Fabrication par Procédé d’Imprégnation Directe de Composites Structuraux à Renfort Textile en Fibres Végétales et à Matrices Thermorétractable ou Thermoplastique

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Evolution of Industrial Applications

Performance → Cost → Environnement

Biomedical

Aerospace

Military

Commercial aircraft

Mass transport

Automotive

Construction

Importance of performance

~20C

Importance of cost

Today

Tomorrow

Environment
Biosourced Composites, Natural Fibers

Flax (lin) and Hemp (chanvre) are comparable with glass fiber in terms of specific stiffness.
State-of-the-Art: “Green Washing!”

- Semi-structural (non-structural) parts
  - PP or PLA (with low $P_{\text{melt}}$)
  - LFRT
  - SFRT

- High performance structural parts
  - Textile

✅ Enhance mechanical properties: textile reinforcement (high fiber length and content)
✅ Avoid thermal degradation (even for engineering polymer with high $T_{\text{melt}}$)
✅ Decrease the manufacturing cost: direct impregnation without semi-products

Clusters: Projets Structurants de Pôles de Compétitivité (PSPC)

- Project SINFONI
  - Hemp Chanvre
- Project FIABILIN
  - Flax Lin

Positioning of each fiber (flax and hemp) as the third technical fiber for industrial applications (after glass and carbon fibers)
Flax Textile Reinforced Thermoset Composites

- Resin Transfer Molding process
  - Resin Injection
  - Heating
  - Mold Filling & Curing
  - Releasing

- Resin flow: Darcy’s law
  \[ \frac{Q}{A} = u_D = -\frac{K}{\mu} \nabla P \]
  - \( Q \): flow rate
  - \( A \): cross section
  - \( u_D \): volume-averaged velocity
  - \( P \): resin pressure
  - \( \mu \): resin viscosity
  - \( K \): permeability of fiber reinforcement

- Continuous textile reinforcement at high \( V_f \): enhance the mechanical properties
- Low processing temperature: avoid the thermal degradation of flax fiber
- Long process cycle time (due to long resin curing time)
Different Scales in Natural Fibers

- **Liquid absorption**
  - Liquid penetration into the fiber

- **Fiber swell**
  - Increase of fiber diameter

*Flax fiber in contact with the liquid resin*
Wetting: Improvement? Characterization!

Capillary radius changes due to fiber swell & liquid absorption into fiber

- Surface tension: 48.29 mN/m
- Contact angle: ~36° (with oil) << 90°
**Permeability of Natural Fiber Textile**

- **Unsaturated permeability**: time-dependent fiber swell, non-uniform permeability

- **Saturated permeability** ($K_{sat}$)

  \[ \bar{u}_{Darcy} = -\frac{K}{\mu} \frac{dP}{dx} \]
  \[ Q = \frac{K}{\mu} \frac{P_1 - P_2}{L} \]
  \[ K_{sat} = \frac{Q \mu L}{A(P_1 - P_2)} \]

**Two test liquids with different fiber swell ratios**

- $f_{sw,oil} < f_{sw,water}$
- $K_{oil} > K_{water}$

<table>
<thead>
<tr>
<th>Fiber Volume Fraction</th>
<th>Permeability [log10(m^2)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.35</td>
<td>-13</td>
</tr>
<tr>
<td>0.4</td>
<td>-12.5</td>
</tr>
<tr>
<td>0.45</td>
<td>-12</td>
</tr>
<tr>
<td>0.5</td>
<td>-11.5</td>
</tr>
<tr>
<td>0.55</td>
<td>-11</td>
</tr>
<tr>
<td>0.6</td>
<td>-10.5</td>
</tr>
</tbody>
</table>

- Model, Water: $k=f(V_f)$
- Model, Oil: $k=f(V_f)$
- Model, Water: $k=0.60$
- Model, Oil: $k=0.6$

Experiment, Water
Experiment, Oil
Permeability of Natural Fiber Textile

\[ K(V_{f,\text{eff}} = f_{sw}^2 V_f) \]

\[ K(V_{f,dry}, f_{sw}) \]

\[ K = \frac{1}{A} \left( 1 - f_{sw}^2 V_f \right)^{n+1} \]

\[ f_{sw} = f(t) \]

\[ n=1.2855 \]
\[ A=1.7967 \times 10^{10} \]
Resin Flow Modeling

Liquid absorption

Mass conservation

\[ Q_{in} = Q_{absorp} + Q_{out} \]

Permeability

\[ \vec{u} = -\frac{K}{\mu} \nabla P \]

Fiber swell

Dry fiber

\[ D_f (\text{small}) \]
\[ h (\text{big}) \]

Wet fiber

\[ D_f (\text{big}) \]
\[ h (\text{small}) \]
Dependence on Fiber Content and Flow Length

Flow front position square vs. time

\[
\frac{\partial}{\partial x^*} \left( \frac{\partial P^*}{\partial x^*} \right) = \mu \frac{x_f^2 V_f}{K(x,t) P_{in} (1-V_f)} \left( \frac{\rho_f}{\rho_l} C_R \frac{\partial f_{so}(t)}{\partial t} - \frac{\partial f_{sw}^2(t)}{\partial t} \right)
\]

\( V_f = 0.34 \)

\( X_f = \frac{(1-V_f) \cdot \mu}{P_{in}} \cdot x_f^2 = 2K \cdot t \)

Mass sink effect (delaying the flow front progress) increases as fiber content and part size increase.

\( V_f = 0.56 \)

Drooping by sink effect

Experiment, \( V_{f,0} = 0.343 \)

Experiment, \( V_{f,0} = 0.345 \)

Experiment, \( V_{f,0} = 0.564 \)

Experiment, \( V_{f,0} = 0.564 \)
Thermoplastic Matrices

- **Decrease the manufacturing cost**
  - Short process cycle time (< 2 minutes for automotive applications)
  - Direct impregnation without semi-products (commingled yarn, powder impregnated fabric, film stacking)

- **Adopt high performance engineering polymers (e.g. Polyamide)**
  - Avoid the thermal degradation of natural fibers: $T_{\text{degrad}} (170 °C) < T_{\text{melt}}$

Air voids or porosities due to poor impregnation, poor fiber-matrix compatibility, resin shrinkage or moisture dissolution

Minimize the fiber exposure to heat

Maximize the local pressure

Temperature & Time

Temperature & Time

Process window

Thermal degradation

Poor impregnation (Void/porosity)
Compression Resin Transfer Molding

Short cycle time

Fiber placing → Resin injection → Compression

RTM
Longitudinal flow

\[ \bar{u} \approx \frac{K_{tow}}{\mu} \left( \frac{P_{inj}}{L} - \frac{P_{cap}}{l} \right) \ll 1, \]
\[ P_{cap} = -\frac{\gamma \cos \theta}{D_{pore}} \]

CRTM
Transverse flow

Maximize resin velocity inside the tow

\[ \bar{u} \approx \frac{K_{tow}}{\mu} \left( \frac{P_{comp}}{H} - \frac{P_{cap}}{l} \right) \gg 1, \]
\[ P_{cap} = -\frac{\gamma \cos \theta}{D_{pore}} \]
Manufacturing Process Scheme

Isothermal mold filling to avoid the temperature drop during the impregnation

1. Preform preheating
   \[ T_{fiber} = T_{high} \]

2. Resin injection and preform placement
   \[ T_{mold} = T_{fiber} = T_{press} = T_{resin} = T_{high} \]

3. Mold closing and holding
   \[ T_{mold} = T_{fiber} = T_{press} = T_{resin} = T_{high} \]

4. Cooling
   \[ T_{composite} = T_{mold} = T_{high} \rightarrow T_{composite} = T_{press} = T_{low} \]

Two presses at different temperature (hot and cool) to reduce the cooling time
Process Conditions

1. Temperature: 195, 205 °C
2. Preheating: 2 min
3. Impregnation time: 60 s, 90 s
4. Pressure condition: 1.5 bar (40 s, 70 s), 21 bar (20 s)
5. Materials: Flax 2×2 twill textile NATTEX 600 + Reactive bio PA
6. Fiber volume fraction ~ 42%, Thickness ~ 1.8 mm
Impregnation Quality

The thermal degradation of flax fiber was not observed.

<table>
<thead>
<tr>
<th>Compression : 60 sec</th>
<th>Compression : 90 sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>Good impregnation</td>
<td>Good impregnation</td>
</tr>
<tr>
<td>Incomplete impregnation</td>
<td>Good impregnation</td>
</tr>
</tbody>
</table>

195°C

205°C
Mechanical Properties

- Mechanical properties obtained by tensile tests
  \(V_f \sim 42\%\), average value of \#1,2 and \#2,1
  - Average tensile modulus: \(13.5 \pm 1.9\) GPa
  - Average tensile strength: \(130 \pm 22\) MPa

<table>
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<tr>
<th>Condition</th>
<th>Cooling</th>
<th>Fiber preheating</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1,1</td>
<td>Slow</td>
<td>In the oven</td>
</tr>
<tr>
<td>#1,2, #2,1</td>
<td>Fast</td>
<td>In the oven</td>
</tr>
<tr>
<td>#2,2</td>
<td>Fast</td>
<td>In the mold</td>
</tr>
</tbody>
</table>

Film stacking, PA 11\(V_f:39\%\): 11.9 GPa ±0.8

Film stacking, PA 11\(V_f:39\%\): 107 ±6.5 MPa
Conclusions

- Natural fiber textile reinforcements should be employed for high performance structural applications.
- The resin flow in the natural fiber reinforcement induces resin absorption and fiber swell: influence on the process cycle time and the final part quality.
- Direct thermoplastic melt impregnation into natural fiber reinforcement within a short cycle time is feasible without the thermal degradation of fibers.
- Industrial applications of natural fiber textile should be carefully selected by considering the manufacturing process, the matrix type and the pros/cons of reinforcement.
  - Aeronautics / Railway / Automotive
  - Process cycle time / Material price (natural fiber textile are not cheap!)
  - Multi-functionality: Aesthetics / Thermal & acoustic isolation / Recyclability