Characterisation of Natural Fibre Thermoplastic Composites using Ultrasonic Longitudinal Sound Wave

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Abstract
Natural fibre thermoplastic composites, NFTC composites, have attractive advantages such as the low density, low cost, carbon neutrality, applicability to the available processing machines of thermoplastics and good mechanical specific properties with respect to the counterparts of glass fibre composites.

The motivation of this work is as follows: Injection moulding of NFTC is a preferable production technique. However, the fibre content definition at different positions of 30 wt.% composite sample shows a deviation up to 30%. This variation is believed to affect the corresponding mechanical properties. The test was carried out by dissolving the host polymer matrix. It is known that the longitudinal sound speed is a function of the material property which in turn is a function of fibre content and adhesion efficiency. Therefore the aim of this article is to study the feasibility of using the ultrasonic longitudinal sound wave and the time of flight TOF instead of the exhausting destructive technique of polymer dissolving. The aim of this work is extended to define the fibre content and distribution in natural fibre composites as well as the effect of fibre length.

Experimental work is executed using different natural fibres namely cellulose, flax, hanf and sisal fibres at two different fibre contents. The selected host thermoplastic is polypropylene with high flow properties. The composite samples are injected moulded in the form of plates with 298*158*3 mm

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Equation (1) shows the sound speed/distance relation based on the sound pulse transmission concept of the longitudinal wave.

\[ C = \frac{2L}{\text{TOF}} = 2Lf \]  

(1)

Previous work [10, 11] shows the relation between the flax fibre content in PP matrix with the longitudinal sound speed by both dry and wet measurements. Figure 1 shows this relation as presented in [1].

![Figure 1: Effect of fibre content on the longitudinal sound speed using USPC 3040 DAC](image)

The objective of this work is to check this fibre content-sound speed technique in the inspection of fibre content homogeneity in PP plates reinforced with different types of natural fibres with different stiffness and geometries. Then to find out the effect of the fibre type on the flow pattern of the composite.

2. Experimental work

The joint project (Natural fibre composites simulation) aims for a full characterisation of NFTC (rheological, thermal and mechanical) in order to simulate the composite behaviour during injection and further under mechanical loading. Thus would result in the implementation of new NFTC materials in high added values industries. Within the activities of the above mentioned project, injection moulded NFTC plates are supplied. The host matrix is polypropylene PP (Moplen EP 500 V) with high flow properties and 0.9 g/cm³. The plates are 298x158x3 mm³. Table 1 shows the used fibres in this study which are supplied by local suppliers. Density of PP and the original granulates are tested according to DIN EN 993-1. The granulates immersed in Decaline solvent and the whole flax is heated in an oil bath for 2 hours at 160°C. The solution is then filtered using pore 3 filter and the extracted fibres are re-weighed and hence the fibre content is calculated.

Table 1: The used fibres in this study

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Type of fibre</th>
<th>Nominal/actual content [wt%]</th>
<th>Original granulates Density [g/cm³]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A30</td>
<td>Flax</td>
<td>30,0 / 30,64</td>
<td>1,0174</td>
</tr>
<tr>
<td>B30</td>
<td>Hanf</td>
<td>30,0 / 28,70</td>
<td>1,0170</td>
</tr>
<tr>
<td>C30</td>
<td>Sisal</td>
<td>30,0 / 27,20</td>
<td>1,0148</td>
</tr>
<tr>
<td>D30</td>
<td>Cellulose</td>
<td>30,0 / 33,11</td>
<td>1,0268</td>
</tr>
<tr>
<td>D10</td>
<td></td>
<td>10,0 / 11,29</td>
<td>0,9379</td>
</tr>
</tbody>
</table>

2.1 Ultrasonic testing

Fibre content estimation using the ultrasonic system is carried out by USPC 3040 DAC. This machine uses distilled water bath as coupling medium. Pulse-echo mode is used. Positioning of the transducer head is adjusted to receive the back signal efficiently. The signal to noise
ratio of up to 60 dB in 0.5 dB steps and the frequency range of 1 kHz to 20 MHz (-3 dB). Time Corrected Gain TCG option is available up to 40 dB. Damping is set at 75 ohm. Pulser frequency is set at 3000 Hz. High and low filters are adjusted at 0 and 6 MHz respectively to optimise the A-scan image. The amplitude is adjusted in the range of 40-45 dB to attain a proper A-scan height (80% of the screen height).

C-scan images are produced for the 5 samples mentioned in Table 1 in addition to a reference plate for the pure PP. Previous work [10, 11] shows the dependence of the speed ‘C’ of the longitudinal wave on the fibre content ‘Wf’ in a linear function in equation 2, as illustrated in figure 1. Based on the constant thickness plate, equation 3 can be deduced. The relation controls the measured thickness w.r.t. the actual thickness is shown in equation 4 in terms of the sound speed ‘C1’ and ‘C2’ at the reference and measured positions respectively. ‘C1’ is found by adjusting the sound speed till the ultrasonic thickness measurement matches the corresponding mechanical thickness of the studied plate middle. Hence ‘Wf’ at the new position can be calculated as in equation 5. Hence a function is established between the fibre content (Wf) and the measured thickness as in the linear trend equation 1 regarding the measured ultrasonic thickness and the nominal Wf as in Table 1.

\[
C = a + bW_f = 2564 + 4.9513W_f \tag{2}
\]

\[
L = C_1 TOF_1 = C_2 TOF_2 \tag{3}
\]

\[
\frac{L_{\text{actual}}}{L_{\text{measured}}} = \frac{C_2}{C_1} = \frac{a + bW_{f_2}}{a + bW_{f_1}} \tag{4}
\]

\[
W_{f_2} = \left[ \frac{L_{\text{actual}}}{L_{\text{measured}}} \right] (a + bW_{f_1}) - a \quad / b \tag{5}
\]

For very small a/b ratio as shown in equation 2, equation 6 can be deduced:

\[
W_{f_2} = \frac{L_{\text{actual}}}{L_{\text{measured}}} W_{f_1} \tag{6}
\]

The plate is scanned in two steps. Firstly, the start-middle region is scanned and then the middle-end region is tested. That is because of the warping present in some plates which affects the accuracy of measurement. Figure 2 presents a sketch of the studied plates. The dashed rectangles represent the two testing steps. The small circles present the points of the experimental validation of fibre content. All samples are left in water bath for 48 hours to avoid the quick water absorption phase and its effect on dimensional stability.

Figure 2 Tested plate showing the regions of scanning and the locations where the fibre content is validated experimentally

2.2 Fibre extraction destructive technique

The fibre content at some selected locations (shown in figure 2) are determined experimentally (destructive test) using the decaline solvent in the above mentioned procedure.
The sample is weighed before and after the test to calculate the fibre content. The extracted fibres from each spot are inserted afterwards in isopropanol solution and pumped in the QICPIC equipment to measure the geometry of the fibres with wet dispersion technique. QICPIC depends on a pulsed light source and telecentric imaging where the fibres are monitored by a high speed mega-pixel camera as shown in figure 3. The experiments are done using two types of lenses namely M8 (20-6820 µm) and M6 (5-1750 µm) according to ISO 13322-2. Each test is repeated twice because bigger lens is suitable for fibre length and the smaller lens is more accurate for the diameter measurement. The pumping cycle of the fibres is repeated three times to check the results reproducibility. Lixell wet dispersion technique can provide high contrast images even for the cellulose fibres. Using wet dispersion technique avoids the dry technique disadvantages where the fibres are agglomerating in many positions leading to discrepancy in the image processing results. The pumped dispersion flows through a cuvette of 1 mm gap where the images are taken. Image recording rate is 100 Hz. For the subsequent image analysis the following settings are considered in order to evaluate the results statistically.

- The fibre lengths of less than 10 µm are disregarded because they are dust most likely.
- The optic concentration of more than 1% is disregarded as it is considered fibres agglomeration site.
- QICPIC Windox software deal with the measured fibres by considering the count of fibres (referred by q1) or by considering the fibre imaged surface area (referred by q2), or by considering their volumes (referred by q3). Q2 is found to have close results with the data sheets of the suppliers.

Figure 3: Scheme of QICPIC measurement [12]

3. Results and discussion

3.1 Fibre content using ultrasonic C-images

Figure 4 shows the C-scan of the plates at the two positions (start-middle) and (middle-end) as mentioned in figure 1. The reference PP is scanned and shown in figure 4a. The longitudinal speed is calibrated at 2700 m/s to achieve plate thickness of 4 mm as mechanically measured. As seen in figure 4a, the measured ultrasonic thicknesses show a deviation of thickness in the range of [3.95-4.05] mm. Some odd sites are present as the green coloured site (above right) which is corresponding to 4.1. The deviation in measurement is attributed to the microvoids introduced during injection as reported by Kas [13]. Therefore this deviation even in the pure PP sample will be considered during the discussion of the composites’ C-scans. Scan axis is the plate symmetrical X-axis and the index axis is the orthogonal one.

In the composites’ C-scans, the thickness meter is correlated with another fibre content meter flow (as explained in figure 1). Figures 4b and 4c show the A30 sample (PP reinforced with
The flow pattern is sketched using line art sketch on the C-scan images. Agglomeration site is evidenced by a dark red region in [220-240] mm scan range. Afterwards the flow pattern retrieves its stable planar flow. The fibre content (according to the colour pattern) in figure 3b is 28.8-31.1% while it is 31.1-31.4% in figure 4c. This great deviation is not obvious in plate B where hemp fibres is used with the same loading percentage. Figure 4d shows fibre content range of 28.5-28.9% as shown in figure 4d and 4e. C30 (sisal sample) shows a range of fibre content starting from 26% to more than 30% as shown in figure 4f and 4g. Most of the above mentioned plates in figures 4b,c,d,f,g shows unsteady planar flow fronts where fountain features are still obvious. Thus indicates the presence of multi-flow system where the fibre content is believed to be non-consistent along the plate. While in the case of cellulosic based plates (D, E), they show steady planar flow as in figure 4h and 4i for the 30 and 10% loadings respectively.
3.2 Fibre content using fibre extraction destructive technique

Table 2 shows the experimental fibre content versus the estimated result using the ultrasonic technique. The results show good matching between the actual and estimated values.

<table>
<thead>
<tr>
<th>Position</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>27.89</td>
<td>27.36</td>
<td>23.47</td>
<td>28.53</td>
<td>10.74</td>
</tr>
<tr>
<td></td>
<td>28.80*</td>
<td>28.50</td>
<td>26.00</td>
<td>30.00</td>
<td>10.70</td>
</tr>
<tr>
<td></td>
<td>29.20</td>
<td>28.02</td>
<td>28.85</td>
<td>29.40</td>
<td>10.83</td>
</tr>
<tr>
<td></td>
<td>30.70</td>
<td>28.90</td>
<td>27.50</td>
<td>29.70</td>
<td>11.20</td>
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<tr>
<td></td>
<td>33.79</td>
<td>29.31</td>
<td>29.36</td>
<td>32.40</td>
<td>11.58</td>
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<tr>
<td></td>
<td>32.40</td>
<td>28.70</td>
<td>30.00</td>
<td>31.00</td>
<td>11.50</td>
</tr>
<tr>
<td>Deviation</td>
<td>5.90</td>
<td>1.95</td>
<td>5.89</td>
<td>3.87</td>
<td>0.84</td>
</tr>
<tr>
<td>[wt.-%]</td>
<td>3.60</td>
<td>0.40</td>
<td>4.00</td>
<td>2.47</td>
<td>0.80</td>
</tr>
</tbody>
</table>

* Values in italics are the estimated values from the ultrasonic test

3.3 Effect of fibre type and geometry

The range of fibre content variation is shown in the last row of Table 1. Composites reinforced with regenerated cellulose and Hanf fibres have the smallest range of fibre content variation (samples B,D,E). Flax and sisal (samples A, C) have bigger variation range. Hanf and flax belong to the same fibres group which is the bast fibre. However the flax has bigger variation range than that of the sisal. This is attributed to the fibre flexibility in case of flax and stiffness in case of sisal. Tangling of flax fibres is more likely to occur resulting in this multi flow phenomenon. The geometry of the extracted fibres are given in Table 3.
Table 3: The used fibres in this study

<table>
<thead>
<tr>
<th>Sample code</th>
<th>A30</th>
<th>B30</th>
<th>C30</th>
<th>D30</th>
<th>D10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type of fibre</td>
<td>Flax</td>
<td>Hanf</td>
<td>Sisal</td>
<td>Cellulose</td>
<td></td>
</tr>
<tr>
<td>Diameter median[µm]</td>
<td>13,29</td>
<td>14,85</td>
<td>22,78</td>
<td>14,44</td>
<td></td>
</tr>
<tr>
<td>Length median[µm]</td>
<td>340,56</td>
<td>970,63</td>
<td>2288,69</td>
<td>386,70</td>
<td></td>
</tr>
</tbody>
</table>

It is obvious that fibres with stiff structure and non-tangling behaviour show better homogeneity in fibre distribution along the plate. The fibre geometry change along the middle and edge sides of the plates is shown in figure 6. There is a parabolic effect of fibre length in case of plate A. In general the shorter fibres accumulate along the flow of the fibres of the plate in case of plate A and D (middle lines).

Figure 6: Fibre geometry along the middle and the edge of the plate

4. Conclusion
- UT proves its efficiency in allocating fibre content.
- Fibres with stiff structure and non-tangling behaviour show the least variation in fibre distribution.
- Fibre’s geometry plays a role in flow where short fibres flow further specially in case of stiff non-tangling fibres

References

1) Bledzki AK, Sperber VE, Faruk O. Natural and wood fibre reinforcement in polymers. Rapra review reports 2002; 13(8)
12) http://www.sympatec.com/ImageAnalysis.html
13) Y O Kas and C Kaynak, ‘Ultrasonic (C-scan) and microscopic evaluation of resin transfer molded epoxy composite plates’, Polymer Testing, Vol 24, pp 114–120, 2005