Economical Carbon and Cellulosic Sheet Moulding Compounds for Semi- and Non-Structural Applications

ABSTRACT

The use of discontinuous carbon fibre plastics (DCFP) in automotive applications is increasing. The material offers a light weight and stiff alternative to conventional short-fibre glass reinforced composites such As Sheet Moulding compounds (SMC), and therefore has a wider deployment potential, in both structural and semi-structural applications   Prohibiting factors to wider utilisation of this material are cost and formability. Carbon moulding compounds available commercially, are expensive not least because of the cost of the fibre, but also these systems contain high fibre volume fractions up to 60% V_f, which hinders flow, and must therefore be unfilled, and involve the use of non- standard resin systems that are relatively expensive compared to standard polyesters. This study examines the performance of cheaper CFSMC grades , produced using a standard SMC production route involving a filled polyester resin to reduce both the volume of fibre reinforcement and resin costs, but also maintaining the flow characteristics and formability found in regular SMC. A particular requirement of modern automotive structures is impact energy absorption for crashworthiness, The impact characteristics was therefore a focus where performance is compared to standard/commercial DCFP grades and with two cellulosic reinforced SMC's manufactured from Rayon and Jute, that offer further cost and weight savings compared to carbon fibre SMC. Mechanical properties were assessed relative to increasing fibre length, and volume fraction. The additional effect of fibre bundling was also investigated, where a strong positive correlation was found between increasing impact energy absorption and increasing bundle diameter, where the toughening mechanism at work is discussed. It was found that although much lower volume fractions (V_f)were employed, the new CFSMC grades offered superior impact strength compared to some higher V_f, DCFP's offered commercially. Cellulosic SMC was also shown to have potential as a a cost effective replacement to commercial carbon fibre SMC grades in non-structural applications where light weighting rather than mechanical performance is paramount.

Keywords: Carbon Fibre, Jute Fibre, Sheet Moulding Compound, Discontinuous

1. INTRODUCTION

Future developments in the automotive sector will be dominated by the requirement for low carbon vehicles, with highly efficient combustion engines or the use of alternative propulsion technologies such as hybrid-electric and all electric power-trains. Whatever the final solution, the light-weighting of vehicle structures will be an important requirement in upping fuel efficiency and range, where the 500kg car is a viable target for the industry[1] . Composite materials are now well established within aviation as a viable light weight alternative to metals, where the next frontier for composites is the automotive industry. However these two industrial sectors differ markedly in their materials and manufacturing requirements. Airframe manufacture is characterized by modest production runs where 10,000 units would represent a large program, where individual aircraft can have a service life of 25+ yrs. Consequently, unit manufacturing cost is not the primary consideration and more importance is attributed to operational cost over the aircraft's life, and hence, the dominating requirement to shave airframe weight as much as possible, and therefore, maximise the payload. In contrast, the automotive industry deals in production runs measured in the 100,000's, where unit production cost is a dominating factor, and the importance of operational cost, has, until recently, been a more secondary issue.

So far, attempts to transfer composite production technologies from aviation into the automotive sector has met with limited success in some niche markets such as , the sports and supercar sectors. This is because the associated production methods are too slow, labour intensive and costly, for wider application in large volume automotive production runs. This situation has prompted research seeking to develop cheaper and faster production methodologies, such as automatic tape laying, robotic fibre placement, rapid resin transfer moulding and rubber forming [2].

Sheet Moulding Compound (SMC) is by far the largest type of thermoset composite used in the automotive industry today. SMC is a glass-reinforced short-fibre composite, incorporating a highly filled formulated matrix –where the thermoset used is typically orthophalic unsaturated polyester. SMC can be as cheap to produce as steel in production runs below 40k [3], offers excellent thermal compatibility with steel, adequate crash management and fatigue resistance but with a 40% weight saving. Because it is a moulded product, manufacture involves short production cycle times, where mouldings can provide a so called "class A" surface finish, and therefore SMC's are frequently used for exterior body panels and other non- and semi-structural components. SMC developers now want to extend the usage of moulding compounds to meet more structural requirements, where high glass content grades have been developed for load carrying frontend structures [4]. Others have tried to develop grades based on carbon fibre to provide stiffer components, where carbon fibres (CF) offer distinct advantages in terms of stiffness and weight saving, with commercial grade carbon fibres offering a modulus of 230 GPa – three times higher than E-glass (70 GPa) and a specific gravity only 70% of E-glass fibres. Previous studies [5, 6] have shown that CFSMC exhibits greatly improved stiffness, where improvements in mechanical properties have been the main interest area for researchers working in the area. A study by Cabrera-Rios and Castro [7] examining the potential of CF reinforced SMC for high stiffness automotive truck parts concluded a statistical improvement in flexural and tensile strengths, and a higher modulus in CF samples compared to glass. The study compared the mechanical properties of SMC test plaques made up of different combinations of reinforcing plies containing 50% weight glass or carbon. Another study by Broderick et al [8] discussed improvements in stiffness and weight reductions in fender support systems, door structures, windscreen bracings and various other small components, when selectively hybridising glass SMC with CF reinforcement. SMC producers are now manufacturing commercial grades to counteract weight gain in vehicles to help them attain government set emissions targets. In a comprehensive study by Stachel and Schäfer [9] examining the use of CF in high volume production (SMC manufacture) and comparing resulting properties to steel/aluminium equivalents, it was shown that CF–SMC could provide a weight reduction potential of up to 60% compared to steel, and would provide for the first time, a technology to utilize carbon fibres in reinforced thermosets at high volume production. The so called "Advanced SMC" formulation debuted on Daimler's Mercedes SLR Silver Arrow sports car in a three-piece scuttle panel. However, of the three original carbon fibre SMC parts on the SLR, two have since been replaced for cheaper alternatives. The price of carbon fibre and concerns about availability continue to limit its use [10, 11].

Other alternatives exist such as "HexMC" - a compression moulding compound manufactured by Hexcel Composites. HexMC consists of chopped bundles of carbon fibre (50mm X 8mm) consisting of 60% discontinuous carbon fibre in an epoxy resin. HexMC is lightweight (1550kg/m^3) and is therefore utilized in aerospace, typically in secondary structures and interiors for aerospace applications such as the window frames on Boeing's new 787 Dreamliner aircraft. HexMC is also a high cost material and demonstrates low flow compared to other moulding compounds where an 80% tool coverage is required [12], making production times slower and more costly. One approach to solving the cost problem is to use recycled carbon fibre grades [13, 14], however, impact performance deficiencies still remain. In summary, Several DCFP's are now available on the market, the main attraction in using carbon fibres is a substantial stiffness improvement [9, 10, 15], and light weighting potential. There are drawbacks, where impact performance is inferior to glass and due to the high V_f of carbon fibres used in these grades, cost is prohibitive, and flow/formability is poor compared to standard SMC. For these reasons the material remains confined to niche applications where the need for stiffness, rather than weight saving is the main driver.

This study compares the performance of carbon fibre with other potential lightweight fibres produced via a standard SMC route, where the fibres are deployed in a standard glass SMC paste formulation. The study also considers the potential offered by other light weight cellulosic fibres. Jute natural fibres and reconstituted cellulose fibres (rayon) could provide a cheap lightweight alternative to carbon in non-structural and semi-structural applications. The fibres examined included a standard carbon fibre (Toray T700SC-12000) (1800kg/m^3), and several types of cellulosic fibres (1500kg/m^3); Cordenka™ 700 high tenacity rayon, and aligned jute bast as a representative natural fibre.

Previous investigations conducted into natural fibre for SMC, have reported shortcomings in mechanical properties, the most serious of these being impact energy absorption. Voorn et al [15] sought to develop flax fibre reinforced SMC, in order to produce lightweight versions that could match the performance of glass. Whilst stiffness and strength could be matched, impact energy absorption was far lower, at 3-7 KJ/m^2 compared to 40-70KJ/m^2 for glass in comparative tests. Huda *et al* [16] in a review of natural fibre composites in the automotive sector, repeatedly details the shortcomings in impact performance demonstrated by natural fibre SMC composites, whereas in a study by Brouwer [17], a factor 5 decrease was observed in flax-reinforced SMC as compared to equivalent glass versions. The results of a recent investigation by the authors [18] has shown that by developing optimized mesostructures, through fibre bundling, the impact performance of mechanically inferior fibres such as natural/cellulosic's, can be substantially improved. The work showed that fibre bundling, (in cellulosic fibres) accounted for a factor of 5 improvement in energy absorption. This study seeks to apply these bundling techniques as a means to improving impact energy absorption in the fibre types listed. The study also, examines other influential factors such as bundle tow size, bundle length and fibre volume fraction. The general objective was to establish the potential usage widow for conventionally formulated high flow CFSMC and cellulosic SMC grades, and given the importance of crashworthiness in modern automotive systems, how far impact performance could be improved in these lightweight low cost materials, compared to currently available DCFP's.

2. METHOD

This section describes the production of three grades of SMC manufactured from carbon fibre, rayon and jute fibre reinforcement. Plaques measuring 228 x 228 mm were formed from each grade using conventional hot compression moulding. The plaques were subsequently sectioned into 25 test pieces complying with ISO 179[19] and ISO 14125[20] for test and measurement of impact and flexural properties.

2.1. Materials

2.1.1. Resin

Unsaturated ortho-phthalate polyester resin was used for SMC manufacture. The properties of the pure cured resin are presented in Table 1:

Elongation at Break (%)	Tensile Strength (MPa)	Tensile Modulus (GPa)
1.7	60	3.8

Table 1: Properties of Unsaturated Ortho-phthalate Polyester Resin [21]

2.1.2. Fibres

Toray T700SC-12000 carbon fibre tow [22] was used as received and after the application of a dilute coating of Baybond PU330, Bayer [23]. Baybond PU330 is an anionic / non-ionic polyester - polyurethane resin dispersed in water designed to improve bonding with unsaturated polyester resins. The application of this coating increased the linear density from 8000 dTex (untreated fibre) to 8080dTex forming non-fibrillating bundles. High tenacity rayon (cellulose II), produced using a proprietary variation of the viscose process, was supplied by Cordenka GmbH [24] in three linear density values of 1220 dTex (720f), 1540 dTex (500f) and 5000 dTex (1100f). The Rayon fibres were used as received, and also after application of a dilute coating solution of Baybond PU330. The application of a coating caused the linear densities of the three tows to increase from 1220dTex, 1540dTex and 5000 dTex to 1260dTex, 1680dTex and 5170 dTex respectively and formed non-fibrillating bundles.

Raw, untreated jute was supplied directly from India [25]. Application of dilute Baybond PU330 was carried out manually to these fibres for comparison with bundled carbon and rayon fibres. These jute fibres do not have a given linear density value, however samples originating from the same strand of bast were compared before and after coating. The weight of jute after the application of coating was approximately 9% w/w greater than an equivalent, untreated strand of jute. After application of coating jute formed a large assembled roving of well bound bundles grouped together similar to assembled E-glass gun roving but with much greater diameter of both bundle and roving.

All natural and regenerated cellulose fibres were oven dried at 80°C for 12hrs prior to SMC compounding and fabrication.

The properties of each of the reinforcing fibre can be found in Table 2,

Fibre	Density (g/cm^3)	Elongation at Break (%)	Tensile Strength (MPa)	Tensile Modulus (GPa)	Weight Saving (% Glass)	Price (£ / Tonne)
Glass	2.53	5	3,500	80	0	1,166
Jute	1.50	1 - 2	336 - 558	31	40	315
Rayon	1.51	12 - 13	483	4	40	600 – 1,800
Carbon Fibre	1.80	2	4,900	230	29	12,500 – 25,000

Table 2: Fibre Reinforcement Price and Properties [24, 26-33]

2.1.3. Fibre Coating (Bundling)

Baybond PU330 is an anionic / non-ionic polyester - polyurethane resin dispersed in water formulated for glass and carbon fibre use with unsaturated polyester resin systems. Supplied PU330 suspension contains a 30% w/w solid which was diluted tenfold with distilled water to achieve a suspension containing 3% w/w solids content. A solids content of 3% w/w was found to prevent bundle separation during compression moulding and was consequently used on all fibre types throughout this study.

Binder was applied to carbon and rayon fibres in continuous tow form using the apparatus illustrated in Figure 1. Fibre tow was drawn from the spool into a coating bath were the fibre is immersed for approximately 10 seconds, through adjustments in the winder speed. The coated tow was dried using the direct application of forced hot air at 120°C before finally being wound onto the collection bobbin. The coated tow was stored in a fan assisted oven at 50°C to finalise drying until being cut into to length. The tension of the tow entering the coating bath was found to control the uptake of coating and therefore the morphology of the dry product. The tension was controlled by way of a swing arm tensioner added directly after the tow spool.

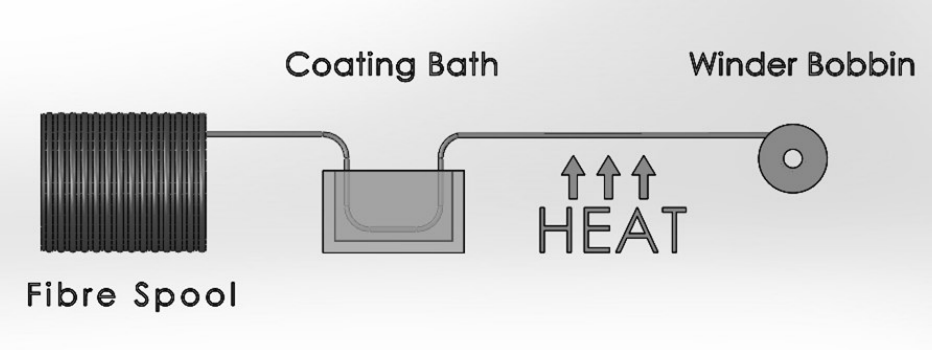

Figure 1: Fibre Tow Sizing Arrangement

Jute fibre was coated by immersing the as-received strands of fibre in a bath of solution. The fibres were steeped for 20 seconds longer than the continuous tow fibre samples due to a greater volume of fibre compared to the continuous coating arrangement. The coated fibres were removed from the binder bath and wound through squeeze rollers to remove excess solution. The fibres were combed in a manner similar to that used to comb and hackle wool fibre. The coated and combed fibres were left to dry at ambient temperature before being placed in an oven at 50°C for the final drying stage.

2.1.4. Fibre Cutting

A Cutex TBC-50 ribbon cutter was used to cut both unbundled and bundled fibre to 25mm lengths initially. The cutting machine is able to vary its cutting length from 1 to 100mm in 1mm increments and was additionally used to produce fibre sample lengths of 10mm, 12mm, 20mm, 30mm, 40mm and 50mm.

2.1.5. Sheet Moulding Compound (SMC)

SMCs can be visualised as being made up of three major components 1) unsaturated polyester resin 2) fibre reinforcement and 3) particulate reinforcement or filler. The SMC paste is a combination of the resin, activators, catalyst and filler which are mixed together to form a viscous paste. All reagents used to produce the SMC paste were provided by Menzolit UK and used as received. A general purpose glass SMC paste formulation was used for all samples. The exact formulation used is the intellectual property of Menzolit UK and therefore it will not be disclosed in detail.

2.1.6. Specimen Manufacture

All samples used an identical SMC paste formulation where the various fibres examined were incorporated at the same volume fraction as a standard glass SMC (19% V_f) unless otherwise stated.

2.1.7. SMC Compounding

Figure 2 is a representation of the SMC production line used throughout this study. The pre-mixed paste containing all the resin and filler components is fed into two doctor boxes before the production run begins. A constant layer of the paste then covers the carrier films as they are drawn through the doctor box system, and a layer of the particular fibres under test, is then sprinkled randomly onto the bottom carrier film by hand, before it and the top paste-covered carrier film are sandwiched together to form the final SMC mouldable sheets. The sheets are then stored at 35°C for 48hrs to allow the so called "thickening process" to proceed (this is characterised by a steep rise in viscosity – which renders the SMC handleable). Thickened SMC sheets are then compression moulded.

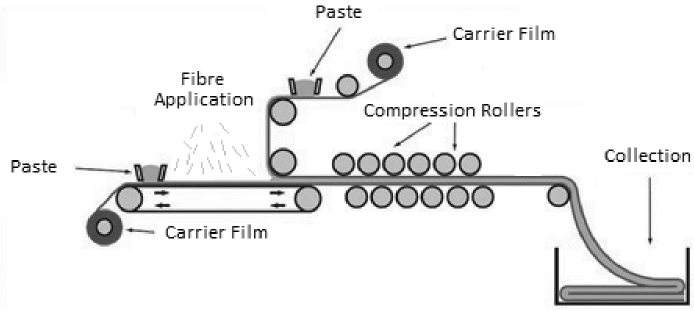

Figure 2: Schematic Diagram of an SMC Manufacturing Line

2.1.8. SMC Moulding

Sample plaques measuring 228x228mm and 4mm thick, were moulded, for each of the different fibre formulations. Two plaques were hot compression moulded from each SMC formulation to better represent the variability commonly found in this class of composites. Green-state SMC was cut into110mm x 110mm squares and made up into so called "charge packs" which consist of multiple layers of green state SMC. The packs were then placed in the centre of the square mould cavity translating to an approximate mould coverage of 33%. Compression moulding was performed with a hot press, where all samples were cured at 145°C and a pressure of 4.1MPa for 3 minutes, -replicating the curing conditions of the standard, industrially produced, SMC composite.

2.2. Testing

2.2.1. Density

The densities of moulded plaques measuring 228 X 228mm were determined by displacement of water in accordance with ASTM D792 [34]. Plaques were weighed on a digital balance in air and then again whilst submerged in distilled and bubble free water. The density was calculated using Eqn. 1

$$D = \frac{a}{a + w - b} \times 997.5 \qquad \textbf{Equation 1}$$

Where D is density of measured sample in Kg/m^3, a is the apparent mass of specimen in air, b is the apparent mass of specimen and suspension apparatus completely immersed in water and, w is the mass of fully immersed suspension apparatus.

Specific properties were calculated using the general equation

$$\varphi_s = \frac{\varphi_e}{\rho} \qquad \textbf{Equation 2}$$

Where φ_s is the specific value of interest, φ_e is the experimentally measure value and ρ is the calculated density. All results have been expressed as Specific values, as this provides the most useful comparison between different composite types.

2.2.2. Test Sample Preparation

ISO 179 and ISO 14125 specify that for samples to comply with specification they must measure 80mm (±2.0mm) x 10mm (±0.5mm) x 4mm (±0.2mm). Samples were cut from moulded plaques using a Denford CNC router fitted with a composite cutting tool bit. The CNC router cut 25 samples from the 228mm x 228mm moulded plaques in both vertical and horizontal orientation relative (Figure 3). A 20mm gap was left at the edge of the panel where fibre bunching is known to occur.

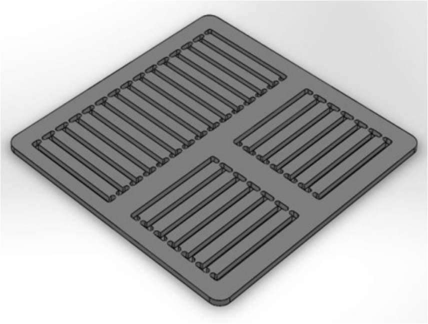

Figure 3: Sample Orientation as Cut by CNC Router

Once milling had completed the 2mm tabs retaining the samples to the plaque were cut using a band saw.

2.2.3. Impact testing

Impact energy absorption (also referred to as Impact Strength) was measured using a Charpy impact test arrangement. Tests were carried out using a Ceast Resil Impactor Junior with a non-instrumented 4 J impact head. Samples were prepared and tested in accordance with ISO179 standard. A total of 30 samples were tested for each reformulation from a random selection of samples cut from two cured panels with the same formulation

2.2.4. Flexural testing

Three-point flexural testing was conducted in accordance with ISO178 using a Lloyd Instrument EZ20 and a 500 N load cell, with tests performed at an extension rate of 1.9 mm/sec. A total of 20 samples were tested for each reformulation from a random selection of samples.

3. Experimental Results and Discussion

3.1. Density

Calculated densities for the produced CFSMC, Rayon SMC and jute SMC are presented in Table 3 and have been used throughout to convert experimental impact and flexural values to specific values to allow easy comparison between SMC formulations.

SMC	**Unbundled / Bundled**	**Volume Fraction (%V$_f$)**	**Density (kg/m^3)**
Commercial Glass	Bundled / Assembled	19	1.9
CFSMC	Unbundled	19	1.601

	Bundled		1.615
	Bundled	28	1.622
	Bundled	37	1.630
Rayon	Unbundled	19	1.598
	Bundled		1.629
	Bundled	28	1.562
	Bundled	37	1.548
Jute	Unbundled	19	1.524
	Bundled		1.533

Table 3: Calculated densities for CFSMC, Rayon SMC and jute SMC [35]

3.2. Carbon Fibre SMC

3.2.1. Impact Strength

Carbon fibre SMC is believed to offer potential property improvements compared to those demonstrated by traditional glass SMC, namely stiffness and reduced material density. Carbon fibre compounded with thermosetting polyester resin was used to benchmark performance of CFSMC against standard glass SMC, and examine additional aspects such as bundling and fibre length effects. Light CFSMC grades were also compared to other lightweight cellulosic fibre alternatives.

Figure 4 presents the specific impact strengths found from CFSMC manufactured from 12mm, 25mm and 50mm bundle lengths at fixed V_f of 19%, 28% and 37%. Unbundled carbon fibre (25mm) with a 19% V_f attained a specific impact strength of 44.3 kJ.m/kg whilst the bundled equivalent reached 50.6 kJ.m / kg. The impact strength of the samples containing bundled fibre increased with increasing bundle length, from 24.3 kJ.m / kg (12mm) to 59.6 kJ.m / kg (50mm). It is believed that the reasons for this lie in the fact that by increasing the length of fibre reinforcement the number of fibre ends are in effect, reduced. This has an important effect, as fibre ends are known [36, 38] to be areas of stress concentration, where failure preferentially occurs. The performance jump seen in CFSMC produced with 12mm and 25mm fibres suggests that samples containing 12mm fibre bundles are below the critical fibre length where the dominant failure mechanism arises from the accumulation of stresses at the fibre ends. Kim (2008) has suggested that when the length of a fibre is below the critical fibre length the end effects become a dominating consideration for composite failure [39]. The findings reported here support this view.

The highest impact energy absorption demonstrated by these polyester / carbon fibre SMC was achieved by samples manufactured from 25mm fibre bundles and a loading volume of 28% V_f.

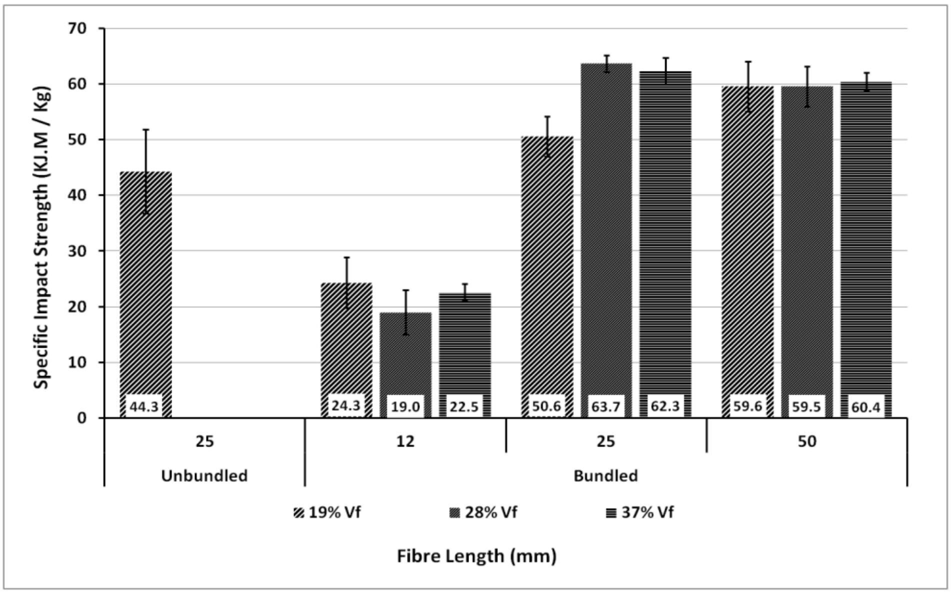

Figure 4: Specific impact behaviour of CFSMC with changing fibre length and volume fraction. (*Error bars signify 1 standard deviation)*

Four DCFP's produced commercially have been used as comparators throughout this study [40-44]. Commercially available DCFP's are known to suffer from poor impact properties, where in one case the reported Charpy impact strength of 40.1 kJ.m / kg is significantly lower than many of the grades produced in this study (Figure 4). This result is interesting because these commercial DCFP grades have been optimised to extract the best properties from the carbon fibre, i.e. higher cost vinyl ester resins are employed, as these are thought to impart superior bonding to CF than polyester, also, these commercial systems contain little or no mineral filler and employ very high volume fractions (up to 60% V_f discontinuous) and the fibres are often present as a continuous unidirectional layer (70% V_f). In contrast, the experimental CFSMCs examined here, were made using a standard SMC manufacturing approach to reduce costs, where polyester resin, heavily loaded with mineral filler was used, where fibre content was kept at a low comparative volume fraction. The fibres had, however, been bundled to reduce loft during manufacturing, this assists wet-out throughout the process and enables the composite meso-structure that manifests in the composite, to be optimised for greater impact energy absorption, as reported by the authors previous studies [18].

3.2.2. Flexural Strength

The flexural properties of CFSMC have been determined from three-point-bend experiments (ISO 179). Figure 5 shows the flexural strength CFSMC manufactured from 12mm, 25mm and 50mm fibre bundles with increasing volume fraction (19%, 28%, 37% V_f). Unbundled CFSMC samples achieved a flexural strength of $76.6x10^6$ N.m/kg whilst the bundled equivalent, demonstrated a somewhat lower strength of $58.7x10^6$ N.m/kg. Unbundled carbon fibre has the ability to fibrillate during moulding providing greater fibre distribution and homogeneity throughout the sample cross-section compared with bundled samples. The greater fibre distribution in unbundled samples providing increased adhesion surfaces within the matrix aiding stress transfer, and tends to avoid the development of fibre-rich and resin-rich phases giving a more homogenised composite. It is expected therefore, that well distributed fibres would have a greater interaction with the matrix, and contribute more to the strength of the composite.

The flexural strength increases with increasing fibre length from $58.7X10^6$ N.m/kg (12mm) to $64.8X10^6$ N.m/kg (25mm) and $82.7X10^6$ N.m/kg (50mm) in 19% V_f CFSMC. Equivalent glass SMC (19%) has a flexural strength of $94.7x10^6$ N.m/kg [35] which is 12.7% greater than the best performing equivalent CFSMC grade. The CFSMC flexural strength results are matrix dominated at these low V_f's, as seen from (Figure 6). It is seen that the large 12K CF bundle does not disperse throughout the sample leading to an inhomogeneous CFSMC.

The increase in volume fraction from 19% - 28% - 37% V_f, results in a general upward trend, as expected from rule of mixtures. Additionally, bundling of the fibres is thought to constrain fibres, limiting fibre matrix interaction and stress transfer. Because the fibres are bundled, the reinforcement is constrained in the composite to discrete areas; this means that matrix properties remain dominant in flexure. Fibre length effects did manifest however, where the samples produced using 50mm fibre bundles outperformed the shorter length bundles at all loading volumes.

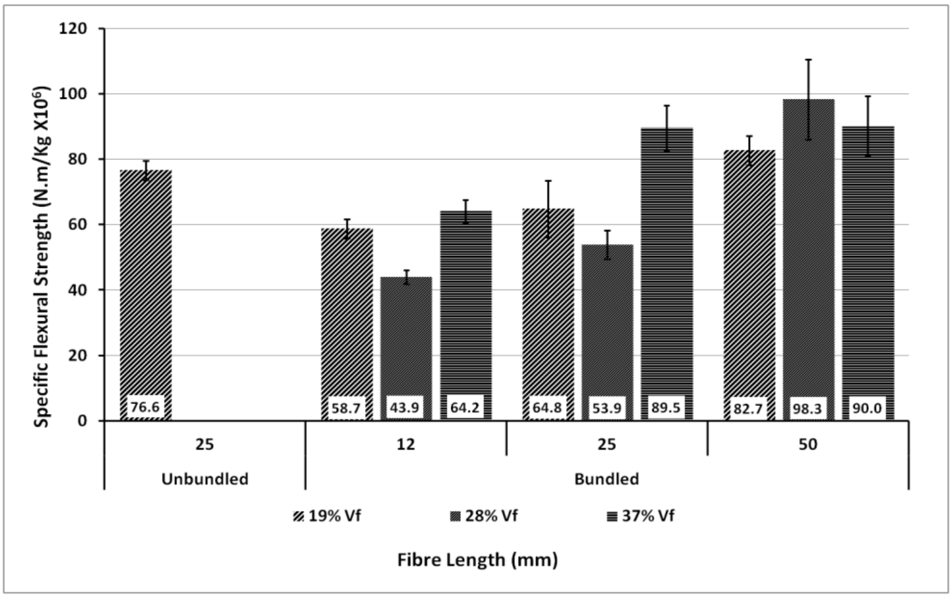

Figure 5: Specific flexural strength of CFSMC with changing fibre length and volume fraction.
(*Error bars signify 1 standard deviation)*

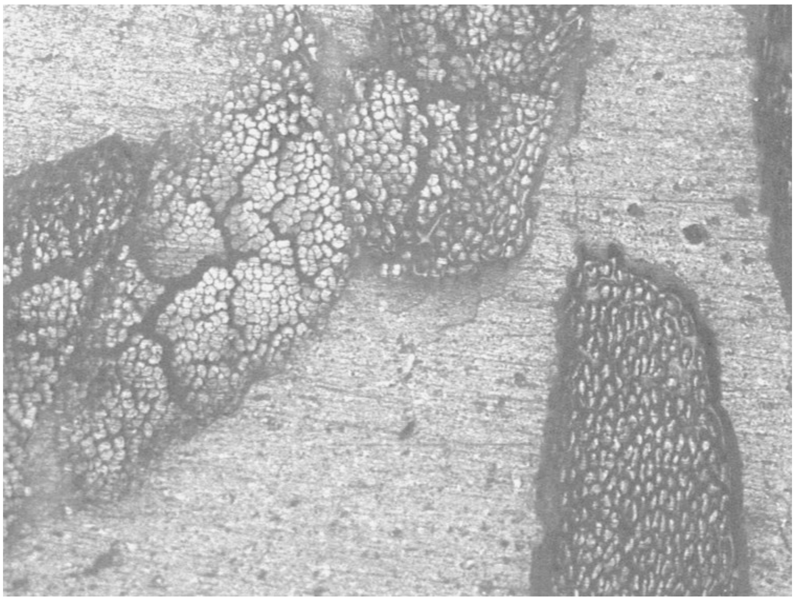

Figure 6: Poor separation of bundled 12k carbon fibre tow in CFSMC

The aforementioned commercial DCFP grades quote specific flexural strengths of $228x10^6$ to 428 $x10^6$ N.m/kg [40-44] which are double the greatest value achieved by the experimental CFSMCs examined here. The disparity is however expected when considering the fundamental differences between the commercial and experimental materials, where the commercial grades use very low levels of mineral filler, which in turn, allows large fibre loadings (60% v/v to 70% v/v ([40-42, 44]). Relatively, low viscosity, resins used in commercial grades also aids in fibre wet-out, a common problem with uncoated, lofty, fibres. Optimised fibre wet-out would not be possible, given the high viscosity, filled, systems used in glass SMC and experimental CFSMC. HexMC has circumvented unforeseen wet-out problems during moulding by pre-wetting the fibre bundles with epoxy resin before assembling the chopped strands into a moulding compound, this has gone some way towards improving performance consistency, however consistent performance of parts remains an issue, fibre rich areas often occur due to resin squeeze out, localised property variations also manifest because of fibre orientation effects.

3.3. Flexural Modulus

Figure 7 shows the specific flexural modulus values obtained from all CFSMCs tested in this study. It is seen that 12mm fibres have greater stiffness than longer fibre CFSMCs containing the same V_f. When the fibre length remained constant but V_f was increased, the modulus did not increase as would be expected from rule of mixtures [27, 41, 42].

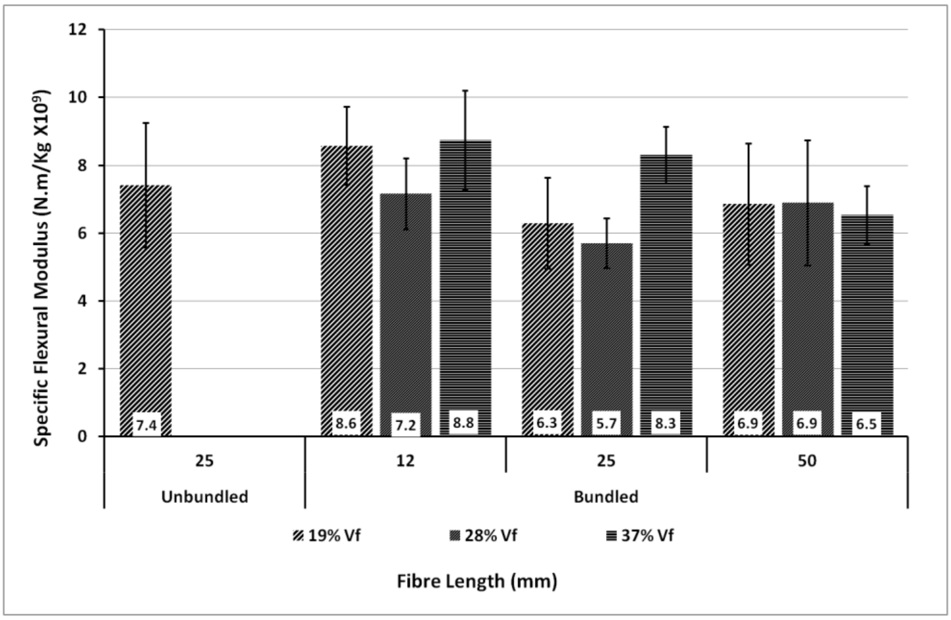

Figure 7: Specific flexural modulus of CFSMC with changing fibre length and volume fraction.
(*Error bars signify 1 standard deviation*)

Comparing maximal flexural strength and modulus results in the CFSMC grades tested, with commercially available DCFP's (60% V_f) and a glass SMC (19% V_f) (Figure 8). It is evident that the commercial DCFPs have superior specific flexural strength and stiffness. This is however expected, given that these DCFPs are designed for high stiffness and strength aerospace/motorsport applications.

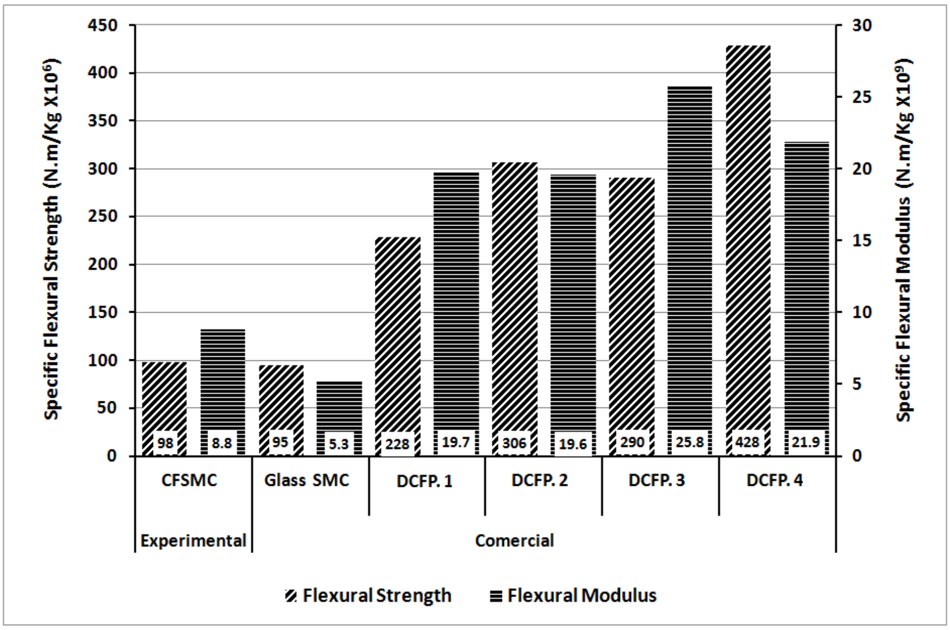

Figure 8: CFSMC with commercially available moulding compounds [35, 40-44]

However these superior properties come at a cost. It should be noted that with these industrial DCFPs, a mould coverage exceeding 80% is very often required, this constrains flow during moulding. A key property of the CFSMC developed in this study, is its capacity to flow during moulding, requiring only 30% mould coverage. This allows complex shapes to be formed, and delivers superior surface finish to the part. This factor is important as it allows the moulding of complex part designs incorporating ribs and folds. In this way, parts can be stiffened considerably, counteracting the comparatively low modulus of the CFSMC grades produced in this study. .In Summary, short fibre polyester CFSMC offers good impact strength combined with high flow levels and reduced material costs making it a suitable material for automotive applications where high flow characteristics allows component stiffness to be realised by design.

3.4. Rayon SMC

The substitution of cellulosic fibres in place of carbon fibre raises the prospect of lightweight fibre reinforcement at a fraction of the cost. Typical comparison figures are detailed in Table 2.

The specific impact behaviours of 1220 dTex (720f) Rayon SMC with 10mm, 12mm, 20mm, 25mm, 30mm, 40mm, 50mm bundle lengths are presented in Figure 9. The impact strength of these samples is seen to lie between 16 kJ.m/kg and 39 kJ.m/kg with changing bundle length. Unbundled Rayon samples (25mm) achieved an impact strength of 16.2 kJ.m/kg whilst the bundled equivalent, reached 25.1 kJ.m/kg. A contributing factor is thought to be the poor wet-out in unbundled fibre SMC due to extensive loft (high air/void volume and spring back) as can be seen in the sample shown in Figure 10. This factor degrades the mechanical performance achievable in the finished composite because fibre/matrix bonding is impaired and the void content is high. Through the application of a coating to Rayon, bundle loft is reduced aiding wet-out during compounding. Evidently, bundling fibres by means of applying a coating increased impact strength in all bundled samples compared to the unbundled grade. Unlike CFSMC, increasing the fibre length does not routinely increase the impact strength although a definite toughening effect was observed at the 50mm fibre length.

Substitution of glass and carbon fibres which are characterised by low elongation to break of 2-4%, (similar to the polyester matrix), with a cellulosic fibre that has an elongation at break almost 5 times greater than the polyester matrix (Table 1 & Table 2) means that failure during impact and flexure is more likely to occur in the matrix.

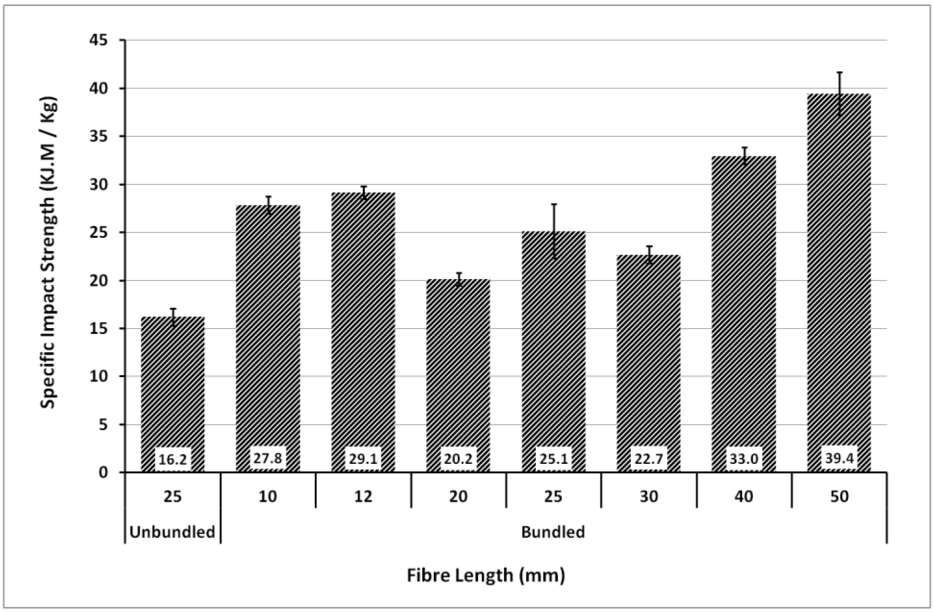

Figure 9: Impact resistance of 1220 dTex (720f) Rayon reinforced SMC (19% V_f) with changing fibre length (*Error bars signify 1 standard deviation)*

Figure 10: Poor fibre wet-out achieved in matured Rayon SMC

Fibre bundling has previously been shown to influence the performance of cellulosic fibres [18]. In order to examine this effect further, the dTex value (and therefore, bundle diameter) was increased and impact performance measured.

Figure 11 shows the influence of increasing dTex value on impact strength of Rayon SMCs. Increasing the fibre dTex value improved the toughness in both unbundled and bundled fibres. A noticeable increase in impact strength was shown in 1220 dTex and 5000dTex samples that had been bundled.

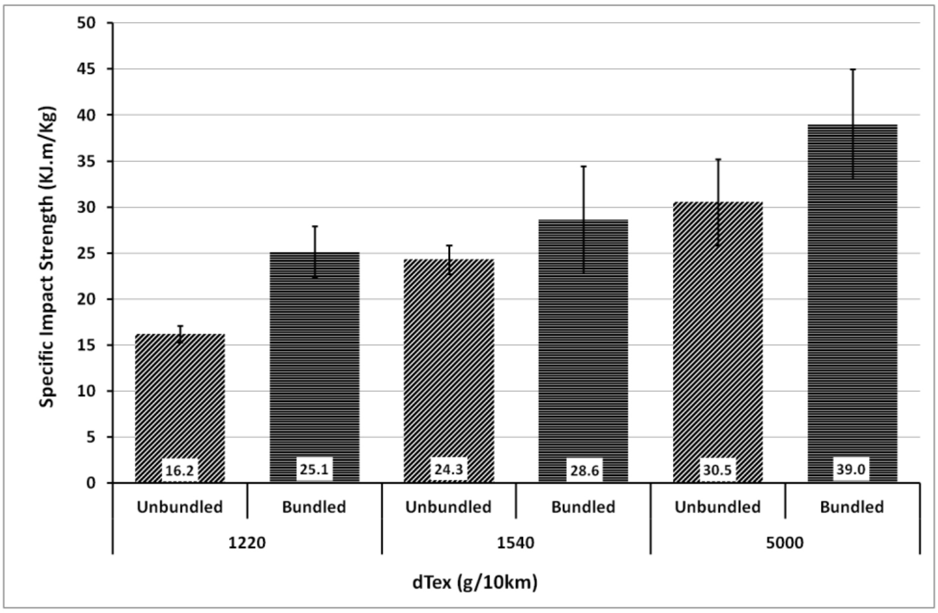

Figure 11: Effect of increasing bundle dTex on impact strength (25mm, 19% V_f)
(*Error bars signify 1 standard deviation*)

Figure 12 plots the effect of increasing volume fraction on the specific impact strength of samples manufactured from 5000 dTex rayon bundles. The impact strength increases from 39.0 kJ.m/kg (19% V_f) to 48.9 kJ.m/kg (28% V_f) and then levels off to 48.1 kJ.m/kg (37% V_f) as was previously seen in the CFSMC results and reported to occur in glass SMCs [45].

This suggests that in cases where the reinforcement is of sufficient strength to deflect cracks back in to the matrix (i.e. not fracture), a point is reached where increasing the V_f value does not automatically lead to tougher composites. This finding supports the crack deflection toughening model proposed, where the central feature of the energy absorption mechanism is reliant on increasing the crack path length and constraining it to the matrix, i.e. the failure mechanism is essentially similar to an inter-laminar fracture, where toughness of composites are known to be derived from the energy dissipation properties of the matrix. Bradley and Cohen [46, 53] stated that fracture toughness in matrix-failure dominated systems (such as here), can be derived simply by the product of the fracture toughness of the matrix, times the volume fraction of the matrix. Though this assessment is simplistic and has since been modified [46, 47] This view would therefore go some way to explaining the levelling off of toughness with increased fibre volume fraction (and hence reduced matrix V_f) as reported . The possible energy absorbing mechanisms at work in Mode I failure have been summarised by previous investigators [47]. These include;

i) The formation of a new crack surface of the main crack tip in the matrix

ii) Plastic deformation and or micro-cracking,

iii) Fibres bridging the crack.

The increasing of crack path length would therefore increase energy absorption due to the increased energy required to form new surfaces. This study highlights how this effect is linked to the fibre architecture, where strong fibres do not fail but defect cracks around themselves. There will however, be a limit to the increase in energy absorption once this effect is fully deployed and the crack is fully confined to the matrix. The reasons for this stem from the fibre architecture commonly found in 2D moulded composites, where all fibres are oriented 90° to the crack path. When the number of bundles is increased (higher V_f) within a typical sample cross-section, two effects will manifest: increased crack deflection due to more bundles, and an increase in the number of fibre ends, hence more opportunities for the crack to propagate vertically, which is the more energetically-favourable behaviour. At some point, these countering effects will level out, where no further increase in toughness could be achieved through increasing the number of bundles. This simple model explains the plateau effect observed, and that reported by others in cases where strong fibres such as carbon and glass are used. Truss *et al* [48], examining fracture mechanisms at work in discontinuous fibre composites, also identified this behaviour, where propagating cracks would travel along fibres in a transverse direction until encountering a fibre end, where the crack would then return to the plane of maximum stress.

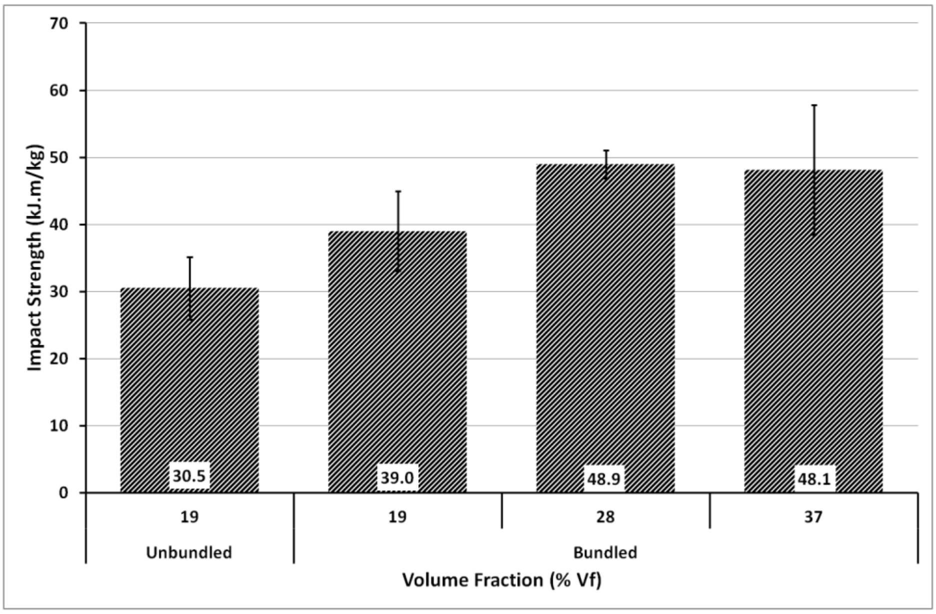

Figure 12: Specific impact strength on Rayon SMC manufactured from 25mm, 5000dTex and increasing V_f (*Error bars signify 1 standard deviation)*

Figure 13 shows typical fracture surfaces found in CFSMC and Rayon SMC samples tested, and schematic representations of the failure path. The left-hand image shows an unbundled sample where fibres were broken and the crack path was directly in line with the point of impact, as depicted in the accompanying schematic. The central image compares the bundled case, where because the bundled fibre do not fail, the crack is deflected horizontally in a transverse direction on meeting a fibre bundle. The only point where the crack can progress vertically is on encountering a fibre bundle end. This behaviour considerably increases the crack length and therefore the required energy to create new surfaces. The right-hand image shows a further case where a fibre bundle happens to be aligned with the impact point. In this case the bundle acts as a weak point giving a low value for fracture energy absorption.

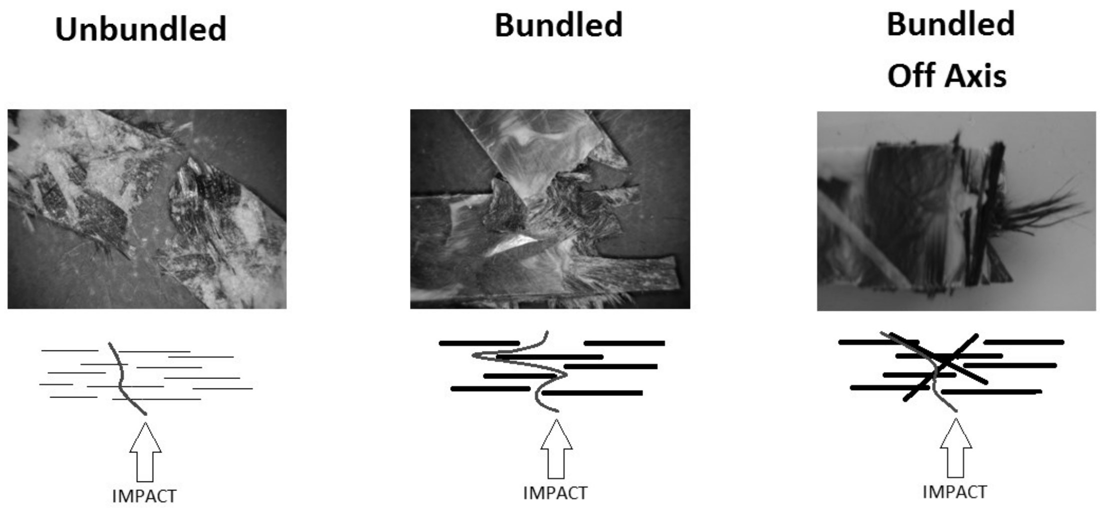

Figure 13: Fracture behaviour observed in typical fracture surfaces seen in CFSMCs and Rayon SMCs

3.5. Jute SMC

Figure 14 summarises the impact properties of Jute SMC with bundled and unbundled carbon fibre, and rayon SMCs manufactured from 25mm fibre bundles with a volume fraction of 19%.

Elementary jute fibres (Table 2) demonstrate substantially higher tensile stiffness compared to elementary Rayon fibres (Jute - 31GPa, Rayon – 4GPa) and is significantly cheaper. Rayon ranges in price between £600-1,800 per tonne whilst jute is £315 per tonne [28, 31, and 32]. Jute is a natural fibre consisting of cellulose I whilst Rayon consists of the lower crystallinity (and hence lower stiffness) cellulose II. [49] The structure of natural jute is complex, consisting of small fibre bundles each only 0.5 – 6.0mm that vary in shape and cross-sectional diameter; the bundles are contained in a weak lignocelluloses polymer (<40%) making extraction of fibre properties complex [50, 51].

Rayon has the advantage over Jute in that the meso-structural aspects such as the bundle size and the exact length of the individual elemental fibres which make up the bundles are controllable. This is important as it allows the fibre architecture to be controlled and thereby higher impact energy absorption in an inherently weaker and less stiff fibre, can be achieved. The higher impact strength of unbundled CFSMC's is thought to be related to the increased ability of the elementary fibre to deflect cracks during impact because of CFs high relative strength, resulting in increased energy absorption.

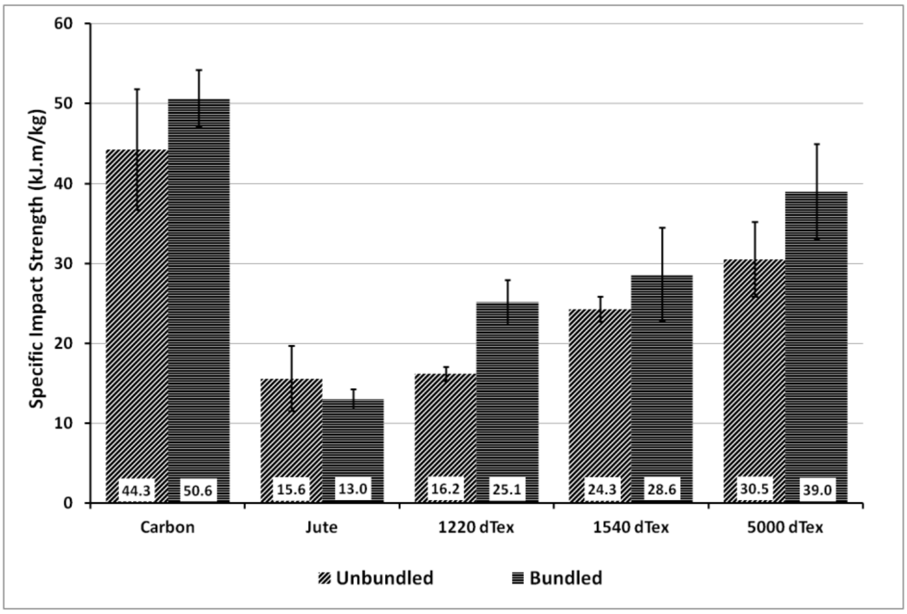

Figure 14: CFSMC, jute SMC and Rayon SMC specific impact property comparison (25mm, 19% V_f)
(*Error bars signify 1 standard deviation)*

Figure 15 compares the flexural strength properties between jute CFSMC and Rayon. The application of a coating to all cellulosic SMCs has the effect of increasing flexural strength whilst the opposite was found for CFSMC. Bundled jute has a comparable specific flexural strength to bundled carbon fibre which suggests a poor load transfer from matrix to fibre in the CFSMC, given the greater specific strength properties of carbon fibre. Glass SMC with comparable V_f has a specific strength of 94 N.m/kg x10^6, - greater than CFSMC and jute SMC, again indicating a relatively poor interface between matrix and reinforcement in these alternative grades.

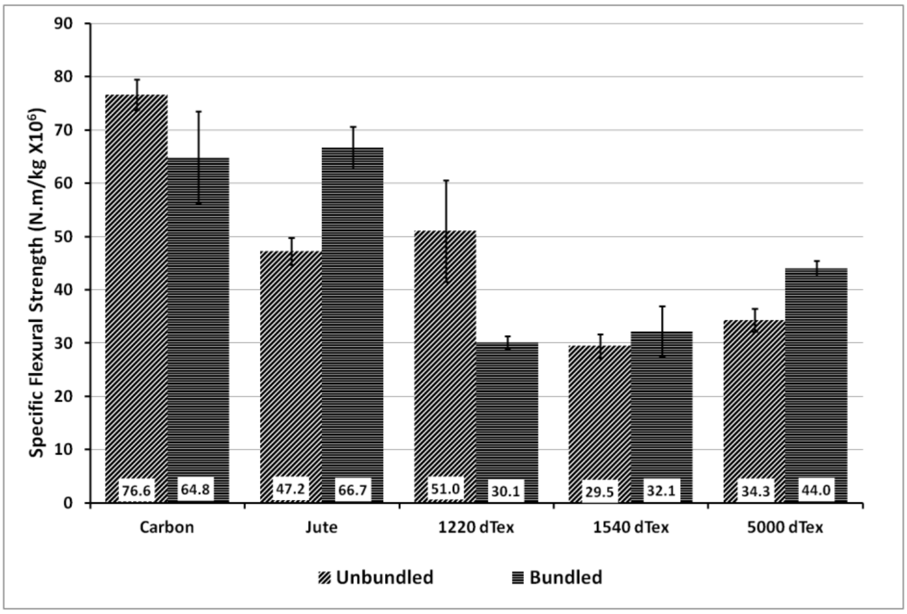

Figure 15: Comparison of flexural strength of CFSMC with cellulosic SMC (25mm, 19% V_f) (*Error bars signify 1 standard deviation)*

Figure 16 compares the flexural modulus figures for the jute, carbon and rayon SMCs. The application of a coating to rayon fibres increased the flexural modulus. This is thought to be due to the accompanying increase in alignment with neighbouring fibres in the bundle itself and in the finished composite due to greater flow induced alignment. The increase in stiffness may also be attributed to the increase in the second moment of area. - Small filaments when bound together to form a larger bundle will be inherently stiffer than single fibres.

The application of a coating in carbon and jute fibre composites has made little significant difference to flexural modulus because in these systems, the increase in bundle stiffness over elementary fibre stiffness would be less pronounced.

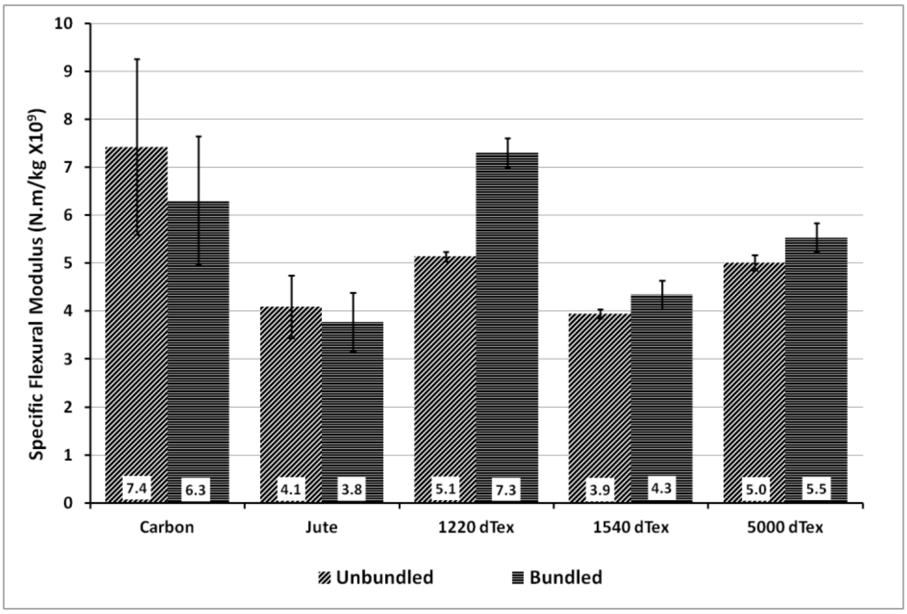

Figure 16: Comparison of flexural Modulus of Carbon Fibre SMC with Cellulosic SMC (25mm, 19% V_f)

(Error bars signify 1 standard deviation)

4. Conclusion

DCFP's such as CFSMC is currently produced commercially as an alternative to glass fibre SMC, to satisfy requirements for higher stiffness and/or lower weight characteristics. CFSMC is currently of much interest, where commercial suppliers are vying to present the market with superior grades. The latest development of a hybrid glass /carbon vinyl ester that boasts a flexural strength and modulus of 307x10^6 N.m/kg (483MPa) and 14.9X10^9 N.m/kg (23.4GPa) respectively (released in February 2014). This material has a cost similar to high performance glass SMC and consists of 20% w/w 12K carbon fibre and 30% w/w glass [52]. The mould coverage is still however, a problem requiring an 80% initial charge area due to low flow and formability characteristics.

This investigation has shown that the lower cost filler and resin system traditionally applied to glass fibre SMC can be suitably adapted for use with carbon fibre where the grades investigated compared favourably with commercial products in terms of impact energy absorption, but with the added appeal of being a "true SMC" material, i.e. demonstrating the ability to form intricate shapes and flow extensively during moulding. Also the low cost, filled polyester resin system and lower volume fraction of fibres used in these experimental grades, presents major potential cost savings over the high volume fraction, unidirectional fibre and vinyl ester resin systems utilised in commercial products.

The use of cheaper, but physically inferior, jute and Rayon fibres, when appropriately arranged and processed, can deliver meso-structures that impart excellent impact energy absorption where several grades were found to outperform commercial DCFPs. The toughening mechanism at work was found to be due to crack deflection – increasing crack path length and energy absorption. This toughening method was embellished via a fibre bundling route, where compounding effects of fibre length and tex value are reported.

Given a design specification for a semi- or non- structural part, cellulosic SMC can offer a weight saving compared with commercial DCFP and a substantial cost advantages, but are also able to demonstrate comparable specific impact energy absorption.

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