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Polylactic acid (PLA) biocomposites reinforced with coir fibres: Evaluation of mechanical performance and multifunctional properties

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Abstract

The effects of fibre content (5-30 wt%) and fibre treatment on surface morphology, tensile, flexural, thermal and biodegradable properties of polylactic acid (PLA)/coir fibre biocomposites were evaluated via scanning electron microscopy (SEM), mechanical testing, differential scanning calorimetry (DSC), thermogravimetric analysis (TGA) and soil burial method. Similar decreasing trends were found for tensile and flexural strengths with higher strength values obtained for PLA/treated coir fibre biocomposites. 20 wt% treated coir fibres were determined to achieve optimum tensile and flexural strengths of biocomposites. Regardless of fibre treatment, the thermal stability of biocomposites is worsened with increasing the fibre content. The decreased cold crystallisation temperatures of biocomposites further confirms the effective nucleating agent role of coir fibres. The biocomposites undergo much faster degradation than PLA, with the maximum weight loss of 34.9% in treated fibre biocomposites relative to 18% in PLA after 18-day burial, arising from the hydrophilic nature of coir fibres.

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1. Introduction

The great potential for using natural fibre reinforced biocomposites lies in their advanced eco-friendliness and wide material applications for automotive, mobile phone and biomedical industries. Natural fibres such as jute, sisal, coconut coirs, hemps, as well as banana and pineapple leaves are generally extracted from renewable sources, fully biodegradable, non-toxic, and can be easily recycled to reduce the materials' carbon footprint. In general, the pre-treatment of natural fibres using typical alkali treatment [1, 2], peroxide treatment [3] or permanganate treatment [4] methods is essential to chemically modify the fibre surface for the improvements of surface roughness and the interfacial adhesion to matrices [5].

Coconut coirs draw great attention due to their low thermal conductivity and bulk density to yield cost-effective and light-weight composite products [6]. Coir is a versatile lignocellulosic fibre extracted from coconut husk with hemicellulose and lignin as fibre bonding materials [7]. Coir fibres have lower cellulose (36-43%) and hemicellulose (0.2%) contents, higher lignin content (41-45%) and microfibrillar angle (30-45°) than other natural fibres [7, 8], which results in their relatively low tensile strength and modulus [7] as well as highest elongation at break among typical natural fibres [9]. Notwithstanding the traditional use of coir fibres as yarn, rope, flooring-furnishing materials or mats [10], more new applications concentrate on acoustic and thermal insulation [11], automobiles for seat cushions and constructions [12, 13]. On the other hand, poly (lactic acid) (PLA), as a widely used commercial biodegradable polymer, is a good material candidate to replace commodity petro-based plastics with the increasing global demand for the agricultural and environmentally sustainable bioproducts. PLA belongs to one of thermoplastic aliphatic polyesters produced from natural resources like corn, rice and sugar beets, etc. with good

stiffness, high strength and low elongation at break [7]. The widespread applications for such biopolymer lie in medical surgery and tissue engineering [14, 15], automotive components [16], food packaging, degradable plastic bags in supermarkets and so forth [17]. As far as the environment and energy are concerned, the development of coir fibre reinforced biocomposites can offer the alternative to the synthetic fibre reinforced counterparts [18, 19]. Furthermore, numerous benefits can be achieved for such biocomposites in terms of enhanced mechanical and thermal properties [20, 21], excellent biodegradability [22] and the net cost reduction resulting from replacement of biopolymers by much cheaper natural fibres. In particular, the use of PLA/coir fibre biocomposites may ease the coir disposal due to their serious environmental problem in landfills, reduce the weight and cost of car interior parts manufactured in automotive industry, and substitute for that of current wood and synthetic materials [23, 24]. Furthermore, such biocomposites may also be applicable to industrial areas where medium to low strengths for reasonable mechanical properties are required. For instance, plant clips used in the agricultural field can benefit from their good biodegradable nature to allow for the easy disposal of clips after finishing the service life.

Extensive research literature has covered the studies of green composites or biocomposites reinforced with coir fibres. Lai et al. [25] investigated the mechanical and electrical properties of polypropylene (PP)/coir fibre composites and found that superior mechanical properties of composites were often achieved for modified coir fibre surfaces as a result of improved adhesion and enhanced polar interactions at the fibre-matrix interfaces. In addition, dielectric properties were influenced by chemical treatment on coir fibres and their values increased with increasing the fibre content. Rosa et al. [10] characterised the mechanical, thermal and structural properties of biocomposites based on starch/ethylene vinyl alcohol (EVOH)/glycerol blends and coir fibres in relation to the fibre content. The increase of fibre content resulted in decreased tensile strengths but significantly improved tensile moduli.

Additionally, the fibre content slightly influenced the thermal stability of biocomposites without major changes in their crystallinity and biodegradability. Asasutjarit et al. [6] reported the enhanced mechanical properties of cement/coir fibre composites in terms of modulus of rupture and internal bond owing to the chemical composition modification and surface modification. The special pre-treatments of washing and boiling coir fibres were detected to change the fibre morphology by creating voids for greater moisture adsorption capacity and more reactivity to aqueous chemical reagents, as well as producing fibre fibrillation. Thus, a better matrix-fibre adhesion could be achieved for better mechanical properties. In the study of low-density polyethylene (LDPE)/coir fibre composites, Brahmakumar et al. [26] demonstrated that the natural waxy surface layer of coir fibres provided a strong interfacial bonding between fibres and LDPE matrices. The removal of such layers led to drastic decrease of fibre pull-out stress, increase of critical fibre length by 100%, and decreases of composite tensile modulus and strength by 60% and 40%, respectively. Jang et al. [27] evaluated the thermal stability and flammability of PLA/coir fibre green composites. The use of plasma treatment offered enhancements of mechanical and thermophysical properties of composites in addition to their decreased shrinkage with increasing the fibre content.

The present study comprises the investigation of effects of fibre content and fibre pre-treatment on mechanical, thermal and biodegradable properties of PLA/coir fibre biocomposites fabricated by compression moulding process, along with a simple composite theoretical modelling to predict their mechanical performance. To our current knowledge, such holistic study has not been comprehended very well especially when focusing on the fully biodegradable PLA/coir fibre composites of which only mechanical properties [11] or mechanical and thermal properties [7, 27] were experimentally researched on the individual basis. It is expected that this work, as the essential guidance, is able to shed light on the

manufacture and characterisation of any potentially commercial products from PLA/coir fibre biocomposites in a more sophisticated manner.

2. Experimental procedure

2.1. Materials and fibre treatment

PLA 3051D pellets with the material density of 1.25 g/cm^3 [28], and as-received coconut coir fibres with the density of 1.15 g/cm^3 [29] and fibre diameter in range of 150-250 μm were purchased from Natureworks LLC, USA and Supercheap Hardware, NSW, Australia, respectively. In biodegradability tests, Hortico compost, obtained from Bunnings Warehouse, Perth, Australia, was used, which normally acts as an ideal soil-quality enhancer to preserve nutrients and water. The alkali solution was prepared by dissolving 2 g NaOH in 100 ml distill water with magnetic stirring. This process led to a 2 wt% homogeneous NaOH solution as suggested by Nayak et al. [1] to achieve the optimum ultimate tensile strength (UTS) and Gu [30], from material economical viewpoint, for better composite strength. As-received coir fibres were chopped in the fibre length of about 6-8 mm, and then were washed by distilled water. Subsequently the prepared fibres were immersed in such a NaOH solution for 1 h at 70°C prior to being rewashed to remove the remaining NaOH. The compression moulding step was not initiated until collected fibres were dried in the convection oven for 24 h at 60°C.

2.2. Fabrication of biocomposite laminates

To remove any moisture, PLA pellets were dried in a vacuum oven for 10 h at 80°C. These polymeric materials were laid out on a 270 ×180 mm^2 aluminium tray, and were heated in the oven for 15 mins at 180°C. After the material removal from the oven, the top of melted PLA was covered with a similar aluminium plate. They were together transferred rapidly to a hydraulic press, which was followed by an applied pressure at 1.0 MPa for the holding time of 15 mins at room temperature. The press was then released, and 5 kg mass bar was placed

on the top of the plate to allow for the material solidification during 30-min cooling time. Afterwards pre-dried coir fibres were evenly distributed between two PLA solid sheets. Stacked materials were heated in the oven again for 15-30 mins and pressed at 1.5 MPa for 15 mins. Similar mass-holding approach was employed to ensure the cooling and solidification of finally prepared biocomposite laminates with fibre contents of 5, 10, 20 and 30 wt%, respectively. A schematic diagram for the layout of laminate fabrication is illustrated in Fig. 1, in which the silicon paper sheets are non-stick material layers to facilitate the laminate release.

2.3. Material characterisation

Strip-like specimens were cut from biocomposite laminates in accordance with ASTM D638 and ASTM D790 for tensile and flexural tests, based on a Lloyds EZ50 universal testing machine, respectively. Both tests were conducted at 2 mm/min (a guage length of 115 mm used in tensile tests). Over four specimens were used for each material batch to ensure the good testing reproducibility.

The surface morphology of fractured tensile specimens, subjected to being sputter coated by platinum, was examined by using a Philips XL20 field emission SEM at 15 kV to investigate the distribution of coir fibres and interfacial bonding between coir fibres and PLA matrices. It is also used to correlate the material structure with mechanical properties accordingly.

Thermogravimetric analysis (TGA) was performed with a Seiko SII Exstar 6000 (TG/DTA 6200) to detect the thermal decomposition effect of PLA/coir fibre biocomposites. About 6-10 mg TGA samples were examined from 40 to 900°C at a heating ramp rate of 10°C/min in nitrogen flow of 200 ml/min.

The thermal properties of biocomposite laminates were measured by using a PerkinElmer DSC 6000 from 30 to 200°C based on two heating/cooling cycles. The DSC samples were also under an isothermal condition at 200°C for 2 mins for removing any previous thermal

history. The associated thermal parameters such as glass transition temperature (T_g), cold crystallisation temperature (T_{cc}), melting temperature (T_m), as well as the heat of fusion (ΔH_m) were determined from the first heating cycle. The resulting degree of crystallinity (X_c) was calculated based on the following Eq. (1) [31, 32]:

$$X_c(\%) = \frac{\Delta H_m}{\Delta H_m^0} \times \frac{100\%}{w} \qquad (1)$$

where ΔH_m^0 is the heat of fusion for a 100% crystalline PLA material (ΔH_m^0 =93 J/g [33]) and w is the weight fraction of PLA matrices in biocomposite laminates.

In biodegradability tests, 20×20 mm^2 specimens were cut from biocomposite laminates, weighed and buried into a deep layer of compost medium. Water was constantly added to the compost container every day for good microbial activity. After three typical periods of 6, 12 and 18 days, the specimens were taken out, washed with tap water and dried in the vacuum oven for 12 h to remove any moisture. The weight loss was calculated based on the following Eq. (2) [34]:

$$W_{loss}(\%) = \frac{W_o - W_t}{W_o} \times 100\% \qquad (2)$$

where W_o and W_t are weights of specimens in the compost soil before and after burial. The final results were reported from the average data based on up to five specimens for each of coir fibre contents.

3. Results and discussion

3.1. Morphological structures

As illustrated in Figs. 2(a)-(d), the apparent voids due to the fibre pull-out were detected despite the existence of certain partial bonding between large microfibres and PLA matrices. These voids inevitably generate the weak zones where the load bearing capacity tends to gradually diminish leading to the lower strength levels. Furthermore, the void size is comparatively small for treated fibre biocomposites as opposed to untreated fibre

counterparts. At the higher fibre content of 20 wt%, saturated fibre bundles were mainly observed in the morphology of untreated fibre biocomposites, which become stress-concentration initiators prone to the crack failure, Fig. 2(e). By contrast, a majority of treated fibres within biocomposites are fractured along PLA matrix planes, and very strong interfacial bonding is presented with little sign of fibre pull-out, Fig. 2(f). When the maximum 30 wt% coir fibres were used in this study, considerably large fibre bundles are demonstrated in Figs. 2(g) and (h) and separated from PLA matrices to form the non-uniform and poor fibre distribution. The actual fabrication of relevant biocomposite laminates appears to be very difficult for the good control of bulk material using compression moulding technique due to the excessive fibre layout.

3.2. Mechanical properties

As seen from Figs. 3(a) and (d), tensile and flexural moduli of treated fibre biocomposites are improved moderately when compared to those of neat PLA at the fibre content levels from 5 to 20 wt%. The maximum tensile and flexural moduli are shown to be 1.47 GPa and 3.07 GPa, respectively, both at the same fibre content of 5 wt%. Nonetheless, at the high fibre content of 30 wt%, significant modulus reduction takes place by 45.7% in tensile modulus and 75.4% in flexural modulus as opposed to corresponding PLA mechanical results. In comparison, untreated fibre biocomposites demonstrate the enhanced moduli by the maximum 25.6% (tensile modulus) and 13.4% (flexural modulus) with 5 and 20 wt% coir fibre reinforcements, respectively. Conversely, it appears that the moduli decline slightly at the fibre content of 10 wt%, but this trend becomes considerably pronounced at 30 wt%. The treated fibres may not consistently lead to the higher composite moduli than untreated fibre counterparts, especially at higher fibre contents of 20 and 30 wt%. This result can be explained by the less sensitivity of tensile modulus of biocomposites to the state of adhesion between fibre reinforcements and polymer matrices than that of tensile strength [27].

On the other hand, tensile and flexural strengths of biocomposites are consistently lower than those of neat PLA regardless of fibre treatment, Figs. 3(b) and (e). However, the substantial strength-regaining phenomenon becomes more manifested for those treated fibre biocomposites, resulting in the optimum tensile/flexural strength taking place at 20 wt% fibre reinforcements. Overall, the strength levels of biocomposites with untreated fibres are inferior to their counterparts with treated fibres, which is in good agreement with the previous literature [30]. This finding may be attributed to the weak interfacial interactions between untreated fibres and PLA matrices [22]. From the perspective of morphology structures, consistently lower tensile strengths of biocomposites relative to that of neat PLA can be predominantly influenced by the fibre voids and pull-out at the low fibre contents of 5-10 wt% and by the fibre agglomeration at the high fibre contents of 20-30 wt%. In addition, lower fibre length associated with small aspect ratios may also adversely affect the coir fibres acting as reinforcements, leading to ineffective stress transfer between fibres and matrices with lower strengths [35, 26].

The elongations at break for biocomposites, as depicted in Fig. 3(c), are relatively low at the fibre contents of 5-20 wt% when compared with that for PLA. This result indicates that more brittle composite characteristic than that of neat PLA is highlighted owing to the well-known rigid reinforcement nature of wetted coir fibres with PLA matrices. More surprisingly, at the high fibre content of 30 wt%, such biocomposites oppositely possess much higher elongation at break as opposed to that of PLA. In general, coir fibres show much greater elongation at break (15-40%) [29, 36] when compared with other typical natural fibres like jute and sisal with relatively small elongation values at 1.16-1.5% and 3-7% [29], respectively. Such different fibre characteristic implies that the enhancement of elongation at break for PLA/coir fibre biocomposites with 30 wt% coir fibres can be ascribed to the existence of excessive unwetted fibre bundles, illustrated in Figs. 2(g) and (h), to facilitate the further longitudinal

deformation of biocomposites under tensile loading. Similar to the previous strength results, the consistently high elongation at break for PLA/treated coir fibre biocomposites is also revealed in good agreement with PP/treated coir composites [30]. The alkali fibre treatment often leads to the removal of the lignin and pectin from coir fibres, and thus the cellulose content can be relatively increased, resulting in the improvement of flexibility and elasticity of coir fibres [30].

Based on the tensile data provided in Fig. 3(b), tensile strengths of PLA/coir fibre biocomposites in terms of fibre content were estimated by using a well-known Nielsen model [37], generally developed to evaluate the stress concentration effect of the fillers within polymer matrices. Its mathematical equation is expressed as follows:

$$\sigma_C = \sigma_m (1 - v_f^{\frac{2}{3}}) K \qquad (3)$$

where σ is the tensile strength, v is the volume fraction and the subscripts c, f and m represent composite, fibre and polymer matrix, respectively. K is a stress concentration factor, which accounts for the weakness in the structure caused by the discontinuity in stress transfer and generation of stress concentration at the filler/polymer matrix interface [38, 39]. There is no stress concentration when K=1, and the lower K value means that stress concentration effect is much greater. The experimental data of treated and untreated fibre biocomposites were fitted to the Nielsen model by using a least square method [40, 41] in order to determine the associated K values. As displayed in Fig. 4, despite the noticeable data deviation, much better estimation has been shown between experimental data of untreated fibre biocomposites and theoretical results of Nielsen model when compared with those of treated fibre biocomposites. This observation further validates even weaker interfacial bonding between untreated fibres and PLA matrices as one of applicable conditions in Nielsen model [38], which is in good accordance with the morphological structures in Figs. 2(a) and (c). The K

values for treated and untreated fibre biocomposites were found to be 0.66 and 0.27, respectively, indicating that more severe stress concentration effect takes place in untreated fibre biocomposites, mainly due to the fibre agglomeration as evidenced in the aforementioned SEM results. The interfacial adhesion is believed to be better improved by the fibre treatment with the validation of theoretical modelling as well.

3.3. TGA results

The TGA results for untreated and treated coir fibres, consisting of both TGA and DTG curves, are demonstrated in Fig. 5. The decomposition profile of untreated coir fibres can be characterised by three distinct peaks [10, 42]. The first-peak phenomenon for the loss of weight at the temperature range from 25 to 150°C is attributed to the evaporation of water. This is followed by the second peak taking place between 190 and 290°C, which corresponds to the hemicellulose degradation. The third sharp peak in accordance with the thermal degradation of cellulose is assigned between 290 and 360°C. Furthermore, the degradation of lignin occurs between 280 and 500°C [10, 42]. On the other hand, alkali treatment appears to considerably enhance the thermal stability of coir fibres, as indicated in Table 1, by increasing maximum degradation temperatures for the second peak from 271 to 310°C for untreated and treated fibres, respectively. Such finding can be explained by the removal of a significant amount of hemicellulose during the alkali treatment as well as easily hydrolysed substances that decompose earlier than those cellulose and lignin [42].

As depicted in Fig. 6 and Table 1, neat PLA indicates the degradation peak at 358°C while PLA/untreated coir fibre composites result in the peak shift to the lower temperatures at 313, 347, 333 and 327 °C for the fibre contents of 5, 10, 20 and 30 wt% accordingly. Similar trend was also detected in Fig. 7 for PLA/treated coir fibre composites apart from a much lower shift level in range from 321 to 293°C. Evidently, increasing the fibre content enables the decrease of composite thermal stability owing to the lower degradation temperature of coir

fibres [10], which is in good agreement with the material characteristics of composites reinforced with sisal fibres [43] and phormium fibres [28].

3.4. DSC thermal analysis

The DSC thermograms and associated thermal parameters for neat PLA and PLA based biocomposites are illustrated in Fig. 8 and Table 2, respectively. The addition of coir fibres does not significantly affect the T_g and T_m values despite the modest decreases of these two critical temperatures. It reveals that interactions between coir fibres and PLA matrices are not strong enough to slow down the chain mobility in relation to the glass transition. T_{cc} of biocomposites decreases considerably from 115 to 110ºC regardless of fibre treatment, as opposed to that of neat PLA (T_{cc} =119 ºC). Accordingly, coir fibres are confirmed to play an effective nucleating role to accelerate the PLA crystallisation process, and thus improve the crystal growth rate. This is a general material characteristic of typical natural fibres as also found in PLA/hemp fibre composites [44], PLA/kenaf fibre composites [45] and PLA/phormium tenax composites [28]. On the other hand, the determined X_c for biocomposites is noticeably higher when compared with that for neat PLA. Treated fibre biocomposites become superior to untreated fibre counterparts in terms of the increasing level of X_c. The higher X_c of polymer matrices often results in better mechanical properties of polymer composites, which is in good accordance with the consistently higher level of tensile/flexural strengths in treated fibre biocomposites depicted in Figs. 3(b) and (e). Besides, the maximum X_c value of 20.64% is determined at treated fibre content of 20 wt%, which also coincides with the optimum strengths obtained at the same content level in this study. The alkali treatment seems to contribute to the improvement of interfacial bonding between coir fibres and PLA matrices to increase their potential hydrogen bonding and mechanical interlocking.

3.5. Biodegradable effect

The biodegradability level of biocomposites was characterised by using the weight loss of material samples after 6, 12 and 18 days, Fig. 9. There is almost no weight loss found for neat PLA until after 18 days with 18.1 % weight deduction. The biodegradation rate of PLA/coir fibre biocomposites appears to be greater in comparison with that of neat PLA, resulting from hydrophilic nature of coir fibres to facilitate the fast water diffusion from compost into composites through fibres or at fibre-matrix interfaces. More obviously, treated fibre biocomposites are subjected to more pronounced weight loss relative to untreated fibre counterparts. The highest level of weight losses (34.9% and 22.6%, respectively) takes place concurrently at the fibre content of 10 wt% in both cases after 18 days. The higher level of weight loss for treated fibre biocomposites may be due to the existence of residual NaOH to break up the PLA polymeric chains leading to a faster degradation rate in compost burial tests. Irrespective of fibre treatment, the coir fibre content of 10 wt% seems to be the saturation level for the biodegradable effect, above which the excessive amount of coir fibres tend to form their individual fibre bundles or aggregates rather than interact actively with PLA and compost for any further fast biodegradation process arising from the bacteria growth.

4. Conclusions

Mechanical, thermal properties and biodegradability of PLA/coir fibre biocomposites with and without alkali treatment have been successfully evaluated. The tensile/flexural strengths of treated fibre biocomposites were better than those of untreated counterparts despite being less than that of neat PLA in both cases. This result can benefit from the alkali treatment to improve the fibre-matrix interfacial adhesion and fibre wettability. In view of the morphological structure, the low strength phenomenon for biocomposites can be associated with the obvious voids due to fibre debonding and pull-out at the low fibre contents of 5-10

wt%, as well as prevalent fibre agglomeration at the high fibre contents of 20-30 wt%. Due to embedded coir fibres with lower thermal degradation temperatures, the thermal stability of biocomposites decreases irrespective of the fibre treatment. The X_c of biocomposites is enhanced as opposed to that of PLA. Furthermore, much higher X_c values have been found for treated fibre biocomposites, which are in good agreement with their relatively good mechanical properties. The typical nucleating agent role of coir fibres is also confirmed in that the embedded coir fibres decrease the T_{cc} of biocomposites substantially to facilitate the PLA rapid crystallisation process. The soil burial tests suggest that good biodegradability has been shown in biocomposites, especially to a greater extent for those with fibre treatment.

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Fig. 9 Biodegradability of neat PLA and PLA/coir fibre biocomposites (UFC= untreated fibre biocomposites and TFC= treated fibre biocomposites).

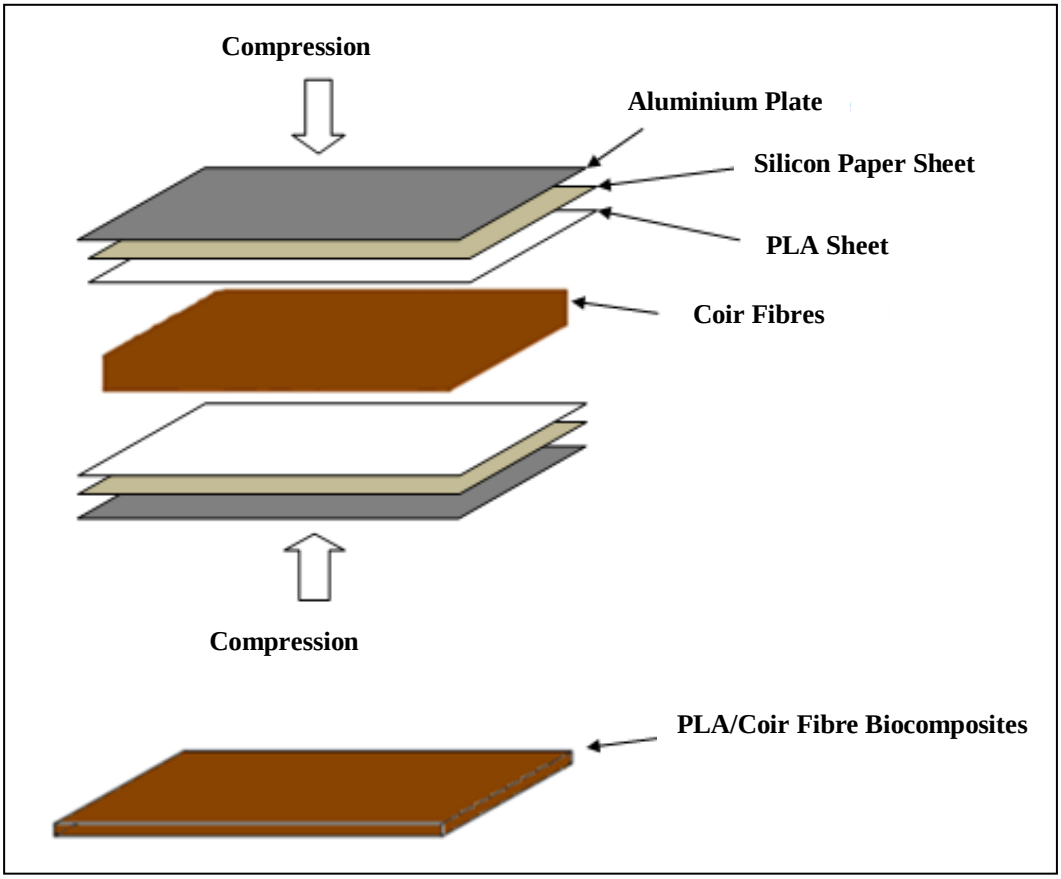

Compression
Aluminium Plate
Silicon Paper Sheet
PLA Sheet
Coir Fibres
Compression
PLA/Coir Fibre Biocomposites

Fig. 1

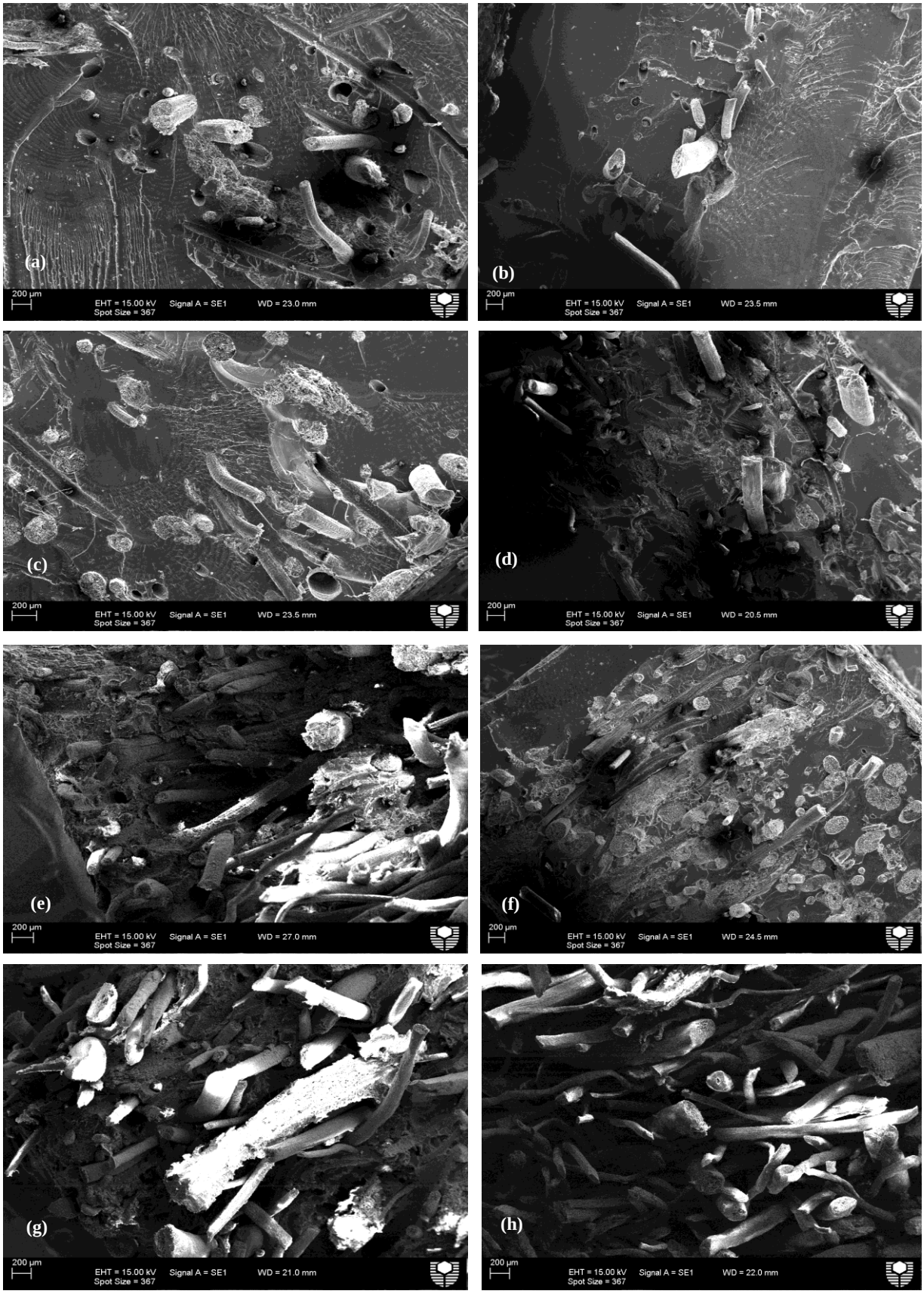

(a)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 23.0 mm
(b)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 23.5 mm
(c)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 23.5 mm
(d)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 20.5 mm
(e)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 27.0 mm
(f)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 24.5 mm
(g)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 21.0 mm
(h)
200 µm
EHT = 15.00 kV
Spot Size = 367
Signal A = SE1
WD = 22.0 mm

Fig. 2

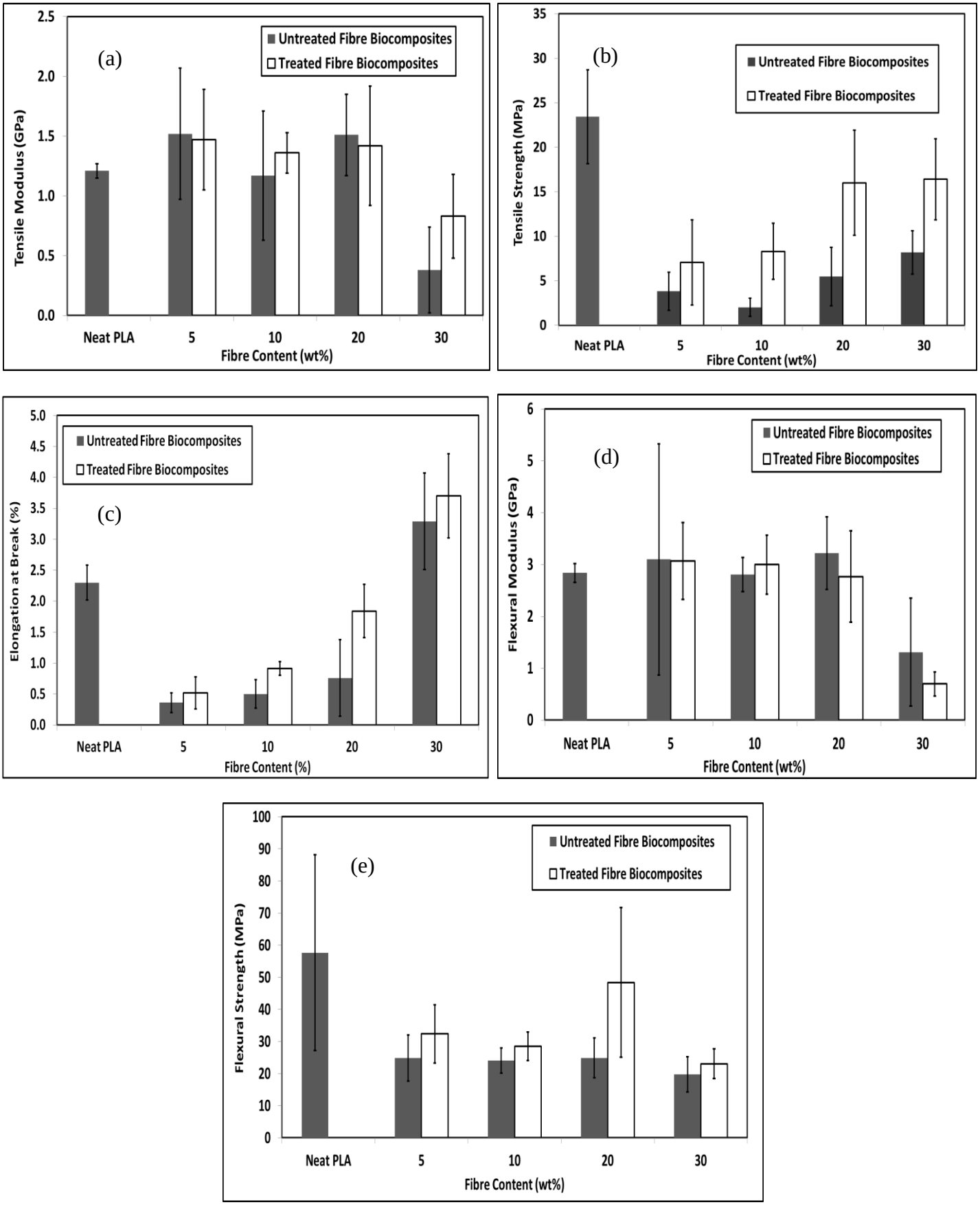

(a)
Untreated Fibre Biocomposites
Treated Fibre Biocomposites
Tensile Modulus (GPa)
2.5
2.0
1.5
1.0
0.5
0.0
Neat PLA
5
10
20
30
Fibre Content (wt%)
(b)
Untreated Fibre Biocomposites
Treated Fibre Biocomposites
Tensile Strength (MPa)
35
30
25
20
15
10
5
0
Neat PLA
5
10
20
30
Fibre Content (wt%)
(c)
Untreated Fibre Biocomposites
Treated Fibre Biocomposites
Elongation at Break (%)
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
Neat PLA
5
10
20
30
Fibre Content (%)
(d)
Untreated Fibre Biocomposites
Treated Fibre Biocomposites
Flexural Modulus (GPa)
6
5
4
3
2
1
0
Neat PLA
5
10
20
30
Fibre Content (wt%)
(e)
Untreated Fibre Biocomposites
Treated Fibre Biocomposites
Flexural Strength (MPa)
100
90
80
70
60
50
40
30
20
10
0
Neat PLA
5
10
20
30
Fibre Content (wt%)

Fig. 3

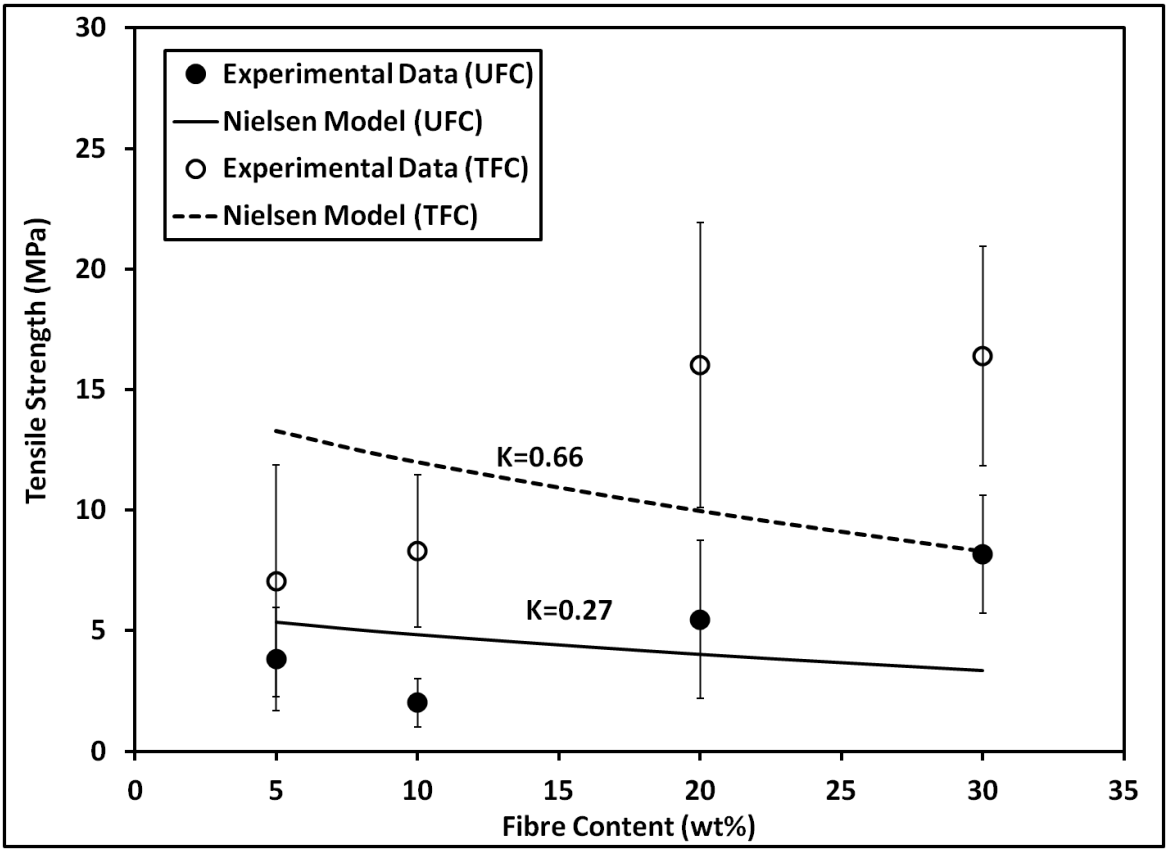

Experimental Data (UFC)
Nielsen Model (UFC)
Experimental Data (TFC)
Nielsen Model (TFC)
K=0.66
K=0.27
Tensile Strength (MPa)
Fibre Content (wt%)
0
5
10
15
20
25
30
35

Fig. 4

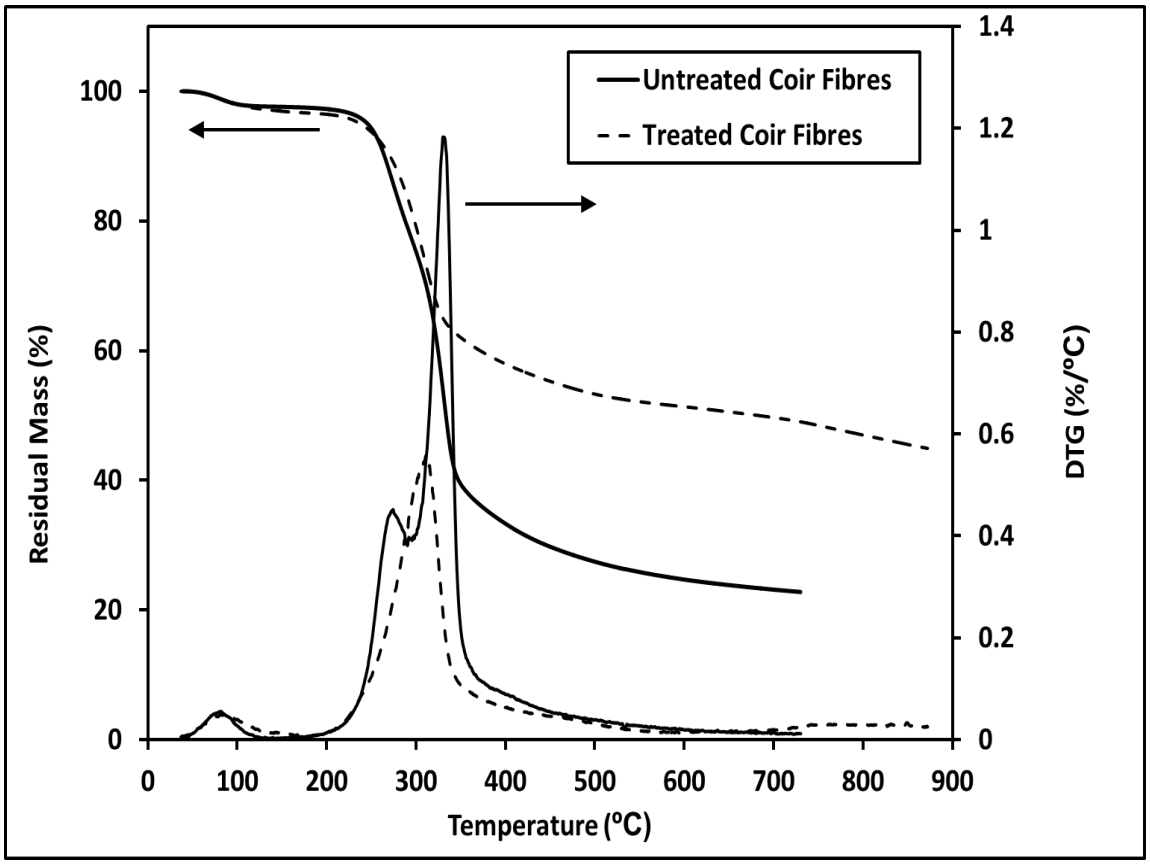
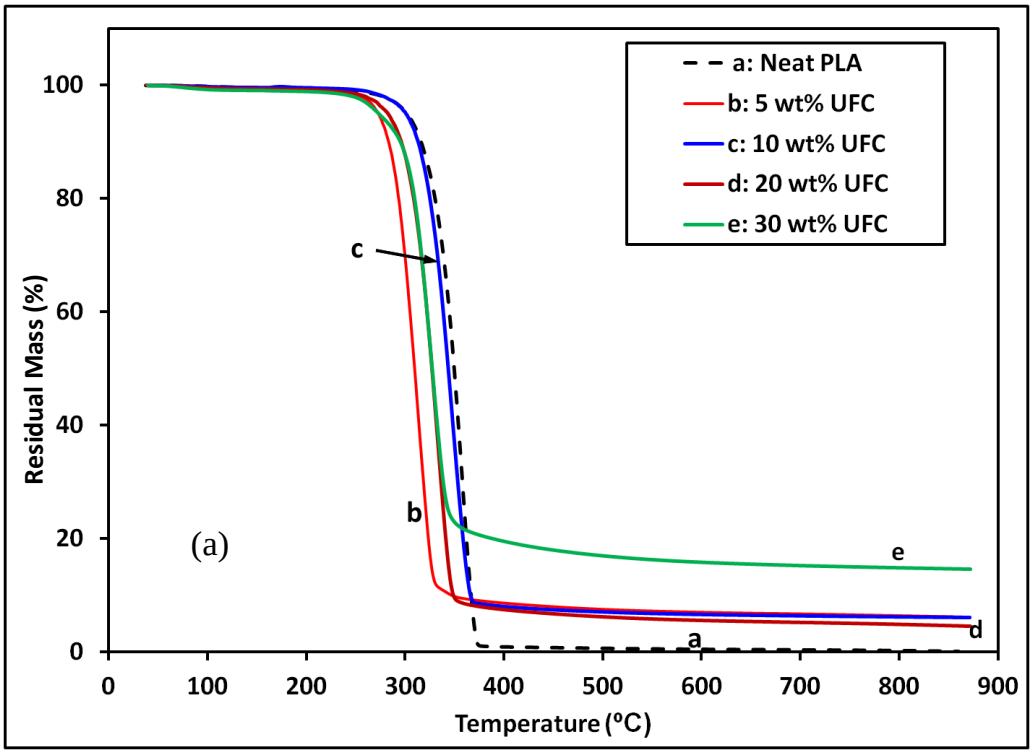

Untreated Coir Fibres
Treated Coir Fibres
Residual Mass (%)
DTG (%/°C)
Temperature (°C)
100
80
60
40
20
0
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0
100
200
300
400
500
600
700
800
900

Fig. 5


a: Neat PLA
b: 5 wt% UFC
c: 10 wt% UFC
d: 20 wt% UFC
e: 30 wt% UFC
Residual Mass (%)
Temperature (°C)
(a)
a
b
c
d
e

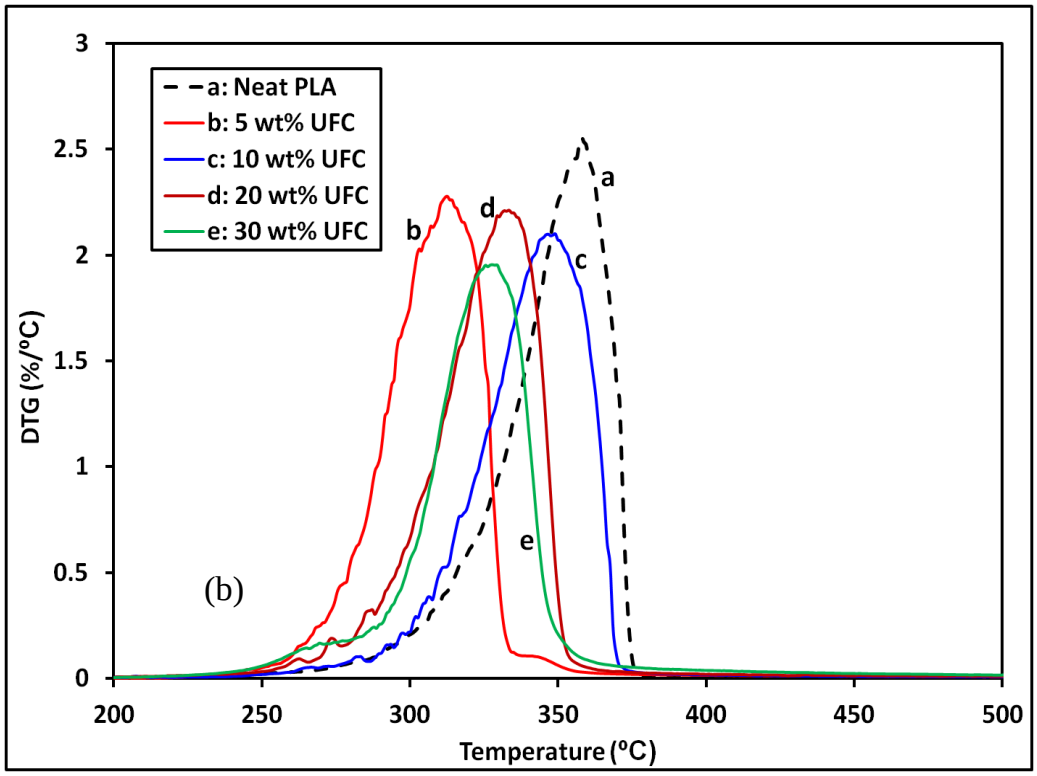

a: Neat PLA
b: 5 wt% UFC
c: 10 wt% UFC
d: 20 wt% UFC
e: 30 wt% UFC
DTG (%/°C)
Temperature (°C)
(b)
a
b
c
d
e

Fig. 6

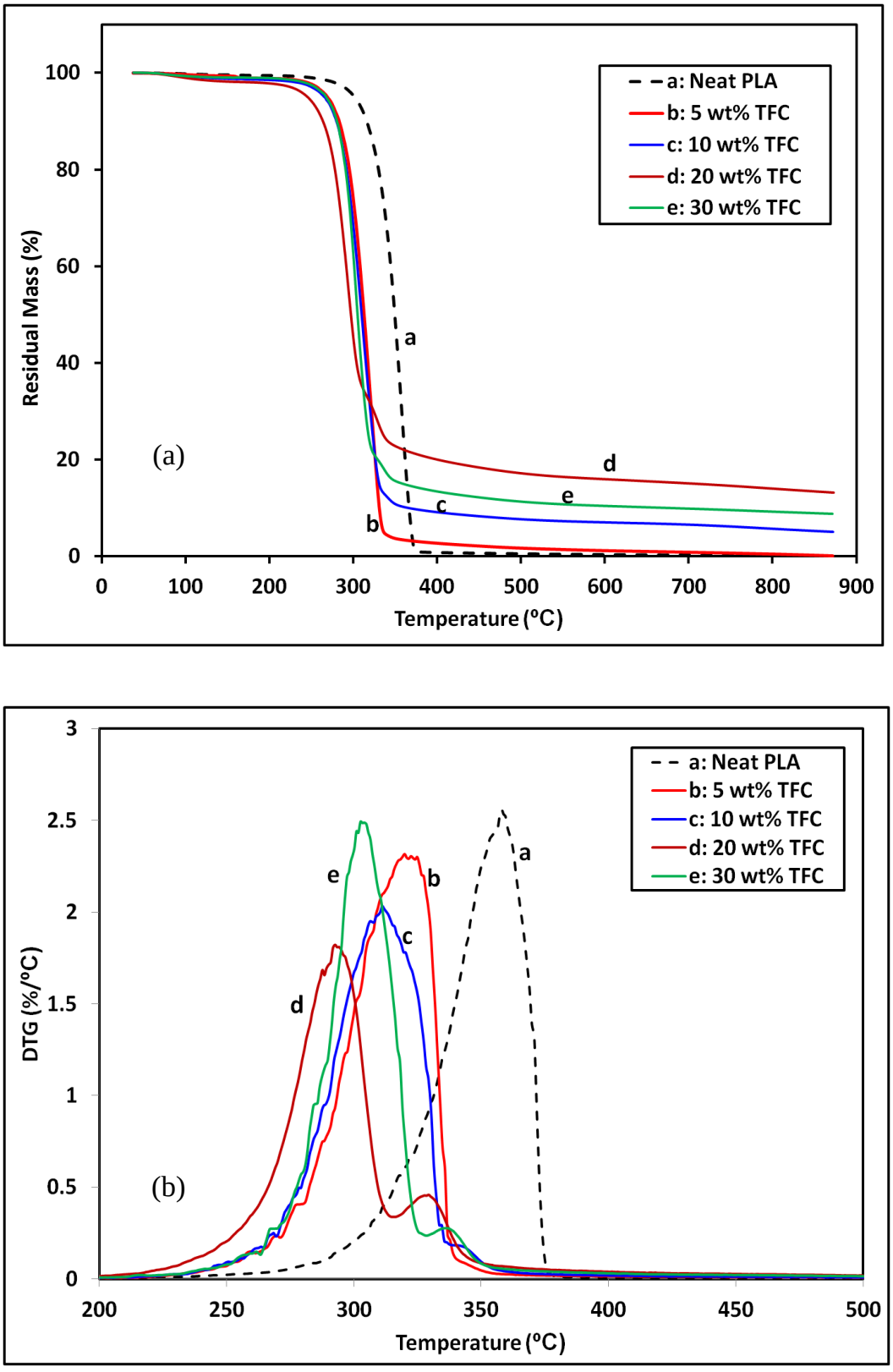

Residual Mass (%)
Temperature (°C)
a: Neat PLA
b: 5 wt% TFC
c: 10 wt% TFC
d: 20 wt% TFC
e: 30 wt% TFC
(a)
DTG (%/°C)
Temperature (°C)
(b)

Fig. 7

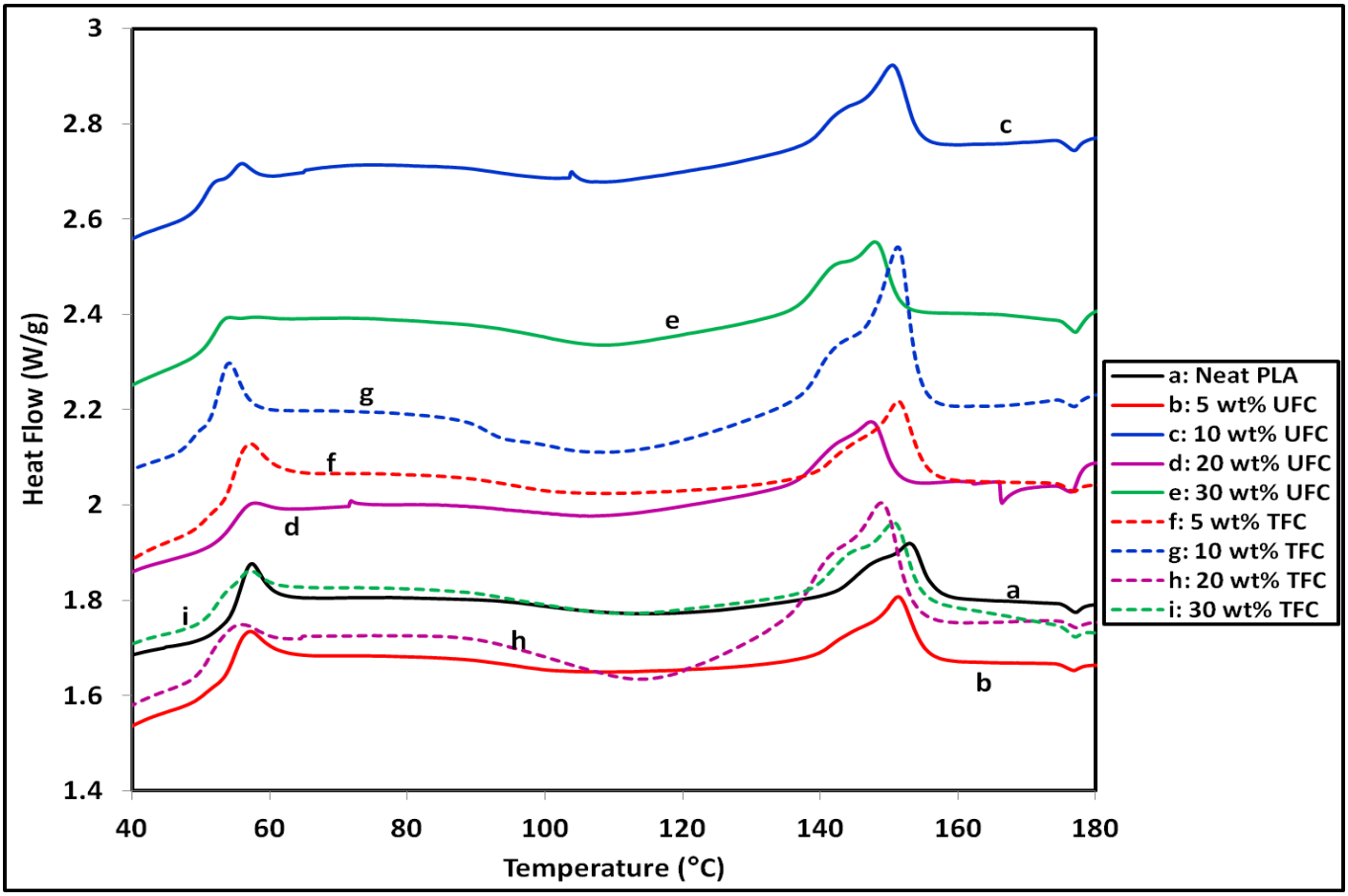

Heat Flow (W/g)
Temperature (°C)
3
2.8
2.6
2.4
2.2
2
1.8
1.6
1.4
40
60
80
100
120
140
160
180
a
b
c
d
e
f
g
h
i
a: Neat PLA
b: 5 wt% UFC
c: 10 wt% UFC
d: 20 wt% UFC
e: 30 wt% UFC
f: 5 wt% TFC
g: 10 wt% TFC
h: 20 wt% TFC
i: 30 wt% TFC

Fig. 8

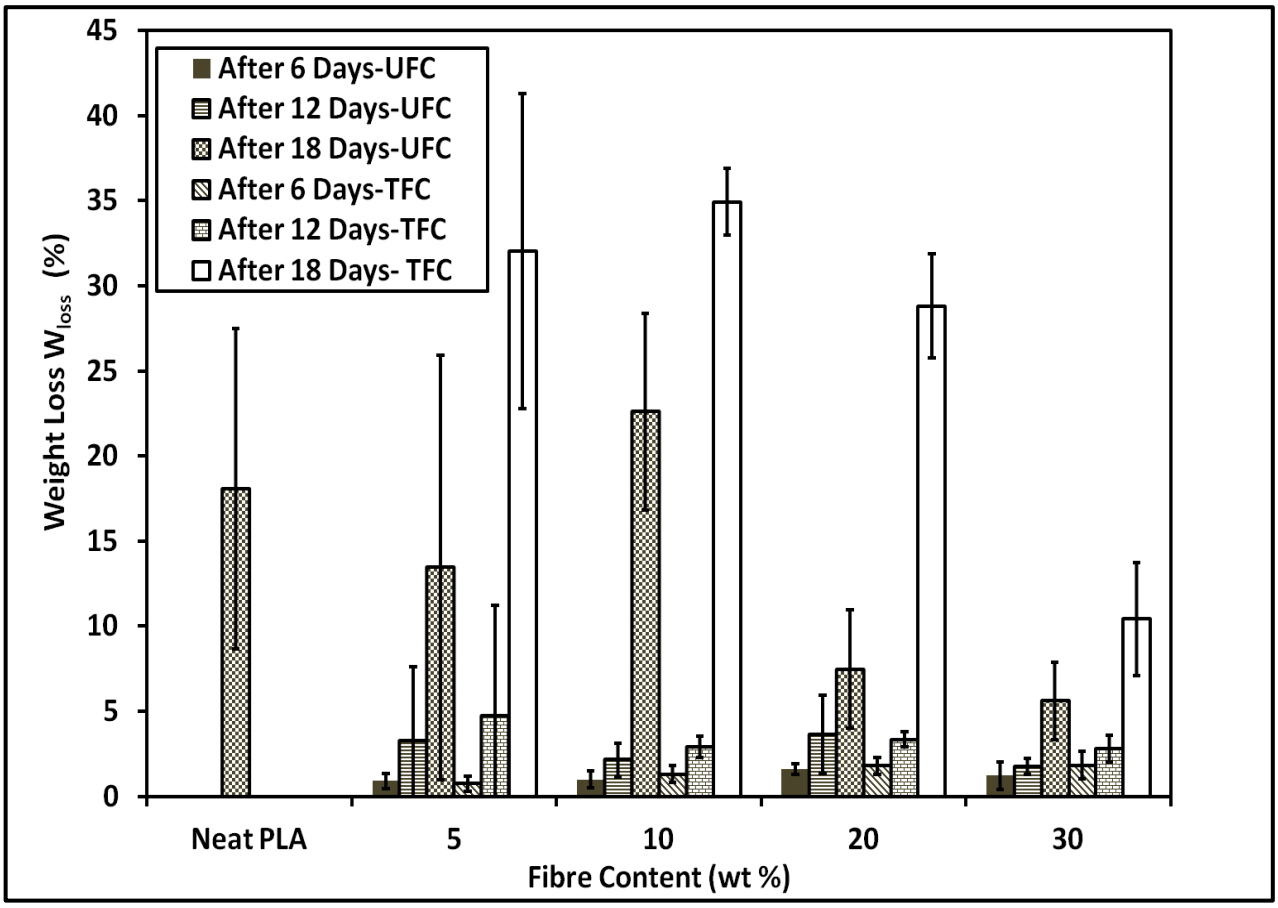

After 6 Days-UFC
After 12 Days-UFC
After 18 Days-UFC
After 6 Days-TFC
After 12 Days-TFC
After 18 Days- TFC
Weight Loss W_{loss} (%)
45
40
35
30
25
20
15
10
5
0
Neat PLA
5
10
20
30
Fibre Content (wt %)

Fig. 9

Table 1. TGA peak data* for coir fibres, neat PLA and PLA/coir fibre biocomposites

Material type	T_{d1} (ºC)	DTG (%/ºC)	T_{d2} (ºC)	DTG (%/ºC)	T_{d3} (ºC)	DTG (%/ºC)
Untreated coir fibres	80.3	0.055	271.0	0.443	331.3	1.183
Treated coir fibres	82.4	0.050	310.3	0.551		
Neat PLA			357.8	2.561		
5 wt% UFC			312.8	2.277		
10 wt% UFC			347.3	2.097		
20 wt% UFC			332.7	2.207		
30 wt% UFC			327.3	1.960		
5 wt% TFC			320.5	2.306		
10 wt% TFC			310.9	2.027		
20 wt% TFC			293.0	1.819	329.0	0.456
30 wt% TFC			303.7	2.493	336.6	0.277

**All TGA peak data are benchmarked against those of untreated coir fibres. T_{d1}, T_{d2} and T_{d3} are temperatures of maximum degradation rate for degradation peaks 1, 2 and 3 in DTG curves, respectively. UFC= untreated fibre biocomposites and TFC= treated fibre biocomposites.*

Table 2. Thermal properties of neat PLA and PLA/coir fibre biocomposites

Material sample	T_g (°C)	T_{cc} (°C)	T_m (°C)	ΔH_m (J/g)	X_c (%)
Neat PLA	54.78	119.00	152.97	6.44	6.93
5 wt% UFC	53.76	112.63	151.42	8.37	9.48
10 wt% UFC	50.83	110.03	150.49	8.65	10.33
20 wt% UFC	54.44	112.44	147.39	6.55	8.81
30 wt% UFC	51.49	109.50	147.79	7.86	12.08
5 wt% TFC	53.64	114.81	150.92	6.52	7.38
10 wt% TFC	49.60	110.02	151.27	16.66	19.91
20 wt% TFC	51.43	114.29	148.86	15.36	20.64
30 wt% TFC	52.77	113.33	150.63	9.42	14.47

Note that UFC= untreated fibre biocomposites and TFC= treated fibre biocomposites.