YARN OPTIMISATION AND PLANT FIBRE SURFACE TREATMENT USING HYDROXYETHYLCELLULOSE FOR THE DEVELOPMENT OF STRUCTURAL BIO-BASED COMPOSITES

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1 Introduction

Low-cost renewable natural fibres [1] have, almost exclusively, been used as short fibre randomly distributed reinforcements in non-structural thermoplastic applications [2] (and references therein). Through this study, the potential of biofibres as reinforcements in load-bearing applications is assessed by evaluating the performance of vacuum infused thermoset unidirectional (UD) plant bast fibre composites (PFCs) against E-glass composites (GFCs).

However, the development of structural PFCs requires specific consideration over traditional composites. Firstly, the lack of composites-applicable biofibres is apparent noting that they require specific consideration over textile industry requirements [3]; where textile yarns are twisted for processability, employing twisted yarns as reinforcements hinders impregnation and compromises orientation efficiency of the resulting composite. This study highlights the significance of reinforcement plant fibre yarn construction (twist and compaction) in composite manufacturing (fill time, void content) and mechanical properties.

Secondly, current research trends highlight the importance of interface engineering in the development of PFCs due to their shortcomings associated with poor fibre-matrix adhesion. Although conventional fibre surface modification techniques improve the interface and composite mechanical properties, they i) are an additional step in PFC manufacture, ii) require expensive or toxic chemicals, and iii) reduce the reinforcing fibre tensile strength by up to 50% (if unoptimised) [4]. This study investigates the use of a cheap, commercially applicable, non-toxic, novel fibre surface treatment technique: hydroxyethylcellulose (HEC) sizing of plant fibre yarns. HEC sizing may not only eliminate the need for introduction of twist in yarns for textile processability, it can also act as a film-former, lubricant, surfactant and binder in the production of aligned fabrics. Moreover, hydrophobic modification of HEC can enhance its performance as a surfactant and even as a compatibiliser to create a better fibre-matrix interface.

2 Methodology

2.1 Materials

For this study, four commercially available plant bast fibre ring spun yarns were chosen (Table 1). J250 (jute) and H285 (hemp) yarns were obtained in high twist. F250 is a low twist flax commingled with a polyester binder yarn while F400 is low twist roving of flax. The three numbers denote datasheet (nominal) linear density in tex. The deviation of the true linear density from the nominal linear density is also presented in Table 1. The significant difference for J250 may be attributable to the 7 – 10 % moisture content of plant fibres [5], particularly as J250 is produced in humid Bangladesh. The measured fibre density, mean yarn twist angle and yarn packing fraction are also presented in Table 1.

2.2 Production of plant fibre UD fabric

A simplified drum-winding facility is used to produce UD mats (Fig. 1). The process involves the automatic winding of a yarn (from a single bobbin) around a rotating and traversing metal drum. To minimize inter-yarn spacing, periodic manual adjustments are necessary. Once the drum length is covered, the monolayer winding is uniformly hand painted with 0.6 wt% aqueous HEC solution and dried at 60 °C for 30 mins. The UD mat is recovered upon drying. HEC was purchased from the Dow Chemical Company under the trade name Cellosize HEC QP-52000H.

2.3 Manufacture of UD composites

Four layers of UD mat were used to produce 250 mm square 3-3.5 mm thick composite plaques. Resin injection was achieved by vacuum infusion. To observe the flow regime, the mould tool included a clear acrylic top face. Reduced race tracking and improved impregnation was achieved by line-gate injection with resin flow perpendicular to the UD mat.

For composite manufacture, two standard thermosetting resin systems were used: *i)* a Reichhold Norpol orthophthalic unsaturated polyester (UP) type 420-100 and *ii)* a Gurit UK Ltd. low viscosity Epoxy Prime 20LV. For both resin systems, post cure was carried out at 50 °C for 6 h after ambient curing for 16 h.

UD and random E-glass composites were also manufactured for comparative purposes.

2.4 Experimental

2.4.1 Yarn tensile properties

To observe the effect of HEC treatment on the mechanical properties of the yarns, tensile properties of the untreated and HEC treated yarns were measured with an Instron 5969 testing machine set up with a 2 kN load cell. Single yarns with a gauge length of 250 mm were tested at a cross-head speed of 200 mm/min. Ten specimen were tested for each yarn, as deemed sufficient by several researchers [3, 6, 7]. Yarn tenacity (cN/tex), stiffness (N/tex) and failure strain (%) were determined from the tensile tests.

2.4.2 Composite physical properties

The fibre volume fraction (v_f), matrix volume fraction (v_m) and void volume fraction (v_v) of the manufactured composites were determined using (1), where W and ρ represent mass and density, respectively while the subscripts f, m and c denote fibres, matrix and composite, respectively. Composite density was determined using helium pycnometry. Optical microscopy was used to qualitatively investigate the types of porosity in the composites. Mould fill times during laminate manufacture were also recorded.

$$v_f = \frac{\rho_c}{\rho_f}\frac{W_f}{W_c};\; v_m = \frac{\rho_c}{\rho_f}\left(1-\frac{W_f}{W_c}\right);$$
$$v_v = 1-\left(v_f + v_m\right) \quad (1)$$

2.4.3 Composite mechanical testing

Tensile tests were conducted according to ISO 527-4:1997 using an Instron 5985 testing machine equipped with a 100 kN load cell and an extensometer. Six 250 mm long and 25 mm wide specimens were tested for each type of composite at a cross-head speed of 2 mm/min. The ultimate tensile strength (UTS) σ_T, tensile modulus E_T (in the strain range of 0.025 - 0.10 %) and the failure strain ε_T of the composite samples were measured.

From the composite properties, fibre tensile strength σ_f, and modulus E_f were back-calculated using the rule of mixtures in (2). The reinforcement efficiency factor (η) for UD composites was assumed to be 1. For E-glass random (R) composites, an efficiency factor of 3/8 was used.

$$E_f = \frac{E_T - v_m E_m}{\eta v_f};\qquad \sigma_f = \frac{\sigma_T - v_m \sigma_m}{\eta v_f} \quad (2)$$

Three-point bending flexural tests were performed according to ISO 178:1997 using a Hounsfield testing machine equipped with a three-point bending fixture and 2 kN load cell to determine the flexural strength σ_F and flexural modulus E_F of the composites. Six specimens (80 mm long and 15 mm wide) were tested for each type of composite at a cross-head speed of 2 mm/min.

Finally, the impact properties of the composites were determined using an Avery Denison pendulum Charpy testing machine according to ISO 179:1997. The un-notched specimens were loaded flatwise with weighted hammers at a point perpendicular to the direction of the UD fabric plane; a 2.7 J hammer was used for PFCs while a 5 J hammer was used for E-glass UD composites. A striking velocity of 3.46 ms^{-1} was used. Six specimens (100 mm long and 10 mm wide) were tested for each type of composite.

3 Results

3.1 Effect of HEC treatment on yarn tensile properties

Although statistically insignificant (at α = 0.05), HEC treatment has a detrimental effect on the tenacity and stiffness of high twist yarns (Fig. 2).

The stiffness of J250 and H285 drops by about 24% while the tenacity decreases by up to 20%. However, low twist yarns show significant improvement in tensile properties. The tenacity and stiffness of low twist F400 increases by 230% and 75%, respectively. An increase of 14% is observed in the tenacity of low twist F250. It is noteworthy that the failure strain of all yarns increases; the increase is in the range of 25 - 125 %. Hence, the significant increase in tenacity, stiffness and failure strain of low twist yarns upon HEC treatment leads to an appreciable increase in yarn toughness. The observed effect is that upon HEC treatment low twist yarns also fail by fibre breakage rather than fibre slippage, as was the case prior to treatment.

The positive effect of HEC treatment on low twist yarns but negative effect on high twist yarns is due to differences in yarn permeability. Measuring the amount of HEC deposited onto the yarn, it was observed that low twist yarns were more responsive to HEC treatment due to lower compaction, easier impregnation and larger exposed fibre surface area. SEM micrographs confirmed extensive HEC deposition onto fibres of low twist yarns due to increased hydrogen bonding opportunities with surface cellulose.

It is thought that HEC treatment also affects the load-bearing situation; in low twist yarns, HEC treatment introduces intra-yarn (inter-fibre) cohesion and generates a hydrogen bonding network. Not only is the force required to overcome this inter-fibre cohesion higher than before, but the transverse bonding of fibres also implies fibres are strained more uniformly when loaded. The load bearing situation is more favourable now leading to an increase in strength and stiffness upon treatment. In high twist yarns, however, HEC cannot impregnate the fibres in the interior of the yarn easily due to higher packing fractions towards the centre [8]. As the fibres on the surface are bonded together and the interior is not, fibres are strained more non-uniformly. The load-bearing situation is now worse. Hence strength and stiffness deteriorate.

HEC treatment basically plays the role of twist in textile engineering; it allows binding of fibres in the staple yarn, increases yarn strength by increasing inter-fibre friction and yet there is no loss in fibre strength from fibre obliquity. This is ideal in the production of composites.

3.3 Composite physical properties

The produced PFCs have fibre volume fractions ranging from 27 - 36 %; GFCs have higher fibre content of 43%. Higher volume fraction PFCs can be manufactured however an upper limit is set by yarn/fabric compaction and packing at about 50 - 60 % [9].

PFCs are observed to be 40 - 50% less dense than GFCs due to the lower density of plant fibres (1.4-1.6 gcm^{-3}) compared to E-glass (2.6 gcm^{-3}). The void content of PFCs is in the range of 0.5 - 4.1 % and is comparable to that found in E-glass UD composites (1 - 3 %); E-glass R composites are observed to have high void fractions of up to 8%.

This study reveals interesting effects of reinforcement yarn twist on composite fill times and void content. Fig 3. shows the relationship between mean yarn twist angle, fill time of PFCs and void content of PFCs.

Although high twist PFCs (J250 and H285) have low fill times which is comparable to GFCs, low twist PFCs (F250 and F400) have significantly higher fill times (Fig. 3). This is because resin flow in high twist PFCs is dominated by inter-yarn (macroscale) flow whereas resin flow in low twist PFCs is dominated by intra-yarn (microscale) flow. In intra-yarn flow, viscous forces are much larger than inertial forces while the opposite is true for inter-yarn flow [10]. Noting the initially established positive correlation between yarn twist and yarn compaction, in high twist tightly compacted yarn PFCs, inter-yarn infusion is dominating and thus resin flow is rapid. Here, intra-yarn spaces are much smaller compared to inter-yarn spaces. In low twist, low compaction yarn PFCs, intra-yarn infusion becomes particularly important and thus resin flow is relatively slow. In these, inter-yarn spaces are comparable to intra-yarn spaces. The difference in length scales of inter-yarn and intra-yarn spacing does not only affect the flow regime, they also affect the homogeneity of the composites. Fibre distribution in low twist PFCs is more homogenous than in high twist PFCs; this would affect stress distribution within the composite upon loading.

Additionally, low twist PFCs are observed to contain fewer voids. In high twist PFCs, resin impregnation is hindered, particularly as yarn compaction increases towards the centre of ring spun yarns [8].

The hindered impregnability and compromised wettability is perceivable in the form of voids.

A qualitative analysis of the type of fibre-related voids [9] concludes that high twist PFCs are troubled with impregnation voids (Fig. 4), where cavities exist between fibre/yarn bundles. This is in agreement with earlier conclusions regarding hindered impregnability from high twist yarns. Low twist PFCs, on the other hand, have a higher content of interface voids, where cavities exist at the fibre/matrix interface (Fig. 4). It should be noted, however, that high twist PFCs also contain interface porosities as interface porosities are related to the fibre-matrix compatibility and not the greatly to the yarn structure. It is just that low twist flax yarns are highly defibrillated (Fig 4.) and thus a greater fibre perimeter is bonding with the matrix resulting in a higher relative amount of interface voids.

3.3 Composite mechanical properties

Specific properties of PFCs and plant fibres relative to E-glass fibre (UD) composites are presented in Fig 5. PFCs have similar strain at UTS compared to GFCs. E-glass UD composites have a UTS in excess of 700 MPa, which is considerably higher than the UTS of UD PFCs, which ranges between 140 - 285 MPa. An advantage of natural fibres over glass fibres for reinforcement is their low density, however even the specific strength of PFCs is only 30 - 50 % that of E-glass UD composites. In terms of stiffness, PFCs perform better. Low-twist F400 composites have a stiffness of 23.4 - 24.6 GPa, while other PFCs have appreciably lower stiffness ranging between 14.2 - 19.0 GPa. The tensile stiffness of F400 PFCs is more comparable to GFCs (34.0 - 36.9 GPa) while the specific stiffness of F400 notably matches that of E-glass UD composites.

Back-calculated fibre properties have also been determined; these are useful as firstly they allow comparison of composite properties for the same fibre content (that is, $v_f = 1$) and secondly they indicate the apparent potential of the reinforcing fibres. The UTS and stiffness of E-glass is in the range of 1650 - 1900 MPa and 75.6 - 81.6 GPa, respectively and matches literature values [1]. Low-twist F400 flax shows a UTS of 898 - 929 MPa and stiffness of 68.8 - 75.1 GPa. Based on these results, F400 flax fibre can effortlessly outperform E-glass in terms of specific stiffness (by 40 - 60 %) and compete with E-glass in terms of specific strength.

The flexural performance mimics the tensile performance of the composites (Fig. 6). E-glass UD composites show the highest strength and stiffness. The specific stiffness of F400 is comparable to that of E-glass UD composites while its specific strength is about half that of E-glass UD composites. Nonetheless, the performance of F400 is best amongst the PFCs. F250, H285 and J250 display similar properties that are marginally better than that of E-glass R in terms of specific strength.

The impact properties of PFCs compare poorly to E-glass UD composites and are lower than the impact strength of E-glass R composites, even when compared in terms of specific strength. Where E-glass UD composites have an impact strength of 300 - 350 kJ/m^2, PFCs have impact strengths five to ten times less at 30 - 60 kJ/m^2. However, the inferior impact properties of PFCs can be used as indicators of possible applications or can be improved by using woven fabrics or even hybrid fabrics. Noticeably, epoxy composites exhibit lower impact strength than UP composites. This indicates better compatibility of plant fibres with epoxy resin and less fibre pull-out from the matrix, as fibre pull-out dissipates more energy compared to fibre fracture [11].

4 Conclusions

The research conducted highlights yarn twist and compaction as yarn physical properties that dictate the role of textile plant fibre yarns as reinforcements in composites. High twist PFCs require shorter fill times with resin flow dictated by inter-yarn flow. Higher void content is also observed with impregnation voids being the primary form of voids. Low twist PFCs require significantly longer fill times and the composites are more homogenous with fewer voids.

It is also observed that low twist PFCs can produce high quality composites that can compete and even outperform GFCs in terms of specific stiffness, without any fibre surface modification. However, PFCs still display poor strength performance (tensile, flexural and impact).

The use of HEC sizing offers the potential to replace 'twist' in textile yarns, which is traditionally used to increase the dry strength and processability of yarns. HEC sizing can increase the tenacity and stiffness of low twist yarns by 230% and 75%, respectively, without reducing composite orientation efficiency or yarn impregnability. The capability of producing UD fabrics in a semi-automatic manner whilst incorporating HEC sizing treatment is seen as a positive step forward towards creating structural PFCs.

Investigations on the use of hydrophobically modified HEC (HMHEC) to improve the surface properties of plant fibre yarns and induce better compatibility between the fibre and matrix will follow this present research.

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Table 1: List of plant fibre material and their properties

ID	Yarn	Supplier	Density [gcm^{-3}]	True linear density [tex]	Mean twist angle [°]	Packing fraction
J250	**Jute**	Janata Ltd. (Bangladesh)	1.4333 ± 0.0047	206 ± 21	14.0 ± 4.2	0.596
H285	**Hemp**	Safilin (Poland)	1.5313 ± 0.0025	278 ± 17	13.2 ± 3.0	0.591
F250	**Flax**	NetComposites (UK)	1.5288 ± 0.0034	229 ± 22	3.3 ± 2.5	0.421
F400	**Flax**	Safilin (Poland)	1.5742 ± 0.0036	396 ± 16	0.3 ± 0.1	-

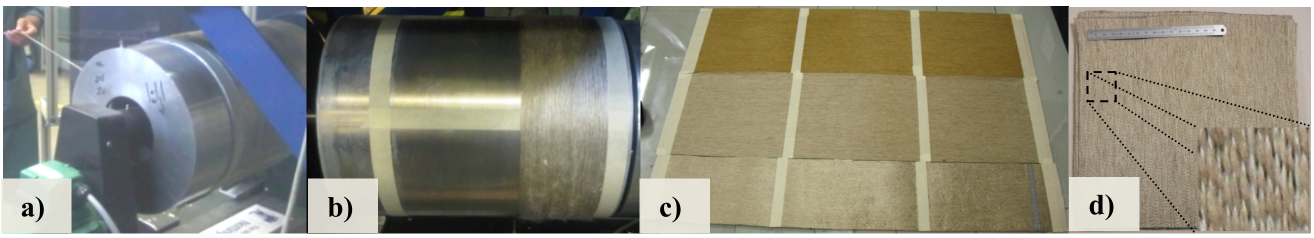

Fig. 1: Developed UD mat fabrication process involving a) winding of yarn around drum, b) periodic manual adjustments to minimise inter-yarn spacing, c) and d) sizing with HEC and drying to produce mat.

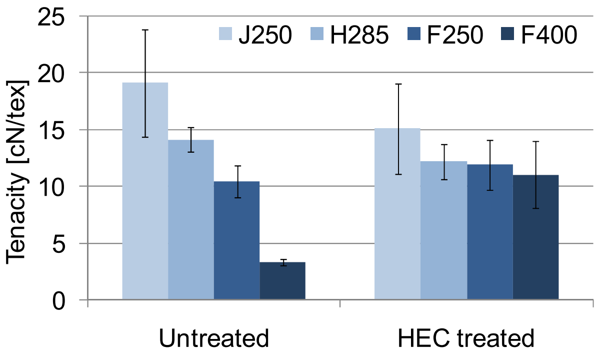

Fig. 2: Effect of HEC treatment on yarn tenacity

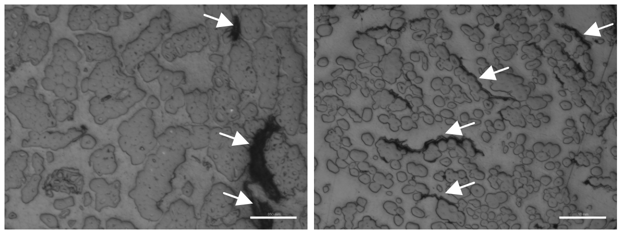

Fig. 4: Optical micrographs of J250 (left, scale bar is 50 µm) and F400 (right, scale bar is 100 µm) epoxy composites. High twist J250 composites have impregnation voids while low twist F400 composites have interface voids (indicated by arrows).

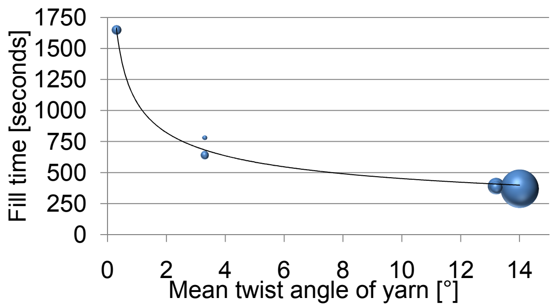

Fig. 3: Composite fill time as a function of mean yarn twist angle. Bubble size represents relative composite void content. Low twist PFCs require longer fill times but have fewer voids.

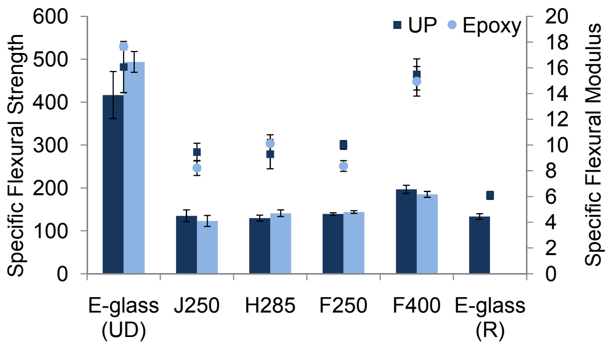

Fig. 6: Specific flexural strength [MPa·cm^3/g] (Bars) and specific flexural modulus [GPa·cm^3/g] (Points) of PFCs compared to GFCs. Polyester composites represented by the darker shade.

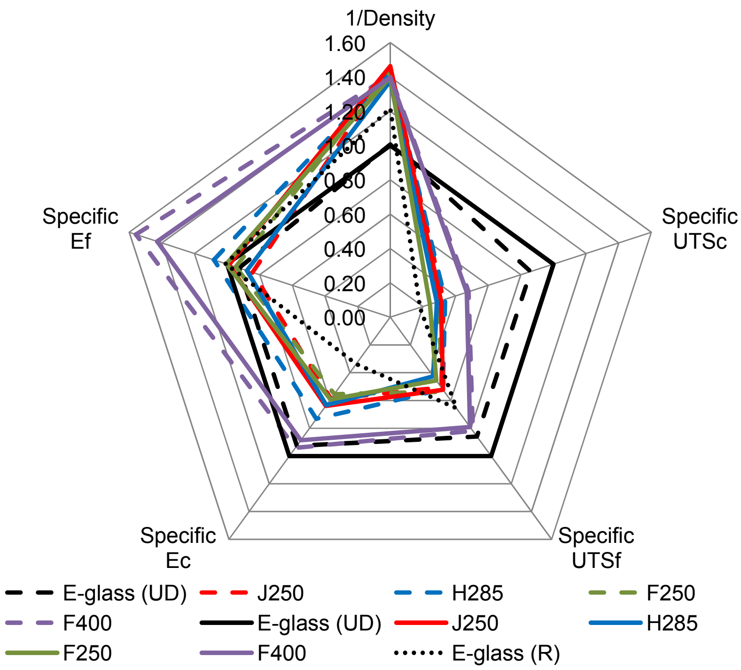

Fig. 5: Specific tensile properties of composites (measured) and fibres (back-calculated) relative to E-glass (UD) polyester. Dashed line represents epoxy composites and continuous line represents polyester composites.