# GREEN COMPOSITES: SUSTAINABILITY AND MECHANICAL PERFORMANCE

J.M. Chard[1,2*], G. Creech[1], D.A. Jesson[2], P.A. Smith[2]
[1]Scott Bader Company Ltd, Wollaston, Northamptonshire, NN29 7RL, UK
[2]Faculty of Engineering and Physical Sciences, University of Surrey, Guildford, Surrey, GU2 7XH, UK
*Corresponding Author (Jonathan_Chard@scottbader.com)

**Keywords**: *natural fibres, mechanical properties, sustainable composites*

## 1 Sustainable Composites

The development of 'green' composite materials that can be (economically) competitive replacements for Glass Fibre Reinforced Polymer (GFRP) composites is an important area of research. For a 'green' composite to be classed as a sustainable alternative, the manufacture, use and disposal phases need to be considered. (The issue is made more complicated by the current flexibility in the definitions of both 'green' and 'sustainable' in this context).

Natural fibre (NF) composites utilise a range of fibres that occur in nature to produce systems that are perceived as 'green'. Here, the focus is on bio-derived organic fibres from vegetative feedstocks, but since there is a range of growing techniques, fibre isolation methods and other treatment processes available, NFs for composites might not be deemed a sustainable option once these economic, energy and environmental costs are taken into account.

As well as the option of using NFs, 'green' polymers are also available: some are promoted for use in packaging because of their ability to bio-degrade, although questions over the sustainability of the manufacture of these polymers are often left unanswered. A common technique for improving the green credentials of a standard thermoset resin is to incorporate a functionalised vegetable oil. Ultimately, this decreases the performance of the base resin since it often interacts with the resin constituents (having a negative impact on the optimum proportions), causing weaknesses in the final polymer structure.

Non-structural NF composites have already been commercialized in a range of applications including NF/bio-derived thermoplastic composites used for car inlays and internal door panels [1]. Since NFs have a lower density than glass, the specific properties of these composites are adequate for automotive applications. For NF composites to fully substitute glass composites - which are commonly used in conjunction with thermosetting resins - for structural components, high performance NF composites need to be developed.

Some of the previously identified issues with NF composites are the lower than expected mechanical performance [2] and the highly hydrophilic nature of the fibre [3]. It has been demonstrated that optimising the resin-fibre interface is one method that has the potential for improving the final composite performance [4], but such optimisation can be expensive (as well as having environmental impact).

## 2 Scope

The current study examines a range of glass and NF composites, utilising matrices of thermosetting resins with improved sustainability (reformulated to include constituents derived from vegetable oils whilst maintaining performance), in order to develop a range of more sustainable composites with useful mechanical properties. Depending on the properties required, the most appropriate sustainable composite can be selected. Hence, whilst attempting to make GFRP composites more sustainable through selection of an appropriate matrix, this work also examines a range of NF composites which have potential as competitors to GFRP composites, by offering reasonable mechanical properties with a reduced environmental impact.

## 3 Materials and Methods

### *3.1 Modified Resin Systems*

For the majority of composites, the largest contribution to the environmental impact of the manufacture of a composite is the resin system used [5]. Therefore, implementing sustainable changes to the resin system would achieve the largest positive impact on the sustainability of the final composite. For sustainable changes to a resin system to be commercially viable the 'more sustainable' resin system will need to achieve similar physical and mechanical performance, and be financially competitive when compared with the standard resin system.

In order for sustainable changes to a resin system to be effective, reformulation of the base resin with sustainable raw materials is required.

Two development thermosetting resins, based upon Crestapol® technology, have been manufactured by Scott Bader Company Ltd. These resin systems substitute some of the petrochemical based raw materials with (vegetable) oils. The first bio-resin utilises 22 % by volume from natural oils (Sustainable #1) and the second bio-resin utilises 40 % from natural oils (Sustainable #2). These development resin systems have similar production costs and are expected to achieve similar mechanical performance compared to standard thermosetting resin systems.

The standard thermosetting resins used in this research for comparison are cold curing unsaturated polyesters (UP), vinyl esters (VE) and urethane methacrylates (UMa). All of the resin systems that were utilized within this research were also supplied by Scott Bader Company Ltd.

Plaques of resin were cast with these resin systems and specimens were manufactured from these plaques. The purpose was to investigate whether by reformulating the resin with a green content it would be possible to achieve similar mechanical performance compared with standard thermosetting resins.

### *3.2 Natural Fibre Composites*

Substituting NFs for synthetic fibres is seen as another method for improving the sustainability of a composite as a result of the lower growth and processing requirements of NFs compared with synthetic fibres.

Depending on the type and growth conditions of the selected NF, and subsequent processing, the diameter of the fibre commonly varies between 5 and 80 μm but diameters of one to two orders of magnitude greater diameter can be observed. Again, depending upon the conditions mentioned above, the length of NFs can vary from 0.4 to 250 mm [6]. As the diameter of E-glass commonly varies between 9 to 25 μm, (within the diameter range of NFs) it means that NFs could be substituted for E-glass within a composite without the need for substantial changes to the composite manufacture process or resin system.

In order to understand the optimum configuration for a NF composite a range of fibre types and architectures need to be investigated.

The range of fibres used within this research varied from low quality, minimally processed to high quality, highly processed.

Hemp CSM (chopped strand mat), supplied by Hemp Technology, is a needle punched non woven fabric. The majority of fibres within this fabric are bast fibre with a low quantity of core fibre and other organic particulate contaminates. From examining the fabric it has been deduced that the fibres are not fully separated. Compared with the other NF fabrics used within this research, this fabric is considered the lowest quality material used within the current study.

A unidirectional hemp fibre fabric, supplied by Engtex, uses yarns of hemp fibre held together in parallel by a cotton cross stitch. The yarns have an approximate diameter of 0.75 mm and a twist of between 50 to 100 turns per metre. The fibres within the yarn are solely bast fibre and the fibres are almost completely separated. From examining this fabric, it was concluded to be of higher quality but requires a higher level of processing during manufacture compared with the hemp CSM.

Viscose rayon (VR) fibre yarn, supplied by Cordenka GmbH, is a continuous fibre of reformed cellulose. Even though this fibre has uniform cross-section, similar to E-glass, the composition of the fibre is completely cellulosic. The structure of the

VR fibre is Cellulose II, where the cellulose unit cell is projected along the (010) lattice plane. The two other NFs used within this research are Cellulose I, where the cellulose unit cell is projected along the (100) lattice plane. This VR fibre is considered the highest quality 'natural' fibre utilized within this research but requires the most extensive processing during manufacture. The first step was to filament wind the yarn around a frame to form a unidirectional layer of reinforcement.

An E-glass CSM fabric was utilized within this research to act as a comparison for the NF fabrics.

The composites were manufactured by a vacuum assisted resin transfer molding process. All of the NF fabrics were dried for two hours at 105 °C immediately before composite manufacture to remove residual moisture. The specimens were produced from these composites. A range of composites were manufactured using all of the resin systems previously discussed.

The fibre volume fraction of the composites was calculated using Archimedes density determination with canola oil for the solid resin, fibres and the final composites.

### *3.3 Residual Moisture With Natural Fibres And Its Effect On Mechanical Performance Of Composites*

Owing to the hydrophilic nature of a NF, the NFs will absorb moisture from the humidity within the surrounding air. This could be an issue for many types of thermosetting resin systems, as the cure of the resin can be inhibited due to the presence of water. The residual moisture within the NF could lead to a weakened fibre-matrix interface leading to a composite with lower mechanical performance.

This investigation examined the amount of residual moisture within the range of NFs and the effect that drying the fibres prior to composite manufacture had on mechanical performance. The fibres were stored at 20 °C with an approximate relative humidity of 40 %.

The composites for this part of the study were manufactured using the unidirectional hemp fabric with the UP and the UMa resin systems. Two sets of fabric were used; one set used the fabric stored under normal conditions and the other set was dried for two hours at 105 °C immediately before composite manufacture.

### *3.4 Fracture Toughness Of Natural Fibre Composites*

One of the critical design considerations for composites is their ability to resist crack propagation. The current understanding of fracture toughness for NF composites utilising thermosetting resin systems is limited. For NF composites to be considered as a structural material the fracture toughness needs to be investigated.

As the fracture toughness of standard thermosetting resin systems is low compared with other engineering materials, it is anticipated that the addition of minimally processed NFs should increase the fracture toughness of a thermosetting resin system through process such as crack deflection and fibre debonding, fracture and pull-out.

This investigation examined the fracture toughness of the hemp CSM composite compared with the unreinforced resin system and the E-glass CSM composite (test method given below).

### *3.5 Mechanical Testing*

The resin systems were mechanically tested in tension according to BS EN ISO 527 – Part 1: 96 [7]. The 25 mm width coupon composite specimens were mechanically tested in tension according to BS EN ISO 527 – Part 4: 97 [8]. The method used for measuring the fracture toughness of the hemp CSM composite and E-glass CSM composite was single edged notched in tension with associated compliance calibration [9].

All of the test procedures were conducted using an Instron quasi-static test machine (3382) with a 50 kN load cell and a clip on extensometer (50 mm gauge length) to measure displacement (strain). The data was gathered using Bluehill2, Instron's propriety software.

## 4 Results and Discussion

### *4.1 Tensile Strength of Resin Systems*

The tension test results for the thermosetting resin systems can be seen in Fig. 1. The two re-formulated

resin systems with the raw materials derived from natural oils (S1 and S2) displayed similar tensile strengths to the standard resin systems whilst decreasing the use of petrochemical based raw materials.

*4.2 Tensile Strength of Natural Fibre Composites*

The tensile strength of composites with the UMa resin system are displayed in Fig. 2, and the associated fibre volume fractions are shown in Table. 1.

The use of the hemp CSM as a reinforcing medium reduced the tensile strength by half compared with the base resin. One possible explanation is that during composite manufacture there were regions that could not be infused, which caused porosity within the composite. This porosity could be causing stress concentrations and so failure at lower stresses. This porosity would also decrease the calculated fibre volume fraction due to buoyancy caused by these voids.

The unidirectional hemp fibre composite achieved a similar tensile strength compared with the E-glass CSM composite, when the NFs were aligned in the loading direction (obviously, being a unidirectional fabric, this peak tensile strength result only occurs when the fibres are aligned to the load direction). This composite also had a lower degree of porosity compared with the hemp CSM composite.

The VR composite achieved approximately double the tensile strength of the unidirectional hemp and E-glass composite. This was to be expected since the VR composite has double the fibre volume fraction compared with the unidirectional hemp composite. This composite achieved the highest tensile strength out of the cellulose based fibres, but this fibre also has the highest processing requirement. In addition, the transverse properties of this VR composite is lower than the E-glass CSM composite.

*4.3 Residual Moisture within Natural Fibres and the Effect on Tensile Strength of Composites*

The results from residual moisture investigation have shown that the two naturally grown fibre fabrics had a moisture content of 8.9 % (hemp CSM) and 9.9 % (hemp UD) (Table 2). Perhaps as a result of its slightly different cellulose structure, the VR composite had a moisture content of 12.6 %.

From drying the unidirectional hemp fibres before composite manufacture, the tensile strength of the composite increased by 25 % with the UP resin system and 60 % with the UMa resin system (Fig. 3). Since the UMa resin is known to be more sensitive to moisture than the UP resin, a more significant improvement in mechanical properties with the UMa composite was to be expected.

These results show that residual moisture within the fibres can have a significant detrimental effect on the mechanical performance of a composite.

*4.4 Fracture Toughness of the Hemp CSM Composite*

The use of the hemp CSM fabric resulted in a fracture toughness value of 2.7 MPa $m^{1/2}$ for the composite. This result represents a factor of six increase compared with the resin system (Table 3). Unfortunately, the NF composite was still a factor of 4.5 lower in fracture toughness compared with the E-glass CSM composite. The fracture surface of the hemp CSM composite and E-glass CSM composites are shown in Fig. 4 and 5 respectively.

Since the fibre volume fraction of the hemp CSM composite was lower by a factor of two than the E-glass CSM composite (Table 1), and the E-glass fibres are longer and of higher stiffness, the results obtained were to be expected. As mentioned previously, there is porosity within the hemp CSM composite which would also reduce the fracture toughness of this material.

The fracture toughness result for the hemp CSM composite shows that even though the tensile strength of the hemp CSM composite is lower, compared with the resin system, it does greatly improve the fracture toughness of the resin system.

## 5 Concluding Remarks

Thermosetting resins, modified with bio-based constituents are seen as a sustainable alternative to standard petrochemical based polymers. The current (and ongoing) research has shown that polymers can be produced that can compete with the mechanical performance of commercially available systems.

This has been achieved through consideration of the formulation of the resin system and consequent incorporation of constituents derived from natural oils at the development stage, rather than treating them as a bio-additive. Hence, the sustainability of commercial resin systems may be improved as the plants, from which the oils are derived, absorb $CO_2$ during their growth. With minimal processing, the raw materials based on natural oils could help offset the $CO_2$ emitted from production of the other raw materials within the resin system leading to a $CO_2$ 'neutral' (or even 'negative') product. The issue with using naturally grown oils is that they could compete with food production, which would incur a negative social impact. Consideration must also be given to the competition with the petrochemical industry to replace both polymers and fuels with 'green' alternatives.

The ongoing research is developing a range of GFRP and NF composites, which utilize more sustainable resin systems. The results presented here show that such composites have the potential to be used in structural applications. One of the issues faced when using NFs is that in order to maximize performance of the composite a high degree of fibre processing is required. This may have a negative impact on the sustainability of the composite. The balance between sustainability and mechanical performance will ultimately govern the selected fibre processing route.

From selecting the correct quality and type of fibre and fabric architecture it could be possible to have NF composites that compete, in some applications, with synthetic fibre composites. It is likely that NF woven architectures are required for NF composites to compete with E-glass CSM composites. As these fibres are naturally grown, care should be taken in selecting and growing the correct fibre in order that fibres do not compete with food crops.

Moisture within a NF can greatly affect mechanical performance of a composite; NFs should be dried before composite manufacture with thermosetting resins to ensure optimum performance. Residual moisture should also be taken into account when comparing fibre treatment methods since the difference in performance between the methods could be down to differences in the residual moisture.

For a green composite to be described as a sustainable alternative it must account for all of the possible environmental, social and economic impacts. The fact that a raw material is bio-derived does not automatically mean that it is a sustainable alternative. However, it is envisaged that sustainable composites that utilize resin based upon natural oils and NFs with some form of treatment will be used in structural and non-structural applications once these issues of confirming sustainability are addressed through life cycle assessment.

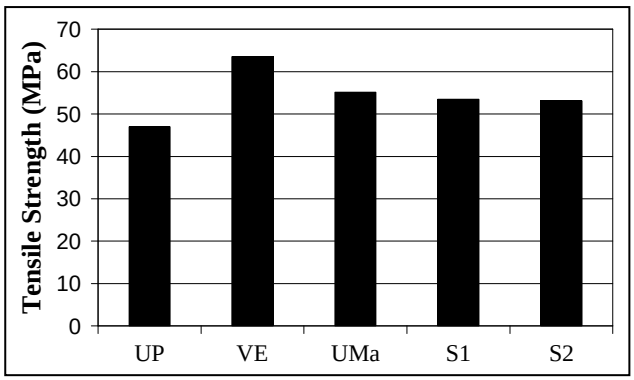


Fig. 1. Tensile strength of thermosetting resins

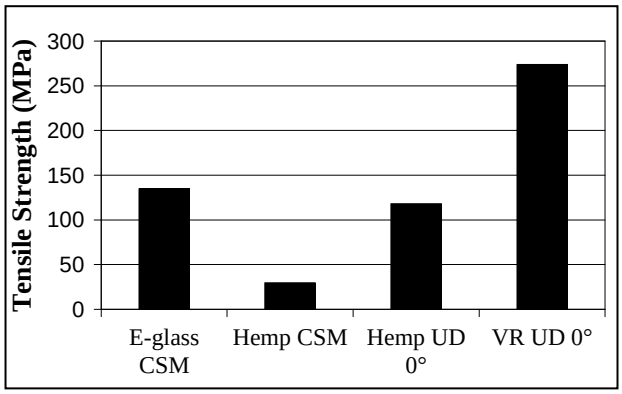


Fig. 2. Tensile strength of UMa/fibre reinforced composites

Table. 1. Fibre volume fractions of tested composites

| Composite Type | Fibre Volume Fraction |
|---|---|
| E-glass CSM | 0.37 ± 0.05 |
| Hemp CSM | 0.16 ± 0.05 |
| Hemp 0° UD | 0.32 ± 0.04 |
| VR 0° UD | 0.65 ± 0.02 |

Table. 2. Fracture toughness of hemp CSM composite, including the base resin and E-glass CSM composite results

| Material Type | Fracture Toughness ($MPa\ m^{1/2}$) |
|---|---|
| UP Resin | 0.41 ± 0.01 |
| Hemp CSM Composite | 2.7 ± 0.1 |
| E-glass CSM Composite | 12.2 ± 0.5 |

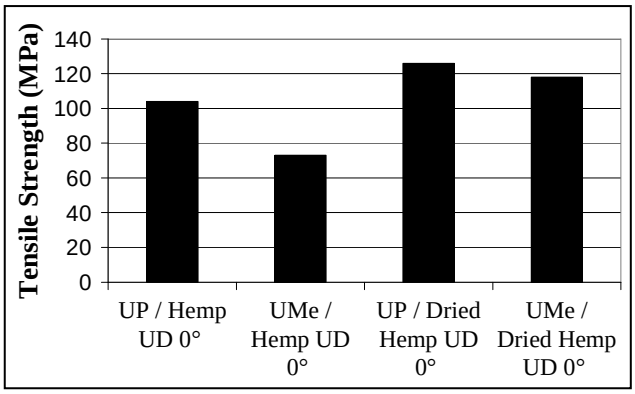


Fig. 3. Tensile strength of standard and dried hemp UD composites

Table. 3. Moisture content within natural fibres

| Natural Fibre Type | Moisture Content (%) |
|---|---|
| Hemp CSM | 8.9 ± 0.1 |
| Hemp UD | 9.9 ± 0.1 |
| Viscose Rayon | 12.6 ± 0.1 |

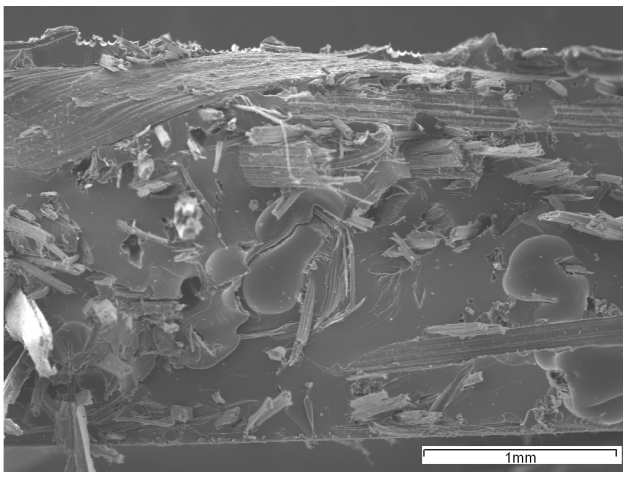


Fig. 4. Fracture surface of a hemp CSM composite

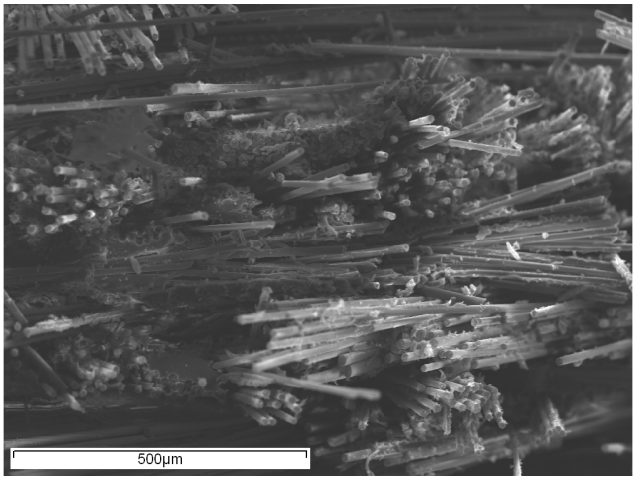


Fig. 5. Fracture surface of an E-glass CSM composite

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