UNDERSTANDING THE LINKS BETWEEN STRUCTURE
AND PROPERTIES IN DIE DRAWN
WOOD POWDER POLYMER COMPOSITES

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Abstract

Wood powder polypropylene composites (WPC) have been produced with 40%w/w of a
soft or a hard wood as filler. The die drawing process developed at the University of Leeds
[1] has been used to produce oriented composites. Aspect ratios (A_r) for both wood
powders were calculated by using a Sysmex image analyser. Prediction of the Young’s
modulus of the oriented WCP was calculated using the Cox-Krenchel model [2]. It was
found that theoretical predictions agree with the experimental data very well. Both isotropic
and anisotropic composites were examined for tensile and flexural properties, and for
thermal expansion. The results show an increase in all mechanical properties as the degree
of orientation was increased; in particular, the tensile Young’s modulus (E) was improved
from 1.9 to 8.2GPa for softwood composites (C120PP), and from 2.9 to 5.4GPa for
hardwood composites (HB120PP). The linear thermal expansion coefficient (α) of the WPC
decreased with increasing nominal draw ratio (λ) and depended on the testing direction;
e.g. for C120PP tested parallel to the orientation direction it fell from 70x10^{-6}/°C for the
isotropic material to about 22x10^{-6}/°C for an oriented composite. In addition, the
morphology (ESEM) of the composites was monitored at various stage of processing.

1. Introduction

Considerable efforts have been made to design inexpensive materials filled with wood
fibres characterized by low density and high stiffness in order to reduce the cost of the
material while at the same time improving mechanical and physical properties. One way to
achieve this goal is to produce structures with a high degree of molecular orientation.
First generation technology refers to the simple processing of WPC. The traditional
manufacture of thermoplastic composites is often a two-step process; after mixing the
composites are formed into a product by compression or injection moulding, or extrusion.
These composites have limited applications due to poor mechanical properties. However,
there is a strong movement in research towards more highly engineered WPC with greater
structural performance and an attempt to create a new, second generation of WPC by
introducing orientation of the polymer molecules [3]. The orientation process increases the

strength and stiffness of these composites significantly. The tests of oriented polymers reinforced by wood fillers have had promising results; with stiffness increased 6-fold in comparison to the first generation WPC [4]. Of particular interest is to die draw the WPC, test and link the structure to their properties via modelling.

2. **Experimental Details**

2.1. **Materials and Composite Manufacturing**

BP polypropylene pellets (PP) with a density of 920 kg/m$^3$ and MFI of 2.10g/10min were used as the matrix for two types of wood powder (WP); C120 and HB120 supplied by Rettenmaier GmbH, Germany.

Two types of WPC, C120PP and HB120PP, containing 40% w/w wood powder were blended by using a co-rotating twin-screw extruder. The temperature profile of the extruder barrel was set between 210°C and 190°C. The screw speed was changed in the range from 200 to 600 rpm to reduce tearing of the extruded strand. After extrusion, the strands were cooled in air and pelletized. The blending process was repeated twice with the same parameters in order to obtain better homogeneity and dispersion of the WP in the PP matrix.

2.2. **Sample Preparation and Characterisation**

The samples were prepared from the compounded pellets by a compression moulding technique using a heated press. The press temperature was set up to 200°C. A thermocouple was used to control the temperature inside the metal mould giving the possibility to prevent thermal decomposition of the organic fillers and the PP. The pressure was applied gradually until its maximum value of 0.8MPa, at a temperature above the melting point of the PP, and kept until end of the process. When the temperature inside the mould reached 200°C, a 3-min annealing time was allowed. The materials were then cooled at a rate of 10°C/min to ~50°C. These isotropic samples were machined into a shape in order to be oriented using the die drawing technique [1].

Because the properties of WPC are related to water content, the samples were conditioned at 65%RH for 2 weeks following the guidelines of ASTM D1037-99 by placing them in a box containing saturated magnesium acetate tetrahydrate which gives an atmosphere at this value.

2.3. **Die Drawing**

The principle of the method is illustrated in Figure1. For further details on the die drawing technique see [1].

Die drawing of the filled and unfilled PP was carried out at nominal draw ratios of 3 ($3\lambda$) and 10 ($10\lambda$). To obtain the higher $\lambda$, the materials were drawn twice because the composites were too weak to withstand the flow stress of a single draw step. In the first stage, the materials were drawn at a temperature of 150°C. In the second stage, the temperature was increased to 160°C and 165°C for C120PP and HB120PP respectively. Before drawing, the samples were kept in the heating chamber for about 20min to obtain temperature equilibrium throughout the material.
Die drawing of both materials was performed using an Instron 5564 machine at a draw speed of 20mm/min for both draw ratios.

3. Results

3.1. Tensile and Bending Properties

Tensile modulus and strength of the PP and its composites in isotropic and oriented stages were measured using an RDP Howden tensile testing machine at 21±2°C. For the oriented materials, the tensile tests were performed on cylindrical samples at diameters of ~5.7 and 3.1mm (the diameters of the die drawn product at 3λ and 10λ respectively). A nominal strain rate of $10^3$ s$^{-1}$ was used for all tests, with a small pre-load applied (up to 7N) in order to ensure that the linear region of the $\sigma$-$\epsilon$ curve was observed. A video extensometer was utilized to measure the displacement generated in the sample. Both the RDP Howden testing machine and the digital camera were linked to a computer running the video extensometer software supplied by RDP Howden to plot the $\sigma$-$\epsilon$ curve and determine the tensile modulus and strength.

Results in Table 1 show the variation in moduli with $\lambda$ for unfilled and filled PP.

<table>
<thead>
<tr>
<th>$\lambda$</th>
<th>Draw temperature ($^\circ$C)</th>
<th>Tensile modulus (GPa)</th>
<th>Specific modulus (GPa m$^3$/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>PP</td>
<td>C120 PP</td>
</tr>
<tr>
<td>1</td>
<td>-</td>
<td>1.4 ± 0.1</td>
<td>1.9 ± 0.1</td>
</tr>
<tr>
<td>3</td>
<td>150</td>
<td>3.5 ± 0.2</td>
<td>3.2 ± 0.2</td>
</tr>
<tr>
<td>10</td>
<td>160</td>
<td>9.4 ± 0.2</td>
<td>8.2 ± 0.3</td>
</tr>
<tr>
<td></td>
<td>165</td>
<td>-</td>
<td>5.7 ± 0.2</td>
</tr>
</tbody>
</table>

Table 1. Tensile Young’s modulus and specific modulus as a function of orientation stage and draw temperature.

The main aim of tensile modulus tests was to establish if, how and which type of the WP reinforce the WPC. As expected, the Young’s modulus for all materials increased with
increasing $\lambda$ because oriented PP chains control and dominate the composite system as shown in Figure 2.

![Figure 2. Transformation of isotropic WPC to highly oriented.](image)

For PP the tensile E-moduli were 1.4, 3.5 and 9.4GPa for the isotropic, $3\lambda$ and $10\lambda$ samples respectively. The C120PP showed slightly lower increase for the oriented states, i.e. 3.2 and 8.2GPa according to the mentioned material state. Isotropic composites C120PP and HB120PP showed higher modulus than the PP indicating the reinforcing effect that vanished for the oriented materials. The HB120 seems to be more effective however when the specific modulus is taken into account the WP types do not reinforce the oriented PP. The HB120PP showed lower modulus than C120PP in the oriented stages. This may be because this material could not be drawn at 160°C, it could only be drawn at 165°C. This enabled it to deform more easily but produced less orientation of the polymer chains resulting in a modulus of 5.4GPa when drawn to 10$\lambda$. When C120PP was drawn under the same temperature it produced a similarly low modulus of 5.7GPa. This suggests that neither the C120 or HB120 show adequate stress transfer nor the stiffness depends more on the draw temperature than the filler type.

Die drawing also affects tensile strength which increased with increasing $\lambda$ as shown in Table 2.

<table>
<thead>
<tr>
<th>$\lambda$</th>
<th>Draw temperature (°C)</th>
<th>PP Tensile strength (MPa)</th>
<th>C120 PP Tensile strength (MPa)</th>
<th>HB120 PP Tensile strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-</td>
<td>15.8 ± 2.0</td>
<td>12.7 ± 0.3</td>
<td>12.8 ± 0.1</td>
</tr>
<tr>
<td>3</td>
<td>150</td>
<td>126.3 ± 4.5</td>
<td>43.2 ± 1.5</td>
<td>41.8 ± 0.6</td>
</tr>
<tr>
<td>10</td>
<td>160</td>
<td>229.3 ± 3.9</td>
<td>126.8 ± 1.8</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>165</td>
<td>125.0 ± 3.6</td>
<td>131.6 ± 2.2</td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Tensile strength and elongation at break.
A small degree of orientation made a considerable increase in tensile strength. For isotropic PP, the tensile strength was $15.8 \pm 2.0$ MPa but for partially oriented material it was almost 8 times higher. Further increase in molecular orientation strengthened the polymeric matrix. Incorporation of 40%w/w of WP into PP results in a reduction of tensile strength for each sample. Orientation of the structure also changed the manner of material failure as shown in Figure 3.

The isotropic materials have linear Hookian region followed by a short plateau before failure. With increasing $\lambda$ the yield point is less marked and the material exhibit a lower failure strain. Although, the stress-strain curves are now almost straight lines the materials do not break in the same way. Failure did not consist of the fracture of the polymer chains, but either in bonding between them or structure defects e.g. filler aggregates or voids leading to shear of the whole layers of the material.

The flexural tests were undertaken following the guidelines of ASTM D790M-93. A three point bending method was chosen to evaluate flexural modulus and strength. All materials were tested as a function of $\lambda$ and span/diameter ratio (L/D). Three ratios were considered: 40, 32 and 16 to 1. A RDP Howden testing machine was employed to take measurements with a strain rate of $10^3$ s$^{-1}$. In general, it can be said that both bending modulus and strength increase with increasing $\lambda$. However when a modulus increases with increasing L/D, the strength decreases as shown in Table 3.

Figure 3. Difference in shape of the $\sigma$-$\epsilon$ curves between unfilled and filled PP as a function of $\lambda$. 

In general, it can be said that both bending modulus and strength increase with increasing $\lambda$. However when a modulus increases with increasing L/D, the strength decreases as shown in Table 3.
The increase in bending modulus with increasing $\lambda$ is especially well seen for PP. For C120PP and HB120PP this feature is recognisable only for highly oriented stage. Surprisingly, the differences in moduli between the isotropic and partially oriented composites are so small for each considered L/D that they might be neglected compared to $10\lambda$. For L/D ratio 40, which should give a value essentially free of shear effects, the flexural moduli are very high i.e. about 6 and 5.5GPa for PP and C120PP respectively. The data indicate that die drawing temperature has also affected the bending modulus. A 5°C increase in the temperature resulted in a large drop in bending modulus; from 5.5 to 3.5GPa for C120PP. As known from the previous results, the addition of C120 or HB120 into PP reduced the strength of the oriented composites. Again, strength increases with $\lambda$, and there are no significant differences in strength between isotropic and partially oriented composites. Moreover, the drawing temperature seems to not have so a large impact on strength and, as for tensile strength, the wood type does not influence the results. During testing, only the isotropic samples broke in a brittle manner. The rest of the materials showed deflections of up to 29mm with permanently deformation their surfaces were free from any trace of failure.

3.2. Thermal Expansion Coefficient ($\alpha$)

Measurement of $\alpha$ for the isotropic and both oriented materials was performed in the temperature range of 15°C to 25°C, parallel ($\alpha_{||}$) and perpendicular ($\alpha_{\perp}$) to the orientation direction. Before the experiment, the samples were conditioned with flowing wet nitrogen for about 30min in order to reach internal equilibrium.
The thermal expansion coefficient of the oriented material depends on the degree of orientation and its direction, however, the latter influences $\alpha$ more significantly as shown in Figure 4. For both materials, it can be seen that $\alpha_\parallel$ remained constant independent of orientation level. However $\alpha_\parallel$ shows dramatic changes as the level of orientation is increased. For the PP it drops from $(9.86\pm0.34) \times 10^{-6}/^\circ C$ for the isotropic material to $(3.41\pm0.26) \times 10^{-6}/^\circ C$ for $3\lambda$ and is too small to be detected using the technique employed here for the material drawn at $10\lambda$. For the C120 filled material $\alpha_\parallel$ dropped from $(6.99\pm0.34) \times 10^{-6}/^\circ C$ for the isotropic stage to $(2.27\pm0.23) \times 10^{-6}/^\circ C$ for the $3\lambda$ and is again too small to detect for the $10\lambda$ material.

![Figure 4](image-url)  
Figure 4. Influence of die drawing direction and C120 on thermal expansion coefficient.

4. Modelling
A key aim of this project is to understand, within the numerical modelling framework, the links between the components as a function of each processing stage and consequently the effect of the wood fibres on the properties of the oriented product. In order to do this and to allow accurate modelling of the composite stiffness, it is necessary to know the bulk density of the wood powders and their aspect ratio at various processing stages.

4.1. Wood Powder Density ($\rho$) and Volume Fraction ($V_f$)
Densities of both particle types were monitored with changing structure. Knowing the proper value of filler density and also its volume fraction is a basic and crucial issue for calculating the composite stiffness.
An approach used to predict the density of the WP consisted in compressing the WP in a cylindrical mould of known dimensions. A piston was pressed into the mould until a pressure of 2000 psi (13.8MPa) was achieved. The volume of the mould was then calculated and densities of $890\text{kg/m}^3$ for C120 and $900\text{ kg/m}^3$ for HB120 respectively were achieved. It can be thought that the densities are too low due to inherent porosity of the particles and the empty spaces between them. However, in a real composite the WP have to be consider in the macroscopic scale because the PP is not able to penetrate the WP structure. It covers the filler and closes (blocks) the voids of the WP. Consequently, the WP looks like a ‘capsule’ with plenty voids inside.
The fibre volume fraction \( V_f \) described by equation (1) can be calculated

\[
V_f = \frac{m_f \rho_m}{m_f \rho_m + \rho_f m_f}
\]  

(1)

where \((m)\) is the mass of the component and \((\rho)\) its density, \((f)\) fibres, \((m)\) – matrix giving the \( V_f \) of 0.402 and 0.398 for the C120 and HB120 respectively.

### 4.2. Aspect Ratio \((A_r)\) of Wood Powder

The \( A_r \) of filler is of great importance to the final product. It is very difficult to evaluate and define by a simple number. This is because the particles are 3D objects and very often irregular in shape. There are a variety of methods for calculating particle size [6] but image analysis is one of the most commonly used. In the present study a Sysmex FPIA 2100 Image Analyzer was used which, in brief, characterises a particle by the diameter of a circle \( D \) with the same area as the area of the particle. In addition, by dividing the particle circumference into the circle circumference it gives a circularity value \( c \). However, the apparatus does not give direct information about \( A_r \). A model developed by us at the University of Leeds was employed to extract this data.

To determine if \( A_r \) is changes with degree of structure orientation, the WP extracted from blended and the 10\( \lambda \) composites were analysed. The investigation has not been finished yet but the indications are that the die drawing process decreases the \( A_r \). For C120 it dropped from 9.7 to 8.1 for blended and drawn materials. For HB120 the tests have to be repeated because the \( A_r \) appeared much lower than for C120, whilst ESEM micrographs indicate that the \( A_r \) is similar to that of the C120.

### 4.3. Cox – Krenchel Model

The Cox-Krenchel model was used to predict the stiffness of the isotropic and oriented WPC. The model follows the rule of mixtures approach and can be describe by equation (2)

\[
Ec = \eta_o \eta_L E_f V_f + E_m (1 - V_f)
\]  

(2)

where

\[
\eta_o = \sum_n a_n \cos^4 \Theta \quad \text{where} \quad \sum_n a_n = 1
\]  

(3)

\[
\eta_o = 3/8 \quad \text{for a random two dimensional (2D) fibre orientation} \quad (3a)
\]

\[
\eta_o = 1/5 \quad \text{for a random three dimensional (3D) fibre orientation} \quad (3b)
\]

\[
\eta_L = \left(1 - \frac{\tanh(\beta A_r)}{\beta A_r}\right)
\]  

(4)

and for the hexagonal packing system of the filler
\[
\beta^2 = \frac{2G_m}{E_f \ln \left( \frac{2\pi V_f}{V_f \sqrt{3}} \right)}
\]

(\eta_o) is Krenchel’s orientation efficiency factor, (\eta_L) is Cox’s length factor, \((E)\) is the modulus, \((V)\) is the volume fraction, and \((m)\) and \((f)\) denote matrix and filler subsequently. Table 4 shows the results of the calculations.

<table>
<thead>
<tr>
<th></th>
<th>isotropic</th>
<th>oriented 3(\lambda)</th>
<th>oriented 10(\lambda)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\rho_f) (kg/m(^3))</td>
<td>890</td>
<td>890</td>
<td>890</td>
</tr>
<tr>
<td>(\rho_m) (kg/m(^3))</td>
<td>897 ± 6</td>
<td>893± 6</td>
<td>896± 5</td>
</tr>
<tr>
<td>(V_f)</td>
<td>0.40</td>
<td>0.40</td>
<td>0.40</td>
</tr>
<tr>
<td>(V_m=1-V_f)</td>
<td>0.60</td>
<td>0.60</td>
<td>0.60</td>
</tr>
<tr>
<td>(E_f) (GPa)</td>
<td>11 ± 1</td>
<td>11 ± 1</td>
<td>11± 1</td>
</tr>
<tr>
<td>(E_m) (GPa)</td>
<td>1.4 ± 0.1</td>
<td>3.5 ± 0.2</td>
<td>9.4 ± 0.2</td>
</tr>
<tr>
<td>(\eta_L)</td>
<td>0.63</td>
<td>0.61</td>
<td>0.59</td>
</tr>
<tr>
<td>(\eta_o)</td>
<td>0.63</td>
<td>0.61</td>
<td>0.59</td>
</tr>
<tr>
<td>(E_c) (GPa)</td>
<td>1.9</td>
<td>3.1</td>
<td>6.6</td>
</tr>
<tr>
<td>theoretical value</td>
<td>(\eta_o = 3/8) (2D)</td>
<td>(\eta_o = 1/5) (3D)</td>
<td>(\eta_o = 0.66)</td>
</tr>
<tr>
<td></td>
<td>(\eta_o = 0.94)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(E_c) (GPa) – experimental value</td>
<td>1.9 ± 0.2</td>
<td>3.2 ± 0.4</td>
<td>8.2 ± 0.3</td>
</tr>
</tbody>
</table>

Table 4. Comparison of the theoretical and experimental moduli for the C120PP.

Using the data presented in Table 4 (Young’s moduli of a solid wood are not presented in this paper, but it was determined as 11GPa) and assuming that the Poisson’s ratio is 0.35 giving the shear modulus for the PP of 0.52GPa, the experimentally determined Young’s modulus of the C120PP could be compared to the predictions given by equation (2). For further details on calculating \(\eta_L\) and \(\eta_o\) in the ‘shear-lag’ analysis see [2]. For the isotropic C120PP there is a very good agreement between the Cox-Krenchel predictions and the experimental data. The isotropic composites showed a random 2D orientation of the WP. For the oriented materials the model understated the theoretical moduli. It has to be mentioned that the experimental modulus for the C120PP oriented at 3\(\lambda\) may be too small because of weak adhesion between the C120 and the PP resulting in the low density of the drawn material.

Ward’s orientation function \((I)\) [5] given as equation (6) was used to predict a degree of the WP orientation in the drawn composite system.
\[ \eta_0 = I = \frac{1}{(1 - K^2)^2} \left[ 1 + \frac{K^2}{2} - \frac{3K}{2(1 - K^2)^{1/2}} \cos^{-1} K \right] \]  

(6)

where \( K = R_A^{-3/2}, (R_A) \) is an actual draw ratio.

\( R_A \), defines as a ratio of the cross-sectional area of the billet to cross-sectional area of the final product, are 3.0±0.4 and 10.2±0.1 for the C120PP drawn at 3\( \lambda \) and 10\( \lambda \) respectively. Using equation (6) the \( I \) orientation function was 0.66 and 0.94 according to the mentioned material state. Substituting these values to equation (2) the theoretical moduli of 3.9GPa for 3\( \lambda \) material and 8.1GPa for 10\( \lambda \) material were achieved. They agree very well with the experimental moduli as shown in Table 3 above.

5. Conclusions

Blending of the C120 and HB120 into polypropylene PP at a loading of 40%w/w has been achieved by twin-screw extrusion. Compression moulded samples were machined into a conical tag used for die drawing. 2-stage orientation was done successfully to 3 and 10\( \lambda \). The orientation of the WP structure has resulted in an increase in tensile and bending properties and decrease in the thermal expansion.

The macroscopic density of 890kg/m\(^3\) and aspect ratio in the rage from 9.1 to 8.1 of the WP were evaluated giving the possibility to use the Cox-Krenchel model to prediction the composite stiffness. For the isotropic C120PP composite the C120 indicated 2D orientation giving the theoretical modulus of 1.9GPa which is in excellent agreement with the experiment value of (1.9±0.2)GPa. For the oriented composites the orientation function \( \eta_0 \) used in the Cox-Krenchel model has not understated the moduli. For this reason, Ward’s orientation function \( I \) was used to link the modulus with the molecular and particles orientation. The function \( I \) is also a very successful tool to predict the stiffness of the WPC in oriented states. The theoretical moduli are 3.9GPa and 8.1GPa for the 3\( \lambda \) and 10\( \lambda \) materials respectively while the experimental values are (3.2±0.3) and (8.2±0.3)GPa.

6. Reference

3. Newson R., Maine F.W., ‘A New Class of Oriented and Expanded Low density Wood Fibre Composites’ in Wood Polymer Symposium, 2005, Bordeaux, France