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ALIGNED VIRGIN AND RECYCLED SHORT CARBON FIBRE HYBRID COMPOSITES

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ABSTRACT

One of the main problems in industrial applications of carbon composite materials is recycling end of life products. Usually chopped recycled fibres are randomly dispersed in materials as low-grade additives for non-structural applications; however, in order to maximize economic and functional viability, these fibres should be reused as reinforcement for high performance structural composites. The fibre alignment level is the key factor to increase the fibre volume fraction, and consequently the performance of recycled composites. Another important factor is the impact of the fibre recovery process on the mechanical properties of carbon fibres. Carbon fibres can be damaged during the fibre recovery process leading to a significant loss of strength, but the modulus is almost the same as that of virgin carbon fibres. The aim of this research is to manufacture intermingled hybrid composites using highly aligned new and recycled discontinuous carbon fibres with the HiPerDiF (High Performance Discontinuous Fibres) method to increase the availability of recycled carbon fibres. This method enables manufacturing of discontinuous fibre hybrid composites with different fibre mixing ratios; the constituents are intimately mixed and the hybridisation is achieved within the ply. In this work, the stiffness and the failure properties of aligned short fibre composites, as a function of the recycled to virgin carbon fibres ratio, are investigated. The HiPerDiF method proved to be a valuable remanufacturing method to achieve high performance short fibre composite from end of life recycled composites.

1 INTRODUCTION

The wide spread of composite material in various engineering sectors over the last decades poses the challenge of dealing with the end-of life products. Their landfill disposal has been made more difficult by the enforcement of strict legislations. Moreover, considering the high production costs of carbon fibres and an increasing demand that is barely met by the supply, recycling would lead to a profitable re-introduction in the market of a raw material that otherwise would represent an expensive waste. However, the recycling process is made difficult by the multi-phase and the cross-linked nature of the thermosetting matrix.

The less structurally interesting way to recycle composite materials is to chop or mill production waste and end-of-life parts into small pieces and burn off the thermosetting matrix, obtaining short fibres, typically below 1 mm. These fibres can be randomly dispersed in materials as low-grade additives for non-structural applications, such as for anti-static and electromagnetic interference shielding as well as for enhanced heat conduction.

However, in order to maximize economic and functional viability, recycled fibres, conveniently separated by the matrix, should be reused as reinforcement for high performance structural composites.

A complete review about the technologies to recycle carbon fibre reinforced polymers for structural applications was presented by Pimenta and Pinho [1]. The recycling process of composite materials, and in particular of carbon fibre reinforced thermosetting resins, can be divided in two stages: the fibre reclamation and the fibre remanufacturing to obtain an intermediate material or a finite product.

Amongst the fibre reclamation processes it is worth mentioning pyrolysis, i.e. thermochemical decomposition of the matrix at elevated temperatures in an inert environment [2], oxidation in a fluidised bed, i.e. the matrix elimination at high temperature in oxygen-rich flow [3], and chemical recycling in various reactive media at moderate temperature, e.g. catalytic solutions [4], benzyl alcohol [5], and supercritical fluids [6-9].

Of greater interest for the presented work is, independently from the fibre recovery process, the remanufacturing of the reclaimed fibres. When the recycling process preserves the reinforcement architecture of the waste this can be used as it is [10]. However, the size-reduction of CFRP waste before reclamation, the fibre breakage during reclamation and the chopping of the fibres after reclamation lead to recycled fibres that are usually fragmented in short length. This makes direct moulding techniques, e.g. injection moulding [11] and bulk moulding compound compression [12], and the compression moulding of intermediate random [13] or aligned mats [14] more interesting for industrial applications. To deliver improved recycled materials a high fibre alignment is the key factor to increase the fibre volume fraction, and consequently the performances of recycled composites [15, 16]. A modified papermaking technique was developed by Pickering [17], Turner et al. [14] and Warrior et al. [18] reaching 80 % of the theoretical alignment value and a fibre volume of 45 % with a moulding pressure of 100 bar. Wong et al. [19] presented a centrifugal alignment rig, which uses a dispersion of fibres in a viscous media accelerated through a convergent nozzle installed in a rotating drum. An alignment level of 90 % was obtained using of 5 mm fibres. The same authors [19] worked on a hydrodynamic spinning process of a viscous fibre suspension. Janney et al. [20] developed the Three Dimensional Engineered Preform Process (3-DEP) adding multiple motions control to a pulp moulding process tool and therefore the control of fibre areal weight and orientation.

The HiPerDiF method, developed at the University of Bristol [21], has proven to be an effective way to manufacture composite materials with high levels of alignment from short fibres. This unique fibre orientation mechanism uses the momentum change of a water-fibre suspension to align the fibres. It was previously noted that tensile modulus, strength and failure strain of aligned discontinuous fibre composites produced with the HiPerDif method were close to those of continuous fibre composites provided that the fibres are accurately aligned and their length is sufficiently long compared to the critical fibre length [22, 23].

The use of the HiPerDiF method allows on one hand the production of high performance recycled carbon fibre composites thanks to a high level of fibre alignment and on the other hand is adequate to deal with the geometric characteristics of recycled carbon fibres. Therefore, this method allows the efficient reclaiming of value from end-of-life products and is well placed in the developing supply and processing chain of recycled carbon fibres.

Reviewing the work on the various fibre recovering processes it can be observed that their strength is sensibly reduced even if in general the stiffness of the fibre is not compromised. In this work, the overall stress-strain response of HiPerDiF aligned short fibre composites, as a function of the recycled to virgin carbon fibres ratio, is investigated.

2 MATERIALS AND MANUFACTURING

2.1 Materials

High tensile strength virgin carbon fibre (C124, TohoTENAX) and recycled carbon fibres (AS4, Hexcel) from a M56 resin composite with the pyrolysis “cycle B” process defined by Pimenta and Pinho in [24] have been used. The mechanical properties are summarised in Table 1.

Fibre properties		Virgin Fibres*	Recycled Fibres**
Diameter	[μm]	7	6.5
Length	[mm]	3	3
Density	[g/cm^3]	1.82	1.79
E_{11}	[GPa]	225	230
Failure σ_{11}	[MPa]	4350	1110
Failure ε_{11}	[%]	1.8	0.5

Table 1: Fibres properties:
*From manufacturer data sheet.
**Measured with single fibre tests a gauge length of 10 mm.

The fibres are impregnated using MTM49-3 epoxy resin.

2.2 Fibre alignment process

In the HiPerDiF process for producing highly aligned discontinuous fibre preforms, developed at the University of Bristol, short fibres suspended in water are accelerated through a nozzle and directed in a gap between two parallel plates. The fibre alignment mechanism relies on a sudden change of the liquid momentum. The water medium is removed by suction and the aligned fibre preform is dried with infrared radiation to allow the resin impregnation process. The process allows the alignment of fibres between 1 and 6 mm in length. A schematic of the HiPerDiF short fibre alignment machine is shown in Figure 1. The virgin and recycled carbon fibres were well mixed in the fibre dispersing process, which resulted in a high degree of hybridisation in composite samples [23].

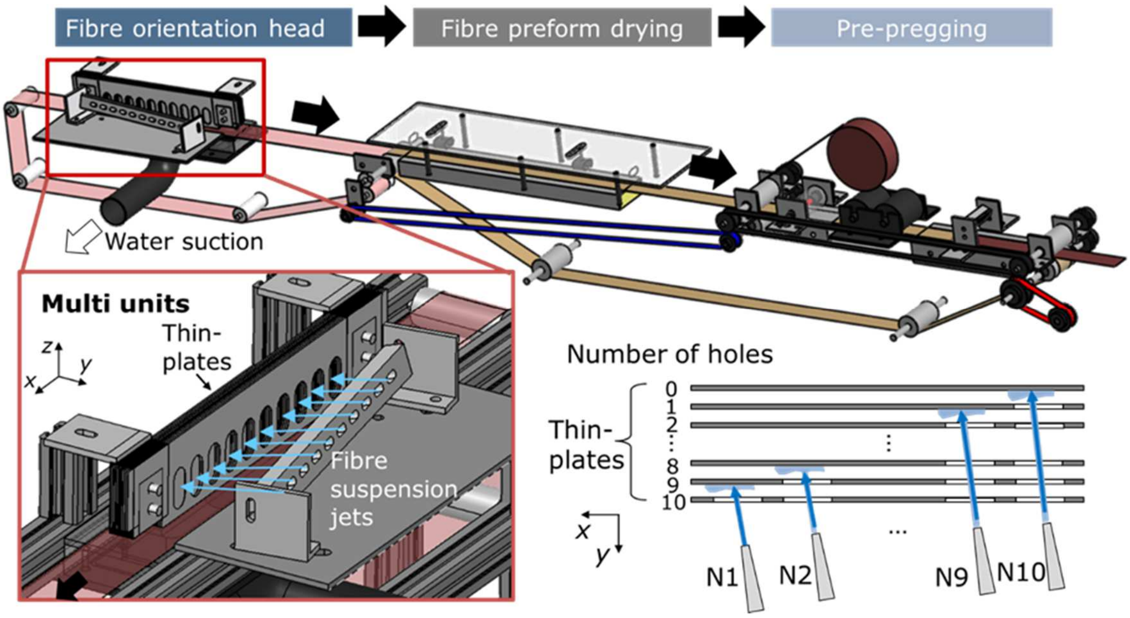

Figure 1: HiPerDiF fibre alignment machine.

It has to be noted that the machine, during the research work presented here, was undergoing a process of development: although this did not affect the fibre alignment level it caused changes in the fibre volume fraction between the different sets of specimens.

2.3 Specimen manufacturing

The specimens were prepared by vacuum bag moulding and cured in autoclave for 135 minutes at a temperature of 135°C and a pressure of 6 bar. Burrs at all edges were removed. GFRP end-tabs were bonded with Huntsmann Araldite 2014-1. A schematic of the specimen is shown in Figure 2. The nominal thickness of the specimens, manufactured with four plies, is 0.22 mm.

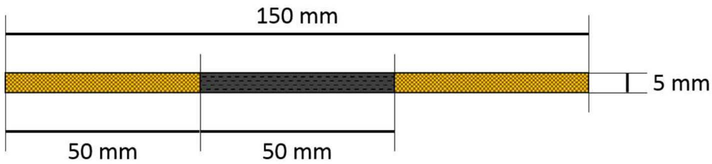

Figure 2: Specimen geometry.

3 EXPERIMENTAL WORK

3.1 Experimental methodology

Tensile tests were performed on an electro-mechanical testing machine with a cross-head displacement speed of 1 mm/min. The load was measured with a 10 kN load cell (Shimadzu, Japan) and the strain was measured with a video extensometer (Imetrum, UK). A white speckle pattern over a black background was spray painted on the specimens to allow the strain measurement with the video extensometer. The gauge length for the strain measurement was around 50 mm. Specimens with 100 % of virgin and recycled fibres as well as blends with 10, 20, 25, 30, 33, 40, 50, 60, 80 % recycled fibres were manufactured and tested.

3.2 Experimental results

The measured tensile stiffness as a function of the recycled fibre amount in the composite is summarised in Figure 3.

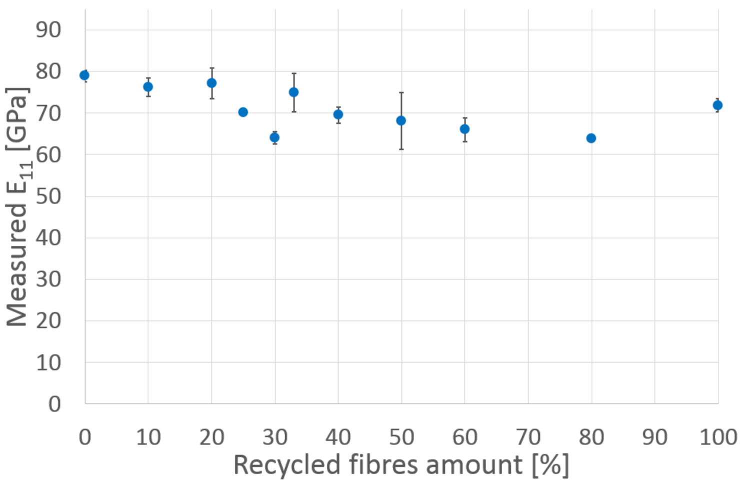

Figure 3: Measured tensile stiffness as a function of the recycled fibre amount in the composite.

By observing Figure 3, it is evident that the stiffness of the materials is independent from the amount of recycled fibres. The average stiffness is 70.4 GPa and the coefficient of variation is 8.3 %. The high coefficient of variation is caused by the different fibre volume fraction of the different specimen sets. The stiffness of the specimens remanufactured with 100 % of recycled fibres is 71.8 and the coefficient of variation 2.2 %; this is amongst the higher values of stiffness that can be found on the available literature about remanufactured recycled composites.

The measured tensile failure strain and strength as a function of the recycled fibre amount in the composite are summarised in Figure 4.

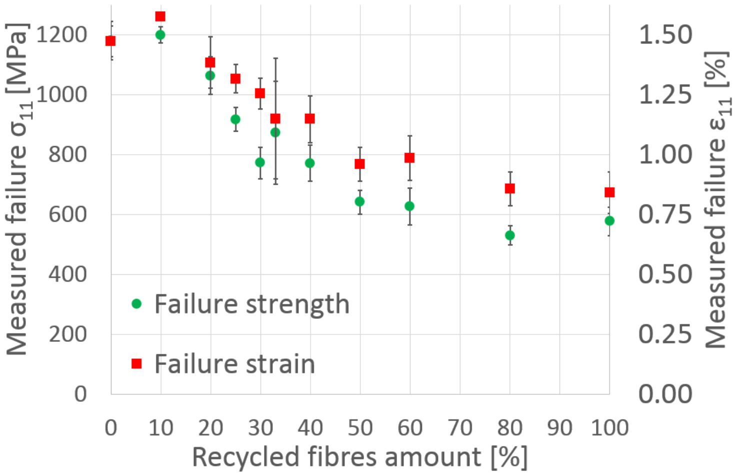

Figure 4: Measured tensile failure strain and stress as a function of the recycled fibre amount in the composite.

In Figure 4, the fact that the 100 % recycled composites present a failure strain (0.84 %) higher than the recycled fibres (0.5 % in Table 1) is noteworthy. This could be partially attributable to the fact that the recycled fibre length (3 mm) is lower than the one used to measure the fibres properties in [24], 10 mm. Therefore, the statistical probability of the defects, introduced by the recycling process, to affect the single fibre strength is reduced, leading to a delay in the overall failure of the composite. Moreover, even if the results are affected by the different fibre volume fraction, a trend can be identified. The failure strain and strength follow a linearly reducing trend between 0 and 50 % of recycled fibre, and after this are substantially constant.

6 RESULTS DISCUSSION

To be able to better interpret the data it was necessary to measure the fibre volume fraction. In a first attempt the resin burn-off method, codified in the ASTMD3171 standard, was used. This proved unreliable as the short fibres were blown away by the internal air circulation in the furnace. Considering the low weight of each specimen, even the loss of a small amount of fibre during the resin burn off compromises the fibre volume fraction measurement. It was therefore decided to calculate a nominal fibre volume fraction V_{fNom} with the formula described in Equation 1:

$$V_{fNom} = \frac{\eta(Q_{fR}E_{11R}+Q_{fN}E_{11N})}{E_{11Mes}} \qquad (1)$$

Where Q_f is the fibre fraction, E_{11} the stiffness, the subscripts R, N and Mes refer to recycled and virgin fibres and measured respectively. The correction factor η, in this case 0.853, takes into account the discontinuity and the misalignment of the fibres and is calculated as the ratio between the nominal and the calculated 100 % virgin composite stiffness.

The strength is normalised to a common value of fibre volume fraction, i.e. 60 %. It is possible to estimate a failure envelope for the strength using a linear or a bilinear rule of mixture (RoM) developed for hybrid composites [25].

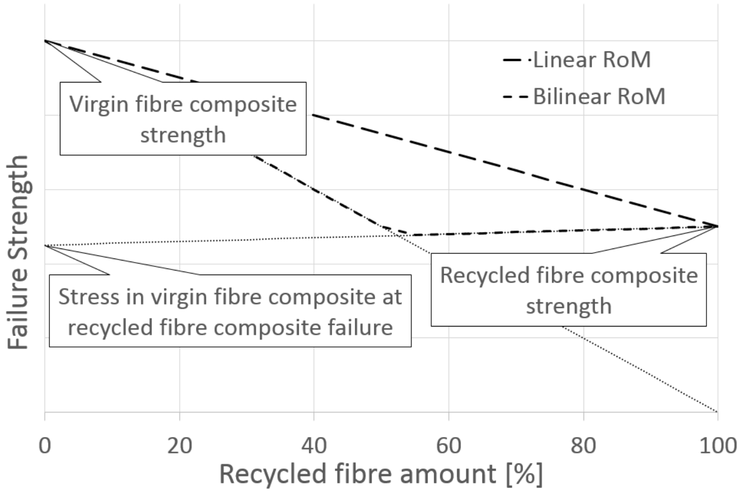

Figure 5: Definition of linear and bilinear Rule of Mixture (RoM), adapted from [26].

The recycled-virgin fibre composite can be considered as a hybrid composite constituted by two types of fibres with the same stiffness, the nominal difference is 2.2 %. However, high difference can be observed in the failure properties, in this case the recycled fibres act as the low elongation and the virgin fibres as the high elongation constituent. In terms of strength, it is commonly accepted that the linear RoM represents the upper bound. While the lower bound is defined by the failure stress of the recycled and virgin fibre composites, represented by the dotted lines in Figure 5; however, if the amount of virgin fibre is sufficient to withstand the load after the recycled fibre failure, the hybrid strength will be higher than the primary strength of the constituents.

The normalised strength along with the estimated envelopes is shown in Figure 6. The values of failure stress, for both the virgin and the recycled fibre composites, input in the envelopes presented below are obtained from the tests results presented above.

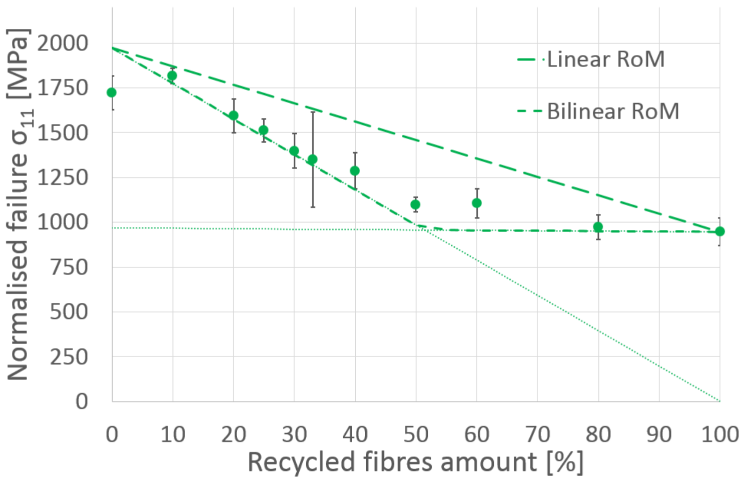

Figure 6: Normalised strength and envelope as a function of the recycled fibre amount in the composite.

Because the composite stiffness as a function of the recycled fibre amount is constant and the stress-strain curves show a linear elastic-brittle behaviour, the same construction described in Figure 5 for the strength can be applied to the failure strain, as shown in Figure 7.

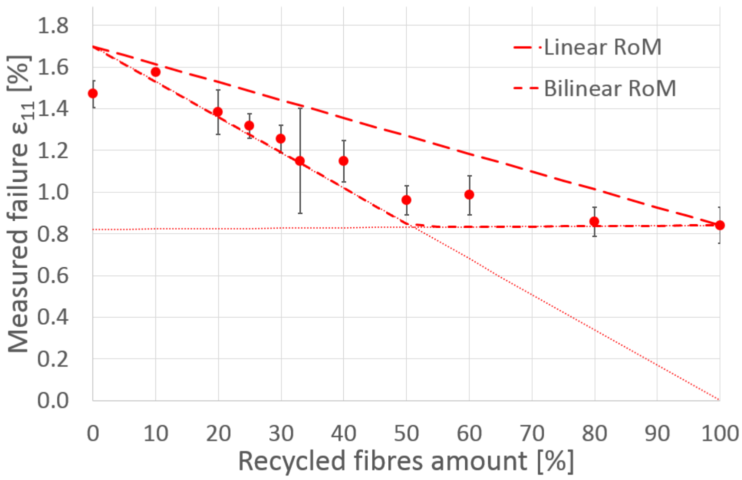

Figure 7: Measured failure strain and envelope as a function of the recycled fibre amount in the composite.

Observing Figures 6 and 7 it is clear how the strength and the failure strain follow the bilinear rule of mixture. In particular, they follow a diminishing trend, dominated by the virgin fibre properties up to 50 % of recycled fibre amount. In the region around the intersection of the two construction lines the experimental data exceeds the prediction of the bilinear rule of mixture. This has been observed also by Parratt and Potter in [27] and can be ascribed to statistical effects caused by the formation of clusters during the mixing of the two fibre types. After this threshold, the values are substantially constant and correspond to the strength of the recycled fibre composite. Increasing the failure values of the recycled fibre will therefore lead to an improvement in the virgin-recycled carbon fibre hybrid composite.

6 CONCLUSIONS

The HiPerDiF method proved to be a valuable instrument to remanufacture recycled carbon fibres and obtain a raw material with high structural and economical value. In particular, the material obtained with the recycled fibres, pure or blended with virgin ones, has a stiffness comparable with that of new material. Moreover, using short fibres between 1 and 6 mm, not only is compatible with the size constrains imposed by the recycling processes, but is also beneficial for the strength of the remanufactured products. However, the strength is reduced by the decay in the mechanical properties caused by the recycling process. On one hand, this can be accepted if the loss in strength is compensated by the economic benefits of using a cheaper raw material, on the other end, the constant development of the recycling techniques is delivering recycled fibres with a minimal loss of strength properties, making recycled fibres composites a viable solution for high performances applications.

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REFERENCES

[1] S. Pimenta, S.T. Pinho, “Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook” Waste Management, Vol. 31, pp. 378-392, 2011.

[2] L.O. Meyer, K. Schulte, E. Grove-Nielsen, “CFRP-Recycling Following a Pyrolysis Route: Process Optimisation and Potentials” Journal of Composite Materials, Vol. 43(9), pp. 1121-1132, 2009.

[3] S.J. Pickering, R.M. Kelly, J.R. Kennerley, C.D. Rudd, N.J. Fenwick, “A fluidised bed process for the recovery of glass fibres from scrap thermoset composites” Composites Science and Technology, Vol 60, pp. 509-523, 2000.

[4] R.E. Allred, J.M. Gosau, J.M. Shoemaker, “Recycling process for carbon/epoxy composites” SAMPE 2001 Symposium & Exhibition, SAMPE, Longbeach, CA, USA, 2001.

[5] M. Nakagawa, K. Shibata, H. Kuriya, “Characterization of CFRP using recovered carbon fibers from waste CFRP” Second International Symposium on Fiber Recycling, The Fiber Recycling 2009 Organizing Committee, Atlanta, Georgia, USA, 2009.

[6] R. Pinero-Hernanz, C. Dodds, J. Hyde, J. Garcia-Serna, M. Poliakoff, E. Lester, M.J. Cocero, S. Kingman, S.J. Pickering, K.H. Wong, “Chemical recycling of carbon fibre reinforced composites in nearcritical and supercritical water” Composites Part A: Applied Science and Manufacturing, Vol. 39, pp. 454-461, 2008.

[7] R. Pinero-Hernanz, J. Garcia-Serna, C. Dodds, J. Hyde, M. Poliakoff, M.J. Cocero, S. Kingman, S.J. Pickering, E. Lester, “Chemical recycling of carbon fibre composites using alcohols under subcritical and supercritical conditions” Journal of Supercritical Fluids, Vol. 46, pp. 83-92, 2008.

[8] G. Jiang, S.J. Pickering, E.H. Lester, T.A. Turner, K.H. Wong, N.A. Warrior “Characterisation of carbon fibres recycled from carbon fibre/epoxy resin composites using supercritical n-propanol” Composite Science and Technology, Vol. 69, pp. 192-198, 2009.

[9] M. Goto, “Chemical recycling of plastics using sub- and supercritical fluids” Journal of Supercritical Fluids, Vol. 47, pp. 500-507, 2009.

[10] J. Meredith, “The role of recycled carbon fibre composites in motorsport applications” Carbon Fibre Recycling and Reuse 2009 Conference, IntertechPira, Hamburg, Germany, 2009.

[11] K.H. Wong, S.J. Pickering, T.A. Turner, N.A. Warrior, “Compression moulding of a recycled carbon fibre reinforced epoxy composite” SAMPE’09 Conference, SAMPE, Baltimore, MD, USA, 2009.

[12] S.J. Pickering, “Recycling technologies for thermoset composite materials - current status” Composites Part A: Applied Science and Manufacturing, Vol. 37, pp. 1206-1215, 2006.

[13] M. Szpieg, M. Wysocki, L.E. Asp, “Reuse of polymer materials and carbon fibres in novel engineering composite materials” Plastics, Rubber and Composites, Vol. 38, pp. 419-425, 2009

[14] T.A. Turner, S.J. Pickering, N.A. Warrior, “Development of high value composite materials using recycled carbon fibre” SAMPE’09 Conference, SAMPE, Baltimore, MD, USA, 2009.

[15] L.T. Harper, T.A. Turner, J.R.B. Martin, N.A. Warrior, “Fibre alignment in directed carbon fibre preforms-A feasibility test” Journal of Composites Materials, Vol. 43(1), pp. 57-74, 2009.

[16] K.H. Wong, T.A. Turner, S.J. Pickering, N.A. Warrior, “The potential for fibre alignment on the manufacture of polymer composites from recycled carbon fibre”, SAE International journal of Aero space, Vol. 2(1), pp. 225-231, 2009.

[17] S.J. Pickering, “Carbon fibre recycling technologies: what goes in and what comes out?” Carbon Fibre Recycling and Reuse 2009 Conference, IntertechPira, Hamburg, Germany, 2009.

[18] N.A. Warrior, T.A. Turner, S.J. Pickering, “AFRECAR and HIRECAR Project results” Carbon Fibre Recycling and Reuse 2009 Conference, IntertechPira, Hamburg, Germany, 2009.

[19] K.H. Wong, T.A. Turner, S.J. Pickering, N.A. Warrior, “The potential for fibre alignment in the manufacture of polymer composites from recycled carbon fibre” SAE AeroTech Congress and Exhibition, SAE International, Seattle, Washington, USA, 2009.

[20] M.E.G. Janney Jr, N. Baitcher, “Fabrication of chopped fiber preforms by the 3-DEP process” Composites and Polycon 2007, American Composites Manufactureres Association, Tampa, FL, USA, 2007.

[21] H.Yu, K.D. Potter, “Method and apparatus for aligning discontinuous fibres”, UK patent, Patent application number 1306762.4, 2013.

[22] H. Yu, K.D. Potter, M.R. Wisnom, "A novel manufacturing method for aligned discontinuous fibre composites (High Performance-Discontinuous Fibre Method)" Composites Part A: Applied Science and Manufacturing, Vol. 65, pp. 175-185, 2014.

[23] H. Yu, M.L. Longana, M. Jalalvand, M.R. Wisnom, K.D. Potter, "Pseudo-ductility in intermingled carbon/glass hybrid composites with highly aligned discontinuous fibres" Composites Part A: Applied Science and Manufacturing, Vol. 73, pp. 35-44, 2015.

[24] S. Pimenta, S.T. Pinho, "The effect of recycling on the mechanical response of carbon fibres and their composites" Composite Structures, Vol. 94(12), pp. 3669-3684, 2012.

[25] Y. Swolfs, L. Gorbatikh, I. Verpoest, "Fibre hybridisation in polymer composites: a review" Composites Part A: Applied Science and Manufacturing, Vol 67, pp. 181-200, 2014.

[26] G. Kretsis, "A review of the tensile, compressive, flexural and shear properties of hybrid fibre-reinforced plastics" Composites, Vol 18(1), pp. 13-23, 1987.

[27] N.J. Parratt, K.D. Potter, "Mechanical behaviour of intimately-mixed hybrid composites" Proceedings of ICCM3-3rd International conference on composite materials. Paris, France, pp. 313–23, 1980.