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

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Evolution of Industrial Applications

Performance → Cost → Environnement

Biomedical

Aerospace

Military

~20C

Commercial aircraft

Today

Mass transport

Automotive

Construction

Tomorrow

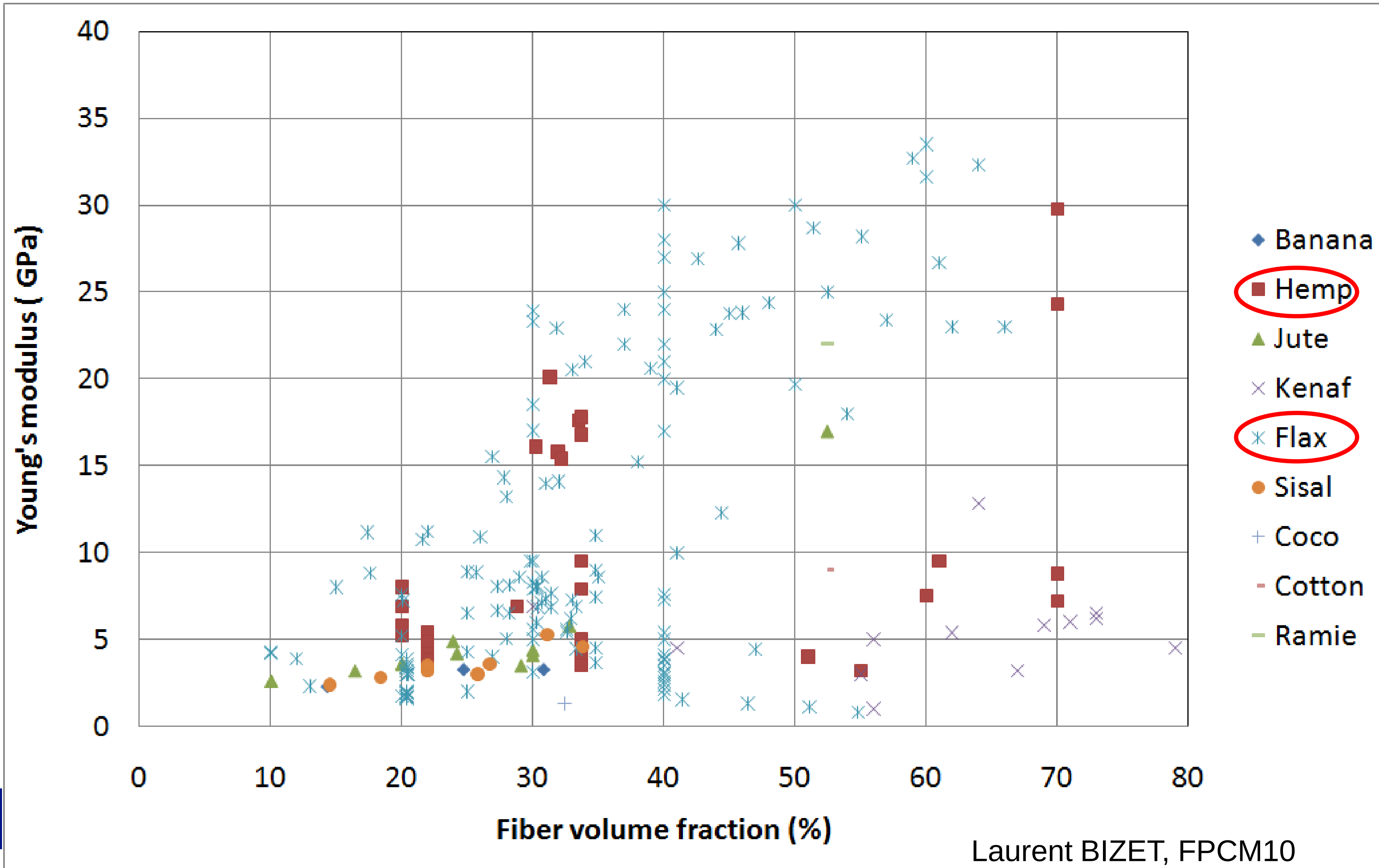
Environment

Importance of performance

Importance of cost

Biosourced Composites, Natural Fibers

Flax (lin) and Hemp (chanvre) are comparable with glass fiber in terms of specific stiffness.

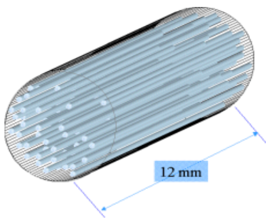


Laurent BIZET, FPCM10

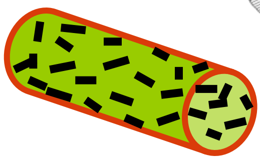
State-of-the-Art : " Green Washing !"

- **Semi-structural (non-structural) parts**

PP or PLA
(with low P_{melt})



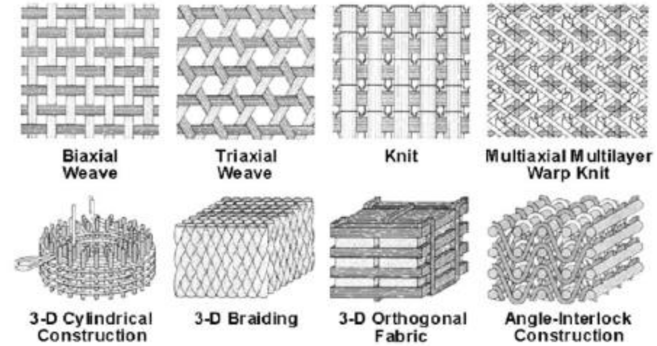
LFRT



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_{melt})
✓ Decrease the manufacturing cost: direct impregnation without semi-products

Clusters: Projets Structurants de Pôles de Compétitivité (PSPC)

INVESTISSEMENTS D'AVENIR

Mines Douai / TPCIM

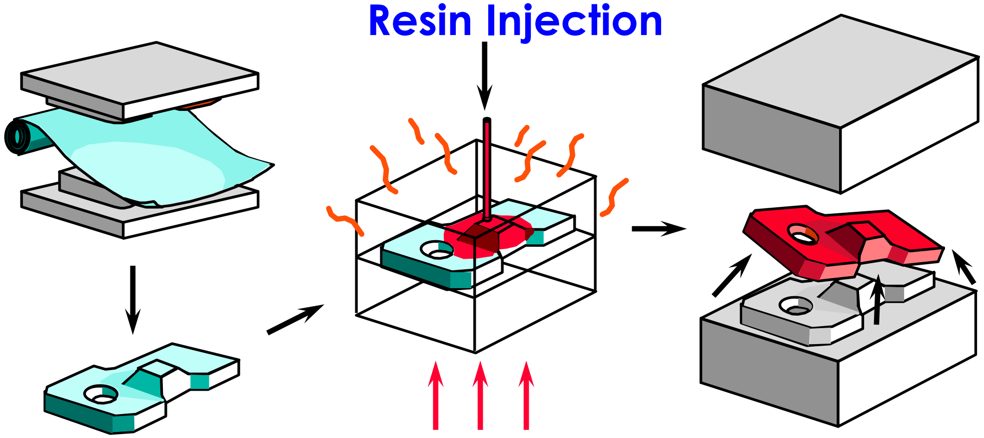
Project SINFONI	**Hemp** *Chanvre*	
Project FIABILIN	**Flax** *Lin*	

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

MINES Douai

Flax Textile Reinforced Thermoset Composites

- **Resin Transfer Molding process**



- **Resin flow: Darcy's law**

$$\frac{Q}{\mathbf{A}} = u_D = -\frac{\mathbf{K}}{\mu}\nabla P$$

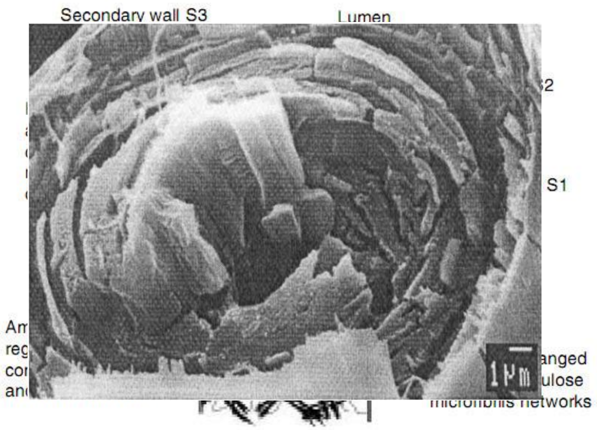
Fiber reinforcement (K)

Resin (μ)

Q: flow rate
A: cross section
u_D: volume-averaged velocity
P: resin pressure
μ: 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

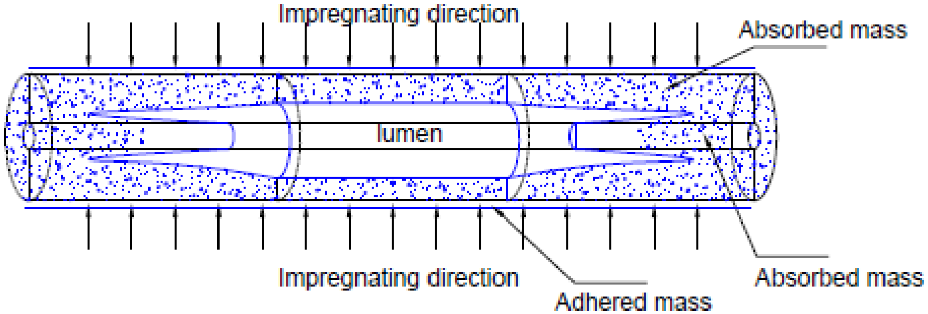


Porous elementary fiber

Flax fiber in contact with the liquid resin

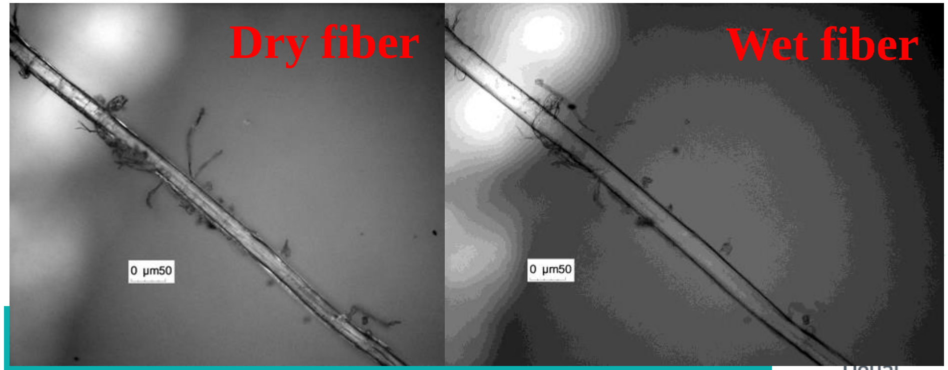
- **Liquid absorption**

Liquid penetration into the fiber



- **Fiber swell**

Increase of fiber diameter

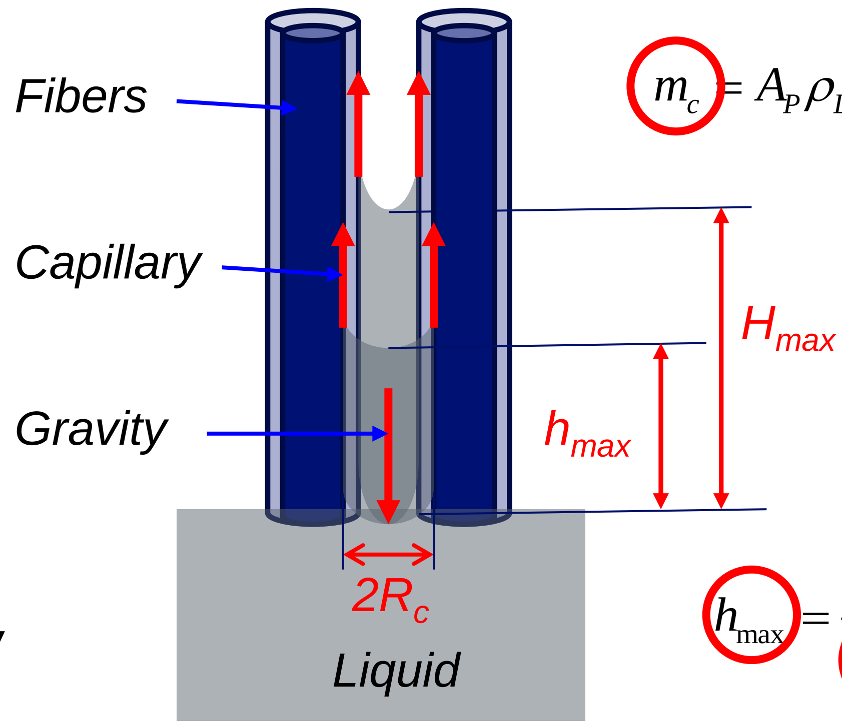


Wetting: Improvement? Characterization!

Poor wetting

Good wetting

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



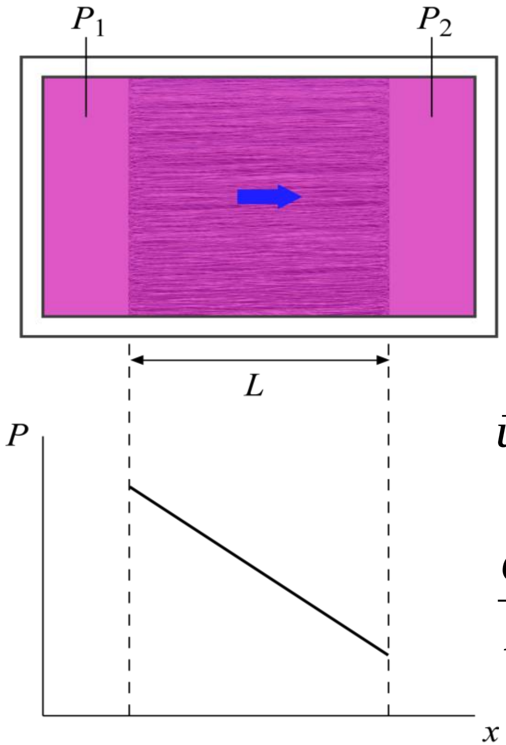
$$m_c = A_P \rho_L \sqrt{\frac{\gamma_L R_c \cos\theta}{2\mu} t}$$

$$h_{max} = \frac{2\gamma_L \cos\theta}{R_c \rho_L g}$$

- ✓ 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})**



Steady flow at maximum fiber swell (uniform in the preform)

$$\vec{u}_{Darcy} = -\frac{\mathbf{K}}{\mu}\frac{dP}{dx}$$

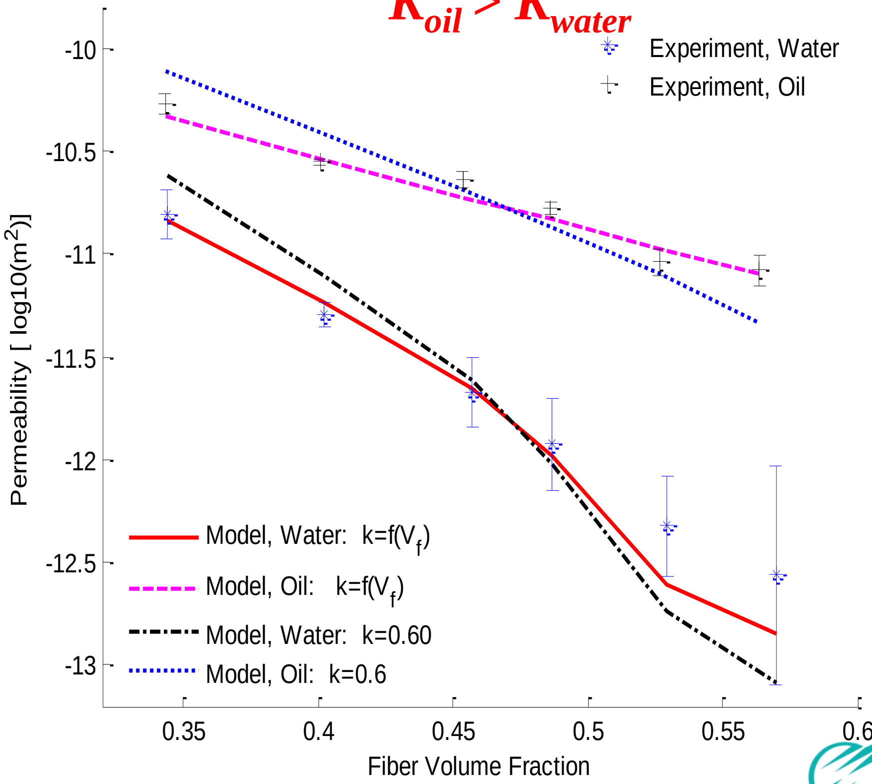
$$\frac{Q}{A} = \frac{\mathbf{K}}{\mu}\frac{P_1 - P_2}{L}$$

$$\mathbf{K}_{sat} = \frac{Q\mu L}{A(P_1 - P_2)}$$

Two test liquids with different fiber swell ratios

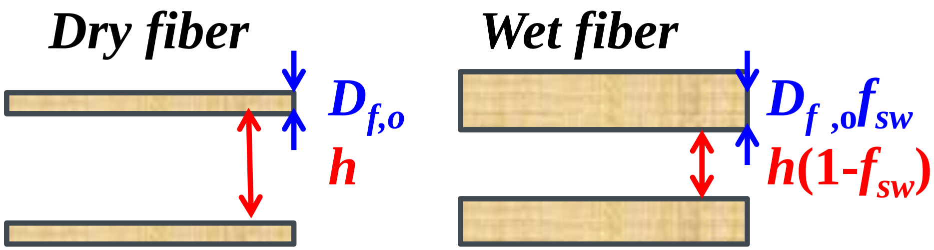
$f_{sw,oil} < f_{sw,water}$

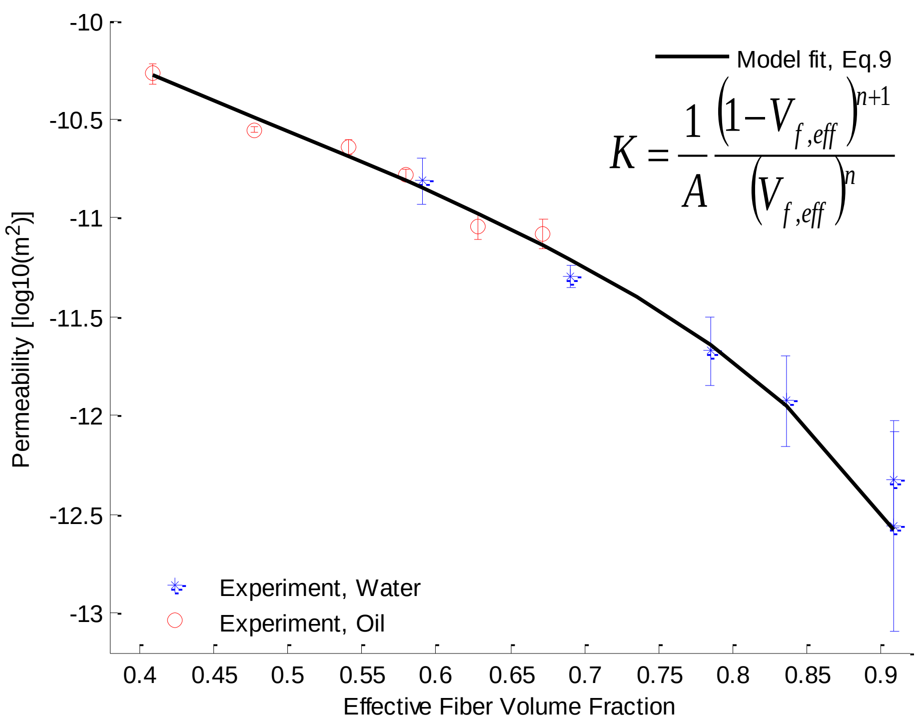
$K_{oil} > K_{water}$



Permeability of Natural Fiber Textile

- $K(V_{f,eff}=f_{sw}^2 V_f)$





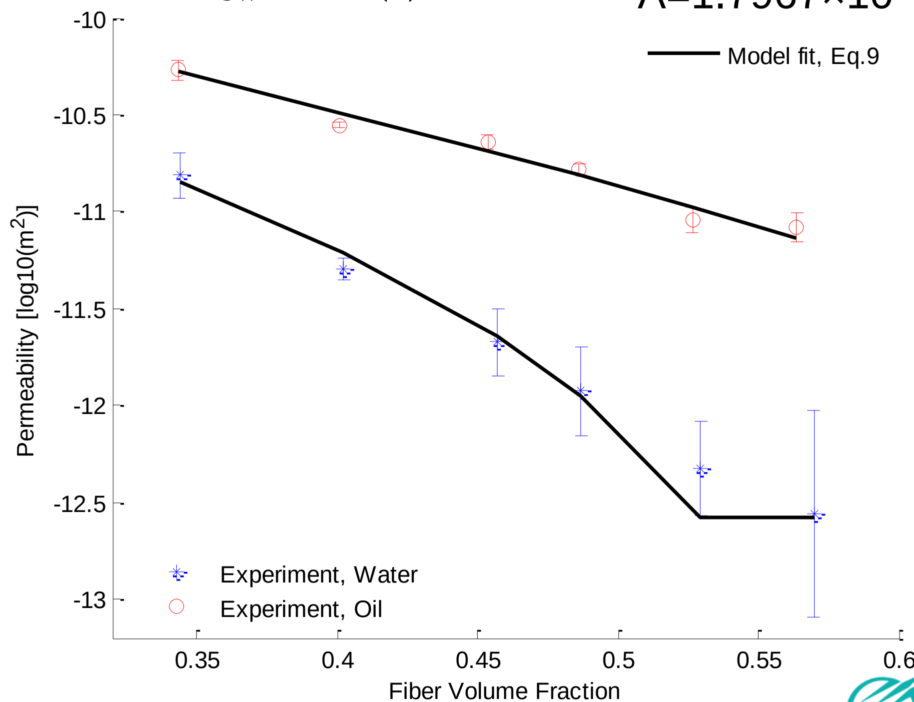
- $K(V_{f,dry}, f_{sw})$

$$K=\frac{1}{A}\frac{\left(1-f_{sw}^{2}V_{f}\right)^{n+1}}{\left(f_{sw}^{2}V_{f}\right)^{n}}$$

$$f_{sw}=f(t)$$

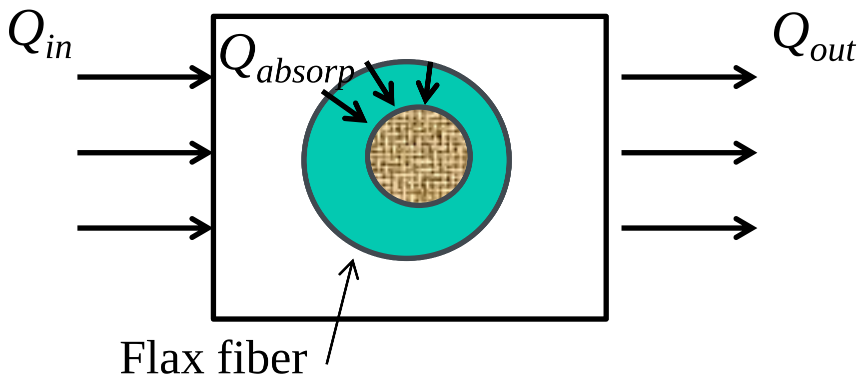
n=1.2855

A=1.7967×10^10



Resin Flow Modeling

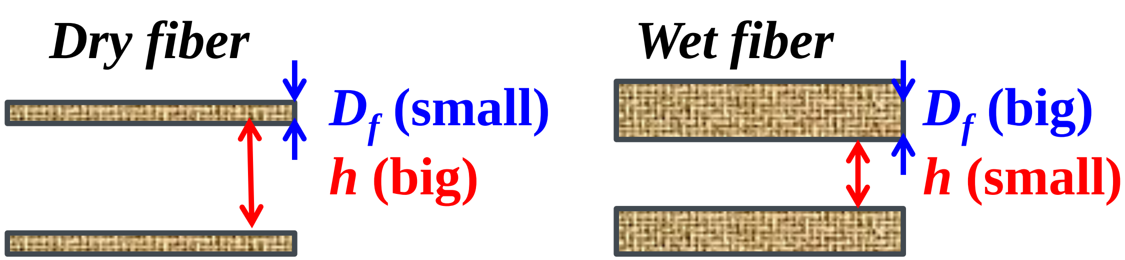
Liquid absorption → *Mass conservation*



$$Q_{in} = Q_{absorp} + Q_{out}$$

$$-\nabla \cdot \vec{u} = s(= \dot{\varepsilon})$$

Fiber swell → *Permeability*



$$\vec{u} = -\frac{\mathbf{K}}{\mu}\nabla P$$

Dependence on Fiber Content and Flow Length

Flow front position square vs. time

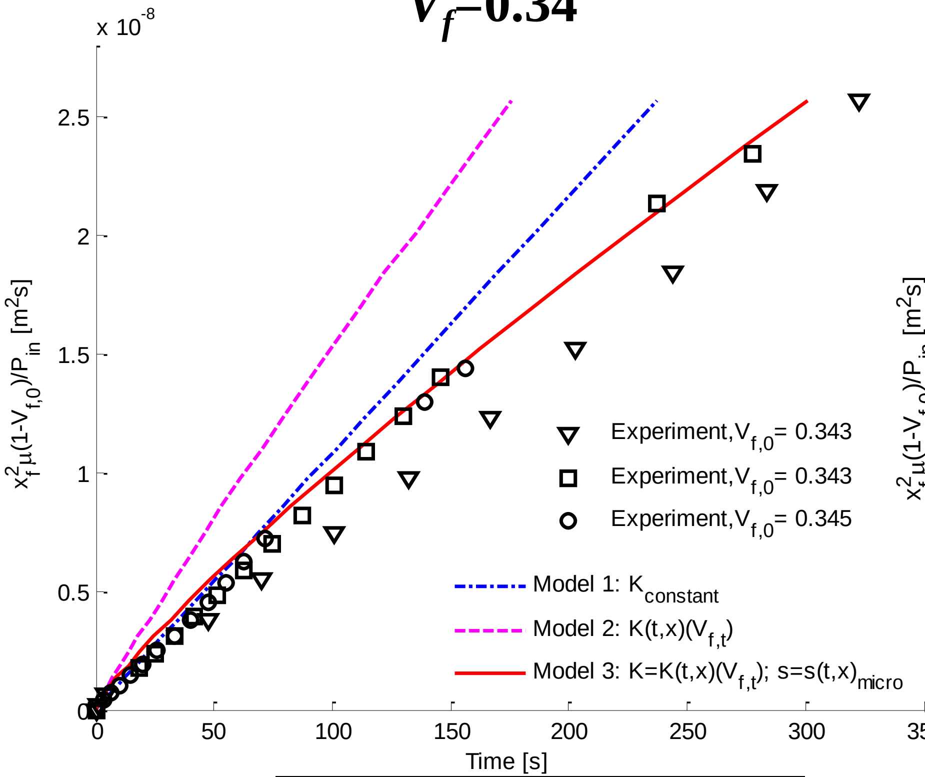
$$X_f = \frac{(1-V_f)\cdot\mu}{P_{in}}\cdot x_f^2 = 2K\cdot t$$

K=const.
K(x,t)
K(x,t) & sink

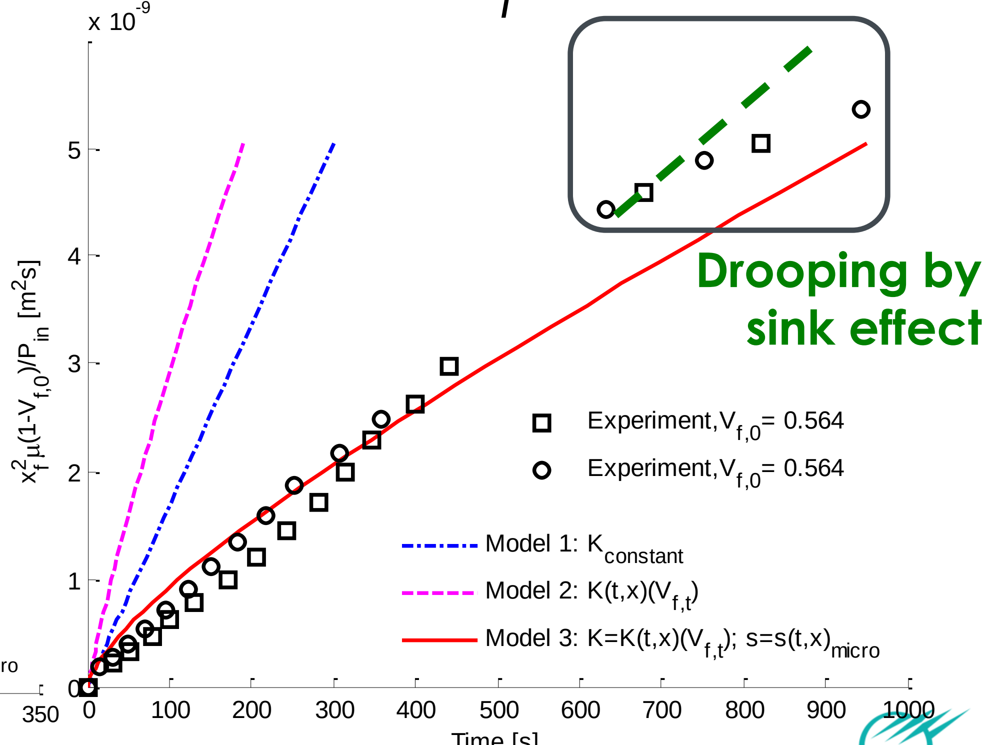
$$\frac{\partial}{\partial x^*}\left(\frac{\partial P^*}{\partial x^*}\right) = \frac{\mu}{K(x,t)\,P_{in}}\frac{x_f^2\,V_f}{1-V_f}\left(\frac{\rho_f}{\rho_l}C_R\frac{\partial f_{so}(t)}{\partial t} - \frac{\partial f_{sw}^2(t)}{\partial t}\right)$$

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

V_f=0.34

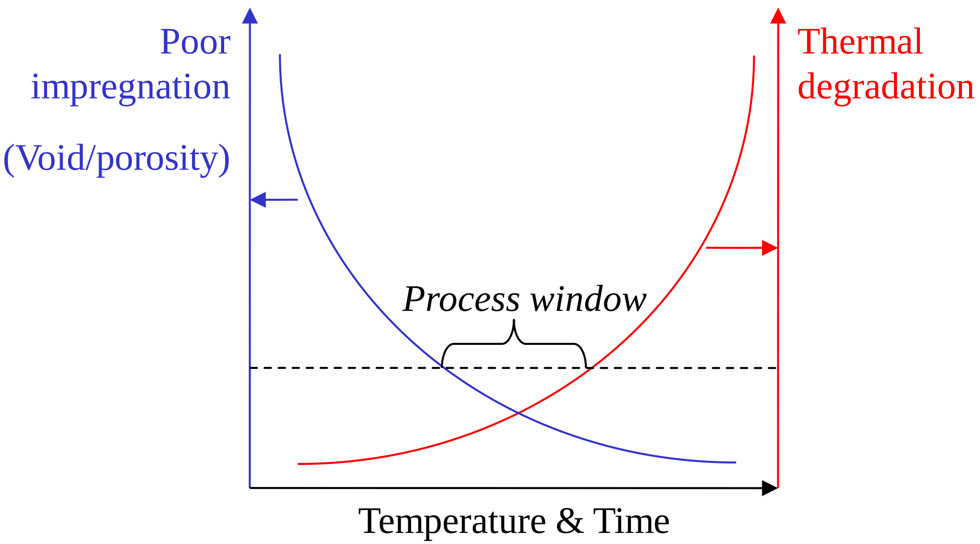


V_f=0.56



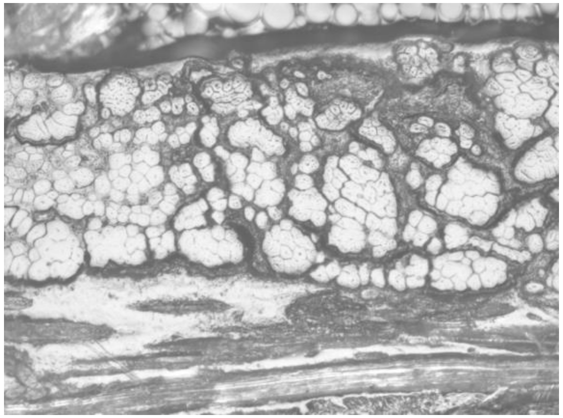
Thermoplastic Matrices

- **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_{degrad} (170 °C) < T_{melt}



Minimize the fiber exposure to heat

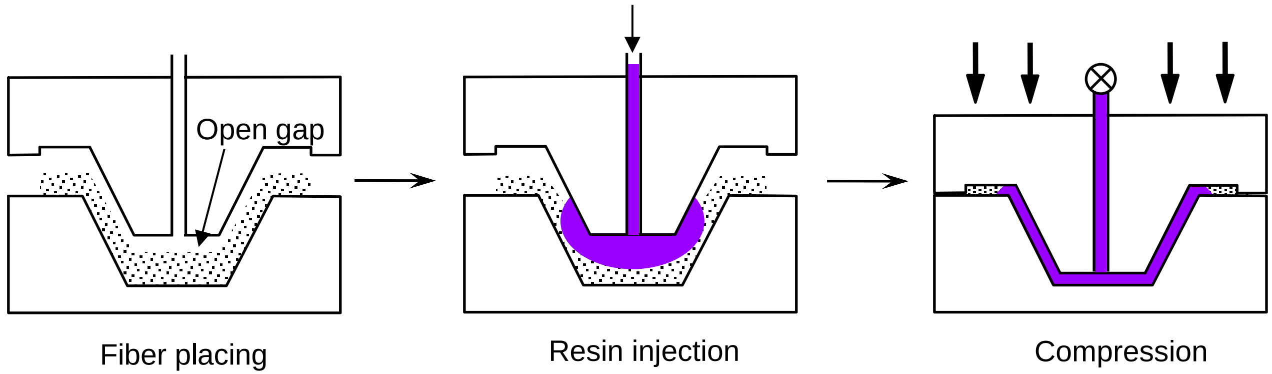
Air voids or porosities due to poor impregnation, poor fiber-matrix compatibility, resin shrinkage or moisture dissolution



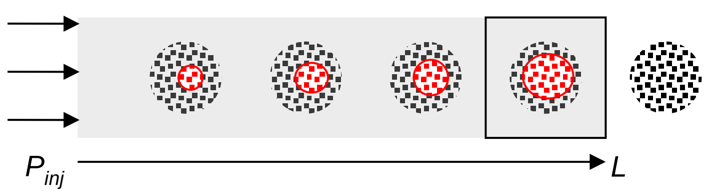
Maximize the local pressure

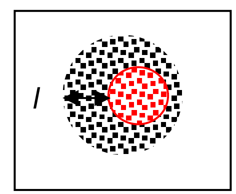
Compression Resin Transfer Molding

Short cycle time



RTM
Longitudinal flow



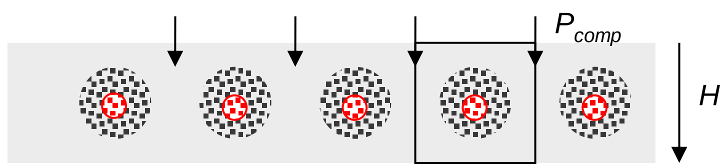


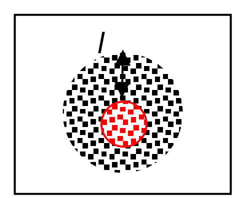
$$\vec{u} \cong \frac{K_{tow}}{\mu}\left(\frac{P_{inj}}{L} - \frac{P_{cap}}{l}\right) << 1,$$

$$P_{cap} = -\frac{\gamma \cos\theta}{D_{pore}}$$

CRTM
Transverse flow

Maximize resin velocity inside the tow





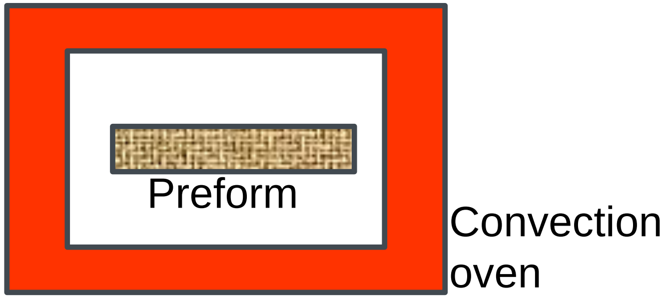
$$\vec{u} \cong \frac{K_{tow}}{\mu}\left(\frac{P_{comp}}{H} - \frac{P_{cap}}{l}\right) >> 1,$$

$$P_{cap} = -\frac{\gamma \cos\theta}{D_{pore}}$$

Manufacturing Process Scheme

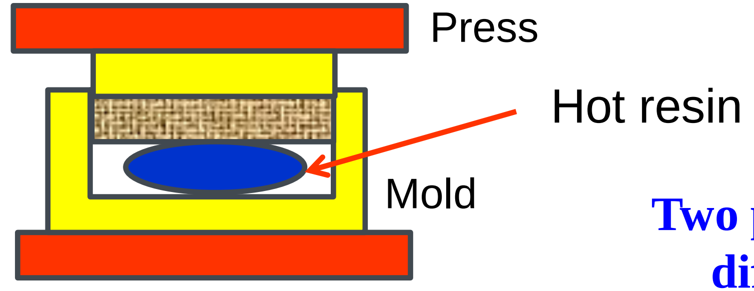
Isothermal mold filling to avoid the temperature drop during the impregnation

1. Preform preheating



$T_{fiber}=T_{high}$

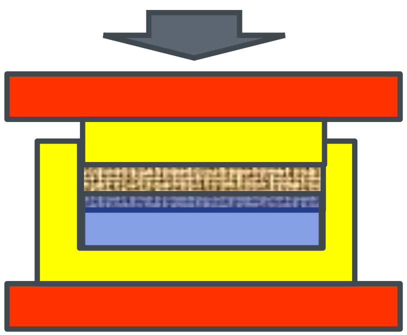
2. Resin injection and preform placement



$T_{mold}=T_{fiber}=T_{press}=T_{resin}=T_{high}$

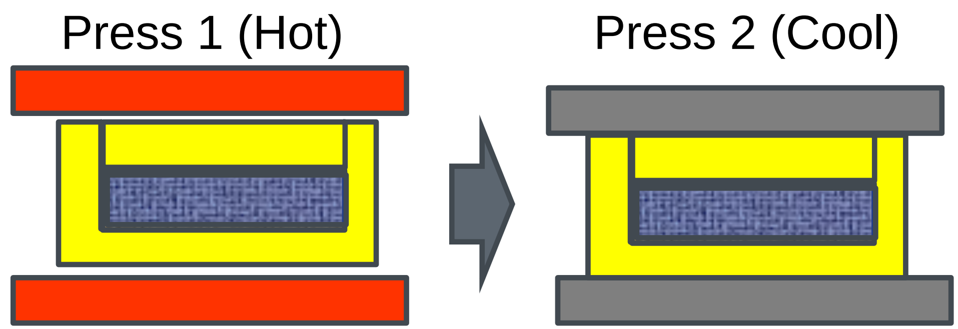
Two presses at different temperature (hot and cool) to reduce the cooling time

3. Mold closing and holding



$T_{mold}=T_{fiber}=T_{press}=T_{resin}=T_{high}$

4. Cooling



$T_{composite}=T_{mold}=T_{high} \rightarrow T_{composite}=T_{press}=T_{low}$

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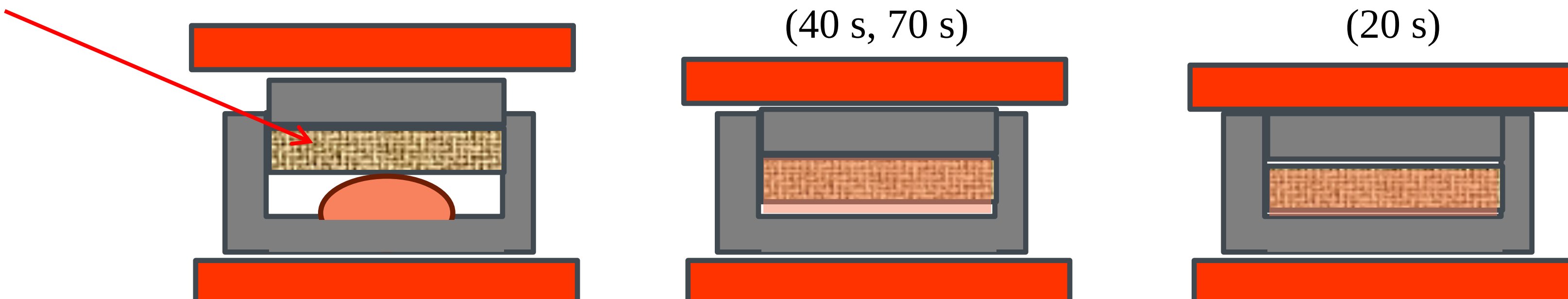
Process Conditions

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

Fiber preheating : 2 min → **Resin injection** → **Compression : 60, 90s**

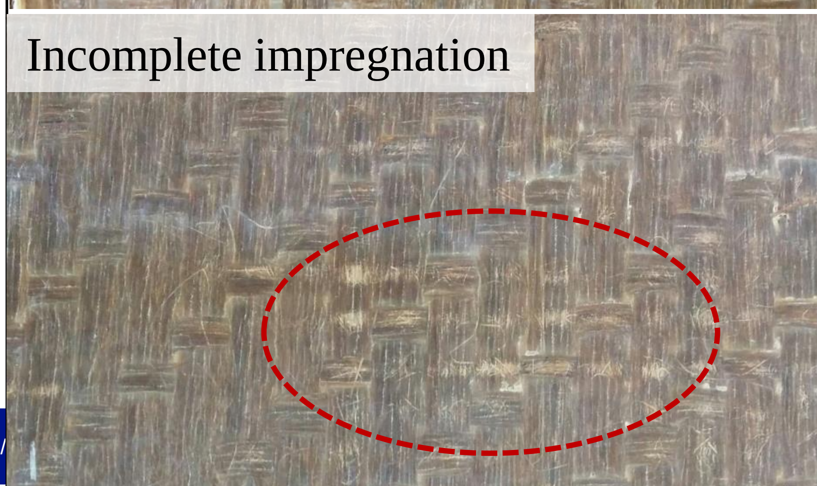
Impregnation at 1.5 bar (40 s, 70 s)

Holding at 21 bar (20 s)



Impregnation Quality

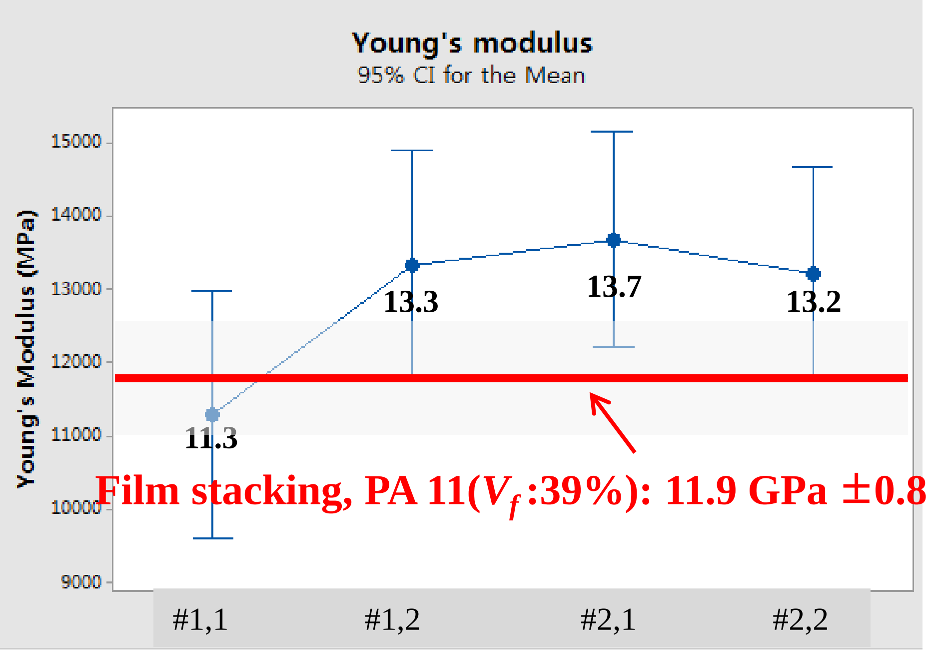
The thermal degradation of flax fiber was not observed.

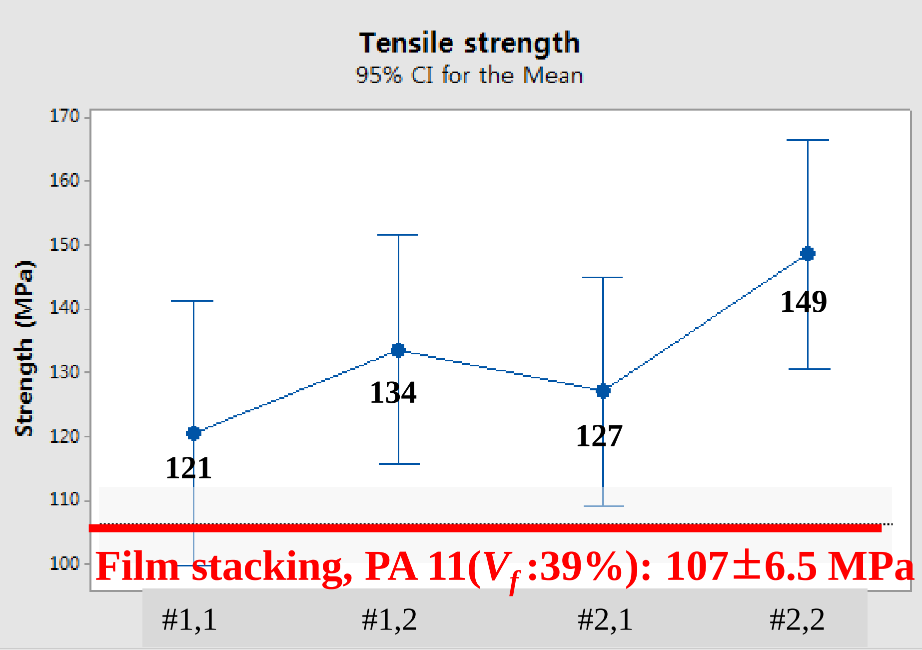
	Compression : 60 sec	Compression : 90 sec
195°C		
205°C		

Mechanical Properties

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

Condition	Cooling	Fiber preheating
#1,1	Slow	In the oven
#1,2 #2,1	Fast	In the oven
#2,2	Fast	In the mold





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