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Title: Development of high performance recycled carbon fibre composites with an advanced hydrodynamic fibre alignment process

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Abstract: Carbon fibre composites have great potential for vehicle lightweighting but the high cost and environmental impact of their production tends to largely undermine the advantages in non-aerospace applications. Recycled fibre has the potential to significantly reduce both cost and environmental impact but has yet it has not been widely accepted by the composites industry due to reduced mechanical performance in components as well as the difficulties in handling and processing caused by the fluffy discontinuous form which is quite unlike any current material formats which suit existing processing methods. The developed alignment process allows discontinuous random recycled carbon fibre to be processed into tapes with a highly aligned orientation distribution. This allows composites with high fibre content to be manufactured at lower moulding pressures with the added benefit of keeping fibre length degradation to a minimum. To evaluate the effects of process factors on fibre orientation, a two-level full factorial experimental plan was designed. This represents the first time a systematic study of input parameters on final part performance has been published in the open literature. With further improvements to the process, it is shown that it is possible to manufacture a composite achieving high fibre volume content (46%) under 7 bar moulding pressure in an autoclave, exhibiting competitive mechanical properties with almost 100 GPa tensile modulus and over 800 MPa tensile strength.

Research Data Related to this Submission

There are no linked research data sets for this submission. The following reason is given:

No data was used for the research described in the article

Declaration of interests

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

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Development of high performance recycled carbon fibre composites with an advanced hydrodynamic fibre alignment process

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29th April 2020

Dr. Kathleen B. Aviso

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Dear Dr Aviso,

Please find attached the revised manuscript “Development of high performance recycled carbon fibre composites with an advanced hydrodynamic fibre alignment process”, following your correspondence of 10th Feb 2020. The authors are grateful to the reviewers for their helpful guidance, which was very comprehensive. We have made the recommended changes as detailed below. In addition, the changes have been tracked and highlighted in the manuscript. The authors are confident that the paper is now a better match for the vision of JCP.

We look forward to a positive response.

Yours sincerely

Dr Zhe Liu



Editor's comments:

Kindly revise the highlights such that each highlight has a maximum of 85 characters in length including the spaces.

The highlights have been modified and each of them contains fewer than 85 characters in length including the spaces.

1. It is recommended that the authors work with a science editor who is proficient in the Native English language to improve the organization and structure of the manuscript.
In accordance with this advice, more thorough proof reading has been conducted with the revised manuscript.
2. Please avoid reference clusters. It is recommended that a short discussion of the relevant contribution of each reference cited be included to clearly differentiate one reference from another.
Following this advice, the manuscript has been revised and clusters of references have been removed
3. The entire manuscript should be written in the third person plural or singular perspective. Please avoid the use of "we" or "our"
Following this advice, the manuscript has been revised and no occurrences of 'we' or 'our' remain.
4. Please avoid having one heading after another with no discussion in between as in the case of section 2 and 2.1.

The following text has been added to the manuscript (between section 2 and section 2.1, Page 7):
"In this research work, a hydrodynamic short fibre alignment process was used to manufacturing highly aligned short fibre tape. A 2-level full factorial experiment was designed to investigate the effect of four operating conditions on fibre orientation. The alignment quality of each tape was evaluated through dry fibre tape compaction testing. After analysing the results, the critical influencing factors were identified. Then the optimal aligned fibre tapes were manufactured and moulded as composites specimens. The mechanical performance of the aligned short fibre composites was evaluated through analysing the experimental results of tensile testing."

The following text has been added to the manuscript (between section 3 and section 3.1, Page 11):

"In this study, the optimal processing factors of the hydrodynamic alignment process has been identified. The mechanical property of aligned fibre composites has been evaluated and analysed."

5. The conclusions can be enhanced and improved by providing the implications of the results of your work towards the development and adoption of cleaner production technologies or towards improving environmental sustainability. Furthermore, the entire manuscript should

clearly discuss how this work is situated within the broader context of cleaner production and environmental sustainability. Kindly provide directions for future work.

The paper has been revised to demonstrate greater alignment with the broader context by modifying several sections (including the abstract and introduction) and including more references to other work in JCP. The authors believe that these changes significantly increase the value of the paper for JCP's readership. In addition, the areas suggested for future work in the conclusions section has been expanded.

6. Please make sure that only relevant references are included in your reference list. Recommended references provided by the reviewers or the editor are only recommendations and should only be included if they add value to your manuscript.
7. Please be reminded that original article submission should have a length of 6000 - 8000 words. Kindly keep this in mind as you conduct your revisions.
8. Please read through the authors guidelines to ensure that the manuscript adheres to them. For example, the reference should be cited in the format (Author, Year) instead of the numbered format used in this submission. All references should then be listed in alphabetical order, according to the surname of the first author, at the end of the manuscript. Following this advice, the format of the references has been revised.

Reviewers' comments:

Reviewer #1:

1. Abstract – The four influencing factors optimisation using two level factorial on the fibre orientation should be mentioned

The following text has been added to the manuscript (Page 2):

“To evaluate the effects of process factors on fibre orientation, a two-level full factorial experimental plan was designed.”

2. Methodology – The Design of Experiments (DOE) using Minitab and its methodology should be explained in this section for readers clarity

The following text has been added to the manuscript (Page 7):

“To explore the effects of processing factors, the Design of Experiments was conducted using Minitab. The four influencing factors to be investigated in this study were fibre length (to avoid the interference from the inconsistent fibre length of recycled carbon fibre, chopped virgin fibre tow was used), fibre volume concentration in suspension, dewatering vacuum level, the ratio of rotating drum speed and the fibre suspension depositing rate. For each of the factors two value levels were selected. The randomised experiment run order

was generated Minitab which includes two replicate measurements for the same factor settings (levels).”

3. Results and Discussion – Please explain/define what do you mean by heterogeneous nature in this context "Due to the heterogeneous nature of the drum surface, if High vacuum level was applied, the fibres will tend to move along the glycerine flow direction during the dewatering process, and the pre-aligned orientation will be damaged"?

The manuscript has been revised as (Page 12):

“The drum contains multiple vacuum slots to enable the residual glycerine flow can be dewatered. If high vacuum level was applied, a locally concentrated high vacuum suction region is generated around the vacuum slots. The fibres will tend to move along the flow direction and be accumulated around the vacuum slots. Then the pre-aligned orientation will be damaged.”

4. Could the authors explain or provide scientific explanation to the readers to why they have gotten 59% of fibres for 3 mm composite and 54% for 12 mm composite for a 3 degree fibre misalignment? Explain the effect of fibre length on the fibre misalignment? And could authors explain the difference in the results obtained in this study with 70% of 4 degree misalignment in reference 27? "In the present work, based on the fibre orientation distribution results in Figure 7, there are 93% of fibres....."

The following text has been added to the manuscript (Page 13):

“Although the optimised processing factors were chosen, the composite specimens manufactured with 12mm fibre have a higher degree of misalignment. The reason was explained by analysing the effects of the fibre length on suspension jet stability and alignment quality (Liu, Turner et al. 2019). For the suspension jets with same fibre volume concentration, the 12mm fibre-containing suspension has 4 times larger fibre aspect ratio compared to that of 3mm fibre-containing suspension which leads to a more significant fibre-fibre interaction. Thus, even though the fibre concentration, suspension viscosity and jet Re number are same, the use of 12 mm fibre can make the jet more unstable and resulting a fibre misalignment.”

In the reference 24, the authors used a filtration process. However, the alignment process used in this study is an optimised centrifugal process. In the reference 27, the author was using a centrifugal process but the details of setup and processing parameters were not clearly described. The fibre misalignment angles stated in reference 24 and 27 were analytically calculated and not validated by experimental data. Thus, in the authors considered opinion the data from literature are not directly comparable (at least to the extent encouraged by the reviewer) with the results in this study.

5. The scale bar in Figure 6 should be re-drawn with a bigger and clearer font
Figure 6 has been revised.

6. For Table 1, the representation is confusing, it is advised to separate it into two columns i.e one on experimental parameter and the other on parameter range
Table 1 has been revised.

Reviewer #2:

1. In Table 3 the standard deviation of sample Tenax-A HT 12 mm is much larger than the other ones Possible explanations?

The following text has been added to the manuscript (Page 15):

“On the other hand, the larger fibre aspect ratio of 12mm fibre-containing suspension results in more significant fibre-fibre interaction which leads to more inconsistent jet stability. Thus, the standard deviation of Tenax-A HT 12 mm composites sample is much greater than the others.”

2. Residuals of glycerine are found to compromise the adhesion between fibres and matrix. Do the authors think that, when using recycled fibres deriving from different processes, other substances present on the surfaces can compromise the final performance of the material?

The authors are in agreement with the reviewer’s concern: If the recycled fibre is not clean (for example, it contains residual char on the surface), the bonding between fibre and matrix will be affected and resulting mechanical performance potentially compromised. The researchers who focus on developing recycling technologies normally use SEM to observe whether there is any residual remain on the fibre surface but this is out of scope for this study which focusses on machine parameters affecting alignment quality.

Reviewer #3:

1. The abstract does not address the novelty of the paper.

The following text has been added to the manuscript (Page 2):

“This represents the first time a systematic study of input parameters on final part performance has been published in the open literature.”

2. Introduction
 - a. Literature review on other techniques for the processing of rCF such as tape, yarn manufacturing and HiPerDiF is not sufficient. Especially the difference between the used method in this paper and HiPerDiF should be clearly mentioned.

The following text has been added to the manuscript (Page 5):

“The former has been manufactured through a patented technology HiPerDiF. In this process, water has been used to disperse the fibres. Then the fibre suspension spreads through a nozzle on a corresponding plate. Then the fibres tend to align parallel to the plates

and deposit on the perforated moving belt. Vacuum suction channel has been attached at the bottom to dewater the residual liquid and maintain the aligned fibre orientation. In the end the fibre tape is dried and resin impregnated by the application of heat and pressure to form prepregs.

The yarn has been produced by an optimised DREF-3000 friction spinning machine. The process consists of three steps: carding (to form card webs), drawing (to form draw frame slivers), and spinning (to manufacture hybrid yarn structures with a core-sheath structure). In this method, the feed material is mainly longer reclaimed fibres >50mm from the dry fabric waste."

- b. The sentence „Traditional textile carding has been used to provide excellent alignment" is not correct. The subsequent processing such as e.g drawing is the main reason for the improvement of the fibre alignment. The alignment in card sliver is generally not adequate.

The authors agree that this statement is misleading. The manuscript has been revised as follows (Page 5):

“In the Traditional textile industry, carding has been widely used to guide the fibres to produce a fibre web which lacks sufficient homogeneous and aligned fibre orientation. In addition, as a very brittle material, carbon fibre can be severely damaged under this intense mechanical action.”

- c. The information must be concised provided in „After the first recycling stage, the virgin carbon fibre composite components will be reused as ASMC (advanced sheet moulding compound) SMC [12] which can only offer lower volume fraction with random fibre orientation and may be acceptable for the manufacture of semi- or nonstructural components."

Unfortunately the authors are not certain what the reviewer is taking issue with here. We are happy to make further clarifications to the text but references are given for interested readers to investigate.

- d. A large portion of the information provided in the last paragraph of introduction should be included in experimental parts.

The following text has been moved to section 2.1 (Page 8):

“The four influencing factors to be investigated in this study were fibre length (to avoid the interference from the inconsistent fibre length of recycled carbon fibre, chopped virgin fibre tow was used), fibre volume concentration in suspension, dewatering vacuum level, the ratio of rotating drum speed and the fibre suspension depositing rate.”

The following text has been moved to section 2.2 (Page 9):

“Compaction testing is a better way of discerning small differences in alignment quality than tensile testing, although the latter clearly provides a definitive guide to performance the results are confounded by several additional factors (such as resin mechanical properties, moulding conditions, errors in the tensile testing process etc.).”

3. Methods and materials: The properties of single fibres such as strength, modulus and elongation are missing.

Table 2 has been added to describe the fibre properties.

4. Aligned fibre tape manufacturing with the hydrodynamic alignment process:
 - a. more information in Figure 1 should be included.

Figure 1 has shown how the centrifugal alignment process working and the details has been explained by text in Section 2.1. This process has not been patented, the authors are unwilling to disclose all the details by a more explicit image or a photo. See also comments below which are also relevant.

- b. What is the dimension of the tapes produced? What is the production rate of the process?

The dimension of tape "(125mm×942mm)" has been stated in the manuscript (Page 8). As the alignment process used in this study is still at lab scale stage (low TRL), the authors are optimising the process through investigating the influencing processing factors. There is significant work still to do before we can progress to a more representative production rate. Primarily given this uncertainty but also considering potential IP issues the authors are unwilling to indicate the production rate.

- c. How do you control the areal density in the process?

The following text has been added to the manuscript (Page 8):

"By gathering the fibre volume concentration in suspension, jet volume flowrate, nozzle moving speed, fibre density and the area of deposited tape (125mm×942mm), the areal density of a single layer of tape can be calculated. The nozzle travel cycle is controlled and monitored by machine controller so the number of deposited layers can be counted and converted to the total areal density of tape."

- d. The parameters given in Table 1 should be discussed in more detail.

The following text has been moved to the Experimental part (section 2.1):

"The four influencing factors to be investigated in this study were fibre length (to avoid the interference from the inconsistent fibre length of recycled carbon fibre, chopped virgin fibre tow was used), fibre volume concentration in suspension, dewatering vacuum level, the ratio of rotating drum speed and the fibre suspension depositing rate."

- e. Why did you remove the sizing of the fibres?

Virgin fibres have been used in this study to simulate recovered fibre as they have consistent and known mechanical properties and fibre length. One disadvantage with this approach is that the surface is arguably enhanced by the sizing when compared to sizing-free recovered

fibres. For this reason, the virgin fibre sizing was removed to improve the analogy with recovered fibres.

5. Dry fibre tape compaction testing:

- a. It is not clear how did you calculate the fibre volume fraction by tape compaction.

The following text has been added to the manuscript (Page 9):

“Then the fibre volume fraction of the tape is equal to areal density divided by fibre density and the thickness of tape.”

6. Manufacture of aligned fibre composite in an autoclave for the tensile testing:

- a. Virgin fibres of 3 and 12 mm were used to analyse the process parameters as mentioned in Table 1. However, 7 mm CF was additionally used for the manufacturing of composites. Why 7 mm fibre was not included for the analysis of experimental factors, though it showed the highest tensile strength?

To identify the effects of fibre length on alignment quality, 3mm and 12mm fibre were considered as they are extremes of the applicable lengths in this process. In terms of establishing main effects there was little perceived benefit in including intermediate lengths. The 7mm T800 fibre type was included as an example of higher performance fibre use in the manufactured composite (i.e. not in establishing fundamental behaviour of the process). The difference between the mechanical performance of the Tenax-A HT C124 fibre vs the TorayT800 is discussed in the Table 2.

- b. Which process parameters were used to produce the tapes applied for the composite manufacturing.

The parameters were stated in Figure 5.

- c. The thickness of the composite was between 0.2 to 0.5 mm, which is very thin and should be at least around 1 mm to get reliable results. I have doubt about the reproducibility of the composite results.

The mistake has been corrected in the manuscript (Table 3)

7. Mechanical properties:

- a. Only theoretical calculation of tensile modulus of composites is included in the discussion. Nothing is discussed about the tensile strength. Why the tensile strengths are different in different composite specimen?

The authors do not consider that a theoretical calculation of tensile strength is of value given the inherent issues with such a calculation. The tensile strength data is provided as-is and a discussion of strength based on such a limited number of repeats with 2 fibre types, fibre length effects and complex surface phenomena related to potential residual glycerine is potentially misleading to readers. The authors have included some text to highlight the strength differences observed experimentally.

Reviewer #4:

1. To sum up, even if the article is well written, and results are clear and interesting, I think that talking about recycled carbon fibre realignment is abusive. Indeed, I am not sure that the photography presented on figure 2 is really representative of your tests. A finer explanation of the passage between long recycled fibre to short ones who are used in suspension is recommended.

Figure 2 is a schematic diagram to describe the manufacturing process of aligned recycled fibre composites and is wholly representative of the process used in this work. The fibre shown in the image labelled "Recycled Carbon Fibre" has the same fibre length as those shown in "Aligned Fibre Tape". There is not a sub-process to convert the long recycled fibre to short ones. Indeed the fibre shown in the image is fibre recovered via our own recycling process. Long fibres can potentially be recovered from recycling processes but it is common (and potentially significantly most cost / energy effective) to reduce the fibre length before fibre recovery. A detailed discussion of the different recycling routes is outside the scope of this paper.

2. Comment on abstract: Just one remark, due to your quite short fibre, I do not agree with the sentence "... length degradation to a minimum." Indeed, I am not sure that you can used much more longer fibre in your process.

It's quite true that significantly longer fibres would not be appropriate for the process. The authors would like to explain the sentence mentioned by the reviewer: "This allows composites with high fibre content to be manufactured at lower moulding pressures with the added benefit of keeping fibre length degradation to a minimum."

To achieve high fibre volume fraction, applying high compression pressure is the widely used method. However, if the laminate has randomised fibre orientation, the fibre length will degrade under high compression pressure due to fibre to fibre contact. Thus, aligning the short fibres can make the composites achieving high volume fraction under relatively low compression pressure thus avoiding widespread fibre breakage and resulting length reduction. This phenomenon is well established in existing literature. See for instance a 2009 conference paper by the authors "Compression moulding of a recycled carbon fibre reinforced epoxy composite" International SAMPE Symposium and Exhibition (Proceedings) Volume 54, 2009, 12p SAMPE '09 Spring Symposium Conference Proceedings; Baltimore, MD; United States; 18 May 2009 through 21 May 2009.

3. Comment on introduction: In the top of page 6, there is an unduly bracket. You explain that you use cut virgin fibre, but in my opinion, the final goal of recycling process of carbon fibre is closely linked with fibre length preservation. And how can you process fluffy recycled fibre? I think that in your introduction, you do not explain enough which recycled route do you envision.

Whilst its true that in an ideal situation the maximum fibre length would be preserved and it is possible to recover fibres from e.g. non-crimp fibre composites that have not been impregnated but there are no recycling processes that can cope with layered (i.e. real laminated) composites and recover e.g. 1m long fibres. In addition, there are no conversion processes that can make use of such long fibres. Of course the exact lower limit of useful length could be debated but the authors would argue that this remains true for, say, 100mm long fibres.

These issues are certainly valid concerns for the future of recovered carbon fibre composites but there is an increasing amount of literature in this area and most of these factors are well understood, the authors intended the current paper to be a systematic and detailed evaluation of processing factors and have attempted not to pontificate too much on the wider issues which are covered elsewhere.

4. Comments on materials and methods section:
 - a. Section 2.3. Does effective length of ELG recycled carbon are representative of high performance composite? Why are you forced to shred this fibre?

The sentence referred to by the reviewer is: "End of life carbon fibre composites was supplied by ELG Carbon Fibre Ltd and then shredded and recycled by the Fluidised Bed process at the University of Nottingham [11] (described as HS recycled in Table 2).".

As explained in the sentence, the End of life carbon fibre composites supplied by ELG had not yet been recycled. Thus, they had to be shredded to ensure the size can feed in to fluidized bed. Normally, for end of life composites pieces there is no clear and detailed information provided regarding fibre length, fibre orientation, fibre type etc. See also comments above.

- b. If a grinding of the composite before their treatment or a grinding of fibre after their treatment is necessary, I am sure that the obtained product will not be economically viable.

This is a valid concern however the removal of manual process steps is thought to be a far more important concern at present. Size reduction has been shown elsewhere to be relatively inexpensive compared to the recovery and conversion processes. As explained above the authors do not wish to be drawn excessively into cost factors outside the scope of the actual investigation.

- c. Can you analyze the human factor in the manual determination of the centre line of each fibre in each picture! There is no image analysis software?

It is difficult to track the complete fibre edge by using image analysis software. The author has improved the measuring method by sketching a line along the fibre length direction and the line is coincident with the fibre edge. Therefore, the human factor can be minimised. The author will involve more advanced characterization method (for example, MicroCT) in the future work.

5. Comments on results and discussion section:

- a. Please, explain us what is a low vacuum and a high vacuum.

The low vacuum level 0.25bar and high vacuum level 0.5bar have been stated in Table 1.

- b. Page 12, end of section 3.1, the sentence " ... five times wire to jet speed ratio ... " is not clear for me.

The manuscript has been revised as (Page 12):

"The manufacturing conditions are 0.15% fibre volume concentration in the suspension, five times higher drum tangential rotating speed relative to the suspension jet velocity and low dewatering vacuum level."

- c. Same page and section, sentence "A tape made from ... with 12 mm fibre." is unnecessary due to previous presented results.

The authors would like to keep this sentence as a short summary of previous presented results to assist with overall clarity.

- d. Section 3.2 - Why don't you really compare your orientation dispersion with references 27 and 24. For example for reference 27, 70% of fibre are in the range +/-4°. In this section, a synthesis table would help.

In the reference 24, the researcher used a filtration process. However, the alignment process used in this study is an optimised centrifugal process. In the reference 27, the author was using a centrifugal process but the details of setup and processing parameters were not clearly described. The fibre misalignment angles stated in reference 24 and 27 were analytically calculated and not validated by experimental data. Thus, the author think the data from literatures are not directly comparable with the results in this study. In addition, the researchers didn't state the full range data of fibre orientation angle. Thus, a synthesis table or graph cannot be scientifically used to compare the fibre orientation distribution.

- e. In your equations, too many parameters are not defined.

The following text has been added to the manuscript (Page 14):

"E and V are Young's modulus and volume fraction; and the subscripts c, f, and m refer to the composite, fibres, and matrix."

- f. On figure 9, you present 4 cases, and in your discussion section, you analyze only two cases. Why?

To identify the effects of fibre length on alignment quality, 3mm and 12mm fibre were considered as they are extremes of the applicable lengths in this process. In terms of establishing main effects there was little perceived benefit in including intermediate lengths. The 7mm T800 fibre type was included as an example of higher performance fibre use in the manufactured composite (i.e. not in establishing fundamental behaviour of the process). Normally, for end of life composites pieces there is no clear and detailed information provided regarding fibre length, fibre orientation, fibre type etc. Thus, including the HS recycled fibre (recycled from the end of life carbon fibre composites supplied by ELG) in the processing factor analysis will mislead the results. However, the difference between the mechanical performance of the Tenax-A HT C124 fibre vs the TorayT800 and HS recycled fibre is discussed in the text.

6. Comments on figures and tables:

- a. Table 3: can you explain the short difference between "Tenax 12 mm" and HS recycled cases on fibre volume fraction and tensile modulus.

As it is explained in section 3.3, the average fibre length of HS recycled fibre is 1.15mm, but 35% of the fibres have a length shorter than the average and in around 3% of cases is below the critical fibre length of 0.38mm. It is challenging to retain the aligned orientation of these very short fibres during the deposition process and resulting a slightly worse alignment quality. Thus, the composites specimen made with HS recycled fibre tape obtained a lower fibre volume fraction and tensile modulus.

- b. Table 4: due to your manual estimation of distribution factor, are you sure of the third digit after decimal point?

As it is explained in section 2.4, the in-plane fibre orientation microscopical image was imported to AutoCAD, the line used to represent each fibre was drawn manually over the image on AutoCAD. Then AutoCAD automatically calculated the fibre orientation angles and the data file was exported as a spreadsheet. Thus, the numbers contain three decimal places and are accurate.

- c. Figure 2: recycled carbon fibre photography is yours or not? If not please precise the reference.

It is the authors' work.

- d. Figure 9: you don't have error bars for Toray T800 tests?

Following this advice, the Figure 6 has been revised.

7. Comments on references: [2] and [10] right alignment problems!

Following this advice, the manuscript has been revised.

CRediT author statement

- **Zhe Liu** – Conceptualization, Writing - Original Draft, Investigation, Validation, Methodology, Formal analysis
- **Thomas A. Turner** – Writing - Review & Editing, Supervision, Project administration, Methodology
- **Kok H. Wong** – Methodology, Software, Supervision
- **Stephen J. Pickering** – Supervision, Project administration, Funding acquisition

29th April 2020

Dr. Kathleen B. Aviso

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Dear Dr Aviso,

Please find attached the revised manuscript “Development of high performance recycled carbon fibre composites with an advanced hydrodynamic fibre alignment process”, following your correspondence of 10th Feb 2020. The authors are grateful to the reviewers for their helpful guidance, which was very comprehensive. We have made the recommended changes as detailed below. In addition, the changes have been tracked and highlighted in the manuscript. The authors are confident that the paper is now a better match for the vision of JCP.

We look forward to a positive response.

Yours sincerely

Dr Zhe Liu



Development of high performance recycled carbon fibre composites with an advanced hydrodynamic fibre alignment process

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Key words: Carbon fibre reinforced composites, Recycling and Reuse, Mechanical properties, Mechanical testing

Word count: 6588

Highlights

- Recycled carbon fibre composites have great potential for vehicle light-weighting
- High-performance materials have been produced through the developed process
- Processing factors influencing the alignment quality are addressed
- A highly aligned fibre orientation distribution is observed enhancing fibre packing
- The observed mechanical properties compete with metals for transport applications

Abstract

Carbon fibre composites have great potential for vehicle lightweighting but the high cost and environmental impact of their production tends to largely undermine the advantages in non-aerospace applications. Recycled fibre has the potential to significantly reduce both cost and environmental impact but has yet it has not been widely accepted by the composites industry due to reduced mechanical performance in components as well as the difficulties in handling and processing caused by the fluffy discontinuous form which is quite unlike any current material formats which suit existing processing methods. The developed alignment process allows discontinuous random recycled carbon fibre to be processed into tapes with a highly aligned orientation distribution. This allows composites with high fibre content to be manufactured at lower moulding pressures with the added benefit of keeping fibre length degradation to a minimum. To evaluate the effects of process factors on fibre orientation, a two-level full factorial experimental plan was designed. This represents the first time a systematic study of input parameters on final part performance has been published in the open literature. With further improvements to the process, it is shown that it is possible to manufacture a composite achieving high fibre volume content (46%) under 7 bar moulding pressure in an autoclave, exhibiting competitive mechanical properties with almost 100 GPa tensile modulus and over 800 MPa tensile strength.

1. Introduction

In the light of steadily increasing global environmental pressure, the European Union has set stringent CO₂ emissions regulations for both the commercial aviation and automotive sectors (EuropeanCommission 2014, Regulation(EU)-2019/63 2019). The CO₂ emissions of all new cars in 2021 is required to be 40% lower than the 2015 average (Regulation(EU)-2019/63 2019), while aircraft produced from 2024 should achieve a 20% - 30% reduction in CO₂ emissions, compared to the 2014 level (EuropeanCommission 2014). Hence, lightweight design has become a major research theme in the aerospace and automotive sectors to reduce emissions and improve fuel efficiency. By virtue of their high specific mechanical properties, carbon fibre reinforced composite materials are widely used in modern transport vehicles. For example, the usage of composites in commercial aircraft has reached about 50 wt.% (Brosius 2007), and a similar trend can be found in automotive applications like the BMW i3 (Starke 2016). With increasing interest in the use of composite materials, their recyclability and the environmental impact of their production becomes an issue. Life cycle analysis for composites has drawn attention from researchers (Li, Bai et al. 2016, Wu, Xia et al. 2018, Cousins, Suzuki et al. 2019, Hagnell and Åkermo 2019), because at least 95% of a new vehicle by weight needs to be recycled at the end of life and 85% of the waste should be reused since 2015 according to the ELV directive (Directive-2000/53/EC 2014, Mudgal, Kong et al. 2014).

Composite materials are complex from a recycling & reuse perspective as the fibres themselves are universally processed into a composite form with other materials which then normally require separation at the recycling stage. It is also not sufficient to produce high quality recycled fibres; they must either be able to be incorporated into normal composites or must be able to be incorporated into an alternative high performance intermediate material. To date, the full potential of recycled carbon fibres has not been realised and efforts are ongoing

to improve the mechanical properties achievable such that closing the loop becomes more realistic. The difference in elastic modulus between virgin and recycled carbon fibre filaments themselves tends not to be significant (Pickering 2015), but some degradation of strength can typically be observed, depending on the nature of the recycling process. Virgin fibre is continuous at the point of manufacture and can be readily woven into the formats that existing high-grade applications demand. In contrast, recycled carbon fibre cannot be a direct substitute for virgin fibre as it lacks sizing, has a short fibre length, is discontinuous in nature and fluffy in form. Thus, one option is to develop further manufacturing processes to improve the orientation distribution and convert the material into an intermediate format that can be processed via conventional routes. A circular closed-loop material cycle of carbon fibres has been examined (Hagnell and Åkermo 2019), the author points out that the reclaimed fibre length is the key factor in determining the potential reuse value of recycled carbon fibre in high-performance product design. After the first recycling stage, the virgin carbon fibre composite components will be reused as ASMC (advanced sheet moulding compound) with fibre length between 25-50mm, which are introduced into a compression moulded structural component for automotive applications. This is then processed through the secondary recycling process with further fibre length reduction (fibre length between 2mm and 25mm). As the fibre length becomes too short for efficient alignment with the current technologies, these recyclates will be converted into non-woven mat or SMC (Oliveux, Dandy et al. 2015) which can only offer lower volume fraction with random fibre orientation and may be acceptable for the manufacture of semi- or non-structural components.

Meanwhile, members of the recycling industry are seeking new processes for higher-value fibre remanufacturing, and the concept of aligning the fibres is considered to be a competitive conversion option as it allows high fibre packing fraction (and thus high mechanical properties) to be achieved even under low compressing pressure which can avoid fibre

breakage and save energy. Several fibre alignment processes have been developed. Under a magnetic field, short carbon fibre with a length less than 500 μ m can be aligned due to its diamagnetic anisotropic property (Matthews M 1996). A similar method has been explored to control the orientation distribution of carbon nanotubes or polymer fibres by electric fields (Park, Wilkinson et al. 2006, Seyam, Cai et al. 2008, Yousefi, Sun et al. 2014). In the traditional textile industry, carding has been widely used to guide the fibres to produce a fibre web which lacks sufficient homogeneous and aligned fibre orientation. In addition, as a very brittle material, carbon fibre can be severely damaged under this intense mechanical action. By combining recycled carbon fibre with glass fibre or thermoplastic fibre, hybrid prepreg tow (Yu, Potter et al. 2014) and yarn (Hasan, Nitsche et al. 2018) has been developed. The former has been manufactured through a patented technology HiPerDiF. In this process, water has been used to disperse the fibres. Then the fibre suspension spreads through a nozzle on a corresponding plate. Then the fibres tend to align parallel to the plates and deposit on the perforated moving belt. Vacuum suction channel has been attached at the bottom to dewater the residual liquid and maintain the aligned fibre orientation. In the end the fibre tape is dried and resin impregnated by the application of heat and pressure to form prepreps. The yarn has been produced by an optimised DREF-3000 friction spinning machine. The process consists of three steps: carding (to form card webs), drawing (to form draw frame slivers), and spinning (to manufacture hybrid yarn structures with a core-sheath structure). In this method, the feed material is mainly longer reclaimed fibres >50mm from the dry fabric waste. Benefitting from the aligned fibre orientation, these two candidates can potentially be used as substitutes for virgin fibre products, but secondary conversion processes are required, e.g. weaving, to convert them to either prepreg or preform before moulding. Though the application of hybrid prepreg tow in 3D printing has been investigated, it still cannot fit the rapid production and high mechanical property requirements of large structural components.

During the late 1960s and 70s, hydrodynamic converging flow alignment processes attracted researchers' attention. Three processes (Parratt 1968, Bagg, Dingle et al. 1971, Bagg 1977) were developed by researchers from ERDE, the Explosives Research and Development Establishment UK. Among these, the filtration process is the most widely published technique. Initially, a batch scale rig was developed to manufacture aligned fibre tape up to size 400 mm x 300 mm. This process was then upgraded into a continuous one for a sheet roll with 152 mm width (Bagg, Evans et al. 1969). A diluted glycerine/water solution was applied to disperse discontinuous fibres. As a Newtonian fluid, the viscosity of glycerine/water solution will remain constant during the dispersing process if the temperature remains constant. Longer fibres and a higher fibre concentration in the suspension are always preferred in order to achieve high mechanical performance and high production rate. With a carefully selected short fibre length, low fibre volume concentration in suspension and the high shear rate offered by the high viscosity Newtonian dispersion medium, a good dispersion quality can be achieved (i.e. undispersed bundles were not observed). The fully dispersed fibre suspension was then conveyed towards a convergent headbox with a narrow-slit exit. The headbox was attached above a transferring forming fabric and travelled backwards and forwards relative to the mesh repeatedly to deposit fibre suspension on a region of the forming mesh. Meanwhile, a vacuum suction was attached under the mesh to dewater the residual dispersing medium. In the end, water spray was applied to gently wash the damp tape, and then binder was applied.

The centrifugal process was built with two significant modifications compared to the filtration process. A series of nozzles was used instead of the converging headbox as the nozzle can offer a two-dimensional convergence contour along the flow direction. A rotating drum with a layer of forming fabric attached at the internal surface was used as the

centrifugal force could accelerate dewatering. It was pointed out that the centrifugal process can offer better alignment quality along with higher throughput (Salariya and Pittman 1980).

In the present study, an improved centrifugal rig has been constructed inspired by the ERDE work. A 2-level full factorial experiment was designed to investigate the effect of four operating conditions on fibre orientation. The volume fraction and fibre orientation distribution within the aligned tape were evaluated by analysing micrographs of both the fibre elliptical cross-sectional geometry and in-plane fibre orientation geometry. Wong (Wong, Turner et al. 2009) surmised that a higher level of alignment implies that the fibres are more tightly packed, which results in a high fibre volume content under the same pressure. Dry mat compaction testing was therefore used as the primary testing method to evaluate the resulting fibre volume fraction in all aligned tapes under different loading pressure, and this data was used to determine the influence of the four factors. The summarised results of the compaction testing allowed the down-selection of the optimised manufacturing conditions of the alignment process. For these tapes, tensile testing was used to evaluate the mechanical properties of aligned discontinued carbon fibre moulded laminates.

2. Methods and materials

In this research work, a hydrodynamic short fibre alignment process was used to manufacture highly aligned short fibre tape. A 2-level full factorial experiment was designed to investigate the effect of four operating conditions on fibre orientation. The alignment quality of each tape was evaluated through dry fibre tape compaction testing. After analysing the results, the critical influencing factors were identified. Then the optimal aligned fibre tapes were manufactured and moulded as composite specimens. The mechanical performance of the aligned short fibre composites was evaluated through analysing the experimental results of tensile testing.

2.1 Aligned fibre tape manufacturing with the hydrodynamic alignment process

The hydrodynamic fibre alignment process, as described in Figure 1, was used in this work. The fully dispersed fibre suspension is gently transferred into an intermediate pressurised tank which ensures that the flow through the conical nozzle is steady. The nozzle is attached on a linear actuator that keeps moving transversely to reach the desired width of fibre tape. By gathering the fibre volume concentration in suspension, jet volume flowrate, nozzle moving speed, fibre density and the area of deposited tape (125mm×942mm), the areal density of a single layer of tape can be calculated. The nozzle travel cycle is controlled and monitored by machine controller so the number of deposited layers can be counted and converted to the total areal density of tape. A forming fabric is placed inside the drum and acting as a filtration medium to detach fibres from the dispersing medium, and it also serves as a vacuum pressure distribution medium to create an even and highly efficient dewatering behaviour. The filtered glycerine is recirculated for reuse. After the desired areal density has been achieved, the dispersion medium remaining in the tape is gently washed away with an atomised warm water, and an epoxy-based binder is then applied to maintain the aligned fibre orientation. To explore the effects of processing factors, the Design of Experiments was conducted using Minitab. The four influencing factors to be investigated in this study were fibre length (to avoid the interference from the inconsistent fibre length of recycled carbon fibre, chopped virgin fibre tow was used), fibre volume concentration in suspension, dewatering vacuum level, the ratio of rotating drum speed and the fibre suspension depositing rate. For each of the factors two value levels were selected. The randomised experiment run order was generated Minitab which includes two replicate measurements for the same factor settings (levels). The details of the 2-level full factorial experimental plan are listed in Table 1. Toho Tenax Europe GmbH supplied carbon fibre staples, Tenax®-A HT C124. The fibre lengths were 3 mm and 12 mm with a nominal filament diameter of 7 microns (Table 2). In

order to remove the sizing, fibres were placed inside a furnace for 15 mins at 550 °C. Univar UK supplied Dow Oleon Glycerine 4810 with a glycerol content over 99.5%wt which was mixed with water to reduce the viscosity to 400 mPas which was measured at 15°C with a Brookfield LVDVII viscometer. Then, the solution was employed as a dispersing medium and was stored in a sealed container to avoid excess absorption of atmospheric moisture. A cooling system in the suspension tank was designed to help keep the viscosity of the suspension constant by controlling the temperature.

2.2 Dry fibre tape compaction testing

Compaction testing is a better way of discerning small differences in alignment quality than tensile testing, although the latter clearly provides a definitive guide to performance the results are confounded by several additional factors (such as resin mechanical properties, moulding conditions, errors in the tensile testing process etc.). A compaction test rig was used to measure the thickness of carbon fibre tape (in order to determine the fibre volume content) with varied pressure. The testing rig consists of two flat platens; the bottom one mounted on the base of the Instron 5969 universal testing machine; while the top one is connected to a 10 kN load cell attached to the machine crosshead. The machine's control software automatically logged the crosshead location during the test. There are two linear variable differential transformers (LVDTs) attached on both sides of the upper platen to record the distance between the platens accurately. The square-shaped specimens with an edge length 35mm were prepared by stamping carbon fibre tape with a cutting template. The areal density of each sample was accurately measured before the test. Afterwards, four layer specimens were stacked together, carefully located at the centre of the bottom platen and then compressed at a rate of 1 mm/min until compression pressure reached to 10 bar. The system compliance was measured by two LVDTs when 10 bar pressure was applied with no

specimen; the compliance curves were curve-fitted and applied as a correction factor during post-processing of the data. The thickness of each tape was obtained by taking the average reading values from the two LVDT units after the compliance was subtracted. Then the fibre volume fraction of the tape is equal to areal density divided by fibre density and the thickness of tape.

2.3 Manufacture of aligned fibre composite in an autoclave for the tensile testing

Not only short virgin fibres (Tenax®-A HT C124_3mm and 12mm, Toray_T800S_7mm) but also recycled fibre was introduced in this work. End of life carbon fibre composites was supplied by ELG Carbon Fibre Ltd and then shredded and recycled by the Fluidised Bed process at the University of Nottingham (Pickering 2015) (described as HS recycled in Table 2 and Table 3). The effective number average fibre length is 1.15mm. As the source of this supplied end life waste piece was not known, the fibre filament properties of HS recycled fibre were assumed to be the same as Tenax®-A HT C124 fibre. The details of laminates which were manufactured from these tapes are listed in Table 3. The laminates were stacked with the MTM57 resin films (supplied by Solvay, Heanor, UK) for subsequent impregnation and vacuum bagged to remove the trapped air. Then the compacted materials were moulded in an autoclave at 120 °C, 7 bar pressure, for an hour. In the end, the composites were cooled down to around 60 °C (with a cooling rate of 3°C/min) under pressure before demoulding. The manufacturing procedure of aligned fibre composites is described as a schematic diagram in Figure 2.

Tensile tests were performed with the specimens made from these composites following the testing standard ASTM D3039. A 250 kN load cell was used (Figure 3), and the testing rate was 1 mm/min. For each type of laminate, four specimens were prepared. An extensometer

was attached on the specimen to record strain. Optical micrographs were used to view and analyse the cross-sectional of the laminates.

2.4 Fibre orientation determination

The surfaces of polished composite specimens were analysed with an optical microscope to investigate the in-plane fibre orientation. The composite samples were placed at the bottom of a plastic container and fibres were parallel to the container's bottom surface. Then, the container was filled with Prime20LV (Gurit, UK) resin. The cured resin containing the composite sample was then polished until the full length of carbon fibre can be observed. An optical microscope captured all sample images then imported into AutoCAD. The fibre orientation was determined by sketching a line along the fibre length direction and the line is coincident with the fibre edge. The data file contains orientation angles of each fibre were exported as a spreadsheet for further analysis. For each case, more than 5000 carbon fibres from five microscope images were processed to identify the orientation distribution. Figure 6 shows the examples of the aligned short fibre composite specimens which were used for in-plane fibre orientation analysing. Finally, the fibre orientation distribution results of 3mm fibre composites and 12mm fibre composites were obtained in Figure 7.

3. Results and discussion

In this study, the optimal processing factors of the hydrodynamic alignment process has been identified. The mechanical property of aligned fibre composites has been evaluated and analysed.

3.1 Dry fibre tape compaction results

In previous research work, with 10 bar compaction pressure, 3mm fibre aligned tape has been shown to achieve 47.7% fibre volume fraction and 12mm fibre aligned tape obtained 32.5%

fibre volume content (Liu, Wong et al. 2015). To improve the process efficiency and achieve higher fibre volume fraction, the processing factors influencing alignment quality are explored in this paper.

Thirty-two discontinuous aligned fibre tapes were manufactured using the described alignment process and tested using the developed compaction test. Based on the compaction test results of the full factorial test, the main effects plots for fibre volume fraction are shown in Figure 4. Fibre volume concentration in the dispersion medium is not a significant factor in determining the alignment of 3mm fibre and 12 mm fibre which is thought to be due to the improved dispersion process where the fibres were distributed more homogeneously in the suspension. Higher wire/jet velocity ratio is beneficial to the fibre alignment for both 3mm and 12 mm fibres, as it can drive fibres along the flow direction and avoids fibres bending as they are deposited on the nylon mesh. The vacuum level applied around the rotating drum is another critical factor: The drum contains multiple vacuum slots to enable the residual glycerine flow can be dewatered. If high vacuum level was applied, a locally concentrated high vacuum suction region is generated around the vacuum slots. The fibres will tend to move along the flow direction and be accumulated around the vacuum slots. Then the pre-aligned orientation will be damaged. Thus, a lower vacuum level is beneficial for 3mm fibre to maintain aligned fibre orientation, which is validated by DoE results plotted with Minitab in Figure 4.

The compaction result curves are plotted in Figure 5. This includes two benchmarks for reference: a 2D planar carbon fibre 100gsm random tape (supplied by Technical Fibre Products Ltd) and a 100gsm unidirectional continuous fibre (UD) tape (supplied by Easy Composites Ltd) as well as two aligned tapes from the full factorial test which are made with 3mm and 12mm Tenax®-A HT C124 virgin fibres respectively. These two obtained highest fibre volume fractions in the compaction test results for their respective fibre length. Under

10bar compression pressure, the fibre volume content of UD tape achieves 68% while the random tape can only obtain 18% fibre volume fraction which indicates that fibre packing efficiency can be significantly improved by the aligned fibre orientation. With 3mm fibre, the tape exhibits 52% fibre volume fraction under 10bar compaction pressure. The manufacturing conditions are 0.15% fibre volume concentration in the suspension, five times higher drum tangential rotating speed relative to the suspension jet velocity and low dewatering vacuum level. For 12mm fibre tape, 41% fibre volume fraction was achieved, and the processing factors are 0.15% fibre volume concentration suspension, five times wire to jet speed ratio and high dewatering pressure. A tape made from aligned 3 mm fibres can achieve a 27% higher volume fraction than the tapes with 12 mm fibres. This is own to a better fibre dispersion quality, which means no fibre bundle remains with shorter fibre.

3.2 Fibre orientation evaluation

In previous research work, 70% of the fibres were found to be in the range $\pm 4^\circ$ misalignment angle for a filtration process with rotating drum (Richter 1980). For the ERDE filtration process, it was reported that 90% of the fibres were in the range of $\pm 15^\circ$ (Salariya and Pittman 1980). In the present work, based on the fibre orientation distribution results in Figure 7, there are 92% of fibres in the range $\pm 10^\circ$ and 60% of fibres are in the range of $\pm 3^\circ$ in the 3mm fibre composite samples which have 46% fibre volume fraction. 86% of fibres in the range $\pm 10^\circ$ and 53% of fibres were in the range of $\pm 3^\circ$ in 12mm fibre composite samples, which have 43% fibre volume content. Although the optimised processing factors were chosen, the composite specimens manufactured with 12mm fibre have a higher degree of misalignment. The reason was explained by analysing the effects of the fibre length on suspension jet stability and alignment quality (Liu, Turner et al. 2019). For the suspension jets with same fibre volume concentration, the 12mm fibre-containing suspension has 4 times larger fibre aspect ratio compared to that of 3mm fibre-containing suspension which leads to

a more significant fibre-fibre interaction. Thus, even though the fibre concentration, suspension viscosity and jet Re number are same, the use of 12 mm fibre can make the jet more unstable and resulting a fibre misalignment. Comparing with the results for 3mm fibre aligned tape obtained in the dry fibre tape compaction test, at the same loading pressure, the fibre volume content of moulded composites is reduced from 49% to 46% which indicates that the fibre alignment quality of the cured composite samples was slightly degraded following the post-processing operations – residual glycerine washing, applying binder and laminating.

3.3 Mechanical properties

In Table 4, the results from the tensile test are listed, which include the average fibre volume fraction of each specimen, mean values and standard deviations of modulus and strength. The fibre lengths investigated in this paper are longer than the critical fibre length. Thus, fibre breakage results rather than matrix or interface failure. Thus, extensive fibre pull-out situation was not witnessed at the fracture surface of both virgin fibre samples and recycled fibre ones. The fracture surface images were obtained using Scanning Electron Microscopy (JEOL 6060LV) and are listed in Figure 8.

For comparison, a modified rule of mixtures for modulus (Sanadi and Piggott 1985, Coleman, Khan et al. 2006) was used to produce an analytical modulus calculation. The expression is given by:

$$E_c = \eta_0 \eta_1 V_f E_f + V_m E_m \quad \text{Equation 1}$$

Where E and V are Young's modulus and volume fraction; and the subscripts c, f, and m refer to the composite, fibres, and matrix.

η_0 = fibre orientation distribution factor

η_1 = fibre length distribution factor

The fibre orientation distribution factor can be calculated by Krenchel's equation (Krenchel 1964):

$$\eta_0 = \sum_{i=0}^{180^\circ} V_{fi} (\cos \theta_i)^4 \quad \text{Equation 2}$$

Where θ is the measured fibre orientation angle.

Based on the shear lag theory (Cox 1952), the fibre length distribution factor η_1 can be described:

$$\eta_1 = 1 - \frac{\tanh(\alpha l/d_f)}{\alpha l/d_f} \quad \text{Equation 3}$$

$$\alpha = \sqrt{\frac{-3E_m}{2E_f \ln V_f}} \quad \text{Equation 4}$$

Where d_f is fibre diameter.

The values of η_0 and η_1 are listed in Table 5. The comparison between the theoretical predictions and experimental results are plotted in Figure 9. Although the moulding pressure is only 0.7MPa which is much lower than the standard compression moulding processes (5–10 MPa), the composites made by the developed hydrodynamic alignment process obtained an excellent fibre packing efficiency due to the highly aligned fibre orientation. In the issue, high tensile modulus and strength were achieved. With 46% fibre volume fraction, the composite made from 3 mm aligned fibre gave a 98 GPa tensile modulus which achieves the closest agreement with the predicted value. With the same curing pressure (7 bar), the laminate made with 12 mm fibre can only achieve 40% fibre volume content and obtain 62 GPa tensile modulus which is 28% lower than the predicted value (Table 5). The reason for this discrepancy is thought to be that the 12mm fibre tape has a higher porosity compared to

that of 3mm fibre aligned tape. Thus, during dewatering, the through-thickness pressure difference of 12 mm fibre tape is lower (due to air leakage) when the tape is not fully covered by glycerine, which leads to the ineffective removal of residual glycerine. On the other hand, the larger fibre aspect ratio of 12mm fibre-containing suspension results in more significant fibre-fibre interaction which leads to more inconsistent jet stability. Thus, the standard deviation of Tenax-A HT 12 mm composites sample is much greater than the others.

Furthermore, although the same washing method was applied for both 12 mm and 3 mm fibre tapes, the 12 mm tape contains a higher level of residual glycerine after washing as the washing process is ineffective in dealing with the higher initial glycerine content. Residual glycerine is likely to affect material properties by reducing fibre: matrix adhesion significantly. The T800 fibre has a smaller diameter (5 μ m), and when the fibre suspension has the same fibre volume concentration, the actual number of fibre filaments will be higher than with 7 μ m diameter fibres. Thus, the fibre to fibre interaction will be more significant and tend to reduce the alignment quality. As a result, T800_7mm fibre laminates exhibit 42% fibre volume fraction, which is 8% lower (4% fibre volume fraction in difference) than that of 3mm fibre laminates. The experimental tensile modulus is 87 GPa, which is 28% lower than the analytical value, and this is thought to be related to residual glycerine. 38% fibre volume fraction and 60GPa tensile modulus are obtained by the composite made by short recycled fibre, the average fibre length is 1.15mm, but 35% of the fibres have a length shorter than the average and in around 3% of cases is below the critical fibre length of 0.38mm. It is challenging to retain the aligned orientation of these very short fibres during the deposition process. These properties are competitive with the alternative structural materials and show the potential of fibre alignment for the manufacture of high-value composites from recycled carbon fibre.

Comparison with existing published results is challenging due to the variety of fibre volume contents and methods of manufacture. By the conventional wet process, Sanadi and Piggott (Sanadi and Piggott 1985) manufactured aligned carbon/epoxy composites with 2 mm long fibre which had 35% fibre volume fraction; these materials achieved 60 GPa tensile modulus and 650 MPa tensile strength. With 12mm long recycled fibre, Wong and Turner (Turner, Pickering et al. 2009, Wong, Turner et al. 2009) manufactured carbon/epoxy composite specimens with 45% fibre volume fraction and obtained 80 GPa tensile modulus which is 23% lower than the predicted value 103 GPa. The HiPerDiF method developed by Yu (Yu, Potter et al. 2014) has demonstrated properties of 115GPa tensile modulus and 1509MPa tensile strength for 3mm fibre tapes, 55% fibre volume fraction was achieved using pressure intensification which appears to apply more than 21bar. In this process where water is used as the dispersion media, any issues related to residual glycerine are avoided, but the viscosity of water may not offer enough shear stress to adequately disperse longer fibres and fluffy recycled fibre from end of life parts.

4. Conclusions

Discontinuous tapes have been shown to exhibit significantly improved mechanical properties when the alignment level of the fibres is high. This improvement comes both from higher fibre volume content in the composite and avoiding fibre breakage in the subsequent moulding process. A hydrodynamic process intending to achieve highly aligned fibre orientation has been developed, and the effects on alignment quality from processing factors have been explored and analysed in this paper. The composite laminates were made with 3mm carbon fibre under 7 bar curing pressure and had a tensile modulus of 98GPa and tensile strength of 826 MPa with a fibre volume content of 46%. Genuine recycled fibre from laminate scrap was also used in this work, and a tensile modulus of 60GPa and tensile

strength of 322 MPa was achieved by 1.15mm mean fibre length recycled fibre laminate composites with a fibre volume fraction of 38%. This indicates the capability of fibre alignment technique for manufacturing of high-value composites with specific mechanical properties that are not only better than other widely applied low-cost composite and metallic materials but are also competitive with virgin carbon fibre composites. Hence, this paper proposes an effective recycling and re-manufacturing solution for composite materials to aircraft and automobile manufacturers, to overcome the challenges of minimum 95% material recycled ratio and 85% material reused ratio from increasingly stringent sustainability regulations (Directive-2000/53/EC 2014, Mudgal, Kong et al. 2014). In addition, the high-value composites manufactured by this process brings us one step closer to closed loop composite material usage. Future research work will focus on improving the commercial viability of the developed alignment processes through investigation of more efficient dewatering methods to reduce energy use and therefore process cost.

Acknowledgements

The authors gratefully acknowledge the support of the Boeing Company under the Boeing / University of Nottingham strategic collaboration in carbon fibre recycling.

Tables and Figures

Table 1 Details of experimental factors

Experimental parameter	Parameter range	
Fibre length, mm	3	12
Fibre volume concentration in suspension, %	0.05	0.15
Wire / Jet velocity ratio	1.5	5
Dewatering vacuum level	Low (0.25bar)	High (0.5bar)
Number of replicates	2	2

Table 2 Details of fibre property

Fibre type	Fibre diameter	Tensile modulus	Tensile strength	Ultimate elongation
	μm	GPa	MPa	%
Tenax®-A HT C124_3mm	7	225	4275	1.9
Tenax®-A HT C124_12mm	7	225	4275	1.9
Toray_T800S_7mm	5	294	5880	2.0
HS recycled_ 1.15mm	7*	225*	4275*	1.9*

*Estimated fibre property

Table 3 Specification of aligned preforms and composite samples

Fibre length	Fibre type	Fibre tape areal density	Fibre volume fraction	No. of layers fibre tape	Tensile testing specimen dimensions	No. of specimens
mm	/	g/m ²	%	/	l × w × t (mm)	/
3	Tenax®-A HT C124	152	46	2	250×25.17×3.7	3
12	Tenax®-A HT C124	99	40	3	250×25.22×4.3	3
7	Toray_T800S	58	42	3	250×25.06×2.3	3
1.15	HS recycled	131	38	3	250×25.03×5.6	3

Table 4 Tensile test results

Fibre type	Fibre volume fraction	Experimental tensile modulus	Modulus SD	Experimental tensile strength	Strength SD
	Vf%	GPa	GPa	MPa	MPa
Tenax®-A HT C124_3mm	46	98	2.58	826	71.9
Tenax®-A HT C124_12mm	40	62	3.02	458	202.9
Toray_T800S_7mm	42	87	0.67	1417	135.6
HS recycled_1.15mm	38	60	4.48	322	25.6

Table 5 Theoretical prediction of composite mechanical properties

Fibre type	Fibre volume fraction	Fibre orientation distribution	Fibre length distribution factor	Predicted tensile modulus	$\left \frac{(E_{Expt.} - E_{Predict})}{E_{Predict}} \right \times 100\%$
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	factor				
	Vf%	η_0	η_1	GPa	%
Tenax®-A HT C124_3mm	46	0.976	0.987	103	5
Tenax®-A HT C124_12mm	40	0.949	0.997	86	28
Toray_T800 _7mm	42	0.960	0.995	121	28
HS recycled _1.15mm	38	0.941	0.965	79	24

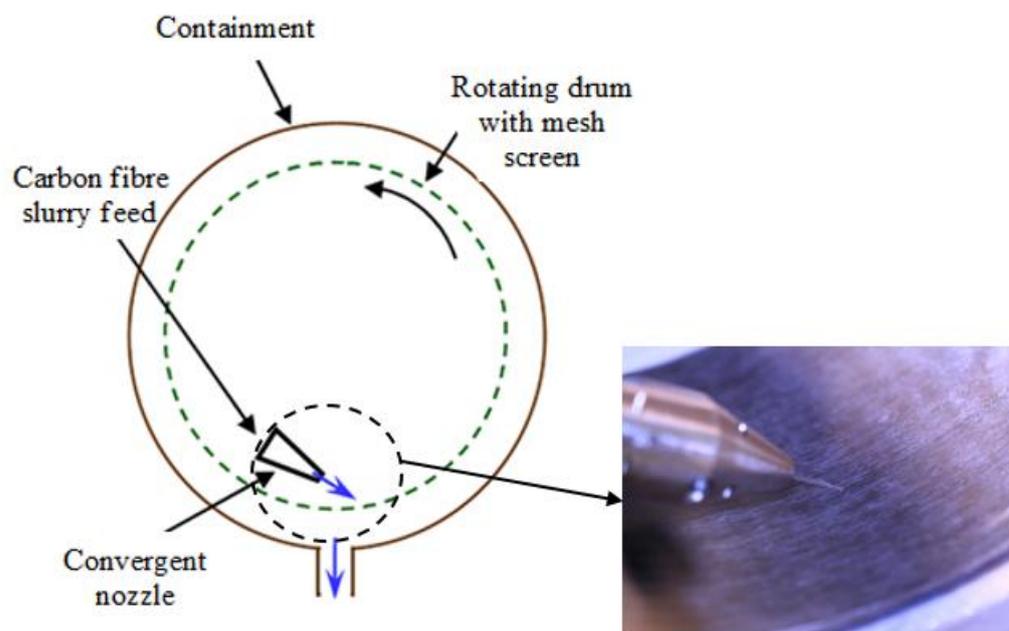


Figure 1 A schematic representation of the hydrodynamic alignment process

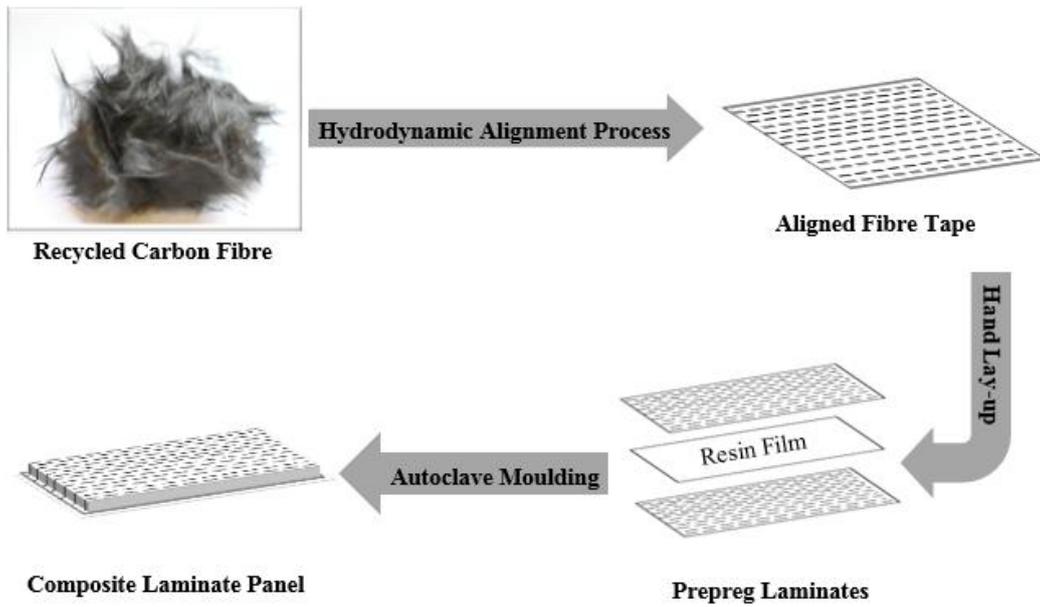


Figure 2 A schematic representation of the manufacturing process of aligned fibre laminate composites

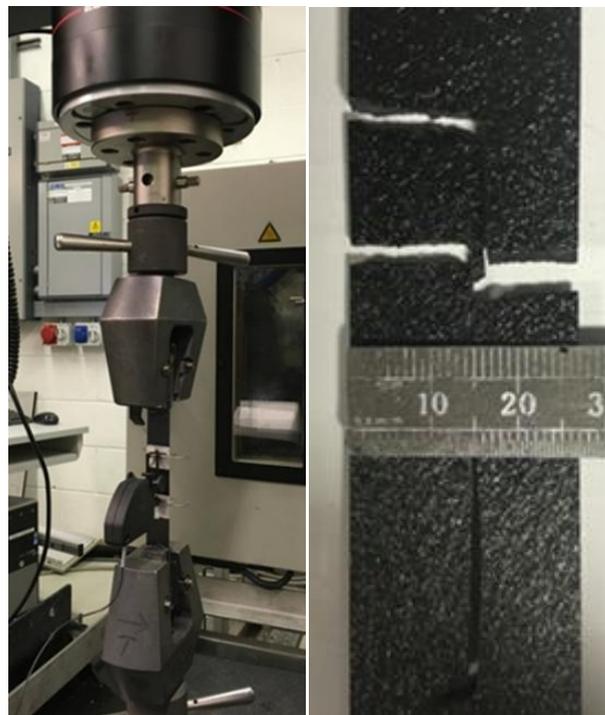
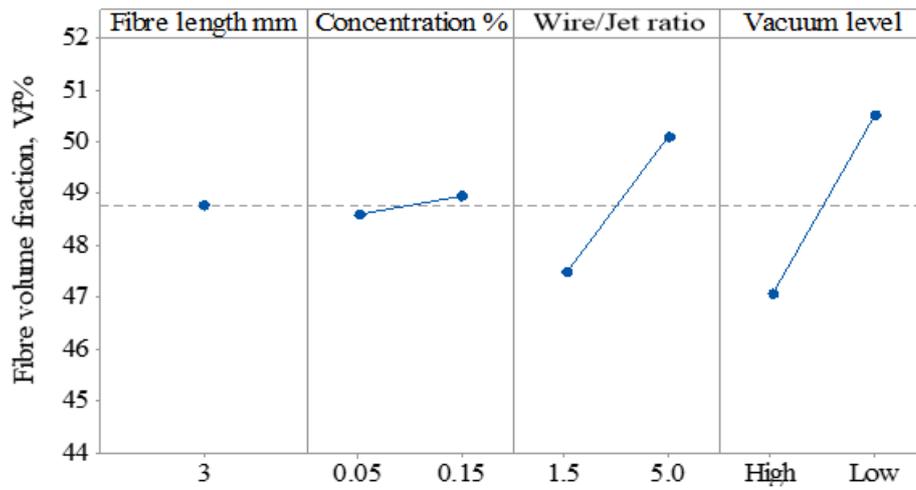


Figure 3 Tensile testing setup and typical fracture cross section of 3mm fibre laminate composite specimen

Main Effect Plot for 3mm Aligned Fibre Mats



Main Effect Plot for 12mm Aligned Fibre Mats

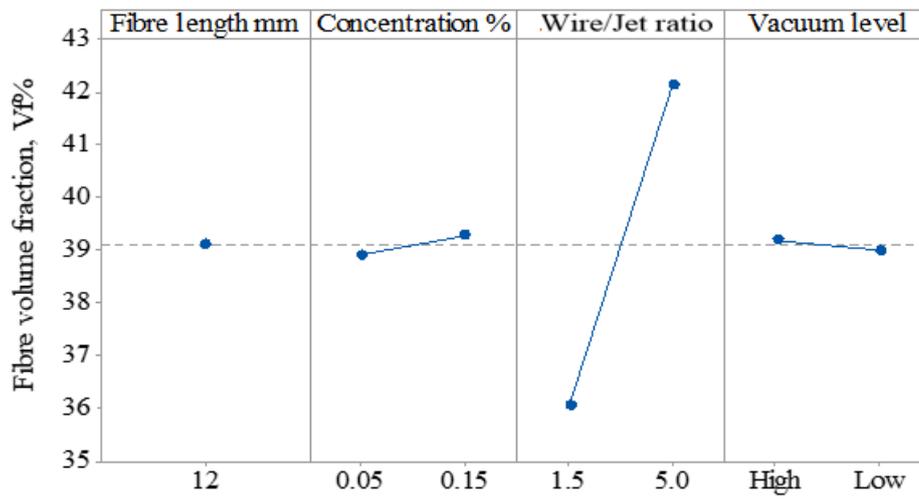


Figure 4 Main effect plots of compaction test results from full factorial DoE

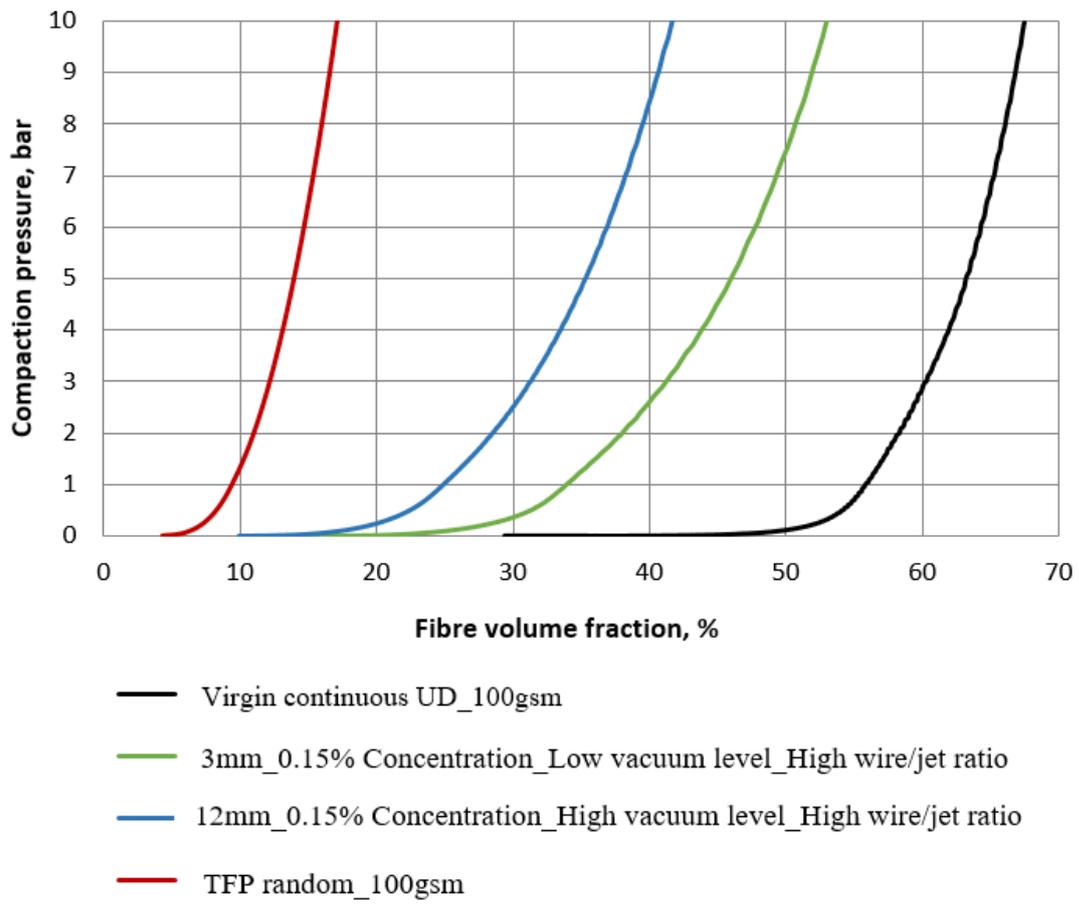


Figure 5 Dry fibre tape compaction test results

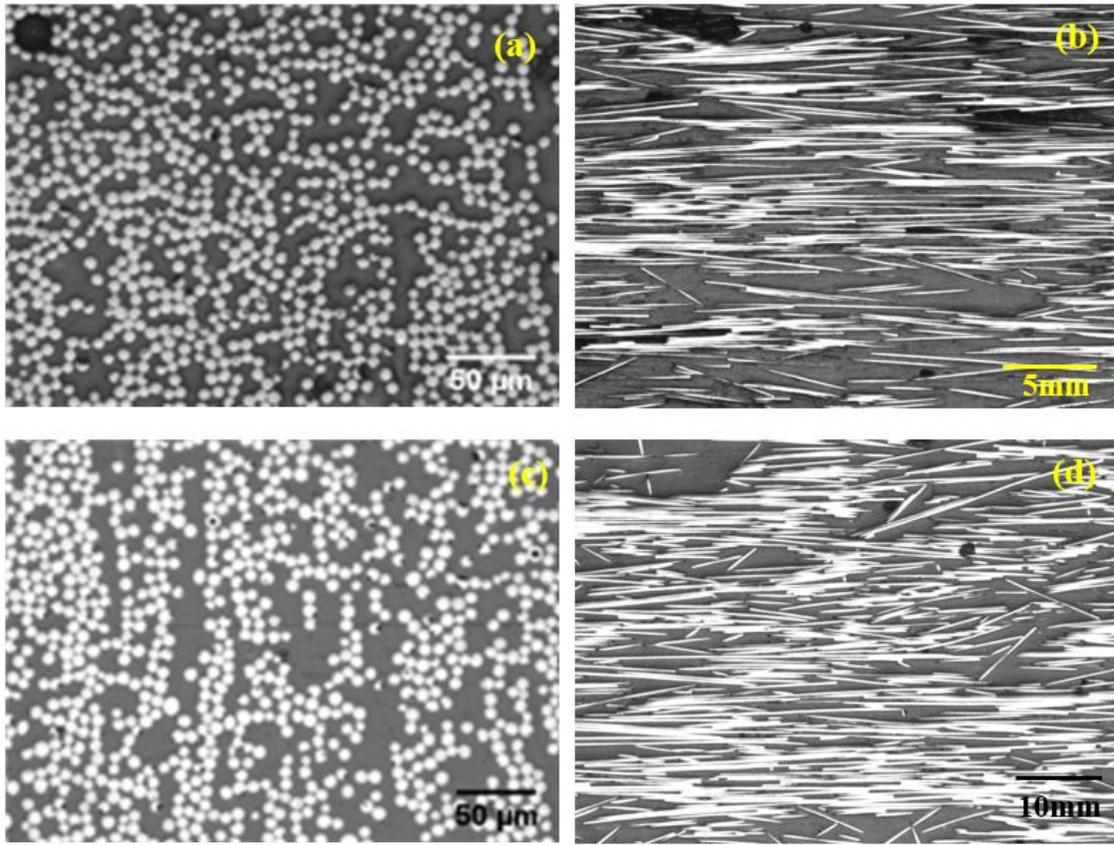


Figure 6 Micrograph of aligned composite cross section and in-plane view: (a)(b)3mm_46% fibre volume fraction; (c)(d)12mm_43% fibre volume fraction

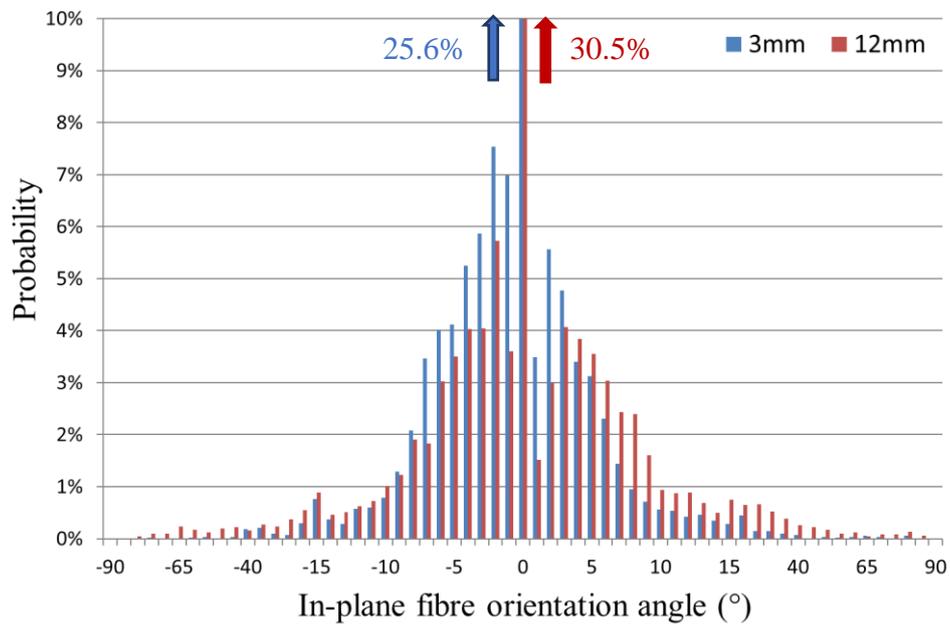


Figure 7 In plane fibre orientation distribution for composite specimens made by 3mm fibre (fibre orientation distribution factor $\eta_0 = 0.977$) and 12mm fibre (fibre orientation distribution factor $\eta_0 = 0.949$). Note that the y-axis is truncated.

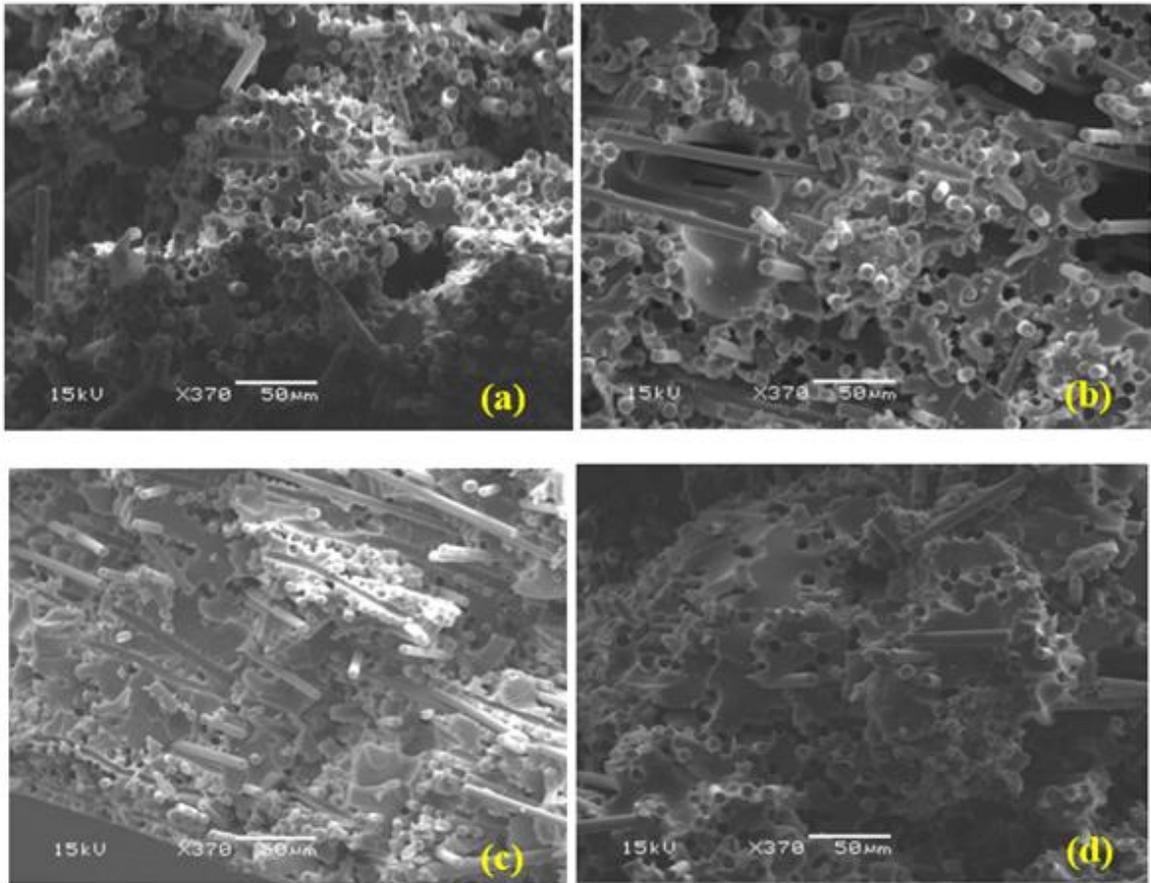


Figure 8 Fracture surface images obtained by SEM: (a) 3mm fibre specimen; (b) 12mm fibre specimen; (c) recycled fibre specimen; (d) 7mm_T800 fibre specimen

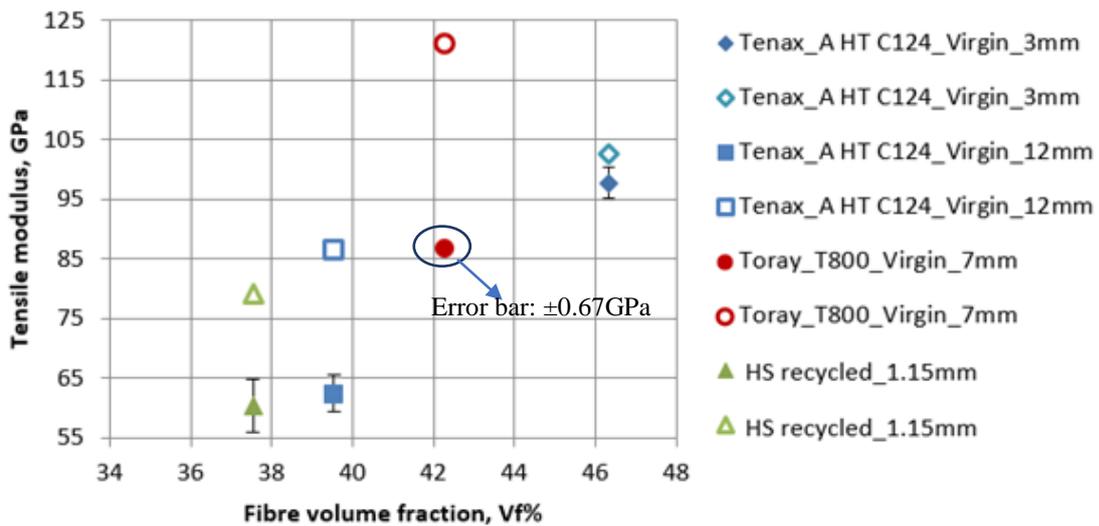


Figure 9 Experimental (solid marker) and predicted (hollow marker) tensile properties of

composites reinforced with 3mm,7mm and 12mm aligned fibre tapes and 1.15mm aligned recycled fibre tapes

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Development of high performance recycled carbon fibre composites with an advanced hydrodynamic fibre alignment process

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Highlights

- Recycled carbon fibre composites have great potential for vehicle light-weighting
- High-performance materials have been produced through the developed process
- Processing factors influencing the alignment quality are addressed
- A highly aligned fibre orientation distribution is observed enhancing fibre packing
- The observed mechanical properties compete with metals for transport applications

Abstract

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4 Carbon fibre composites have great potential for vehicle lightweighting but the high cost and
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6 environmental impact of their production tends to largely undermine the advantages in non-
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8 aerospace applications. Recycled fibre has the potential to significantly reduce both cost and
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10 environmental impact but has yet it has not been widely accepted by the composites industry
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12 due to reduced mechanical performance in components as well as the difficulties in handling
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14 and processing caused by the fluffy discontinuous form which is quite unlike any current
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16 material formats which suit existing processing methods. The developed alignment process
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18 allows discontinuous random recycled carbon fibre to be processed into tapes with a highly
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20 aligned orientation distribution. This allows composites with high fibre content to be
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22 manufactured at lower moulding pressures with the added benefit of keeping fibre length
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24 degradation to a minimum. To evaluate the effects of process factors on fibre orientation, a
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26 two-level full factorial experimental plan was designed. This represents the first time a
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28 systematic study of input parameters on final part performance has been published in the
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30 open literature. With further improvements to the process, it is shown that it is possible to
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32 manufacture a composite achieving high fibre volume content (46%) under 7 bar moulding
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34 pressure in an autoclave, exhibiting competitive mechanical properties with almost 100 GPa
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36 tensile modulus and over 800 MPa tensile strength.
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1. Introduction

In the light of steadily increasing global environmental pressure, the European Union has set stringent CO₂ emissions regulations for both the commercial aviation and automotive sectors (EuropeanCommission 2014, Regulation(EU)-2019/63 2019). The CO₂ emissions of all new cars in 2021 is required to be 40% lower than the 2015 average (Regulation(EU)-2019/63 2019), while aircraft produced from 2024 should achieve a 20% - 30% reduction in CO₂ emissions, compared to the 2014 level (EuropeanCommission 2014). Hence, lightweight design has become a major research theme in the aerospace and automotive sectors to reduce emissions and improve fuel efficiency. By virtue of their high specific mechanical properties, carbon fibre reinforced composite materials are widely used in modern transport vehicles. For example, the usage of composites in commercial aircraft has reached about 50 wt.% (Brosius 2007), and a similar trend can be found in automotive applications like the BMW i3 (Starke 2016). With increasing interest in the use of composite materials, their recyclability and the environmental impact of their production becomes an issue. Life cycle analysis for composites has drawn attention from researchers (Li, Bai et al. 2016, Wu, Xia et al. 2018, Cousins, Suzuki et al. 2019, Hagnell and Åkermo 2019), because at least 95% of a new vehicle by weight needs to be recycled at the end of life and 85% of the waste should be reused since 2015 according to the ELV directive (Directive-2000/53/EC 2014, Mudgal, Kong et al. 2014).

Composite materials are complex from a recycling & reuse perspective as the fibres themselves are universally processed into a composite form with other materials which then normally require separation at the recycling stage. It is also not sufficient to produce high quality recycled fibres; they must either be able to be incorporated into normal composites or must be able to be incorporated into an alternative high performance intermediate material. To date, the full potential of recycled carbon fibres has not been realised and efforts are ongoing

1 to improve the mechanical properties achievable such that closing the loop becomes more
2 realistic. The difference in elastic modulus between virgin and recycled carbon fibre
3 filaments themselves tends not to be significant (Pickering 2015), but some degradation of
4 strength can typically be observed, depending on the nature of the recycling process. Virgin
5 fibre is continuous at the point of manufacture and can be readily woven into the formats that
6 existing high-grade applications demand. In contrast, recycled carbon fibre cannot be a direct
7 substitute for virgin fibre as it lacks sizing, has a short fibre length, is discontinuous in nature
8 and fluffy in form. Thus, one option is to develop further manufacturing processes to improve
9 the orientation distribution and convert the material into an intermediate format that can be
10 processed via conventional routes. A circular closed-loop material cycle of carbon fibres has
11 been examined (Hagnell and Åkermo 2019), the author points out that the reclaimed fibre
12 length is the key factor in determining the potential reuse value of recycled carbon fibre in
13 high-performance product design. After the first recycling stage, the virgin carbon fibre
14 composite components will be reused as ASMC (advanced sheet moulding compound) with
15 fibre length between 25-50mm, which are introduced into a compression moulded structural
16 component for automotive applications. This is then processed through the secondary
17 recycling process with further fibre length reduction (fibre length between 2mm and 25mm).
18 As the fibre length becomes too short for efficient alignment with the current technologies,
19 these recyclates will be converted into non-woven mat or SMC (Oliveux, Dandy et al. 2015)
20 which can only offer lower volume fraction with random fibre orientation and may be
21 acceptable for the manufacture of semi- or non-structural components.
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51 Meanwhile, members of the recycling industry are seeking new processes for higher-value
52 fibre remanufacturing, and the concept of aligning the fibres is considered to be a competitive
53 conversion option as it allows high fibre packing fraction (and thus high mechanical
54 properties) to be achieved even under low compressing pressure which can avoid fibre
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breakage and save energy. Several fibre alignment processes have been developed. Under a magnetic field, short carbon fibre with a length less than 500µm can be aligned due to its diamagnetic anisotropic property (Matthews M 1996). A similar method has been explored to control the orientation distribution of carbon nanotubes or polymer fibres by electric fields (Park, Wilkinson et al. 2006, Seyam, Cai et al. 2008, Yousefi, Sun et al. 2014). In the Traditional textile industry, carding has been widely used to guide the fibres to produce a fibre web which lacks sufficient homogeneous and aligned fibre orientation. In addition, as a very brittle material, carbon fibre can be severely damaged under this intense mechanical action. By combining recycled carbon fibre with glass fibre or thermoplastic fibre, hybrid prepreg tow (Yu, Potter et al. 2014) and yarn (Hasan, Nitsche et al. 2018) has been developed. The former has been manufactured through a patented technology HiPerDiF. In this process, water has been used to disperse the fibres. Then the fibre suspension spreads through a nozzle on a corresponding plate. Then the fibres tend to align parallel to the plates and deposit on the perforated moving belt. Vacuum suction channel has been attached at the bottom to dewater the residual liquid and maintain the aligned fibre orientation. In the end the fibre tape is dried and resin impregnated by the application of heat and pressure to form prepregs. The yarn has been produced by an optimised DREF-3000 friction spinning machine. The process consists of three steps: carding (to form card webs), drawing (to form draw frame slivers), and spinning (to manufacture hybrid yarn structures with a core-sheath structure). In this method, the feed material is mainly longer reclaimed fibres >50mm from the dry fabric waste. Benefitting from the aligned fibre orientation, these two candidates can potentially be used as substitutes for virgin fibre products, but secondary conversion processes are required, e.g. weaving, to convert them to either prepreg or preform before moulding. Though the application of hybrid prepreg tow in 3D printing has been investigated, it still cannot fit the rapid production and high mechanical property requirements of large structural components.

1 During the late 1960s and 70s, hydrodynamic converging flow alignment processes attracted
2 researchers' attention. Three processes (Parratt 1968, Bagg, Dingle et al. 1971, Bagg 1977)
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4 were developed by researchers from ERDE, the Explosives Research and Development
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6 Establishment UK. Among these, the filtration process is the most widely published
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8 technique. Initially, a batch scale rig was developed to manufacture aligned fibre tape up to
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10 size 400 mm x 300 mm. This process was then upgraded into a continuous one for a sheet roll
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12 with 152 mm width (Bagg, Evans et al. 1969). A diluted glycerine/water solution was applied
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14 to disperse discontinuous fibres. As a Newtonian fluid, the viscosity of glycerine/water
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16 solution will remain constant during the dispersing process if the temperature remains
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18 constant. Longer fibres and a higher fibre concentration in the suspension are always
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20 preferred in order to achieve high mechanical performance and high production rate. With a
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22 carefully selected short fibre length, low fibre volume concentration in suspension and the
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24 high shear rate offered by the high viscosity Newtonian dispersion medium, a good
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26 dispersion quality can be achieved (i.e. undispersed bundles were not observed). The fully
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28 dispersed fibre suspension was then conveyed towards a convergent headbox with a narrow-
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30 slit exit. The headbox was attached above a transferring forming fabric and travelled
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32 backwards and forwards relative to the mesh repeatedly to deposit fibre suspension on a
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34 region of the forming mesh. Meanwhile, a vacuum suction was attached under the mesh to
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36 dewater the residual dispersing medium. In the end, water spray was applied to gently wash
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38 the damp tape, and then binder was applied.

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49 The centrifugal process was built with two significant modifications compared to the
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51 filtration process. A series of nozzles was used instead of the converging headbox as the
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53 nozzle can offer a two-dimensional convergence contour along the flow direction. A rotating
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55 drum with a layer of forming fabric attached at the internal surface was used as the
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1 centrifugal force could accelerate dewatering. It was pointed out that the centrifugal process
2 can offer better alignment quality along with higher throughput (Salariya and Pittman 1980).
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5 In the present study, an improved centrifugal rig has been constructed inspired by the ERDE
6 work. A 2-level full factorial experiment was designed to investigate the effect of four
7 operating conditions on fibre orientation. The volume fraction and fibre orientation
8 distribution within the aligned tape were evaluated by analysing micrographs of both the fibre
9 elliptical cross-sectional geometry and in-plane fibre orientation geometry. Wong (Wong,
10 Turner et al. 2009) surmised that a higher level of alignment implies that the fibres are more
11 tightly packed, which results in a high fibre volume content under the same pressure. Dry mat
12 compaction testing was therefore used as the primary testing method to evaluate the resulting
13 fibre volume fraction in all aligned tapes under different loading pressure, and this data was
14 used to determine the influence of the four factors. The summarised results of the
15 compaction testing allowed the down-selection of the optimised manufacturing conditions of
16 the alignment process. For these tapes, tensile testing was used to evaluate the mechanical
17 properties of aligned discontinued carbon fibre moulded laminates.
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39 **2. Methods and materials**

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42 In this research work, a hydrodynamic short fibre alignment process was used to manufacture
43 highly aligned short fibre tape. A 2-level full factorial experiment was designed to investigate
44 the effect of four operating conditions on fibre orientation. The alignment quality of each tape
45 was evaluated through dry fibre tape compaction testing. After analysing the results, the
46 critical influencing factors were identified. Then the optimal aligned fibre tapes were
47 manufactured and moulded as composite specimens. The mechanical performance of the
48 aligned short fibre composites was evaluated through analysing the experimental results of
49 tensile testing.
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2.1 Aligned fibre tape manufacturing with the hydrodynamic alignment process

The hydrodynamic fibre alignment process, as described in Figure 1, was used in this work. The fully dispersed fibre suspension is gently transferred into an intermediate pressurised tank which ensures that the flow through the conical nozzle is steady. The nozzle is attached on a linear actuator that keeps moving transversely to reach the desired width of fibre tape. By gathering the fibre volume concentration in suspension, jet volume flowrate, nozzle moving speed, fibre density and the area of deposited tape (125mm×942mm), the areal density of a single layer of tape can be calculated. The nozzle travel cycle is controlled and monitored by machine controller so the number of deposited layers can be counted and converted to the total areal density of tape. A forming fabric is placed inside the drum and acting as a filtration medium to detach fibres from the dispersing medium, and it also serves as a vacuum pressure distribution medium to create an even and highly efficient dewatering behaviour. The filtered glycerine is recirculated for reuse. After the desired areal density has been achieved, the dispersion medium remaining in the tape is gently washed away with an atomised warm water, and an epoxy-based binder is then applied to maintain the aligned fibre orientation. To explore the effects of processing factors, the Design of Experiments was conducted using Minitab. The four influencing factors to be investigated in this study were fibre length (to avoid the interference from the inconsistent fibre length of recycled carbon fibre, chopped virgin fibre tow was used), fibre volume concentration in suspension, dewatering vacuum level, the ratio of rotating drum speed and the fibre suspension depositing rate. For each of the factors two value levels were selected. The randomised experiment run order was generated Minitab which includes two replicate measurements for the same factor settings (levels). The details of the 2-level full factorial experimental plan are listed in Table 1. Toho Tenax Europe GmbH supplied carbon fibre staples, Tenax®-A HT C124. The fibre lengths were 3 mm and 12 mm with a nominal filament diameter of 7 microns (Table 2). In

1 order to remove the sizing, fibres were placed inside a furnace for 15 mins at 550 °C. Univar
2 UK supplied Dow Oleon Glycerine 4810 with a glycerol content over 99.5%wt which was
3 mixed with water to reduce the viscosity to 400 mPas which was measured at 15°C with a
4 Brookfield LVDVII viscometer. Then, the solution was employed as a dispersing medium
5 and was stored in a sealed container to avoid excess absorption of atmospheric moisture. A
6 cooling system in the suspension tank was designed to help keep the viscosity of the
7 suspension constant by controlling the temperature.
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19 **2.2 Dry fibre tape compaction testing**

21 Compaction testing is a better way of discerning small differences in alignment quality than
22 tensile testing, although the latter clearly provides a definitive guide to performance the
23 results are confounded by several additional factors (such as resin mechanical properties,
24 moulding conditions, errors in the tensile testing process etc.). A compaction test rig was
25 used to measure the thickness of carbon fibre tape (in order to determine the fibre volume
26 content) with varied pressure. The testing rig consists of two flat platens; the bottom one
27 mounted on the base of the Instron 5969 universal testing machine; while the top one is
28 connected to a 10 kN load cell attached to the machine crosshead. The machine's control
29 software automatically logged the crosshead location during the test. There are two linear
30 variable differential transformers (LVDTs) attached on both sides of the upper platen to
31 record the distance between the platens accurately. The square-shaped specimens with an
32 edge length 35mm were prepared by stamping carbon fibre tape with a cutting template. The
33 areal density of each sample was accurately measured before the test. Afterwards, four layer
34 specimens were stacked together, carefully located at the centre of the bottom platen and then
35 compressed at a rate of 1 mm/min until compression pressure reached to 10 bar. The system
36 compliance was measured by two LVDTs when 10 bar pressure was applied with no
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1 specimen; the compliance curves were curve-fitted and applied as a correction factor during
2 post-processing of the data. The thickness of each tape was obtained by taking the average
3 reading values from the two LVDT units after the compliance was subtracted. Then the fibre
4 volume fraction of the tape is equal to areal density divided by fibre density and the thickness
5 of tape.
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11 **2.3 Manufacture of aligned fibre composite in an autoclave for the tensile testing**

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14 Not only short virgin fibres (Tenax®-A HT C124_3mm and 12mm, Toray_T800S_7mm) but
15 also recycled fibre was introduced in this work. End of life carbon fibre composites was
16 supplied by ELG Carbon Fibre Ltd and then shredded and recycled by the Fluidised Bed
17 process at the University of Nottingham (Pickering 2015) (described as HS recycled in Table
18 2 and Table 3). The effective number average fibre length is 1.15mm. As the source of this
19 supplied end life waste piece was not known, the fibre filament properties of HS recycled
20 fibre were assumed to be the same as Tenax®-A HT C124 fibre. The details of laminates
21 which were manufactured from these tapes are listed in Table 3. The laminates were stacked
22 with the MTM57 resin films (supplied by Solvay, Heanor, UK) for subsequent impregnation
23 and vacuum bagged to remove the trapped air. Then the compacted materials were moulded
24 in an autoclave at 120 °C, 7 bar pressure, for an hour. In the end, the composites were cooled
25 down to around 60 °C (with a cooling rate of 3°C/min) under pressure before demoulding.
26 The manufacturing procedure of aligned fibre composites is described as a schematic diagram
27 in Figure 2.
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51 Tensile tests were performed with the specimens made from these composites following the
52 testing standard ASTM D3039. A 250 kN load cell was used (Figure 3), and the testing rate
53 was 1 mm/min. For each type of laminate, four specimens were prepared. An extensometer
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1 was attached on the specimen to record strain. Optical micrographs were used to view and
2 analyse the cross-sectional of the laminates.
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6 **2.4 Fibre orientation determination**

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9 The surfaces of polished composite specimens were analysed with an optical microscope to
10 investigate the in-plane fibre orientation. The composite samples were placed at the bottom of
11 a plastic container and fibres were parallel to the container's bottom surface. Then, the
12 container was filled with Prime20LV (Gurit, UK) resin. The cured resin containing the
13 composite sample was then polished until the full length of carbon fibre can be observed. An
14 optical microscope captured all sample images then imported into AutoCAD. The fibre
15 orientation was determined by sketching a line along the fibre length direction and the line is
16 coincident with the fibre edge. The data file contains orientation angles of each fibre were
17 exported as a spreadsheet for further analysis. For each case, more than 5000 carbon fibres
18 from five microscope images were processed to identify the orientation distribution. Figure 6
19 shows the examples of the aligned short fibre composite specimens which were used for in-
20 plane fibre orientation analysing. Finally, the fibre orientation distribution results of 3mm
21 fibre composites and 12mm fibre composites were obtained in Figure 7.
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42 **3. Results and discussion**

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45 In this study, the optimal processing factors of the hydrodynamic alignment process has been
46 identified. The mechanical property of aligned fibre composites has been evaluated and
47 analysed.
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52 **3.1 Dry fibre tape compaction results**

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55 In previous research work, with 10 bar compaction pressure, 3mm fibre aligned tape has been
56 shown to achieve 47.7% fibre volume fraction and 12mm fibre aligned tape obtained 32.5%
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1 fibre volume content (Liu, Wong et al. 2015). To improve the process efficiency and achieve
2 higher fibre volume fraction, the processing factors influencing alignment quality are
3 explored in this paper.
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8 Thirty-two discontinuous aligned fibre tapes were manufactured using the described
9 alignment process and tested using the developed compaction test. Based on the compaction
10 test results of the full factorial test, the main effects plots for fibre volume fraction are shown
11 in Figure 4. Fibre volume concentration in the dispersion medium is not a significant factor in
12 determining the alignment of 3mm fibre and 12 mm fibre which is thought to be due to the
13 improved dispersion process where the fibres were distributed more homogeneously in the
14 suspension. Higher wire/jet velocity ratio is beneficial to the fibre alignment for both 3mm
15 and 12 mm fibres, as it can drive fibres along the flow direction and avoids fibres bending as
16 they are deposited on the nylon mesh. The vacuum level applied around the rotating drum is
17 another critical factor: The drum contains multiple vacuum slots to enable the residual
18 glycerine flow can be dewatered. If high vacuum level was applied, a locally concentrated
19 high vacuum suction region is generated around the vacuum slots. The fibres will tend to
20 move along the flow direction and be accumulated around the vacuum slots. Then the pre-
21 aligned orientation will be damaged. Thus, a lower vacuum level is beneficial for 3mm fibre
22 to maintain aligned fibre orientation, which is validated by DoE results plotted with Minitab
23 in Figure 4.
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47 The compaction result curves are plotted in Figure 5. This includes two benchmarks for
48 reference: a 2D planar carbon fibre 100gsm random tape (supplied by Technical Fibre
49 Products Ltd) and a 100gsm unidirectional continuous fibre (UD) tape (supplied by Easy
50 Composites Ltd) as well as two aligned tapes from the full factorial test which are made with
51 3mm and 12mm Tenax®-A HT C124 virgin fibres respectively. These two obtained highest
52 fibre volume fractions in the compaction test results for their respective fibre length. Under
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1 10bar compression pressure, the fibre volume content of UD tape achieves 68% while the
2 random tape can only obtain 18% fibre volume fraction which indicates that fibre packing
3 efficiency can be significantly improved by the aligned fibre orientation. With 3mm fibre, the
4 tape exhibits 52% fibre volume fraction under 10bar compaction pressure. The manufacturing
5 conditions are 0.15% fibre volume concentration in the suspension, five times higher drum
6 tangential rotating speed relative to the suspension jet velocity and low dewatering vacuum
7 level. For 12mm fibre tape, 41% fibre volume fraction was achieved, and the processing
8 factors are 0.15% fibre volume concentration suspension, five times wire to jet speed ratio
9 and high dewatering pressure. A tape made from aligned 3 mm fibres can achieve a 27%
10 higher volume fraction than the tapes with 12 mm fibres. This is own to a better fibre
11 dispersion quality, which means no fibre bundle remains with shorter fibre.
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28 **3.2 Fibre orientation evaluation**

29 In previous research work, 70% of the fibres were found to be in the range $\pm 4^\circ$ misalignment
30 angle for a filtration process with rotating drum (Richter 1980). For the ERDE filtration
31 process, it was reported that 90% of the fibres were in the range of $\pm 15^\circ$ (Salariya and
32 Pittman 1980). In the present work, based on the fibre orientation distribution results in
33 Figure 7, there are 92% of fibres in the range $\pm 10^\circ$ and 60% of fibres are in the range of $\pm 3^\circ$
34 in the 3mm fibre composite samples which have 46% fibre volume fraction. 86% of fibres in
35 the range $\pm 10^\circ$ and 53% of fibres were in the range of $\pm 3^\circ$ in 12mm fibre composite samples,
36 which have 43% fibre volume content. Although the optimised processing factors were
37 chosen, the composite specimens manufactured with 12mm fibre have a higher degree of
38 misalignment. The reason was explained by analysing the effects of the fibre length on
39 suspension jet stability and alignment quality (Liu, Turner et al. 2019). For the suspension
40 jets with same fibre volume concentration, the 12mm fibre-containing suspension has 4 times
41 larger fibre aspect ratio compared to that of 3mm fibre-containing suspension which leads to
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1 a more significant fibre-fibre interaction. Thus, even though the fibre concentration,
2 suspension viscosity and jet Re number are same, the use of 12 mm fibre can make the jet
3 more unstable and resulting a fibre misalignment. Comparing with the results for 3mm fibre
4 aligned tape obtained in the dry fibre tape compaction test, at the same loading pressure, the
5 fibre volume content of moulded composites is reduced from 49% to 46% which indicates
6 that the fibre alignment quality of the cured composite samples was slightly degraded
7 following the post-processing operations – residual glycerine washing, applying binder and
8 laminating.
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21 **3.3 Mechanical properties**

22 In Table 4, the results from the tensile test are listed, which include the average fibre volume
23 fraction of each specimen, mean values and standard deviations of modulus and strength. The
24 fibre lengths investigated in this paper are longer than the critical fibre length. Thus, fibre
25 breakage results rather than matrix or interface failure. Thus, extensive fibre pull-out situation
26 was not witnessed at the fracture surface of both virgin fibre samples and recycled fibre ones.
27 The fracture surface images were obtained using Scanning Electron Microscopy (JEOL
28 6060LV) and are listed in Figure 8.
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41 For comparison, a modified rule of mixtures for modulus (Sanadi and Piggott 1985, Coleman,
42 Khan et al. 2006) was used to produce an analytical modulus calculation. The expression is
43 given by:
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$$49 \quad E_c = \eta_0 \eta_1 V_f E_f + V_m E_m \quad \text{Equation 1}$$

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52 Where E and V are Young's modulus and volume fraction; and the subscripts c, f, and m
53 refer to the composite, fibres, and matrix.
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58 η_0 = fibre orientation distribution factor
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η_1 = fibre length distribution factor

The fibre orientation distribution factor can be calculated by Krenchel's equation (Krenchel 1964):

$$\eta_0 = \sum_{i=0}^{180^\circ} V_{fi} (\cos \theta_i)^4 \quad \text{Equation 2}$$

Where θ is the measured fibre orientation angle.

Based on the shear lag theory (Cox 1952), the fibre length distribution factor η_1 can be described:

$$\eta_1 = 1 - \frac{\tanh(\alpha l/d_f)}{\alpha l/d_f} \quad \text{Equation 3}$$

$$\alpha = \sqrt{\frac{-3E_m}{2E_f \ln V_f}} \quad \text{Equation 4}$$

Where d_f is fibre diameter.

The values of η_0 and η_1 are listed in Table 5. The comparison between the theoretical predictions and experimental results are plotted in Figure 9. Although the moulding pressure is only 0.7MPa which is much lower than the standard compression moulding processes (5–10 MPa), the composites made by the developed hydrodynamic alignment process obtained an excellent fibre packing efficiency due to the highly aligned fibre orientation. In the issue, high tensile modulus and strength were achieved. With 46% fibre volume fraction, the composite made from 3 mm aligned fibre gave a 98 GPa tensile modulus which achieves the closest agreement with the predicted value. With the same curing pressure (7 bar), the laminate made with 12 mm fibre can only achieve 40% fibre volume content and obtain 62 GPa tensile modulus which is 28% lower than the predicted value (Table 5). The reason for this discrepancy is thought to be that the 12mm fibre tape has a higher porosity compared to

1 that of 3mm fibre aligned tape. Thus, during dewatering, the through-thickness pressure
2 difference of 12 mm fibre tape is lower (due to air leakage) when the tape is not fully covered
3 by glycerine, which leads to the ineffective removal of residual glycerine. On the other hand,
4 the larger fibre aspect ratio of 12mm fibre-containing suspension results in more significant
5 fibre-fibre interaction which leads to more inconsistent jet stability. Thus, the standard
6 deviation of Tenax-A HT 12 mm composites sample is much greater than the others.
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14 Furthermore, although the same washing method was applied for both 12 mm and 3 mm fibre
15 tapes, the 12 mm tape contains a higher level of residual glycerine after washing as the
16 washing process is ineffective in dealing with the higher initial glycerine content. Residual
17 glycerine is likely to affect material properties by reducing fibre: matrix adhesion
18 significantly. The T800 fibre has a smaller diameter (5 μ m), and when the fibre suspension
19 has the same fibre volume concentration, the actual number of fibre filaments will be higher
20 than with 7 μ m diameter fibres. Thus, the fibre to fibre interaction will be more significant
21 and tend to reduce the alignment quality. As a result, T800_7mm fibre laminates exhibit 42%
22 fibre volume fraction, which is 8% lower (4% fibre volume fraction in difference) than that of
23 3mm fibre laminates. The experimental tensile modulus is 87 GPa, which is 28% lower than
24 the analytical value, and this is thought to be related to residual glycerine. 38% fibre volume
25 fraction and 60GPa tensile modulus are obtained by the composite made by short recycled
26 fibre, the average fibre length is 1.15mm, but 35% of the fibres have a length shorter than the
27 average and in around 3% of cases is below the critical fibre length of 0.38mm. It is
28 challenging to retain the aligned orientation of these very short fibres during the deposition
29 process. These properties are competitive with the alternative structural materials and show
30 the potential of fibre alignment for the manufacture of high-value composites from recycled
31 carbon fibre.
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1 Comparison with existing published results is challenging due to the variety of fibre volume
2 contents and methods of manufacture. By the conventional wet process, Sanadi and Piggott
3 (Sanadi and Piggott 1985) manufactured aligned carbon/epoxy composites with 2 mm long
4 fibre which had 35% fibre volume fraction; these materials achieved 60 GPa tensile modulus
5 and 650 MPa tensile strength. With 12mm long recycled fibre, Wong and Turner (Turner,
6 Pickering et al. 2009, Wong, Turner et al. 2009) manufactured carbon/epoxy composite
7 specimens with 45% fibre volume fraction and obtained 80 GPa tensile modulus which is 23%
8 lower than the predicted value 103 GPa. The HiPerDiF method developed by Yu (Yu, Potter
9 et al. 2014) has demonstrated properties of 115GPa tensile modulus and 1509MPa tensile
10 strength for 3mm fibre tapes, 55% fibre volume fraction was achieved using pressure
11 intensification which appears to apply more than 21bar. In this process where water is used as
12 the dispersion media, any issues related to residual glycerine are avoided, but the viscosity of
13 water may not offer enough shear stress to adequately disperse longer fibres and fluffy
14 recycled fibre from end of life parts.

35 **4. Conclusions**

36 Discontinuous tapes have been shown to exhibit significantly improved mechanical
37 properties when the alignment level of the fibres is high. This improvement comes both from
38 higher fibre volume content in the composite and avoiding fibre breakage in the subsequent
39 moulding process. A hydrodynamic process intending to achieve highly aligned fibre
40 orientation has been developed, and the effects on alignment quality from processing factors
41 have been explored and analysed in this paper. The composite laminates were made with
42 3mm carbon fibre under 7 bar curing pressure and had a tensile modulus of 98GPa and tensile
43 strength of 826 MPa with a fibre volume content of 46%. Genuine recycled fibre from
44 laminate scrap was also used in this work, and a tensile modulus of 60GPa and tensile
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1 strength of 322 MPa was achieved by 1.15mm mean fibre length recycled fibre laminate
2 composites with a fibre volume fraction of 38%. This indicates the capability of fibre
3 alignment technique for manufacturing of high-value composites with specific mechanical
4 properties that are not only better than other widely applied low-cost composite and metallic
5 materials but are also competitive with virgin carbon fibre composites. Hence, this paper
6 proposes an effective recycling and re-manufacturing solution for composite materials to
7 aircraft and automobile manufacturers, to overcome the challenges of minimum 95% material
8 recycled ratio and 85% material reused ratio from increasingly stringent sustainability
9 regulations (Directive-2000/53/EC 2014, Mudgal, Kong et al. 2014). In addition, the high-
10 value composites manufactured by this process brings us one step closer to closed loop
11 composite material usage. Future research work will focus on improving the commercial
12 viability of the developed alignment processes through investigation of more efficient
13 dewatering methods to reduce energy use and therefore process cost.
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35 University of Nottingham strategic collaboration in carbon fibre recycling.
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Tables and Figures

Table 1 Details of experimental factors

Experimental parameter	Parameter range	
Fibre length, mm	3	12
Fibre volume concentration in suspension, %	0.05	0.15
Wire / Jet velocity ratio	1.5	5
Dewatering vacuum level	Low (0.25bar)	High (0.5bar)
Number of replicates	2	2

Table 2 Details of fibre property

Fibre type	Fibre diameter	Tensile modulus	Tensile strength	Ultimate elongation
	μm	GPa	MPa	%
Tenax®-A HT C124_3mm	7	225	4275	1.9
Tenax®-A HT C124_12mm	7	225	4275	1.9
Toray_T800S_7mm	5	294	5880	2.0
HS recycled_ 1.15mm	7*	225*	4275*	1.9*

*Estimated fibre property

Table 3 Specification of aligned preforms and composite samples

Fibre length	Fibre type	Fibre tape areal density	Fibre volume fraction	No. of layers fibre tape	Tensile testing specimen dimensions	No. of specimens
mm	/	g/m ²	%	/	l × w × t (mm)	/
3	Tenax®-A HT C124	152	46	2	250×25.17×3.7	3
12	Tenax®-A HT C124	99	40	3	250×25.22×4.3	3
7	Toray_T800S	58	42	3	250×25.06×2.3	3
1.15	HS recycled	131	38	3	250×25.03×5.6	3

Table 4 Tensile test results

Fibre type	Fibre volume fraction	Experimental tensile modulus	Modulus SD	Experimental tensile strength	Strength SD
	Vf%	GPa	GPa	MPa	MPa
Tenax®-A HT C124_3mm	46	98	2.58	826	71.9
Tenax®-A HT C124_12mm	40	62	3.02	458	202.9
Toray_T800S_7mm	42	87	0.67	1417	135.6
HS recycled_1.15mm	38	60	4.48	322	25.6

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Table 5 Theoretical prediction of composite mechanical properties

Fibre type	Fibre volume fraction	Fibre orientation distribution factor	Fibre length distribution factor	Predicted tensile modulus	$\left \frac{(E_{Expt.} - E_{Predict})}{E_{Predict}} \right \times 100\%$
	Vf%	η_0	η_1	GPa	%
Tenax®-A HT C124_3mm	46	0.976	0.987	103	5
Tenax®-A HT C124_12mm	40	0.949	0.997	86	28
Toray_T800_7mm	42	0.960	0.995	121	28
HS recycled_1.15mm	38	0.941	0.965	79	24

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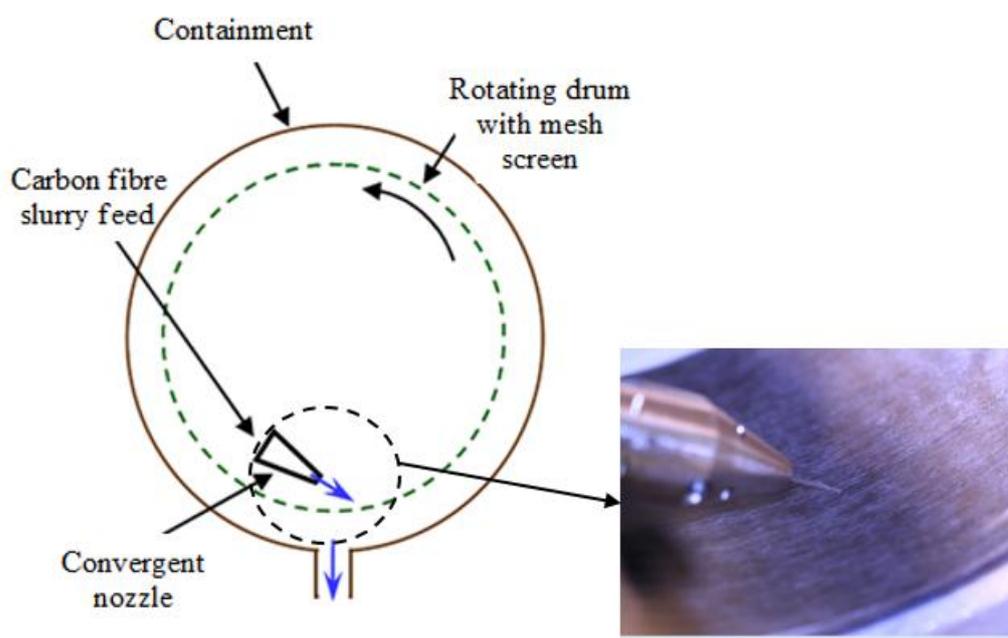


Figure 1 A schematic representation of the hydrodynamic alignment process

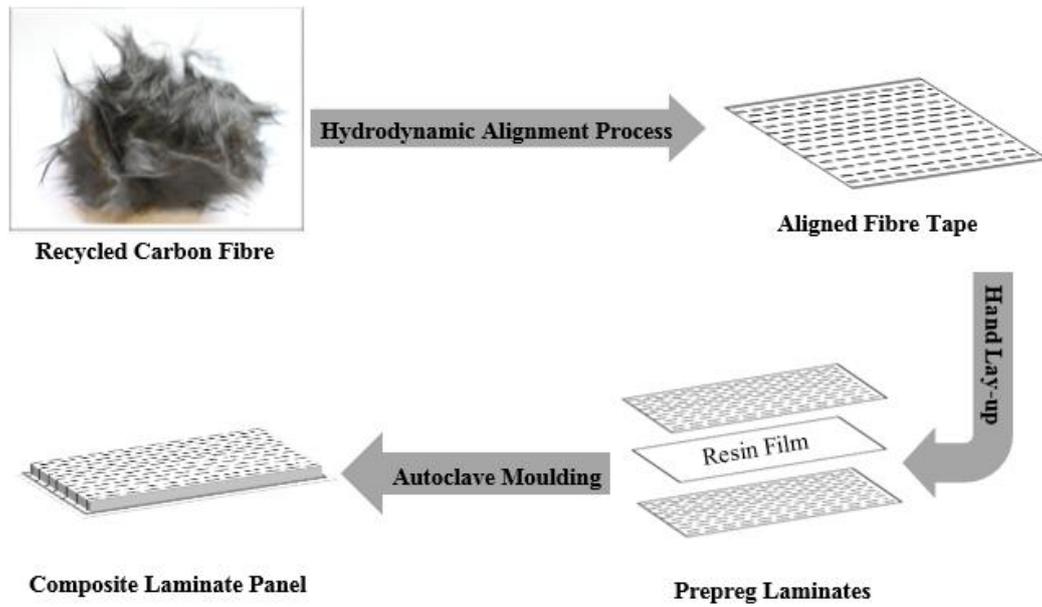


Figure 2 A schematic representation of the manufacturing process of aligned fibre laminate composites

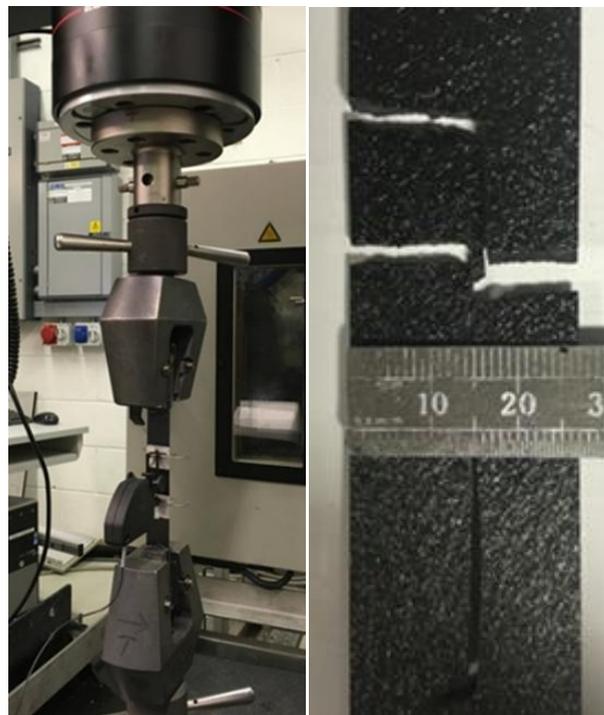


Figure 3 Tensile testing setup and typical fracture cross section of 3mm fibre laminate composite specimen

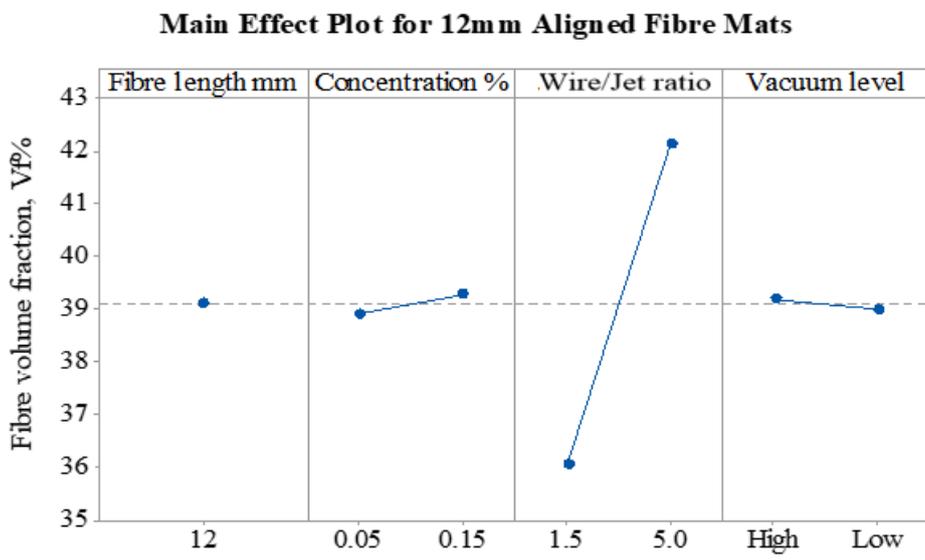
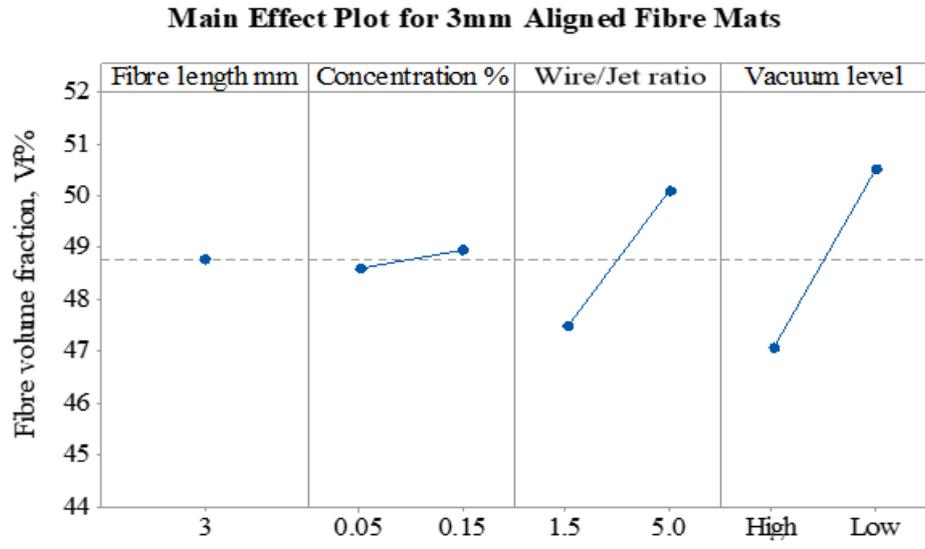


Figure 4 Main effect plots of compaction test results from full factorial DoE

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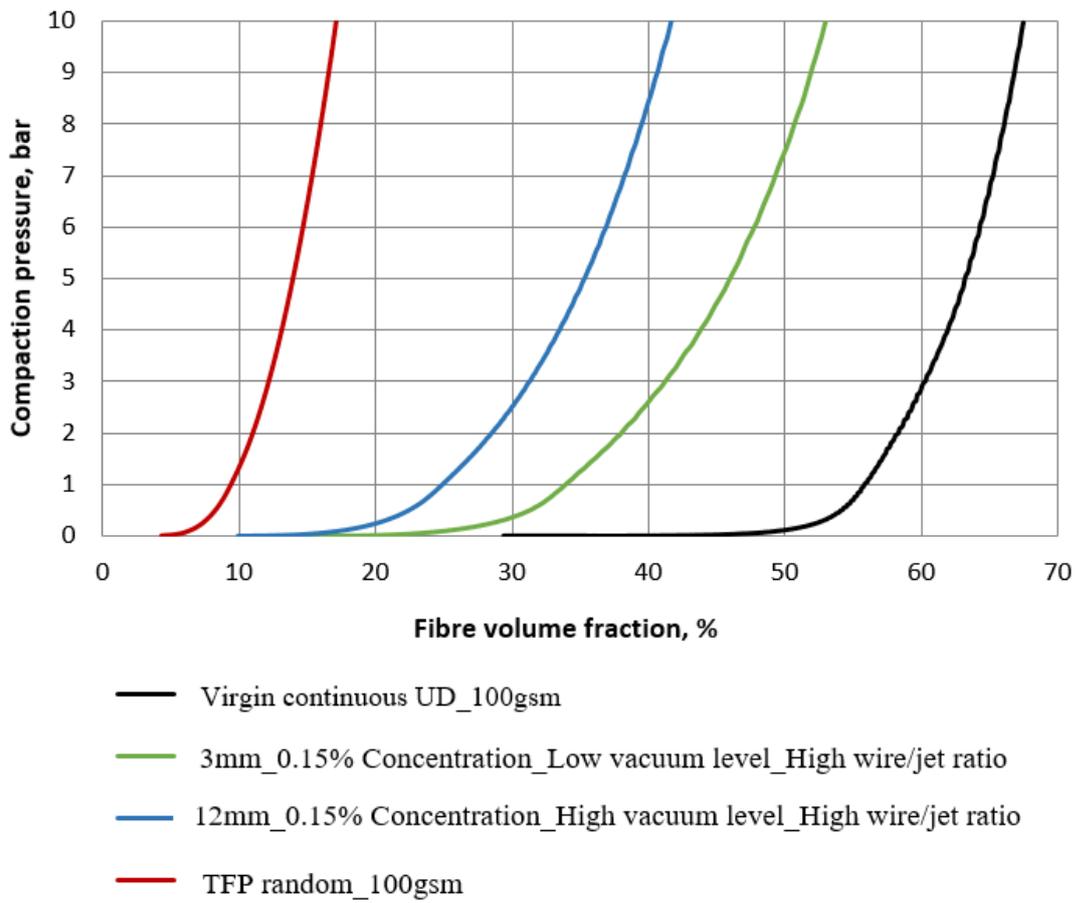


Figure 5 Dry fibre tape compaction test results

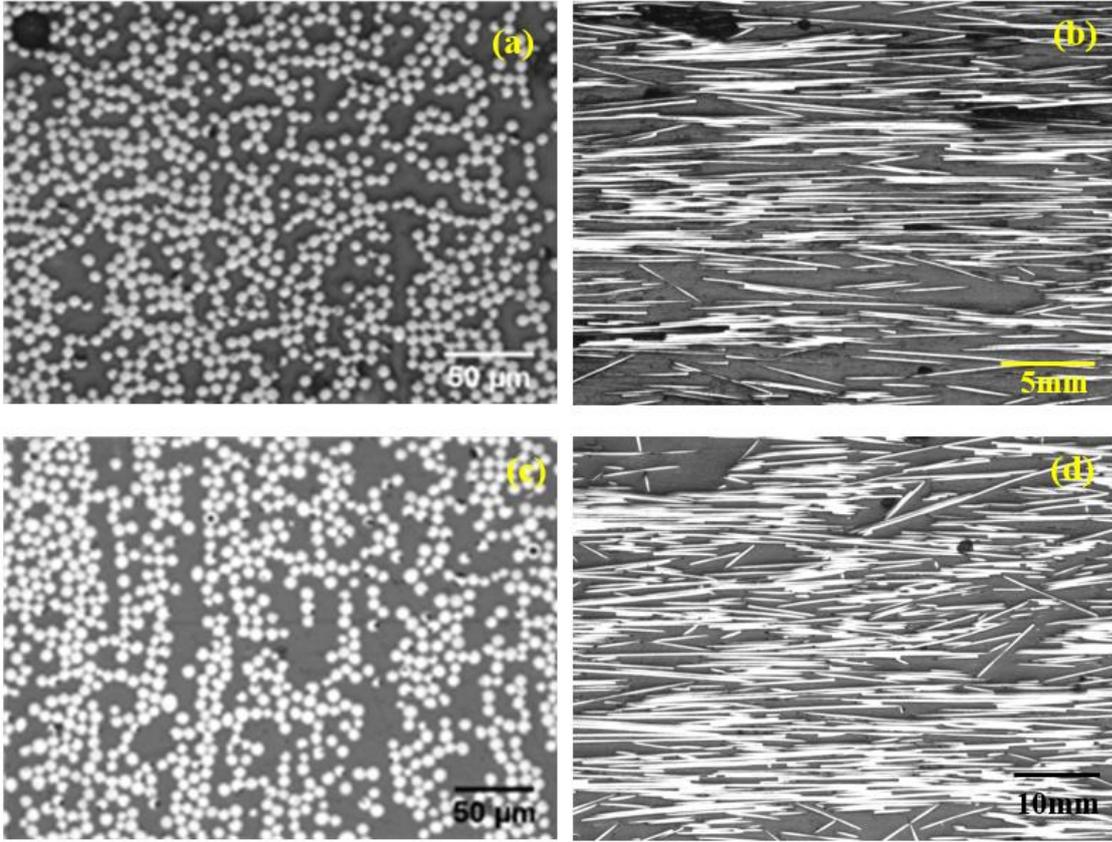


Figure 6 Micrograph of aligned composite cross section and in-plane view: (a)(b)3mm_46% fibre volume fraction; (c)(d)12mm_43% fibre volume fraction

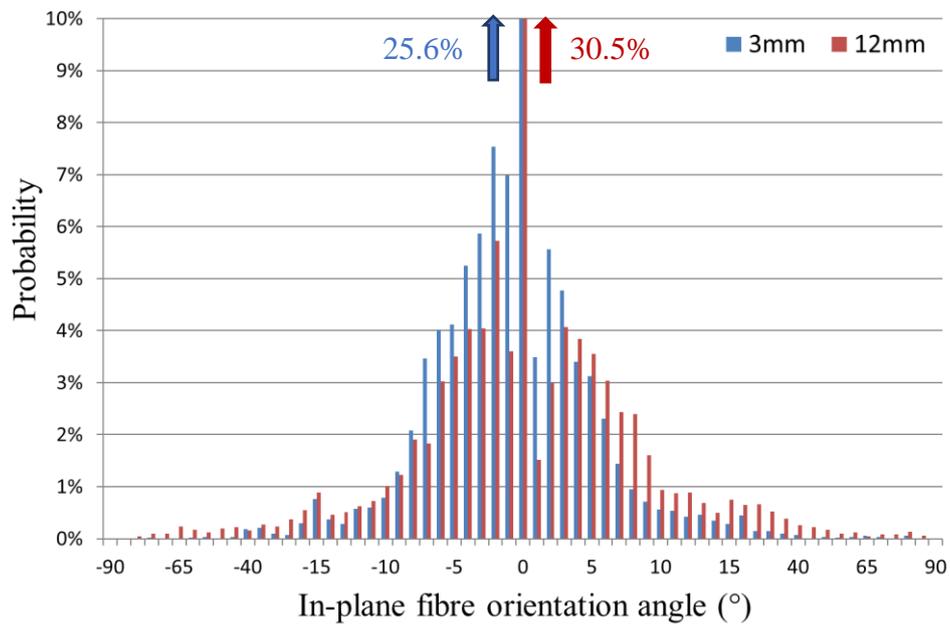


Figure 7 In plane fibre orientation distribution for composite specimens made by 3mm fibre (fibre orientation distribution factor $\eta_0 = 0.977$) and 12mm fibre (fibre orientation distribution factor $\eta_0 = 0.949$). Note that the y-axis is truncated.

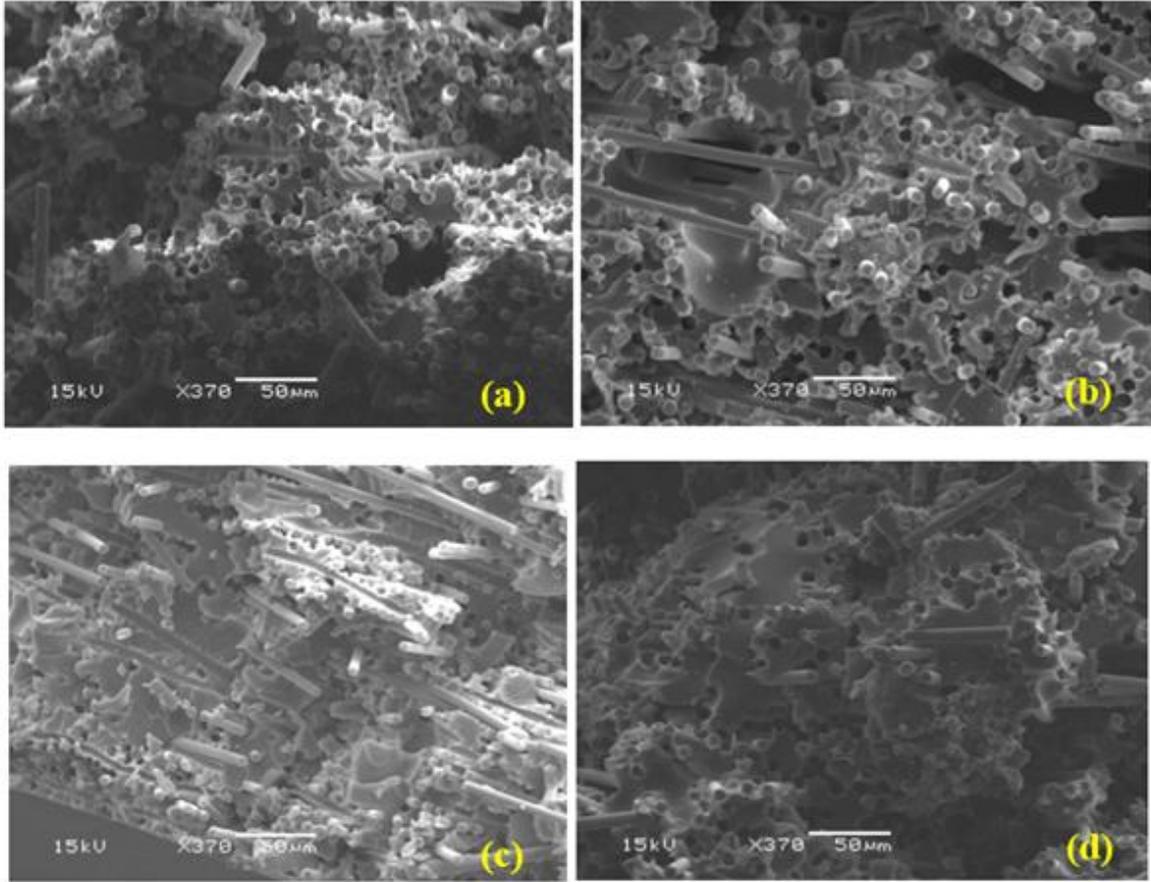


Figure 8 Fracture surface images obtained by SEM: (a) 3mm fibre specimen; (b) 12mm fibre specimen; (c) recycled fibre specimen; (d) 7mm_T800 fibre specimen

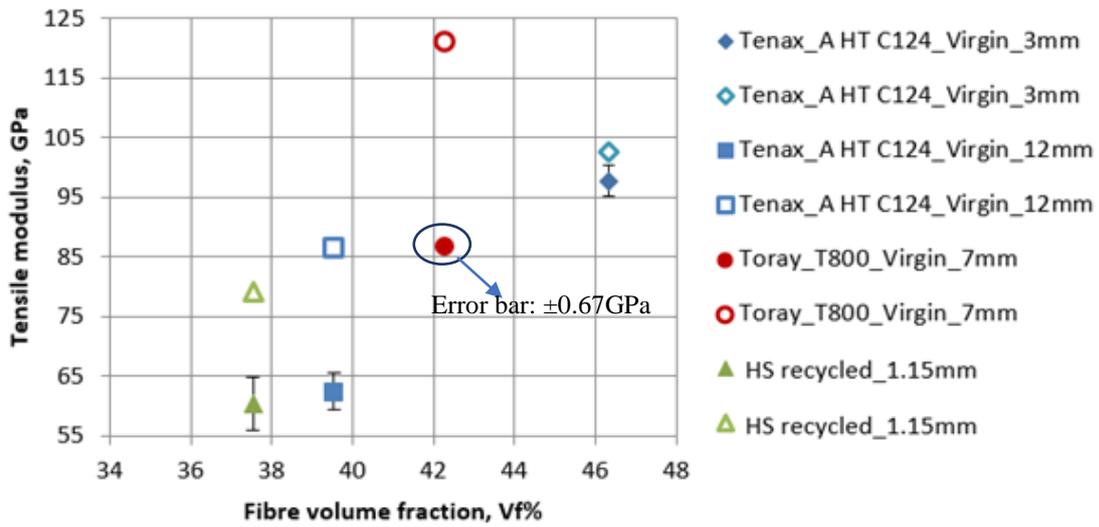


Figure 9 Experimental (solid marker) and predicted (hollow marker) tensile properties of

composites reinforced with 3mm,7mm and 12mm aligned fibre tapes and 1.15mm aligned recycled fibre tapes

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