Supporting Information

Ultrahigh Performance of Nanoengineered Graphene-Based Natural Jute Fiber Composites

Forkan Sarker,[1,2] Prasad Potluri,[1,2]* Shaila Afroj,[3,4] Vivek Koncherry,[1,2]

Kostya S Novoselov,[3,4] and Nazmul Karim[4]*

[1]School of Materials, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK

[2]Northwest Composites Centre, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK

[3]School of Physics & Astronomy, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK

[4]National Graphene Institute (NGI), The University of Manchester, Booth Street East, Manchester, M13 9PL, UK

*Corresponding E-mail: mdnazmul.karim@manchester.ac.uk and prasad.potluri@manchester.ac.uk;

Supporting Information1: Design and Manufacturing of Jute Fibre Composites

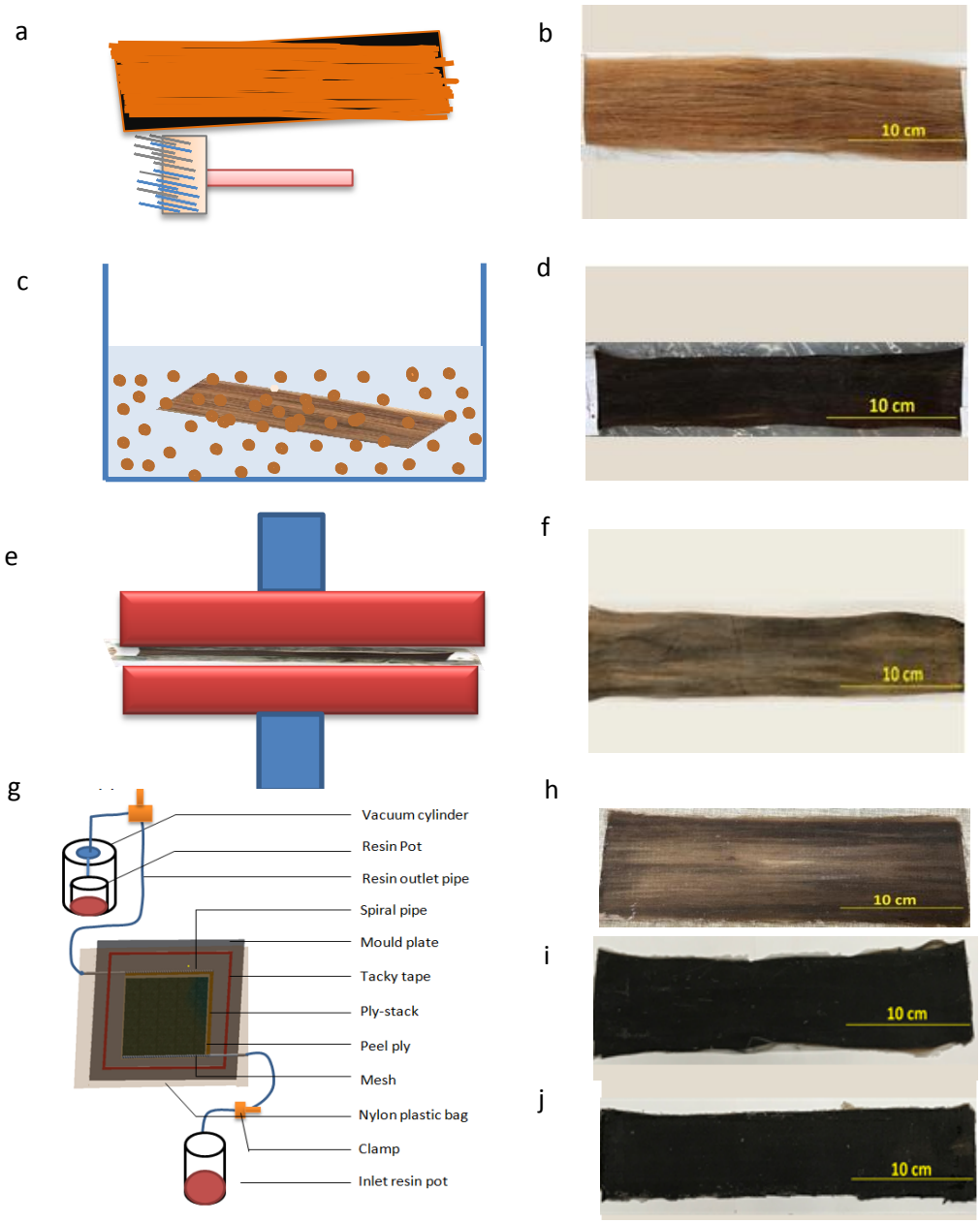

Figure S1. Design, manufacturing of jute fibre preform and its composites: (a) Hand combing of jute sliver; (b) fibrillated jute fibre; (c) Graphene coating on jute fibre; (d) Unidirectional preform after graphene coating; (e) hydraulic press to pack the preform; (f) pressed UD preform; (g) vacuum infusion process for making composites; (h) untreated UD; (i) GO coated; and (j) G flakes coated jute fibre/epoxy composites.

Supporting Information 2: Preparation of Jute and Glass Fibre Preform

Figure S2 shows the fibrillated preform obtained before and after hand combing. Figure S2a shows the sliver bundles cut into 300 mm in size to process. After combing we arranged the fibrillated fibres in unidirectional so that the fibres are parallel to each other (Figure S2b).

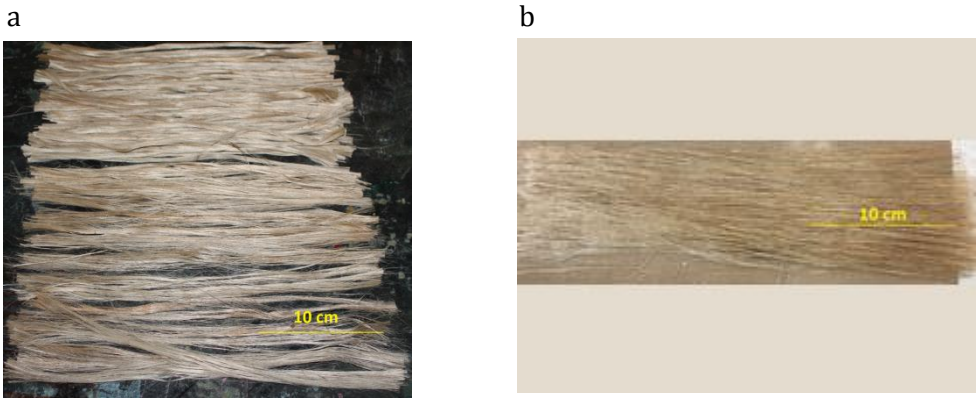

Figure S2. Digital image of new fibre architecture preform with no pressing: (a) without combing; and (b) with hand combing

In order to coat the jute fibre preform with graphene oxides, we first collected the fibrillated fibre. We then sealed the edge of fibre with both sided tape so that fibre alignment never change after the coating (Figure S3a). Following this, we slowly dipped the preform into the coating solution bath and left for 30 mins time, allowing enough time so that the fibres are homogenously coated. Fibres remained into the coating solutions can be found in the (Figure S3b).

a b

Figure S3. Dip coating of unidirectional jute fibre preform: (a) preform before dipping in the coating bath, and (b) preform dipped in the coating bath.

a b

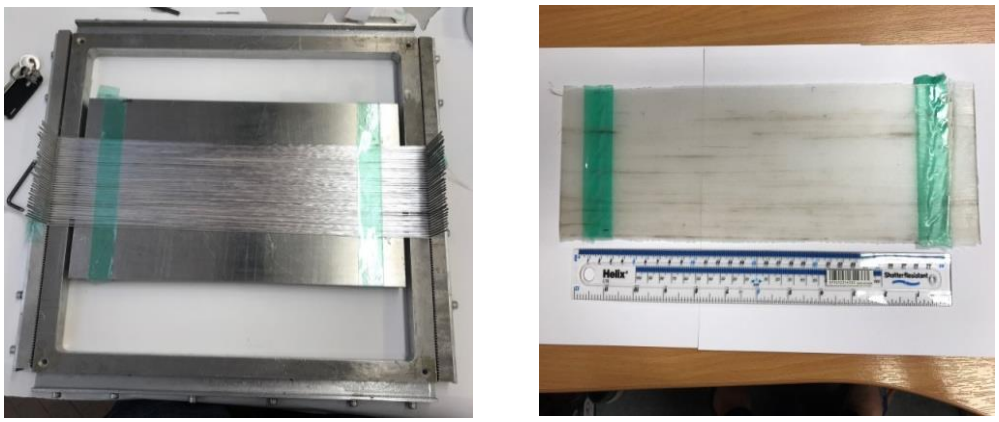

Figure S4. Manufacturing of glass fibre preform: (a) straight segment of glass fibre in the pin board; and (b) digital image of vacuum infused glass fibre composites.

Figure S4 shows the digital image of manufacturing glass fibre preform and its composites. We placed the glass fibre roving parallel to each other with the help of pinboard to obtain UD glass fibre preform. Flash tape was used to hold the fibre together as shown in the Figure S4a. We then carefully collected the UD preform and made the composites (Figure S4b) by vacuum infusion process.

Supporting Information 3: Single Fibre Properties

Table S1 shows the comparison on tensile properties of untreated single jute fibre (UT) with bundle of jute fibres(BF). Untreated fibre has a diameter of ~55µm whereas bundle of fibres shows comparatively higher diameter of the fibre which is ~180µm. From the Table S1 we also observe that tensile properties of bundle of jute fibre are 55% less of Young's modulus and ~39% less of tensile strength than the untreated elementary fibre which is an agreement of previous studies on flax fibre.[1]

Table S1. Tensile properties of single jute fibre

Sample	**Fibre diameter**	**Young's modulus**			**Tensile strength**			**Force at break**	**Extension at break**
	D	E_f(GPa)	β	α	σ_f(MPa)	β	α	F(N)	*E* (%)
BF	*180(25)*	*13.5*	*1.62*	*14*	*180*	*1.55*	*192*	4.25 N	*0.75(0.31)*
UT	55(±12)	30(±19)	1.86	35	295(±188)	1.74	333	0.68(±0.34)	1.0(±0.40)

Supporting Information 4: Microscopy and Density of the Composites

Figure S5 shows SEM images of untreated and treated jute fibre in single fibre and bundles form. For untreated fibre we observe elementary fibres are attached in the technical fibre (Figure S5a). Further to alkali treatment elementary fibres separate from the technical fibre (Figure S5b). The micro-voids present in the alkali treated jute fibre due to the partial removal of hemicellulose, which was present in the inter-fibrillar region of jute fibres. These Graphene flakes can fill those microvoids in the elementary fibre when fibrillated jute fibre is coated by graphene oxides (Figure S5c). After jute fibre is coated with graphene flakes we observe flakes are mechanically interlocked and uniformly covered in the fibre surface.

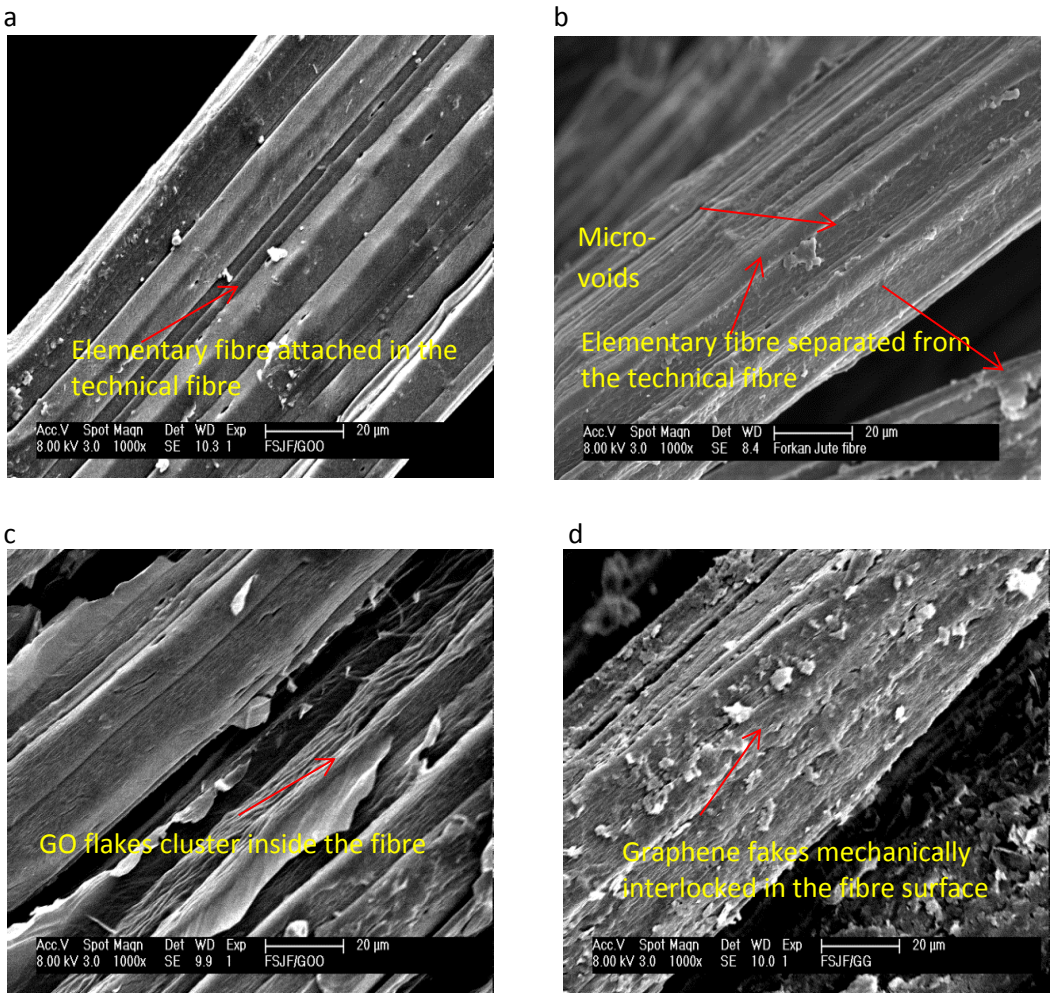

Figure S5. Scanning electron microscopic images of fibre bundles: (a) untreated jute fibre (1000X); (b) alkali (HA) treated jute fibre (1000X); (c) GO coated jute fibre (1000X); (d) G flakes coated jute fibre (1000X).

Optical microscopy was used to qualitatively measure the image of fibre packing arrangement and porosity of the composites. For doing this, three sections from each of the composites were cast by using epoxy resin (resin to hardener ratio 100:10 by mass) and cured for 48 hrs. The samples were ground (using 240, 400, 600, 800, 1200 grit paper) and polished by using diamond grit paper of 6µ and followed by 1µ the samples were polished. Finally, the polished samples were viewed under a microscope (Keyence digital microscope VHX-500F, UK) with 500x magnification. Images were processed using ImageJ software.

Supporting Information 5: Tensile Test of the Composites

Figures S6 shows the image of machine set up for conducting tensile test of the composites. We conduct tensile test of the composites based on ASTM-D3039 standard. We use specimen protector in order to protect the specimen from damage. We use pressure bar to grip the specimen and a video camera to measure the strain of the composites.

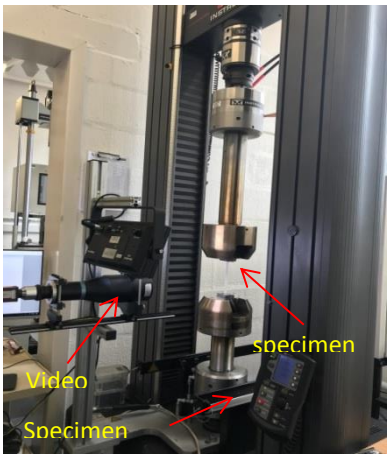

Figure S6. Machine sept up for measuring tensile test of the composites.

Table S2 provide the results of tensile test both in longitudinal and transverse direction. It shows that a mechanical property of the jute fibre/epoxy is largely varied on the type of treatment and also concentrations of graphene in the fibre treatment. Density of jute fibre composites is almost 73% less than the s-glass composites which offer the opportunity to compare the specific properties of natural fibre composite and glass fibre composites. the modulus of graphene coated composites is almost equal of high performance glass fibre composites.

Table S2. Mechanical properties of untreated, graphene oxides and G flakes treated jute fibre /epoxy composites (standard deviation).

Sample code	**Volume fractions**	**Composite density**	**Young's modulus GPa**		**Tensile strength MPa**		**Strain %**	
			Longitudinal	***transverse***	***Longitudinal***	***transverse***	***Longitudinal***	***transverse***
UT	*0.24*	*1.23 (0.09)*	*10.5 (2.8)*	*0.45 (0.08*	*180(13)*	*2.05 (0.2)*	*0.80(0.05)*	*0.85(0.1)*
FC	*0.37*	*1.24(0.10)*	*20.5(3.5)*	*0.48(0.06)*	*202(17)*	*2.1(0.15)*	*0.8(0.03)*	*0.9(0.12)*
FCP	0.48	1.23(0.18)	27.6 (2.2)	0.46(0.07)	232 (18.9)	2.4(0.6)	0.79 (0.06)	0.76(0.15)
HT	50	1.25(0.15)	26.5(3.45)	0.58(0.06)	230(19.7)	4.5(1.6)	0.85(0.09)	1.2(0.2)
HA 0.5	0.54	*1.25(0.12)*	30.6 (3.4)	0.54(0.12)	282 (15.4)	5.28(2.2)	0.86 (0.09)	1.5(0.33)
GO 0.25	0.54	1.30(0.09)	36.9 (± 4.4)	0.76(± 0.14)	295 (±33.9)	6.64(± 2.2)	0.84 (± 0.07)	1.44(± 0.42)
GO 0.50	0.54	1.3(0.11)	42.8(±1.7)	0.80(±0.11)	337 (±53.3)	7.28(±2.5)	0.94(± 0.10)	1.52(±0.49)
GO 0.75	0.56	1.31(0.07)	44.6 (± 3.6)	0.78(±0.21)	379 (±(33)	9.62(±3.01)	0.93(± 0.05)	2.20(±0.25)
GO 1.0	0.56	1.32(0.08)	37.8 (± 1.6)	0.93(±0.11)	292.7 (±8.4)	15.26(±2.5)	0.78 (± 0.01)	2.36(±0.5)
G 1.0	0.55	1.32(0.14)	35.8 ±(2.2)	0.81 ±(0.15)	290 ± (25)	6.5 ± (1.5)	0.84± (0.08)	1.52±(0.3)
G 10.0	0.55	1.32(0.16)	38.1 (± 4.0)	0.82(±0.05)	294 ± (21)	5.43(±1.75)	0.80 (± 0.07)	1.31(±0.46)
Flax-UD[3]	0.55	-	26(1)	1.2 (0.2)	284(8)	21(4)	1.2(0.2)	-
E-glass	0.55	2.25(0.24)	33.5(3.89)	3.5	777(7)	35.6(1.5)	2.59(0.1)	2.2(0.2)
S-glass	0.56	2.23(0.18)	45.1(4.25)	-	1189(35)	-	2.89	-

[3] –means value taken from the literature and – means value not given

Figure S7 provides the specimen used in the longitudinal tensile test of jute fibre/epoxy composites. After the tensile test G0.75 coated composites (Figure S7) shows fibre splitting instead of catastrophic failure due to the strong interface between the GO coated jute fibre and epoxy matrix. On the other hand uncoated (UT) specimen have catastrophic failure mode in the tensile test (Figure S7).

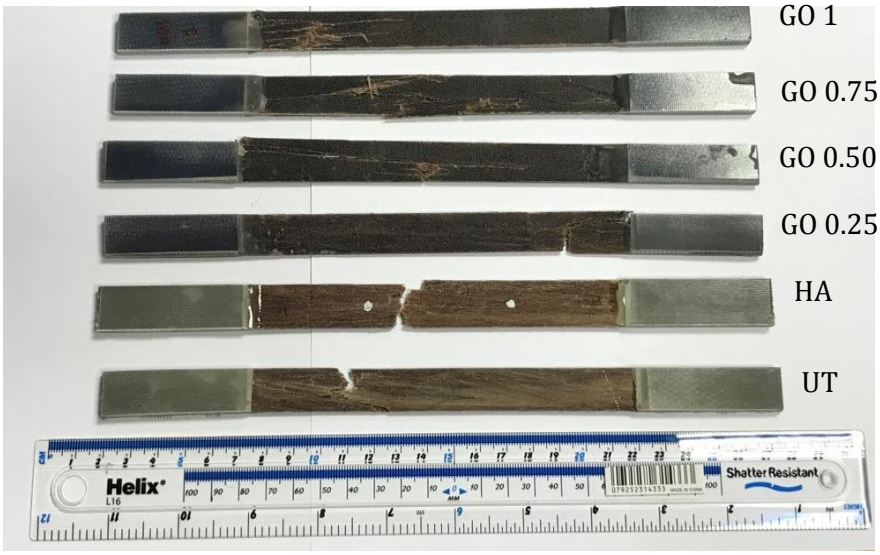

Figure S7. Fracture surface of coated and uncoated specimen after the tensile test.

Figure S8 (a, b) presents the SEM fracture surface of alkali treated jute fibre composites after the longitudinal and transverse tensile test. Figure S8a reveals that alkali treatment reduces the amount of fibre pull out by promoting interfacial shear strength in the composites. We find a linear rate of fibre breakage in the composites, which is an evidence of increasing the mechanical properties of the composites. On the other hand Figure S8b shows the fracture surface of alkali treated transverse tensile test specimen. The grooved appearance in the image indicates the improvement of interface after alkali treatment of jute fibres.

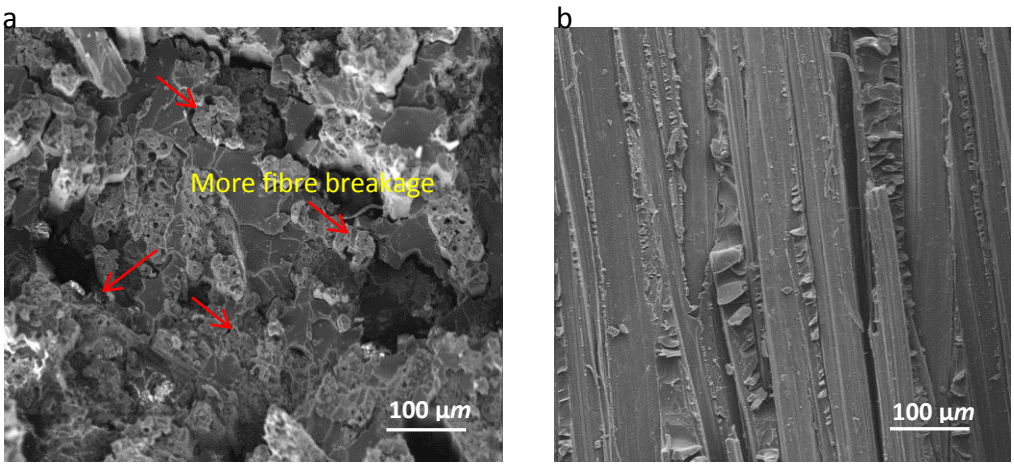

Figure S8. Fracture surface of alkali treated composites: (a) longitudinal tensile test (250X) and (b) transverse tensile test (250X)

Supporting Information 6: FTIR Analysis

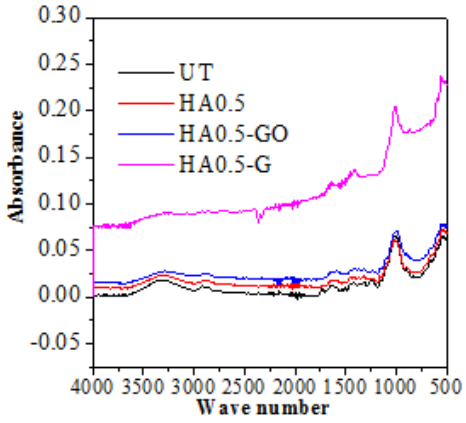

Figure S9. FTIR spectroscopic results of treated and untreated jute fibre.

Figure S9 shows FTIR transmittance spectra for the untreated and the alkali-treated jute fibre bundles. The obtained FTIR spectra for the jute fibres treated with NaOH solution at 0.5-1% are similar to those reported in the literature for cellulose fibres. Cellulose peaks at 3200-3600 cm^{-1} are prominent for the untreated fibres, and are caused by the O-H stretching of the hydrogen bonding network. This peak intensity reduces upon alkali treatment, as is visible in

Figure S9. This reduction is due to the breaking of a hydrogen bond between the O-H groups of cellulose and hemicellulose molecules. Peaks at 1742 and 1242 cm^{-1} are assigned to C=O and C-O stretching modes respectively and are present in the graphs of the UT and HA fibres. The peak at 1742 cm^{-1} is the characteristic peak for the carbonyl stretch of carboxylic groups in hemicellulose and pectin. The peak at 1242 cm^{-1} corresponds to C-O stretching in the acetyl groups in hemicellulose. Following alkali treatment of the fibres, HA0.5 do not provide either of the two peaks. The disappearance of these two peaks after alkali treatment indicates that either the carboxylic acid and acetyl groups were removed or dissolved by the alkali solutions.

References

(1) Coroller, G.; Lefeuvre, A.; Le Duigou, A.; Bourmaud, A.; Ausias, G.; Gaudry, T.; Baley, C. Effect of Flax Fibres Individualisation on Tensile Failure of Flax/epoxy Unidirectional Composite. *Compos. Part A Appl. Sci. Manuf.* **2013**, *51*, 62–70.

(2) Orue, A.; Jauregi, A.; Unsuain, U.; Labidi, J.; Eceiza, A.; Arbelaiz, A. The Effect of Alkaline and Silane Treatments on Mechanical Properties and Breakage of Sisal Fibers and Poly(lactic Acid)/sisal Fiber Composites. *Compos. Part A Appl. Sci. Manuf.* **2016**, *84*, 186–195.

(3) Van de Weyenberg, I.; Chi Truong, T.; Vangrimde, B.; Verpoest, I. Improving the Properties of UD Flax Fibre Reinforced Composites by Applying an Alkaline Fibre Treatment. *Compos. Part A Appl. Sci. Manuf.* **2006**, *37* (9), 1368–1376.