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Facing up to the Challenges of Natural Fibres as “Potential” Engineering Composite Reinforcements

Jim Thomason

Composites Week @Leuven
September 16-20th 2013
Leuven, Belgium
Natural Fibre Composites

- Jim – why are you so down on Natural Fibre?
  - Some personal NF History
- Some of the NFC Challenges
  - NF Anisotropy
  - NF cross section
- Some Conclusions
Natural Fibre some Personal History

ECCM - 8
EUROPEAN CONFERENCE
ON
COMPOSITE MATERIALS

SCIENCE, TECHNOLOGIES and APPLICATIONS

3 - 6 JUNE 1998  NAPLES - ITALY

Half Day Symposium on Natural Fibre Composites
ECCM-8, Naples 1998

Prof Verpoest also attended

Held standard team meeting
ECCM-8 NF Symposium
The Natural Fibre Conundrum

NF – green, cheap, great properties can replace glass fibre

Hey – Lets make some NF composites and replace glass fibre

Hmmm – my NF composites are nowhere near what I predicted
## Natural Fibre some Personal History

### Potential Advantages of Natural Fibres

- Potentially low cost
- Low density (1.45 g/cc vs 2.6 g/cc for GF)
- Very “green” image
- Incinerable - thermally recyclable (with no net increase in CO2 balance)
- Modulus range exceeds that of E-glass
- Non-abrasive - low wear of processing equipment
- No skin irritation problems during handling

### Identified Disadvantages of Natural Fibres

- Fibre properties dependent on level of processing (high properties require more fibre processing = cost penalty)
- Properties dependent on seasonal conditions
- High levels of water adsorption and poor dimensional stability
- Low strength compared to E-glass
- Anisotropic structure - low transverse properties = poor flex & compression performance
- Composites generally require higher fibre loading resulting in high processing viscosities.
- Surface treatments and polymer coupling agents required for best composite properties
- Odour problem after composite processing
- Potentially Bioactive
Natural Fibre some Personal History

1999-2001 Owens Corning NFC Project

- Based on Long Fibre PP Process Technology
- 12 mm pultruded pellets, 20-50% NF-PP
- Pilot Plant capability 500kg/day
  - plan for first production plant in India
- New Sizings developed - some patented for Natural Fibre and Regenerated Cellulose Fibre (Rayon)
- Multiple Demonstrator Parts Moulded and Tested
- Huge automotive OEM, Tier 1 and Tier 2 interest
## Natural Fibre Composite - Demonstrators

<table>
<thead>
<tr>
<th>Organizer / Moulder</th>
<th>Part</th>
<th>Method</th>
<th>Part Wt. (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>OC, MIG Plastics</td>
<td>Buick - Door handle pull</td>
<td>Inj. Mould</td>
<td>0.14</td>
</tr>
<tr>
<td>JCI</td>
<td>Jeep Grand Cherokee - Door Inner</td>
<td>Inj. Mould</td>
<td>0.91</td>
</tr>
<tr>
<td>Mayco Plastics</td>
<td>Chrysler - Air deflector</td>
<td>Inj. Mould</td>
<td>0.31</td>
</tr>
<tr>
<td>Delphi, Proto Plastics</td>
<td>GM – Instrument panel retainer</td>
<td>Inj. Mould</td>
<td>2.6kg</td>
</tr>
<tr>
<td>SEG Kunststoff technik</td>
<td>Audi A2 - Fender stiffener</td>
<td>Inj. Mould</td>
<td>0.45</td>
</tr>
<tr>
<td>Pelzer, Clion GmbH</td>
<td>DCX PT Cruiser - Underbody shield</td>
<td>Extrusion Compress</td>
<td>1.35</td>
</tr>
</tbody>
</table>
## Typical Properties of PP Based Compounds

<table>
<thead>
<tr>
<th></th>
<th>30% Talc</th>
<th>30% Jute-A</th>
<th>30% Jute-B</th>
<th>30% Glass*</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Modulus (GPa)</strong></td>
<td>3.6</td>
<td>4.1</td>
<td>4.4</td>
<td>6.5</td>
</tr>
<tr>
<td><strong>Tensile (MPa)</strong></td>
<td>30</td>
<td>35</td>
<td>45</td>
<td>45-90</td>
</tr>
<tr>
<td><strong>Flex Str (MPa)</strong></td>
<td>56</td>
<td>56</td>
<td>76</td>
<td>65-145</td>
</tr>
<tr>
<td><strong>N Izod (J/m)</strong></td>
<td>27</td>
<td>59</td>
<td>48</td>
<td>80-110</td>
</tr>
<tr>
<td><strong>Un Izod (J/m)</strong></td>
<td>203</td>
<td>198</td>
<td>177</td>
<td>260-750</td>
</tr>
<tr>
<td><strong>HDT (°C)</strong></td>
<td>92</td>
<td>120</td>
<td>135</td>
<td>155</td>
</tr>
<tr>
<td><strong>Density</strong></td>
<td>1.13</td>
<td>1.01</td>
<td>1.01</td>
<td>1.13</td>
</tr>
</tbody>
</table>
Comparison PP Composite Performance

**Short Glass**

- Modulus
- Strength
- Notched
- Unnotched

**Long Jute**

- Modulus
- Strength
- Notched
- Unnotched

**Long Glass**

- Modulus
- Strength
- Notched
- Unnotched

**Long Rayon**

- Modulus
- Strength
- Notched
- Unnotched

<table>
<thead>
<tr>
<th>Fibre Content (% weight)</th>
<th>Relative Performance (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>10</td>
<td>0</td>
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<tr>
<td>20</td>
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<td>30</td>
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<td>40</td>
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<td>50</td>
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<td>60</td>
<td>0</td>
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<tr>
<td>70</td>
<td>0</td>
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<td>80</td>
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<td>90</td>
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<td>350</td>
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<td>360</td>
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<td>370</td>
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<td>380</td>
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<tr>
<td>390</td>
<td>0</td>
</tr>
<tr>
<td>400</td>
<td>0</td>
</tr>
</tbody>
</table>

The graphs illustrate the relative performance of different materials under various conditions, showcasing the Modulus, Strength, Notched, and Unnotched performance across different fibre contents.
Comparison Composite Cost/Performance

**Strength**

- Jute
  - Notched Impact: $0.01$ to $0.02$
  - Unnotched Impact: $0.03$

- Natural Fiber Input:
  - 20% NF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$
  - 40% NF: $0.05$, $0.10$, $0.15$, $0.20$, $0.25$, $0.30$

- Natural Fiber Input:
  - 20% Talc: $0.4$, $0.5$, $0.6$, $0.7$, $0.8$
  - 40% Talc: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% GF: $0.10$, $0.15$, $0.20$
  - 40% GF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

**Unnotched Impact**

- Natural Fiber Input:
  - 20% NF: $0.10$, $0.15$, $0.20$
  - 40% NF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% GF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% Talc: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

**Modulus**

- Natural Fiber Input:
  - 20% NF: $0.10$, $0.15$, $0.20$
  - 40% NF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% GF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% Talc: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

**Notched Impact**

- Natural Fiber Input:
  - 20% NF: $0.10$, $0.15$, $0.20$
  - 40% NF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% GF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% Talc: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

**Talc>2**

- Natural Fiber Input:
  - 20% NF: $0.10$, $0.15$, $0.20$
  - 40% NF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% GF: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$

- Natural Fiber Input:
  - 20% Talc: $0.4$, $0.6$, $0.8$, $1.0$, $1.2$, $1.4$
Natural Fibre some Personal History
1999-2001 Owens Corning NFC Project

Project shelved 2002,
Natural Fibre Composites are not
Performance-Cost competitive with existing materials
Some Philosophy

- If you know your enemies and know yourself, you can win a hundred battles without a single loss.

- If you only know yourself, but not your opponent, you may win or may lose.

- If you know neither yourself nor your enemy, you will always endanger yourself.

_The Art of War_, Sun Tzu
Some of the Challenges of Working with NF

Fibre natural variability
Fibre highly anisotropic
Low transverse and shear reinforcement performance
Fibres mostly non-circular
Fibre lumen = composite voids
Fibre cross section non-uniform along length
Fibre “diameter” often much larger than man-made fibres
High moisture content in fibres at ambient RH – processing issues
Temperature sensitivity – in particular odour issues in processing
Many forms of NF not suitable for use in standard industry processes

Poor (often negative) performance in composites
Composite fibre content measurement?
Moisture sensitivity in composite
Bio-activity (rotting, fungus, mould)
Fibre-matrix interaction - poor
Fogging/Emission issues in Automotive applications
Why Natural Fibre Composites?

Some typical fibre properties are shown in the Table below.

<table>
<thead>
<tr>
<th></th>
<th>Sisal</th>
<th>Jute</th>
<th>Flax</th>
<th>Glass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modulus (GPa)</td>
<td>17-28</td>
<td>20-45</td>
<td>27-70</td>
<td>75</td>
</tr>
<tr>
<td>Strength (GPa)</td>
<td>0.1-0.8</td>
<td>0.2-0.9</td>
<td>0.3-0.9</td>
<td>&gt;1.5</td>
</tr>
<tr>
<td>Density</td>
<td>1.3</td>
<td>1.3</td>
<td>1.5</td>
<td>2.6</td>
</tr>
<tr>
<td>Specific Modulus</td>
<td>13-21</td>
<td>15-35</td>
<td>18-47</td>
<td>29</td>
</tr>
</tbody>
</table>

So some natural fibre may have the potential to replace glass fibres???

\[ E_C = \eta_0 \eta_L V_f E_f + V_m E_m \]
Typical Specs for Automotive Application

There are very few applications where only modulus is required!!!
A typical automotive spec sheet will need –

- Melt Flow Rate (ISO 1133, ASTM D1238)
- Glass Fibre Content (ISO 3451/1)
- Density (ISO 1183, ASTM D792)
- Tensile Strength (ISO R527, ASTM D638M)
- Flexural Modulus (ISO 178, ASTM D790M)
- Shear Modulus (ASTM D4065)
- Impact Strength, Izod (ISO 180, ASTM D256)
- Heat Deflection temperature (ISO 73, ASTM D648)
- Heat Aging Performance (ISO 188, ASTM D573)
- Flammability (ISO 3795)
- Fogging (FLTM BO 116-03)
- Mould Shrinkage (ISO 2577)
- Coeff. of Linear Thermal Expansion (ASTM D696)
Comparison Predicted Composite Modulus

For injection moulded long fibre polypropylene

Remember – comparison on weight content (i.e. specific fibre properties) means NO weight saving advantage!

- Glass Fibre
- NF 20 GPa (Sisal)
- NF 40 GPa (Jute)
- NF 60 GPa (Flax)
Thermoelastic Anisotropy of Flax and Sisal Fibres

• Goal
  – Quantify anisotropy of Flax & Sisal fibres
  – Full thermoelastic characterisation

• Measure
  – UD fibre-epoxy laminates $E(\theta,T), G_{12}, \nu_{12}, \nu_{21}, \alpha(\theta,T)$
  – Epoxy matrix $E_m(T), \nu_m, \alpha_m(T)$
  – Laminate fibre volume fraction ?
  – Flax & Sisal fibre $E_{1f}(T)$ (fibre cross section ?)

• Calculate
  – $E_{1f}(T), E_{2f}(T), G_{12f}(T), \nu_{12f}(T), \alpha_{1f}(T), \alpha_{2f}(T)$
Composite DMA Results

![Graph showing Log Storage Modulus (Pa) vs Temperature (°C) for different Sisal and Resin compositions.](image)
Anisotropy of Fibre Modulus

Temperature (°C)

Modulus (GPa)

-60 -40 -20 0 20 40 60

Flax E1
Flax E2
Sisal E1
Sisal E2
Flax G12
Sisal G12
Composite Thermal Strain

Temperature (°C)

Thermal Strain (mm/m)

- Epoxy
- Flax 90
- Flax 65
- Flax 45
- Flax 25
- Flax 0
Fibre Expansion Coefficients

Fibre Transverse

Fibre Axial

CLTE mm/m°C vs Temperature (°C)

- Sisal Transverse
- Flax Transverse
- Sisal Axial
- Flax Axial
## Summary Thermo-Mechanical Properties NF

<table>
<thead>
<tr>
<th></th>
<th>Glass</th>
<th>Flax</th>
<th>Sisal</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Longitudinal Modulus (GPa)</strong></td>
<td>75</td>
<td>61.5</td>
<td>24.9</td>
</tr>
<tr>
<td><strong>Transverse Modulus (GPa)</strong></td>
<td>75</td>
<td>1.2</td>
<td>1.6</td>
</tr>
<tr>
<td><strong>Shear Modulus (GPa)</strong></td>
<td>30</td>
<td>1.7</td>
<td>1.1</td>
</tr>
<tr>
<td><strong>Axial LCTE (μm/m.°C)</strong></td>
<td>5</td>
<td>-7.3</td>
<td>-2.7</td>
</tr>
<tr>
<td><strong>Transverse LCTE (μm/m.°C)</strong></td>
<td>5</td>
<td>71</td>
<td>73</td>
</tr>
</tbody>
</table>
Fibre Stress = Load/Area = $P/A_f \ ( = 4P/\pi D_f^2 \ ???)$
Single Fibre Cross Section Area

- $A_f$ in single fibre testing is almost universally evaluated from $D_f$ using a transverse image of fibre and assumption of circular cross-section
- Is this acceptable for Natural Fibres??
Single Fibre CSA Measurements

1. Single fibre “diameter” determined by averaging 4 transverse measurements at 2 mm intervals

2. Fibres embedded, cut and polished

3. “true” cross sectional area determined at approximately the same position on fibre

4. Sample ground down 2 mm and polished

5. Steps 3-4 repeated 10x
Single Fibre Modulus

\[ \frac{1}{E^*_f} = \frac{1}{E_f} + C \frac{A_f}{L_0} \]

Sisal, \( \frac{1000}{33} = 30 \) GPa

Flax, \( \frac{1000}{14.1} = 71 \) GPa
Natural Fibre CSA Evaluation

“Diameter” CSA/True CSA vs. Average “Diameter” (mm)

- Flax
- Sisal
Natural Fibre CSA Evaluation

- “Diameter” method significantly overestimates CSA
- Underestimates single fibre modulus and strength
- Magnitude of error is “diameter” dependent
### Effect CSA on Single Fibre Properties

<table>
<thead>
<tr>
<th>CSA method</th>
<th>Diameter</th>
<th>Actual</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flax Strength (MPa)</td>
<td>293</td>
<td>688</td>
</tr>
<tr>
<td>Sisal Strength (MPa)</td>
<td>255</td>
<td>530</td>
</tr>
<tr>
<td>Flax Modulus (GPa)</td>
<td>36</td>
<td>71</td>
</tr>
<tr>
<td>Sisal Modulus (GPa)</td>
<td>20</td>
<td>30</td>
</tr>
</tbody>
</table>
Effect of “Diameter” CSA on Apparent NF Modulus

Assume a diameter independent modulus

- Flax, $E_{1f} = 71.0$ GPa
- Sisal, $E_{1f} = 30.5$ GPa
Simple Model of NF CSA “Diameter” Errors

NF non-circular – simplest model is oval X-section
Simple Model of NF CSA “Diameter” Errors

Due to NF natural twist the oval cross section will be viewed differently at different positions along the fibre.

Transverse view from microscope
Parameteric Ellipse Analysis

True CSA = 0.25\(\pi\)AB

"Diameter" CSA = 0.25\(\pi\)D^2

\[X(t) = 0.5ACos(t)Cos(\phi) - 0.5BSin(t)Sin(\phi)\]

Can solve for \(X_{\text{max}}\) for any \(\phi\) and then average over \(\phi=0-90^\circ\) for different A:B ratios.
CSA Ratio from Ellipse Analysis

Av CSA Ratio
- A/B 5 2.6
- A/B 3 1.7
- A/B 2 1.3
- A/B 1 1.0

Ellipse Major Axis Orientation Angle

CSA Ratio (D^2/AB)
Natural Fibre CSA Evaluation

'Diameter' CSA/True CSA

Average Fibre "Diameter" (mm)

- Flax measured
- Sisal measured
Natural Fibre CSA Evaluation

Lines of fixed CSA and varying ellipse A:B ratio

'\text{Diameter}''\text{CSA}/\text{True CSA}

Average Fibre "Diameter" (mm)

- Flax measured
- Sisal measured
- Flax thin
- Flax average
- Flax thick
- Sisal thin
- Sisal average
- Sisal thick
Other Fibres

Abaca

Coir

Kenaf

Jute
Similar issues probable in CSA estimation from fibre “diameter”
### Natural Fibre CSA Evaluation

<table>
<thead>
<tr>
<th></th>
<th>Average CSA (mm²)</th>
<th>Diameter CSA / True CSA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sisal</td>
<td>0.0272</td>
<td>1.97</td>
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<tr>
<td>Flax</td>
<td>0.0125</td>
<td>2.55</td>
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<td>Jute</td>
<td>0.0032</td>
<td>1.58</td>
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<td>Hemp</td>
<td>0.0058</td>
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<td>Kenaf</td>
<td>0.0061</td>
<td>1.71</td>
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<td>Abaca</td>
<td>0.0213</td>
<td>1.31</td>
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<td>Coir</td>
<td>0.0283</td>
<td>1.41</td>
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</table>
What does this anisotropy mean for the reinforcement performance of natural fibres?

\[ E_C = \eta_0 \eta_L V_f E_f + V_m E_m \]

• Comparison NF and GF often “assumes” isotropic fibre

• Hence simple Krenchel analysis for \( \eta_0 \)

\[ \eta_0 = \cos^4(\theta) \]

• NF is more like an orthotropic composite material

• Apply laminate theory to model reinforcement performance
Engineering Stiffness, Off-axis Orthotropic Lamina

\[ E_x = \frac{\sigma_x}{\varepsilon_x} \quad \varepsilon_{xy} = \bar{S}\sigma_{xy} \quad \text{set } \sigma_{xy} = \{\sigma_x \ 0 \ 0\} \]

\[
\begin{bmatrix}
\varepsilon_x \\
\varepsilon_y \\
\gamma_{xy}
\end{bmatrix} =
\begin{bmatrix}
\bar{S}_{11} & \bar{S}_{12} & \bar{S}_{13} \\
\bar{S}_{21} & \bar{S}_{22} & \bar{S}_{23} \\
\bar{S}_{31} & \bar{S}_{32} & \bar{S}_{33}
\end{bmatrix}
\begin{bmatrix}
\sigma_x \\
0 \\
0
\end{bmatrix}

\text{hence } \varepsilon_x = \bar{S}_{11}\sigma_x
\]

and for all \( \theta \), \( E_x = \frac{1}{\bar{S}_{11}} \)

\[ \bar{S}_{11} = S_{11}\cos^4\theta + (2S_{12} + S_{33})\sin^2\theta\cos^2\theta + S_{22}\sin^4\theta \]

The terms \( S_{11} \), etc., are found from \( S = \)

\[
\begin{bmatrix}
\frac{1}{E_{11}} & -\frac{v_{21}}{E_{22}} & 0 \\
-\frac{v_{12}}{E_{11}} & \frac{1}{E_{22}} & 0 \\
0 & 0 & \frac{1}{G_{12}}
\end{bmatrix}
\]
Offaxis Stiffness Contribution of Anisotropic Fibre

Fibre Modulus Contribution vs. Off-axis Angle (°)

- Flax Krenchel
- Flax "Laminate"
- Sisal Krenchel
- Sisal "Laminate"
Off-axis Stiffness Contribution of Anisotropic Fibre

Integration of curves gives an average orientation factor for RoM random in-plane “GMT” Krenchel \( \eta_0 = 0.375 \) “Laminate” \( \eta_0 = 0.2 \)
Comparison Predicted Composite Modulus

For Randon Inplane moulded long fibre polypropylene “GMT”

- Glass Fibre
- NF 20 GPa (Sisal)
- NF 40 GPa (Jute)
- NF 60 GPa (Flax)

Modulus (GPa) vs Fibre Content (% weight)

Krenchel
Comparison Predicted Composite Modulus

For Randon Inplane moulded long fibre polypropylene “GMT”

- **Glass Fibre**
- **NF 20 GPa (Sisal)**
- **NF 40 GPa (Jute)**
- **NF 60 GPa (Flax)**

The graph shows the comparison of predicted composite modulus for different fibre contents in terms of percentage weight. The modulus values range from 0 to 6 GPa for glass fibre, and from 0 to 4 GPa for other fibres. The fibre content is marked from 0% to 40% on the x-axis.
NF Anisotropy Challenge

OK - Let's just make Unidirectional Composites?

Actual contribution 40-50% less than expected at only 10°
- how "unidirectional" and non-wavy can you make your UD NF composites?
Conclusions (1)

• Estimation of natural fibre cross section area via the ‘diameter’ method leads to significant overestimation of CSA.
  – results in significant underestimation of mechanical properties obtained by single fibre testing.
  – also contributes significantly to the variability observed in the measurement of natural fibres properties.
  – since the magnitude of the CSA error is “diameter” dependent – single fibre properties will appear to be diameter dependent.
Conclusions (2)

- Flax and Sisal fibres exhibit very high levels of mechanical and thermomechanical anisotropy.

- Ignoring natural fibre anisotropy and using only the axial modulus of natural fibres in estimating their composite reinforcing ability will significantly overestimate their potential in any off-axis composite loading scenario.
Announcement
Sustainable Composites

In August 2013 the Advanced Composites Group at the University of Strathclyde filed its first patent application in the area of

Glass Fibre Recovery

covering cost effective, industrially applicable, treatments to regenerate the strength of thermally recycled glass fibres.