



Laboratory of Sustainable Materials and Product Design

LEPAMAP-PRODIS

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*AMADE Winter Day, Girona
January 23, 2026*

Laboratory of Sustainable Materials and Product Design

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**Cellulose-based
materials and
products**



**Biocomposite
materials**



**Bio-based
nanomaterials**



**Product design &
modelling**

***Designing a sustainable future through bio-based
innovation and circularity***

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Cellulose-based
materials and
products

- **Engineered paper products:** wet strength, high-barrier performance, electrically conductive, paper-based sensors, properties enhancement.
- **Optimized pulp and paper processes:** enzyme-based fractionation, DES-pulping, surface & morphological modification, biorefinery approaches.
- **Beyond paper:** molded pulp, cardboard, other paper & board products.
- **Tuning electrostatic interactions** in pulp slurries & computational methods.
- Focus on packaging and special papers (e.g., security).
- Certified lab (ISO 17025:2004) for food contact materials.



PACKAGING
X CLUSTER

Cepi

ASPAPEL

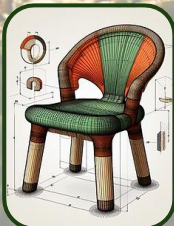
TAPPI



Biocomposite
materials



Bio-based
nanomaterials



Product design
& modelling

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**Biocomposite
materials**

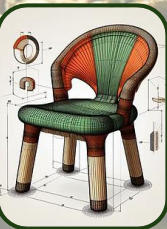
- **Industrial & advanced processing for novel biocomposites:** high-fiber content composites, injection molding, extrusion, hot-pressing, fused deposition modelling, electrospinning
- **Our approach:** enhancing fiber-matrix adhesion via interface tuning and modelling. Valorization of side-streams for high-performance biocomposites. Beyond mechanical performance: functional biocomposites.
- **Micromechanical modelling:** randomly oriented reinforcement (discontinuous). Addressing modelling challenges for predictive approaches.
- **Beyond thermoplastic matrices:** binderless all-cellulose composites.



**Cellulose-based
materials and
products**



**Bio-based
nanomaterials**



**Product design
& modelling**

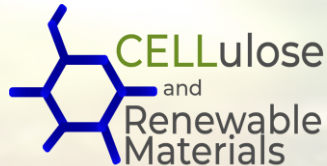
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**Bio-based
nanomaterials**

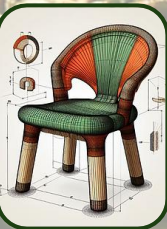
- **Advanced nanopolysaccharides production and characterization:** nanocellulose, nanochitin, alginate complexes, hemicellulose-derived biopolymers. + lignin nanoparticles
- **Electrostatic interactions:** extensive knowledge on electrostatic interactions and their fine-tuning for colloidally stable systems.
- **Fit-for-purpose nanostructured cellulose products:** advancing beyond laboratory scale. Equipped with pilot production lines for high TRL projects.
- **Multiphasic fully bio-based systems:** colloidally stable Pickering emulsions, dispersions, hydrogels, rheology modifiers.



**Cellulose-based
materials and
products**



**Biocomposite
materials**



**Product design
& modelling**

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Product design & modelling

- **Product ecodesign:** integration of end-of-life at the early stages of product design, optimization of design as function of material properties. Not relying on generic libraries, but integrating intrinsic material properties back-calculated from micromechanical modelling.
- **Ergonomics and use analysis:** Shape-function integration based on ergonomics and user needs.
- **Modelling:** finite element analysis, machine learning for property prediction (Bayesian optimization, ANNs, RFs, GNNs), structure-property-process relationships for advanced nanomaterials.
- **Process simulation:** predicting the influence of anisotropy in the final part and the influence over mechanical properties.



Cellulose-based materials and products



Biocomposite materials



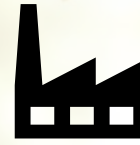
Bio-based nanomaterials

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~120 JCR articles from 2021 to 2025 (87% Q1)



Several university-industry collaborations.
TECNIO Group.



Multiple national and international projects involving other universities, research centers and industries.



Collaborations around the globe (> 20 countries).
Members of several international networks and committees.



Over 30 members (incl. permanent, tenured professors, postdoctoral researchers, PhD candidates and technicians).

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Engineering of cellulose-based multiphasic systems: bridging colloidal systems, interface science and bio-composites

Marc Delgado-Aguilar

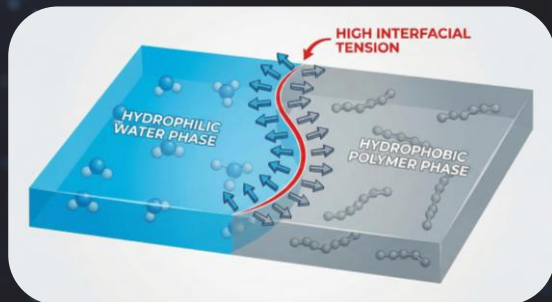


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THE CONTEXT

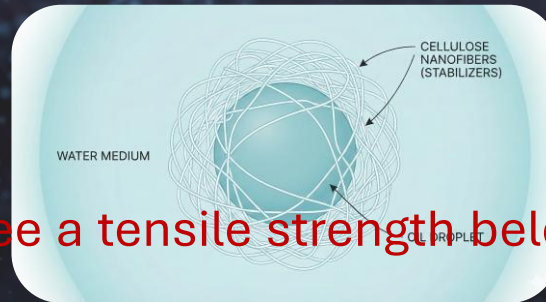


Nature's perfect multiphasic system

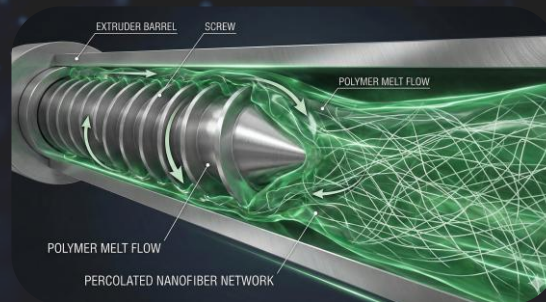


The thermodynamic challenge

THE STRATEGY

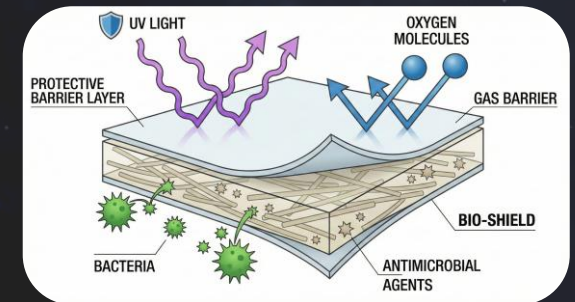
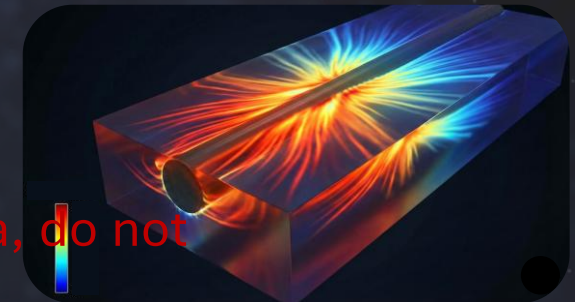


Colloidal stability and Pickering effect



Melt transition: preserving the network

THE OUTCOME



Beyond mechanical performance

DISCLAIMER: If you see a tensile strength below 100 MPa, do not panic. It is not a measurement error. It is simply what happens when you design materials to save the planet rather than to survive re-entry!

THE CONTEXT



*Natural
multiphase*

THE STRATEGY



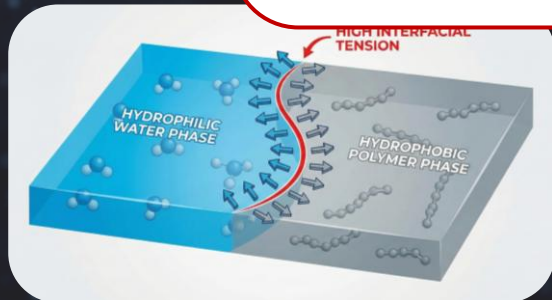
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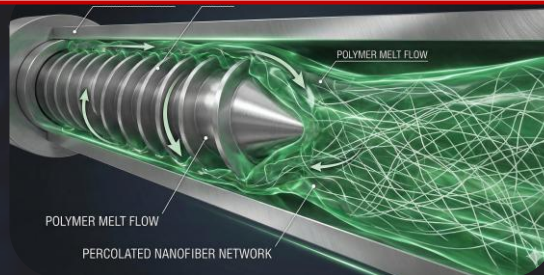
THE OUTCOME



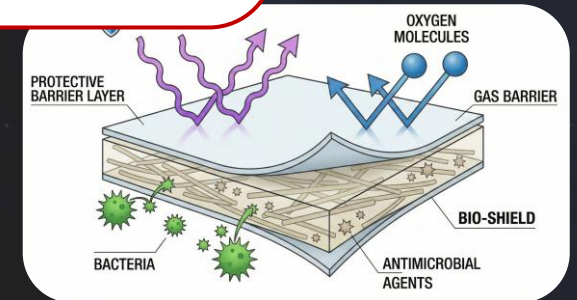
*Cellulose as
reinforcement*



*The thermodynamic
challenge*



*Melt transition: preserving
the network*



*Beyond mechanical
performance*

Content

1. Introducing the world's most sustainable biocomposite
2. Liquid state: pre-structuring the interface in colloidal systems
3. The transition: interface engineering & fiber-reinforced composites
4. Concluding remarks

Content

1. Introducing the world's most sustainable biocomposite
2. Liquid state: pre-structuring the interface in colloidal systems
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4. Concluding remarks

Construction & structural applications

- Buildings.
- Infrastructure.
- Transport.

Household & daily items

- Furniture.
- Kitchenware.
- Stationery.
- Decor.

Energy & fuel

- Heating.
- Energy source.

Recreation & arts

- Musical instruments.
- Sports equipment.
- Toys.
- Art.

Industrial & other uses

- Pulp products.
- Chemicals.
- Tools.

Construction & structural applications

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Energy & fuel

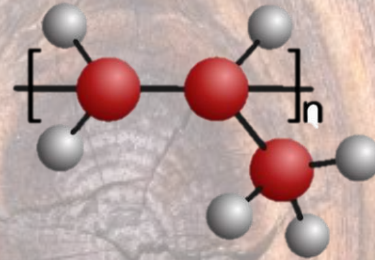
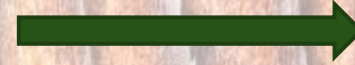
- Heating.
- Energy source.

Recreation & arts

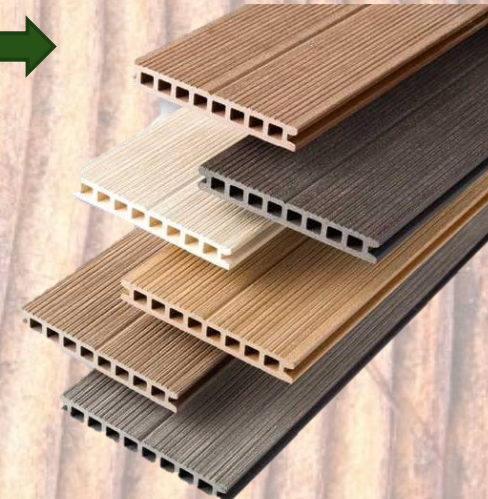
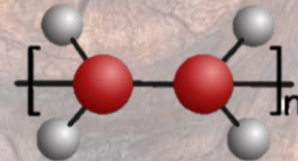
- Musical instruments.
- Sports equipment.
- Toys.
- Art.

Industrial & other uses

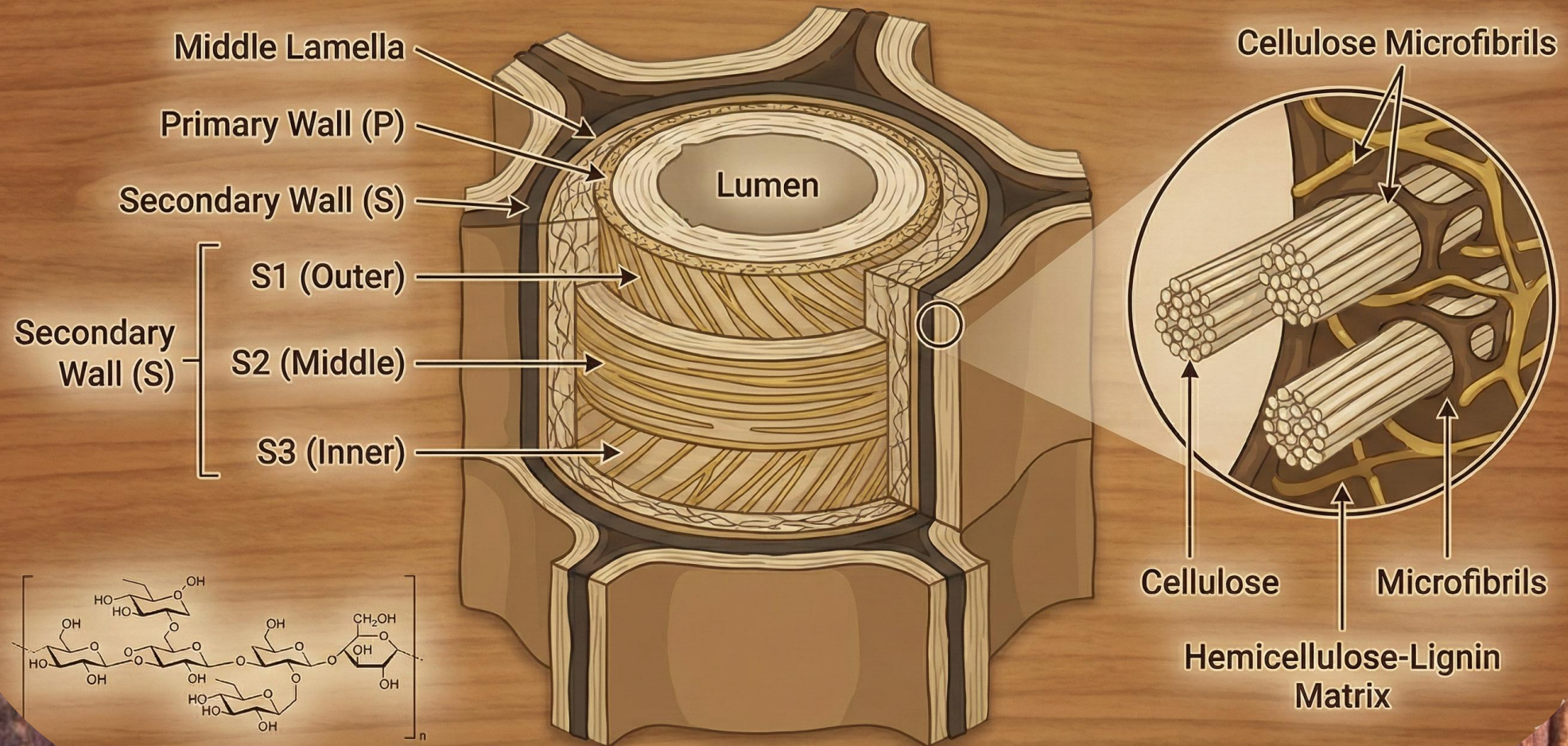
- Pulp products.
- Chemicals.
- Tools.



or

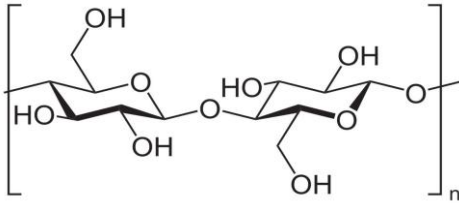


WOOD FIBER CELL WALL STRUCTURE & ORGANIZATION



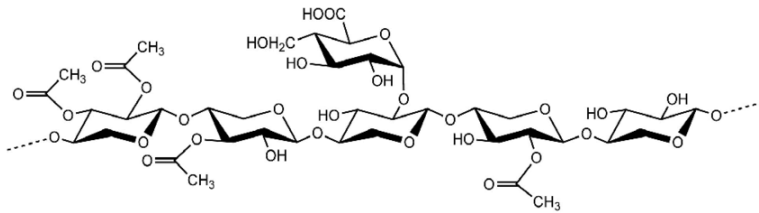
Cellulose, hemicellulose and lignin

Cellulose



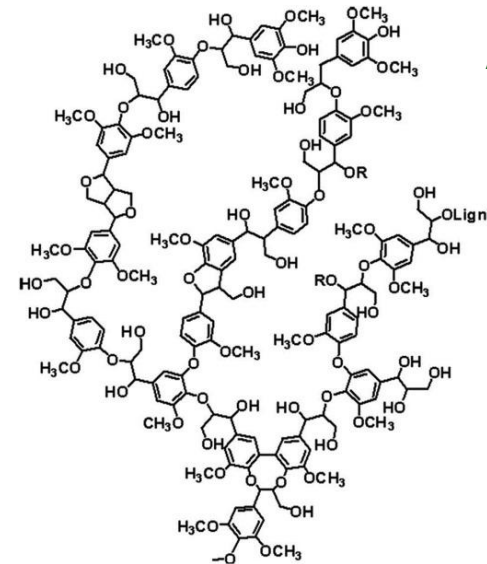
Linear, crystalline homopolysaccharide.
Strength (structural matrix).
Mostly hydrophilic.

Hemicellulose



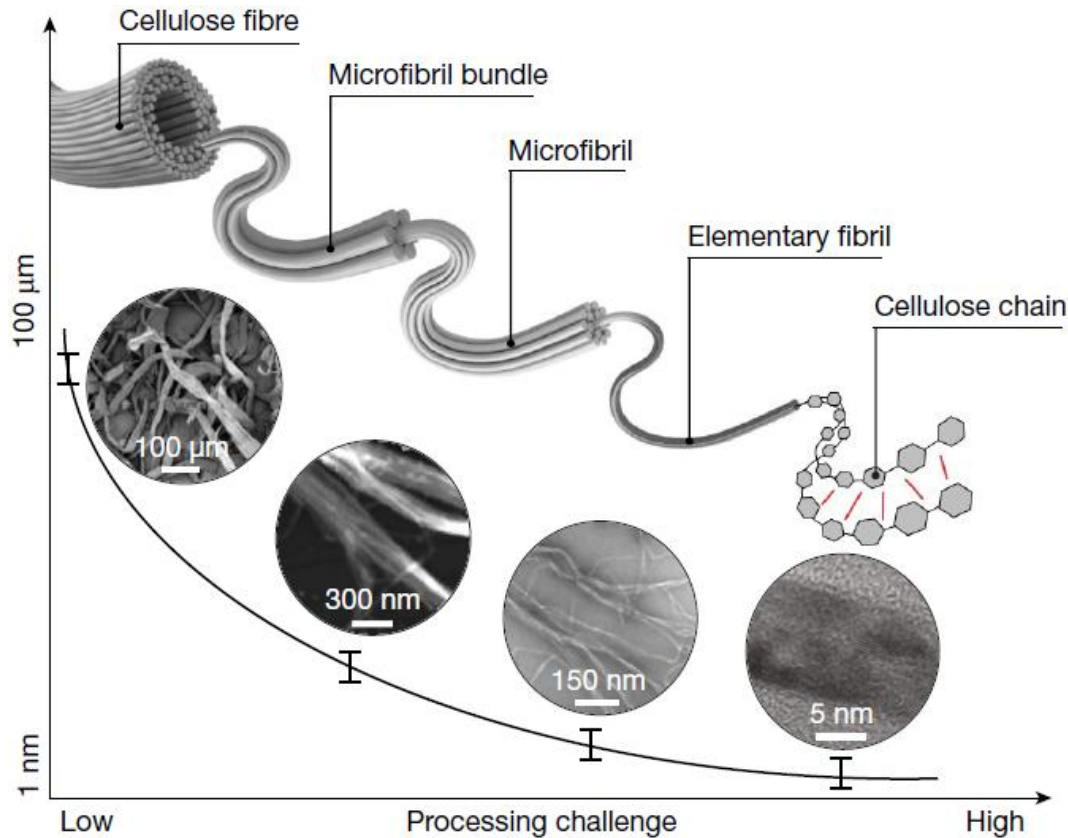
Heterogeneous, branched polysaccharides with lower Mw. Partially water soluble.
Amphiphilic in some extent.

Lignin



Amorphous, highly aromatic biopolymer.
Hydrophobic due to non-polar groups.
Introduces hydrophobic features.

The isolation challenge



- As we process the fibers, we increase the surface area.
 - Cellulose fiber (pulp fibers): $\sigma \approx 1 \text{ m}^2 \text{ g}^{-1}$
 - Elementary fibril (nanofiber): $\sigma > 200 \text{ m}^2 \text{ g}^{-1}$
- Higher surface area leads to higher water absorption capacity. This poses several challenges during composite production and processing (dispersion, hydrolysis, self-entanglement).
- Hemicellulose and lignin are still present and provide several attributes to the fiber structure and properties.
- There is room for surface modification and morphology tuning, which directly impact the resulting composite properties.

The polarity: we are trying to mix oil and water

Cellulose fibers

- They are polar due to the presence of hydroxy groups.
- They tend to agglomerate due to Van der Waals forces and hydrogen bonding.
- Their isolation is performed in aqueous media: they need to be dried prior compounding (agglomeration!!).
- They are an excellent platform for grafting functional groups.
- Cellulose starts to degrade at around 200 °C.

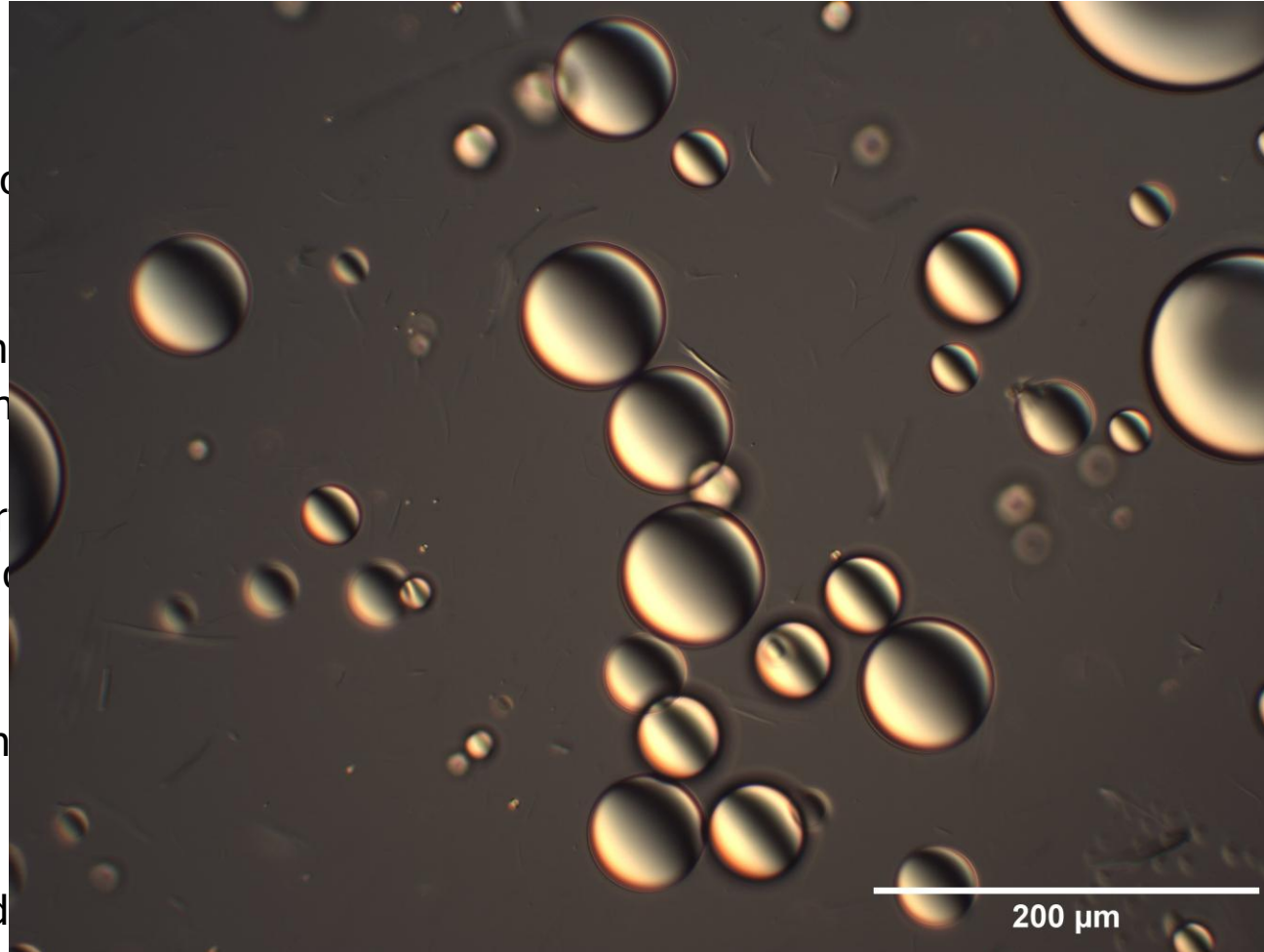
Thermoplastics (most of them)

- They are non-polar (they hate water).
- They do not wet properly hydrophilic surfaces (such as cellulose fibers).
- They require high temperature to be processed (above T_m). In most of the cases, this temperature is close to 200 °C.

The polarity: we are trying to mix oil and water

Cellulose fibers

- They are polar due to hydroxyl groups.
- They tend to agglomerate due to van der Waals forces and hydrogen bonds.
- Their isolation is performed by drying (agglomeration!!).
- They are an excellent source of functional groups.
- Cellulose starts to decompose at high temperatures.



Most of them)

(they hate water).

on hydrophilic surfaces (e.g., glass).

temperature to be processed in the cases, this temperature

Content

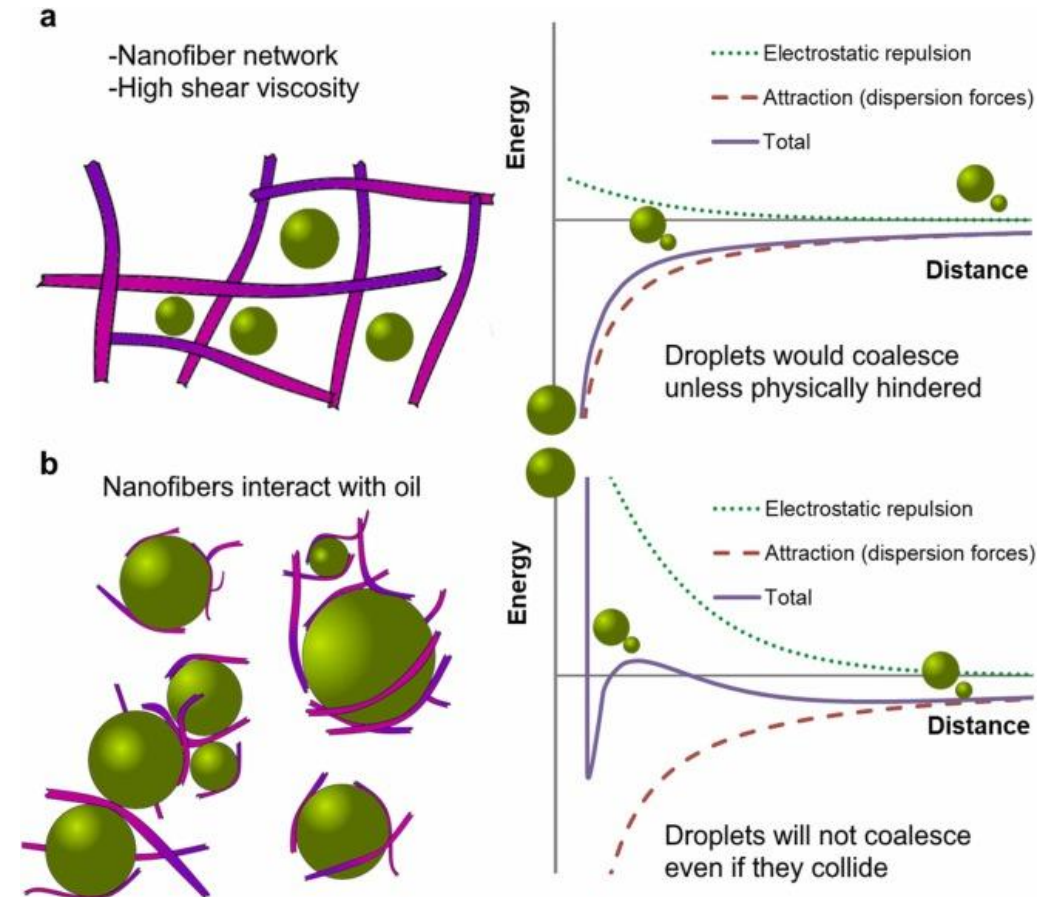
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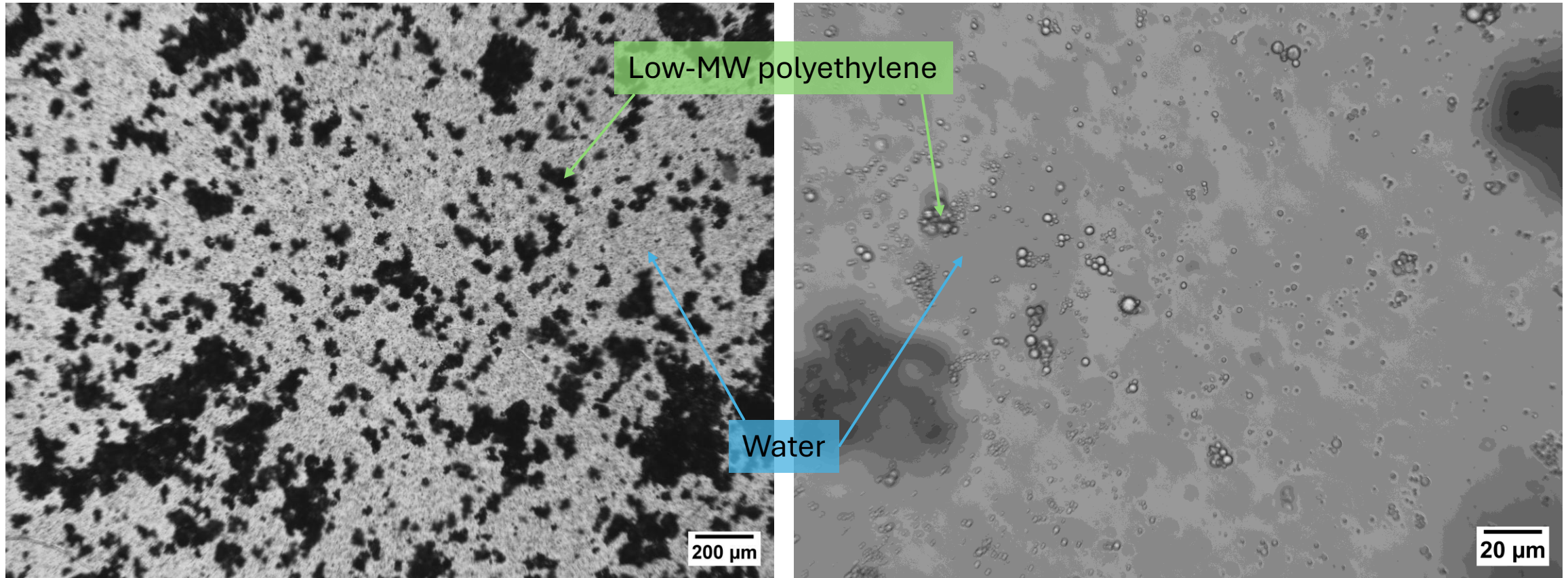
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Pre-structuring the interface

- Controlling the colloidal state (e.g., preventing reinforcement aggregation) is the only way to achieve a 3D-structured network required for mechanical reinforcement in the solid state.
- We can learn from colloidal systems such as Pickering emulsions: the same principles of surface energy and interfacial tension that stabilize emulsions dictate how a polymer melt will “wet” the fiber.



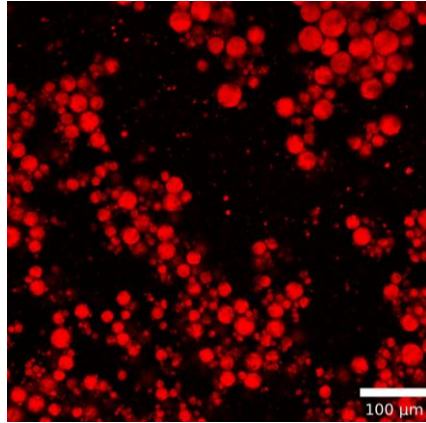
Pre-structuring the interface



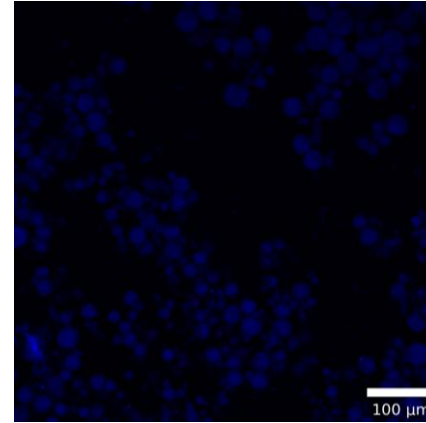
Ongoing stability analysis: 2 months since production at RT

Pre-structuring the interface

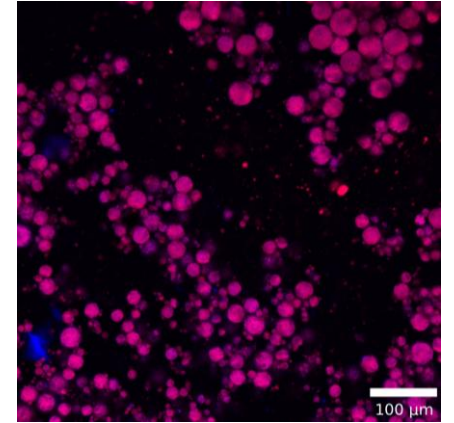
- Adsorption of cellulose fibers (nanofibers) onto the oil phase).
- Mostly driven by electrostatic interactions and rheological stabilization.



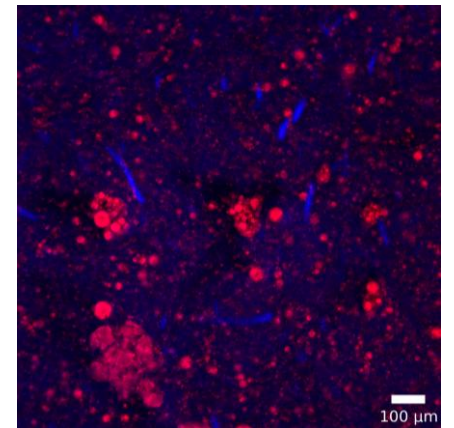
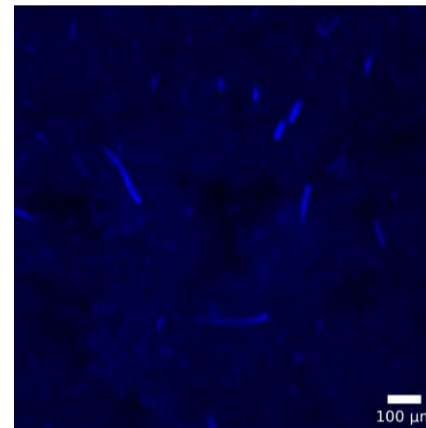
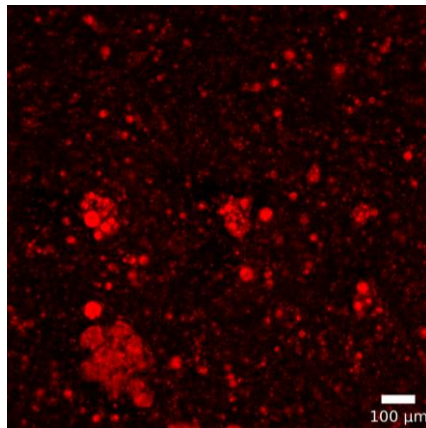
Oil phase: Nile red



CNF phase: Calcofluor white



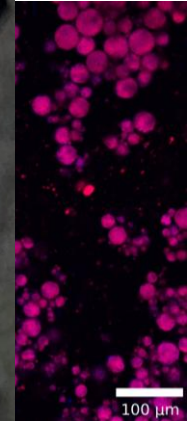
Composite image



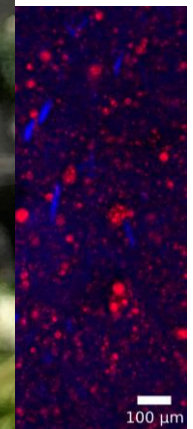
Pre-s

- Adsorption of fibers (nanofibers) in oil phase
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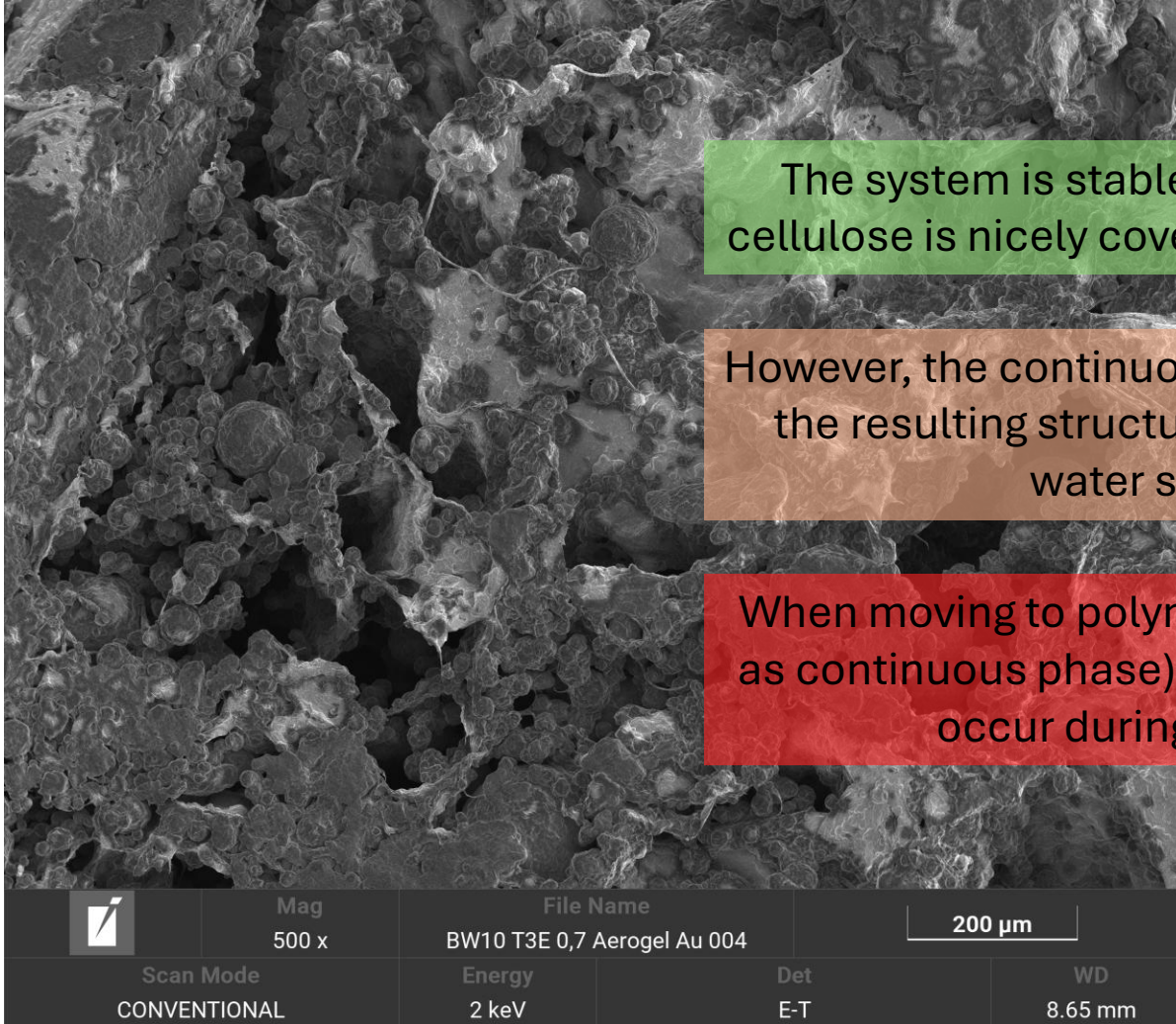
Credits: Giulia Herbst and Gabriela Bastida



image



Pre-structuring the interface



The system is stable, in the solid state, and cellulose is nicely covering the polymer droplets

However, the continuous phase is cellulose, and the resulting structure is highly porous after water sublimation

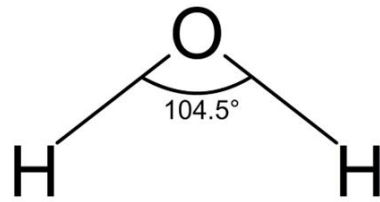
When moving to polymer melts (where they act as continuous phase), fiber agglomeration may occur during water removal



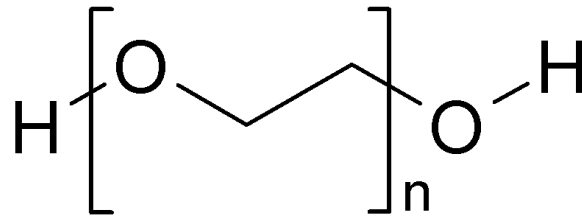
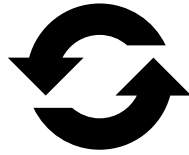
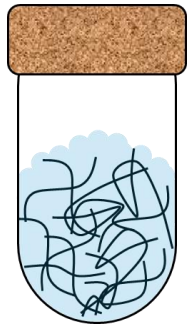
The permanent solvent approach

How do we keep this network structure within a polymer melt and preserve it once solidified?

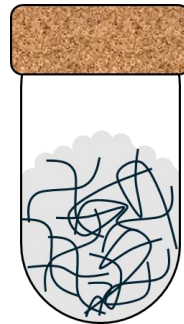
- Solvent exchange.



Water



Polyethylene glycol (PEG1000; $n \approx 22$)

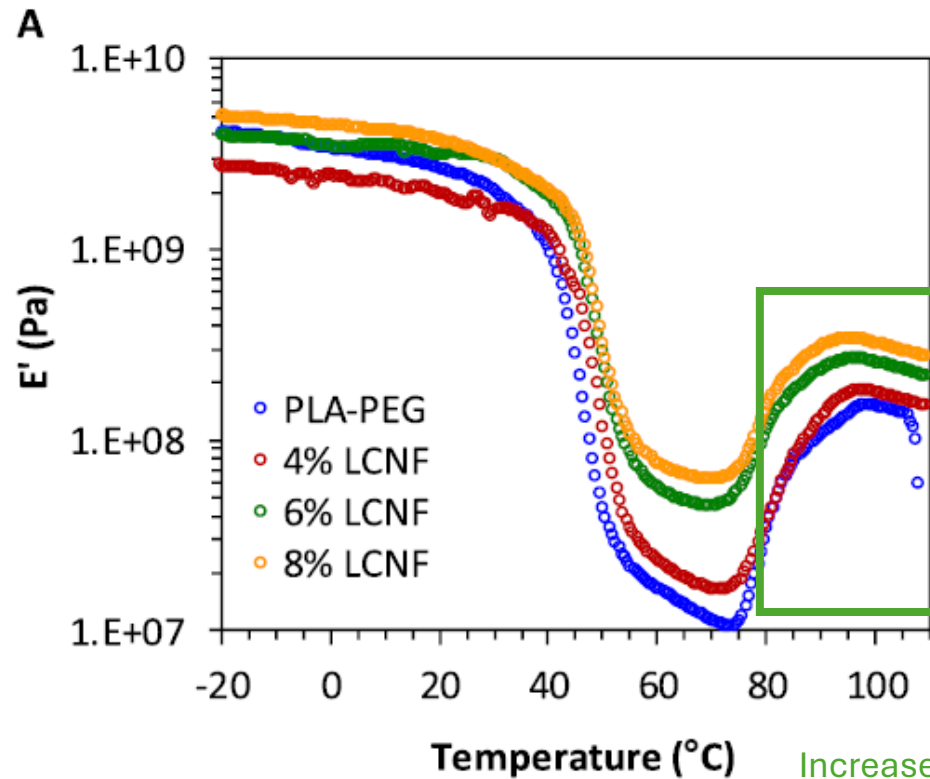


- No boiling point.
- Decomposition occurs before (around 250°C).

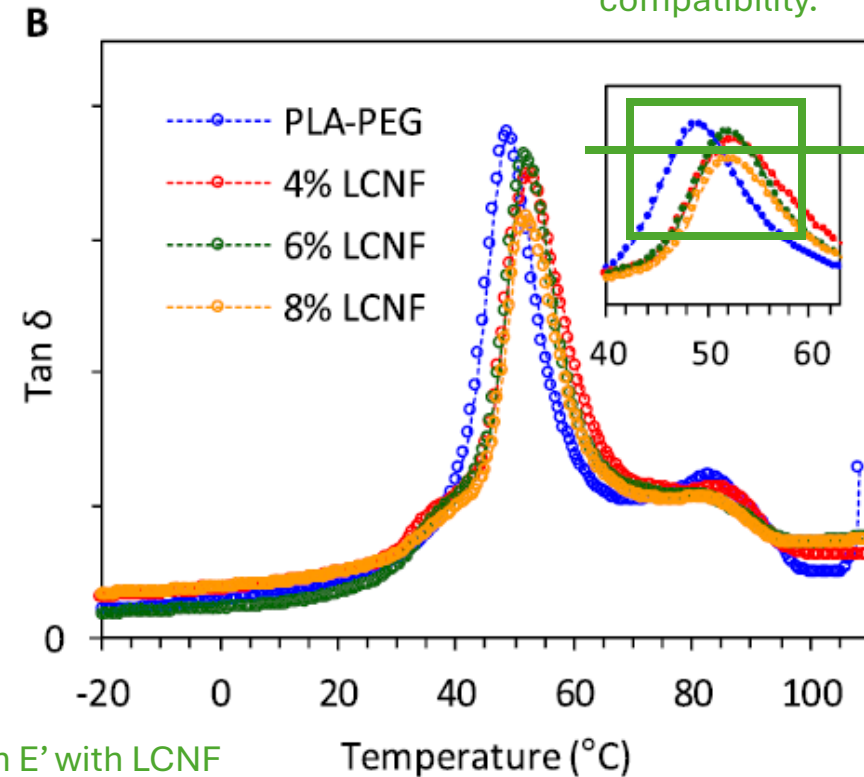
- *PEG acts as a carrier.*
- *Promotes the stabilization of the phases in the melt state, even under shearing and temperature.*
- *Highly entangled network within the matrix.*



The permanent solvent approach



Increase on E' with LCNF content in the rubbery state. Good compatibility between reinforcement and matrix.

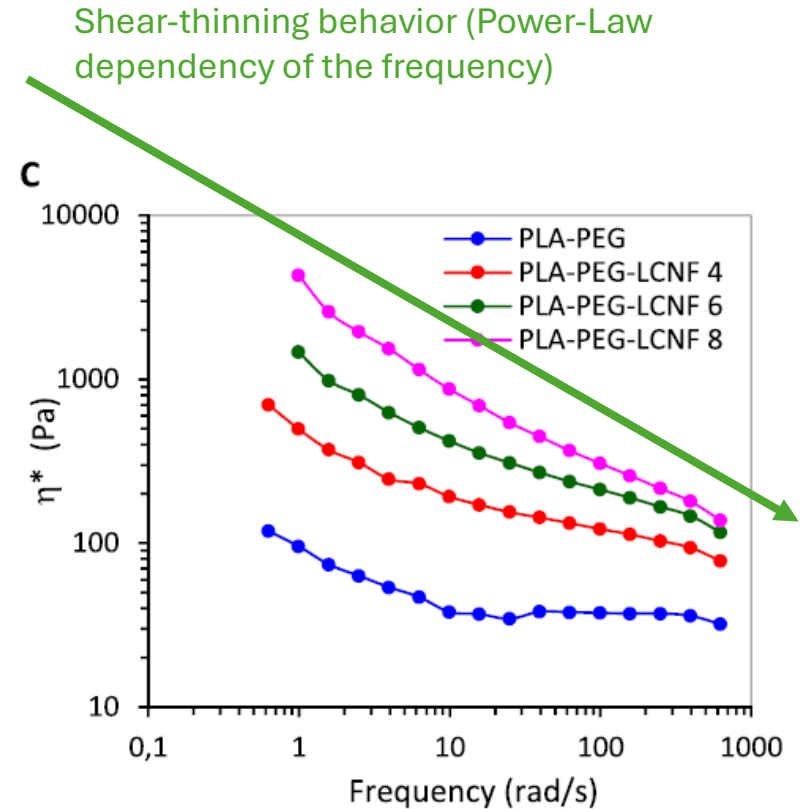
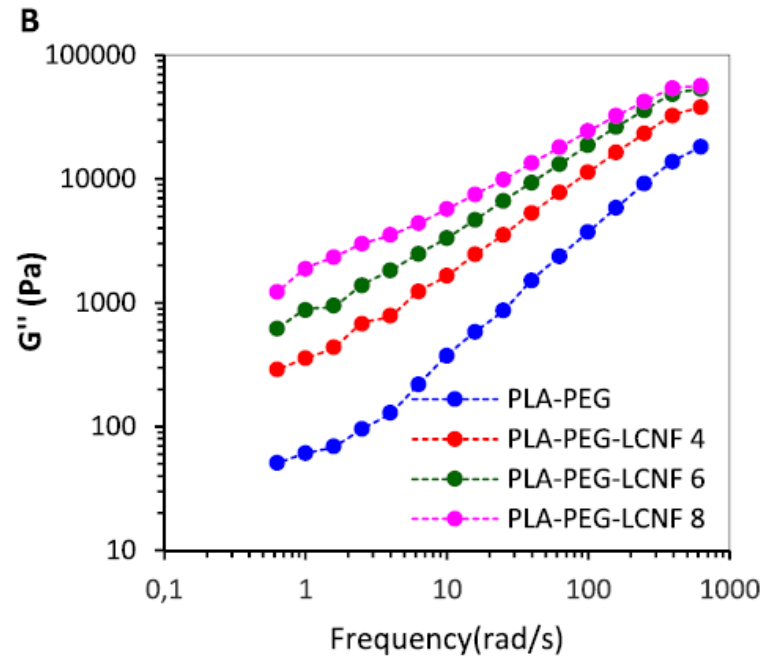
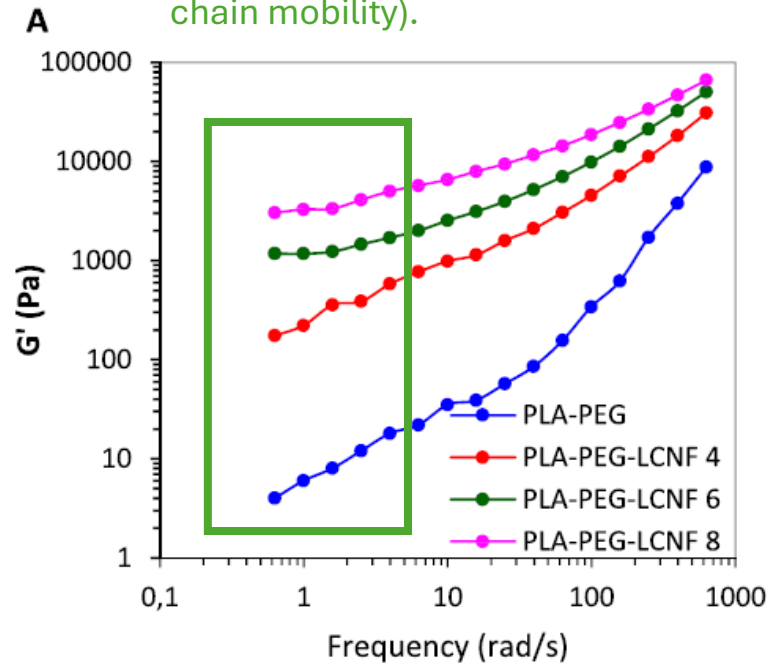


The presence of one single peak supports good PLA/PEG compatibility.

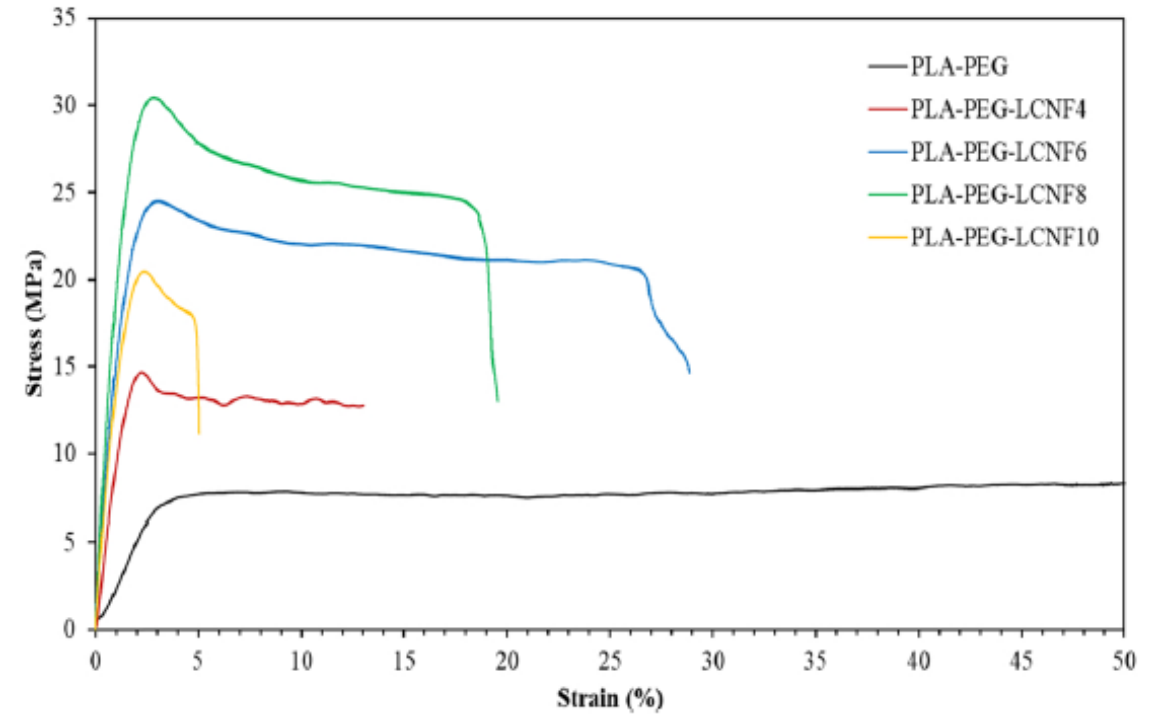
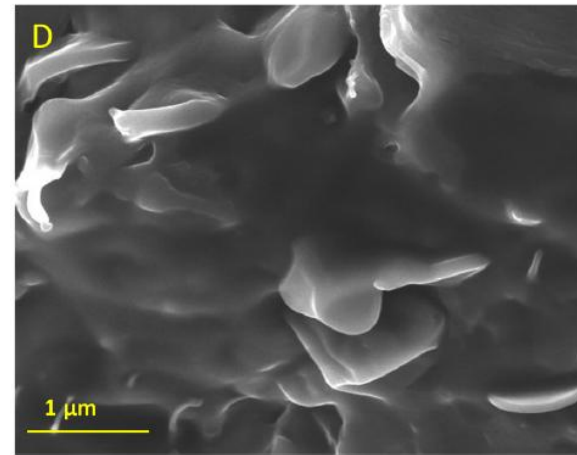
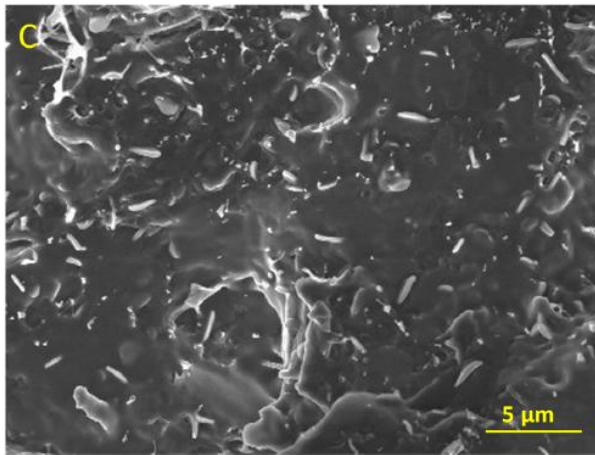
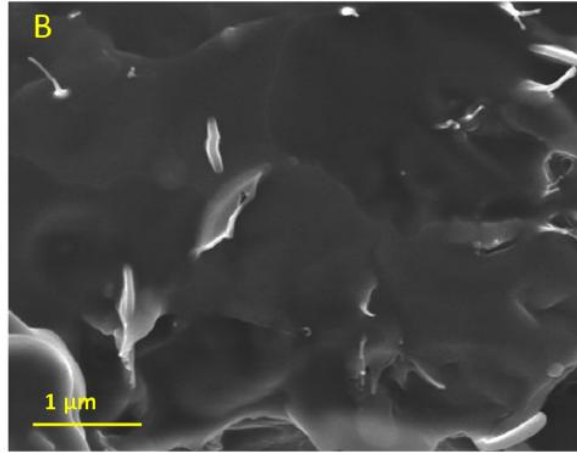
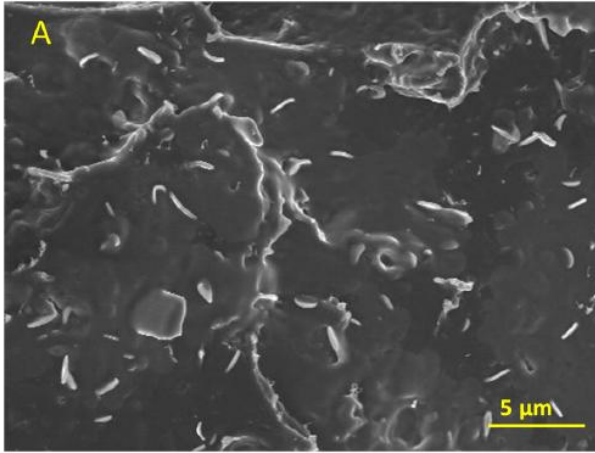
Slight increase on temperature: restriction in segmental mobility of the polymer chains.

The permanent solvent approach

Significant increase of G' at low frequency (restriction of chain mobility).



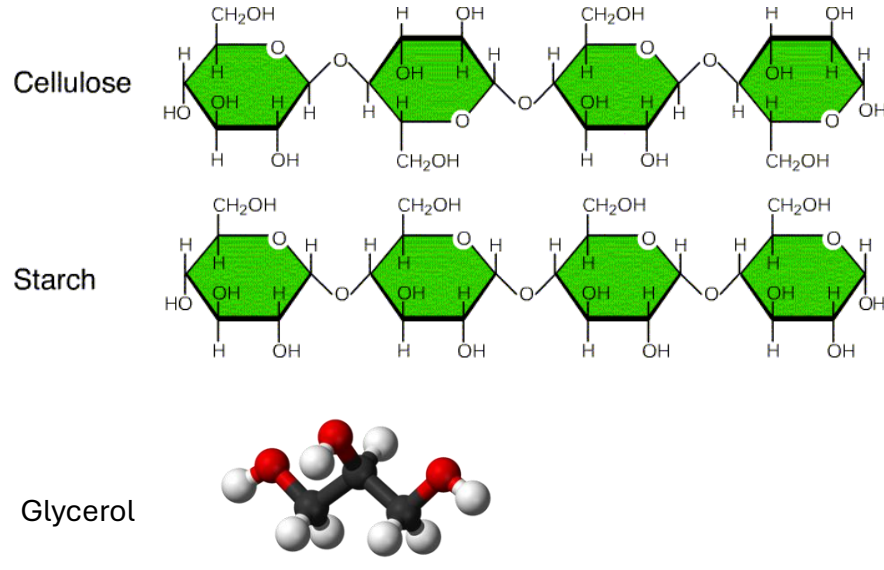
The permanent solvent approach



Stabilizing polymer blends

How do we keep this network structure within a polymer melt and preserve it once solidified?

- Solvent exchange.
- Polymer blends: using thermoplastic starch as carrier for PBAT composites.



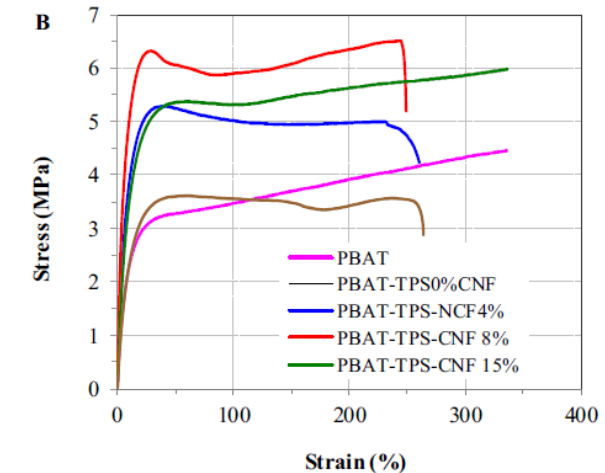
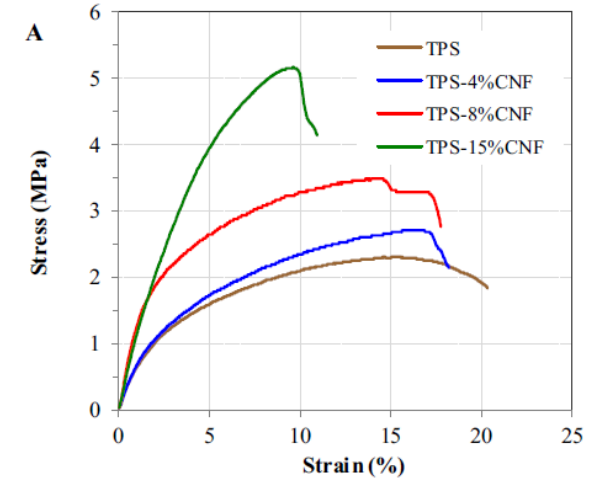
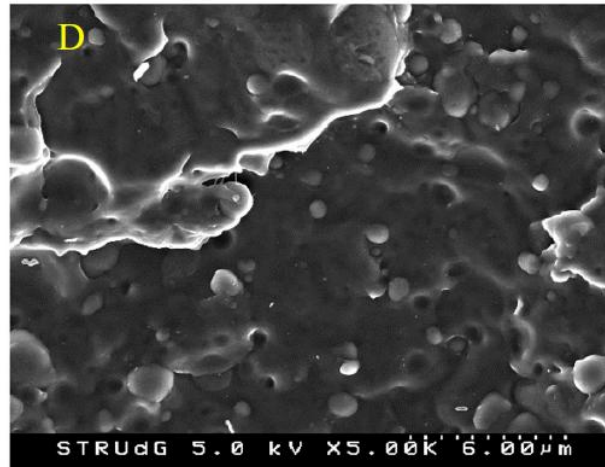
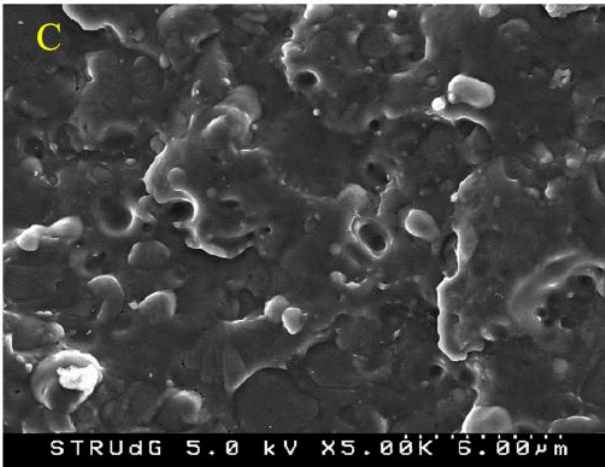
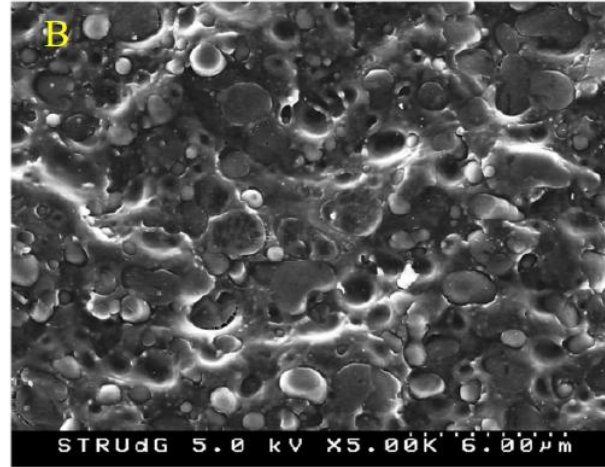
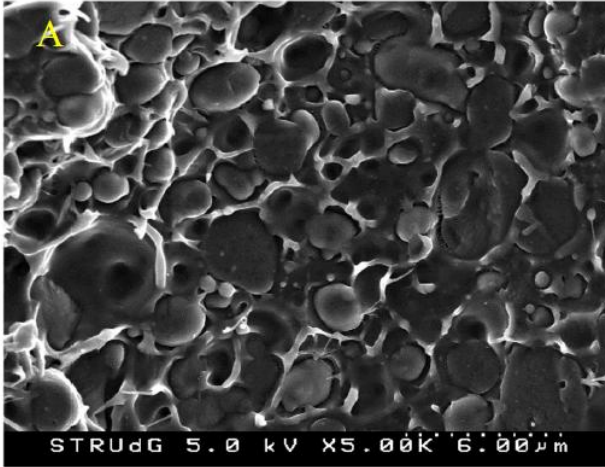
Reinforced
thermoplastic
starch (TPS)



Polybutylene adipate
terephthalate (PBAT)



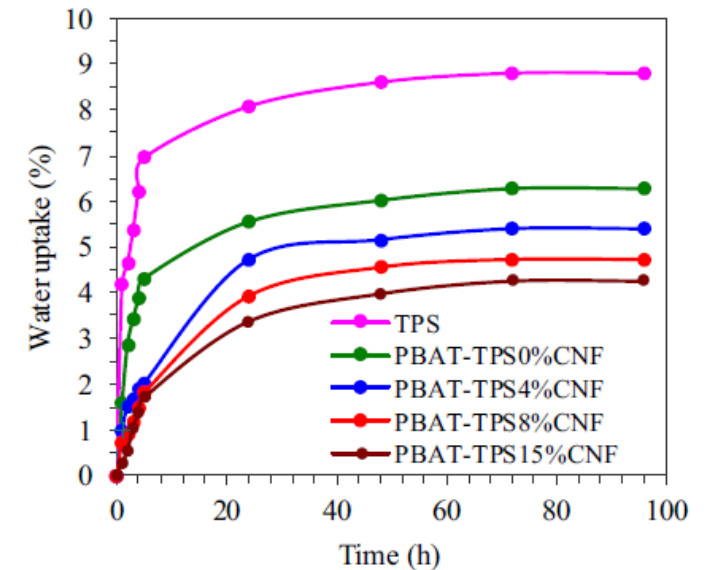
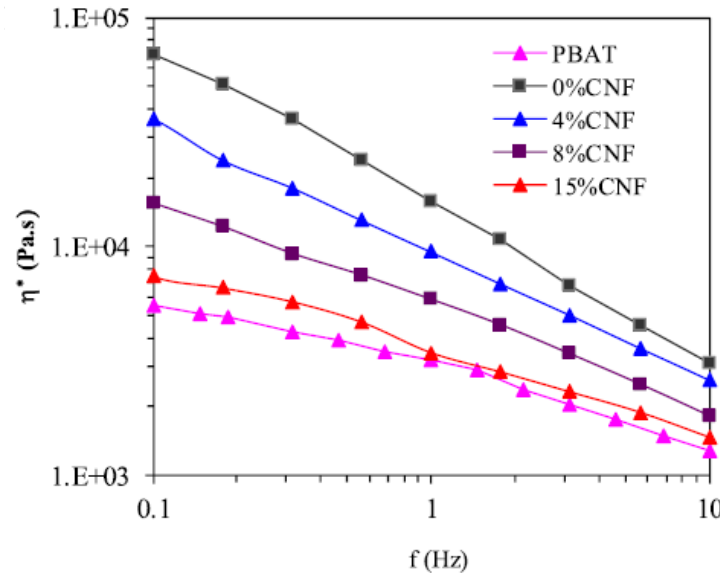
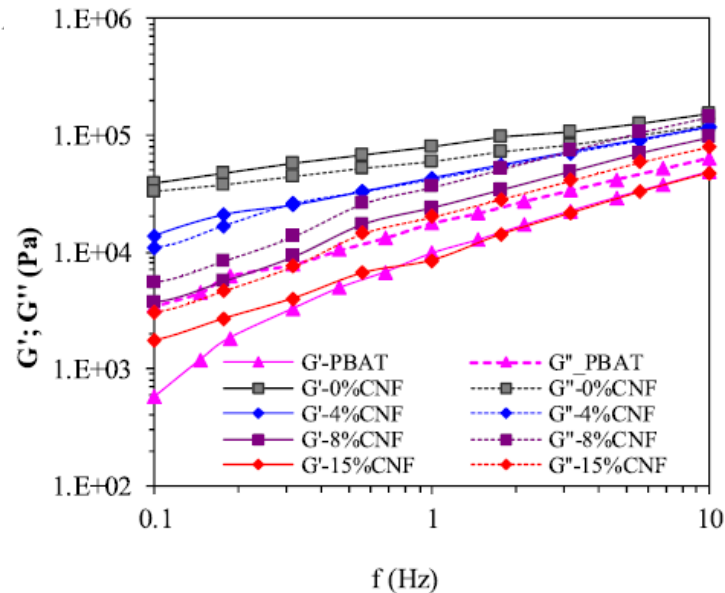
Stabilizing polymer blends



- Cellulose has a dual role: stabilizing the immiscible matrices and reinforcement.
- Co-continuity of the matrices thanks to the percolated CNF network.

Stabilizing polymer blends

Temperature: 150 °C

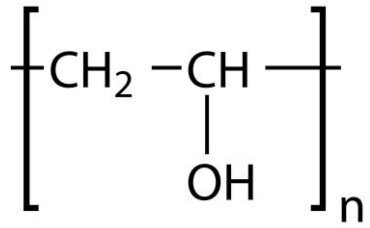


A highly percolated CNF network provides viscous-like materials, indicating lower stiffness and property recovery (compared to neat PBAT).

While hydrophilic phases are included into the PBAT, water uptake decreases:

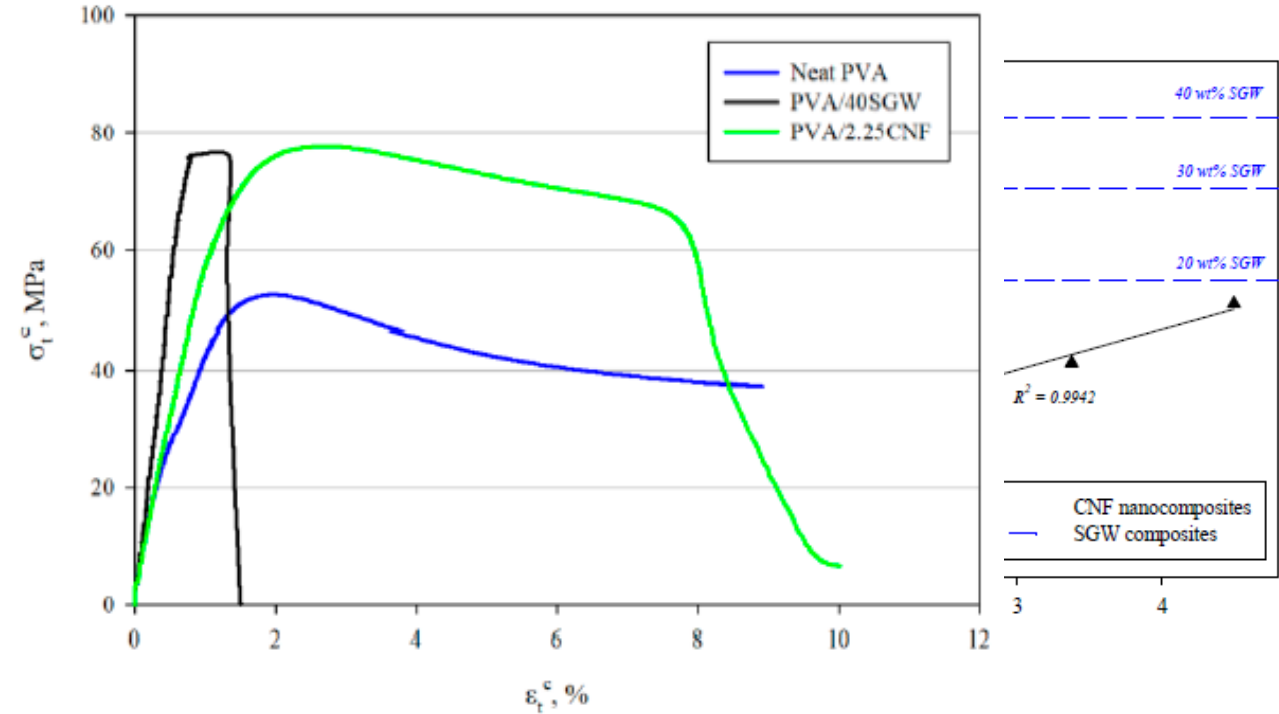
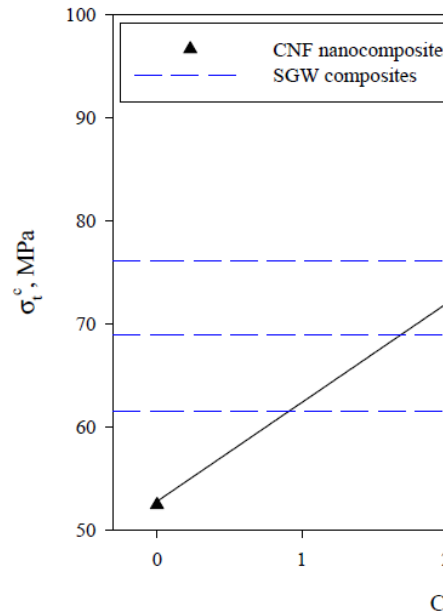
- Homogeneous distribution of the reinforcement and the co-matrix.
- Closing of the structure, followed by potential densification.

Effectively reinforcing polymer matrices with nanocellulose



Polyvinyl alcohol

- Highly hydrophilic (depending on the hydrolysis degree, even water-soluble).
- Excellent interactions with cellulose.



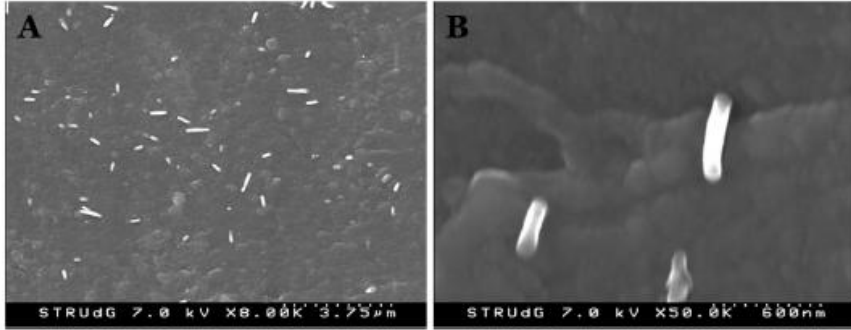
2.25% CNF provides the same enhancement on strength than 40% micro-sized fibers



Residual variations on density



Effectively reinforcing polymer matrices with nanocellulose



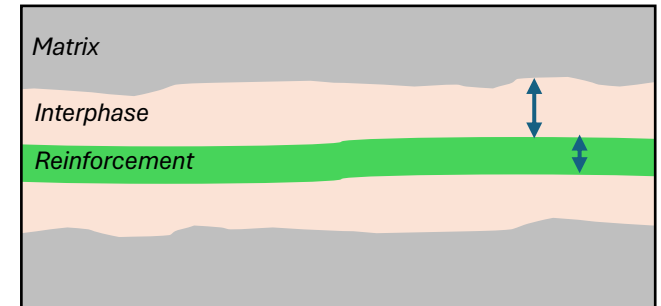
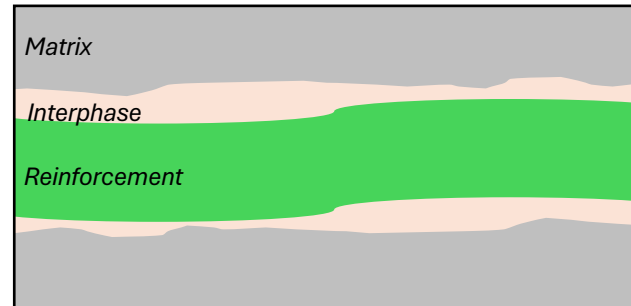
CNF % w/w	V^{NF} v/v	τ (MPa)	t (nm)	σ_t^I (MPa)	σ_t^{NF} (MPa)	f_c
2.250	0.0185	31.25	120.6	94.83	7622.5	0.21
3.375	0.0280	27.40	93.4	104.23	6825.6	0.19
4.500	0.0370	25.28	96.2	97.56	6297.5	0.19

$$\sigma_t^R = \frac{\sigma_t^C}{\sigma_t^m} \quad B = \frac{\ln \left(\sigma_t^R \cdot \frac{1 + 2.5 \cdot V^F}{1 - V^F} \right)}{V^F}$$

$$\tau = \frac{\sigma_t^m \cdot (B - 2.04)}{\chi_1 \cdot \alpha}$$

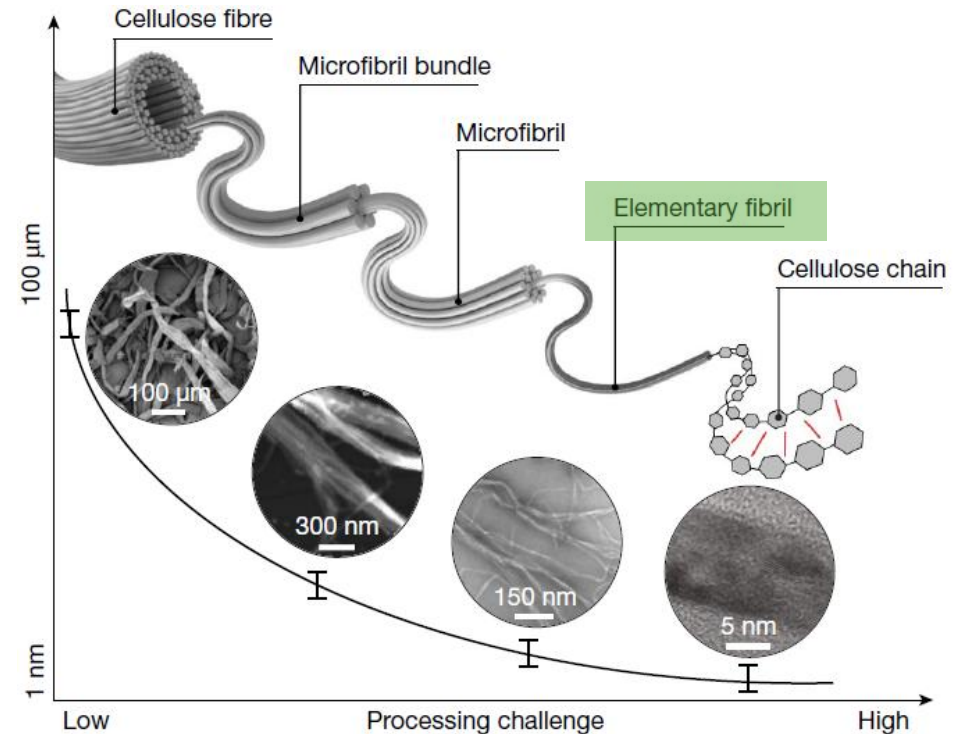
$$\sigma_t^I = \sigma_t^m \cdot e^{\left(\frac{B}{1 + \frac{2 \cdot t}{d^F/2}} \right)}$$

$$\sigma_t^C = \chi_1 \cdot \alpha \cdot \tau \left(1 + \frac{t}{d^F/2} \right) \cdot V^F + \sigma_t^m \left[1 - \left(1 + \frac{t}{d^F/2} \right)^2 \cdot V^F \right]$$



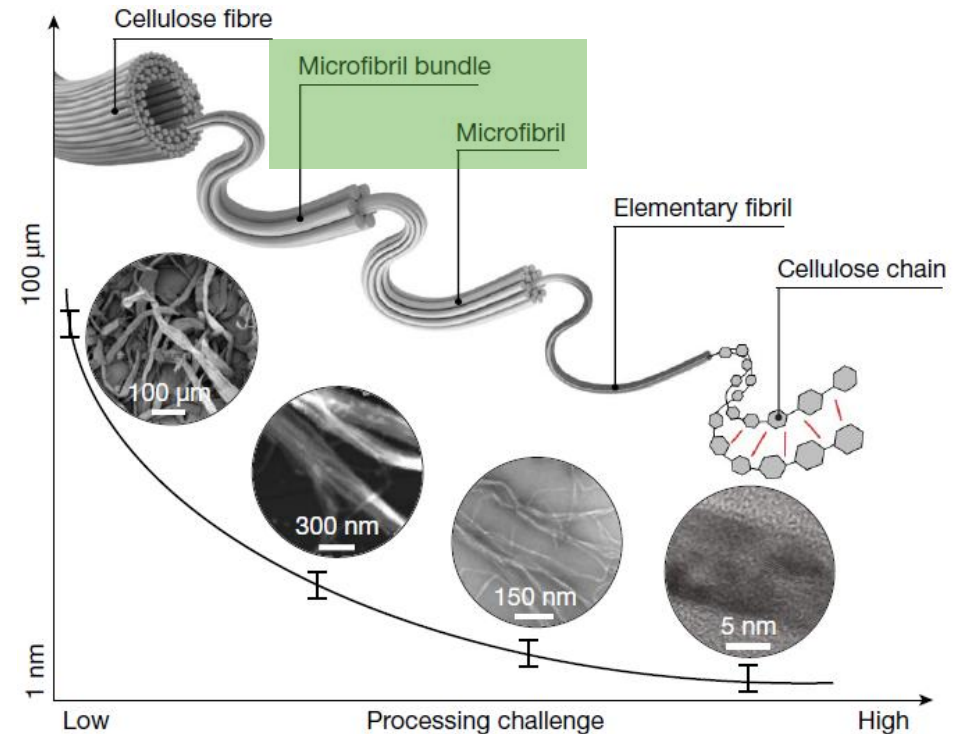
Using nanocellulose is nice, but...

- The high hydrophilicity of nanocellulose hinders their inclusion into hydrophobic polymer matrices.
- Usually, carriers are required, which will impart negative effects to mechanical properties.
- The stability of these systems in the wet and melt state is hard to preserve when moving to the solid state. Mostly based in electrostatic interactions and rheology: disrupted due to shearing and temperature.
- Surface chemical modification is usually required, as well as removing water prior compounding. A lot of research going on, but the cost-property trade-offs remain unexplored due to lack of scale.



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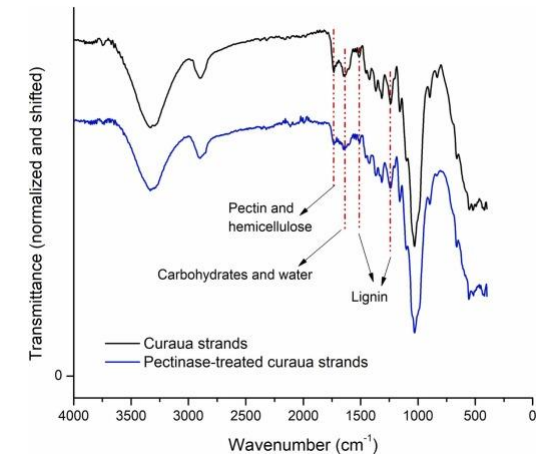
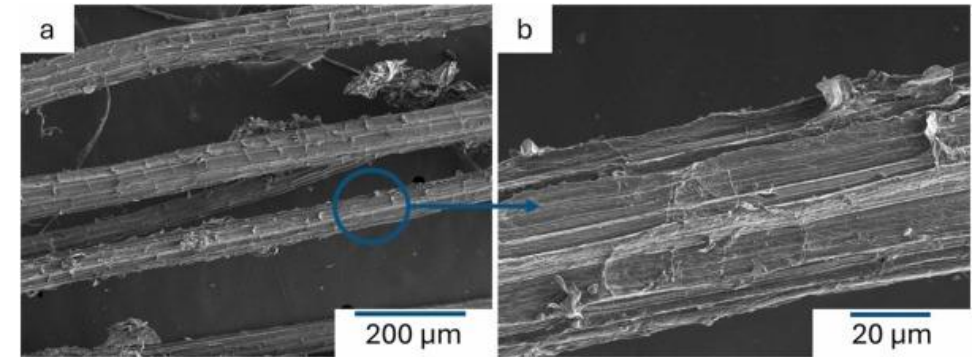
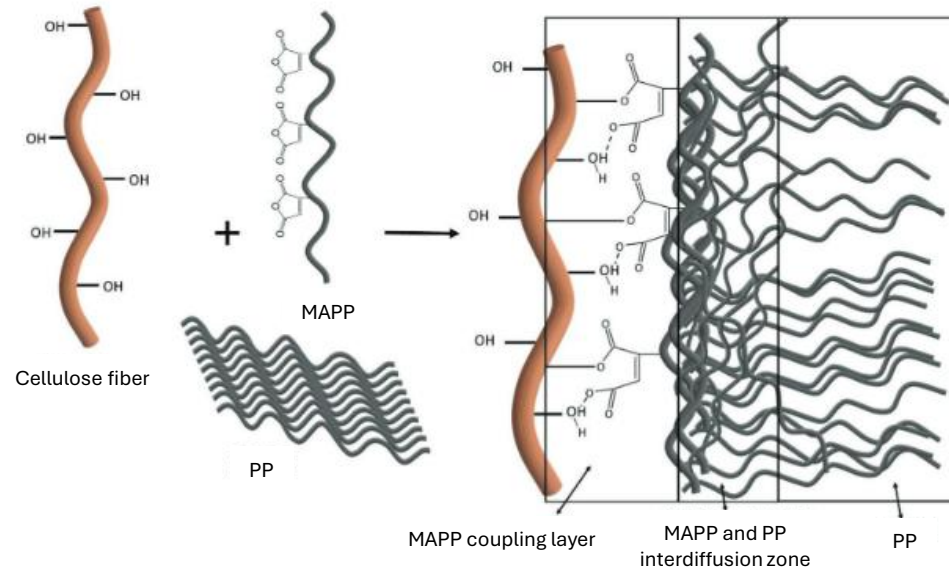
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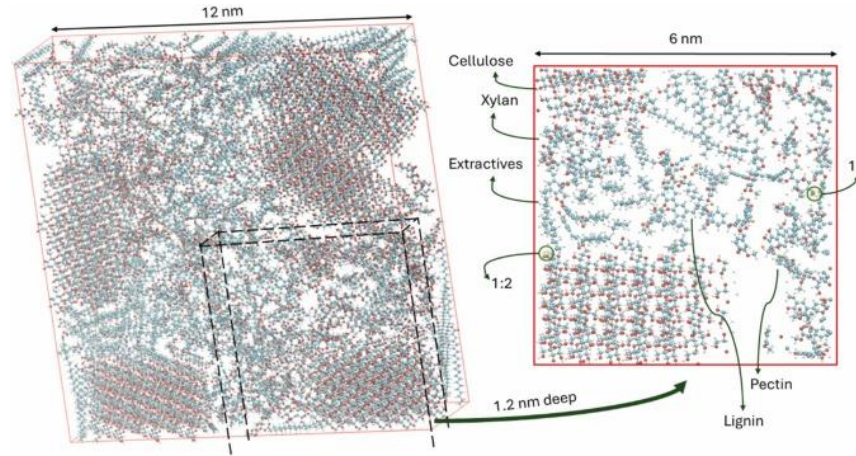
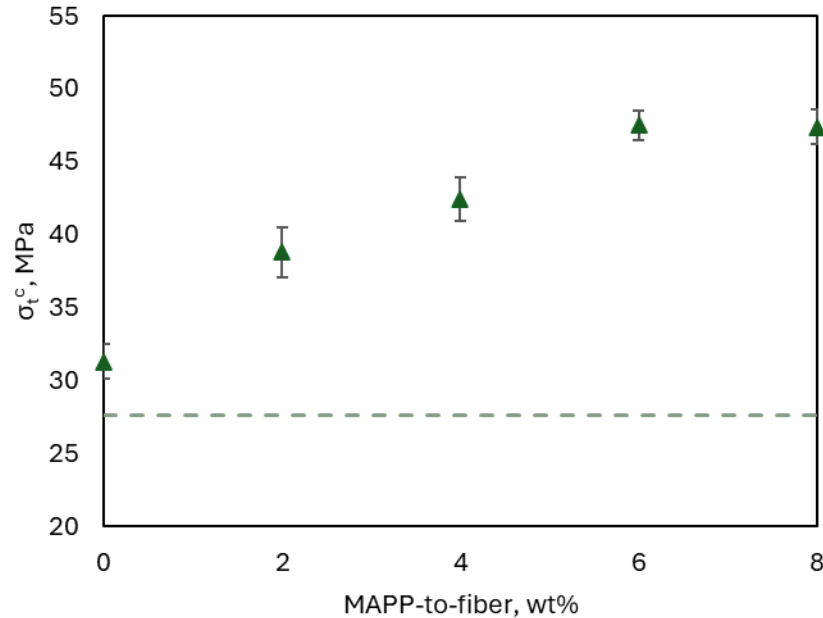
Reinforcing polyolefins: commercially available coupling agents

MA-g-polyolefin approach

Example: MAPP (maleic anhydride-*grafted*-polypropylene)



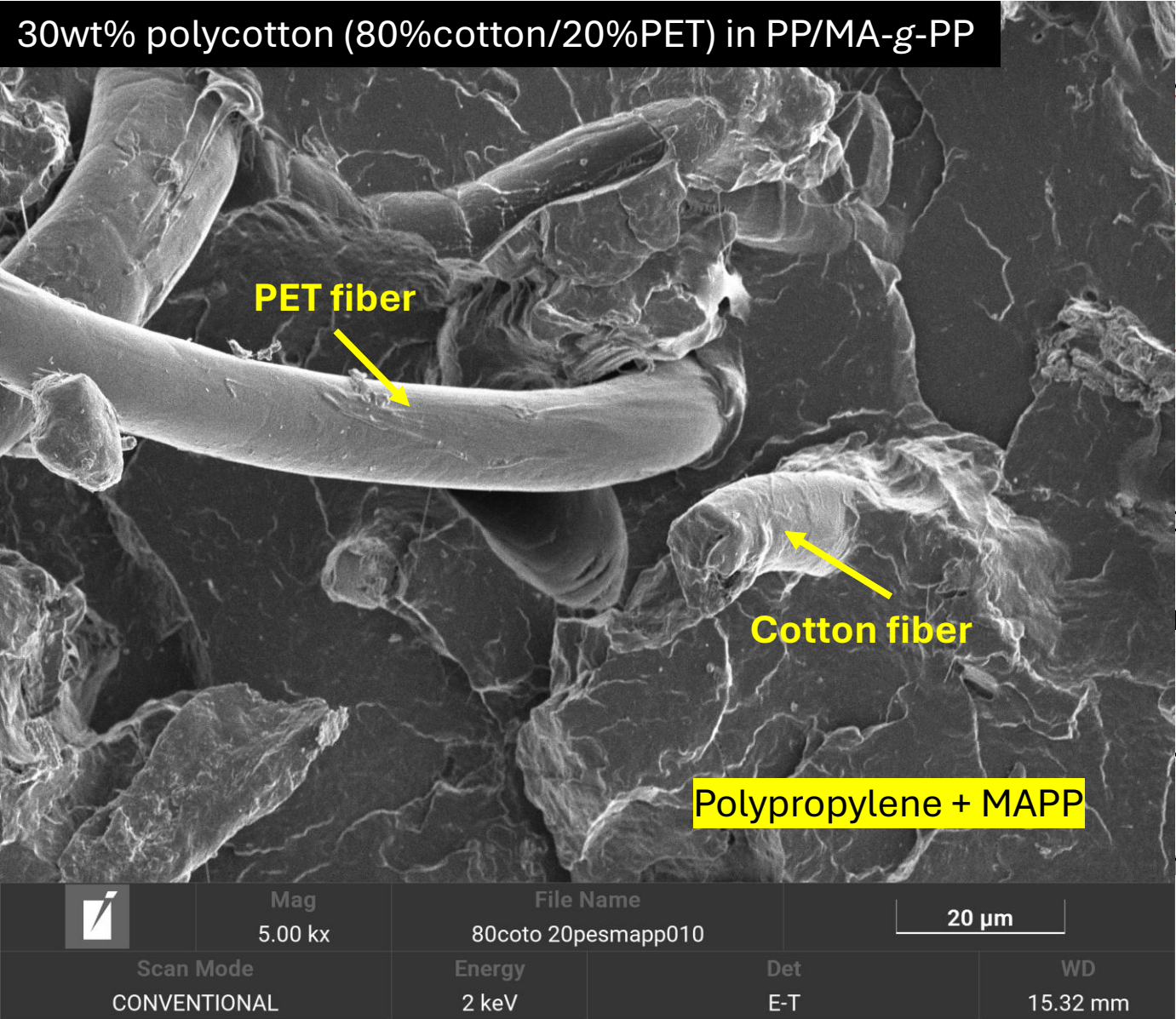
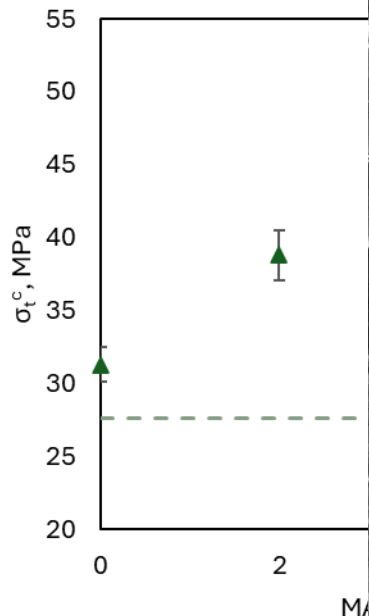
Reinforcing polyolefins: commercially available coupling agents



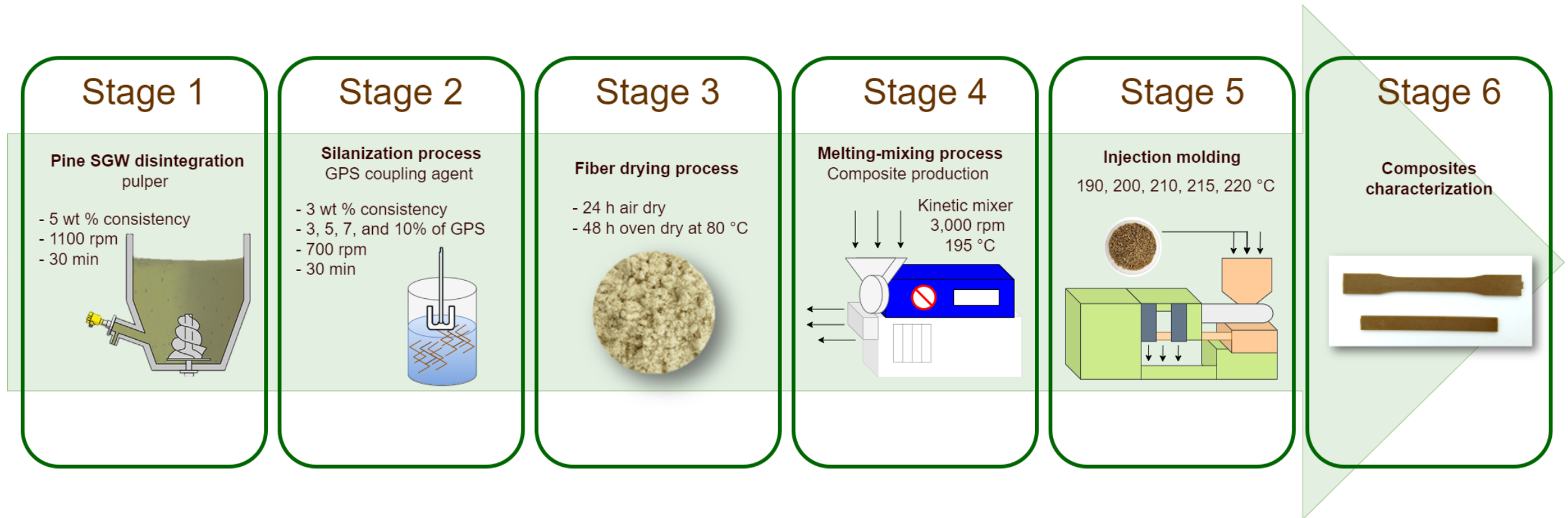
- Available hydroxy groups: $3.5 \cdot 10^{-6}$ mol/g
- Esterification capability of MAPP (15 mg KOH/g acid number): $2.68 \cdot 10^{-4}$ mol -OH per g of MAPP (1:2)
- At a MAPP-to-fiber dosage of 6 wt%: $1.6 \cdot 10^{-5}$ mol -OH/g fiber.

This can lead to MAPP self-enganglement and concentration capping.

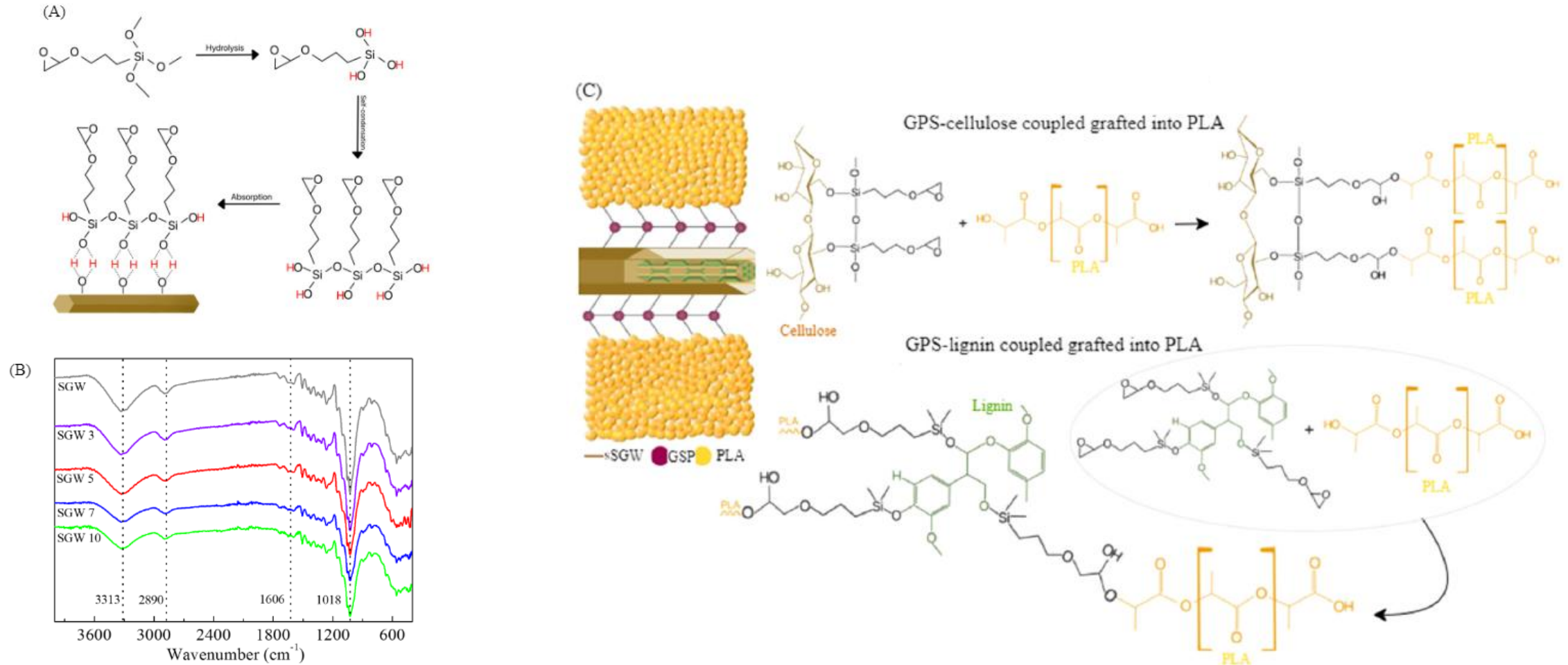
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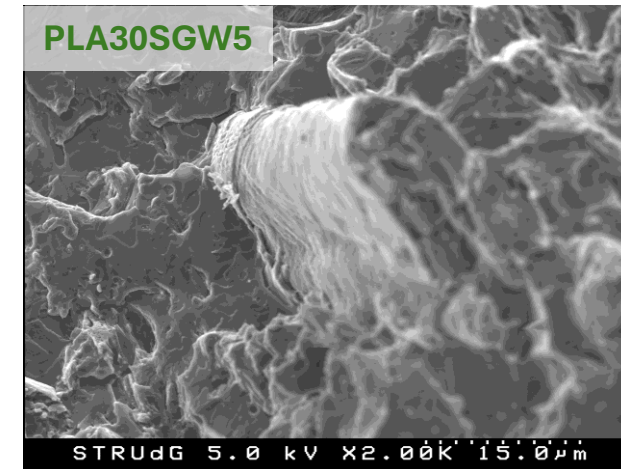
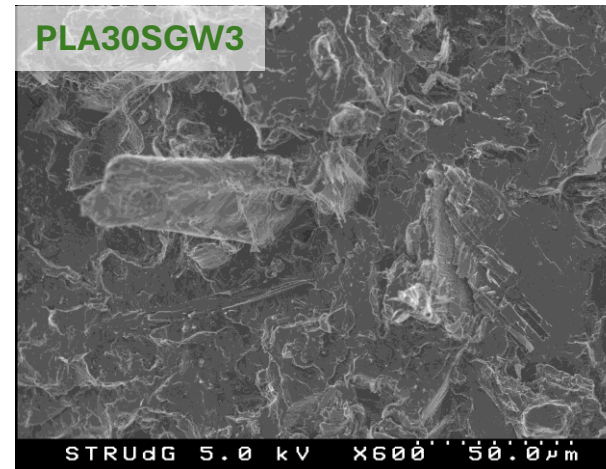
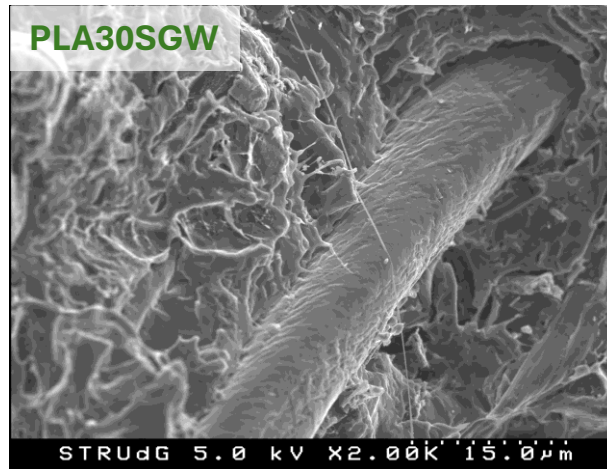
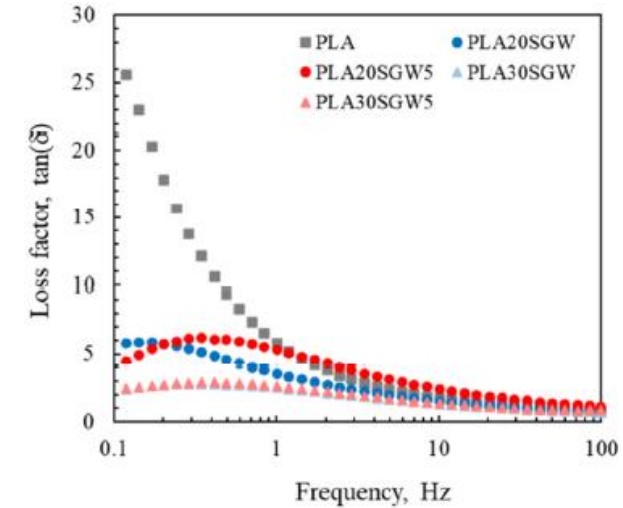
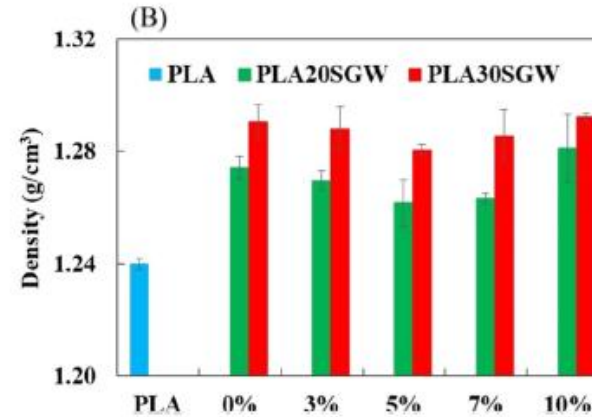
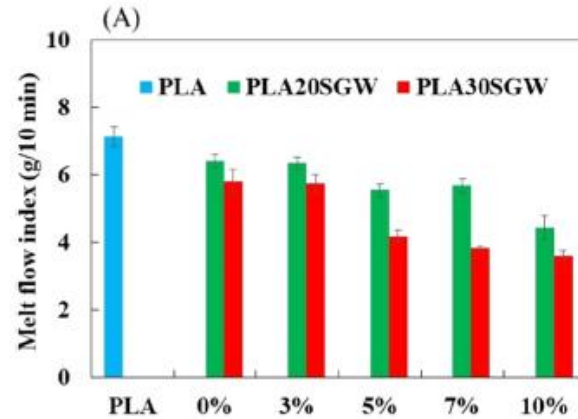
Silane-coupling for PLA biocomposites



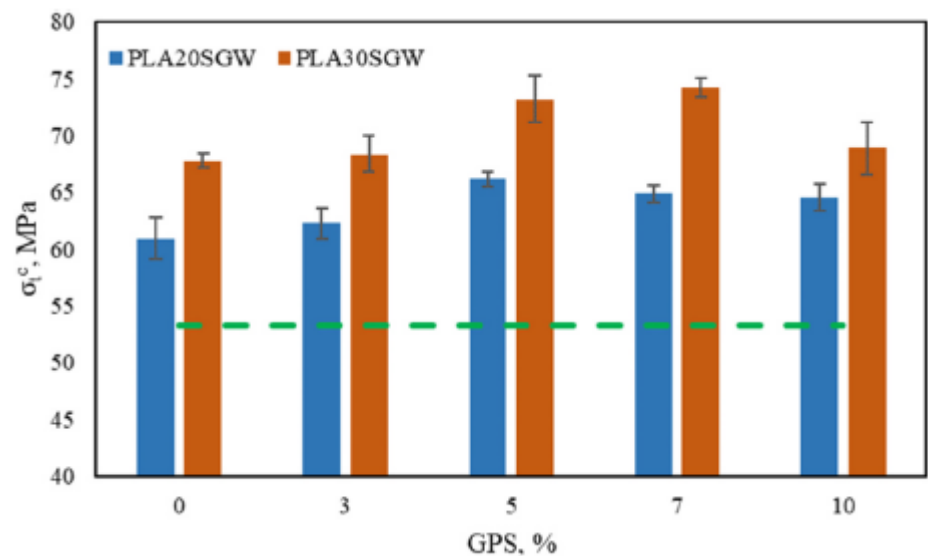
Silane-coupling for PLA biocomposites



Silane-coupling for PLA biocomposites



Silane-coupling for PLA biocomposites



Mean FTSF, E_t^F , σ_t^F and f_c computed by the mRoM and the Hirsch models.

GPS (wt%)	E_t^F (GPa)	σ_t^F (MPa)	FTSF (MPa)	f_c
0			117.52	0.15
3			125.66	0.16
5	27.18 ± 4.01	768.91 ± 37.39	144.61	0.19
7			144.64	0.19
10			134.09	0.17

Micromechanical parameters of PLA/SGW composites at increasing SGW and GPS content.

GPS (wt%)	V^F	τ_T (MPa)	τ_{VM} (MPa)	L_c^F (μm)	χ_1^*	χ_2^*
0	0.151	30.47	35.18	312	0.344	0.432
	0.229	33.89	39.13	276		
3	0.151	31.13	35.94	296	0.386	0.379
	0.229	34.18	39.47	280		
5	0.151	33.08	38.19	283	0.388	0.442
	0.229	36.60	42.26	243		
7	0.151	32.43	37.45	294	0.376	0.523
	0.229	37.11	42.85	249		
10	0.151	32.30	37.29	290	0.353	0.542
	0.229	34.45	39.77	271		

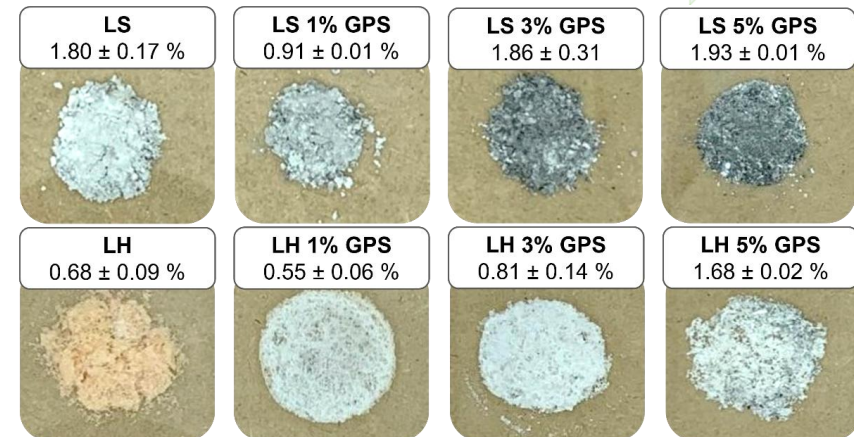
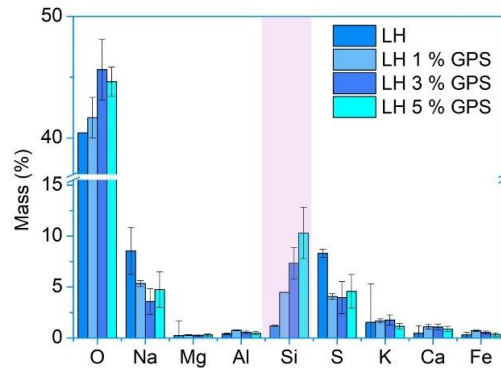
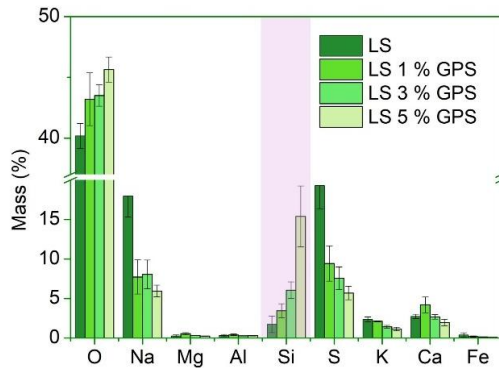
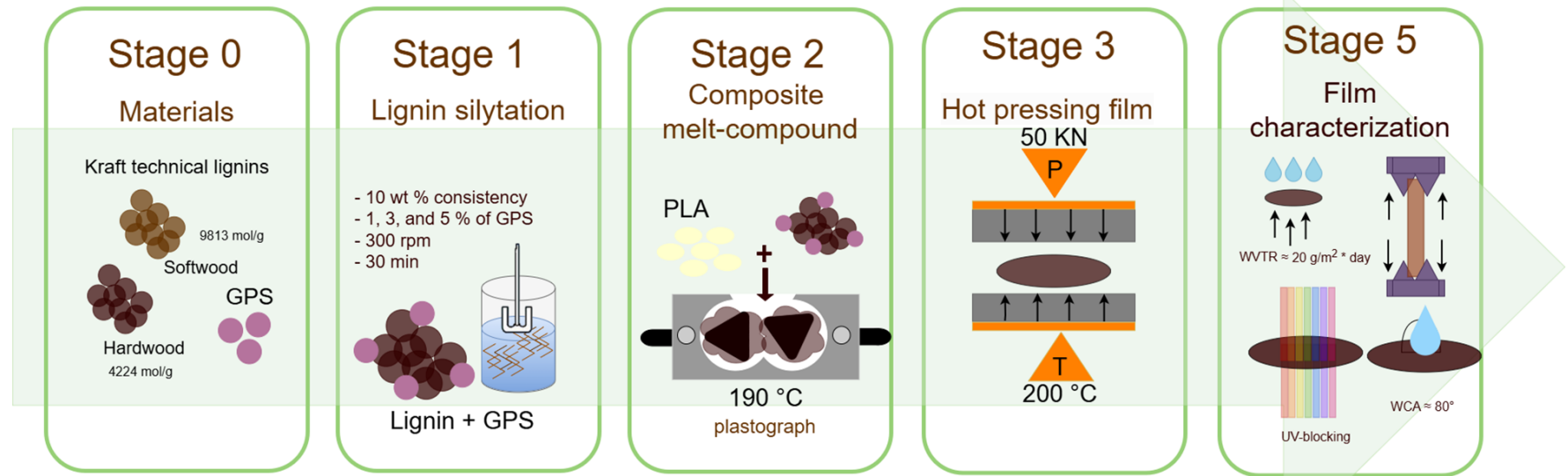
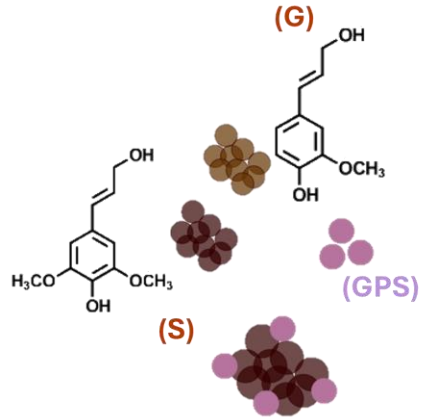
* Calculated using the Von Misses criterion.

$$\sigma_t^c = FTSF \cdot V^F + (1 - V^F) \sigma_t^{m*}$$

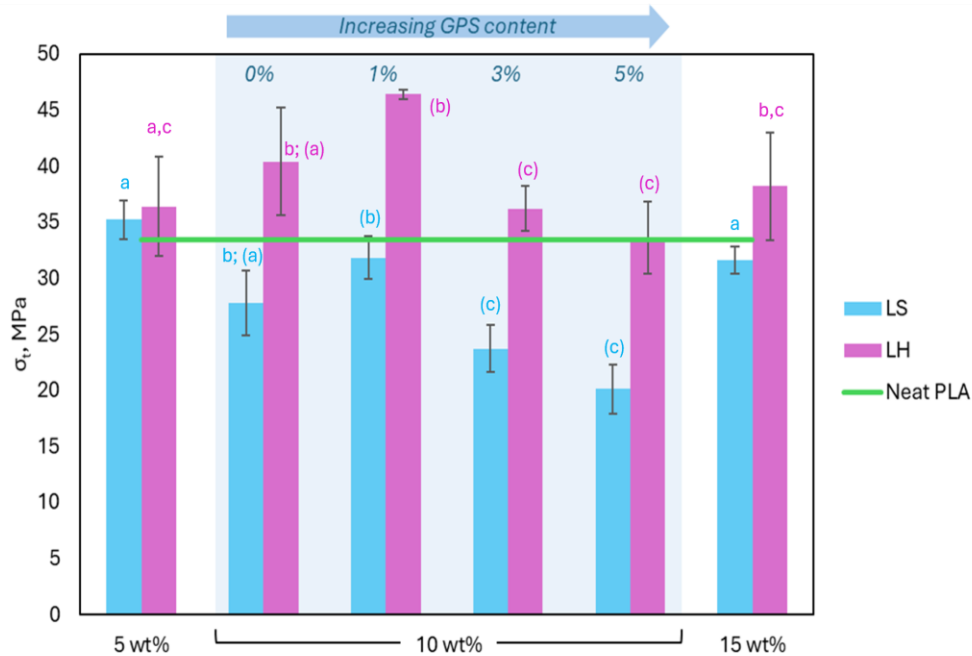
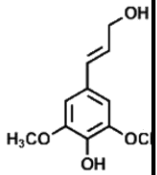
$$FTSF = f_c \cdot \sigma_t^F$$

$$E_t^c = \beta \left[E_t^F \cdot V^F + E_t^m (1 - V^F) \right] + (1 - \beta) \frac{E_t^F \cdot E_t^m}{E_t^m \cdot V^F + E_t^F (1 - V^F)}$$

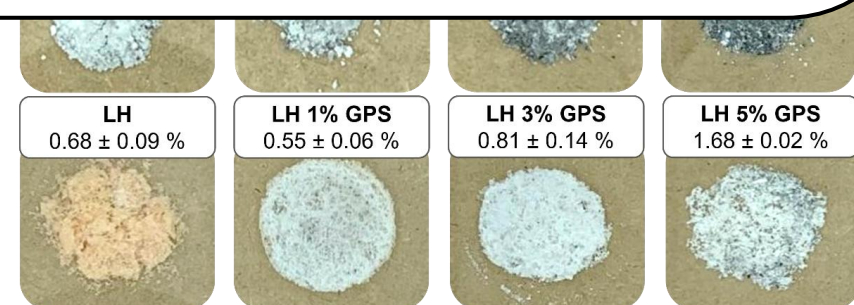
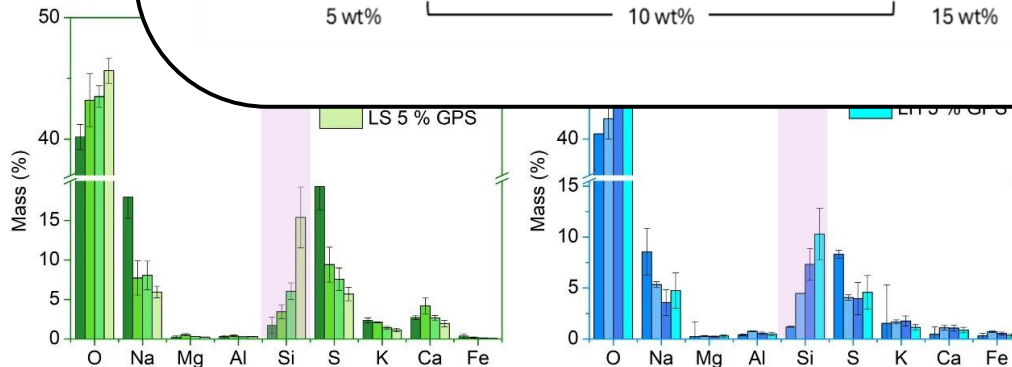
Beyond mechanical performance



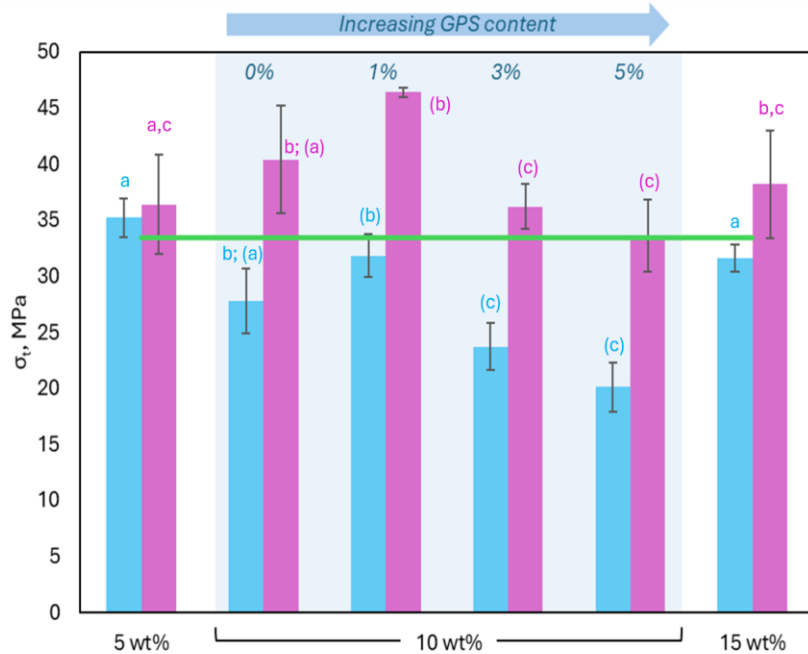
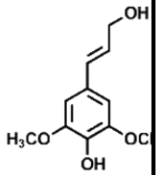
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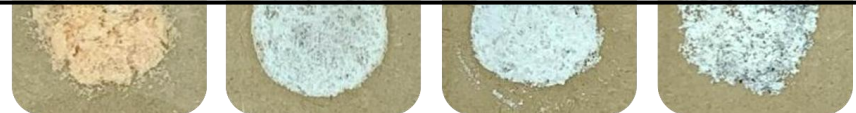
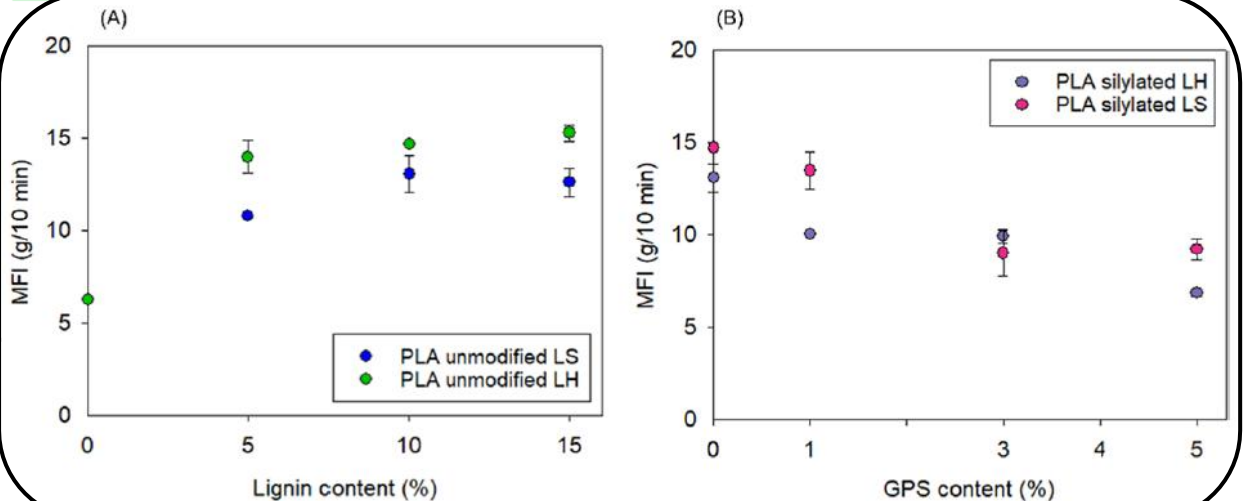
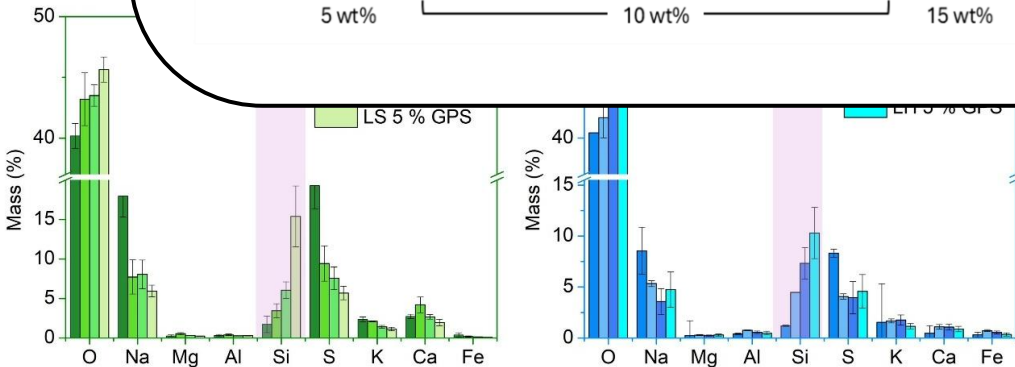
- The source of lignin has an influence on mechanical performance.
- In softwood-derived lignin (LS), mechanical interlocking may be driving reinforcement.
- In hardwood-derived lignin (LH), interfacial bonding appears to be promoted, as well as a better dispersion within the PLA matrix.



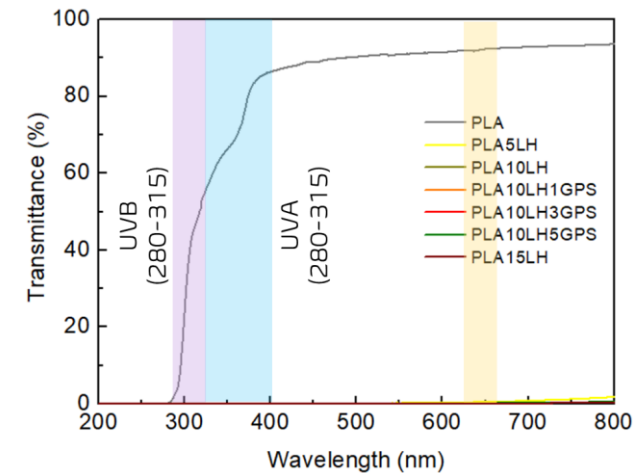
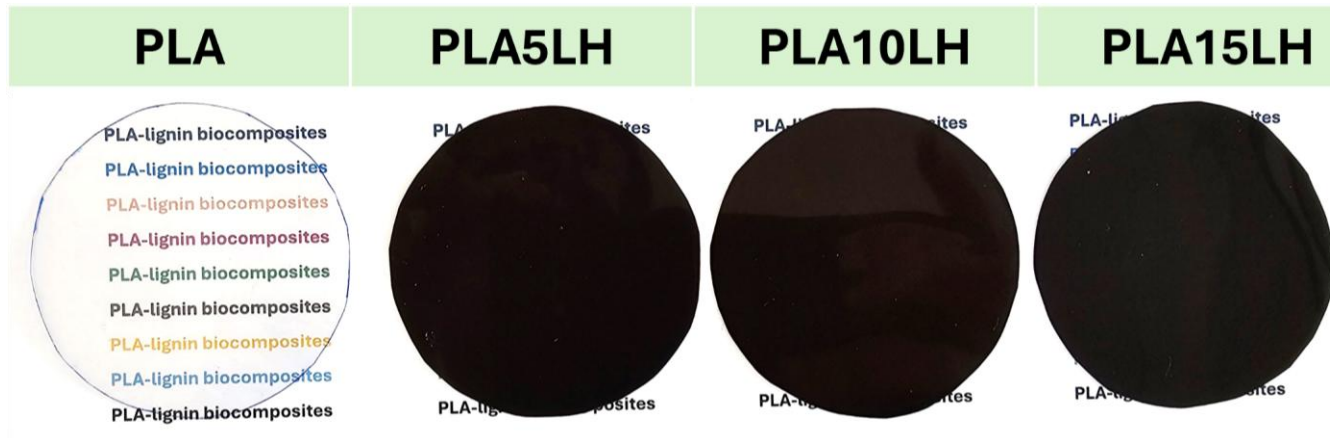
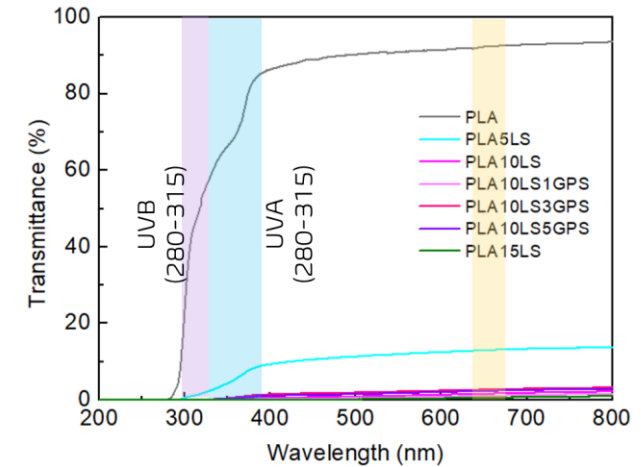
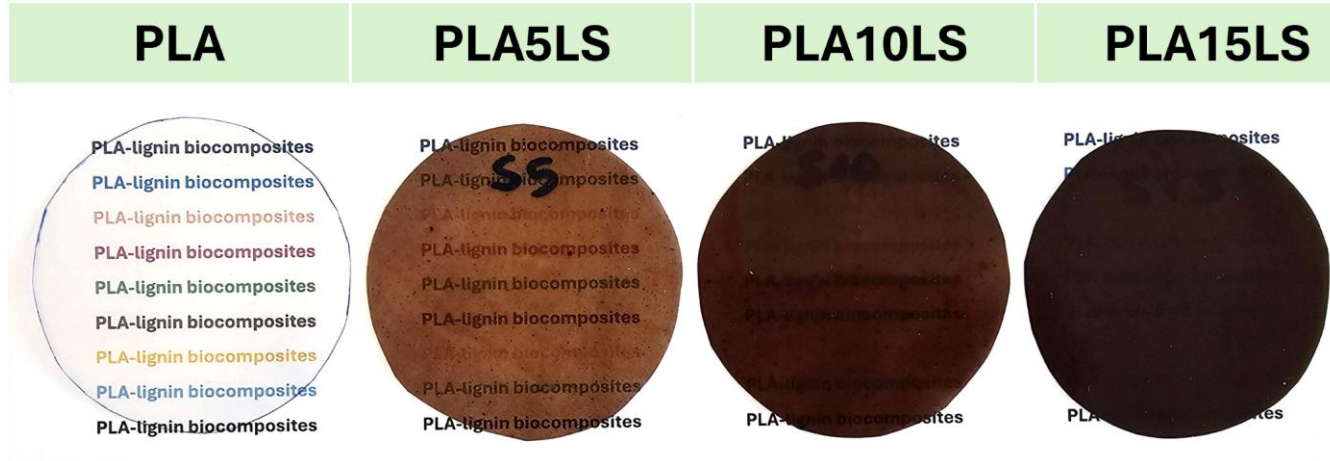
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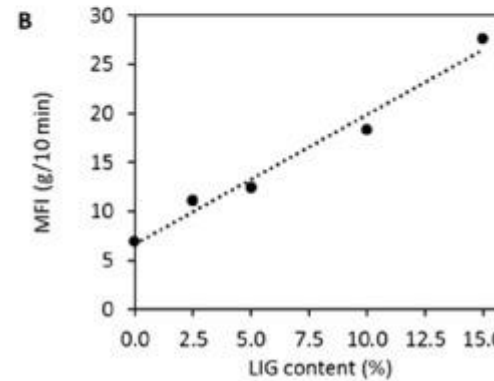
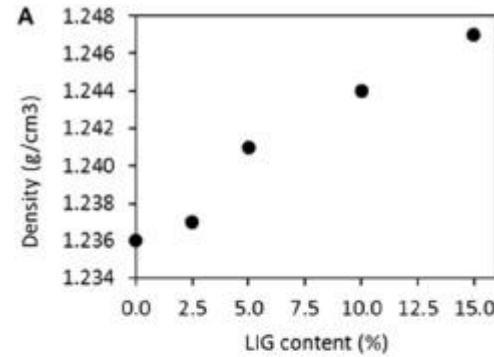
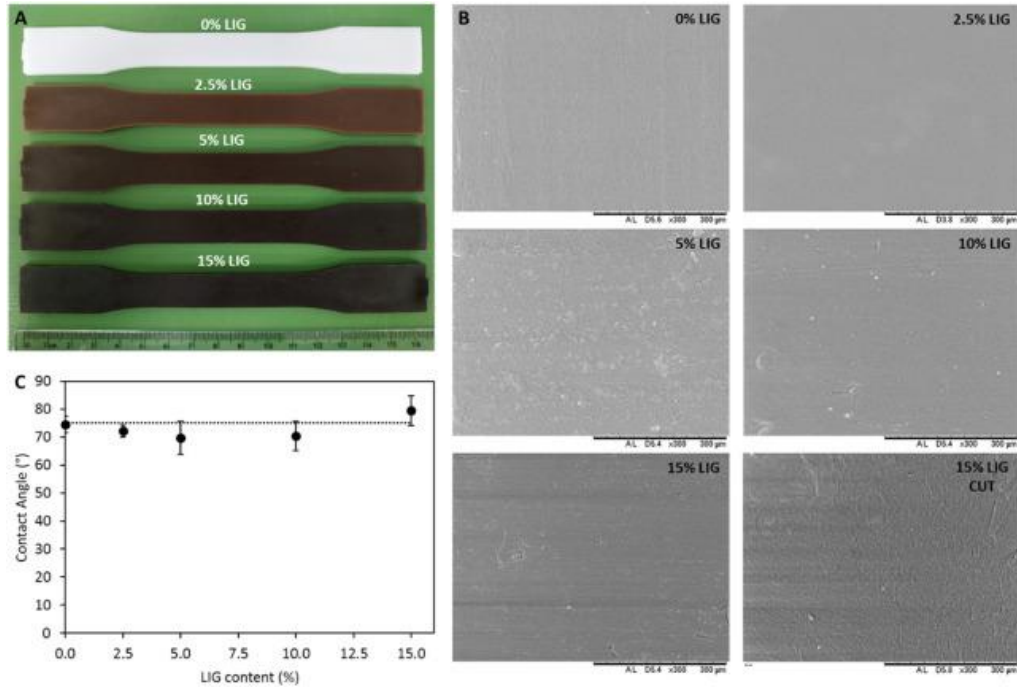
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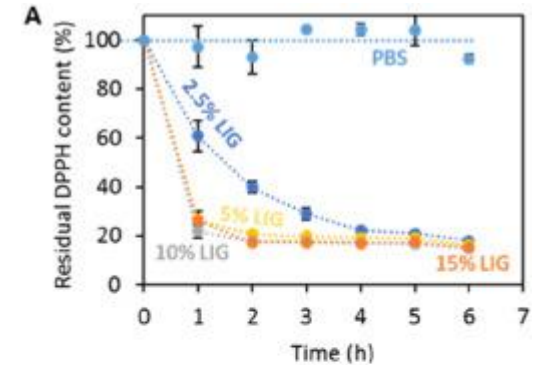
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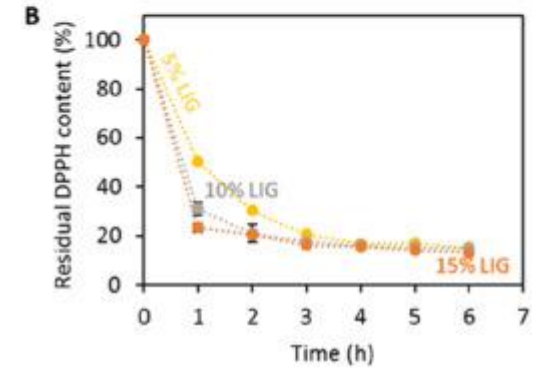
Beyond mechanical performance



Antioxidant activity



Low DPPH concentration



High DPPH concentration

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In a nutshell:

Nature as the ultimate template

- Wood proves that structural performance does not require homogeneous materials: it requires hierarchical and multihasic organization.
- Sustainable engineering is not about simplifying the material, but about mastering its heterogeneity.

The liquid legacy determines solid performance

- The macroscopic strength of a biocomposite is often decided before the material ever solidifies.
- Colloidal strategies (e.g., Pickering emulsions, PEG carriers) are essential to pre-structure the interface and prevent reinforcement collapse, especially in high-surface reinforcements.

Interface engineering is non-negotiable

- Thermodynamic incompatibility is the primary failure mode.
- Coupling agents and brute chemical force are the most common strategies to enhance IFSS.

Beyond mechanical performance

- Biocomposites can also be functional: UV shielding, moisture barrier, antioxidant, antimicrobial...

Laboratory of Sustainable Materials and Product Design

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Thank you

Reach us at:



lepamap.prodis@udg.edu



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Engineering of cellulose-based multiphasic systems: bridging colloidal systems, interface science and bio-composites

Marc Delgado-Aguilar

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*AMADE Winter Day, Girona
January 23, 2026*