental

### 5.2.1 Materials

DREF yarns consisted of S2-GF (SCG75, AGY) and PET fibres (staple fibre, Huvis Corporation). One DREF tow, was composed of a GF core of 66 tex covered by discontinuous spun PET fibres of 25 tex. The DREF yarns were made by friction spinning (Filspec Inc.). Table 5-1 displays properties of the PET fibres. The melting point, glass transition and degradation temperatures were measured by DSC (MDSC 2000, TA instruments). Solid density was determined using a Pycnometer (Accupyc 1340, Micrometrics) and the melt density was measured by PVT analysis (pvT 100, ThermoHaake) between 270°C and 285°C. GF density was of 2.46 g/cm<sup>3</sup> as per AGY's technical datasheets. To form a single DREF yarn, eight tows were grouped together without twist. Resulting in 728 tex as the linear density of one DREF yarn, with 528 tex of GF and 200 tex of PET. The DREF yarn's nominal fibre volume fraction ( $v_{n,f}$ ) was at 60%.

Table 5-1 PET material properties

| Parameter                      | Unit              | Value |
|--------------------------------|-------------------|-------|
| <b>Melting point</b>           | °C                | 257   |
| <b>Glass transition</b>        | °C                | 80    |
| <b>Degradation temperature</b> | °C                | 295   |
| <b>Solid density</b>           | g/cm <sup>3</sup> | 1.43  |
| <b>Melt density</b>            | g/cm <sup>3</sup> | 1.06  |

### 5.2.2 Simulation of the pultrusion by a compression moulding tool

Figure 5-1(a) illustrates a pultrusion die with two sections of the pultrudates represented in a compression mould. The pultrusion die is presented in orange and the compression mould is presented in blue. The fibre tows are presented in white and the resin in grey. A small section of the pultrudate in a pultrusion die corresponds to half the composite in a mould compressing unidirectional fibres. It is assumed that the polymer backflow obtained during pultrusion is equivalent to the polymer flow from the centre of the compression mould towards the edges. Figure 5-1(b) presents a cross-sectional illustration of one fibre tow subjected to micro-impregnation flow towards its centre. The image shows the tow area ( $A_{tow}$ ), the void area ( $A_{void}$ ) and the micro-impregnation ratio ( $R_{imp}$ ). The image displays a A-A section, which provides a longitudinal view of the fibre tow. The A-A section reveals the impregnated and un-impregnated fibres. Figure 5-1(c) presents a sectional view along the axis of a thermoplastic pultrusion die. The section illustrates the tapered angle  $\alpha$ , the effective length ( $L_{eff}$ ) and the effective diameter ( $\emptyset_{eff}$ ) in a pultrusion die. The  $L_{eff}$  start point corresponds to the point where incoming material intersects the die taper at entrance, as described by Lessard *et al.* [45]. The  $L_{eff}$  end point occurs when the material ceases to be subject to pressure and is considered to take at the end of the tapered section of the die, or slightly after. The  $\emptyset_{eff}$  is the diameter of the circle when the material intersects with the die taper at entrance.

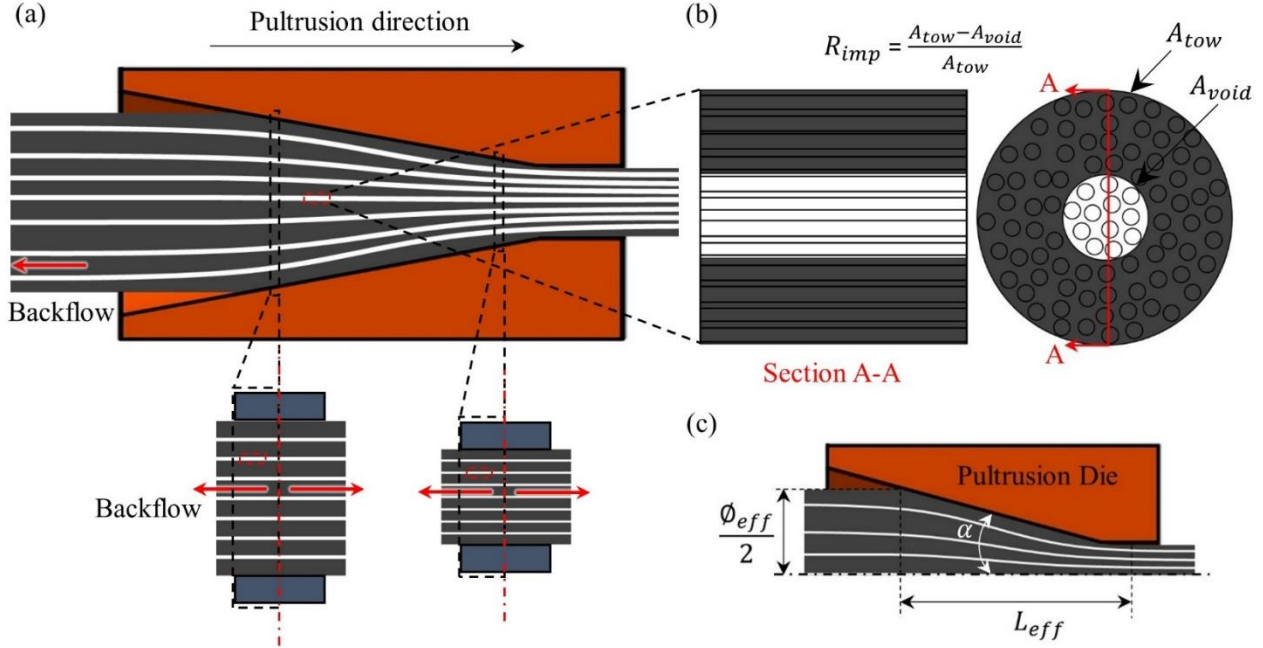


Figure 5-1(a) Illustration of a pultrusion die with sections of the pultrudate corresponding to half the composite in a compression mould. Red arrows indicate the polymer backflow during pultrusion and compression moulding experiment. (b) Axial and longitudinal (A-A) section view of a fibre tow and its micro-impregnation. (c) Illustration of a pultrusion die showing the angle  $\alpha$  of the tapered sections and pultrudate intersecting the die wall with dimensions of  $L_{eff}$  and  $\phi_{eff}$ .

For replicating a pultrusion by a compression moulding experiment, the theoretical fibre volume fraction ( $v_{f,th}$ ) of the final manufactured composite needs to be equivalent. However, due to the cross-sectional area difference of a pultrusion die and a compression mould, a conversion factor  $\delta$  must be calculated by the equation,

$$\delta = A_{mould} / A_{exit,die i} \quad (5.1)$$

where  $A_{mould}$  and  $A_{exit,die i}$  are the cross-sectional area of the compression mould and the  $i$ th pultrusion die's exit area respectively. The theoretical fibre volume content for the pultrudate is calculated according to,

$$v_{f,th,die i} = N_{Y,die} \times A_f / A_{exit,die i} \quad (5.2)$$

where  $N_{Y,die}$  is the number of yarns used during a pultrusion experiment and  $A_f$  is the GF cross sectional-area and is determined by,

$$A_f = \frac{F_f}{\rho_f} \quad (5.3)$$

where  $F_f$  is the linear density of the GF (Tex) and  $\rho_f$  is the solid density of the GF. The theoretical fibre volume content for the moulded composite ( $v_{f,th,mould}$ ) is calculated according to,

$$v_{f,th,mould} = N_{Y,mould} \times A_f / A_{mould} \quad (5.4)$$

where  $N_{Y,mould}$  is the number of yarns used during a compression moulding experiment. By equating (5.2) and (5.4),  $N_{Y,mould}$  is then calculated according to,

$$N_{Y,mould} = \delta \times N_{Y,die} \quad (5.5)$$

### 5.2.3 Pultrusion experiments to be replicated

The top part of Figure 5-2(a) presents a transverse cross-section of a multi-die (MD) pultrusion apparatus to be replicated by a compression moulding experiment. This MD configuration was employed in a recently published article that showcased good impregnation quality of the composite [12]. The heated dies are represented in orange. The pultrusion process had four dies, each separated by a 5 mm gap. All pultrusion dies had a conical tapered section of 31.75 mm and a cylindrical straight section of 6.35 mm. The cone taper angle was 5°. The last die had a thin-walled tube for cooling purposes, as detailed in [12]. The pultrusion dies had exit diameters ( $\phi_{exit,die i}$ ) of 5.13, 5.00, 4.90 and 4.76 mm respectively. The bottom part of Figure 5-2(a) displays a graphical representation of the evolution of the pultrusion area ( $mm^2$ ), i.e., the cross-sectional area of the die, on the left vertical axis and the corresponding compression moulding cavity thickness ( $mm$ ) on the right vertical axis, with respect to time ( $s$ ). The gap area and cavity thickness were considered to be equal to the next die entrance area and corresponding cavity thickness. Figure 5-2(b) displays a graphical representation of the evolution of the pultrusion area ( $mm^2$ ) and the corresponding compression moulding cavity thickness ( $mm$ ) with respect to time ( $s$ ) of all single-die (SD) experiments.

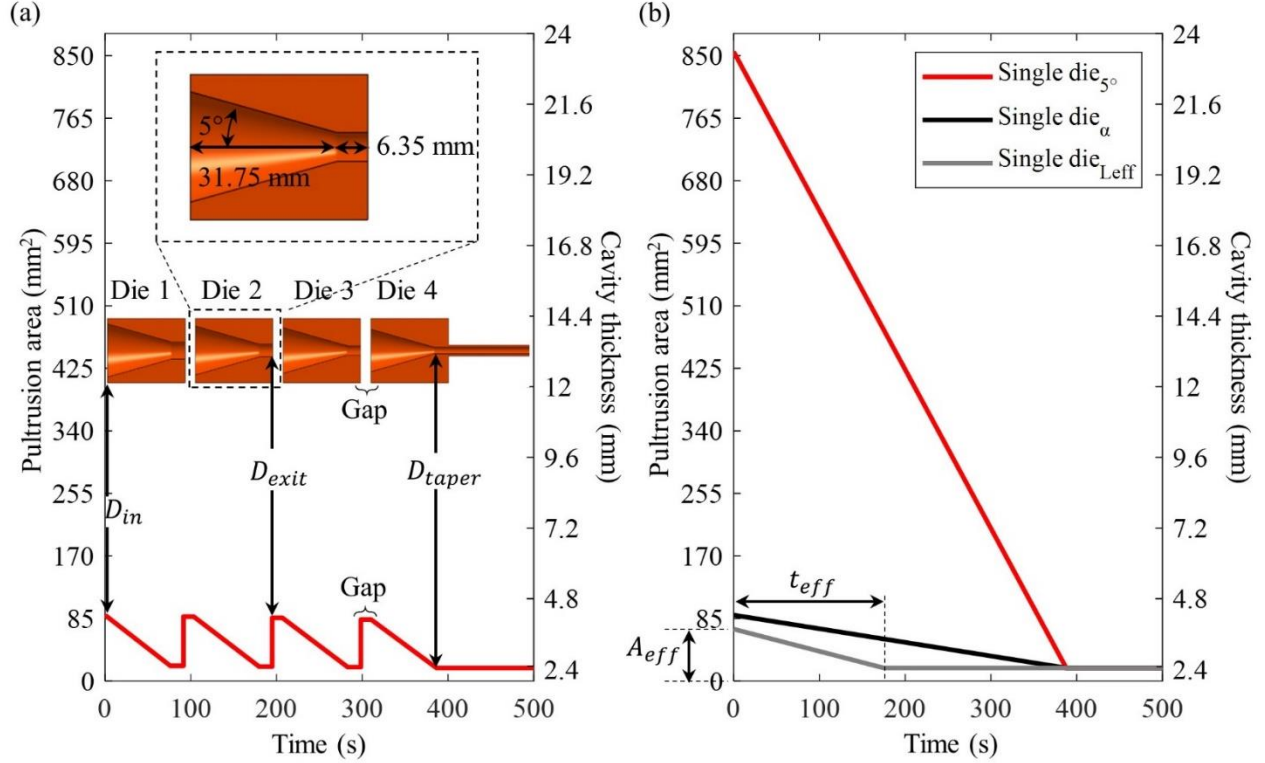


Figure 5-2(a) Top: Transverse cross-section of a multi-die pultrusion apparatus to be replicated by a compression moulding experiment. Bottom: Evolution of the pultrusion area and the corresponding compression moulding cavity thickness with respect of time for the replicated MD experiment. (b) Evolution of the pultrusion area and the corresponding compression moulding cavity thickness with respect of time for all replicated SD experiments.

The die's entrance diameter ( $\varphi_{in,die i}$ ) was calculated according to the relation,

$$\varphi_{in,die i} = \varphi_{exit,die i} + 2 L \tan \alpha \quad i = 1,2,3,4 \quad (5.6)$$

where  $L$  was the length of each die and  $\alpha$  was the taper angle.  $i$  corresponded to the sequence in which the dies were arranged. Given the conical shape of the pultrusion dies the exit area ( $A_{exit,die i}$ ) and entrance area ( $A_{in,die i}$ ) are calculated with  $\varphi_{exit,die i}$  and  $\varphi_{in,die i}$  of each die respectively.

The MD pultrusion experiment consisted of using 46 DREF yarns to achieve a  $v_{f,th}$  of 60%. With this number of yarns, a filling ratio ( $Rf$ ) of 104% was calculated in Die 4 according to,

$$Rf = \frac{N_{Y,die} (A_f + A_m)}{A_{exit,die 4}} \quad (5.7)$$

where  $A_f$  and  $A_m$  are the GF and PET cross-sectional area respectively.  $A_m$  was determined by,

$$A_m = \frac{F_m}{\rho_m} \quad (5.8)$$

where  $F_m$  are the linear density of the PET and  $\rho_m$  is the melt density of the PET. The compression moulding exit thickness  $c_{exit}$  was selected so that the moulded composite was equal to the radius of the Die 4 with an  $\varphi_{exit,die 4}$  of 4,76 mm. That thickness was deemed acceptable for short beam shear testing per ASTM D2344. Consequently,  $A_{mould}$  was calculated by multiplying  $c_{exit}$  of 2.4 mm to the width of the compression mould  $l_{mold}$  of 40 mm. By applying equation (1),  $\delta$  was equal to 5.35. Knowing  $N_{Y,die} = 46$  yarns, to achieve equivalent  $v_{f,th}$ ,  $N_{Y,mould}$  was calculated per equation (5) and equalled 246 DREF yarns.

Subsequently,  $A_{exit,die i}$  for all dies' area was converted in a compression moulding exit thickness  $c_{exit,i}$  according to,

$$c_{exit,i} = \frac{\delta A_{exit,die i}}{l_{mold}} \quad (5.9)$$

The pultrusion die circular entrance area was converted in a compression moulding entrance thickness  $c_{in i}$  using,

$$c_{in i} = c_{exit i} - \frac{A_{exit,i} - A_{in,i}}{l_{mold}} \quad (5.10)$$

Table 5-2 lists the position in mm along the length of the die systems, the time where the pultrudate reaches these positions for a speed of 25 mm/min, the area of the pultrusion dies and the corresponding compression moulding thickness computed using equations (9) and (10). The parameters are displayed for each of the pultrusion dies' geometric features. The features correspond to the die entrances ( $D_{in}$ ), taper exits ( $D_{taper}$ ), straight section exits ( $D_{out}$ ) and the GAP.

Table 5-2 Features of the MD and SD systems with their corresponding position, time, pultrusion area and compression moulding cavity thickness.

| Features      | Position<br>(mm) | Time<br>(s) | Pultrusion Area<br>(mm <sup>2</sup> ) | Cavity thickness<br>(mm) |
|---------------|------------------|-------------|---------------------------------------|--------------------------|
| MD            |                  |             |                                       |                          |
| $D_{in} 1$    | 0.0              | 0.0         | 89.7                                  | 4.5                      |
| $D_{taper} 1$ | 31.8             | 76.2        | 20.7                                  | 2.8                      |
| $D_{exit} 1$  | 38.1             | 91.4        | 20.7                                  | 2.8                      |
| GAP           | 38.1             | 91.4        | 87.6                                  | 4.3                      |
| $D_{in} 2$    | 43.1             | 103.4       | 87.6                                  | 4.3                      |
| $D_{taper} 2$ | 74.9             | 179.6       | 19.7                                  | 2.6                      |
| $D_{exit} 2$  | 81.2             | 194.9       | 19.7                                  | 2.6                      |
| GAP           | 81.2             | 194.9       | 85.9                                  | 4.2                      |
| $D_{in} 3$    | 86.2             | 206.9       | 85.9                                  | 4.2                      |
| $D_{taper} 3$ | 118.0            | 283.1       | 18.9                                  | 2.5                      |
| $D_{exit} 3$  | 124.3            | 298.3       | 18.9                                  | 2.5                      |
| GAP           | 124.3            | 298.3       | 83.6                                  | 4.0                      |
| $D_{in} 4$    | 129.3            | 310.3       | 83.6                                  | 4.0                      |
| $D_{taper} 4$ | 161.1            | 386.5       | 17.8                                  | 2.4                      |
| $D_{exit} 4$  | 208.3            | 500         | 17.8                                  | 2.4                      |
| SD-5°         |                  |             |                                       |                          |
| $D_{in}$      | 0.0              | 0.0         | 855.3                                 | 23.3                     |
| $D_{taper}$   | 161.1            | 386.5       | 17.8                                  | 2.4                      |
| $D_{exit}$    | 208.3            | 500         | 17.8                                  | 2.4                      |
| SD- $\alpha$  |                  |             |                                       |                          |
| $D_{in}$      | 0.0              | 0.0         | 89.7                                  | 4.2                      |
| $D_{taper}$   | 161.1            | 386.5       | 17.8                                  | 2.4                      |
| $D_{exit}$    | 208.3            | 500         | 17.8                                  | 2.4                      |
| SD- $L_{eff}$ |                  |             |                                       |                          |
| $D_{in}$      | 0.0              | 0.0         | 70.6                                  | 3.7                      |
| $D_{taper}$   | 76.3             | 183         | 17.8                                  | 2.4                      |
| $D_{exit}$    | 208.3            | 500         | 17.8                                  | 2.4                      |

Three SD experiments were designed for the comparison with the MD experiment. The first SD was a long die with a 5° taper angle. This single die's exit diameter and length were conceptualized considering the same exit diameter and the total length of the MD process. Thus,  $\varphi_{exit,SD-5^\circ} = \varphi_{exit,die 4} = 4.8$  mm,  $L = 161.1$  mm and  $\alpha = 5^\circ$ . The SD-5° entrance diameter,  $\varphi_{in,SD-5^\circ}$ , was determined to be 33 mm using equation (5.6). The replicated cavity thicknesses were 23.3 mm at entrance and 2.4 mm at exit.

The second SD was a long die with a small taper angle. This single die's entrance and exit diameter were conceptualized considering the same entrance and exit diameter of the MD process. This die's length was also equal to the total length of the MD process ( $L = 161.1$  mm). Accordingly,  $\varphi_{in,SD-\alpha} = \varphi_{in,1} = 10.7$  mm and  $\varphi_{exit,SD-\alpha} = \varphi_{exit,die\ 4} = 4.8$  mm. Equation (5.9) and (5.10) were then executed to transition from pultrusion to a starting compression cavity thickness of 4.2 mm then reducing to 2.4 mm. The equivalent die taper angle  $\alpha$  computed using equation (5.6), was  $\alpha = 1.05^\circ$ .

The third SD experiment was conceptualized using the summation of the effective lengths of each die of the multi-die pultrusion. The exit cavity thickness was again  $c_{exit,SD-L_{eff}} = 2.4$  mm. During the MD compression moulding experiment, the material was assumed to be in contact with the mould when the load reached 120N. It was characterised to be at 3.7 mm. Thus, the starting compression cavity thickness ( $c_{in,SD-L_{eff}}$ ) was 3.7 mm reducing to a  $c_{exit,SD-L_{eff}}$  of 2.4 mm. Given that process was evaluated over a time span, an effective duration ( $t_{eff}$ ) was identified for each die. This duration was defined as the period from the initial application of 120N until the release of the 120N load from the material. The four  $t_{eff}$  measured from each die of the MD replication were added to obtain a  $t_{eff}$  of 183 seconds. The conditions for the SD- $L_{eff}$  experiment corresponded to the following pultrusion die geometry.  $\varphi_{in,SD-L_{eff}} = 9.5$  mm,  $\varphi_{exit,SD-L_{eff}} = 4.8$  mm,  $L_{eff} = 76.3$  mm and  $\alpha = 1.8^\circ$ .

## 5.2.4 Compression moulding experiments

Figure 5-3 presents the different stages of the compression moulding process. The moulds, tensioning fixtures and winding equipment have been featured in previous work [85]. A video of the compression moulding experiment representing the MD process is available at <https://youtu.be/vJCyBYggRt8>. Figure 5-3(a) shows DREF yarns installed on a tensioning fixture. DREF yarns were wound using a digitally controlled winding machine that spread the yarns equally across the desired width. The yarn was tied to one of the rods and the assembly was then rotated to the required number of turns to achieve the correct yarn count. Figure 5-3(b) shows the open-ended heated mould mounted on an electrodynamic test system (Acumen 12, MTS). The test fixture was placed in the heated mould which was then closed without putting pressure on the yarns for five minutes to simulate the preheater found on the pultrusion line. Figure 5-3(c) shows the DREF yarns

being compressed according to a predefined process. The position and displacement rate of the mould were precisely controlled. The force applied to the mould was also recorded throughout the process using the 12 kN rated load cell (662.30H-04, MTS). The fixtures had water-cooling channels to insulate the load cell. To accelerate the cooling of the sample, a fan was installed on the backside of the Acumen and was turned on at the end of the experiment (at  $t = 387$  s). The machine was also equipped with an optical extensometer tracking two white lines on each half of the mould (as shown in Figure 5-3(c)). It was used to accurately track the movement of the mould. Figure 5-3(d) shows a consolidated composite ready for characterization. To prevent the composite from sticking to the moulds, a layer of polyimide tape (Kapton® Polyimide Plastic) was applied to the surfaces of the mould before each experiment. For both the MD and SD configurations, the process was repeated four times. Data from the compression moulding experiments were employed and analysed to compare the applied compression loads (kN), the mould cavity thickness (mm) and consolidation duration (s).

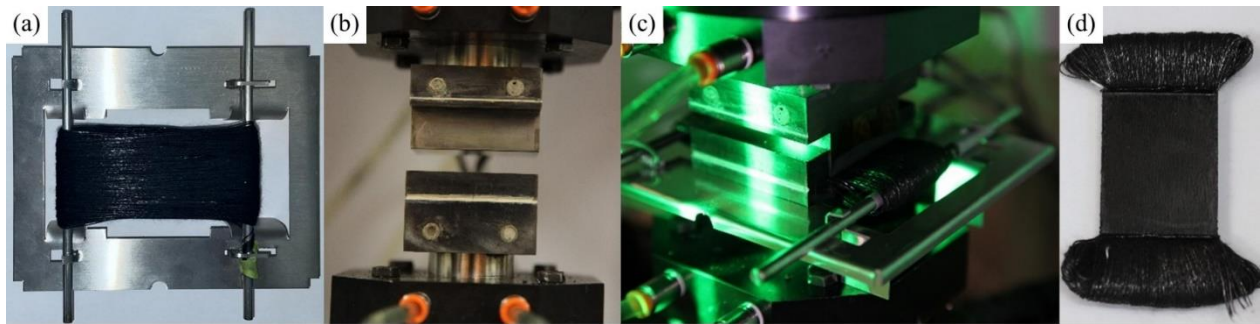


Figure 5-3 Images of the compression moulding process. a) Unidirectional DREF yarns wound on a test fixture. b) Compression moulding tool assembled onto the MTS Acumen. c) Test fixture assembled to conduct consolidation experiment. d) Consolidated sample

### 5.2.5 Characterisation

Impregnation was characterised by optical microscopy. Coupons of approximately 14 mm in length and 4.5 mm in width were cut from the moulded plates with an advanced precision cutting machine (Secotom-20, Struers). These coupons were cut at the middle part of the moulded plates. All coupons were then dried for moisture removal. The coupons were embedded in epoxy resin (Epofix, Struers) then polished and analysed under a microscope (VHX 7000, Keyence). One coupon was selected from each experiment for impregnation characterisation, for a total of four coupons per routine. Images were then analysed using an image processing suite (ImageJ). Six

fibre tows were selected from each coupon for image analysis, resulting in 24 fibre tows analysis for each process. The selection process was detailed in Annexe B. Using ImageJ's ROI (region of interest) manager tool, tow and void area statistics were obtained.  $R_{imp}$  was assessed for each tow. ILSS was characterised following the standard ASTM D2344. Coupons of approximately 14.5 mm in length and 4.8 mm in width were cut from the moulded plates. Coupons were then dried for moisture removal. Four coupons were selected from each experiment, for a total of 16 coupons per routine. Coupons were then tested by a universal testing machine (1362, MTS) equipped with a 5 kN load cell (2525-805, MTS).

### 5.3 Results and discussions

Thermoplastic composite plates were manufactured by compression moulding for both MD and SD processes. There was no fibre damage or visible fibre deviation during the cyclic moulding process. Excess polymer was found on the sides of the mould, indicating excess polymer flow replicating pultrusion backflow. Figure 5-4 presents the variation of the compression load (kN) and cavity thickness (mm) of the moulding processes over time for MD (a), SD-5° (c), SD- $\alpha$  (e) and SD- $L_{eff}$  (g). The multi die process reached  $5.2 \pm 1.5$  kN, while the SD-5° process exhibited the highest consolidation load in the range of  $8 \pm 1.4$  kN. The SD- $\alpha$  process exhibited the lowest consolidation load at  $2.7 \pm 1.2$  kN, while the SD- $L_{eff}$  process reached  $5.8 \pm 0.1$  kN. Lower compaction load means that the reinforcement fibres are less compacted and more permeable to resin flow. The low compaction load observed in the SD- $\alpha$  process was a result of the low compaction rate attributed to the low angle of 1.05° compared to the 5° in the MD replicated experiment. Additionally, the close-up views in Figure 5-4b)-d)-f)-g) shows that the consolidation forces exhibited a decline but does not reach zero once the replicated pultrusion straight section was reached. This was mainly caused by the elastic nature of the reinforcement. The GF are compacted together and exert a reaction force onto the moulds. In the multi-die process, when the compression was removed, the elastic energy accumulated in the compacted fibres was released since the composite was no longer constrained, leading to deconsolidation. The presence of gaps promoted the deconsolidation of the composite. The deconsolidation led to the early contact of the composite to the moulding surface in the next die. This phenomenon was not possible in the SD process.

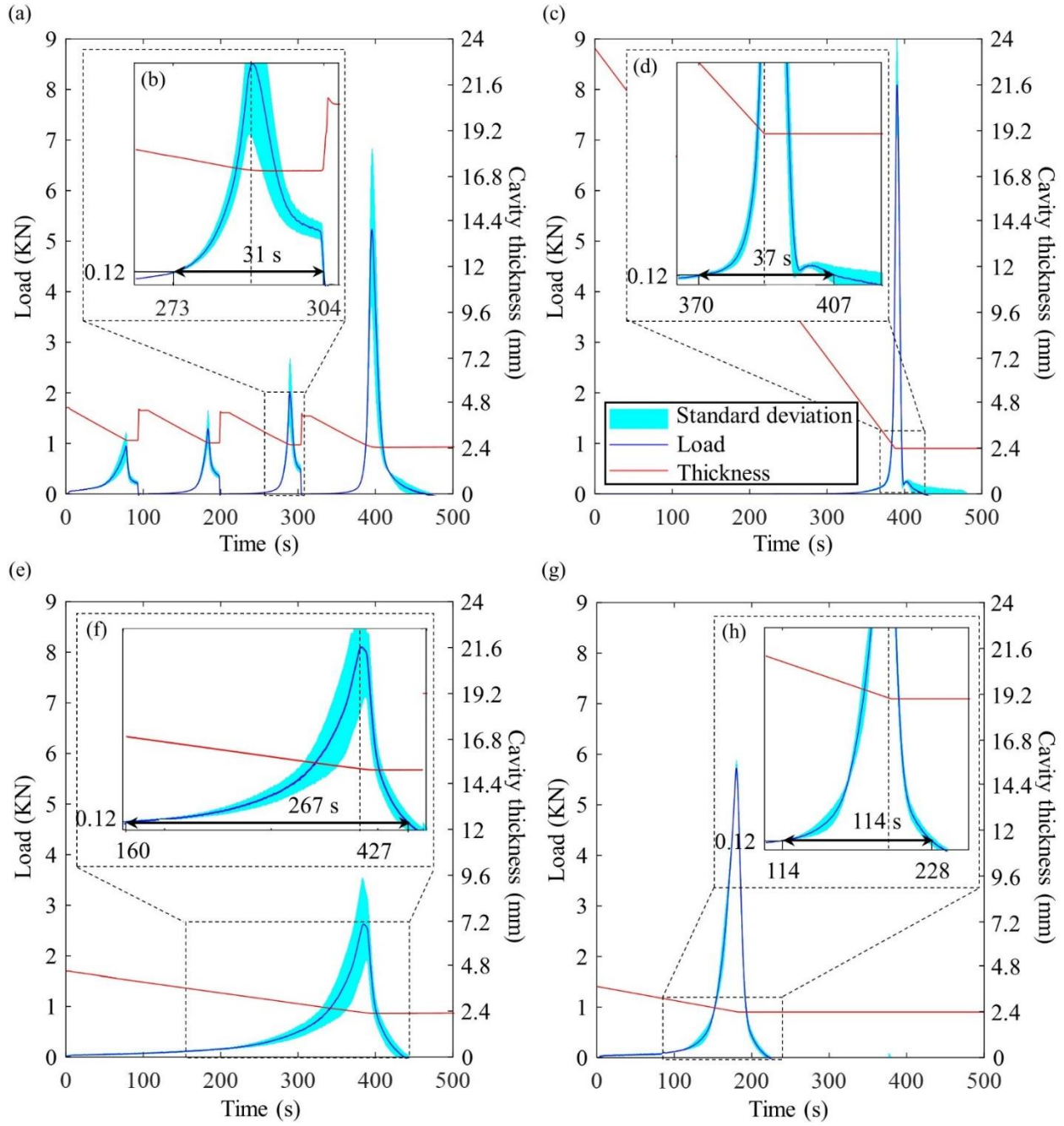


Figure 5-4 Experimental data of the variation of the measured load (kN) and cavity thickness (mm) of the moulding process over time (s). a) Consolidation in a multi-die process. c) in a SD-5° process. e) in a SD- $\alpha$  process. g) in a SD- $L_{eff}$  process. b) d) f) h) Close-up views showing  $t_{eff}$ .

### 5.3.1 Impregnation assessment

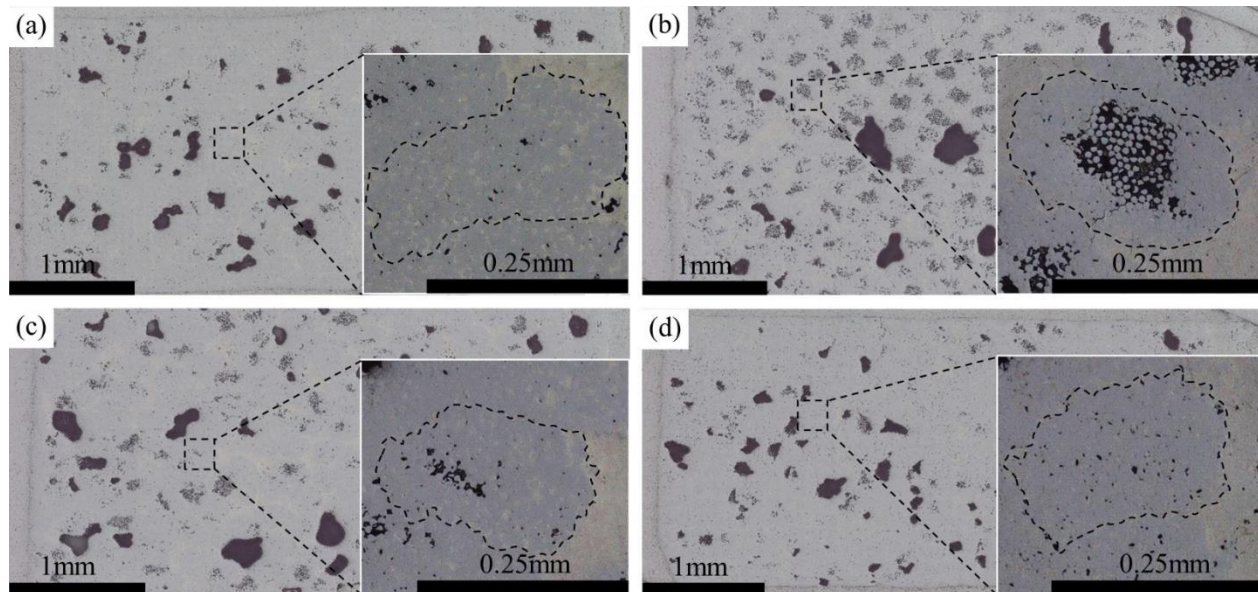


Figure 5-5 Cross-sectional images of coupons from (a) MD, (b) SD-5°, (c) SD- $\alpha$  and (d) SD- $L_{eff}$ . Insets show enlarged views of a yarn cross section encircled by a black dashed line.

Figure 5-5 presents microscopic images of the cross-section of coupons from (a) MD, (b) SD-5°, (c) SD- $\alpha$ , and (d) SD- $L_{eff}$ . Black areas represent voids. The presence of macro-voids was seen in all coupons taken from each experiment. The macro-voids were mostly present towards the centre of the coupons. The macro-voids are attributed to the 20 min.-long cooling duration of the composite plate under fixed cavity thickness. The plate's surfaces have cooled and shrunk before the centre of the plates. Since the plates adhered to the moulds surface, the plate thickness could not reduce under material shrinkage. That led to the macro-voids in the plate's centre. In thermoplastic pultrusion, profile cooling happens in the order of seconds and the profile detach from the pultrusion cavity wall due to shrinkage. This different cooling behaviour prevents the creation of macro-voids in pultruded profiles, as observed in multiple studies [12], [42], [56]. The insets show a fibre tow. Figure 5-6 presents the average tow area ( $A_{tow}$ ) for fibres from MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$ . The lowest  $A_{tow}$  was for fibres from SD-5°. This indicated that fibre tows were more compacted in the SD-5° experiment, compared to those from other experiments. This was explained by the high compaction load demonstrated in the previous section. According to the literature, elevated pressure could compact the fibre tows locally, thereby reducing their permeability [20], [72]. Additionally, resin-rich areas were more visible in all SD experiments in

contrast to the multi-die experiment. This suggested that the deconsolidation and cyclic compaction happening during the MD experiment, aided in the dispersion and relaxation of the fibre tows.

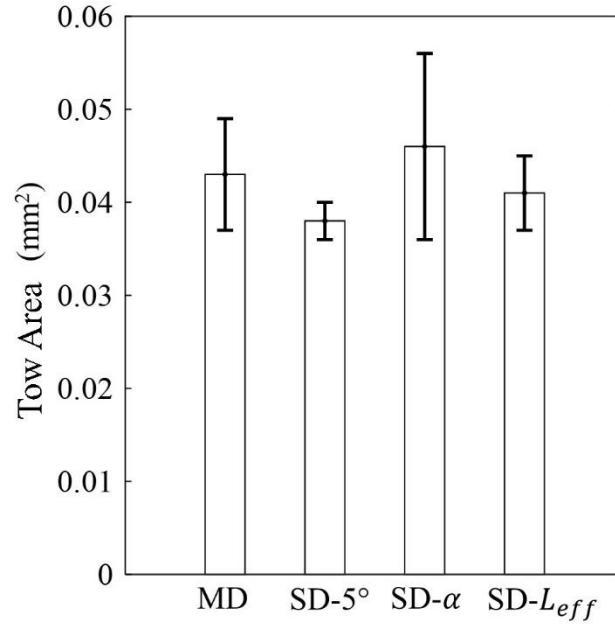


Figure 5-6 Average tow area ( $A_{tow}$ ) for fibres from MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$ .

To further evaluate the compression load of each process, a cumulative load ( $A_{load}$ ) was calculated. The cumulative load is defined as the integral of the load with respect to time curves (kN.s). Figure 5-7(a) presents the cumulative load and Figure 5-7(b) presents the  $t_{eff}$  for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$ . Displayed error bars for each process are the standard deviations on four repeated experiments.  $A_{load}$  was calculated at  $115 \pm 3$  KN.s,  $64 \pm 14$  KN.s,  $176 \pm 54$  KN.s and  $147 \pm 9$  KN.s for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$  respectively.  $t_{eff}$  was calculated at  $183 \pm 9$  s,  $37 \pm 2$  s,  $267 \pm 25$  s and  $114 \pm 6$  s for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$  respectively. Figure 5-7(c) presents the average impregnation ratio ( $R_{imp}$ ) for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$ . Displayed error bars for each experiment are the standard deviations on 24 analysed fibre tows.  $R_{imp}$  was measured at  $96 \pm 4$  %,  $73 \pm 12$  %,  $95 \pm 5$  % and  $89 \pm 10$  % for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$  respectively. SD-5° exhibited the lowest  $R_{imp}$  and lowest  $A_{load}$  and  $t_{eff}$ . Even though this experiment registered the highest peak compression load, as seen in Figure 5-4(c), there was an insufficient time to adequately consolidate the composite. Additionally, it is assumed that the high compression load in the SD-5° compacted the fibre tows and lowered their permeability. Similar observations were

reported in Babeau *et al.* [20] and Bechtold *et al.*[72]. SD- $L_{eff}$  had higher  $A_{load}$  and  $t_{eff}$  than SD-5°, which lead to an increase in its  $R_{imp}$ . MD and SD- $\alpha$  displayed equivalent  $R_{imp}$  values and coinciding standard deviations. However, SD- $\alpha$  had higher  $A_{load}$  and  $t_{eff}$  than multi-die. Notably, MD differed from SD- $\alpha$  process in terms of its cyclic compaction and deconsolidation of the composite. This suggested that the cyclic compaction and deconsolidation were the reason for the high  $R_{imp}$ . Indicating that for multi-die process there is no need for a higher  $A_{load}$  and  $t_{eff}$  due to the cyclic compaction and deconsolidation that enhanced the permeability of the wetted fibres. This confirms the conclusions of Betanzos *et al.* [82] and Lessard *et al.*[45].

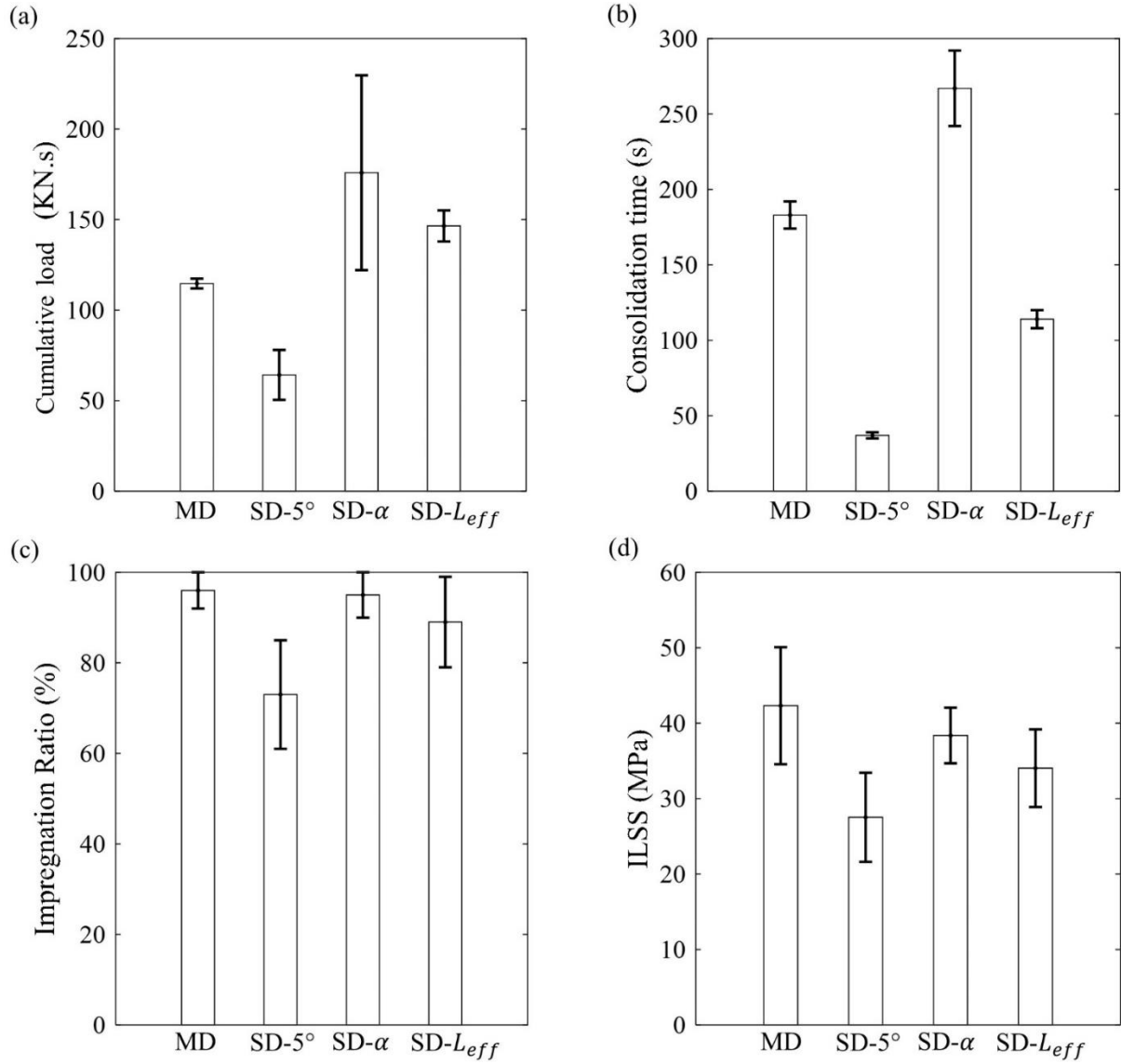


Figure 5-7 Average cumulative load  $A_{load}$  (KN.s), (b) average consolidation time  $t_{eff}$  (s), (c) average impregnation ratio  $R_{imp}$  (%) and (d) ILSS (MPa) for MD, SD-5°, SD-α and SD- $L_{eff}$ .

### 5.3.2 Composite mechanical properties

Figure 5-7(d) presents the ILSS of the GF/PET composite coupons. Displayed error bars for each experiment are the standard deviations on 16 samples. The ILSS was  $42.3 \pm 7.8$  MPa,  $27.5 \pm 5.9$  MPa,  $38.4 \pm 3.7$  MPa and  $34.0 \pm 5.2$  MPa for MD, SD-5°, SD-α and SD- $L_{eff}$  respectively. The bar graph demonstrates a noticeable decrease in the ILSS from multi-die to SD-5°. This was consistent with the decrease in  $R_{imp}$  shown in Figure 5-7(c). Supporting the literature's argument that ILSS decreases with increasing porosity [70], [84]. Although the average values of the ILSS

vary between MD, SD- $\alpha$  and SD- $L_{eff}$  composite coupons, the error bars implies that the values are equivalent. However, the presence of the resin-rich areas observed in Figure 5-5 and the difference in  $R_{imp}$  in Figure 5-7(a) could be the cause of the decrease in the average values of the ILSS detected in SD-  $\alpha$  and SD- $L_{eff}$ . These results suggests that the MD process could result in manufacturing composites with higher mechanical properties.

## 5.4 Discussion

Table 5-3 presents a summary of the results for the maximum load,  $A_{load}$ ,  $t_{eff}$ ,  $R_{imp}$  and ILSS for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$ . MD and SD- $\alpha$  displayed similar  $R_{imp}$  and ILSS results. Suggesting that the cyclic compaction and deconsolidation enhanced the permeability of the wetted fibres in the MD process. However, it is important to note that although the  $t_{eff}$  was the highest for the SD- $\alpha$  process, employing a long pultrusion die could potentially have negative consequences. A narrow entrance and extended taper length may lead to a high shear rate flow against the pulling direction, ultimately resulting in excessive pulling forces, as referenced in the literature [44], [72]. During pultrusion, the polymer backflow increases gradually overtime. In a long pultrusion die, this backflow keeps accumulating during continuous pultrusion for long durations. However, in the case of a short pultrusion die with relatively high taper angle 5° the backflow is quickly evacuated. Furthermore, variations in temperature along a long pultrusion die can result in an increase in thermoplastic polymer viscosity. Suggesting that the accumulation of polymer backflow can obstruct the flow path for subsequent material, potentially causing fibre damage and thus irregularities in the pultruded product. In summary, the MD process provides better micro-impregnation while also lowering the risk of pultrudate damage.

Table 5-3 Summary of the results for the maximum load,  $A_{load}$ ,  $t_{eff}$ ,  $R_{imp}$  and ILSS for MD, SD-5°, SD- $\alpha$  and SD- $L_{eff}$ .

|               | Max load (KN) | $A_{load}$ (KN.s) | $t_{eff}$ (sec) | $R_{imp}$ (%)  | ILSS (MPa)     |
|---------------|---------------|-------------------|-----------------|----------------|----------------|
| MD            | $5.2 \pm 1.5$ | $115 \pm 3$       | $183 \pm 9$     | $96 \pm 4 \%$  | $42.3 \pm 7.8$ |
| SD-5°         | $8.0 \pm 1.4$ | $64 \pm 14$       | $37 \pm 2$      | $73 \pm 12 \%$ | $27.5 \pm 5.9$ |
| SD- $\alpha$  | $2.7 \pm 1.2$ | $176 \pm 54$      | $267 \pm 25$    | $95 \pm 5 \%$  | $38.4 \pm 3.7$ |
| SD- $L_{eff}$ | $5.8 \pm 0.1$ | $147 \pm 9$       | $114 \pm 6$     | $89 \pm 10 \%$ | $34.0 \pm 5.2$ |

## 5.5 Conclusion

A compression moulding tool was implemented to replicate the consolidation process happening during pultrusion. A design methodology of the replicated experiments was shown. Three concepts of SD were suggested, two of them being long pultrusion dies and one short pultrusion die. The moulding process demonstrated that using four consecutive short dies with  $5^\circ$  taper angle lowered the consolidation force and extended the consolidation period compared to a long single die with a  $5^\circ$  taper angle. The MD process exhibited an increase of 32% and 54% in  $R_{imp}$  and ILSS respectively, compared to SD- $5^\circ$ . This indicated that multi-die process resulted in a higher impregnation quality compared to SD- $5^\circ$  due to the restored fibres permeability between each die as opposed to the tightly packed fibre bundles in the single die. However, the die with a  $L_{eff}$  length demonstrated that the consolidation time alone was unable to achieve similar composite impregnation and mechanical properties without deconsolidation. The long single die with a  $1.05^\circ$  taper angle experiment achieved similar composite impregnation and mechanical properties to multi-die process. Yet, it was noted that a long die with a narrow entrance may lead to issues at the die entrance potentially causing fibre damage and irregularities in the pultruded composite. Moreover, it was observed that the deconsolidation and cyclic compaction occurring during the MD experiment may have facilitated the dispersion and relaxation of the fibre tows. In conclusion, MD process proved to enhance composite impregnation without requiring excessive consolidation time. Indicating that thermoplastic pultrusion using multi-die process has the capacity of producing high quality composites. In future studies, experiments with faster cooling will be carried out to eliminate the presence of macro-voids. In conclusion, the approach to replicate the consolidation process of pultrusion by compression moulding developed in this study would generate the potential for optimizing the number of successive dies and their respective geometry for desired composite quality in a cost-effective manner.

### Credit authorship contribution statement

M. El-Jakl: Conceptualization, Methodology, Investigation, Analysis, Writing – original draft.

L. Laberge Laberge: Conceptualization, Writing - Review & Editing, Supervision, Project administration, Funding acquisition.

### Declaration of Competing Interest

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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## **CHAPITRE 6     Article 3: Braid-trusion of hollow thermoplastic composites using an expanding mandrel approach.**

Maissaloun El-Jakl, Louis Laberge Lebel

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### **Abstract**

Braid-trusion involves the combination of a braiding machine with a pultrusion line to continuously produce composite structures with a constant cross-section and angle-oriented fibres. During the braid-trusion process, braids are subjected to tension and compression, which leads to braid deformation and fibre orientation variations. Successful braid-trusion of tubular structures need a braid design methodology that accounts for braid deformations during its passage in the pultrusion die. We present here a braid design methodology for square hollow pultruded beams reinforced by triaxial braids. It is proposed that the internal perimeter of the braid, constrained by the braiding and pultrusion mandrels, must expand during pultrusion to account for braid consolidation. Two braid design are proposed. The first has a constant braiding and pultrusion mandrel perimeter. The second has a braiding mandrel perimeter being 0.9 times of the pultrusion mandrel's, referred to the expanding mandrel approach. The design demonstrated the effect of an expanding mandrel in reducing fibre waviness by achieving similar braiding yarn lengths. Glass fibre/polyethylene terephthalate composites were pultruded with the constant and expanding mandrels. The constant mandrel braid-trusion failed while the expanding mandrel succeeded.

### **6.1 Introduction**

Hollow composite structures find extensive use across various industrial sectors, including aerospace, automotive, marine, sports, and others, primarily due to their lightweight nature and remarkable strength-to-weight ratio [86]. However, the production of these structures presents a unique manufacturing challenge, particularly in relation to the internal surface support and consolidation of the hollow structure [87]. Typically, hollow composite structures are manufactured by using thermosetting prepreg tapes and an inflatable bladder moulding technique [88]. This process involves wrapping the material around an inflatable bladder. The assembled components are then placed within a mould to define the external shape of the hollow structure. By pressurizing the bladder, the prepregs are consolidated outward, ensuring proper consolidation. After curing, the bladder pressure is released, collapsed, and removed. This manufacturing process

requires skilled labour, and the use of elastomers as bladder materials, which generally have limited temperature tolerance [87]. Thus, a demand arises to develop an automated manufacturing process suitable for efficiently producing hollow composite products with excellent mechanical properties.

Pultrusion involves the continuous production of composite material structures with constant cross-sections [11]. This manufacturing process is highly automated and allows for the mass production of structures capable of reaching high fibre volume fractions. Pultruded profiles have shown their ability to fulfil cost, mechanical, physical, and environmental durability criteria while delivering outstanding performance [17]. In recent decades, profiles manufactured through the pultrusion process have garnered growing attention across various industries [17]. Thermoset composite pultrusion is well-established and widely utilized, unlike pultrusion with thermoplastic matrices. Yet thermoplastic composites offer more advantages, as they have higher toughness and damage tolerance, are recyclable and can be joined by welding [11]. In thermoplastic pultrusion, the challenge lies in impregnation due to the molten viscosity being several orders of magnitude higher than that of thermoset polymers, typically ranging from 500 to 5000 Pa·s [18]. To address the challenge posed by the high viscosity of thermoplastics, multiple hybrid yarn technologies have been developed to be used as input materials in thermoplastic pultrusion [2]. These yarns entail blending the polymer with the reinforcement fibres. Thereby, the distance for the polymers to flow and impregnate the reinforcement fibres is reduced. Examples of thermoplastic hybrid yarns input material are prepregs, towpregs, commingled, co-wound and DREF yarns. Unlike other input material, DREF yarns have not been widely employed in thermoplastic pultrusion. DREF yarns consist of a continuous fibre core onto which discontinuous polymer fibres are spun using the friction spinning process [33]. DREF yarns offers benefits such as superior polymer/fibre distribution and improved handling compared to other types of hybrid yarns. To the author's knowledge, DREF yarns were not used in a pultruded braid structure.

In a hybrid yarn input material thermoplastic pultrusion, input materials are wound onto bobbins are placed in a creel that controls the unwinding tension [28]. Subsequently, yarns are drawn through a preheater, heated die, then a cooling system, by the means of a puller. At the end of the manufacturing line, a saw is positioned to cut the finished composite products. The heated die is where melting and impregnation happen. At that stage, the material in the die is called a pultrudate. In a recent development, a series of heated dies arranged sequentially with a decreasing final cross-

section have been utilized to enhance the impregnation of thermoplastic composites. This process is called multi-die pultrusion [12], [28], [29], [56], [66].

Braid-trusion is a production method for composite materials, where a braiding machine is linked with a pultrusion die to produce structures with constant cross-sections and off-axis fibre orientation in a continuous manner [5]. Braiding is a textile process in which three or more yarns are interlaced to produce a continuous structure [50]. This process has received great attention in composites manufacturing due to the braid's mechanical properties, such as good fatigue and crush resistance properties [89]. Michaeli *et al* [25] was one of the first to introduce braiding to thermoplastic pultrusion and produced polypropylene/glass and polyether-ether-ketone/carbon structures. It was later reported that the orientation of braided fibres changes between the before and after pultrusion state [5], [56]. Few experimental studies have explored manufacturing of hollow pultruded thermoplastic composites, primarily focusing on tube structures [58]–[60], [62], with one study introducing a square configuration [59]. However, none of them appear to have utilized a braid design methodology. It is suspected that the braid designs for pultrusion in the aforementioned studies relied on a trial-and-error approach. One of the first publications to introduce braid design for the braid-trusion process was by Laberge Lebel and Nakai [5]. The design was based on the circumferential deformation of a solid triaxial braid during consolidation. It was demonstrated that when subjected to compression within the pultrusion die, the braid's thickness decreases, leading to a reduction in the amplitude of the braiding yarns' oscillation within the braid structure. These oscillations characterize the yarn crimp, which is the actual length of the yarn over the fabric's length along the yarn path [90]. It was found that the braid structure needed to expand in its circumferential direction to adjust to reduction of yarn crimp during braid consolidation [5]. Ghaedsharaf *et al.* [53] introduced a finite element model (FEM) of the 3D geometry of biaxial braids during pultrusion. Contrary to triaxial braids, biaxial braids can also deform along their length under tensile load since there are no axial yarns running along the braid. Using their FEM approach, Ghaedsharaf *et al.* [53] successfully designed a biaxial braid that could be pultruded with prediction of the braid's elongation and final diameter. Both models suggested that the braid-trusion process must account for the lengthening of yarns due to reduction in the yarn's crimp. However, models presented are complex, involving substantial computational time, and have not been validated for tubular braids. Braid-trusion of hollow thermoplastic composites using hybrid yarns was not thoroughly explored. Furthermore, all consolidation methods for the

aforementioned tubular braid-trusion studies employed a straight section mandrel. Although braid-trusion of a solid structure is a complex process, braid-trusion of hollow structures using simultaneous internal and external tooling presents an even greater challenge. To the authors' understanding, no studies have developed hollow braid-trusion design methodology using the alternative approach of an expanding mandrel.

The objective of this study is to develop a simplified hollow braid design methodology incorporating an expanding mandrel. A simple braid geometrical model is first introduced. The braid-trusion design methodology is described. Then, the design method was applied for braid-trusion of a square tubing on a constant and expanding cross-section mandrel. Braid-trusion experiments were conducted according to the design and the braid-truded tubes were characterized.

## 6.2 Braid geometrical model.

Figure 6-1(a) shows the geometrical model of a tubular triaxial braid comprising 24 braiding yarns (BY) and 12 axial yarns (AY) laid onto a rectangular mandrel with rounded corners. The braid is modelled using a 3D Computer-Aided Design (CAD) software (CATIA V5R19). A first set of 12 BYs, in blue on Figure 6-1, spirals in a clockwise direction (CBY). The second set of 12 BYs, in green on Figure 6-1, spirals in a counterclockwise direction (CCBY). The braid pitch ( $p$ ) represents the distance for one BY to complete one full rotation around the braid axis. The pitch is controlled by the braiding machine, that is, by the ratio of the pulling speed of the braid to the rotation speed of the braiding yarns carriers around the braider head [5]. Braids are commonly described with their braid angle ( $\alpha$ ). The braid angle depends on the braid's perimeter ( $C$ ) and  $p$ , as,

$$\tan \alpha = C/p \quad (6.1)$$

$$\tan \alpha = C/p \quad (1)$$

Therefore, the higher the  $C/p$  ratio is, the higher the braid angle will be. Braids are typically made with angles  $\alpha$  between  $10^\circ$  and  $80^\circ$  [91]. The BY path is modelled as a spline having definition points equally spaced onto a one turn spiral, (*i.e.*, for one pitch) around the mandrel. The radial position of the definition points is determined according to the mandrel surface and thickness of the yarns. Figure 6-1 (b) presents the cross-section of the braid showing a representation of the yarns' path. The mandrel has a width ( $W_{mandrel}$ ), a depth ( $H_{mandrel}$ ) and rounded edges radius ( $R_{mandrel}$ ). For this braid architecture of 24 BY and 12 AY, it can be seen on Figure 6-1 that the

CBY pass above all the corners' AY, whereas the CCBY pass under them. This means that if the AY yarns have all the same thickness, the length of the CBY ( $L_{CBY}$ ) passing over the corners will be significantly longer than the length of the CCBY ( $L_{CCBY}$ ) passing close to the mandrel's corner surfaces ( $L_{CBY} > L_{CCBY}$ ). To avoid this difference in length, the braid was deliberately modelled with six large AY yarns, (i.e., the LAY), and six small AY yarns (the SAY) in alternating positions. While yarns are typically represented by an elliptical or lenticular cross-section in kinematic textile modelling [5], it was decided for CAD simplicity to model yarns with circular cross-sections. The diameter of the modelled circular yarns is equal to the yarn's thickness in the structure. The BY's thickness is  $b_{BY}$ . The thickness of the LAY and SAY are  $b_{LAY}$  and  $b_{SAY}$ , respectively. The thickness of the braid ( $t_{braid}$ ) is the summation of two  $b_{BY}$  and one  $b_{LAY}$ . In a braid architecture, the yarns path oscillation is characterized by the crimp ratio ( $R_c$ ) as

$$R_c = L/l \quad (6.2)$$

where  $L$  is the actual length of the fibre and  $l$  is the fabric length along the yarn path. In Figure 6-1,  $l$  is depicted in light blue.  $L$  and  $l$  can be measured on the CAD model. As  $R_c$  increases, the waviness of the fibres increases and the axial mechanical properties of the composite decreases [90].

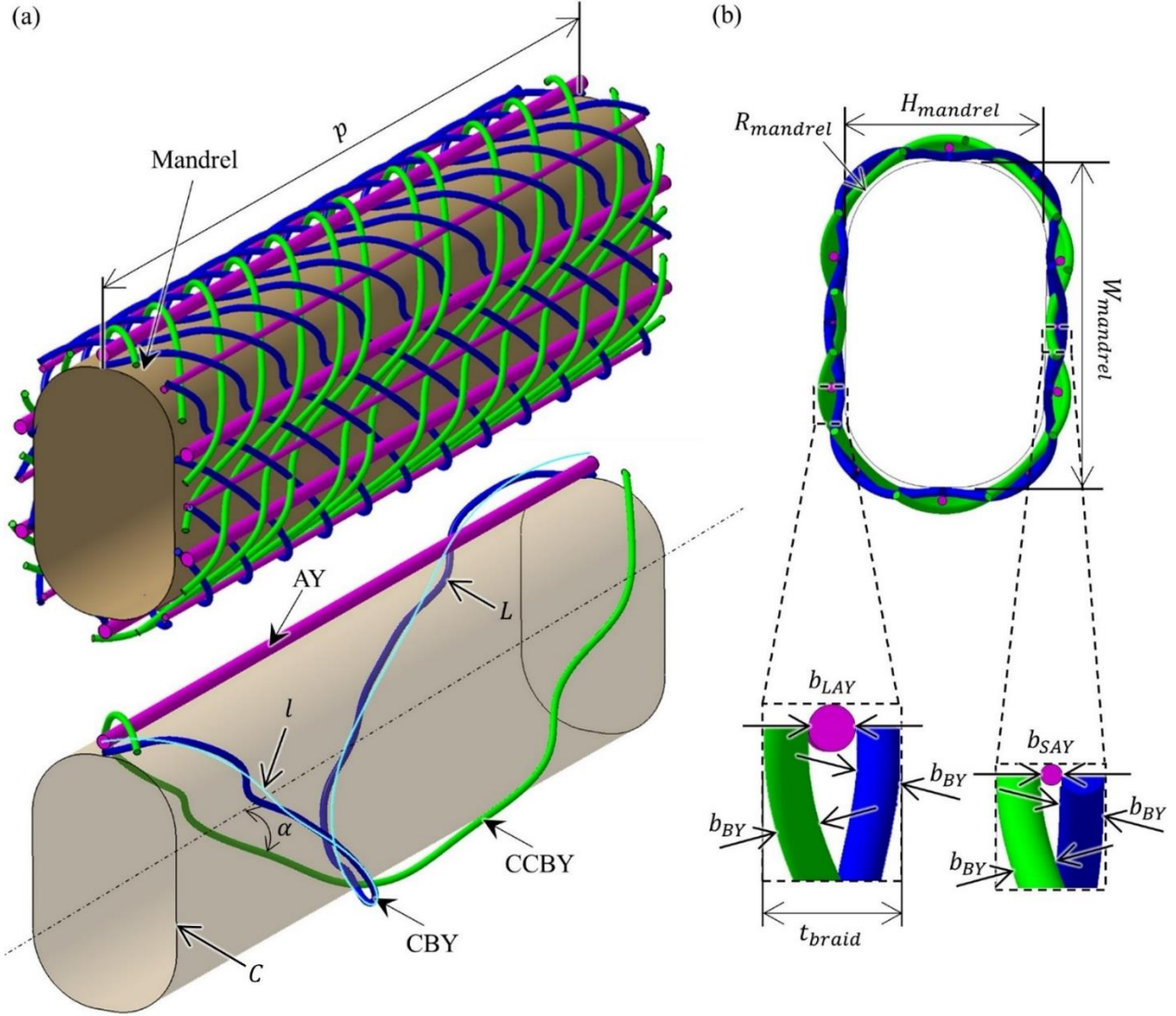


Figure 6-1. (a) Tubular triaxial braid architecture on a rectangular mandrel presenting  $C$ ,  $p$  and  $\alpha$ . CBY and CCBY yarns are presented in blue and green respectively. AY are presented in purple (b) Braid's cross-section showing a representation of the yarns' path. The enlarged view shows the yarns' diameter set to the yarns' thickness  $b_{BY}$ ,  $b_{LAY}$ , and  $b_{SAY}$ .

### 6.3 Braid-trusion design methodology.

Figure 6-2 illustrates the braid on a mandrel before and after pultrusion. Both braids are created with the model introduced in Section 2. The mandrel has the length of three braid pitches. The first pitch has mandrel and braid dimensions before pultrusion, the last pitch has mandrel and braid dimensions after pultrusion. The middle pitch section presents the braid during the pultrusion process which consists of the mandrel expansion in the dies. The before and after conditions and

parameters of the braid will be referred to as in and out. One CBY and CCBY were isolated on top and bottom of Figure 6-2 to showcase the length of the BY before ( $L_{CBY,in}$ ,  $L_{CCBY,in}$ ) and after pultrusion ( $L_{CBY,out}$ ,  $L_{CCBY,out}$ ). The left-hand side of Figure 6-2 shows view of the width of the mandrel before pultrusion ( $W_{in}$ ) illustrating the CBY and CCBY braid angle before pultrusion ( $\alpha_{CBY,in}$ ,  $\alpha_{CCBY,in}$ ). The right-hand side shows view of the width of the mandrel after pultrusion ( $W_{out}$ ) illustrating the braid angles after pultrusion ( $\alpha_{CBY,out}$ ,  $\alpha_{CCBY,out}$ ). The mandrel perimeters before ( $C_{in}$ ) and after ( $C_{out}$ ) pultrusion are shown. While the mandrel dimensions after pultrusion are selected according to the product's requirements, the dimensions of the mandrel before pultrusion (*i.e.*, the braiding mandrel's dimensions) can be proportionally reduced by a reduction ratio ( $R_m$ ) defined as,

$$R_m = \frac{C_{in}}{C_{out}} \quad (6.3)$$

Since reinforcement yarns of glass or carbon fibres are inextensible, the braid perimeter must be expanded to account for the reduction of the crimp during consolidation in the pultrusion die. In other words, the braiding mandrel must be smaller than the pultrusion mandrel. The  $R_m$  can be selected using the CAD model of Figure 6-2. The selection of the proper  $R_m$  for achieving a successful braid-trusion of hollow structure is determined by having the modelled length of the yarns before ( $L_{CBY,in}$ ,  $L_{CCBY,in}$ ) and after pultrusion ( $L_{CBY,out}$ ,  $L_{CCBY,out}$ ) equal. In case that  $L_{CBY,in} > L_{CBY,out}$  or  $L_{CCBY,in} > L_{CCBY,out}$ , the yarns in the braid before pultrusion are longer than the available space in the braided structure. The extra yarn length will then wrinkle and lead to detrimental fibre waviness for the mechanical properties. With time, the extra yarn length would accumulate at the pultrusion die entrance and could lead to blockage stopping the pultrusion. The alternative case that  $L_{CBY,in} < L_{CBY,out}$  or  $L_{CCBY,in} < L_{CCBY,out}$ , the BY will apply a wrapping force on the pultrusion mandrel causing high friction, also leading to a stop of the pultrusion, since the BY cannot elongate.

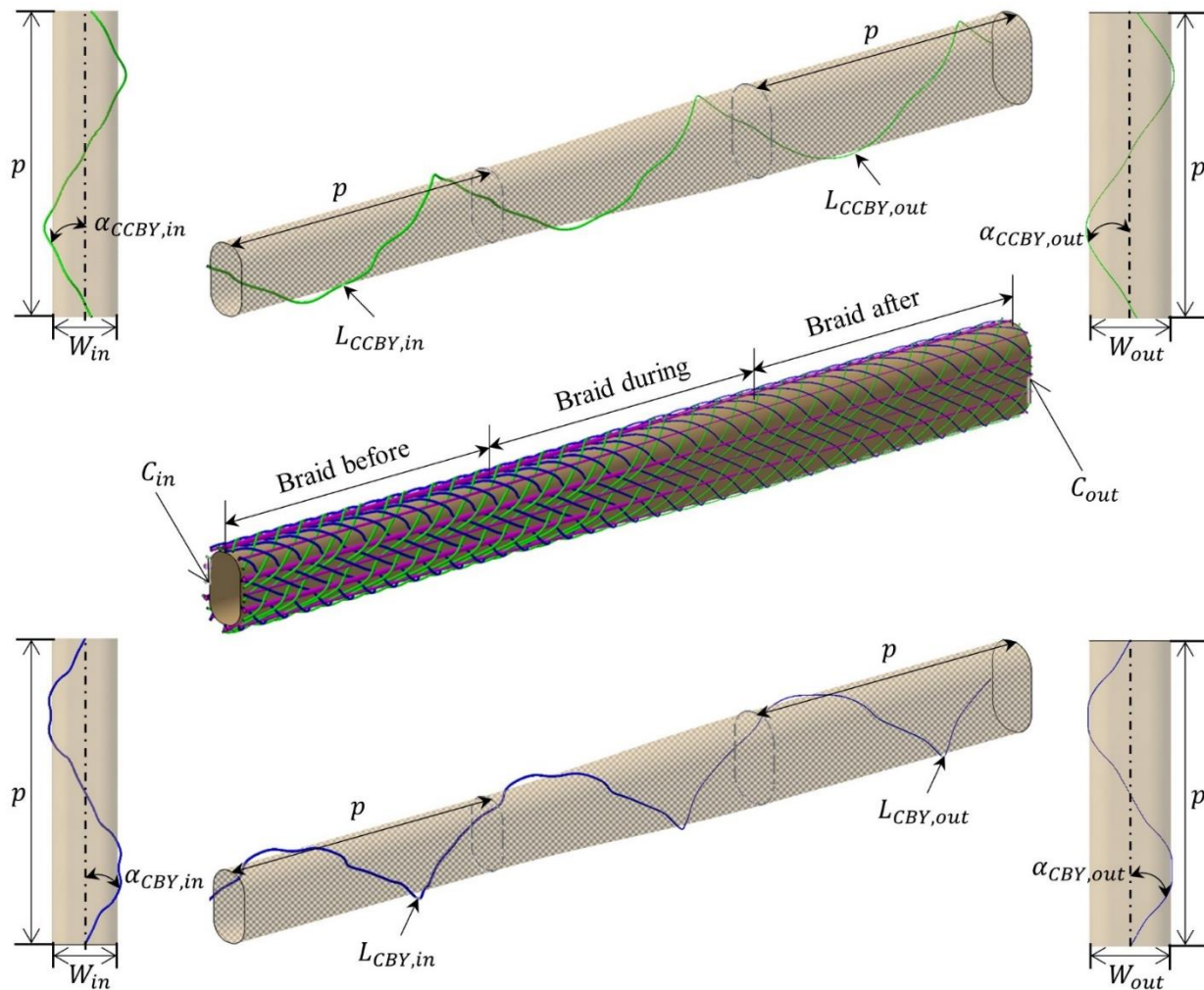


Figure 6-2. Braid CAD presenting braid before, during and after pultrusion. On the top and bottom sections, one CBY and CCBY were isolated to showcase the length of the BY before and after pultrusion. On the left side, section cuts of  $W_{in}$  along  $p$  illustrate CBY and CCBY angles before pultrusion. On the right side, section cuts of  $W_{out}$  along  $p$  illustrate CBY and CCBY angles after pultrusion.

Figure 6-3 shows a flow chart outlining the braid-trusion design methodology. The design methodology focuses initially on generating the CAD of the braid after pultrusion. The methodology has several inputs, some can be modified during the design, while others are fixed. The fixed set of inputs, include firstly the braid structure which is determined by the braiding machine configuration. Secondly, the braid internal geometry after pultrusion and thickness ( $t_{out}$ ) are determined by the braid-trusion die and mandrel geometries and dimensions. The braid parameters are the set of inputs that can be modified during the design. This includes the braid pitch which is defined initially to reach the desired fibre angle for the product performance. The

number of braid layers ( $N_{layer}$ ) and the thicknesses of BY ( $b_{BY,out}$ ) and LAY ( $b_{LAY,out}$ ) are defined to satisfy equation 6.4. Unidirectional (UD) yarns can be added between braid layers. The thickness  $b_{UD,out}$  can be defined to fill the vacant space between layers. Yarns are defined by their yarn count (i.e., the yarn's mass per unit length) and the material type density. At the starting point, all axial yarns are considered as LAY. The first step is to determine the model's yarn thickness after pultrusion. The braid thickness after pultrusion,  $t_{out}$ , is known and expressed by,

$$t_{out} = N_{layer} [2b_{BY,out} + b_{LAY,out}] \quad (6.4)$$

To solve equation 6.4, the ratio between  $b_{BY,out}$  and  $b_{LAY,out}$  is assumed to be the ratio of their yarn counts of the reinforcement ( $F_{f,BY}$ ,  $F_{f,LAY}$ ) and reinforcement material density ( $\rho_{f,BY}$ ,  $\rho_{f,LAY}$ ), such that,

$$\frac{b_{BY,out}}{b_{LAY,out}} = \frac{F_{f,BY} \times \rho_{f,LAY}}{\rho_{f,BY} \times F_{f,LAY}} \quad (6.5)$$

At this point, the braid after pultrusion can be modelled in the CAD software and the yarn lengths can be measured. The condition  $L_{CBY} = L_{CCBY}$  is checked. If it is not verified, SAY are iteratively selected and strategically placed into the braid until the condition is verified. The  $b_{SAY,out}$  is determined as a proportion of the  $b_{LAY,out}$ , analogous to equation 6.5 using,

$$b_{SAY,out} = \frac{F_{f,SAY} \times \rho_{f,LAY}}{\rho_{f,SAY} \times F_{f,LAY}} b_{LAY,out} \quad (6.6)$$

where  $F_{f,SAY}$  is the yarn count and  $\rho_{f,SAY}$  is the density of the reinforcement of the SAY.

Once the braid after pultrusion is modelled, the fibre volume fraction ( $v_f$ ) of the braid can be calculated using the area of reinforcement fibres ( $A_f$ ) on the area of the pultruded product as,

$$v_f = \frac{A_f}{(A_{Die} - A_{mandrel})} \quad (6.7)$$

The  $A_f$  is calculated by,

$$A_f = \sum_{layer\ 1}^N \left[ N_{CBY} \frac{F_{f,CBY}}{\rho_{f,CBY}} \frac{L_{CBY,out}}{p} + N_{CCBY} \frac{F_{f,CCBY}}{\rho_{f,CCBY}} \frac{L_{CCBY,out}}{p} + N_{LAY} \frac{F_{f,LAY}}{\rho_{f,LAY}} \right. \\ \left. + N_{SAY} \frac{F_{f,SAY}}{\rho_{f,SAY}} \right] + N_{UD} \frac{F_{f,UD}}{\rho_{f,UD}} \quad (6.8)$$

Equation 6.8 is solved using the reinforcement yarn counts ( $F_f$ ), density ( $\rho_f$ ) and quantity of yarns ( $N$ ) for all yarn types (CBY, CCBY, LAY, SAY, and UD). Note that the yarns are assemblies of several DREF tows. The  $L_{CBY,out}$  and  $L_{CCBY,out}$  are measured on the CAD model.

The calculated  $v_f$  can be compared to a target value for the product. In case it is judged too far from the target value, the design of the braid after pultrusion can be adjusted by modifying any of the input braid parameters.

The next verification is the die filling condition. For successful pultrusion, it is recommended that the filling ratio ( $R_f$ ) of the die be above 100%.  $R_f$  is calculated according to,

$$R_f = \frac{A_f + A_m}{(A_{Die} - A_{mandrel})} \quad (6.9)$$

In equation 6.9,  $A_m$  is the matrix area. This area is calculated using equation 7 with replacing the reinforcement yarn counts ( $F_f$ ) and density ( $\rho_f$ ) by the polymer's ( $F_m, \rho_m$ ). If  $R_f < 100\%$ , input material composition should be changed to get a  $R_f$  superior to 100%.

The braid-trusion design methodology then proceeds to the modelling of the braid before pultrusion. The important information needed are the yarn thicknesses. Since they are unconstrained, and without the use of advanced textile modelling methods, the yarn thickness must be measured in an actual braid. This braid must be formed onto a mandrel of the same dimensions as the pultrusion mandrel. The yarns'  $b_{BY,in}$ ,  $b_{LAY,in}$  and  $b_{SAY,in}$  are measured. Once the braid before pultrusion is modelled, the  $L_{CBY,in}$  and  $L_{CCBY,in}$  are measured on the CAD. They are compared to the  $L_{CBY,out}$  and  $L_{CCBY,out}$ . If they are not similar,  $R_m$  is then reduced iteratively until reaching  $L_{CBY,in} = L_{CBY,out}$  and  $L_{CCBY,in} = L_{CCBY,out}$ .

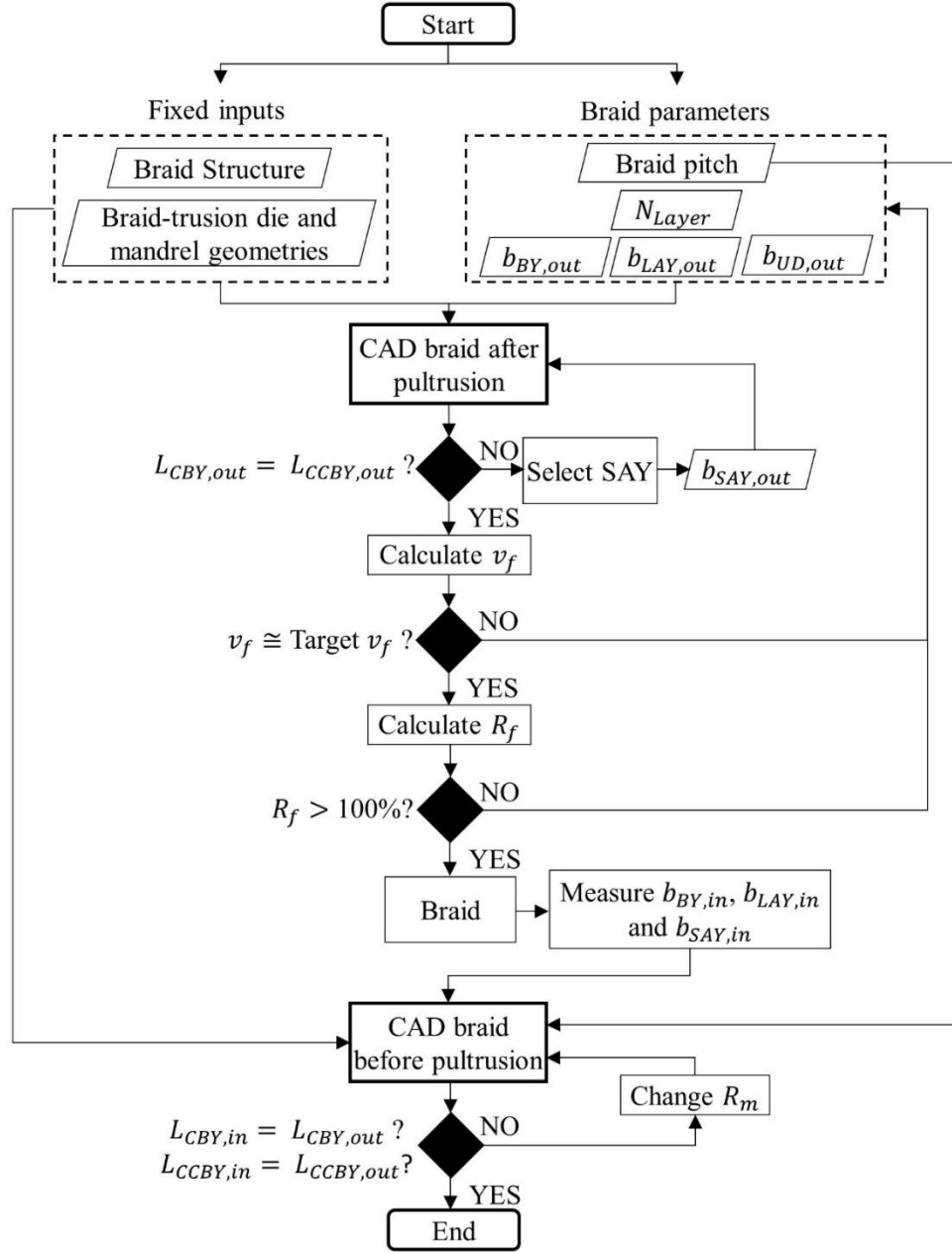


Figure 6-3. Braid-trusion design methodology flow chart starting from the definition of the pultruded braid to get the braid parameters before pultrusion.

## 6.4 Experimental

### 6.4.1 Materials

DREF yarns were used as the hybrid yarn input material for this study. The reinforcement fibre consisted of S2-GF (SCG75, AGY) and the thermoplastic polymer was PET (staple fibre, Huvis

Corporation). The DREF tows were obtained by friction spinning (Filspec Inc.). The DREF tow was composed of a GF core covered by discontinuous spun PET fibres. GF linear density ( $F_f$ ) was 66 tex and PET linear density ( $F_m$ ) was 38 tex. The DREF tow had a nominal volume fraction ( $v_{n,f}$ ) of 50%. A number of these small DREF tows were co-wound together in bobbins to form the larger yarns needed for the braid structure. Table 6-1 displays properties of the PET fibres used in DREF. The melting point ( $T_m$ ), glass transition ( $T_g$ ) and degradation temperatures were acquired using a DSC (MDSC 2000, TA instruments). Solid density was determined using a Pycnometer (Accupyc 1340, Micrometrics) and the melt density was measured by PVT analysis (pvT 100, ThermoHaake) at 270-285°C. GF density was of 2.46 g/ cm<sup>3</sup> as per AGY's technical datasheets.

Table 6-1 PET material properties

| Parameter                      | Unit              | Value |
|--------------------------------|-------------------|-------|
| <b>Melting point</b>           | °C                | 257   |
| <b>Glass transition</b>        | °C                | 80    |
| <b>Degradation temperature</b> | °C                | 295   |
| <b>Solid density</b>           | g/cm <sup>3</sup> | 1.43  |
| <b>Melt density</b>            | g/cm <sup>3</sup> | 1.06  |

#### 6.4.2 Braid-trusion design application

The braid-trusion design mentioned above was used for a particular case. The fixed inputs are initially set as follows. A triaxial braid comprising 24 braiding yarns (BY) and 12 axial yarns (AY). This braiding machine arrangement produce a regular braid structure (2 over 2) [92]. Figure 6-4(a) and (b) presents the mandrel and die cross-section geometry. Both the mandrel and die had a rectangular cross-section with rounded corners. The mandrel depth, width and radius were 26,7 mm, 16.3 mm, and 7,25 mm. The mandrel perimeter was 84.3 mm. The die depth, width and radius were 29.7 mm, 19.3 mm, and 4.5 mm. With the mandrel inserted in the die, the pultruded profile wall thickness  $t_{out} = 1.5$  mm and its area was  $A_{Die} - A_{Mandrel} = 164mm^2$ . The different radius values between the mandrel and the die creates a zone with increased thickness in each of the corners. Figure 6-4(c) shows the modelled braid structure after pultrusion. Two layers of braid were selected with four UD yarns added in the corners to fill the space created by the different mandrel and die radii.

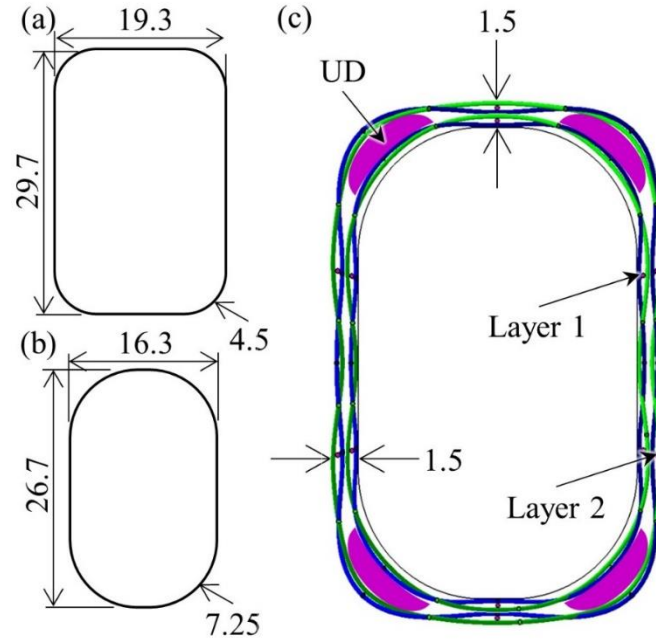


Figure 6-4(a) Die geometry, (b) mandrel geometry and (c) braid-truded structure after pultrusion showing its total thickness, 2 braid layers and 4 UD yarns at the corners. All dimensions are in mm.

Table 6-2 Parameters of the braid after and before pultrusion as determined by the braid-trusion methodology.

| Yarns | DREF tow amount | $\rho_f$ (c/cc) | $\rho_m$ (c/cc) | $F_f$ (tex) | $F_m$ (tex) | $b_{out}$ (mm) | $b_{in}$ (mm) |
|-------|-----------------|-----------------|-----------------|-------------|-------------|----------------|---------------|
| BY    | 40              | 2.46            | 1.08            | 2640        | 1520        | 0.25           | 1.10          |
| LAY   | 40              | 2.46            | 1.08            | 2640        | 1520        | 0.25           | 1.10          |
| UD    | 80              | 2.46            | 1.08            | 5280        | 3040        | 0.5            | n/a           |
| SAY   | 8               | 2.46            | 1.08            | 528         | 304         | 0.05           | 0.22          |

Secondly, the braid-trusion design methodology was applied. The braid pitch was chosen at 105 mm for the first layer and 140 mm for the second layer. Table 6-2 shows the yarns selected for the braids. The BY and LAY were composed of 40 DREF tows. The UD were composed of 80 DREF tows to fill the corners. The braid after pultrusion was then modelled with the verified condition of  $L_{CBY,out} = L_{CCBY,out}$ . To reach that condition, six LAY were replaced by six SAY containing 8 DREF tows. The  $v_f$  was calculated using equation 6.7 and 6.8 and the parameters of Table 6-2 with  $N_{CBY} = N_{CCBY} = 12$ ,  $N_{LAY} = N_{SAY} = 6$  and  $N_{UD} = 4$ . The calculated  $v_f$  was 55%, which corresponded to the target  $v_f$ . Following the design methodology,  $R_f$  was calculated and resulted in 128%. To proceed with the braid methodology, a braid was made on a tube with a circumference

in the same range than the pultrusion mandrel's perimeter. The tube had a diameter of 24 mm, its circumference was 75.3 mm. Subsequently, the yarn thicknesses in the braid were measured to be  $b_{BY,in} = b_{LAY,in} = 1.1$  mm and  $b_{SAY,in} = 0.22$  mm. The braid before pultrusion was then modelled. Table 6-3 presents the modelled braids after and before pultrusion. There were two cases modelled. The first case modelled with a constant section mandrel ( $R_m = 1$ ), while the second was modelled with an expanding cross-section mandrel ( $R_m = 0.9$ ). The two layers pitches are reported at 105 mm and 140 mm. After pultrusion, and for both cases, the first layer's mandrel perimeter was the mandrel tooling perimeter of after pultrusion, that is 73.6 mm. The second layer internal perimeter was estimated to be a rounded rectangle with the dimensions calculated with,

$$W_{Layer2} = W_{Die} - 2(2b_{BY,out} + 2b_{LAY,out}) = 17.8 \quad (6.10)$$

$$H_{Layer2} = H_{Die} - 2(2b_{BY,out} + 2b_{LAY,out}) = 28.2 \quad (6.11)$$

$$R_{Layer2} = R_{Die} = 4.5 \quad (6.12)$$

The rationale for this estimation was to consider that the second layer's external surface was matching the die's surface. The second layer internal perimeter was 84.3 mm. The  $L_{CBY,out}$  and  $L_{CCBY,out}$  are equivalent for each braided layers after pultrusion since these yarn lengths were adjusted by changing six LAY by six SAY containing 8 DREF tows each in the braid modelling. Moreover, both  $R_m = 1$  and  $R_m = 0.9$  cases have identical yarn length for CBY and CCBY after pultrusion. This is because both cases' braids after pultrusion are modelled on the same mandrel. The last 3 columns of Table 6-3 presents the modelled braids before pultrusion. The perimeters were calculated with mandrel dimensions modulated by the  $R_m$  factor. The  $L_{CBY,in}$  and  $L_{CCBY,in}$  are equivalent for each braided layer before pultrusion. This is expected since the braid structure after and before pultrusion is identical and that the yarn thicknesses are proportionally larger. For the two braided layers modelled for case  $R_m = 1$ , it is seen the modelled yarn lengths before pultrusion are larger than the yarn lengths after pultrusion. The average difference between yarn lengths before and after pultrusion is 6.00 mm. Therefore, the extra yarn length in the braid before pultrusion will buckle in the pultrusion die and could result in detrimental fibre waviness. For the braid designed with  $R_m = 0.9$ , both layers show that  $L_{CBY,CCBY,in}$  and  $L_{CBY,CCBY,out}$  had an average difference of 1.25 mm. Therefore, braid-trusion on this expanding cross-section would exhibit less fibre waviness. The comparison of these two braid designs showed that applying the braid design

methodology presented in Figure 6-2 can result in a braid before pultrusion (i.e., at  $R_m = 0.9$ ) that accounts for the reduction of the crimp during consolidation in the pultrusion die.

Table 6-3 Pitch, mandrel perimeters, measured length of CBY and CCBY after and before pultrusion, for the layers of braids with a constant section mandrel ( $R_m = 1$ ) and an expanding cross-section mandrel ( $R_m = 0.9$ ).

| $R_m$<br>(-) | Layer<br>(-) | $p$<br>(mm) | After Pultrusion  |                       |                        | Before Pultrusion |                      |                       |
|--------------|--------------|-------------|-------------------|-----------------------|------------------------|-------------------|----------------------|-----------------------|
|              |              |             | $C_{out}$<br>(mm) | $L_{CBY,out}$<br>(mm) | $L_{CCBY,out}$<br>(mm) | $C_{in}$<br>(mm)  | $L_{CBY,in}$<br>(mm) | $L_{CCBY,in}$<br>(mm) |
| 1            | 1            | 110         | 73.6              | 131                   | 131                    | 73.6              | 137                  | 138                   |
|              | 2            | 140         | 84.3              | 163                   | 162                    | 84.3              | 168                  | 167                   |
| 0.9          | 1            | 110         | 73.6              | 131                   | 131                    | 66.2              | 132                  | 133                   |
|              | 2            | 140         | 84.3              | 163                   | 162                    | 75.9              | 164                  | 163                   |

### 6.4.3 Braiding

Two braids were manufactured following the braid design of Table 6-3. The braiding mandrels were circular tubing. For the case  $R_m = 1$ , the tubing diameter was 24 mm, and its circumference was 75.4 mm. The tubing circumference was approximately equal to the  $C_{in}$  of 73.6 mm of the modelled first layer braid. For the case  $R_m = 0.9$ , the tubing diameter was 22 mm, and its circumference was 69.1 mm. The tubing circumference was approximately equal to the  $C_{in}$  of 66.2 mm of the modelled first layer braid. The braiding machine (KBL-24-1-110, Xuzhou Henghui Braiding Machine Co., Ltd.) was set to braid with pitches of 105 mm for the first layer and 140 mm for the second layer. After braiding the first layer, the four AX were welded on their intended position over the SAY (See Figure 6-4). The second layer was then over-braided to form a braided preform. The braided preforms were approximately 2 m long. The braid pitches and outside circumference were measured.

### 6.4.4 Pultrusion apparatus and process

Figure 6-5(a) illustrates the pultrusion apparatus used for the pultrusion of the braided preforms. The braided preforms were removed from their mandrels and slid on their respective pultrusion mandrels. The pultrusion mandrel was cantilevered from a fixed plate. The total mandrel length was 2 m. The preheating chamber was wrapped with a controlled heating tape set at 220°C. To keep the mandrel centred into the die's cavities, rollers were positioned before the preheating chamber. Five heated pultrusion dies were used and set at 280°C. The pultrusion dies were heated by cartridge heaters that were controlled using the feedback from type J thermocouples. Fans were

installed to cool down the thin-walled tube of the last die to 160°C. The braid was pulled through a puller system along the line at 25 mm/min. Figure 6-5(b) shows a cross-sectional view of the dies and mandrel with a constant cross-section. This mandrel was used for the pultrusion of the braided preform of case  $R_m = 1$ . Figure 6-5(c) shows a cross-sectional view of the dies and mandrel with an expanding cross-section. The mandrel initial perimeter was 0.9 times its finale one. This mandrel was used for the pultrusion of the braided preform of case  $R_m = 0.9$ . Figure 6-5(d) presents a cross-sectional view of one pultrusion die. All pultrusion dies had had a 6° taper with a 17.4 mm length and 8 mm straight section. The dies exit decrease gradually with an area decrease ratio of 4%. The last die presented in Figure 6-5(e) had a 2° taper angle and a 1.6 mm thin-walled rectangular tube of 42.8 mm long.

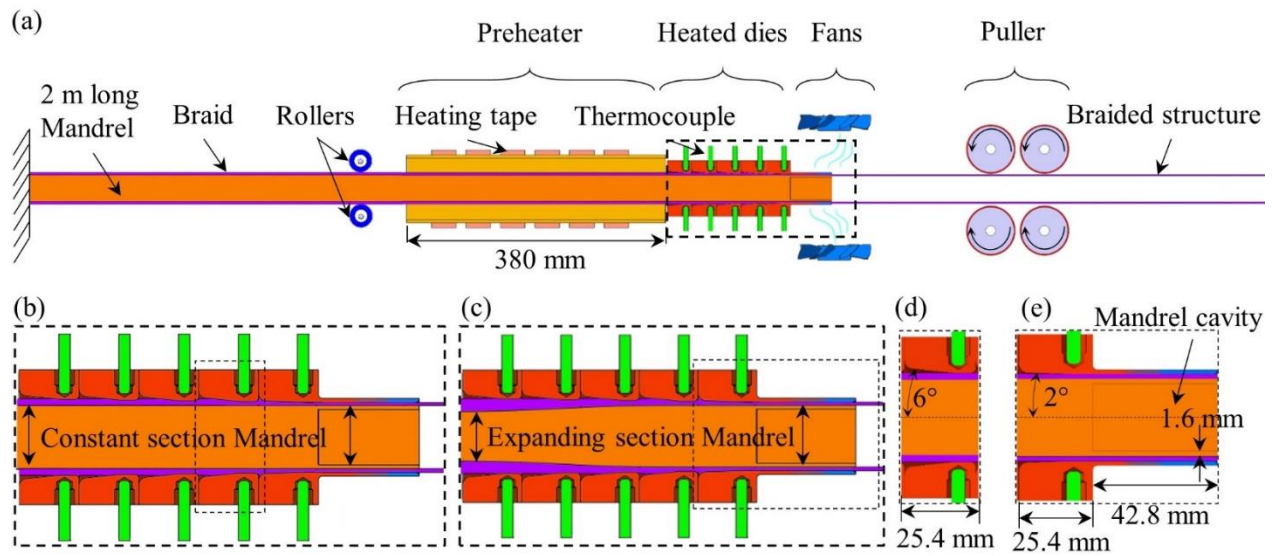


Figure 6-5. (a) Pultrusion apparatus used for the discontinuous braid-trusion manufacturing. (b) Cross-sectional view of the dies and mandrel set up for the external consolidation only. (c) Cross-sectional view of the dies and mandrel set up for the internal consolidation. (d) Cross-sectional view of one pultrusion die. (e) Cross-sectional view of the cooling die.

### 6.4.5 Characterisation

Samples were taken from each braid-truded structure for the evaluation of both internal and external surfaces. The rectangular structures were cut with a precision saw, then polished with a 320-grit to improve fibres' visibility. The images were analysed under a microscope (VHX 7000, Keyence). For each braid-truded structure, a sample was cut at the cross-section for impregnation

assessment. The samples were embedded in epoxy resin (Epofix, Struers) then polished and analysed under a microscope using 200x magnification.

## 6.5 Results and discussions

Table 6-4 presents the braid pitch ( $p$ ) and outside braid circumference ( $C$ ) for both layers for cases  $R_m = 1$  and  $R_m = 0.9$ . The average measured pitch of layers 1 indicated a difference of 5 mm on the braiding machine settings. The standard deviation on three measurements was above 5 mm. The average measured pitch of layers 2 matched the braiding machine settings of 140 mm. The braids outside perimeters of the first layers of each case do not match the modelled  $C_{in}$  of the second layer of each case as per Table 6-3. The outside perimeters of the first layers were  $103 \pm 7$  mm for case  $R_m = 1$  and  $98 \pm 8$  mm for case  $R_m = 0.9$ . The modelled  $C_{in}$  for layer 2 per Table 6-3 were 84.3 mm and 75.9 mm, respectively. Moreover, the measured outside perimeters did not account for the UD welded on the first layer before over-braiding the second layer. The discrepancy is explained by the rather simple modelling of the yarn and braids before pultrusion. This highlights the need for a more advanced modelling method for the braid before pultrusion.

Table 6-4 Pitch and circumference measured on Braid 1 and Braid 0.9.

| Case        | Layer | $p$ (mm)    | $C$ (mm)    |
|-------------|-------|-------------|-------------|
| $R_m = 1$   | 1     | $110 \pm 7$ | $103 \pm 7$ |
|             | 2     | $140 \pm 4$ | $127 \pm 7$ |
| $R_m = 0.9$ | 1     | $110 \pm 5$ | $98 \pm 8$  |
|             | 2     | $140 \pm 6$ | $123 \pm 7$ |

Figure 6-6 presents both braid-truded structures. Braid-truded structure  $R_m = 1$  was pultruded with the constant section mandrel. Braid-truded structure  $R_m = 0.9$  was pultruded with the expanding mandrel. Braid-trusion of 2 m long hollow thermoplastic composite structures using a multi-die pultrusion system was achieved. At the end of the pultrusion of case  $R_m = 1$ , the structure began to exhibit a stop-and-go motion due to slippage occurring between the braid-truded structure and the rollers of the puller. It was also observed an accumulation of a larger diameter braid at the die entrance. These observations support the braid-trusion design methodology introduced above. As shown in Table 6-3, for the braid designed with  $R_m = 1$ , the measured braiding yarn lengths on the CAD models were larger before than after pultrusion, such that  $L_{CBY,in} > L_{CBY,out}$  and  $L_{CCBY,in} > L_{CCBY,out}$ . Moreover, the actual braid circumferences (See Table 6-4) were larger than

the modelled ones (See Table 6-3) indicating an even larger actual  $L_{CBY,in}$  and  $L_{CCBY,in}$ . The start-and-stop motion is attributed to the extra length of braided yarns that are compressed into the dies. The extra length probably created wrinkles and elevated friction resistance, as well as material accumulation at the die entrance. If the pultrusion with a constant section mandrel was continuous, the accumulation of braid at the entrance of the dies would have led to its interruption. On the other hand, the pultrusion for case  $R_m = 0.9$  was done without stop-and-go, nor slippage in the puller, nor interruptions.

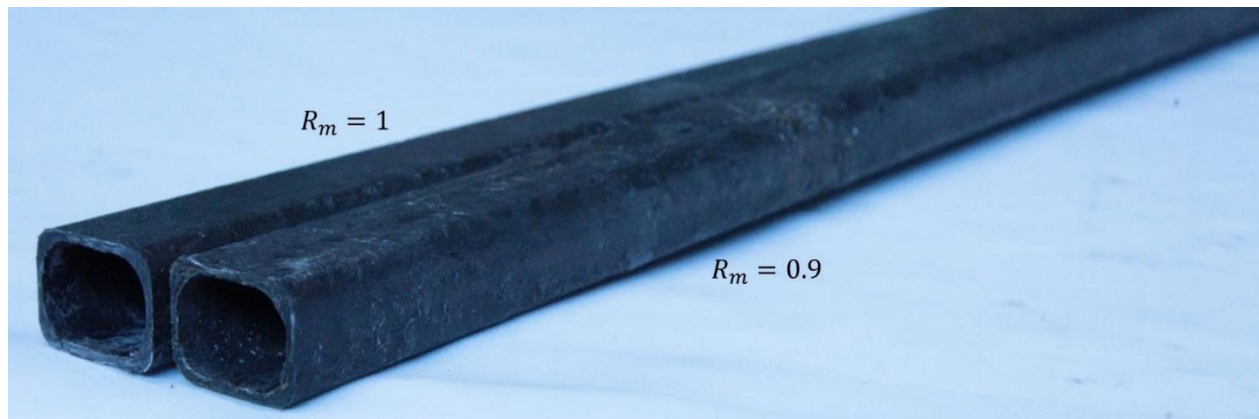


Figure 6-6. Braid-truded structures for the cases  $R_m = 1$  and  $R_m = 0.9$ .

The top two images of Figure 6-7 shows the external (a) and internal (b) surfaces of braid-truded structure for case  $R_m = 1$ . Figure 6-7 (a) shows waviness and squashed fibres at the surface. The braid structure is barely recognizable. The braid pitch after pultrusion could not be measured. Waviness appeared less prominent on the internal surface shown in Figure 6-7 (b). The internal surface's measured braid pitch was  $154 \pm 2$  mm, which was 44 mm longer than the pitch of  $110 \pm 7$  mm measured before pultrusion on layer 1 for case  $R_m = 1$  (See Table 6-4). A longer pitch indicates that braiding yarns slid onto the axial yarns during the passage into the dies. It also explains the braid accumulation at die entrance during pultrusion. These observations confirm that it is not recommended to use a constant mandrel size before and after pultrusion for braid-trusion of tubular structures using bulky hybrid yarns such as the GF/PET DREF yarns used in this study. The bottom two images of Figure 6-7 shows the external (a) and internal (b) surfaces of braid-truded structure for case  $R_m = 0.9$ . Both images show a braided structure that had a repeatable pattern and hardly any apparent fibre waviness. The measured pitches on the braid-truded structure

for case  $R_m = 0.9$  was  $143 \pm 4$  mm on the external surface and  $119 \pm 3$  mm on the internal surface. When accounting for the standard deviations of the measured pitches before (See Table 6-4) and after pultrusion, it is most probable that the pitch did not change during pultrusion. These rather similar pitches before and after pultrusion indicated that the braided yarns did not slip onto the axial yarns during pultrusion. This is also supported by the fact that no braid accumulation and no stop-and-go motion was observed during pultrusion of the case  $R_m = 0.9$ . The fibre waviness reduction indicated on the braid-truded structure for case  $R_m = 0.9$  compared to case  $R_m = 1$  was attributed to the expanding mandrel. As the PET fibres melted in the DREF yarns, the GF loosened, and the expanding mandrel exerted tension on the GF present in the BY yarns. Based on the observations above, it can be supposed that the expanding mandrel approach in a braid-trusion of hollow thermoplastic composites effectively reduced fibre waviness.

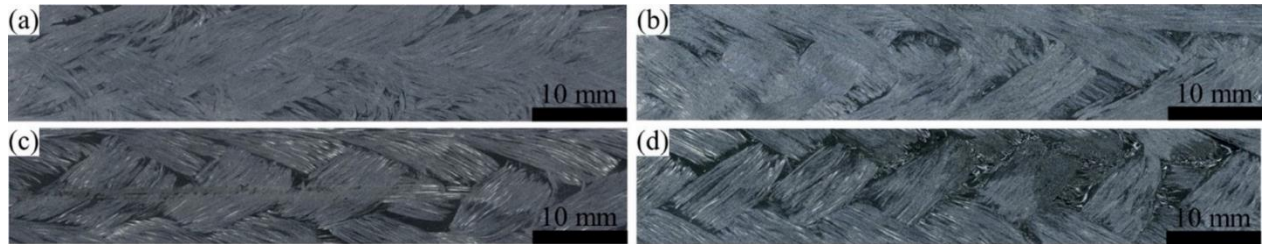


Figure 6-7. (a) External surface of the braid-truded 1, (b) internal surface of the braid-truded 1, (c) external surface of the braid-truded 0.9 and (d) internal surface of the braid-truded 0.9.

Figure 6-8(a) and (b) displays the cross-sectional microscopic images of braid-truded structure the cases  $R_m = 1$  and  $R_m = 0.9$ , respectively. Black areas represent voids. In general, both structures presented a good impregnation quality. Enlarged images taken from the corners of both braid-truded structures showed unimpregnated fibre tows. These unimpregnated fibre tows are the ones composing the UD yarns. The explanation for this difference in impregnation between UD yarns and braided ones may reside in the different axial (*i.e.*, along the pultrusion direction) and radial (*i.e.*, along the tow radius) permeabilities of both regions. More investigation should be done to explain the lower impregnation of the UD compared to the braided yarns.

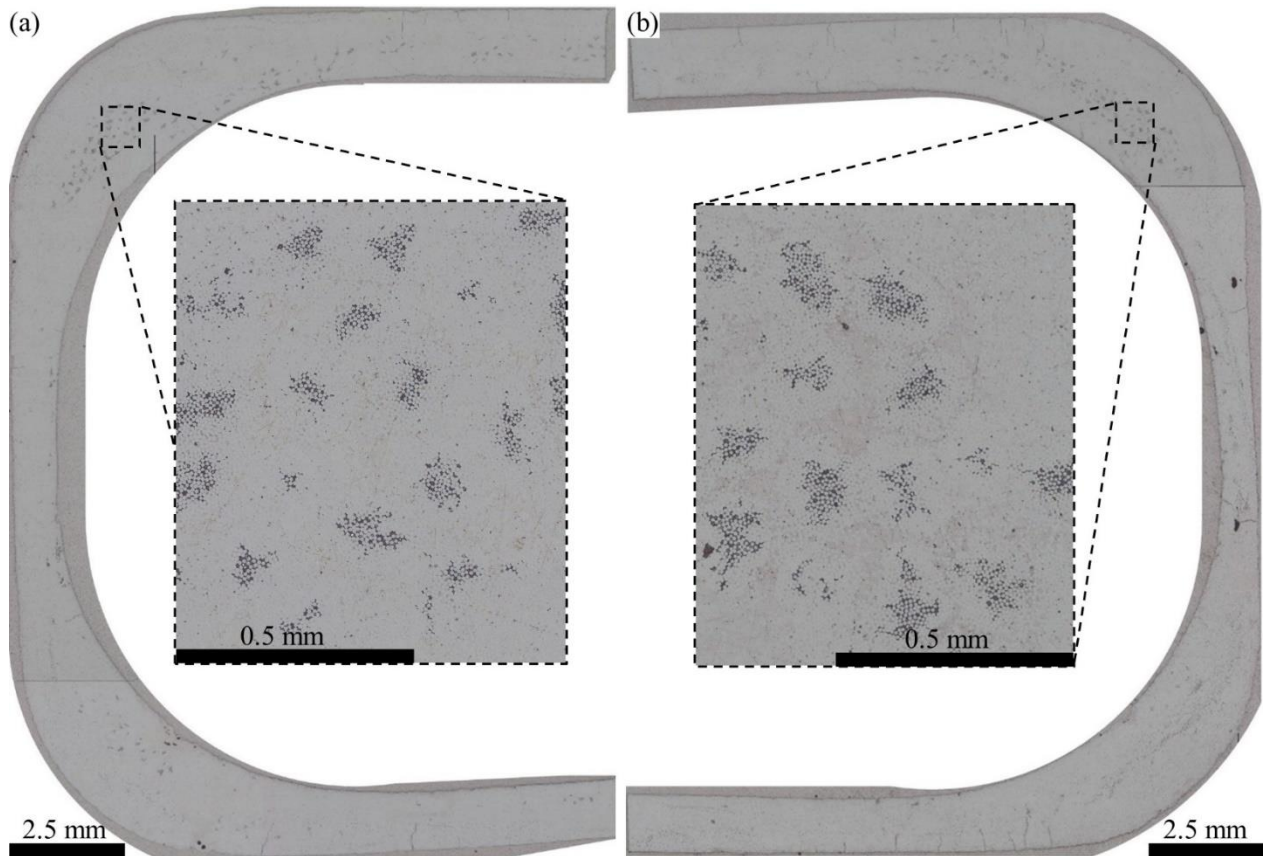


Figure 6-8. Cross-sectional microscopic images of (a) braid-truded structure 1 and (b) braid-truded structure 0.9.

## 6.6 Conclusion

This study proposes and demonstrates a braid-trusion design methodology for hollow structures. The methodology makes use of simple CAD models the braid before and after pultrusion. To achieve a successful braid-trusion of tubular structures, it was suggested that the BY lengths before and after pultrusion needed to be equal. This braid design methodology shows that an expanding section mandrel ensures that the braid adjusts for its deformation while passing through the pultrusion dies. Braid-truded hollow thermoplastic composite structures using a multi-die pultrusion system were manufactured as an example. The absence of major braid deformation at the die's entrance of the braid-truded structure with an expanding mandrel indicated that the braid design was adequate. However, to achieve better alignment between the design and experimental outcomes, a more advanced modelling method for the braid before pultrusion is required. Due to the lack of existing models on this subject, this model serves as an initial exploration, paving the

way for future advancements. Subsequently, despite its simplicity, the model demonstrated promising results, suggesting that with refinement, continuous braid-trusion of hollow composites with an expanding mandrel could be feasible. A precise fibre waviness assessment by CT scan will soon be conducted to measure the actual length of the BY yarns after pultrusion and compare with the designed length. Nonetheless, the design procedure developed in this study could help with the production of high-performance braid-truded thermoplastic composites across various fields.

### **Credit authorship contribution statement**

M. El-Jakl: Conceptualization, Design, Methodology, Investigation, Analysis, Writing – original draft.

L. Laberge Laberge: Conceptualization, Design, Methodology, Writing - Review & Editing, Supervision, Project administration, Funding acquisition.

### **Declaration of Competing Interest**

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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## CHAPITRE 7 DISCUSSION GÉNÉRALE

Les travaux présentés regroupent les trois aspects fondamentaux pour la fabrication de bâtons de hockey en composite thermoplastique par pultrusion tressage.

La première étude se concentre sur la mise en œuvre du fil hybride ‘DREF’ dans la pultrusion thermoplastique. L'originalité de cette étude réside dans sa première prise en compte de fils DREF comme matière d'entrée durant la pultrusion de composites thermoplastiques. Les résultats obtenus illustrent une différence majeure de la réaction à la chaleur entre les fibres de polymère continues et discontinues présentes dans les fils CW et DREF respectivement. L'étude ainsi met en évidence l'amélioration des propriétés mécaniques des produits pultrudés due au fait de la limitation des ondulations de fibres en utilisant le fil DREF. Indiquant donc que les fils DREF présentent un potentiel prometteur pour être utilisés dans la pultrusion thermoplastique de composites à haute performance dans divers domaines. Cette étude ouvre des perspectives pour d'autres opportunités de recherche, telles que l'exploration de la modélisation et d'autres méthodes de fabrication de composites, mettant en évidence l'utilisation du fil DREF. Ce matériel d'entrée a été utilisé pour tous les travaux réalisés dans cette thèse. La réalisation de l'objectif 1 a permis d'établir les bases nécessaires à la poursuite des objectifs. Cela a permis d'explorer le comportement du fil DREF en moulage par compression ainsi que le tressage. La deuxième étude présente une approche de modélisation pour reproduire la pultrusion à travers le moulage par compression. Cette méthodologie a conduit à la conception de quatre scénarios d'analyse de consolidation différents : l'un impliquant le processus multi-filière, et les trois autres utilisant des filières simples. L'un des processus reproduit la longueur effective du système multi-filière. Cela a conduit à la conclusion que l'amélioration de la qualité d'imprégnation n'était pas seulement due à l'augmentation du temps de consolidation, mais également au phénomène de déconsolidation survenant entre les filières de pultrusion. Cette étude, ouvre la voie à la possibilité d'optimiser le nombre de filières successives et leurs géométries respectives avant d'entreprendre une conception complète des filières pour la pultrusion. Cette approche permet d'assurer une rentabilité tout en garantissant la qualité attendue du composite. L'atteinte de l'objectif 2 a conduit à l'application du système multi-filières dans le cadre de l'étude de l'objectif 3.

La troisième étude présente un modèle géométrique de tresses creuses. L'étude propose un modèle simple novateur pour la pultrusion de tresses creuses avec l'utilisation d'un mandrin ascendant. Ce

modèle met en évidence les variations de longueur des fils de tressage avant et après la pultrusion. Ce modèle offre une méthodologie pour la sélection du mandrin approprié pour la *braid-trusion* en fonction de la circonférence d'entrée et de sortie. Le modèle présenté dans cette étude présente un potentiel de contribution à la pultrusion de composites thermoplastiques de tresses creuses à haute performance dans divers domaines. Ainsi, cette étude offre des opportunités de progrès dans les industries nécessitant des structures légères mais robustes, telles que l'aérospatiale, l'automobile, la construction et le sport, tel que des bâtons de hockey.

En regroupant ces trois études, dans le projet de fabrication de bâtons de hockey en composite thermoplastique par pultrusion tressage impliquant trois partenaires industriels, chacun bénéficie de retombées économiques. Pour Filspec, spécialisé dans les matériaux, l'étude du chapitre 4 ouvre la porte à divers marchés offrant des applications variées dans le domaine des matériaux composites. Pour PTI, un fabricant de pultrusion, les études du chapitre 5 et 6 peuvent mener à maîtriser la pultrusion des structures tubulaires tressées. Pour Bauer, fabricant de bâtons de hockey, les études du chapitre 4, 5 et 6, peuvent créer le potentiel de la fabrication automatisée de bâtons réutilisables, réparables et recyclables, localisés au Canada.

### 7.1.1 Détails supplémentaires

La première étude inclut l'utilisation de deux fils, tel que le CW et DREF. Les fibres de polymère utilisés ne sont pas similaires pour les deux types de fils. Par conséquent, des mesures par DSC ont été réalisées afin de comparer les deux polymères. Un calcul de cristallinité a également été effectué pour les deux matières. Ces informations sont présentées dans l'Annexe C.

La deuxième étude inclut l'utilisation d'un moule de compression. Ce moule a été conçu et adapté à une machine d'essai universelle (Acumen 12, MTS). Ce moule est fait en acier AISI 4140 avec un volume total de 40 x 50 x 15 mm<sup>3</sup>. Le plan du moule est présenté dans l'Annexe D. Afin de réaliser les expériences de moulage par compression, la matière a été placée dans la partie du moule convexe (bas). Seule la partie du moule concave (haut) pouvait être déplacé.

## CHAPITRE 8 CONCLUSION ET RECOMMANDATION

La recherche présentée explore le développement de processus de *braid-trusion* thermoplastique. L'objectif de recherche principal de cette thèse était de développer un système de fabrication de bâtons de hockey en composite thermoplastique par pultrusion tressage de bâton de hockey. La réalisation de cet objectif a été accomplie grâce à l'atteinte des sous-objectifs établis au début de cette thèse. La réalisation des trois sous-objectifs est décrite dans trois articles soumis dans des revues scientifiques avec comité de lecture.

L'objectif 1 étant de caractériser le fil hybride 'DREF' pour limiter les ondulations des fibres en pultrusion thermoplastique, a été atteint avec l'étude présentée au chapitre 4. L'étude investigate le comportement sous chaleur des fibres de polymère PET continues et discontinues dans des fils CW et DREF respectivement. L'étude a confirmé que les fibres polymères continues, ont tendance à rétrécir, provoquant l'ondulation des fibres de renforts, contrairement aux fibres de polymère discontinues. Ceci a aussi été confirmé par des images tomographiques des composites pultrudé. Il a été démontré que la tension du fil peut induire une augmentation de défauts de surface. L'étude montre que le fil DREF permet d'obtenir une imprégnation élevée sans aucune application de tension lors de la pultrusion.

L'objectif 2 étant de démontrer l'effet de la pultrusion multi-filières sur la micro-imprégnation des composites thermoplastiques, a été atteint avec l'étude présentée au chapitre 5. En développant une méthodologie de réplcation de la consolidation durant la pultrusion thermoplastique par moulage par compression, plusieurs concepts de filières ont été suggérés. Le processus de moulage a démontré que l'utilisation de quatre filières courtes consécutives avec un angle de  $5^\circ$  induits à la réduction de la force de consolidation et la prolongation de la période de consolidation par rapport à une filière unique longue avec un angle de  $5^\circ$ . Cependant, une filière longue avec un angle de conicité de  $1,05^\circ$  a permis d'obtenir une micro-imprégnation et des propriétés mécaniques similaires à celles du processus à filières multiples. Notant qu'une filière longue a entrée réduite peut potentiellement endommager les fibres, le système multi-filières était considéré comme supérieure. L'étude a démontré que la pultrusion thermoplastiques multi-filières améliore l'imprégnation des composites sans nécessiter un temps de consolidation excessif.

L'objectif 3 étant de développer un modèle pour la pultrusion de tresses creuses en composite thermoplastique en utilisant une approche de mandrin à section ascendante, a été atteint avec

l'étude présenté au chapitre 6. Le modèle développé a montré la variation de longueur des fils de tressage avant et après la pultrusion et la nécessité d'utiliser un mandrin a section ascendante pour remédier à cette variation. La validation du modèle a été faite par la pultrusion thermoplastique a multi-filières de différentes tresses creuses. L'absence de déformation majeure de la tresse à l'entrée de la filière indique que la conception de la tresse était adéquate et que le processus de pultrusion pouvait être effectué en continu.

L'objectif principal de la thèse, soit de développer un système de fabrication de bâtons de hockey en composite thermoplastique par pultrusion tressage de bâtons de hockey a été atteint. Des bâtons de hockey fabriqués par *braid-trusion* de composite thermoplastique ont été produit et testé chez Bauer et ont réussi les tests mécaniques.

## 8.1 Limitations et amélioration futures

**Fil DREF :** Une limitation du fil DREF réside dans la production de fils avec fibres de carbone comme renforts. Dans ce processus, qui implique l'enroulement de fibres discontinues de polymère autour de fils continus, il est essentiel que les fils continus soient fins (nombre de fibres par fils) pour permettre une bonne micro-imprégnation. Pour les fils de verre, cette exigence est accessible et abordable comparé aux fibres de carbone. Les fibres de verres continus sont largement disponibles avec un nombre de filaments relativement faible contrairement aux fibres de carbone.

**Système de refroidissement :** Les études expérimentales présentées ne bénéficiaient pas de système de refroidissement adéquat. Par la suite, cela a entraîné une limitation de la vitesse du processus de pultrusion à l'étude du chapitre 4 et 6. Par rapport à l'étude au chapitre 5, le manque de système de refroidissement adéquat a causé la présence de macro-vides. Le refroidissement inadéquat a induit au collage des deux surfaces des plaques de composite thermoplastique aux surfaces des moules. Lors du retrait du thermoplastique, des macro-vides ont été créé au centre de ces plaques. Par conséquent, une amélioration future serait l'implémentation d'un système de refroidissement permettant de refroidir le composite thermoplastique plus rapidement.

**Caractérisation de l'épaisseur de la tresse avant pultrusion :** L'étude du chapitre 6 propose une méthode expérimentale pour la caractérisation de l'épaisseur de la tresse avant pultrusion. Avec cette méthode, la modélisation ne reproduit pas correctement la tresse avant pultrusion. Une caractérisation plus précise à l'aide d'une méthode de modélisation avancée pour la tresse avant la pultrusion renforcera la confiance dans le modèle proposé.

**Validation de la réduction des ondulations de fibres :** L'étude du chapitre 6 montre la possibilité de réduction des ondulations de fibres grâce au modèle de tresse présenté. Cependant, une validation adéquate n'a pas pu être effectuée en raison des limitations de résolution pouvant être atteintes par la micro-tomographie. Une durée considérable était nécessaire pour obtenir une résolution permettant de caractériser les fibres. La raison pour cette durée prolongée est la taille des bâtons pultrudés. Par conséquent, un travail futur consisterait à réaliser une évaluation précise des ondulations des fibres par micro-tomographie afin de mesurer la longueur des fils de tressage après la pultrusion.

**Validation de la réduction de la force de tirant :** En raison de problèmes techniques, un niveau de bruit élevé a été noté dans les mesures de force de tirant de l'étude expérimentale présentée au chapitre 6. Ce qui a empêché l'analyse des données de cette force. L'étude a proposé que lorsque les longueurs de fils BY sont équivalentes avant et après pultrusion, la force de tirant est plus basse que si ces longueurs sont différentes. En revanche, la validation n'a pas été possible. Pour de futurs travaux, il serait nécessaire d'installer un système de mesure de force adéquat afin de confirmer de manière fiable la diminution de la force de tirant.

**Production continue :** L'étude expérimental présentée dans le chapitre 6 met en évidence un processus de *Braid-trusion* discontinu. Cela est dû au fait que la machine de tressage n'était pas connectée à la ligne de pultrusion. Cependant, l'étude a démontré la possibilité d'une production continue. Un travail futur consisterait d'intégrer la tresseuse à la ligne de pultrusion thermoplastique.

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## ANNEXE A      Matériau d'entrée extrait de la chambre de préchauffe à la fin des expériences de pultrusion.



Figure A1 Image montrant les fils CW (a) et les fils DREF (b) extraits de la chambre de préchauffe à la fin de l'expérience de pultrusion. Les fils CW présentent un retrait du polymère entraînant des ondulations des fibres, tandis que les fils DREF n'en présentent pas.

Après la réalisation des expériences CW-0N et DREF-0N, les fils ont été tirés de la chambre de préchauffe pour montrer leur disposition. La Figure A1 affiche une image des (a) fils CW et (b) fils DREF extraits de la chambre de préchauffe à la fin de l'expérience de pultrusion. La Figure A1 (a) montre les ondulations des fils CW. Cela indique que lorsque la tension n'était pas appliquée, le retrait des fibres de polymère continues présent dans fils CW s'est produit et a entraîné l'entrelacement et l'ondulation des fibres de verre. La Figure A1 (b) montre les fils DREF avec des fibres de verre droites. Ce qui indique que lorsque les DREF ont été exposés à la chaleur dans la chambre de préchauffe, le retrait des fibres discontinus de polymère n'a pas conduit à l'ondulation des fibres de verre, même en l'absence de tension.

## ANNEXE B      Demonstration of the image processing for the calculation of tow and void area to achieve the impregnation ratio.

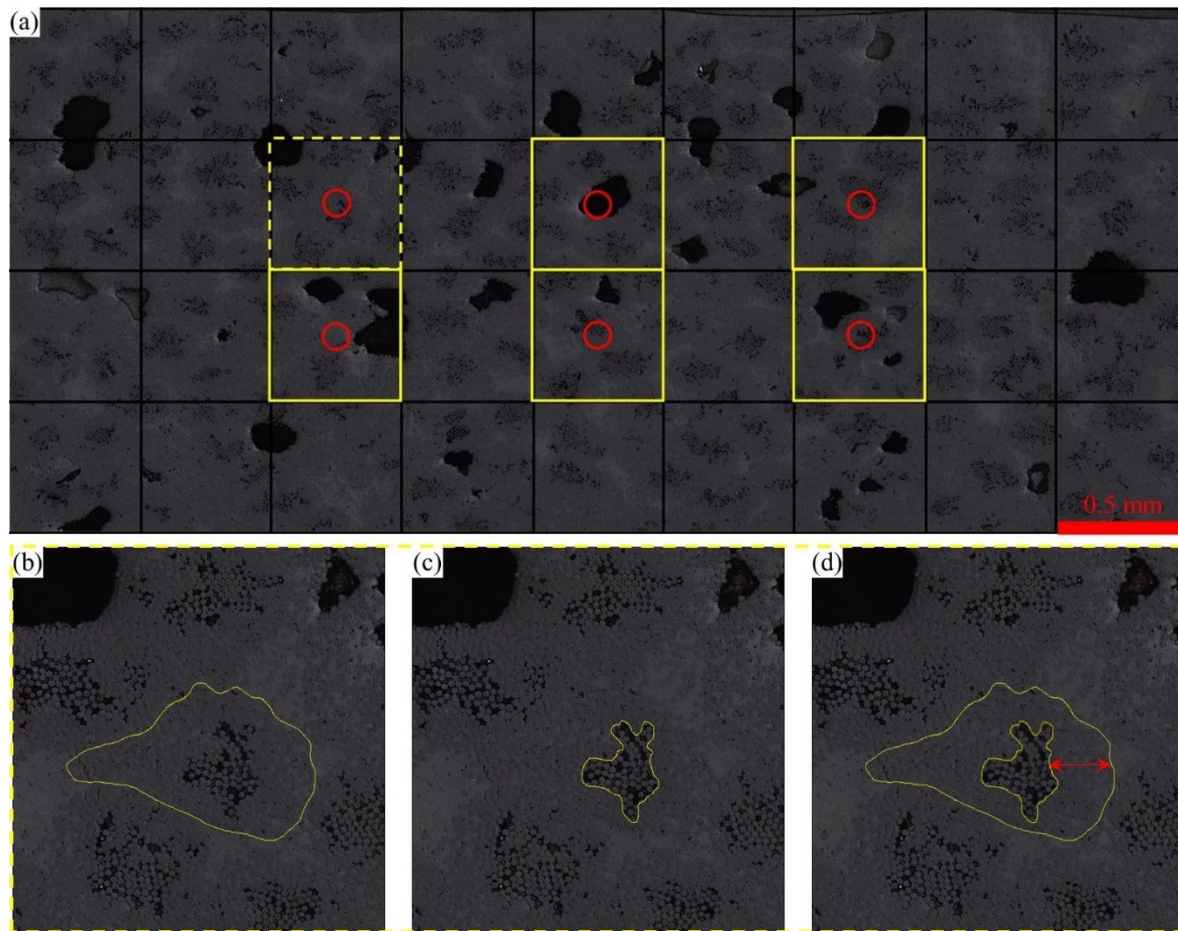


Figure B1 Image montrant la caractérisation de la micro-imprégnation d'un faisceau de fibres. (a) Méthode de sélection de faisceau de fibres à caractériser par traitement d'image pour un coupon issu d'une expérience à filière unique ( $5^\circ$ ). (b) Image de la section 1 montrant l'aire d'un faisceau de fibres (c) Image de la section 1 montrant l'aire non imprégnée du faisceau de fibres. (d) Image de la section 1 montrant l'aire imprégnée du faisceau de fibres avec la flèche rouge.

Figure B1 présente la caractérisation de la micro-imprégnation d'un faisceau de fibres. Figure B1 (a) la méthode de selection d'un faisceau de fibres à caractériser. La figure représente une section transversal d'un coupon provnet d'une expérience à filière unique ( $5^\circ$ ). Un processus de mise à l'échelle standardisé a été appliqué à toutes les images en coupe transversale des coupons caractérisés. Une section de 2 x 4,5 mm a été extraite du centre de chaque coupon. Ensuite, une grille de 0,5 mm a été superposée, divisant les coupons en 36 sections égales. Six sections du centre

ont été sélectionnées (comme indiqué en jaune sur la Figure B1(a)), et le faisceau de fibres le plus proche du point central de chaque section a été choisi pour la caractérisation (comme indiqué en rouge sur la Figure B1 (a)). La figure B1 (b) montre une image agrandie de la section 1 de la Figure B1 (a) montrant l'aire d'un faisceau de fibres ( $A_{tow}$ ). La figure B1 (c) montre une image agrandie de la section 1 de la Figure B1 (a) montrant l'aire non imprégnée du faisceau de fibres ( $A_{void}$ ). Les surfaces ont été obtenues par ImageJ qui a calculé le nombre de pixels inclus dans les sections jaunes. La Figure B1 (d) montre une image agrandie de la section 1 de la Figure B1 (a) montrant l'aire imprégnée du faisceau de fibres avec la flèche rouge. Un taux d'imprégnation ( $R_{imp} = \frac{A_{tow} - A_{void}}{A_{tow}}$ ) a ensuite été calculé.

## ANNEXE C Analyse des mesures de DSC des deux fibres de PET utilisées dans les fils CW et DREF.

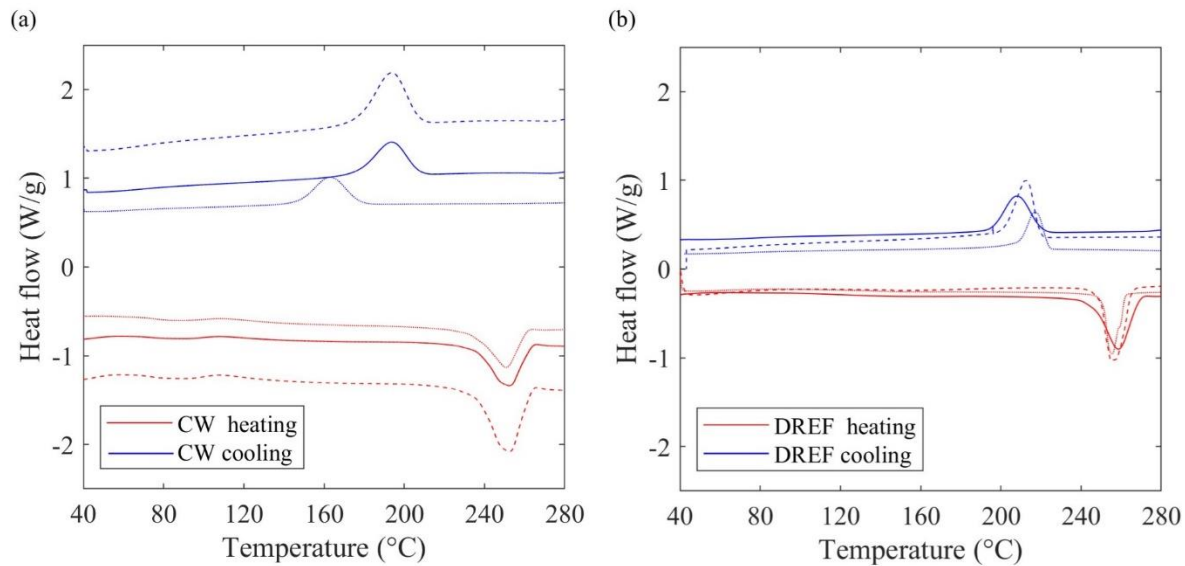


Figure C1 Thermogrammes DSC montrant les courbes de trois échantillons pour le cycle de chauffage (rouge) et le cycle de refroidissement (bleu) (a) pour le PET de CW. (b) pour le PET de la DREF.

Figure C1 montre les thermogrammes DSC pour les échantillons de PET de chaque fil hybride. Trois échantillons ont été testés pour chaque matériau et sont présentés sur la Figure C1 dans différents styles de lignes. Les cycles de chauffage sont représentés en rouge et les cycles de refroidissement sont représentés en bleu. Les échantillons étaient d'environ 4 mg. Le matériau a été soumis à un chauffage puis à un refroidissement à 10°C/min de 40 à 280°C. Le pic endothermique et le pic exothermique représentent respectivement la fusion et la cristallinité à froid du PET. Le point de fusion ( $T_m$ ) était de  $251 \pm 2$  °C et de  $259 \pm 3$  °C pour le PET du CW et le PET du DREF, respectivement. La température de cristallisation ( $T_c$ ) était de  $185 \pm 17$  °C et de  $204 \pm 14$  °C pour le PET du CW et le PET du DREF, respectivement. Le taux de cristallinité  $X_c$  (%) a été calculé selon,

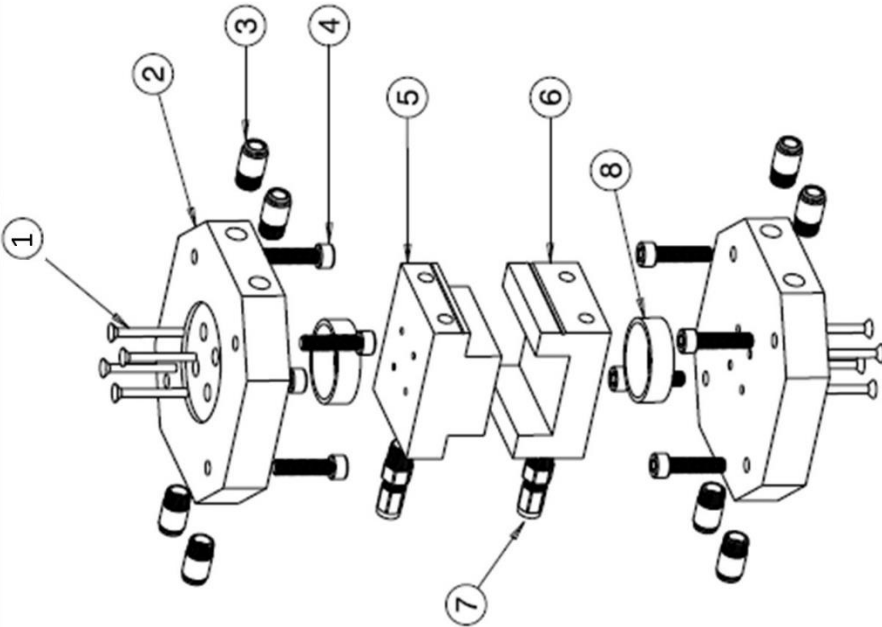
$$X_c = \frac{\Delta H_f}{\Delta H_f^0} \times 100$$

où  $\Delta H_f$  représente l'enthalpie de fusion en J/g et  $\Delta H_f^0$  représente l'enthalpie de fusion théorique du PET cristallin à 100 % et a été considérée comme étant de 120 J/g [1].  $\Delta H_f$  a été déterminé par un logiciel d'analyse (Universal Analysis 2000, TA instruments) utilisant le thermogramme DSC.

Pour le PET du CW, la moyenne  $\Delta H_f$  était de  $52.71 \pm 10.47$  J/g, indiquant un  $X_c$  de  $43.92 \pm 8.73$  %.

Pour le PET du DREF, la moyenne  $\Delta H_f$  était de  $56.58 \pm 5.25$  J/g, indiquant un  $X_c$  de  $47,15 \pm 4,37$  %.

ANNEXE D      Plan du moule utilisé pour les expériences de moulage par compression.

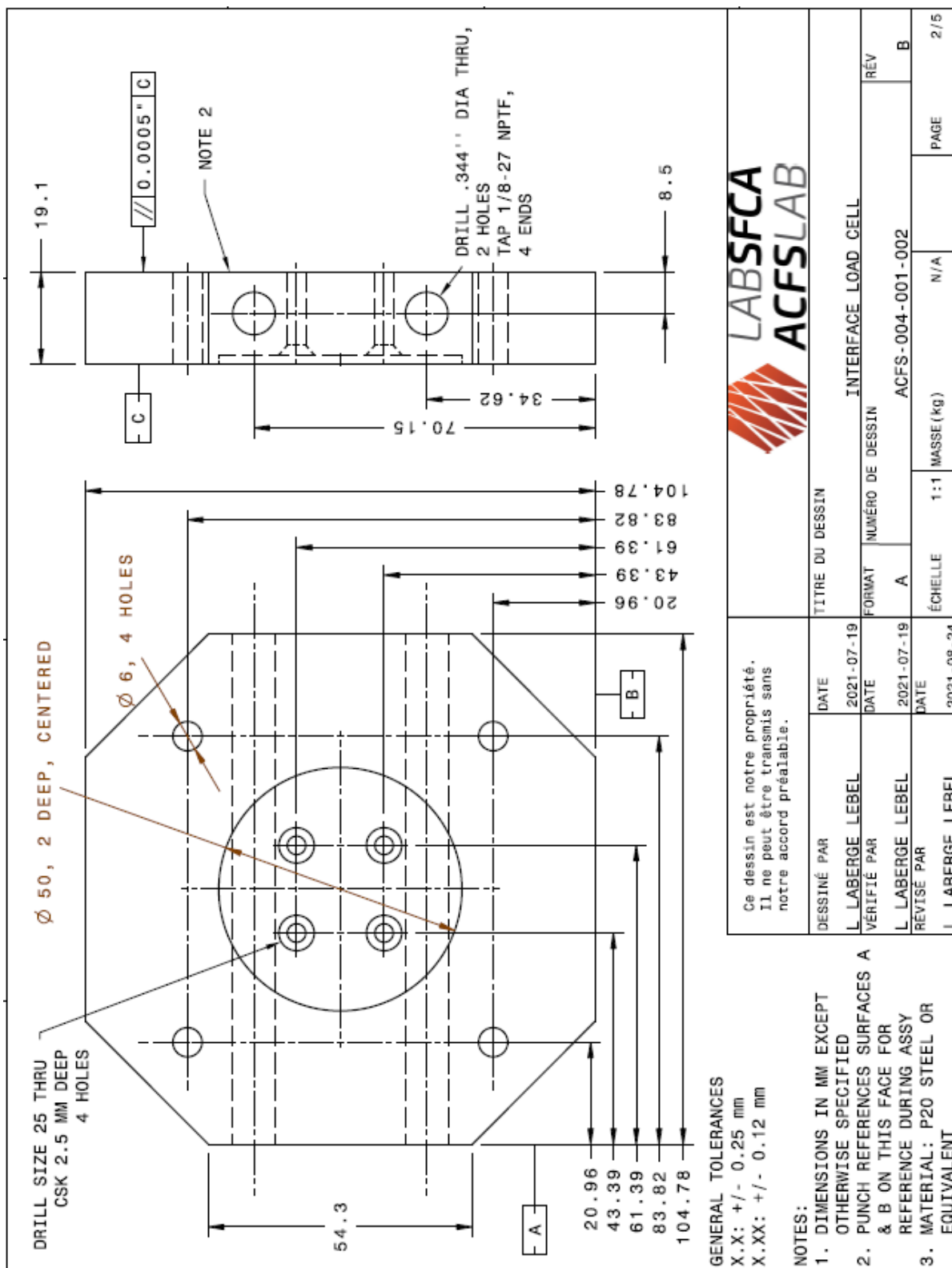


| No | Qty | Name                    | Page    |
|----|-----|-------------------------|---------|
| 1  | 8   | Interface to tool screw | See BOM |
| 2  | 2   | Interface Load Cell     | 2       |
| 3  | 8   | Cooling tube fitting    | See BOM |
| 4  | 8   | Load Cell Screw         | See BOM |
| 5  | 1   | Male Tool               | 3       |
| 6  | 1   | Female Tool             | 4       |
| 7  | 2   | Thermocouple Fitting    | See BOM |
| 8  | 2   | Thermal Barrier Spacer  | 5       |

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notre accord préalable.

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|                 |            |                                 |                  |      |
|-----------------|------------|---------------------------------|------------------|------|
| DESSINÉ PAR     | DATE       | TITRE DU DESSIN                 |                  |      |
| L LABERGE LEBEL | 2021-07-19 | Acumen 12 Material Tooling Assy |                  |      |
| VÉRIFIÉ PAR     | DATE       | FORMAT                          | NUMÉRO DE DESSIN | RÉV  |
| L LABERGE LEBEL | 2021-07-19 | A                               | ACFS-004-001     | B    |
| REVISÉ PAR      | DATE       | ÉCHELLE                         | 2:5 MASSE(kg)    | PAGE |
| L LABERGE LEBEL | 2021-08-24 |                                 | N/A              | 1/5  |

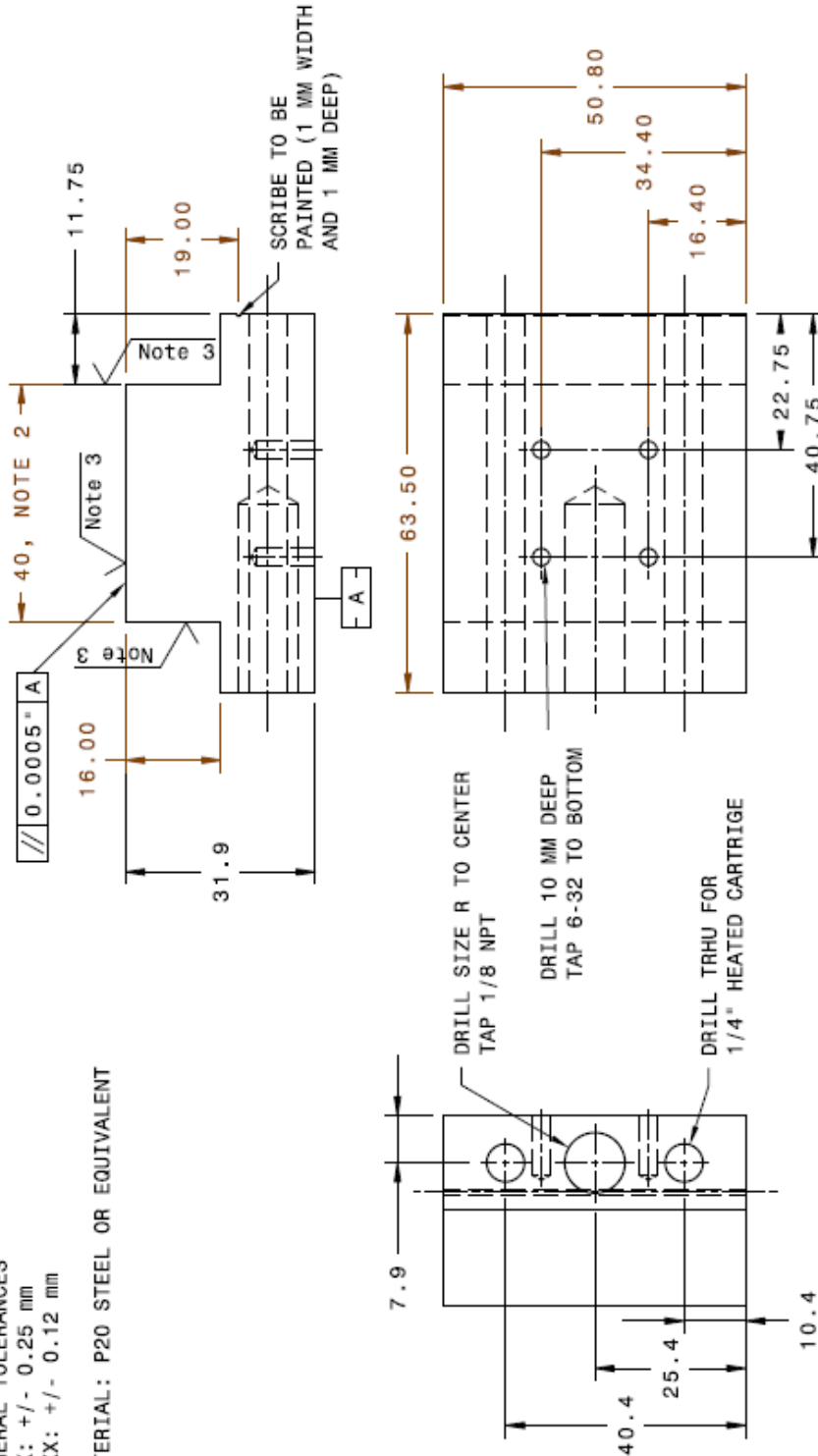


GENERAL TOLERANCES

X.X: +/- 0.25 mm

X.XX: +/- 0.12 mm

MATERIAL: P20 STEEL OR EQUIVALENT



NOTES:

1. DIMENSIONS IN MM EXCEPT OTHERWISE SPECIFIED
2. INSURE ANSI RC3 FIT WITH FEMALE TOOL, CLEARANCE OF 0.0010" TO 0.0026", CONSIDER NOTE 3
3. POLISH SPI A-2 FINISH AND CHROME

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| DESSINÉ PAR      | DATE       | TITRE DU DESSIN |                  |
|------------------|------------|-----------------|------------------|
| L. LABERGE LABEL | 2021-07-19 | MALE TOOL       |                  |
| VÉRIFIÉ PAR      | DATE       | FORMAT          | NUMÉRO DE DESSIN |
| L. LABERGE LABEL | 2021-07-19 | A               | ACFS-004-001-005 |
| RÉVISÉ PAR       | DATE       | ÉCHELLE         | 1:1 MASSE (kg)   |
| L. Laberge Label | 2021-08-24 | N/A             |                  |
|                  |            |                 | PAGE 3/5         |

