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Perspectives on the robustness of the mechanical properties assessment of biocomposites

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ABSTRACT

Biocomposite materials made of natural plant fibers are becoming a viable alternative to the use of synthetic ones such as glass fibers thanks to many economic, ecological, and technical benefits. However, their massive use in the industry requires optimal control of their mechanical performances, which constitutes a real scientific issue to be overcome. Indeed, biocomposites suffer from an important variability in their mechanical properties because of their multiscale structure, the natural growth conditions, the various processing parameters in addition to eventual chemical treatments. Biocomposites are also highly sensitive to the surrounding environment in terms of humidity and temperature because of the hydrophilic properties of natural plant fibers. In this context, this perspective paper aims to provide a critical look at the influence of the main factors that affect the mechanical properties of biocomposites in order to suggest some possible research outlooks that could contribute to optimizing the control of these mechanical properties and expanding the use of biocomposites in industry.

KEYWORDS

Biocomposites; Plant fibers; Moisture; Mechanical properties; Variability; Humidity; Hygrometry; Temperature.

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1. Introduction

Global warming and climate change are nowadays the concern of the political and economic world. The main challenge is to reduce greenhouse gas emissions through the development of eco-friendly materials and sustainable manufacturing processes [1]. To reach this objective, many industrial sectors are moving toward a bio-economy by using biomaterials such as biocomposites instead of conventional synthetic composites made with glass or carbon fibers [2,3]. Biocomposites are elaborated with natural plant fibers such as flax, hemp, or jute fibers. Biocomposites can be called "green composites" if the polymer matrix is also biobased such as the polylactic acid (PLA). The resulting green composites are then 100% biobased and compostable [4].

Natural plant fibers can mechanically compete with synthetic glass fibers in terms of stiffness. Indeed, bast fibers such as flax or hemp are characterized by an elastic modulus comparable to that of glass fibers while being lighter [5]. This results in a specific elastic modulus of bast fibers greater than that of glass fibers, which is a significant parameter in choosing materials that meet the new environmental constraints in terms of energy efficiency and CO_2 emission.

However, the assessment of the mechanical properties of biocomposites is still challenging because of the complex structure of plant fibers. Indeed, natural plant fibers suffer from different issues, typically the variability of their mechanical properties resulting from their natural characteristics, their growth conditions, and their multiscale complex cellulosic structure, which implies the consideration of the analysis scale for the mechanical characterization [6]. Furthermore, the hydrophilicity of plant fibers due to the presence of hydroxyl groups in their cell walls leads to poor interface generation with polymer matrices and moisture absorption into the fiber structure from a humid environment [7]. This moisture absorption contributes in general to a modification in the mechanical properties of the resulting composite depending on the analysis scale due to different mechanisms such as plasticization, creeping of microfibrils, and polymer hydrolysis [8]. The moisture diffusion into the composite is also accelerated by the increase in the surrounding temperature. Therefore, considering the hygrothermal aspect in the mechanical characterization of biocomposites at different scale levels is mandatory to conduct an efficient analysis and to be able to improve correctly the mechanical properties of these eco-friendly materials.

In this context, this perspective paper aims first to give a critical overview of the multiscale mechanical properties of biocomposites, the hygro-mechanical properties in addition to the hygrothermal coupling effect. Then, a discussion will be carried out on the possible solutions to analyze efficiently the mechanical properties of biocomposites by considering the multiscale hygrothermal effect in order to master, control, and improve the functionalities of biocomposite parts and meet the industrial specifications.

2. Multiscale mechanical properties of biocomposites

As shown in Figure 1, natural bast fibers are extracted from plant stems in the form of bundles that are located at the periphery [9], playing the role of mechanical support for the stem. The extraction is in general carried out mechanically by scutching to separate the bundle from the rest of the stem [10]. Fiber bundles are then subjected to hackling for separating them into smaller bundles that are called technical fibers which are the conventional form of natural fibers used in polymer composites. Each technical fiber is composed of a few elementary fibers gathered together via pectic interfaces [9].

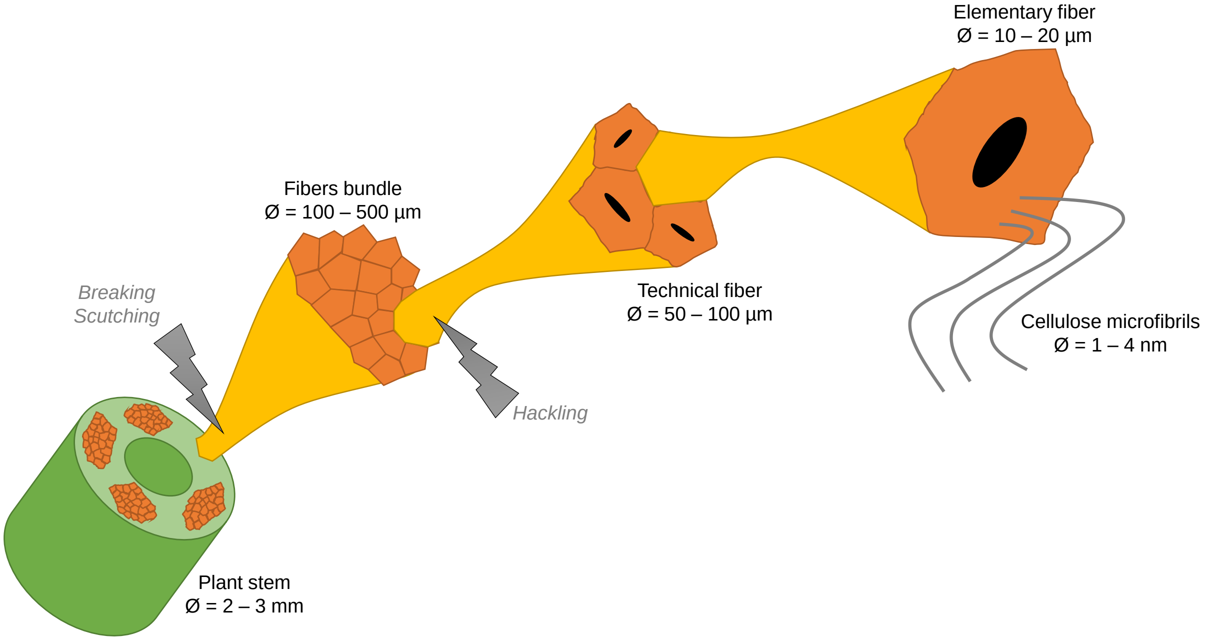

Figure 1: Multiscale structure of natural bast fibers from the plant stem to cellulose microfibrils. Reproduced with permission from Encycl. Mater. Compos. 3, 168–185 (2021) [11]. Copyright 2021 Elsevier

Elementary fibers present a multiscale complex structure composed of different cell walls as shown in Figure 2. The cell wall S2 is the most relevant as it is the thickest. The cell wall S2 can be considered as a natural composite at the nanoscale with cellulose microfibrils as reinforcement and hemicellulose with lignin as a polymeric matrix [12]. Cellulose microfibrils are oriented helicoidally in the cell wall S2 with an angle θ called the microfibrillar angle (MFA).

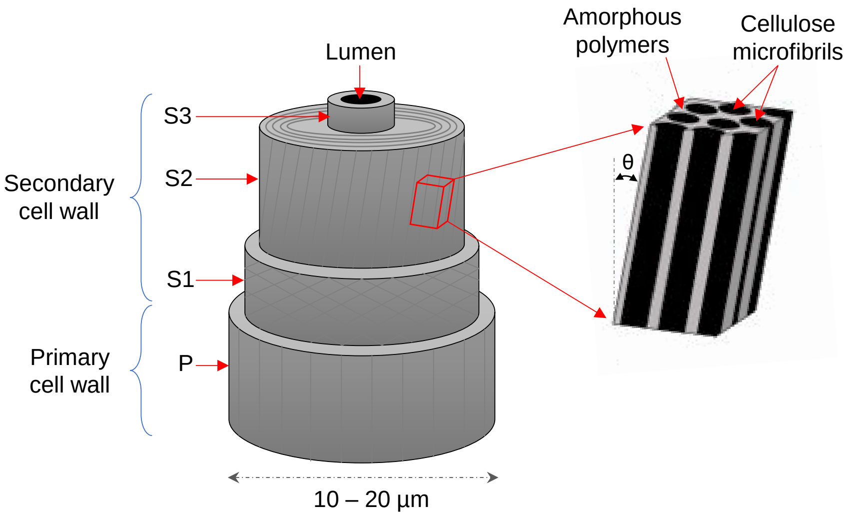

Figure 2: Multiscale cellulosic structure of an elementary bast fiber. Reproduced with permission from Int. J. Adv. Manuf. Technol. 105(1–4), 1549–1561 (2019) [13]. Copyright 2019 Springer Nature

Therefore, many key factors can influence the mechanical properties of the resulting biocomposites, among which we can find first the growth conditions of the plants, the multiscale cellulosic structure of the plant fibers, the extraction conditions from the plant stems, the eventual chemical treatment of the fibers, the environmental conditions in terms of humidity and temperature, and the processing parameters of the biocomposite parts. All these factors and their effects lead to induce a high variability in the mechanical properties of plant fibers and complicate hence an efficient assessment of the mechanical properties of the resulting composites. Indeed, the complexity in the case of natural plant fiber lies in the fact that the main components of an elementary fiber (cellulose, hemicellulose, lignin) are present in variable proportions for the same fiber type [14], which impacts the quality of the resulting fibers. It is dependent on many factors such as the climatic condition of growth (rainfall, sunshine, …), the cultivation conditions (compost, fertilizer, …), and the harvesting process [15]. This variability in the composition contributes to the variability in the mechanical properties obtained by conventional tensile tests because the mechanical properties of cellulose, hemicellulose, lignin, and pectin show a highly significant difference [14,16]. For example, it has been shown in a literature review [5] that the tensile modulus of flax fibers varies from 28 to 100 GPa while their tensile strength varies from 343 to 1035 MPa. For hemp fibers, the tensile modulus varies from 32 to 60 GPa while the tensile strength varies from 310 to 900 MPa. For Jute fibers, the tensile modulus varies from 25 to 55 GPa while the tensile strength varies from 393 to 773 MPa. The variability is among the important issues that the composite industry can face because the mechanical properties of biocomposite parts must meet the industrial standard that cannot be adapted each season to the updated mechanical properties of the cultivated plant fibers. This issue limits their massive use in industrial applications.

On the other side, the multiscale structural organization of biocomposites from cellulose microfibrils of elementary fibers to the whole macrostructure of the composite leads to an important variability of the mechanical properties in the same composite material depending on the testing method and the analysis scale. It has been demonstrated in previous work [6] that the increase of the geometrical contact scale when performing nanoindentation experiments on flax fibers (i.e. the increase of the tip indenter radius) contributes to an important increase of the elastic modulus due to the increase of the cellulose microfibrils involved by compression on the mechanical contact as shown in Figure 3, while the elastic modulus of polypropylene matrix (PP) has not revealed a significant variation when increasing the edge radius of the tip indenter. Tensile tests provide higher elastic modulus values because all cellulose microfibrils of the elementary fiber are involved by tension to mechanical contact.

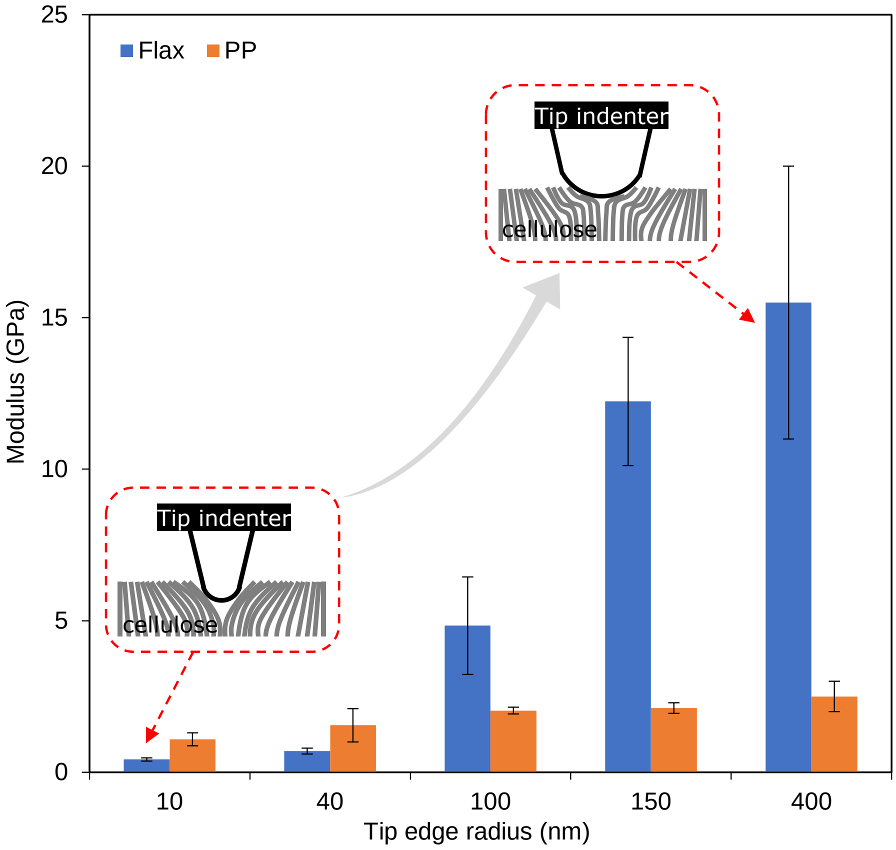

Figure 3: Elastic modulus obtained by nanoindentation of flax fibers and PP matrix with different tip edge radii. Reproduced with permission from Encycl. Mater. Compos. 3, 149–158 (2021) [17]. Copyright 2021 Elsevier

By going toward the mesoscale, the mechanical properties of technical fibers will depend on the number of elementary fibers in the bundle which is randomly variable as shown in the example of Figure 4 for a composite made with unidirectional flax fibers and polypropylene matrix. The mechanical properties of technical fibers differ from those of elementary fibers because technical fibers are bonded assemblies of elementary fibers with a hierarchy of interfaces [15]. At the macroscale, the mechanical properties of the composite could be affected by the size of the technical fibers which is variable because of the variability of elementary fibers' shape and diameter as shown in Figure 4. It can be seen in this figure that flax fibrous reinforcement can also be present in the form of elementary fibers, which depends on the quality of the hackling process during the extraction step (see Figure 1).

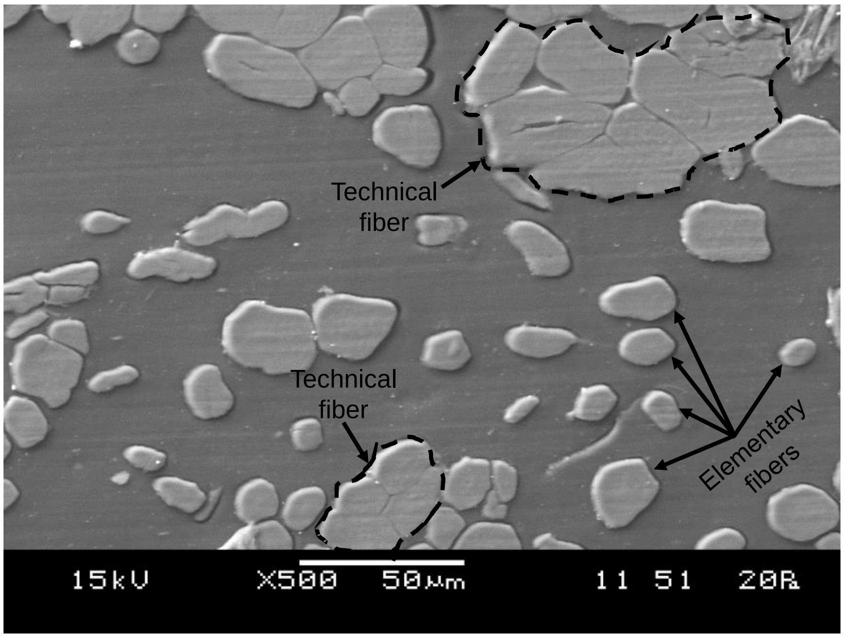

Figure 4: Example of a flax fibrous reinforcement structure in a composite, showing the random distribution and shape of flax fibers

Mechanical properties of biocomposites are then influenced by the natural character of plant fibers and the mechanical analysis scale. The natural growth process of plant fibers is difficult to control because it depends on the climatic and earth conditions of growth. Therefore, it cannot be guaranteed that plant fiber cultivation of the current year will be similar to that of the previous year or next year [18]. For all these reasons, plant fiber production industries must harmonize and master the mechanical properties of plant fiber to be able to provide standard technical fibers with controlled specifications. Mixing the cultivation of several successive seasons could reduce the apparent variability in the mechanical properties. However, this variability will still exist at the microscale in the fibrous structure and will consequently increase the random anisotropy of the biocomposite.

In our point of view, in-depth scientific work should be carried out first to determine if there is a functional relationship between the growth conditions of plant fibers and their resulting mechanical properties and/or their chemical composition by keeping constant the extraction process. If so, this will indicate that maximum control of growth conditions of plant fiber must be carried out and this is obviously not an easy task to achieve. Controlling efficiently the growth conditions means applying artificial climatic conditions in terms of temperature, humidity, and watering instead of natural ones by carrying out the cultivation in greenhouses, which could raise considerably the production cost. Moreover, sun radiation is complicated to reproduce artificially and cannot be controlled similarly each year, which may induce variability in the microstructure of plant fibers and their mechanical properties. Therefore, the effect of sun radiation on the microstructure and mechanical properties of plant fibers must be investigated in depth to determine if this natural parameter could have a significant impact on the variability.

To conclude about the effect of growth conditions, we think that the first mandatory research work to do is the determination of the optimal growth conditions in terms of humidity, temperature, watering, sun radiation, and fertilizers to reach the optimal mechanical properties of plant fibers at the different specific scale levels discussed earlier. The results of these optimizations could be then used to develop a smart cultivation system with artificial intelligence (AI) techniques. This smart system should be able to adapt the growth parameters to the optimal values depending on the actual climatic conditions of each season and each year. In this way, the mechanical properties of plant fibers should be not only stable but also optimized.

The extraction conditions as well as possible treatments before extraction can also affect the mechanical properties of plant fibers and the resulting composite. Indeed, and as shown in [19], chemical treatment such as aqueous ammonia has shown its potential to reduce fiber damage during the extraction process, which leads to improving the mechanical properties of plant fibers. Ammonia treatment can modify the cellulose crystalline packing and dissolve lignin in the biomass [20]. The question here is: is there any functional relationship between the treatment parameters and the variation of the chemical composition as well as the microstructure of plant fibers? Answering this question can bring some perspectives to use the chemical treatment with the aim of adjusting the micro-composition of plant fibers to control their mechanical properties.

Chemical treatment has been also used to improve the interface quality between plant fibers and polymer matrix. Indeed, the effectiveness of the interface between plant fibers and the polymer matrix is subjected to different issues related to the presence of hydrophilic hydroxyl groups on the surface of plant fibers that are not compatible with the hydrophobic nature of the matrix [21]. Several chemical treatments, such as Alkaline treatment, have been developed and investigated on plant fibers to improve fiber adhesion properties with the polymer matrix [21,22]. As an example, it has been shown that an alkaline treatment on flax fibers can increase the mechanical properties of flax fiber reinforced epoxy composites in terms of tensile modulus and tensile strength by around 30% [23].

3. Hygromechanical and hygrothermal characteristics of biocomposites

As noticed in the introduction, elementary plant fibers are characterized by a multiscale cellulosic structure with cellulose microfibrils embedded in natural amorphous polymers of hemicellulose, lignin, and pectin [7,24]. These natural components give plant fibers a hydrophilic character with the ability to absorb water molecules due to the presence of hydroxyl groups, which increases the water content of plant fibers when they are in a humid environment [25,26]. Plant fibers are mainly responsible for water absorption in the biocomposites as shown in the examples of Figure 5(a) for long fiber reinforced thermoset composites and Figure 5(b) for short fiber reinforced thermoplastic composites. Indeed, the moisture content absorbed by the neat polymers is lower than that of polymers reinforced with flax fibers. Moreover, the moisture content increases significantly with the increase of the volume fraction of flax fibers.

Several literature studies demonstrated that the moisture content leads to modifying the mechanical properties of biocomposites, and this hygrometric impact has shown a scale effect. At a macroscopic scale, the moisture content seems to deteriorate the mechanical properties of biocomposites in terms of elastic tensile modulus as shown in Figure 5(c,d), which is attributed to the fact that water molecules act as a plasticizer when they are bonded to the hydroxyl groups in the hydrophilic components of plant fibers [27–29]. Regarding the effect of moisture content on tensile strength, the different works in the literature show contradictory results. Some experiments conducted by different lab groups found that the moisture content leads to a decrease in the tensile strength of the biocomposite because of interface weakening, fiber plasticization, and polymer hydrolysis that contributes to the breakage of its molecular chain [26,27,30–34]. On the other hand, other works have demonstrated that the tensile strength increases with the increase of the moisture content [35–37]. This particular behavior was explained by the fiber swelling due to moisture absorption, which could strengthen the interfacial bonding between plant fibers and polymer matrix because of the radial expansion of elementary fibers [29,36,38]. However, this suggestion is uncertain because it has been shown in other work that the interlaminar shear strength decreases with the increase of water content [35]. These divergent literature findings prove that the hygromechanical behavior of biocomposites is highly complex to master because different phenomena can be involved at different scale levels depending on different factors such as the fiber processing parameters, the fiber treatment, the matrix choice, and the processing parameters of the composite.

To discuss these complex hygromechanical behaviors, it is important to notice that all these considered factors can determine the properties of the interfaces between plant fibers and the polymer matrix. Therefore, depending on the quality of the resulting interfaces, the moisture diffusion rates through the fibers and through the interfaces will be modified, which will hence modify the mechanisms by which the moisture impacts the mechanical performance of the biocomposite. Indeed, if the processing of the biocomposite generates high-quality interfaces, moisture diffusion will be carried out through the fibers because there will be less damage on the interfaces, such as voids or micro-cracks, from which the water molecules could be diffused. In this case, the radial swelling of fibers due to water uptake will strengthen the adhesion between fibers and the matrix in the interface zone, which will lead to an enhancement of the mechanical tensile strength of the biocomposite at the macroscale. However, if the processing parameters are not optimized for the biocomposite, the interfaces will be generated with damage, which will facilitate water diffusion through the interfaces. As a consequence, more degradation of the interfaces will occur with the presence of water due to the hydrolysis process, and the swelling of fibers will intensify this degradation because of the induced compressive stresses. This will result in a decrease in the mechanical tensile strength of the biocomposite at the macroscale.

For more understanding of the hygromechanical behavior of biocomposites, it is important to look at the microscale toward experimental investigations that have been carried out on isolated plant elementary fibers and have highlighted that their mechanical properties show in general an increase when rising the relative humidity and the water content [39,40]. This specific hygromechanical behavior has been explained by the fact that the water content contributes to a rearrangement of cellulose microfibrils toward the fiber axis, which increases consequently the mechanical properties by hygro-mechanical hardening. Indeed, it has been explained that water absorption could induce the plasticizing of the amorphous matrix of plant fibers (because of water adsorption) and the creep of their cellulose microfibrils in the relaxed amorphous matrix, leading to their re-arrangement, with more parallel orientations to the fiber axis, which contributes to the increase of the fiber stiffness [39].

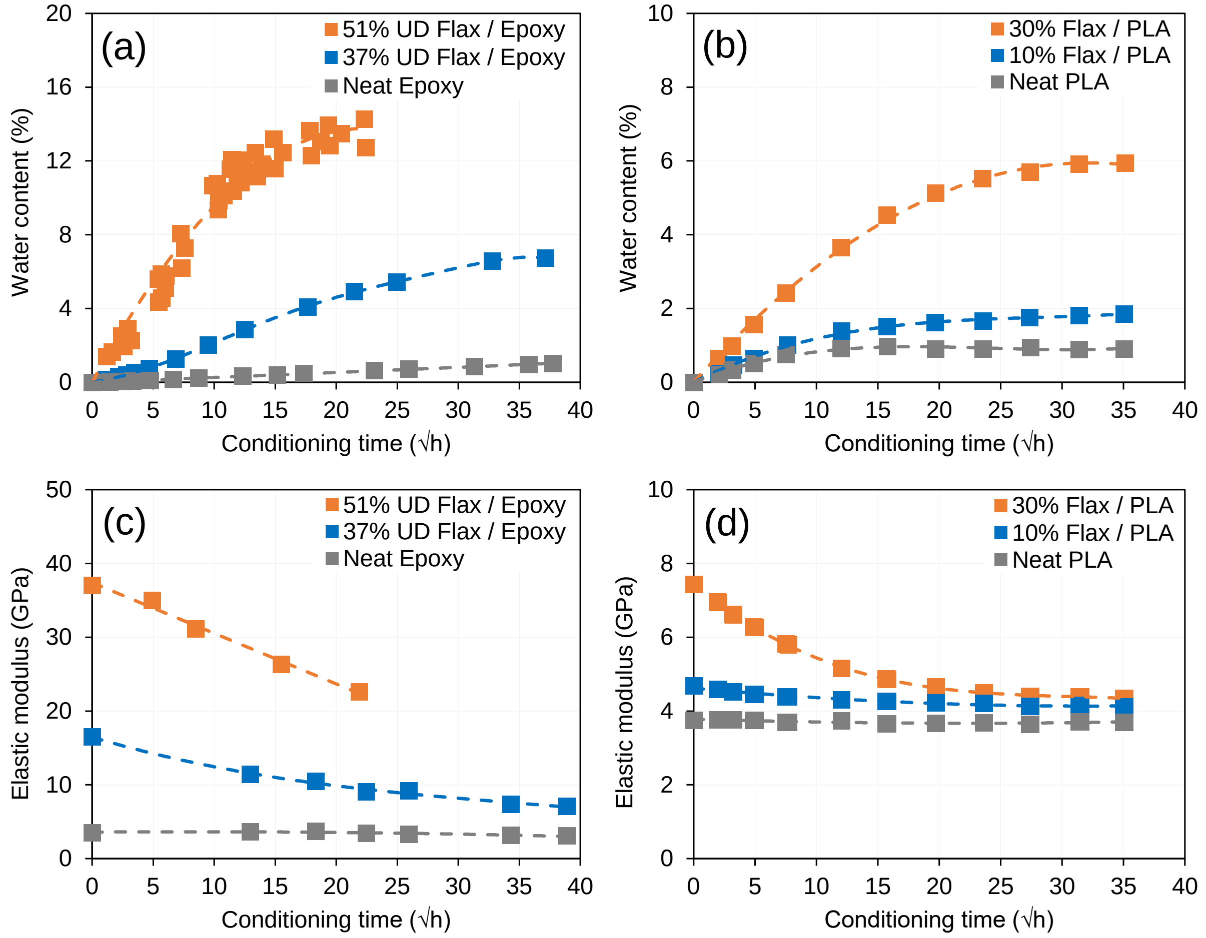

Figure 5: Effect of a humid environment on water content and mechanical properties of biocomposites. (a) and (c) are for long flax fiber composites, while (b) and (d) are for short flax fiber composites. Reproduced with permission from Compos. Part B Eng. 211, 108660 (2021) [8]. Copyright 2021 Elsevier

The hygromechanical behavior of biocomposites becomes more complex when considering also the thermal effect, especially the conditioning temperature. Indeed, increasing the temperature of the hygrometric conditioning leads to an acceleration of the water diffusion into the biocomposite structure [35,41,42]. Figure 6 shows an example of unidirectional flax fiber reinforced epoxy composites with 44% of fiber volume fraction [42]. At 90% of relative humidity (RH), it can be seen from Figure 6(a) that increasing the hygrometric conditioning temperature from 20°C to 40°C can raise the water content at saturation from 4% to 7%. However, the impact of the conditioning temperature on the mechanical properties is not significant as shown in Figure 6(b-c). Only the exposure time to the humid environment induces a decrease of the tensile modulus and tensile strength until reaching the hygrometric saturation of the moisture content. Therefore, since the moisture content should control the mechanical properties of biocomposites [43], why does an increase in the moisture content due to the conditioning temperature have no significant influence on the mechanical properties in Figure 6?

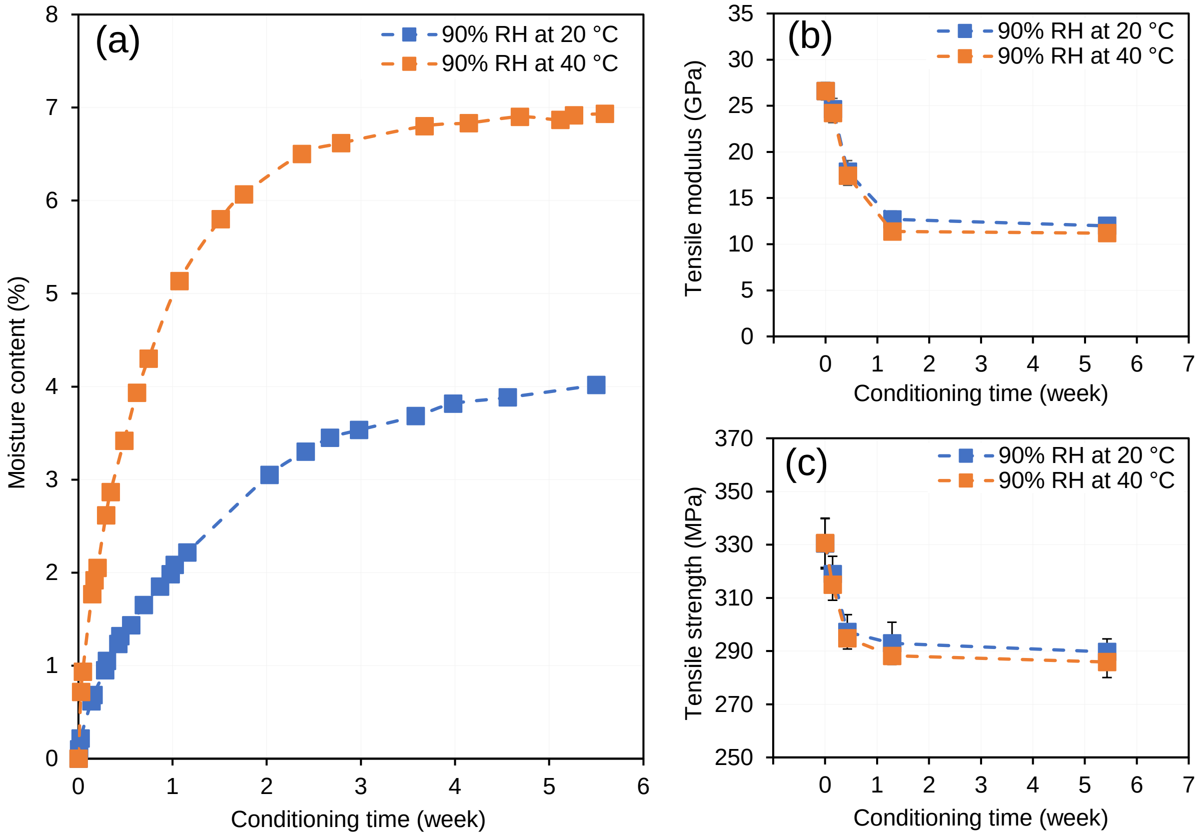

Figure 6: Evolution of (a) moisture content, (b) tensile modulus, and (c) tensile strength in the function of the hygrothermal conditioning time for flax fiber reinforced epoxy composites. Reproduced with permission from Compos. Part B Eng. 48, 51–58 (2013) [42]. Copyright 2013 Elsevier

This specific hygrothermal behavior can be explained when looking into the hygrothermal properties of flax elementary fibers at the microscale. To this aim, Figure 7 has been created using the experimental results of Thuault et al. [44]. It shows the evolution of the tensile modulus and the tensile strength of flax fibers subjected to a temperature increase with two different hygrometric conditions: an initial room RH of 60% at the beginning of the thermal conditioning and a constant RH of 50% during the thermomechanical tests using a climatic chamber [44]. Flax fibers without controlled RH show a drastic decrease of their mechanical properties in the function of temperature increase until reaching 60°C, while flax fibers with a controlled RH show a slight decrease of both tensile modulus and tensile strength. Hence, the presence of a humid environment reduces the thermal effect on plant fibers, which could explain the fact the thermal effect is not significant in the results of Figure 6(b-c). As explained previously, the moisture content in plant fibers contributes to an increase in their mechanical properties due to the rearrangement of cellulose microfibrils. The hygrometric hardening of plant fibers could offset the decrease of their mechanical properties due to the thermal softening of their natural polymeric components.

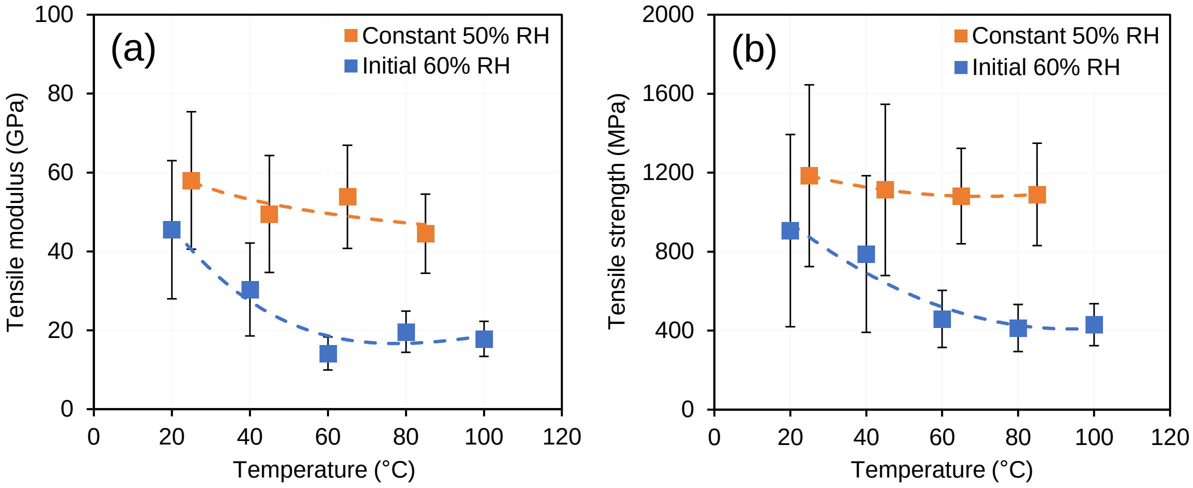

Figure 7: Evolution of (a) tensile modulus and (b) tensile strength of elementary flax fibers subjected to different hygrothermal conditioning.

It is important to note that the effect of the polymer matrix properties should not be ignored. Indeed, the biocomposites used for the results of Figure 6 are made with epoxy, which is a thermoset resin that is not highly sensitive to temperature after its polymerization. Nevertheless, when using a thermoplastic matrix such as polylactic acid (PLA), the thermal softening of the PLA when increasing the conditioning temperature contributes to a significant decrease in the mechanical properties of the resulting biocomposites despite the presence of a humid environment as shown in [41] with flax fibers reinforced PLA composites. Moreover, the hygrothermal conditioning type is a significant parameter that can change the hygrothermal behavior of the biocomposite by affecting the failure mode of the matrix, even for thermoset resin such as epoxy: If total water immersion is used instead of a climatic chamber to accelerate the water diffusion process, it contributes to change the failure mode of epoxy from brittle to ductile after a large water immersion time at 60°C [35], which affects the mechanical properties of the polymer matrix and, consequently, that of the resulting biocomposite.

It can be concluded that biocomposite materials are highly sensitive to hygrothermal conditions and their hygrothermal behavior is highly complex with different physical couplings at each characteristic scale level (Cellulose microfibrils, Interfaces, elementary plant fibers, fiber bundles, the polymer matrix, and the whole biocomposite structure). The hygrothermal characteristics of biocomposites should not be always considered as a negative aspect regarding the mechanical properties. Moisture absorption can improve the mechanical performances of biocomposites via the hygromechanical hardening of plant fibers. This finding has been verified in our previous work in the case of extreme mechanical solicitations such as machining [8]. The moisture content absorbed in plant fibers leads to improving their shear efficiency during the cutting operation, which is a sign of the increase in the cutting contact stiffness. The hygrothermal properties of plant fibers have also shown an advantage in biocomposite processing via their self-shaping ability in the presence of a humid environment due to the moisture-induced bending actuation, which could be used as a driving force in autonomously self-shaping materials facing humidity variations [45,46]. Many innovative applications can be derived from this self-shaping ability of biocomposites in a humid environment such as smart adaptive systems to regulate humidity or to activate a given function (temperature, electrical charge, etc.) at a given level of humidity.

However, the exposure time to the humid environment must not exceed the transient regime threshold because, otherwise, interface damages occur due to polymer hydrolysis and contribute to the deterioration of the biocomposite structure [8]. The threshold between the transient regime and saturated regime in terms of moisture content and/or exposure time is highly dependent on many parameters related to the natural fibrous structure and the exposure environment such as the structure of the fabric (unidirectional or bidirectional) [47], the fiber volume fraction [41], the fiber orientation [32], the conditioning temperature [41,42], and the hygrometric conditioning process (water immersion or relative humidity) since the water immersion process accelerates significantly the water diffusion [47].

In our point of view, the hygrothermal factor should be considered as a means of transient improvement of the mechanical performance of biocomposites during manufacturing processes and industrial applications. To this aim, scientific studies should be carried out on this subject in order to define how to master the hygrothermal behavior of biocomposites at the transient regime without reaching the saturated regime where the mechanical properties start to be degraded.

4. Conclusion

Mechanical properties of biocomposites result from a very complex mixture of factors that interact at different scales. Figure 8 summarizes the main factors affecting the properties of biocomposites from plant cultivation to composite processing. To strengthen the mechanical properties assessment of biocomposites, several scientific studies are required at different scale levels to reach a fine understanding of the physical mechanisms involved with each factor in the biocomposite structure.

Mastering the effect of these main factors will lead to not only reducing the variability of mechanical properties but also to making these properties at their optimum level. Investigating the functional relationships between the main factors and the properties of biocomposites will also contribute to developing models with the capacity to predict the effect of these factors and their interactions. These research outlooks could significantly allow the standardization of technical specifications of biocomposite parts for massive industrial use.

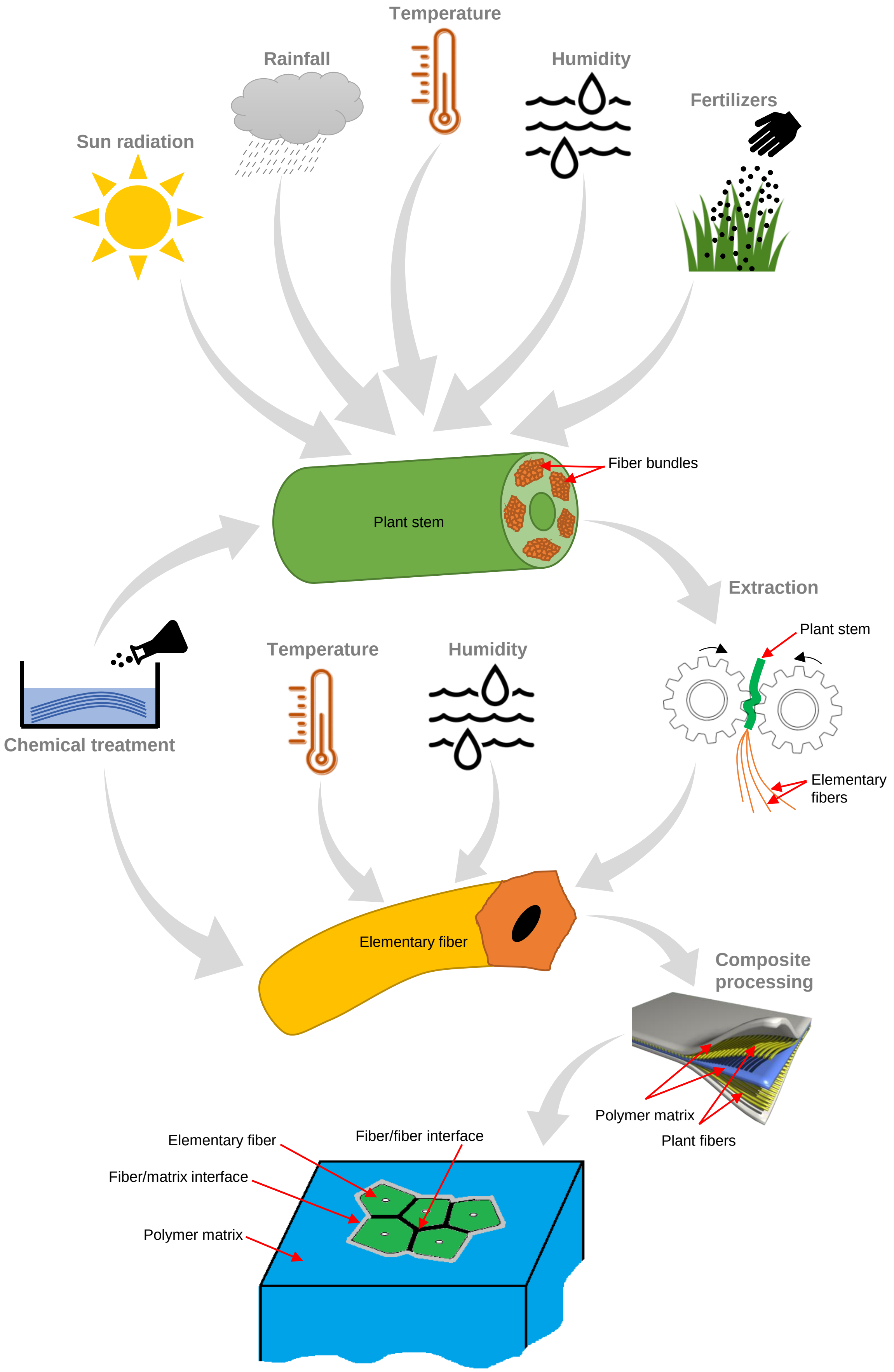

Figure 8: Schematic illustration showing the main factors affecting the properties of biocomposites from plant cultivation to composite processing

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