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Applications for carbon fibre recovered from composites

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Abstract. Commercial operations to recover carbon fibre from waste composites are now developing and as more recovered fibre becomes available new applications for recovered fibre are required. Opportunities to use recovered carbon fibre as a structural reinforcement are considered involving the use of wet lay processes to produce nonwoven mats. Mats with random in-plane fibre orientation can readily be produced using existing commercial processes. However, the fibre volume fraction, and hence the mechanical properties that can be achieved, result in composites with limited mechanical properties. Fibre volume fractions of 40% can be achieved with high moulding pressures of over 100 bar, however, moulding at these pressures results in substantial fibre breakage which reduces the mean fibre length and the properties of the composite manufactured. Nonwoven mats made from aligned, short carbon fibres can achieve higher fibre volume fractions with lower fibre breakage even at high moulding pressure. A process for aligning short fibres is described and a composite of over 60% fibre volume fraction has been manufactured at a pressures up to 100 bar with low fibre breakage. Further developments of the alignment process have been undertaken and a composite of 46% fibre volume fraction has been produced moulded at a pressure of 7 bar in an autoclave, exhibiting good mechanical properties that compete with higher grade materials. This demonstrates the potential for high value applications for recovered carbon fibre by fibre alignment.

1. Introduction

Carbon fibre is finding increasing numbers of applications in lightweight structures due to its high specific mechanical properties. High growth rates are already being observed in several major civil aerospace programmes, such as Boeing 787 Dreamliner and Airbus A350. Optimistic levels of growth are also predicted for the automotive, industrial and power generation sectors. Current levels of carbon fibre usage are in excess of 100,000 tonnes per annum with growth forecast to be between 10% and 20% per annum. Automotive remains an interesting potential future user of considerable amounts of fibre with global production now in excess of 90,000,000 vehicles per annum. Even a small amount of fibre in a small fraction of total production would make a significant impact on demand.

Several processes are now operating commercially for the recovery of carbon fibres from end of life components, such as aircraft parts and tooling, as well as in-process scrap such as out of shelf-life prepreg and ply cuter offcuts. The high energy use associated with the production of virgin carbon fibre leads to a high market value and gives the potential for the production of recovered fibre at a significantly lower cost than that associated with virgin fibre. Process scale is of course a dominant factor with virgin fibre production lines showing significantly increased efficiency through improved process monitoring and hardware, whereas recovered fibre production is typically smaller scale thus struggling to make use of the efficiencies of larger scale operation.

Currently it is difficult to understand the impact of legislation on the recovered carbon fibre market; increasing pressure to manufacture fuel efficient vehicles drives car manufacturers towards lightweighting methodologies but often these are somewhat at odds with consumer desires which ultimately drive production. The End of Life Vehicle Directive serves to put pressure on vehicle makers but often has the effect of prohibiting the use of materials which will save energy over the use phase of the product lifecycle. No corresponding regulation exists in aerospace where the use phase dominates and lightweighting through the use of carbon fibre is overwhelmingly compelling. Landfill tax remains as a motivating factor for fibre recovery with increasing difficultly experienced with the disposal of both uncured and cured waste and a growing impetus for cost saving at a business level.

Although fibres may be recovered at a cost which is lower than the corresponding grade of virgin fibre, the form of the recovered fibres is inevitably different from virgin and this factor has to-date limited the penetration of recovered fibres into virgin fibre markets. The challenge therefore is to change the material into a form which competes with virgin materials (whether on the basis of cost or performance) without incurring so much cost that the price is not competitive with virgin fibres.

1.1. The nature of recycled carbon fibre

In pyrolysis or thermal fluid recycling processes, the physical form of the fibres recovered is basically that of the material fed in with the polymer removed. If scrap prepreg materials are processed it may be possible to recover the carbon fibre in the form of a textile, in which the fibres are continuous, ready for re-impregnation. However, in most cases the scrap being processed would be in a variety of physical forms, as shown in figure 1, and so a short fibre product could be produced by either chopping the waste before processing or by chopping the carbon fibre recyclate after recycling. The fibre products are in a fluffy form as there is no longer any size holding the fibres bundles together. The fluidized bed process also produces a recyclate with a fluffy form, as shown in figure 2, in which the carbon fibre is in the form of individual filaments with a distribution of fibre length. All the processes produce a clean fibre recyclate with no substantial residue on the surface, as shown in figure 3**.**

Figure 1. End of life laminate scrap after primary size reduction (left) and secondary size reduction (right).

Figure 2. Recycled fibre in raw form showing fluffy, discontinuous, 3D random and highly entangled structure with areas of higher fibre packing due to retained bundle integrity.

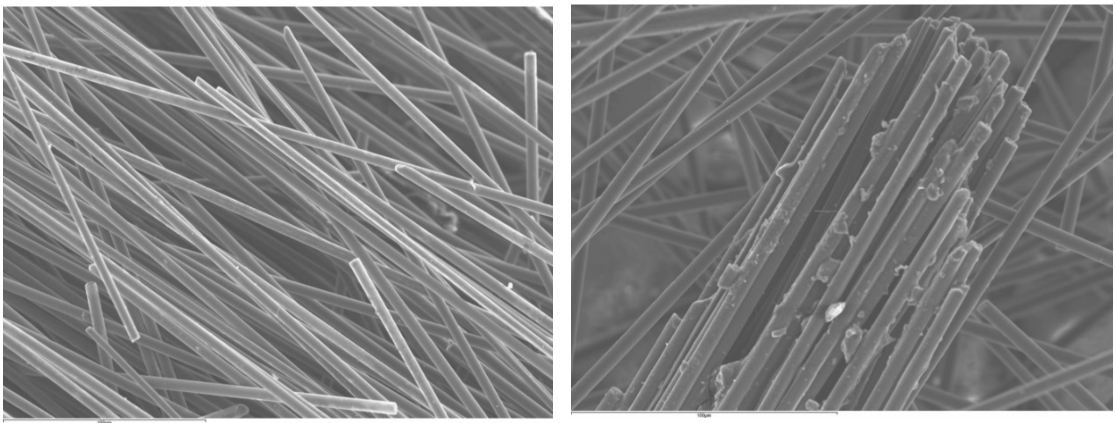

Figure 3. Recycled fibres, showing a clean surface free from polymer residue (left) and with some char remaining (right) causing bundle integrity to be maintained.

The mechanical properties of recycled carbon fibre are generally good and compare well with virgin fibre. Measurements of stiffness show that the recycling processes generally gives recycled fibres with a stiffness similar to that of virgin fibre. However, there is some degradation in tensile strength. The thermal fluid processes only show a reduction of a few percent. The pyrolysis processes are reported to show slightly more strength reduction with up to 10% loss in strength. However, the fluidized bed process shows a loss of strength of up to 50%. This may be due to the increased mechanical agitation in the process and also the effect of the oxidizing atmosphere. Though process is particularly suitable for contaminated end-of-life waste which may not be appropriate for other recycling processes.

The electrical conductivity of the recycled fibre has been found to be similar to that of virgin fibre and analysis of the surface chemistry shows that after recycling there are still active oxygenated species on the surface and the recycled fibres have been found to bond well to epoxy resin.

1.2. Applications for recycled carbon fibre

There are a many potential applications for recycled carbon fibre and it can be used as a structural reinforcement in composites with a wide range of mechanical properties and hence value. Broadly there are several approaches for reuse:

1. Direct competition with virgin materials (composite or metallic)
   - Reduced mechanical performance may be offset by reduced cost
   - At what scale should the recycled materials be compared with other materials: tow, mat, or compound?
2. Development of or penetration into new markets
   - New material forms may be appropriate for use of recycled fibre (e.g. commingled with other fibres)
   - Replacement for glass as well as virgin carbon fibre.
3. Increased functionality
   - Discontinuous fibres may be used in applications to give enhanced drape properties for easier manual or automated forming
   - Exploiting the conductivity of the recycled fibre in applications where reduced mechanical strength is of secondary importance.
   - Manufacturing low fibre areal mass materials, in which discontinuous recycled fibre may be preferred.

Currently the outlook is unclear and many routes for market development are under consideration. Milled fibre is an obvious end use application but it has low mechanical properties as a reinforcement and the market is not significant when compared to the volumes of fibre that may become available. Pellets for injection moulding processes are one potential avenue and highly aligned mat is another area showing promise.

1.3. The case for high value applications for recycled carbon fibre

The key driving force for future recycled carbon fibre-based materials will be a business case whereby the fibre recovery provider can sell the fibres to an end user or intermediate product manufacturer. A high value end use application enables a more viable supply chain for recovered fibres rather than a low value, high volume re-use route. Penetration into the automotive market is seen as a significant enabler for carbon fibre recycling in general and the costs proposed by automotive vehicle manufacturers provide a good target for recovered carbon fibre-based materials. The challenge is to manufacture materials from the recovered fibre without significantly increasing the cost. Aluminium is a good mechanical property target as the automotive industry already performs high levels of research into weight saving through the use of this material. These two cost and performance requirements therefore define the area of applicability for recovered fibre products.

2. Achieving high fibre volume fraction with fluffy discontinuous fibre

Composite structural performance depends largely on the mechanical properties and volume content of the reinforcement and how these structural properties can be attained will define the market that the composite will be targeted toward. At an initial level, the mechanical property of virgin and recycled carbon fibres don't appear very different: both of them have insignificant difference in tensile stiffness and interfacial shear strength; in the case of tensile strength, it depends on the chosen recycling process and the commercially available pyrolysis processes can offer fibre with high degree of strength retention. However, a marked difference is in their physical form: virgin fibre is continuous and can readily be weaved into the trademark woven pattern that marks it out as belonging to high grade applications, for instance found in superlight bicycles, Formula One and aerospace structure; whereas recycled fibre is short and discontinuous, without sizing and in a fluffy form, preventing it from being a direct substitute for virgin fibre and requiring further conversion processes to improve handling and processability. What they have in common is that both are valuable materials that can be processed specifically for specific applications. For recycled fibre, having economically viable conversion processes is pivotal for them to be established away from markets associated with a perception of lower quality, such as being used as a filler in injection moulding, and to reach out to wider markets offering higher grade applications.

Using milled recycled fibre in injection moulding is the earliest conversion process to enable the recycled fibre to be used in high volume basis. This application will have a long term presence but wider routes are required as the market will become saturated. Attention is focusing on seeking out established technology in which recycled fibre can be used directly, and the papermaking industry may be able to offer not only a process for direct use of recycled fibre but also an economically viable process for turning the fluffy fibre into a nonwoven form. Work is still ongoing to optimise the process and to establish markets for the nonwovens. Meanwhile, the recycling industry is continuing to develop new processes for higher value usage and fibre alignment is considered one an important conversion options as it not only offers higher fibre volume fractions to be achieved with lower moulding pressure, but it will also reduce high pressure fibre-to-fibre contact points and preserve fibre length for much needed structural application development. The following two sub-sections report the use of nonwoven mats in reinforcing thermoset composites, and consider the pros and cons as well identifying the need to lead to the development of an aligned fibre structure.

2.1. Use of random wet lay non-wovens for composites manufacture

In a paper presented at the SAMPE conference in 2009 [1], the feasibility of using recycled fibre in a nonwoven form for a flame-retardant epoxy composite was reported. The fibre was of grade T300 from Toray and was recycled via a pyrolysis process by ELG Carbon Fibre Ltd, Coseley, UK (formerly Recycled Carbon Fibre Ltd) . The average input fibre length before moulding was 12 mm and it was converted into a 100 g/m^2 nonwoven mat 520 mm wide using a papermaking process at Technical Fibre Products Ltd, Kendal, UK. The nonwoven mats were collated with 200 g/m^2 resin film, MTM56-2FRB, supplied by Cytec (formerly Advanced Composites Group), Heanor, UK and compression moulded at 120 °C under a pressure varying from 2 MPa to 14 MPa to make a composite of fibre volume content ranging from 20% to 40%. The moulding pressure required to achieve a required fibre volume fraction was determined by performing a compression test on a stack of dry non-woven mats between two platens attached to a universal testing machine with distance between the platen and load were being constantly recorded. To evaluate the mechanical performance of the composite, tensile and flexural tests were performed on at least 5 specimens for each fibre loading. The change in fibre length due to the moulding pressure was measured by matrix burn-off, fibre separation, digital image capture via a microscope and finally length measurement via the Image Pro Plus image analysis software, provided by MediaCybernetics, Cambridge, UK.

Figure 4 and figure 5 show that the nonwoven mat has been successful in enhancing the tensile and flexural moduli of the composite and a linear correlation between the moduli and fibre content can be observed. However, for the case of composite strength, such a linear correlation only applies for fibre content up to 30% as beyond which the composite performance starts to drop. Several factors can affect the strength of a fibre reinforced composite, including the inherent fibre strength, shape, orientation and interface with the matrix. All these factors are considered common between all samples tested and the drop in strength is attributed to the degradation in fibre length distribution and increase in composite void content and these are shown in figure 6and figure 7 respectively. For full utilisation of the fibre reinforcement potential, the fibre must be longer than L_c, the critical fibre length, which is given by $L_c = r_f\sigma_f/\tau_c$, where r_f, σ_f and τ_c are the fibre radius (3.6 µm), fibre ultimate tensile strength (4.16 GPa) and composite shear strength respectively. Shear strength varies with the chosen type of carbon fibre and for a typical epoxy composite reinforced with recycled carbon fibre, the value varies between 60.0 MPa and 80.0 MPa [2]. Thus, the estimated critical length is between 0.19 mm and 0.25 mm. It can be seen in figure 6 that at 20% fibre content, about 70% of the fibre population is longer than 0.25 mm. However, at 40% fibre content, there is only about 10% and thus the fibre reinforcing potential is greatly reduced. As mentioned earlier, another factor that could contribute to the decrease in strength is the presence of high a void content, as depicted in figure 7.

Nonwoven mat has a high degree of loft as it comprises filamentised fibres with random planar orientation and a wide range of length distribution. This suggests a very irregular flow channels between the fibres and the permeability very much depends on the availability of interstitial free space. It is hypothesized that under high moulding pressure, the creation of short fibres promotes nesting and reduces the interstitial free space, disrupting or locally blocking up the flow channels, resulting in higher void in the resultant composite.

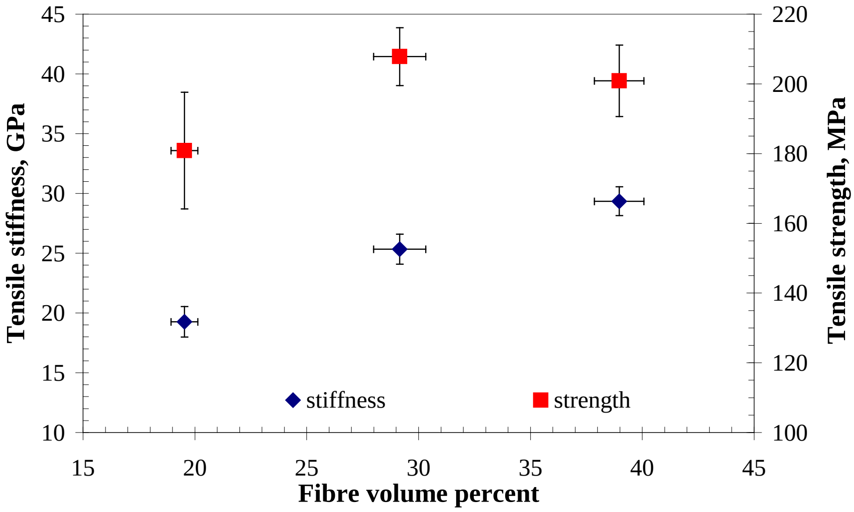

Figure 4. Tensile properties of composite reinforced with recycled carbon fibre nonwoven mats at various fibre volume content [1].

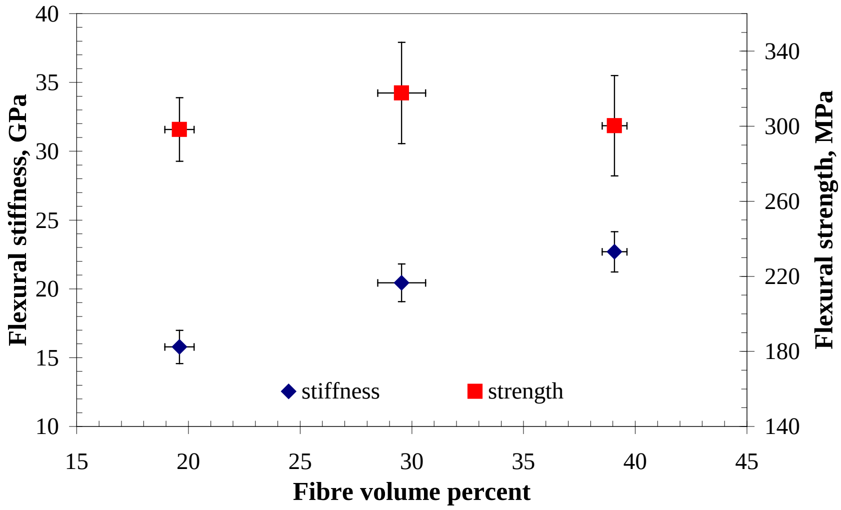

Figure 5. Flexural properties of composite reinforced with recycled carbon fibre nonwoven mats at various fibre volume content [1].

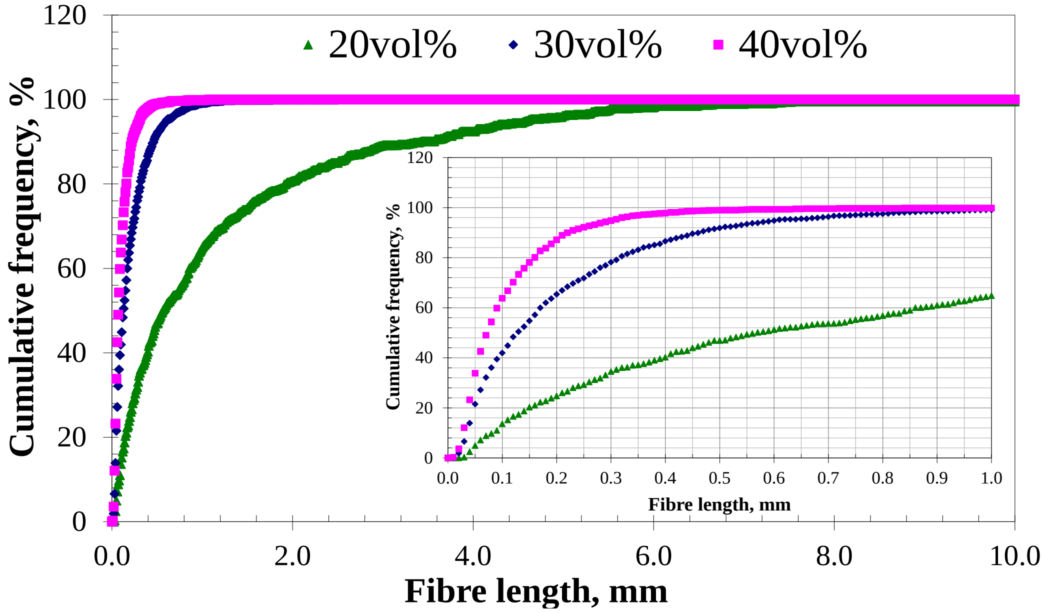

Figure 6. Fibre length distribution from composites at various fibre volume contents [1].

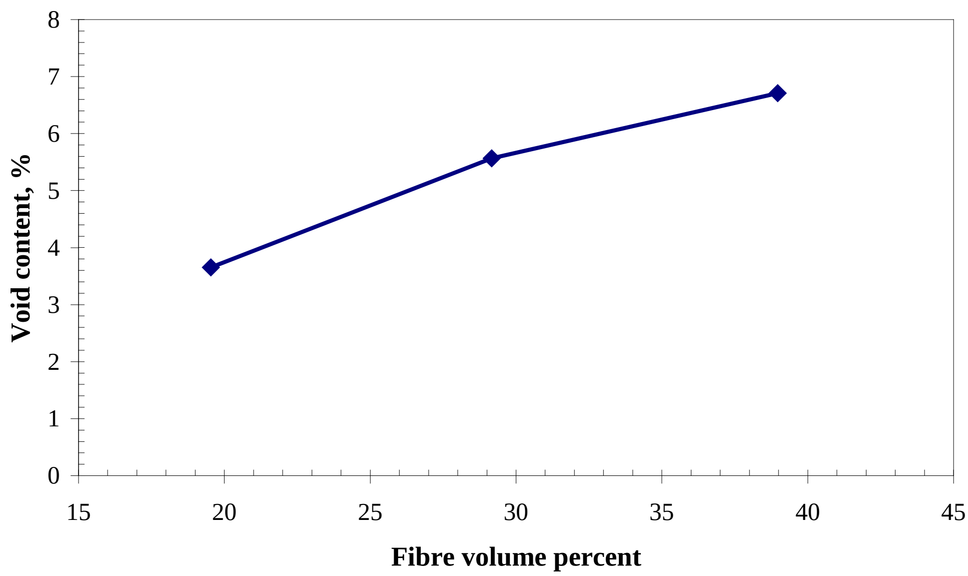

Figure 7. Composite void content [1].

2.2. The need for fibre alignment

Research and development in nonwoven mat manufacture continues to be of importance to the composite industry as it is built on an established know-how and is capable of lending itself to existing moulding technology, such as resin film infusion and compression moulding for semi-structural applications. Thermoplastic material, whether in the form of powder, filament or film, can potentially be included in the nonwoven mat, producing a commingled structure ready for thermoforming. However, with a random, filamentised fibre architecture, the maximum practical volume content is about 30% in order to minimize excessive length degradation. Higher volume fraction can only be realised through fibre alignment, a conversion process to arrange individual filament to a preferential direction to form a close-packed structure.

3. Technology for fibre alignment

Much work has been done in the past to control orientation of discontinuous fibre. Conventional textile processes such as carding, combing and gilling have been successful in improving orientation for cotton yarn, which is generally long, tough and flexible. Unfortunately, for the recycled carbon fibre, which is unsized, entangled and brittle, these intense mechanical actions are likely to cause severe damage to the fibre itself. Injection moulding is a process capable of introducing anisotropy to composite mechanical property by affecting the fibre orientation. A 3-layer structure is generally produced across the thickness of moulded sample with two skin layers having fibre preferentially aligned to the flow direction, but fibre at the core section has major orientation distribution normal to the flow. Besides the high shear stress inside the melting and dispersion zones of the moulding machine, the interaction between fibres and machine parts have a significant effect on the resultant fibre length of the moulded part. Practically, 35 vol% is the maximum fibre content this process can offer without causing too much damage to the fibre length. Ideally, interaction between fibres should be kept to minimum, or in other words, fibres are allowed to rotate and translate freely toward the preferential direction and this implies that alignment is best taken place with low fibre volume content and then the aligned fibres are densified to obtain a close-packed structure. These requirements can be met by wet hydrodynamic processes, which subject a dilute or semi-dilute fibre suspension toward a convergent device for alignment and after which the liquid medium is removed to obtain a layer of well aligned fibres. Further details of the hydrodynamic processes and their process parameters that govern the alignment efficiency are reported in the subsequent section.

3.1. Techniques for alignment of discontinuous fibre

Several approaches have been developed using different means to align discontinuous fibre. The diamagnetic anisotropic property of carbon fibre has been exploited under magnetic field for alignment development but this is limited to fibre length less than 500 micron [3, 4]. Electric fields have also been used for orientation control but generally for carbon nanotube [5] or polymer fibres [6]. Another approach that has drawn attention is via hydrodynamic force created by fluid flow. Of these approaches, the hydrodynamic process is considered to the most practical as the working principal is based on well-established and mature hydrodynamic technology and it also offers versatility for pilot scale research and can readily be scaled up for commercial application. Between the late sixties and the seventies, the Explosives Research and Development Establishment (ERDE), UK devised and developed three patented hydrodynamic alignment techniques based on extrusion [7], filtration [8] and centrifugal [9] process.

The extrusion process was designed for aligning asbestos and silicon carbide whiskers in an ammonium alginate solution [10]. The fibre suspension was extruded through an orifice of 2mm diameter into an acid precipitating bath. The extrudate gelled immediately in the bath and hence the alignment was maintained. After reeling up on a winding drum, it was then washed and dried. Then the residual alginic acid was burned off. In the pilot plant, the nozzle traversed across a winding drum to build up several complete layers of extrudate sheets. The whole sheet was then cut across to give a large flat sheet. The level of alignment was not presented but high volume packing up to 50% was reported. However, this process was deemed to be impractical and uneconomical as the ammonium alginate itself was not recyclable and it required a lengthy combustion stage for complete removal of the alginic acid [11].

The filtration approach is the most widely published ERDE fibre alignment process. It started from a batch scale, which was capable of making aligned fibre sheets up to 40 cm x 30 cm. It was later developed into a continuous process for a 152 mm wide sheet roll [11, 12]. Discontinuous fibre was dispersed in glycerine to form a suspension. A good dispersion quality was obtained if there were no undispersed bundles in the bulk suspension and this could be achieved with the selection of short fibre length, low fibre loading and the presence of a high shear rate within the viscous medium. Air entrapment should be kept to minimum as fibres deviating around air bubbles would affect the alignment quality. The suspension was then pumped gently toward a convergent device with a narrow slit exit. Fibres were aligned inside the convergent device through a velocity gradient across the suspension streamlines. The device was located above a fine conveying mesh and travelled repeatedly depositing suspension over a region on the mesh. A vacuum was applied under the mesh to promote draining. The damp mat was then washed with a gentle water spray and binder applied. The degree of alignment attained from this process was good and more than 90% of the fibre population was within ±15° of the desired direction [13]. However, when the mat thickness increases, it's through-thickness permeability would decrease, resulting in a longer dwell period to allow completion of glycerine drainage before the next deposition was applied and leading to an economically unacceptable production rate [14]. In addition, due to the reciprocating motion of the convergent slit, a lower degree of alignment was seen at the end regions of the reciprocating path.

The centrifugal process was built on the strength of the filtration approach but equipped with two significant modifications i.e. the narrow slit exit was replaced with a series of convergent nozzle as the latter offered an all-round converging contour along the nozzle axis and the conveying mesh was replaced with a rotating drum lined with a fine mesh internally as the drainage could be accelerated with the aid of centrifugal force. It had been claimed that a better alignment was achieved with higher throughput rate [15].

3.2. Initial development of high fibre volume fraction composites from recycled carbon fibre

A bench-top centrifugal rig was built according to the principal outlined by the ERDE process [9]. The capability of the rig was investigated via a $2^{(6-1)}$ fractional factorial experiment design to provide a better insight into the effect of operating conditions on alignment quality. The six factors under consideration were fibre length, fibre loading in the slurry, glycerine viscosity, nozzle diameter ratio, aligned mat areal density and relative speed between the rotating drum and slurry deposition rate. Orientation of the fibre within the aligned mat was measured through image analysis of the fibre elliptical cross-sectional geometry and this orientation data was used as an output response to assess the effect of changes from the six input factors. Virgin Toho Tenax HTA 5131 carbon fibre with its sizing removed by heating at 550 °C for 10 minutes inside a furnace was used. Once the best operating condition was identified from the test, a 3mm long recycled Toray T800 carbon fibre, supplied by ELG Carbon Fibre Ltd, was then used for making the mats, which was later compression moulded with epoxy resin films into a composite for mechanical testing. A summary of a more detailed analysis [16] of the test result is presented here. A schematic representation of the alignment rig is shown in figure 8. After the deposition process, the rotational speed of the perforated drum was set to 1500 rpm and this speed was maintained for 1 minute in order to further remove glycerine from within the mat. The mat was then placed on a vacuum tray for removal of the residual glycerine through water spraying and later was followed with application of a general grade PVA binder, sourced from Hobbycraft, Dorset, UK. The mat was then dried at room temperature overnight.

From the results of the test, it was found that there were four main effects: alignment quality improved with a higher relative speed ratio and nozzle diameter ratio but there was a negative correlation with mat areal density and fibre length. The only interaction effect was between the fibre length and veil area density. Figure 9 shows the fibre orientation distribution of the best and the worst aligned mats from the tests. The former had 94% of the fibres laid within ±10° from the preferred orientation, whereas the latter case only 78%.

The results of the test allowed the formulation of the best operating conditions for aligning recycled carbon fibre. It is found that a fibre slurry with 0.1 vol% of recycled fibre was required in order to prevent entanglement of the fibre filaments. The glycerine was diluted with 5 wt% of tap water to lower its viscosity in order to accelerate filtration rate during the fibre deposition stage. A higher relative speed was selected for getting a better alignment by further stretching the extruded fibre slurry before it reached the rotating mesh. The designated areal density for the recycled fibre mats was 35g/m^2. The mats produced were collated with resin film and pressed to 100 bar 3 times to increase the packing by a nesting effect. Then a moulding pressure of 80 bar was applied at 120 °C to fabricate a composite of thickness of around 2 mm and with a targeted fibre volume content of 60%. The mechanical properties of the composite were evaluated by performing a 3-point bending test along and normal to the alignment direction and the result is shown in table 1. It can be seen that the flexural strength measured along the alignment direction is a magnitude higher than the value perpendicular to the alignment and similarly for the flexural stiffness. This clearly indicates that a good fibre alignment has been achieved and this is further supported by referring to the micrograph shown in figure 10, which presents a close-packed fibre architecture. Image analysis of these micrographs revealed that the local fibre volume content was between 60% and 63%, which was in agreement with the targeted value of 60%. The void content of the composite was found to be around 1%. The fibre length distribution of the composite was determined by matrix burn off to allow individual fibre lengths to be measured digitally. The length distribution based on number fraction is shown in figure 11and a peak can be seen near the 3.1 mm region. Weight average and number average of the fibre lengths are also included in the figure 11 and when compared to the corresponding input fibre length, no significant degradation in fibre length is observed, despite the mats had been subjected to 100bar pressure in the early stage of the moulding process. The outcome of this test clearly highlighted the advantages of having a well aligned fibre structure compared to the nonwoven mat reported early.

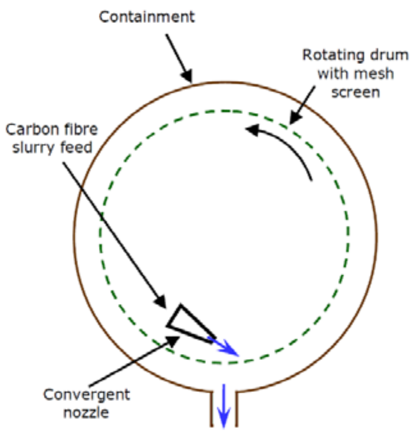

Figure 8. A schematic representation of the fibre alignment rig [16].

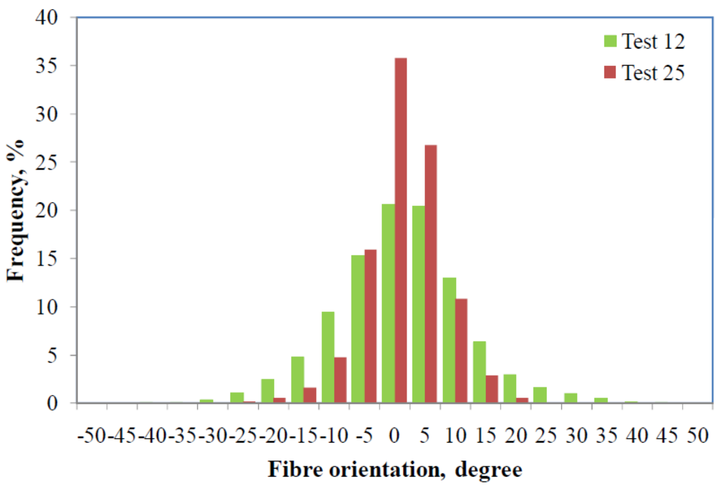

Figure 9. Fibre orientation distribution obtained from the best and worst samples from the factorial test [16].

Table 1. Average flexural properties of composite containing recycled and aligned carbon fibre [17].

Specimen	Flexural strength, MPa	Flexural stiffness, GPa
Along the alignment direction	1240 ± 69	81.8 ± 5.0
Normal to the alignment direction	107 ± 2	7.2 ± 0.2

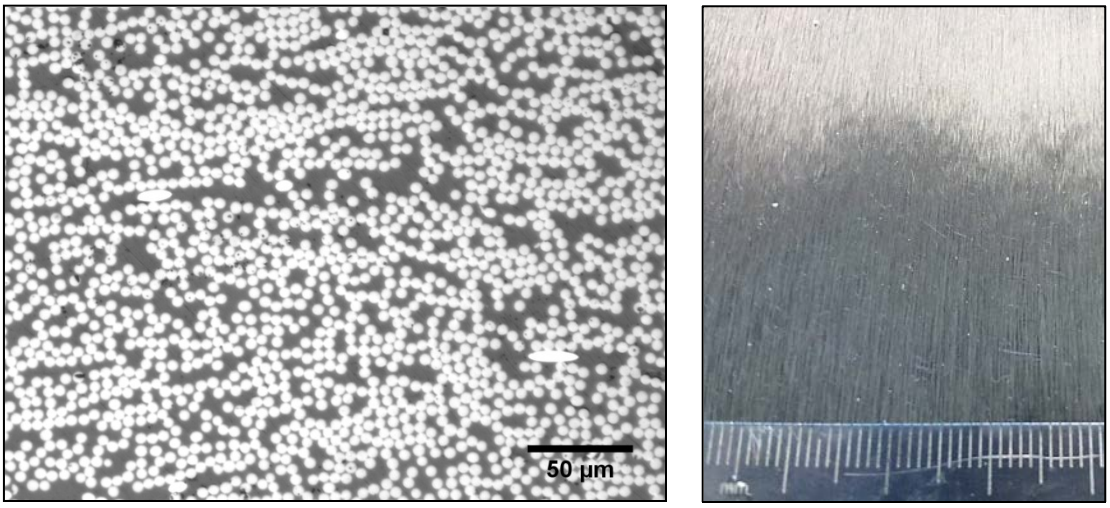

Figure 10. Micrograph of a composite containing 60% (vol) recycled, aligned fibre mat (left) and a top view of the mat before moulding (right) [16].

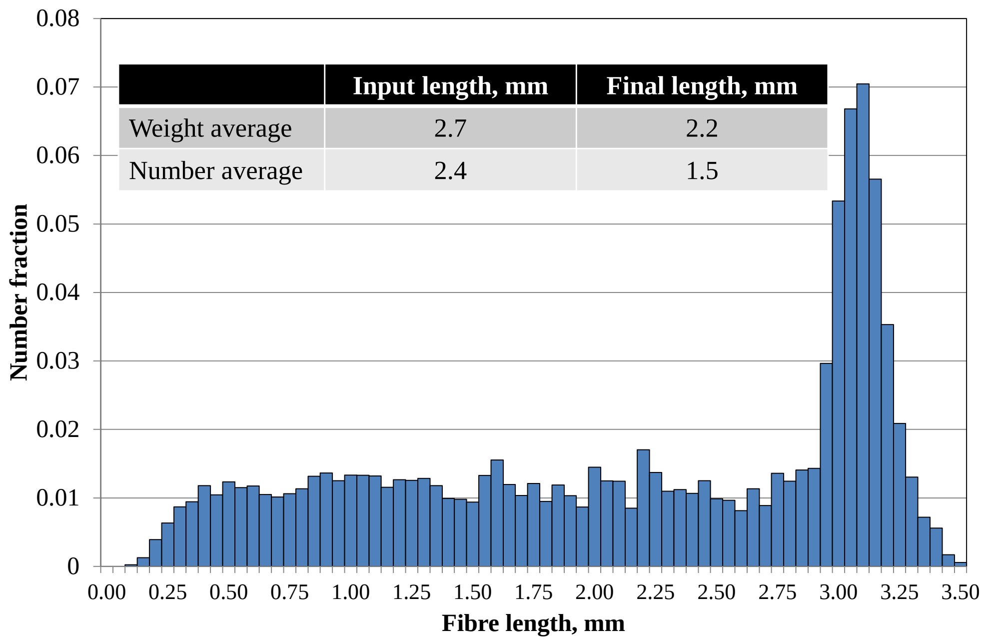

Figure 11. Length distribution of fibre obtained from composite reinforced with recycled, aligned carbon fibre with 60% volume fraction [16].

4. High fibre volume fraction composites moulded at lower pressures

Within the composites industry, the best mechanical properties are achieved in composites manufactured from unidirectional continuous fibre where a fibre volume fraction of about 60% can be achieved by moulding a pressures of less than 10 bar in an autoclave. Whilst a composite fibre volume fraction of 60% (vol) from aligned recycled carbon fibre has been achieved, processing pressures of up to 100 bar were required. Moulding at high pressure is inevitably more expensive and further work is currently being undertaken to gain a better understanding of the process of fibre alignment with the aim of being able to achieve better alignment at lower moulding pressures. In this section, work on liquid jet geometry and its dependency on fluid properties and suspended fibre is reported. Modifications were then made to the test rig and this resulted in the production of an aligned mat able to give a higher fibre content at a lower moulding pressure. Full size composite coupons were then fabricated and the tensile properties are presented.

4.1. Improving fibre alignment – understanding convergent jet behaviour

This section introduces the use of ink jet technology to provide an insight into the understanding of factors that governing the geometry of jet emerging from a given nozzle.

4.1.1. Jet geometry influencing alignment quality. The fundamental principle of the hydrodynamic alignment process is that short fibres are aligned by the velocity gradient along the flow direction in a convergent nozzle. Then, the jet of fibre suspension is deposited on a nylon mesh and the liquid drained to form an aligned fibre mat. The alignment process has been further developed through experience gained from the paper manufacturing industry. It was found through careful observation of the fibre suspension jet that an unstable jet (figure 12a) which splashes during deposition process will reduce fibre alignment. A varicose jet and air bubbles (figure 12– b, c) can misalign fibres.

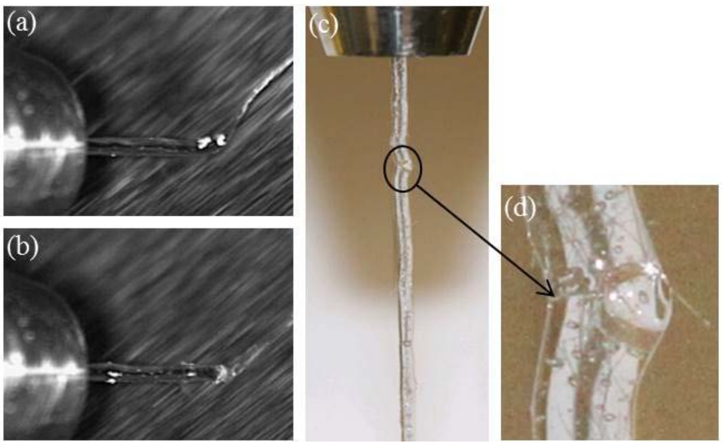

Figure 12. Photos of unsatisfactory fibre suspension jets – (a) unstable jet; (b) varicose jet; (c) jet containing air bubbles.

4.1.2. Investigating influencing factors on fibre suspension jet stability. Liquid jets are known to break up because of the interfacial forces between the jet and the surrounding air. When a liquid jet is discharged into air, disturbances on the jet surface will be augmented by the aerodynamic interactions between the jet and the surroundings. The most commonly accepted jet disintegration classification in fluid mechanics literature was developed by Ohnesorge (1936) who combined the Reynolds and Weber numbers and proposed a dimensionless Ohnesorge number Z (or Oh) [19].

$$Z = \frac{\mu_L}{\sqrt{\rho_L \sigma d_j}} = We_L^{0.5} Re_L^{-1} \tag{1}$$

where We_L is liquid's Weber number, Re_L is liquid's Reynolds number and d_j is the liquid jet diameter at nozzle exit. Based on Ohnesorge's work, Reitz suggested that there are four liquid jet breakup regimes which are presented in figure 13and table 2 [19] and Derby stated that the criterion for a drop to possess sufficient kinetic energy to be ejected from the nozzle is $We_{crit}\geq 4$ or $Re\leq 2/Z$ [20]. Therefore, a stable jet can be developed when $4\leq We_L \leq 8$.

Table 2. Expressions for breakup regimes [19].

Regime	Range
Rayleigh (varicose)	$We_L \geq 8$ and $We_g < 0.4$
First wind-induced (sinuous)	$1.2+3.41Z^{0.9} < We_g < 13$
Second wind-induced	$13 < We_g < 40.3$
Atomization	$We_g > 40.3$

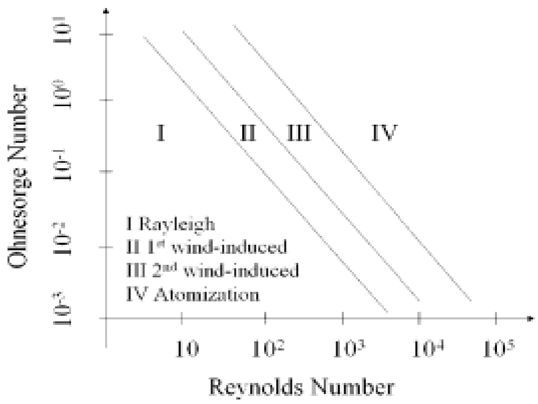

Figure 13. Classification of the modes of liquid jet disintegration [19].

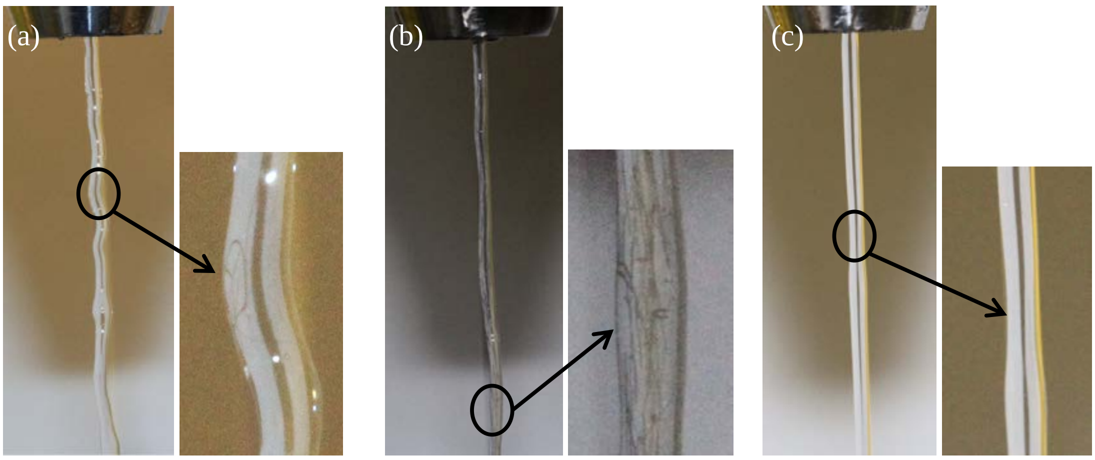

Figure 14. Images of fibre suspension jets and glycerine/water jets all at the same Weber number: (a) 12mm length fibre_0.05% volume fibre concentration jet – the fibre becomes disturbed in the wavy jet; (b) 3mm length_0.15% volume fibre concentration - fibre suspension jet and fibres are aligned with flow direction; (c) glycerine/water solution jet behaves varicose slightly.

In a two phase flow jet (liquid/fibre), the jet breakup regimes are likely to be different to the liquid jets described in figure 13 and table 2. Figure 14 shows that the fibre–liquid interaction and fibre–fibre interaction give jets different unstable effects although the Weber numbers are the same. Future work will focus on exploring the effects of different fibre lengths and fibre volume concentrations on suspension jet stability.

4.2. Exploring hydrodynamic alignment process - experimental investigations

This section reports the mechanical properties of composite, which contained aligned mats that were produced frm the modified alignment process. A Design of Experiment (DoE) approach was implemented to investigate correlation between factors that affected the alignment quality.

4.2.1. Materials. Tenax®-A HT C124 carbon fibre staples coated with a water-soluble sizing, was supplied by Toho Tenax Europe GmbH. The fibre lengths were 3 mm and 12 mm and the individual fibre nominal diameter is 7 μm. Fibre sizing was removed by placing the staples inside an ashing furnace at 550 °C for 15 mins. Oleon Glycerine 4810 with glycerol content ≥99.5% was supplied by Univar UK. The glycerine was diluted with tap water until a viscosity of around 400 mPas was measured at room temperature (15 ℃) using a Brookfield LVDVII viscometer. The diluted glycerine was used as a dispersion medium.

4.2.2. Manufacture of aligned mat and Design of Experiment (DoE). The hydrodynamic alignment process, as depicted in figure 8, was used for this study. During the alignment process, the fibre dispersion is pumped into a pressure pot and is pressurized to create a steady flow to a concentric conical nozzle that is located above a rotating drum. A nylon mesh positioned inside the drum is employed as a filter to separate fibre from dispersion medium. A vacuum suction is applied under the mesh to increase the liquid draining rate. The nozzle is fixed to a linear actuator and continuously moves forward and backward to create a certain width fibre mat. The filtered glycerine is separated from air by a cyclone and is pumped back to a storage tank for reuse. Once the target veil areal density has been met, the slurry deposition process will be halted. The mat is then washed with warm water to remove the remaining dispersion medium and an epoxy based binder is applied to maintain the aligned fibre orientation.

A 2-level full factorial experimental work plan was designed to explore the effects of processing factors on alignment quality of discontinuous carbon fibre in hydrodynamic alignment process. Details of the factors and levels are listed in table 3. The dimensions of the aligned mats made were 135mm in width by 900mm in length.

Table 3. Details of full factorial DoE.

Fibre length, mm	Fibre volume concentration in suspension, %	Wire / Jet transvers velocity ratio	Dewatering vacuum level	Number of replicates
3	0.05	1.5	Low	2
12	0.15	5	High	2

4.3. Dry fibre mats compaction results

A compaction test rig for measuring carbon fibre mat thickness (used to determine the average fibre volume fraction) under different compression pressure is shown in figure 15. The rig essentially contains two flat platens; the bottom platen is fastened to the base of a 5969 universal testing machine from Instron; the upper platen is connected to a 50 kN load cell, which is mounted to the machine crosshead. System compliance of the entire rig was first determined by closing the two platens together at a crosshead speed of 1mm/min until a compression pressure of 10 bar was registered by the load cell. The crosshead location was automatically logged by the machine's control software during the test. The data was used to measure the distance between the two platens. Two linear variable differential transducers (LVDTs) were also mounted on the top platen to provide further readings of the distance between the platens. To measure the thickness and volume fraction of the mats, test specimens 35mm length square were obtained by stamping carbon fibre mat using a cutting template. Each mat was weighed before the test. Then 4 samples were stacked together and carefully transferred to the bottom platen and were compressed at 1 mm/min until 10 bar pressure was attained. The thickness and fibre volume fraction of the mats were determined by taking average of the readings from the two LVDT units.

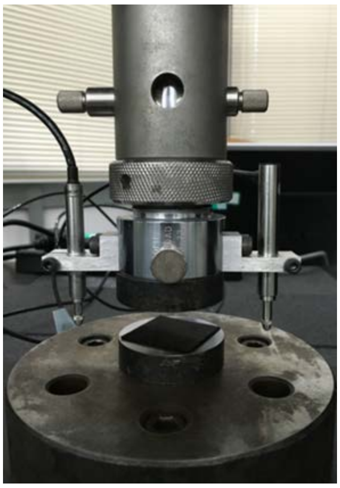

Figure 15. Dry fibre mat compaction testing apparatus.

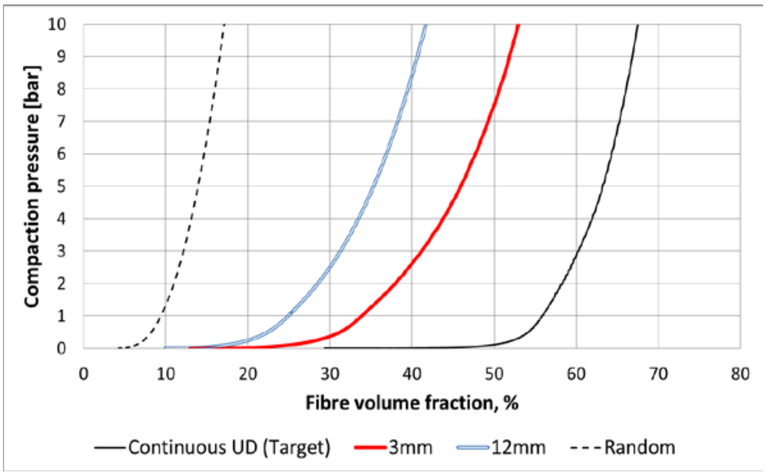

Figure 16. Dry fibre mat compaction test results.

The compaction result is presented in figure 16, includes two benchmarks: a 2D planar random mat and a unidirectional continuous fibre (UD) mat as well as two aligned mats of different fibre lengths obtained from the full factorial test. Under 10bar compaction pressure, the fibre volume fraction of UD mat is 68% though the random mat can only give 18% fibre volume content at the same pressure. This clearly indicates that an aligned fibre orientation improves fibre volume fraction. For discontinued short fibre (especially the fluffy recycled fibre), to achieve good mechanical properties, high fibre volume fraction is required. Thus, alignment is a necessity. It can be seen that with 0.1% of fibre concentration in the suspension, the 3 mm fibre aligned mat can achieve a volume content of 47.7% at 10 bar, which is 32.5% higher than the 12 mm fibre mats and this is attributed to having a better fibre suspension quality, which is free of fibre bundles for the shorter fibre. Further improvement was made to the rig and process operating parameters and this allows better alignment performance to be attained from a suspension with higher fibre concentration. At 10 bar pressure, the achievable fibre volume content for the 3 mm and 12 mm mats are 52.8% and 41.6% respectively.

4.4. Manufacture of aligned fibre composite in an autoclave and tensile testing

Laminates were made from discontinuous aligned fibre mats (details given in table 4) and MTM57 resin films supplied by Cytec Industrial Materials, UK. The materials were vacuum bagged to remove trapped air. The compacted materials were then moulded at 120 °C, 7 bar pressure, for 1 hour. Finally, it was cooled down to around 60 °C under pressure before demoulding.

Table 4. Specifications of aligned preforms and composite samples.

Fibre length	Fibre mat areal density	Fibre volume fraction	No. of layers fibre mat	MTM57 resin film areal density	No. of layers resin film	Moulded laminate dimensions
mm	g/m^2/layer	%	/	g/m^2/layer	/	$l \times w \times t$(mm^3)
3	72	46	24	83	16	280×135×1.87
12	71	43	20	83	19	280×135×1.93

Tensile tests were performed with a 250 kN load cell which was attached to the crosshead (figure 15), and a test speed of 1 mm/min was applied for all specimens, i.e. 4 specimens from each laminate. Extensometer was attached on each specimen to record strain. Micrographs of the cross-sectional view of the laminates are shown in figure 17 and a close-packed fibre structure is observed. Image analysis software was used on figure 17 and showed that the fibre volume content of 3 mm fibre composite is 46% and the fibre volume content of 12 mm fibre composite is 43%.

Figure 15. Tensile testing setup.

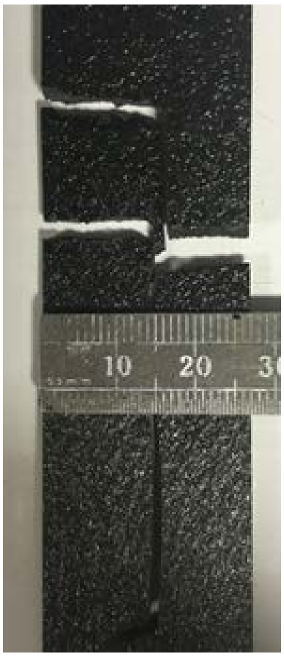

Figure 16. Fracture cross section of 3mm fibre laminate composite.

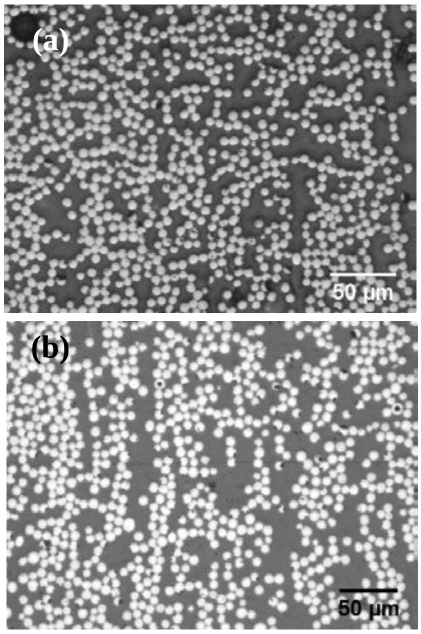

Figure 17. Micrograph of aligned composite: (a) 3mm_46% fibre volume fraction; (b) 12mm_43% fibre volume fraction.

4.5. Results

The results obtained from the tensile test are presented in figure 18. It can be seen that with 46% fibre volume fraction, 3 mm fibre laminate composite gave a 15.15 GPa higher tensile modulus than the laminate made with 12 mm fibre although the fibre length is much smaller. This indicates that good fibre alignment has been achieved and the aligned fibre structure can make a significant improvement to the mechanical properties. The void fraction in the composites was estimated, by image analysis, to be less than 2% for the composite made with 3mm fibre and less than 0.5% for the composite made with 12 mm fibre. The properties achieved for the 46% volume fraction composite give a specific tensile modulus of 0.057 GPa/kg/m^3 and a specific tensile strength of 0.42 MPa/kg/m^3, given a composite density of 1.5 kg/m^3. These specific properties are greater than alternative competing structural materials with the exception of carbon fibre composites and show the potential of fibre alignment for the manufacture of high value composites from recovered carbon fibre.

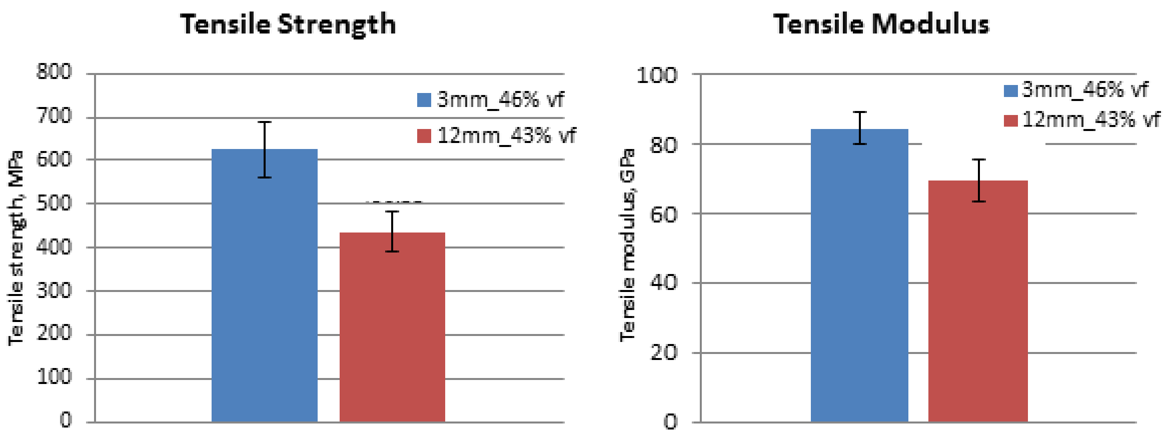

Figure 18. Tensile properties of composite reinforced with 3mm and 12mm aligned fibre mats.

5. Conclusions and future directions

New applications for increasing quantities of recovered carbon fibre produced by emerging commercial recycling activities are sought and use as structural reinforcement utilises the excellent mechanical properties of this material. Wet lay processing to produce nonwoven mats lend themselves to discontinuous recovered carbon fibre that typically has a fluffy form. However, random nonwoven mat structures cannot be processed into composites with mechanical properties better than readily available low cost metallic and composite materials. Nonwoven mats with aligned discontinuous fibres allow significantly better mechanical properties to be achieved, through higher fibre volume fractions in the composite and reduced fibre breakage during moulding processes. A process for achieving high levels of fibre alignment has been developed, based on existing technology, and this demonstrates the potential of fibre alignment for the production of high value composites that give specific mechanical properties better than other widely used low costs metallic and composite materials and approaching those of virgin carbon fibre composites.

Future work must now focus on developing alignment processes further, considering in particular process capital and operating costs, so that commercially viable processes are available to support future expansion of the emerging carbon fibre recycling industry.

Acknowledgment

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