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DOI
10.1016/j.jcomc.2024.100550

Publication date
2025

Document Version
Final published version

Published in
Composites Part C: Open Access

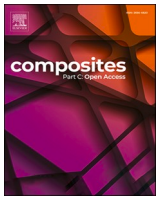
Citation (APA)
Joustra, J., Brans, K., Fernandez Villegas, I., Sinke, J., & Teuwen, J. (2025). Reverse forming thermoplastic composites: Design and process development. *Composites Part C: Open Access*, *16*, Article 100550. https://doi.org/10.1016/j.jcomc.2024.100550

Composites Part C: Open Access 16 (2025) 100550

Contents lists available at ScienceDirect

Composites Part C: Open Access

journal homepage: www.sciencedirect.com/journal/composites-part-c-open-access



Reverse forming thermoplastic composites: Design and process development

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ARTICLE INFO

Keywords:
Reverse forming
Thermoplastic composites
Circular economy
Design
Structural reuse

ABSTRACT

Structural reuse is a promising alternative to recycling of composite materials; it preserves material composition while liberating the materials for reuse in secondary applications. Thermoplastic reinforced composite materials have the potential to expand reuse opportunities by adapting their shape, or reversing them to a laminate blank. In this study, we evaluated reverse forming of glass fibre-polypropylene (GF-PP) laminates by developing a processing method, testing material properties and the effect of three design parameters: forming strain, laminate architecture and material type. Forming strain relates to the deformation mechanism of inter-ply slip, and is imposed through varying the contour depth and bending radius. Laminate architecture relates to resin redistribution, and is imposed by using an orthogonal as well as quasi isotropic layup. Finally, the material type affects both Inter-ply slip as well as resin redistribution, and is imposed by using plain and twill weaves. GF-PP blanks were prepared using a heated platen press and subsequently formed and flattened using convection heating (<165 °C) and vacuum pressure in a novel moulding process. The samples had typical values for flexural strength of 91 - 113 MPa and flexural modulus of 9–16 GPa. Using a Design of Experiments analysis the process was deemed robust for the given boundary conditions. These results demonstrate the feasibility of reverse forming for cases where inter-ply slip is the governing deformation mechanism. The presented reverse forming process and design parameters can be used to create new thermoplastic composite parts, anticipating for structural reuse through reverse forming.

1. Introduction

Fibre reinforced polymers are considered durable, allow for highly optimised structures, and are often used to reduce fuel consumption in transport applications through lightweight design with high strength [1]. Moreover, glass fibre reinforced composites are a key element in the energy transition, because of their use in wind turbine blades [2]. Composites are also increasingly used in the automotive, aerospace and other industries [3]. This results in a correspondingly growing amount of composite waste to be processed at end of use. For wind turbine blades alone, for example, 43 million tonnes of waste are expected globally by 2050 and the recycling capacity is expected to be insufficient to process these volumes [4,5]. Thus, closing the resource loop, the focal point of the circular economy, remains challenging for these composite materials.

Recycling processes for composites can be categorised in mechanical, thermal or chemical treatments. Mechanical recycling downsizes the material in subsequent shredding and grinding steps. The resulting fractions can be sorted on for example density or particle size, allowing separating resin-rich and fibre-rich fractions [6]. Mechanical processing can be a pre-treatment to subsequent thermal or chemical processing. Thermal recycling exposes the material to high (>450 °C) temperatures to decompose the matrix fraction and extract fibres [3]. Depending on process conditions, the resin fraction is either used to fuel the process, or extracted as “pyrolysis oil” [7]. Chemical recycling uses solvents, acids or other solutions, possibly combined with elevated heat and pressure, to dissolve the matrix and extract “clean” fibres [3]. A notable different approach is recycling composite materials into moulding compounds through e.g. vitrimerisation or using cleavable thermosets [8,9]. However, recycling composite materials into moulding compounds or

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https://doi.org/10.1016/j.jcomc.2024.100550

Available online 8 December 2024

separate fibre and matrix fractions comes at the expense of laminate architecture, material composition and most often also fibre length [3,10]. As such, recycling often yields materials with reduced mechanical properties which is reflected in their value [3,11].

Structural reuse on the other hand, recaptures value by directly cutting parts from a decommissioned product. Directly harvesting construction elements, such as panels and beams, maintains the material composition and laminate architecture, thus retaining material quality at relatively low energy investment. Although demonstrated for wind turbine blades [12], reuse of thermoset-based composite parts remains limited by the original product geometry and materials composition. These characteristics cannot be altered, and are therefore decisive for the reuse scenario. Thermoplastic composites (TPC) however present additional opportunities.

Thermoplastic matrix composites such as glass reinforced polypropylene have found widespread adoption due to their versatility, performance and low cost. In addition to established industrial applications, recent publications report on developing hybrid polymer-metal parts and using recycled fibres in additive manufacturing [13,14]. TPC are generally considered recyclable [6], but the challenge lies in maintaining fibre length without compromising mouldability [15,16]. Instead of reprocessing TPC through granulating and remoulding, we focus on re-shaping to preserve fibre length and composite layup. Typically, TPC parts are produced by thermoforming preconsolidated laminate blanks into functional parts [17]. The process may involve multiple sequential forming actions, adapting the part to its final shape [18]. We propose to employ these thermoforming processes to support reuse of TPC parts by adapting the part, or reclaiming its material as laminate blank through reverse forming.

Reverse forming inverses the forming process of a TPC part. Reverse forming is a thermoforming process, where elevated temperature and pressure are used to impose a shape onto a TPC part. But, rather than forming a product, the goal is to revert an existing (used) part into a flattened laminate [19]. This procedure could preserve the material in its original composition including fibre length and layup, thus preserving quality and the value of the material. Despite the potential, reverse forming, or re-shaping, has received little attention up to now .

Initial experiments explored the processing procedures for reverse forming and its effect on material properties. Cousins et al. flattened out a thick, unidirectional layminate with minor curvature and was able to preserving fibre continuity [20]. Obande et al. (2023) flattened out L-shaped samples, finding minor fibre defects, but no significant effect on the acrylic matrix [21]. On the other hand, Sitohang et al. employed the process to intentionally introduce fibre waviness in CF/PEEK samples. So, reverse forming parts while preserving material quality requires development of a processing method, understanding its associated parameters and addressing these in the initial product design.

In this study, we aim to explore the potential of reverse forming and its implementation in product design. To evaluate the potential, we develop a method for reverse forming thermoplastic composite parts, and determine the change in material properties through processing. To design for reverse forming, we determine the associated design parameters and their effect on material quality.

In the following section, we first provide background on the governing deformation mechanisms and defects in thermoforming TPC's. In Section 3 we describe the materials, experimental setup, mechanical testing and analysis procedure. In Section 4, we present the observations on deformation, strength and modulus of reverse formed laminates. In Section 5, we analyse the results by means of a Design of Experiments approach and evaluate the observed failure modes and deformation mechanisms. Here we also elicit insights into how to anticipate for reverse forming by design intent and provide recommendations for further development of the reverse forming process. The insights are summarised in the conclusion, Section 6.

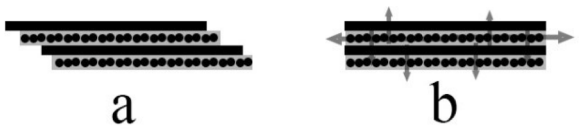


Fig. 1. Deformation mechanisms for single-curved TPC laminates: a)inter-ply slip and b) resin redistribution.

2. Background

To understand the forming process and identify design parameters, we briefly introduce the deformation mechanisms in thermoforming TPC. The main deformation mechanisms in the forming process are inter-ply slip, ply bending and, in case of double curved shapes, intra-ply shear [17,19]. In addition, the imposed temperature, pressure and fibre movement may allow resin redistribution within the laminate [22]. In this study we limit the number of acting deformation mechanisms by focusing on single-curved shapes. This scopes the study to the mechanisms of inter-ply slip and resin redistribution (Fig. 1). The acting deformation mechanisms need to be addressed in the design of the shape, laminate architecture and processing conditions.

The deformation mechanisms allow laminates to conform to an imposed shape through relative displacement between plies and redistribution of resin. Inter-ply slip accommodates the difference in path lengths by creating a book-ending effect [23]. This movement is facilitated by resin redistribution: at elevated temperatures resin viscosity lowers, facilitating shear in the resin-rich interlayer between plies. Inter-ply slip may be hindered by friction between the plies, imposed deformation strains and differences in deformation between adjacent plies [24,25]. Such deformation behaviour is induced by material, design and process parameters like resin melt viscosity, contour depth, laminate thickness, forming speed, -temperature and membrane tensile forces.

Inhibiting deformation mechanisms, such as inter-ply slip and resin redistribution, may lead to excessive loads on the compressive face of the laminate, causing defects like fibre waviness and out of plane buckling [19]. Waviness can cause a significant reduction in compressive strength due to early kinking failure [19]. The strength reduction is related to the extent of waviness but not yet fully understood. Taking into account that a composite's compressive strength is a governing design factor, this additional uncertainty means TPC parts are designed with a high safety factors or even preliminary rejected [26]. In both cases, this leads to overly high consumption of high-end materials.

To test defect formation in a TPC laminate during reverse forming, we conducted a pilot study. A sample of 100 × 100 mm was produced using Glassfiber-Polypropylene (GF-PP) fabric in a quasi-Isotropic layup. The laminate had a wall thickness of 2 mm and was curved with a radius of 40 mm. This semi-cylindrical sample was placed on an aluminium plate and covered with a vacuum foil. The setup was heated to 165 °C and pressure was applied using a vacuum pump. Then, the setup was cooled and the laminate released from the mould. This procedure delivered a reverse-formed, flattened, laminate with notable defects.

After reverse forming, fibres on the compressive face had shifted, introducing both in-plane and out-of-plane defects. In-plane, we

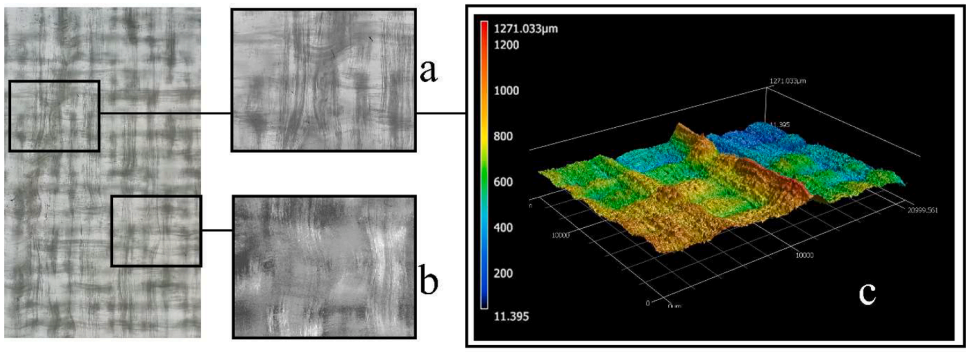


Fig. 2. (a) Buckling, (b) waviness in both fibre directions and (c) height map of buckling area, observed after subsequent forming and flattening of a GF-PP laminate blank.

Table 1
Material properties of polypropylene/glass fibre fabrics used in this study. V_f calculated from sample density.

Material type	Delcotex 86072	Delcotex 86028
Fibre	E-glass 600 tex	E-glass 600 tex
Weave	Plain	Twill
Areal weight	235 g/m^2	490 g/m^2
Threads (weft & warp)	2 threads/cm	4 threads/cm
V_f	58 % ± 3 %	58 % ± 3 %

observed fibre waviness both orthogonal and parallel to the bending axis (Fig. 2). Out-of-plane, we observed a notable buckling line parallel to the bending axis. Similarly, Kiss et al. (2020) observed waviness, yarn slippage and buckling after flattening a formed TPC part. However, these observations were made on double-curved geometries and as such it was unclear to which deformation mechanisms the observed defects could be attributed. Nonetheless, we expect the major challenge for fully restoring the original material properties is to maintain fibre architecture in all process stages.

Based on the given deformation mechanisms and identified design parameters, we aim to develop a reverse forming procedure for thermoplastic composite laminates based on the following requirements:

1. Reverse form an existing part into a flat laminate
2. Maintain material properties; prevent introduction of defects and process degradation
3. Identify the boundary conditions for reverse forming design

3. Materials & methods

To determine design parameters for reverse forming we developed a reverse forming process, analysed reprocessed material properties and evaluated the effect of individual design parameters. The materials, design parameters and the experimental setup are further explained in the following sections.

3.1. Materials

In this experiment we used glass fibre reinforced polypropylene (GF-PP). This material has good impact and fatigue resistance and is mainly used in the automotive and transport industry such as truck bodies and underbody panels [27–29]. The relatively low melting temperature of PP, facilitates experimentation and ensures relatively low energy investments for upscaling. Compared to carbon fibre, glass fibres are less sensitive to fibre misalignment [30]. These properties are expected to make the reverse forming process robust. We used two types of GF-PP prepreg fabrics, produced by Delcotex and supplied by Lange+Ritter (Table 1). The fabrics are woven from an impregnated yarn, enabling high fibre volume fractions and a constant ratio of fibre and matrix [31]. The plain woven fabric's lower density was reflected in its lower thread count per cm., allowing more fibre movement in processing.

3.2. Design parameters

We focused on three design parameters: forming strain, laminate architecture and fabric weave (Table 2). The first, forming strain, is defined by the product shape and as such represents the geometric design space. It is supported by inter-ply slip, which will shift plies over the full material thickness, creating a "bookending" effect [22,23]. If however inter-ply slip is obstructed during forming, compressive loads may cause fibre failure. Therefore, we imposed deformation strains within the maximum strain for E-glass (5 % [32,33]), thus testing at 3 % and 5 % forming strain.

Table 2
Experiment parameters.

#	Description	setting 1	setting 2	Intended deformation mechanisms	Processing factors
P1	Forming strain	2.9 %	5.0 %	Inter-ply slip	Contour depth Forming rate
P2	Laminate architecture	Orthogonal $[0/90]_{nS}$	Quasi Isotropic $[0/\pm45]_{nS}$	Resin redistribution	Laminate stiffness Interlaminar shear
P3	Material type	Plain 235 g/m^2	Twill 490 g/m^2	Inter-ply slip Resin redistribution	Fibre movement Inter-ply friction Ply stiffness

Table 3
Description of blanks, samples and specimen in relation to experiment stages.

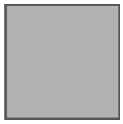
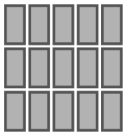
Material form	Experiment stage	Procedure	Process parameters
	Production, Blanks (4 total)	4 laminates produced: 2 Laminate architectures (P2) 2 Material types (P3)	Heating at 8 °C/min., 2 Bar. 165 °C for 10 min., 20 Bar. Cooling at 8 °C/min.
	Forming experiment, Samples (12 total)	3 sample conditions: Pristine Reverse formed Process witness	Heating at 8 °C/min., 1 Bar. At 145 °C, 10 mBar. 165 °C for 10 min., 1 mBar. Cooling at 8 °C/min.
	Testing, Specimen (60 total)	5 test specimen from each sample: Thickness Flexural properties	Heating at 8 °C/min., 1 Bar. At 145 °C, 10 mBar. 165 °C for 10 min., 1 mBar. Cooling at 8 °C/min.

Table 4
Sample design parameters and resulting thickness mean and (standard deviation) in mm . Significant ($\alpha < 0.05$) changes indicated in bold.

Sample	Design parameters			Sample thickness [mm]		
No.	P1: Strain	P2: Laminate	P3: Fabric	Pristine	Reverse formed	Witness
1	2.9 %	CP	Plain	1,87 (0)	1,83 (0,05)	1,88 (0,05)
2	2.9 %	QI	Twill	2,39 (0,01)	2,44 (0,05)	**2,43 (0,01)**
3	5 %	CP	Twill	2,39 (0,01)	**2,48 (0,03)**	**2,44 (0,01)**
4	5 %	QI	Plain	1,85 (0,02)	1,86 (0,01)	1,86 (0,01)

The laminate architecture defines the directional materials properties in use as well as processing. Quasi isotropic layups are often used because of their near-homogenous behaviour in-plane. But, QI laminates need to be formed at a lower forming rate to prevent defect introduction, because of the higher shear stress developed during forming [34]. For forming a cross-ply layup (0/90) could be more suitable as it allows intra-ply shear [19,24].

Both layups were tested, to capture the effect of two major laminate layups on part performance and moldability. Quasi Isotropic followed a $[0/\pm45]_{nS}$ stacking sequence, and Orthogonal $[0/90]_{nS}$ [35]. We chose

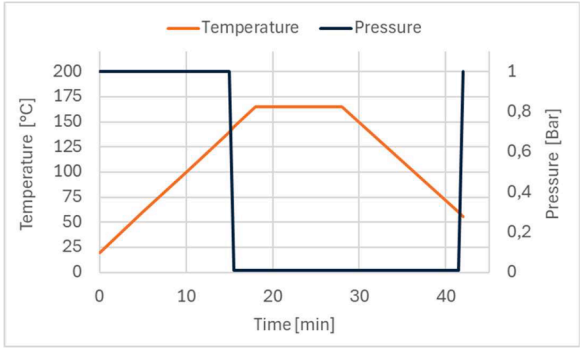


Fig. 3. Temperature and pressure plot of the forming and reverse forming process.

the number of plies to produce a symmetric laminate with a nominal thickness of 2 mm to comply with testing standards [36]. The total number of plies depended on the areal density and ply thickness of the woven fabric, thus arriving at 12 and 8 layers for respectively Delcotex 86072 and 86028.

The material type can also affect inter-ply friction, fibre movement, and individual ply stiffness. The number plies also determined the number of resin interlayers and distribution of resin shear forces. The fabric weave, twill or plain, and thread density affects the laminate drapeability in forming and potentially off-axis fibre movement. Consequently, we included the material type as third design parameter, to evaluate the effect on reverse forming the TPC part (Table 2).

Systematic variation allowed evaluating the effect of specific design parameters on the quality of the reverse formed laminate. The effects of forming strain, laminate architecture and fabric weave were evaluated using a design of experiments approach. Forming rate was kept constant in the experiment to limit the number of design parameters to three. The design parameters were systematically varied using a L4 Taguchi orthogonal array [37], limiting the number of samples to 4 (Table 4). This setup enabled isolating the effect the respective design parameters had on the process through statistical analysis of sample properties [38].

3.3. Experimental setup

We manufactured pre-consolidated blanks into curved material samples and subsequently flattened them. We distinguished three sample conditions: pristine, reverse formed and process witness. The pristine sample was not exposed to the forming process to represent the original laminate blank. We exposed the reverse formed samples to a forming and subsequent flattening process. The process witness samples were co-processed on a flat section of the same mould to collect identical pressure and temperature history. These three conditions allowed observing the effects of reverse forming, while isolating the effects of processing conditions from acting deformation mechanisms.

After the forming experiment we tested sample flatness, thickness, flexural properties and observed the failure mode through visual inspection. Flatness served as an indicator for residual internal stresses after forming, which would manifest themselves as springback after release from the mould. Thickness, measured before and after forming, reflected the consolidation quality and was as such a measure for the processing conditions and potential laminate decompaction.

We tested flexural properties using a four-point-bending setup [36]. A flexural test was preferred over tensile or compression testing because of the nature of the expected defects. Defects such as waviness have little effect on tensile properties, but cause unstable loading conditions in compression, leading to buckling. In a flexural test however, the stable loading condition and simultaneous tensile and compression loading of the test specimen were expected to show the effect of potential defect introduction, while providing reproducible results.

Finally, timelapse photos of the flexural test were used to observe defect formation and failure type. These results enabled evaluating the effects of reverse forming on the laminate quality and flexural properties, as well as relating those to each individual design parameter.

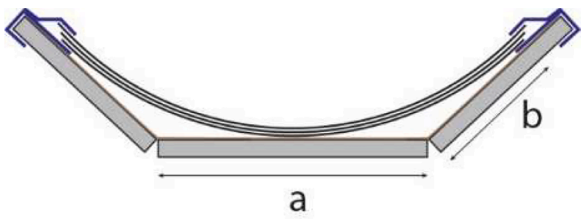


Fig. 4. Three piece folding mould used in flattening, dimensions a and b chosen to inverse strains according to design parameter P1.

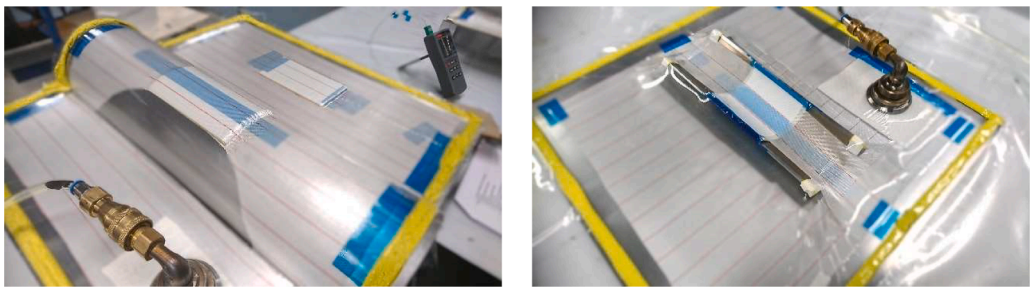


Fig. 5. Process setup for a) forming over a concave mould and b) reverse forming using the folding mould mechanism.

Table 3 shows the experiment stages in relation to the material form.

3.3.1. Press consolidation

We prepared 250 × 250 mm laminates according to the test matrix (Table 4). We chose the laminate layup to have a 2 mm nominal thickness to conform to the testing standard [36]. The laminates were symmetric and balanced to prevent internal stresses which may otherwise have interfered with the forming and flattening processes [22]. Potential variations in areal density or thread count in the fabric were evened out through laminate stacking.

For consolidation, we placed a dry laminate stack in a Joos heated platen press at a 2 bar closing pressure. The temperature was increased from room temperature to 165 °C at 8 °C/min and the press had a dwell time of 10 min at 165 °C. Then, the pressure was increased over 3 min up to 20 bar, and kept constant for 10 min. Following, the material cooled down at a rate of 8 °C/min to 35 °C, after which the consolidated laminate blank was released from the press.

From each blank we then cut three samples, one for each processing condition. The sample size of 125 × 75 mm was chosen to accommodate a minimum of five flexural test specimen of 60 × 15 mm [36]. We cut the samples from the blanks using a water-cooled diamond saw, thus ensuring consistent sample size and squared-off edges. We assessed sample quality through visual inspection on defects such as fibre misalignment, cross-sectional microscopy to check for porosity and thickness measurement to verify consolidation quality.

3.3.2. Forming and reverse forming process

We thermoformed samples from flat to curved using two cylindrical aluminium moulds. The radii of the moulds were calculated to impose the required forming strain. Accounting for inter-ply slip over the full sample thickness, the maximum normal strain was calculated by dividing material thickness by radius (Eq. (1)). This resulted in mould radii of 40 mm and 70 mm, respectively.-

$$\varepsilon = \frac{t}{r}$$

Equation 1 Strain (ε) as function of thickness (t) and mould radius (r)

We positioned the samples on the mould, a 0.5 mm PTFE caul plate on the sample topside distributed pressure to facilitate even consolidation and a good surface quality. The stack was covered with a release- and breather cloth and sealed off with a Wrightlon 5400 nylon bagging film connected to a vacuum pump. For forming a flat sample over a concave mould, we were cautious to prevent bridging of the foils and leaks occurring in the corners. For reverse forming we ensured the foils would not obstruct the flattening process and fold-out evenly across the sample. Fig. 5 shows the process setups. During each experiment, we monitored the pressure to ensure no leaks occurred. When taking the mould from the oven, we inspected the setup under vacuum, to verify that the foils behaved as intended.

The setup was heated in a convection oven and laminate temperature was monitored using a thermocouple type K on the process witness sample. We then placed the assembled mould in a convection oven, set to increase temperature with 8 °C /min (Fig. 3). When the laminate started to soften at 145 °C, we lowered the vacuum-pressure to 10 mbar to impose forming pressure on the material. Heating continued up until 165 °C, at which the laminate was left to dwell for 10 min to allow relaxation of internal stresses, and maintain laminate consolidation [39]. The laminate then cooled down at -8 °C/min until 60 °C, and the formed parts were released from the mould.

Finally, we reverse formed the formed parts. To maintain fibre alignment during the reverse forming process, we applied membrane tensile forces using a folding mould mechanism (Fig. 4). The mould consisted of three aluminium strips, connected lengthwise by PTFE-tape, dimensional drawings are included in Supplement 1. The PTFE tape acted as hinge and ensured clean release after processing. The mechanism was dimensioned to inverse the strain during initial forming. The strain imposed by the folding mechanism was calculated by dividing sample arc length by the mechanism width. The blank was clamped all along the upper edges of the folding mould using 3M 8991 tape, thus fixating both opposing sides of the laminate. The tape remains intact during processing, but allows the laminate to slide out of the clamps in case of excess membrane tensile loads. Again, a PTFE caul plate was placed on top to distribute pressure and a good surface quality.

Reverse forming occurred under the same process conditions as forming. The mould and sample were placed onto an aluminium base plate, covered with release and breather cloth and sealed off with nylon bagging film. The setup was heated in a convection oven and connected to a vacuum pump, following the same temperature and pressure parameters as in the forming process. The flattening process reproduced the forming process conditions of temperature and pressure, but inversed the forming direction while applying membrane tensile forces.

3.3.3. Testing

The specimens for flexural testing were prepared according to ISO14125. We cut five test specimen from each sample using a water cooled diamond saw. Before proceeding to flexural testing, we measured the thickness of all specimen using a Mitutoyo micrometer.

We tested the flexural properties of the test specimen in a four-point bending test (Fig. 6). The advantage of a four-point setup over a three point bending was the absence of compression on the laminate surface in the area that was most prone to deformation and defect formation: the sample centre. The flexural tests were performed at a Zwick-100 universal static bench equipped with a 500 N loadcell, following the ISO14125 testing standard. The four-point bending test setup had a support span of 45 mm and a load span of 15 mm, both the supports and loading members had a 6 mm diameter. The specimen were placed maintaining orientation from reverse forming; the compression side (mould-side in Fig. 4) corresponded to the bottom side in the flexural test. The testing speed was calculated from the sample thickness. We took timelapse pictures to observe defect progression in the specimen during the test (Fig. 7). Taken every 5 s, the intervals corresponded to 0.2 mm vertical travel of the loading member. Using these cross-

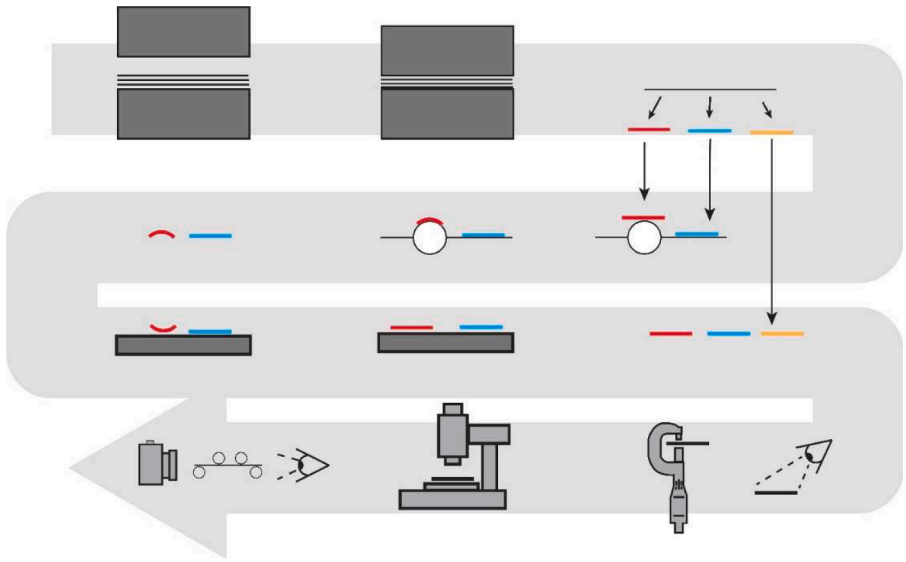


Fig. 6. Experimental procedure with three samples: reverse formed (Red), process witness (Blue) and pristine (Orange).

sectional photographs we identified the failure modes according to ISO14125.

We calculated the modulus between 0.05 and 0.25 % strain in accordance with ISO14125. For flexural strength, we considered 90 % of the maximum stress within the region of conventional deflection (1.5 times sample thickness, [36,40]). This point represents the elastic limit, after which the material transitions from elastic to plastic deformation, causing irreversible deformation to the laminate. This makes the 90 % limit more relevant to practical applications.

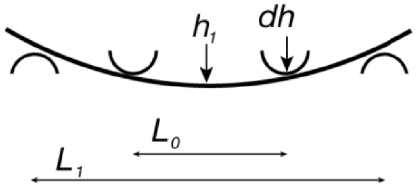
The midpoint deflection was calculated from the loading member travel using the geometric relation in Eq. (2) [41]. Where h_1 is the specimen midpoint deflection, *dh* is the loading member travel, L_0 is the loading member span, and L_1 the support span. This relation assumes circular deflection of the sample under the imposed load, which holds for small deflections.

No adverse effects of repeated heating, such as discolouration of the laminate were observed. The mould (bottom) side showed minor imprints of the folding mechanism in the resin. Visual inspection did not reveal apparent surface defects such as waviness or void formation. The fibre stack returned back to square edges, the edges showed chamfering due to resin squeeze flow and the vacuum-bagging pressure constraint (Fig. 8). These edges were trimmed-off when preparing the test specimen for testing.

To assess the effect of reverse forming on material properties we compared the test results of the reverse formed and process witness samples to those of the pristine laminate. We used a statistical T-test used to check for significant ($\alpha < 0.05$) changes in thickness, flexural strength and flexural modulus. This analysis indicated whether reverse forming significantly changes the mechanical properties of the laminates.

Equation 2 Midpoint deflection

$$h_1 = dh - \left(\frac{dh}{1 - \left(\frac{L_1^2}{L_0^2} \right)} \right)$$



4. Results

Following the test matrix we produced four blanks which were cut into three samples each for the processing conditions, adding up to twelve samples in total. The reverse formed samples were thermoformed over cylindrical moulds and subsequentially flattened. The process witness samples were co-processed on a flat section of the same mould. After forming (Fig. 5a), the reverse formed sample curvature matched that of the mould and we observed no spring-back. Then, after flattening (Fig. 5b), the samples showed no spring-back when released from the mould. In both instances, the sample top surface was smooth owing to the PTFE caul plate.

Table 4 lists the laminate thicknesses of the samples. Variations in thickness and mechanical properties between samples 1–4 can be directly attributed to the systematic variation of design parameters. Samples 1 and 3 have a higher strength and modulus than samples 2 and 4, which directly relates to their CP and QI laminate architectures (P1). Sample 2 and 3 have a higher thickness than sample 1 and 4, which can be attributed to the type of material (P2) and the number of layers in the laminate to fulfil the symmetry and balance requirements. To discern the effect of the reverse forming process on sample properties, we need to compare the results of the three processing conditions: pristine, reverse formed and process witness.

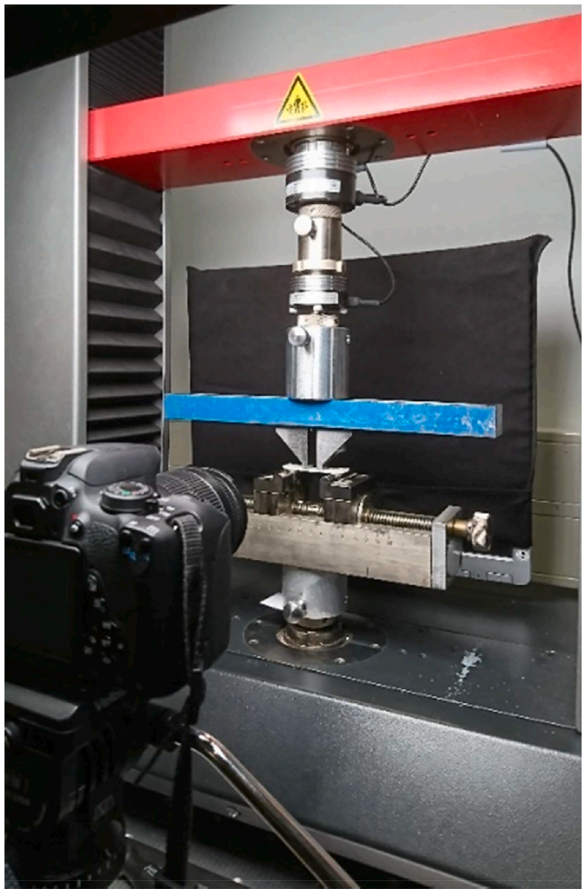


Fig. 7. Four-point bending test setup with photo camera.

4.1. Processing and reverse forming effects

Fig. 9 shows the stress/strain curves for the four samples in three processing conditions: pristine, reverse formed and process witness. For all values, we calculated the average over 5 test specimen. The curves for individual test specimen are included in Supplement 2. For each sample, the curves show high alignment in the elastic region and elastic limit, indicating preservation of material characteristics after multiple forming actions. The plastic behaviour shows some variation, with more ductile behaviour for sample 2. For consistency, and as relevant parameters for product design, we focus on evaluating the flexural modulus and 90 % ultimate strength.

Fig. 10 and Table 5 show the strength and modulus of the four samples in the three processing conditions. Overall, the standard deviations remain close to 10 % of the calculated means, except for the flexural modulus of pristine sample 3 (12 %). This generally small scatter in results allows for good comparison between the samples as well as their processing conditions.

Fig. 8. Cross-section of the laminate after subsequent forming and reverse forming.

To evaluate the effect of reverse forming, we compare the reverse formed to the pristine samples. For thickness we observe significant, yet minor changes in sample 3 (4 %) (Table 4). Regarding flexural properties, reverse formed sample 2 shows an increase of 8 % in strength, but a decrease of 9 % in modulus. The other samples show no significant change to the pristine condition. Thus, for flexural properties, we observe minor thickness increase, but no significant effects on flexural properties from reverse forming for 3 out of 4 samples. Yet, part of these changes might be attributable to the processing conditions.

To evaluate the effect of the processing conditions we compare the process witness samples with the pristine samples . For thickness we observe small, yet significant, changes in sample 2 and 3 (both gaining 2 %). Considering flexural properties, we observe an increase in strength for three samples. Sample 1 exhibits a large increase (10 %), but also a relatively large standard deviation in comparison to the other process witness samples. For samples 2 and 3, the increase (4 %) as well as standard deviations are relatively small. Still, the flexural modulus remained unaffected by processing for all process witness samples.

4.2. Failure type

Table 6 shows photos of the samples during the flexural test at the onset of failure. The overall dominant occurring defect is matrix cracking under the loading members, which presents itself as a the sudden occurrence of a white area in the laminate. This was often followed by the formation of a buckling line above the cracking point close to the loading member. The observed defects indicate compressive fracture including interlaminar shear [36]. Failure mode occurrence will be further detailed per processing condition.

The pristine samples consistently show matrix cracking under the loading member, indicating compressive fracture including interlaminar shear. In the reverse formed samples we observe both matrix cracking as well as buckling on the compressive face, although the extent of damage varies per specimen. The process witness samples consistently show matrix cracking, in two cases followed by buckling on the compressive face of the test specimen. Overall, while all samples exhibit matrix cracking to some extent, buckling behaviour was most notable in the reverse formed samples.

Overall, reverse forming and processing have a minor effect on the sample properties. The majority of observable changes are smaller than 5 %. Only the flexural properties of sample 2R and strength of 1 W show a notable change, although both remain within 10 % of the pristine value. Next to that, the reverse formed and process witness samples exhibit buckling behaviour more often. These results do not unequivocally indicate to what extent the changes in flexural properties can be attributed to either processing conditions or reverse forming. Nonetheless, the samples largely retain their properties through multiple successive processing actions.

4.3. Design parameters

The effect of each design parameter (forming strain, laminate architecture and fabric type) on changes in flexural strength and flexural modulus was determined using a Design of Experiments approach. The standardised effect of each parameter was calculated using a T-test, where T-values below 2.57 were considered insignificant [38,42]. We visualised the effects and significance of each individual design parameter using Pareto plots.

The systematic variation in sample compositions enabled evaluating the effect of the design parameters on the change in sample properties. The Pareto charts in Fig. 11 show the effect of deformation strain, laminate architecture and fabric type on the change in strength and modulus compared to the pristine values. For both modulus and strength, the imposed strain has the largest effect, followed by fabric

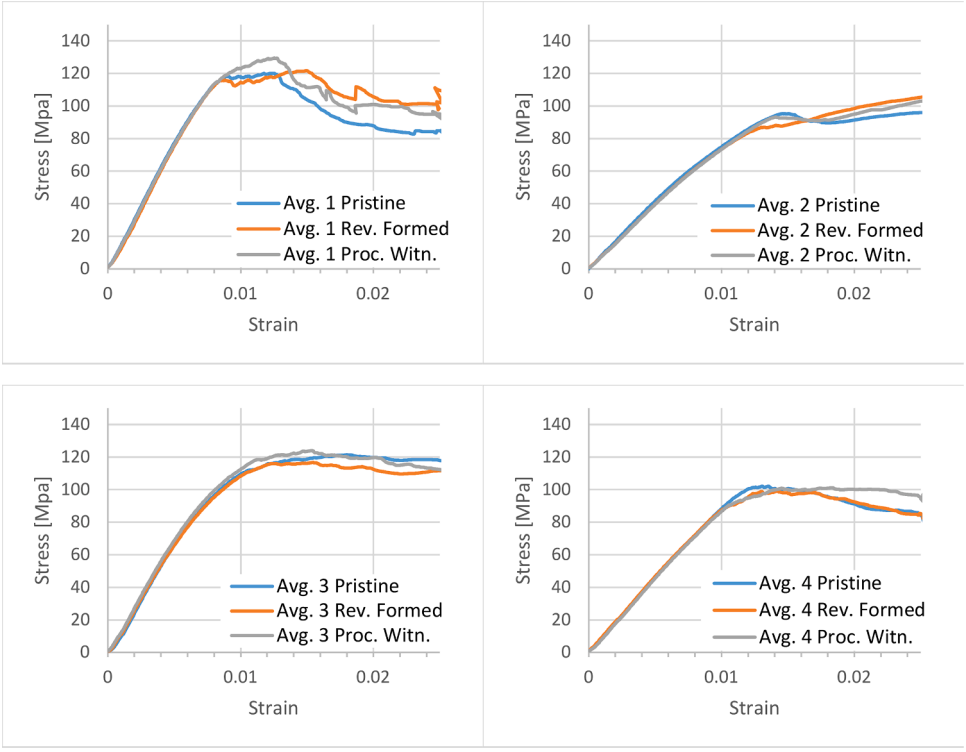


Fig. 9. Average flexural stress/strain curves for Samples 1–4 in three processing conditions.

type and laminate architecture. The magnitude of the effect is larger for strength than for modulus. However, all effects fall below the significance level of 2.57. Thus, even though imposed strain has the largest effect, it still does not contribute to the observed change in material properties.

5. Discussion

None of the three design parameters showed a significant effect on the materials strength and modulus after reverse forming. The imposed forming strain was expected to affect the compressive load on the fibres during forming and flattening, and as such relate to fibre alignment defects such as (in plane) waviness and (out of plane) buckling. The laminate architecture was expected to affect on the ply stiffness and off-axis fibre movement through subsequent forming actions. The fabric weave was expected to affect the laminate drapeability in forming and potentially also off-axis fibre movement, as the plain fabric was much lower-packed. Together, the imposed strain, architecture and fabric were expected to affect the reverse forming process. However, the results indicate that the reverse forming process is robust for the particular (simple) shapes used in this study or for deformation cases involving only inter-ply slip. These results apply to both cross-ply and quasi isotropic laminates with a plain or twill weave and imposed forming strains up to 5 %.

5.1. Deformation mechanisms

In the forming as well as the flattening process, the imposed deformations inflicted both tension and compression stresses on the sample. These manifested themselves in both the laminate as well as in individual plies. In the laminate, the imposed deformation induced inter-ply slip. This deformation mechanism facilitated conforming to an imposed shape and alleviates internal stresses on the individual plies. Indeed, no spring-back was observed after forming, nor flattening.

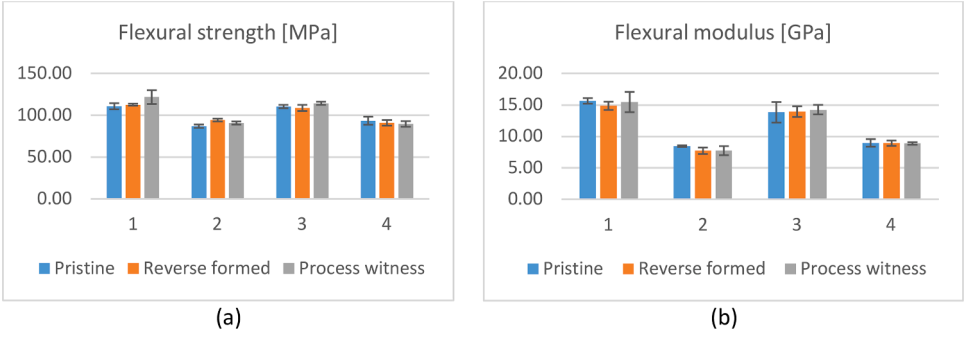
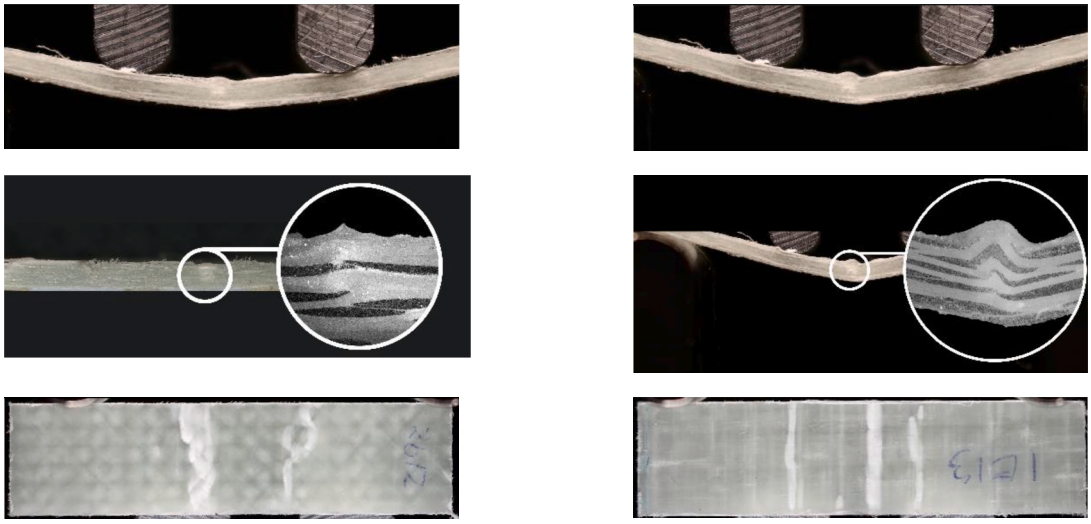


Fig. 10. Mean flexural strength (a) and flexural modulus (b) for samples 1–4 in pristine, reverse formed and process witness condition.

Table 5
Mean and (standard deviation) of strength (a) and modulus (b) for Samples 1–4 in pristine, reverse formed and process witness condition. Significant ($\alpha < 0.05$) changes noted in bold.

(a) 90 % Flexural strength (MPa)			
#	Pristine	Reverse formed	Process witness
1	110,7 (3,7)	112,7 (1,2)	**121,8 (8,3)**
2	87 (1,9)	**94,3 (1,7)**	**90,9 (1,8)**
3	110,4 (2)	108,7 (3,7)	**114,3 (1,9)**
4	93,5 (4,8)	91 (3,3)	89,6 (3,3)
(b) Flexural modulus (GPa)			
#	Pristine	Reverse formed	Process witness
1	15,6 (0,4)	14,9 (0,6)	15,5 (1,6)
2	8,5 (0,1)	**7,7 (0,5)**	7,7 (0,7)
3	13,8 (1,6)	13,9 (0,8)	14,2 (0,8)
4	9 (0,6)	8,9 (0,4)	8,9 (0,2)

Table 6
Failure types observed in 4-point bending test.



	Matrix cracking	Buckling
Pristine	1, 2, 3, 4	3
Rev. formed	1, 3	1, 2, 3, 4
Proc. Witness	1, 2, 3	3, 4

In-ply compressive loads can however still lead to defect formation. The pilot study showed how relatively low forming pressure and unconstrained flattening resulted in formation of waviness and out-of-plane buckling. In the developed flattening procedure, the in-plane compression loads were counteracted by imposing a tensile strain. This setup effectively prevented off-axis fibre movement and defect formation in the reverse forming process.

5.2. Process and design implications

From the deformation mechanisms and failure modes, we can see that the important process parameters for reverse forming correspond to a conventional thermoforming process [29]; temperature, forming force and laminate/membrane tensile forces.

In this study, the temperature for reverse forming was the same as that of the original thermoforming process. This temperature allowed

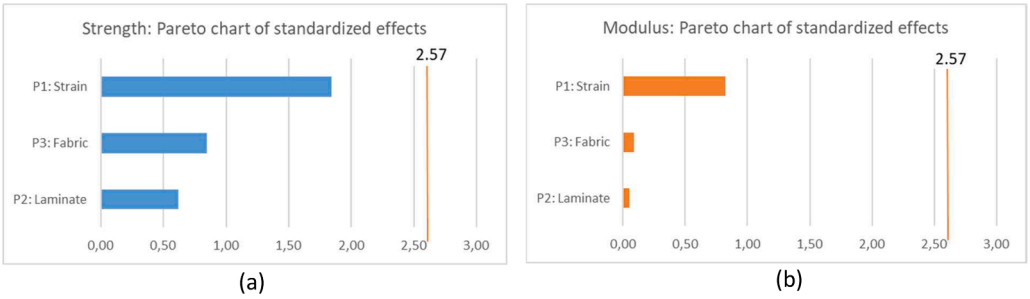


Fig. 11. Pareto chart of standardised effects for the change in a) Strength and b) Modulus. Significance threshold (2.57) indicated.

material deformation and movement and no adverse effects of repeated heating were observed. The forming force and, at the same time, the laminate compaction force in forming and reverse forming were similar but lower than in blank production. This low forming pressure was due to the possibilities regarding test setup. However, such a low pressure can have consequences for the occurrence of buckling, voids or decompaction [43].

Minor decompaction was observed in both reverse formed as well as process witness samples. This is most likely caused by the relatively low processing pressure. During forming and flattening, the applied pressure was a factor of 20 lower than during blank consolidation, allowing the fibre rovings to gradually rearrange to their original, unconsolidated state. As a result, some reverse formed and process witness samples showed a minor (<4 %) increase in thickness compared to the pristine samples. The effect is less pronounced in the reverse formed samples, possibly because the imposed membrane tensile forces partially counteracted the fibre relaxation. On the other hand, the absence of major decompaction indicates that vacuum pressure sufficed to prevent laminate deconsolidation [22]. We did observe strong deconsolidation when a vacuum bag failed in early testing. Then, a sample 3 laminate increased in thickness from 2,4 mm to 3,6 mm. Thus, processing without vacuum pressure does lead to thickness increase and deconsolidation.

Expressing the shape factor as imposed strain allows expanding these results beyond the cylindrical shapes used in this experiment. We expect similar results could be obtained for other laminates and shapes that would involve inter-ply slip within the given conditions. As such, these design parameters can be adopted in designing new thermoplastic composite parts for reverse forming.

The economic viability of reverse forming is challenged by the investment in energy and value of the reclaimed material. Heating and pressurising could use a substantial amount of energy per formed unit. Moreover, materials like GF-PP used in this study are relatively low-cost, which could challenge the return on investment. Although reverse forming has the potential to reduce use of primary raw materials, the energy demand should be carefully monitored to not outweigh the gains made in avoided environmental impacts and recaptured material value.

5.3. Contributions

The goal of the study was to evaluate the potential of forming and subsequent flattening and its implementation in product design. We therefore monitored the material properties through processing. Maintaining material properties is not a given, as demonstrated by the waviness and buckling defects observed in the pilot study on reverse forming. A reverse forming process was developed and a statistical T-test was used to evaluate the change in material properties through subsequent forming and reverse forming, finding that most samples did not show a significant change in properties. The observed changes were all within 10 % of pristine values, indicating that the presented methodology successfully preserves material properties.

The findings in this study contribute to both research and practice in the field of thermoplastic composite reprocessing and design. Earlier studies included a wide range of materials and processing conditions. The processing temperatures in these studies varied according to matrix type, but where Kiss et al. processed GF-PP samples at 220 °C, we successfully formed GF-PP at 165 °C. Processing times ranged from 60 s [19, 43] up to 8 h [20]. The presented method has a total process time of 41 min, which is close to the 35 min reported by [21]. Altogether, the presented method adds a potentially interesting method to the processing options for reverse forming TPC.

The reverse forming process effectively returned a formed component to pristine blank condition, opening up additional reuse opportunities. The folding mould mechanism prevented defect formation and can be readily adapted to comply to the curvature and strain required by the part geometry. The process was robust for the given design parameters, which can thus be used to enable reverse forming of thermoplastic composite parts by design intent. Finally, the observed failure modes add to the knowledge base on failure behaviour of thermoplastic composite laminates.

Reverse forming could stimulate reuse of thermoplastic composite parts and laminates through various circular economy strategies. Flattened out parts could be reused as reclaimed blanks, ready to be thermoformed into another part. This allows parts and materials to be reused outside of their original context. Such reclaimed materials could be used to substitute use of primary raw materials and thus reduce resource consumption. Potentially, the process could be adapted to allow for upgrading of parts, for example, to adapt the style of aircraft or automotive interior panels. This would enable multiple product lifecycles, thus prolonging part and material lifetime. Moreover, subsequent forming and reusing existing parts while maintaining material properties preserves the energy and value embedded in the material.

5.4. Limitations

Compound curvatures have been intentionally excluded from this study to avoid introduction of intra-laminar (Trellis) shear. Double curved laminates introduce a more complex arrangement of forming loads and intra-ply shear, complicating the reverse forming process accordingly. Focusing on single-curved shapes allowed evaluating the process and design parameters for reverse forming of laminates dominated by inter-ply slip.

The used processes lend themselves well to initial lab experimentation, but do not reflect industrial production. We used a quasi-isothermal process where the process time is largely dictated by the thermal inertia of the moulds and the indirect application of heat through a convection oven. This made the process time and energy consuming, both undesirable in an industrial process setting. Also, the clamping force during reverse forming is hard to define for this arrangement and conditions. Based on the tensile strength of the tape and the application in this setup, we find a maximum membrane tensile load of 1270 N, or 5.1 MPa. The precise membrane tensile loads should be defined to industrialise the process.

The material on the other hand, may have benefited from the relatively slow process. In the forming process, the pressure on the laminate was applied close to the glass transition temperature, while the heating continued until melting temperature. In this resin-dominated process, the sample will only deform once the matrix is sufficiently softened. The low forming speed reduced the risk for matrix defects and the 10 min dwell time at melting temperature allowed for healing through self-diffusion [22]. The following low cooling rate facilitated crystallisation in the polymer, benefiting its mechanical properties. The risk of fibre failure was prevented by imposing maximum 5 % normal strain and the dwell allowed for relaxation of shear stresses in the laminate resulting from the imposed deformation as well as thermal expansion.

The study remains inconclusive about the factor significantly affecting the flexural properties of the samples through processing and forming. One reason could be interaction between the selected design parameters. However the chosen experimental design limits analysis of such interaction effects. Interaction effects cannot be calculated from a fractional factorial design, because the interaction is confounded. i.e. the interaction of P1xP2 is confounded with the setting of P3 in this experiment.

5.5. Recommendations

In this study we tested a reverse forming process for TPC parts focussing on three design parameters. Based on the experimental results we expect the reverse forming concept can be further developed in terms of materials, processing and design. Regarding materials, the plain weaving pattern is known to be less compliant to imposed double-curved contours, compared to e.g. a twill weave [44]. To better understand the effect of weaving pattern on forming and subsequent flattening

processes, we recommend further experimentation on increasingly complex geometries.

The tests in this study supported exploring potential for reverse forming in processing and design. To gain more insight into the influence on material properties by reverse forming, we recommend more extensive material characterisation, including impact resistance and fatigue life. To evaluate the effect of compression and tensile loaded faces in the forming process, the samples could be flipped in testing. To detail the failure behaviour, fractures could be further investigated with SEM micrography. To evaluate potential long term effects such as fatigue or cumulative damage, we recommend to test materials that have been subjected to multiple successive cycles of forming and reverse forming.

Glass fibres and polypropylene are both tolerant to processing conditions and imposed deformations strains. We expect that materials with narrower processing and alignment tolerances will affect the process setup and allowable geometries for reverse forming accordingly. In addition, we expect such materials to be more susceptible to material property degradation through reprocessing and thus lower the robustness of the overall process. Glass fibre rovings have a low friction coefficient, which allow them to move in a dry fabric. Such fibre movement is constrained in prepreg materials, and even more so in preconsolidated blanks. This limited freedom of movement in the original part production may have supported the reverse forming process. Conversely, dry fibre placement-based parts (e.g. infusion-based thermoplastics) could be more complex to reshape or flatten when fully cured.

We recommend further development of the heating and pressure parameters in forming and flattening. Concerning heating, non-isothermal (e.g. infrared) heating to reduce process time and energy consumption. However, this presents additional challenges in heat distribution as well as transferring the pre-heated sample to the moulding stage. Increasing processing pressure may lower the risk of defect formation and laminate decompaction. It is expected that optimising these factors will significantly increase the forming rate and future studies may explore the impact of varying pressure levels on material performance.

Reverse forming requires further analysis to determine the forces needed for flattening a product, especially when more elaborate shapes are used. Kiss et al. (2020) reverse formed a pyramid shape by exerting a distributed load on the sample edges to apply membrane tension. Although the shape was successfully flattened, fibre misalignment caused by yarn slippage and decompaction reduced the laminate properties. We used membrane forces to prevent buckling and waviness. Compound curvatures however might need tension forces in fibre directions of the product rather than the bias directions which are used in forming operations. Overall, we recommend investigating the configuration and level of tensile strain required for different laminate configurations.

Introduction of membrane tensile strains should be further developed to gain increased control over ply movement in reverse forming. Depending on shape, the folding mould mechanism could be elaborated to progressively introduce strain in multiple directions. Alternatively, springs could be used to introduce membrane tensile forces [19]. Both solutions need clamping spots or tabs to introduce the membrane forces. Although these could be attached afterwards, using those present on the part itself would be desirable. Clamping spots are usually trimmed off after production [43], but leaving these intact could be a good starting point to redesign a part to facilitate reverse forming.

Reverse forming could be anticipated for in the initial design of the part. Next to accounting for process needs, the design of a part could aim to alleviate compressive loads in the laminate during reverse forming. For example by designing in opposite forms to even out tensile and compression loads in the laminate or limiting deformations by constraining form complexity through e.g. curvature or depth. Next to modifying the part shape, additional layers could be added to change the laminate design.

6. Conclusion

In this study we reversed the forming process of thermoplastic composite laminates while preserving their material properties. Samples with systematically varied design parameters were subjected to a forming and subsequent flattening process. Flexural testing of the samples before and after processing revealed no significant change in flexural strength and modulus. Thus demonstrating the feasibility of reverse forming and the developed procedure for cases dominated by inter-ply slip.

Taking a design of experiments approach enabled analysing the effect of three predefined design parameters on the reverse forming process: imposed deformation strain, laminate layup and fabric type. The imposed strain is a function of the imposed form and laminate thickness, and thus represents the overall part geometry, while layup and fabric type relate to the laminate architecture. All effects fell below the statistical significance threshold. Consequently, the reverse forming process is considered robust for these design parameter settings.

During flattening, we imposed a membrane tensile strain onto the curved laminates which was equivalent to the forming strain. The process effectively enforced inter-ply slip, alleviated or even eliminating compression loads in the plies and thereby prevented formation of defects such as waviness or out-of-plane buckling.

The presented procedure and parameters can be used to anticipate for reverse forming in the design of new thermoplastic composite parts. We expect this will diversify potential reuse applications by allowing parts to be reshaped and reused through multiple use cycles.

CRediT authorship contribution statement

Jelle Joustra: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Karel Brans:** Investigation, Formal analysis, Data curation. **Irene Fernandez Villegas:** Writing – review & editing, Conceptualization. **Jos Sinke:** Writing – review & editing, Resources, Conceptualization. **Julie Teuwen:** Writing – review & editing, Resources, Methodology, Conceptualization.

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.

Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 730456. The funding had no involvement on the study execution and supports open access publication.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jcomc.2024.100550.

Data availability

Data will be made available on request.

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