

The Potential for Fibre Alignment in the Manufacture of Polymer Composites from Recycled Carbon Fibre

Kok H. Wong, Thomas A. Turner, Stephen J. Pickering and Nicholas A. Warrior
University of Nottingham

Copyright © 2009 SAE International

ABSTRACT

This paper studies the feasibility and potential benefits of aligning recycled carbon fibres, in the form of short individual filaments, to manufacture fibre reinforced polymer composites. A review of fibre alignment processes is presented to provide insight into the different alignment technologies. The main focus is on wet hydrodynamic processes, which offer a high degree of alignment for discontinuous fibres. The process parameters that govern the alignment efficiency are also reported. The effect of alignment on fibre packing efficiency in the manufacture of composites is included, together with a report of preliminary fibre alignment results obtained from three different alignment processes.

INTRODUCTION

Carbon fibre reinforced thermosetting composites have been widely used in different applications. This is motivated by their high specific mechanical performance, ease of part integration and corrosion resistance. However, due to the cross-linked nature of the polymer matrix, the composites are not readily biodegradable and thus conventional disposal in landfill is not encouraged. In addition, the cost of landfill is continually increasing due to both a shortage of capacity and the implementation of various regulations on the type of materials allowable for landfill. Incineration is another widely used disposal route. Despite the recovery of some heat energy, there is environmental concern over release of toxic components from polymers containing halogenated flame retardants, but more importantly the valuable carbon fibres are completely destroyed in the high temperature oxidation process. From the aspect of recovery of valuable materials, together with the expansion of global environmental awareness and the

mission of sustainable management, significant effort are now being made towards recycling both the polymer and carbon fibre [1]. Recycling also offers further benefits such as a reduction the demand for new resources and significant energy savings as carbon fibre manufacture is very energy intensive.

Unlike metals recycling, there is little infrastructure for recycling thermoset composite waste as it is in the early stages of development. The waste may be granulated and milled into powder to be used as a low cost substitute for fillers, such as calcium carbonate or to provide a degree of reinforcement from the fibres. However, as the fibres are still covered in polymer that has been mechanically damaged during size reduction, this limits the value that can be realised from the carbon fibre. Another issue in mechanical recycling is the chemical compatibility between the recycle and the host matrix. A further barrier includes the difficulty in identifying the recycle composition, due to the presence of heterogeneous types of thermosetting resins, additives, contaminants, coating and painting.

Alternatively, a fluidised bed thermal process, which is highly tolerant of contamination, has been shown to be capable of recycling carbon fibres which exhibit satisfactory mechanical property retention [2]. This process is particularly robust in dealing with end-of-life composite parts, which usually contain foam cores and metallic inserts for fasteners.

More recently, the feasibility of complete material recovery from pyrolysis processes has been widely pursued. The processes have been claimed to be self-sustainable [3] and to operate without causing significant detrimental effect to the fibre mechanical performance [4, 5].

Applications for reusing the recycled materials must be developed in order to make the recycling strategy economically sustainable. Carbon fibre is a valuable commodity and thus a considerable financial opportunity is available for exploitation from the use of the recycled fibre. However, reuse of these fibres poses a significant challenge because of the physical form of the material. The recycled fibres are generally short, filamentised and with a random 3D orientation, as a result of the size reduction of the end-of-life waste for ease of transportation, separation and processing. These fibres can be milled and used as a low-grade additive for non-structural applications, such as for anti-static and electromagnetic interference shielding as well as for enhanced heat conduction. However, to maximise economic and functional viability, the recycled fibre should be reused as a reinforcement in high performance structural engineering applications. This can best be realised by the achievement of high levels of fibre alignment in order to give the maximum strength and fibre content in the finished composite. The intention of this paper is to provide an insight into the feasibility and benefits of fibre alignment for short recycled carbon fibre, so that an intermediate product is generated to be used for high-grade applications. The paper also reports some preliminary results obtained from our alignment processes.

FIBRE ALIGNMENT

Fibre reinforced composites are highly anisotropic materials. High mechanical performance is attained when all the fibres are subjected to a load parallel to their major axis. Greater performance is further accomplished when more fibres are packed into the composite. In the case of continuous fibre, the aligned and close-packed structure is readily acquired. For example, a continuous unidirectional fibre prepreg usually contains a fibre content of 55-60% by volume. On the contrary, it is more difficult to obtain a high fibre volume fraction from short, filamentised recycled fibres. If the fibres are used in their raw form, for example, in bulk moulding compounds, the fibre volume fraction that can be practically achieved is around 10% [6]. An intermediate step is required to pre-compact the fibres in order to achieve higher fibre content. Papermaking is a well established process to convert fluffy fibres into a 2D non-woven mat. And, as a binder is used the also greatly reduces the airborne hazards from free fibres and improves handling. Ideally, composites made of this non-woven veil have isotropic properties. However, unlike unidirectional prepreg, the attained fibre volume fraction is low, due to the nonwoven 2D random orientation of the fibres together with a low apparent bulk density and high degree of loft. With a vacuum bag moulding setup, the volume content attained is around only 9% [7]. A substantial amount of pressure is required to obtain higher fibre volume content from the non-woven structure. For example to achieve 30% and 40vol%, a compression pressure of 70bar and 140bar respectively is required [7]. Unfortunately, such a high pressure results in severe fibre length degradation, limiting the

fibre reinforcing potential. In order to fully harness the reinforcing potential of the recycled fibres, they should be adequately packed at low pressure and this can be realised only by a substantially parallel fibre alignment. Other advantages of having an aligned short fibre structure include high formability and drapability.

Much work has been done to control the orientation of short fibres. Conventional textile processes such as carding exert a mechanical force on the fibres and a good degree of alignment can be obtained provided the fibres are long, tough and flexible. However, as carbon fibre is a very brittle material, it can be severely damaged under this intense mechanical movement. Injection moulding and extrusion are the other areas where the effect of process parameters on the degree of fibre orientation has been studied. It has been widely reported that a 3-layer structure is observed across the thickness of a composite fabricated by the injection moulding process. Fibres that located at the top and bottom layers are preferentially aligned parallel to the flow direction. However, those at the centre show that their major orientation distribution is perpendicular to the flow. In the case of an extrusion process, a more uniform aligned structure is seen. A convergent die or nozzle is usually used in the extrusion process to produce a velocity gradient between the stationary surface layers and the accelerated centre streamlined flow to give fibre alignment. Composites produced from these processes typically have a fibre volume less than 35vol% because of processing difficulties arising from greater interaction between fibres and machine parts. These high interactions also result in degradation of fibre length. This degradation increases with higher fibre volume fraction. To achieve a better degree of orientation with minimum length degradation, the distance between two immediate fibres should be large enough for them to have free rotation and translation. This suggests that a very dilute fibre suspension is required. The excessive fluid content is then removed in order to increase the fibre volume content. These requirements are met by an innovative hydrodynamic process, developed by Explosives Research and Development Establishment (ERDE), UK in the 1960s. In this process, a dilute fibre suspension stream was directed to a tapered, profiled convergent head ending in a slit of variable width. The head travelled repeatedly over a flat fine wire mesh on which the aligned fibres were deposited. Vacuum suction was applied underneath the mesh to increase the liquid filtration rate. Several other approaches have also been developed using different forces for alignment: magnetic field [8] and electric field [9, 10]. Of the processes mentioned above, the ERDE one is considered to be the best as the working principle is based on well established and mature hydrodynamic technology. It also offers versatility for pilot scale research and can readily be upgraded to a continuous process.

EDRE WET FIBRE ALIGNMENT PROCESSES - ERDE devised and developed three patented fibre alignment techniques based on extrusion [11], filtration [12] and

centrifugal processes [13]. Each process will be discussed individually in the subsequent sections.

Extrusion process - The extrusion process was also known as an alginate process. It was developed for aligning asbestos and silicon carbide whiskers in an ammonium alginate solution [14]. The fibre suspension was extruded under slight pressure through an orifice of 2mm diameter and 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. Further heat-cleaning was required to remove the residual alginic acid. 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 [15].

Filtration process – Fibre filtration is the most widely published ERDE fibre alignment process. It started with a batch process capable of making aligned fibre sheets up to 40 x 30cm but was later developed into a continuous process for a 152mm wide sheet roll [15, 16]. The process can be resolved into three separate operations: Fibre dispersion, alignment and separation.

Dispersion stage - In the first fibre dispersion stage, the fibres must be individually separated from the agglomerate in a glycerine medium. The degree of dispersion depends upon the fibre properties (length, concentration and stiffness), suspension medium viscosity and the design of the dispersion unit. Fibre concentration has to be carefully controlled in order to minimise interaction between adjacent fibres. Theoretically, this threshold varies inversely with the square of the aspect ratio of the fibre [17]. Generally, fibres with higher stiffness are preferable as they will not tangle while in the suspension [16]. High viscosity is desirable for achieving good fibre dispersion. A special impeller is needed to ensure high shear rate with low turbulence for fibre dispersion without entraining air bubbles which can have an adverse effect on the orientation [15].

Alignment stage - In this stage, the suspension is pumped gently into a convergent alignment head, in which the fibres are aligned due to a velocity gradient across the fluid streamlines. The alignment head travels repeatedly above a fine mesh on which the aligned fibres are filtered out from the solution, aided by vacuum suction. Murty and Modlen observed that the alignment started even before actually entering the convergent region [18]. In order to get a high degree of alignment, the fibres must be straight and completely dispersed during the dispersion stage. The alignment head must be designed in such a way that the top entrance should

have a large angle of convergence and the angle is then reduced to a small value at the end slit opening. A curved-sided contour is thus recommended [17]. The alignment head reciprocating speed should be slightly greater than the speed of the suspension leaving the end slit in order to stretch and lay the suspension satisfactorily on a mesh without resulting in suspension accumulation. As quoted by Dingle [17], the degree of alignment is independent of viscosity but dependent on fibre length. However, high viscosity was required to provide a combined effect of good dispersion and flow stability, which in turn did improve the alignment. A stiffer fibre was also preferred as it will remain straight during the alignment stage [16].

Separation stage – The glycerine medium must be removed rapidly without distorting the aligned fibre structure deposited on the mesh. This is done by using a mesh with a low resistance and an efficient positive displacement pump. The filtration process takes up most of the cycle time and the filtration rate can be increased by reducing the glycerine viscosity by the application of heat [16]. If the filtration rate is slow, then the slit width can be reduced to produce a thinner film in order to preserve the alignment [19]. However, this might lead to fibre blockage at the smaller exit slit. After the separating process, the carrier medium removal was completed achieved by washing with a gentle water spray. By replacing the stationary wire filter mesh with a moving endless belt, the ERDE process could produce a continuous dry fibre mat and prepreg of 152mm width [15].

The degree of alignment attained from this filtration process is good and more than 90% of the fibre population is in the range $\pm 15^\circ$ of the desired direction [20]. However, a high pressure difference must be supplied across the filter bed to remove glycerine efficiently. As the mat thickness increases, the filtration process becomes more difficult due to reduction in through-thickness permeability. Thus, an increasing dwell period is required between each reciprocation of the alignment head, leading to an economically unacceptable production rate [21]. 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. As a result, a improved centrifugal process was suggested, as discussed in the next section.

Centrifugal process – The centrifugal process has the same fibre dispersion stage, but different design for the converging head and fibre filtration [13, 21]. Instead of a narrow slit, a circular converging nozzle with an inlet and exit diameter of 14mm and 0.7mm respectively is used. The nozzle reciprocates axially across the inside a rotating drum made of stainless steel perforated sheet on which the fibre suspension is sprayed at a speed of 4.55m/s. Instead of a vacuum suction, the dispersion medium was extracted by a centrifugal force. It was reported that the centrifugal force increased the fibre filtration rate. For optimum alignment, the acceleration g-