Agro-Fiber Based Composites With Use in Automotive Industry

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***Abstract* – Natural reinforcement (for example: flax and jute) have already shown that it can be applied in composite parts to decrease the weight (up to 30 %) and the cost of vehicle production (by approximately 20 %), ultimately reducing the environmental burden. Their early utilization in the automotive sector dates to the 1990s in Europe. Natural fibers constitute an adequate substitute for E-glass due to their lower specific gravity, which means that natural fibers can provide more specific strength over stiffness when combined with a plastic matrix. These advantages help replace non-biodegradable (synthesis) composites and are beneficial for consumers. Taking advantage of the "environmentally friendly composites", car manufacturers utilize them in the manufacture of components such as headliners, dashboards, door panels, and trunk-liners. The present paper summarizes research findings on natural fiber properties and their utilization in natural-fiber composites (NFC, or NFRC – natural-fiber reinforced composite) with a focus on the field of automotive production.**

***Keywords* – Eco-friendly materials, composite materials, natural fiber, jute, flax.**

DOI: 10.18421/TEM133-70
https://doi.org/10.18421/TEM133-70

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Received: 01 February 2024.
Revised: 29 May 2024.
Accepted: 21 June 2024.
Published: 27 August 2024.

1. Introduction

The pressure to reduce the carbon footprint (CO_2) is leading scientists and materials engineers to develop new materials from renewable sources.These represent a new group of materials, and much research is in progress. In NFCs, natural fibers are blended with a polymer matrix (made of thermosets, thermoplastics, and biodegradable polymers). NFCs can be applied in various technical-engineering fields such as building and construction, sound insulations, packaging, handicrafts, bio-medical, sports, automotive sectors (Audi group, Ford, Opel, Mercedes, Daimler Chrysler, Toyota, Volvo, Seat, Peugeot, etc.) and so on. It is a suitable alternative for applications where the weight of the material is a major problem [1].

Natural fibers (NFs) are materials derived from renewable resources of plant (lignocellulosic), animal and mineral origin. Whereas the animal fibers' main component is protein, plant fibers comprise components as follows [2]:

- cellulose – cellulose is an essential building constituent of the cell wall skeleton, is a polysaccharide composed of β-D-glucopyranose. It is insoluble in organic solvents. Its dissolution occurs in concentrated acids due to a reduction in the degree of polymerization. The individual fibers are interconnected by hydrogen bridges in the cell wall – providing the necessary stiffness and strength (specific gravity: 1.25 $g.cm^{-3}$). It decomposes at temperatures above 200 °C by dry distillation at normal pressure.
- hemicellulose – this component is not a cellulose form. Hemicellulose comprises a group of heteropolysaccharides, being hydrophilic and soluble in alkalis.
- lignin – it is organic polymer with three complex compounds. It is amorphous and hydrophobic, soluble only in hot alkalis. Pectin and waxes are also present.

Natural fibers have low density compared to synthetic fibers and can provide reinforcement capable of imparting high specific mechanical properties to the final composite. Fig. 1 shows the breaking length / elongation ratio of natural fibers versus E-glass.

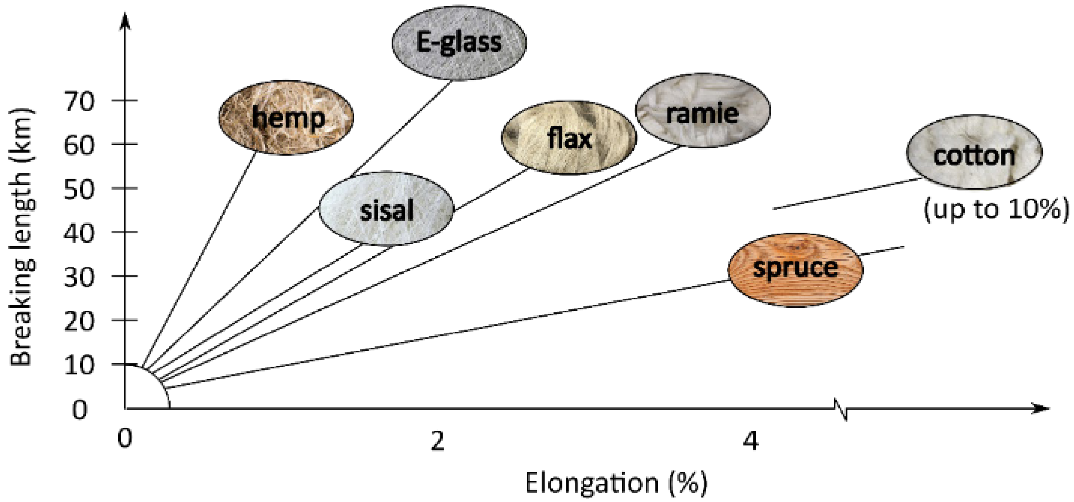

Figure 1. Breaking length / elongation of natural fibers versus synthetic fiber: E-glass [3]

The density of NFs range from 1-1.5 g.cm^{-3}, except for bamboo fibers, where the value is ρ = 0.8 g.cm^{-3}. In comparison, the density of glass fibers ρ = 2.6 g.cm^{-3} means that natural fibers can provide more specific strength over stiffness when used with the plastic matrix. Added benefits of natural fibers include lightweight, non-toxicity and biodegradability [4].

Disadvantages of natural fibers include: poor fire resistance (changes in molecular structure occur), low impact strength, durability, thermal stability, lack of polymer wettability and resulting poor compatibility at the component interface, limited choice of the matrix due to NF degradation at temperatures in the 170-200 °C range, and last but not least - greater susceptibility to moisture (resulted from the strong propensity of cellulose to form hydrogen bonding in the open air). The properties of these composites (abbreviated as NFRP or NFC) depend on the components and adhesion between them (Fig. 2). Natural fibers are naturally susceptible to moisture due to amorphous regions, hydroxyl, and other polar groups. The presence of moisture reduces mechanical properties, biodegradability, dimensional instability, and lack of adhesion at the component: matrix versus fiber interface. By targeted treatment of the fiber surface, it is possible to achieve better wetting of the fiber by the polymer and, thus, a better adhesion rate. Generally, fibers can be modified by chemical and physical methods or a combination thereof [1], [5].

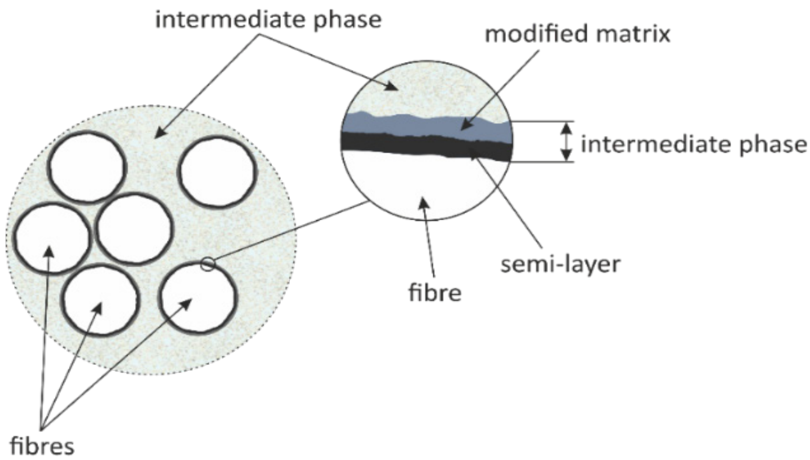

Figure 2. Schematic illustration of the interphase in a composite material [6]

The most readily available types of lignocellulosic fibers are flaxseed and jute. World production of flax is 830·10^{3} tones and jute: 2300·10^{3} tones – data from source [1], [7].

Flax (Lat.: *Linum Usitatissimum*) is the most frequently applied fiber in the clothing industry. It is grown in temperate climates. The mechanical properties depend on the diameter and location in the original stalk. The length of the technical fibers is equal to 30-80 cm. Harvesting is completed approximately in 10 days, or once the colour of the flower has changed to yellow. Flax has the capacity to absorb H_2O, which means that the fibers can take up to twice the amount of water compared to their original weight. Flax has an excellent heat conduction property. The best-producing countries are China, France, and Belarus.

Jute (Lat.: *Corchorus Olitorius / Corchorus Capsularis*) is grown in warm and humid areas. The fibers are extracted from the stem in running water (in 120-150 days, it grows to a height of 2.5-3.5 m), and the dry weight of the fibers is in the range of 4.5-8 % of the original weight of the plant. Jute fiber is considered one of the strongest lignocellulosic vegetables bast fibers with less moisture, acid, and UV light resistance. The advantages of jute fiber: exceptional isolate and antistatic properties. Bangladesh and India are top-producing countries. Characteristics of jute / flax fibers are in Table 1 [1], [8], [9], [10], [11].

Table 1. Characteristics of jute and flax fibers [8], [9], [10], [11], [12]

Properties of fibers	FLAX (origin: leaf)	JUTE (origin: stem)
Density [g / cm^3]	1.5	1.3
Elongation [%]	2.7-3.2	1.5-1.8
Tensile strength [MPa]	500-1500	393-773
Cellulose [%]	27.6	26.5
Hemicellulose [%]	71	60-72
Lignin [%]	18.6-20.6	12-24
Diameter [μm]	2.2	25-200
Microfibril angle [°]	–	6.2

Figure 3. Flax and jute fibers (properties are depending on aspects, for example: harvest time, region, etc.) , [13], [14]

2. Jute Fiber-Reinforced Polymer Composites

The carbon footprint in producing 1 kg of glass fiber is 1.7-2.2 kg of carbon dioxide while applying any natural fiber can reduce this value to about 30 % [15].

Safety regulations must be observed when handling glass fiber since mechanical irritation occurs in direct contact with the skin. Furthermore, increasing ecological concerns are being raised among companies concerning fibers' environmental safety and recyclability. Jute fibers are an alternative to glass fibers. They have a ↓ density yet ↑ specific modulus and almost identical tensile modulus. They can be applied with matrices of thermosets, thermoplastics, and biopolymers. The group of biopolymers is dominated by polylactide acid, an oil-based polymer. Using an epoxy matrix with jute fibers forms strong bonds concerning the good adhesion of the components. Raghavendra [16] published SEM images of the matrix versus fiber interface and states that the fiber-matrix interfacial bonding between the NF / polymer is more than that of the matrix / glass fiber [9]. Subsequently, the mechanical properties are compared, exhibiting 55 % tensile strength of jute-fibre composites and 61 % flexural strength of glass-fibre composites. The authors conclude that applying jute composites in commercial applications is appropriate for balancing cost and properties [16]. The study [17] deals with the relationship between the weight gain of jute fibers (with epoxy) and the resulting properties. Hardness, tensile properties, and impact strength increase with increasing fiber / flexural strength ratio and are largely influenced by the porosity of the composite. Maximum tensile strength value = 110 MPa, flexural strength = 4.45 GPa, flexural modulus = 3.02 GPa and impact energy = 4.88 J (for the alternative composite material with 48 wt. % of jute fiber loading –Fig. 4 and Fig. 5).

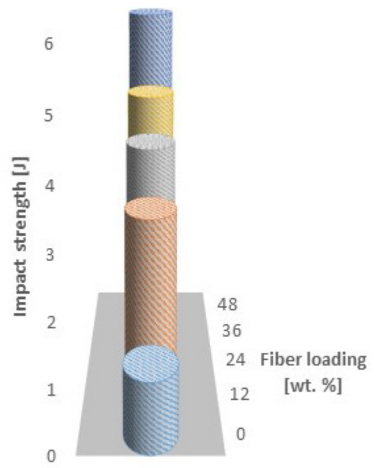

Figure 4. Effect of fiber loading on selected property (ASTM D 256) [17]

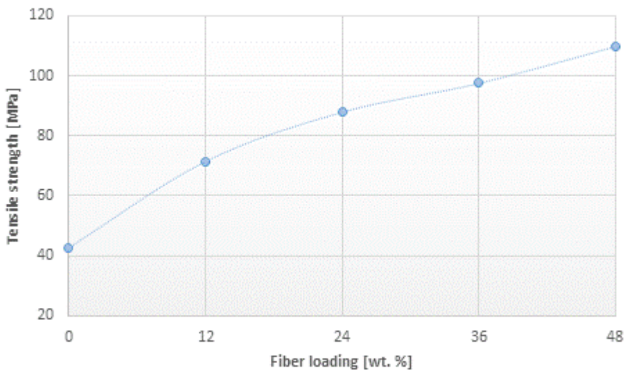

Figure 5. Effect of fiber loading on selected property (ASTM D 3039-76) [17]

Kumar and Srivastava [18] report a similar result in terms of tensile strength and flexural strength. As the proportion of fibers in the composite increases, the tensile and flexural strength properties increase (applied: the hand lay-up technique, which is suitable for construction purposes and prefabricated buildings used in natural calamities).

The study [19] applies thermogravimetric analysis (thermal behaviour of the unidirectional composites) and tensile and flexural testing. It compares two types of composites with epoxy resin → 1. in combination with jute fibers and 2. in combination with bamboo fibers, both made using the vacuum technique. It is revealed that jute + epoxy resin showed better thermal behaviour. Study [20] compares flexural and compression properties after immersion of composite material samples (polyester matrix + jute reinforcement) in distilled H_2O, seawater, and acid solutions at laboratory temperature (immersion: up to 21 days). It shows a deterioration of the above-mentioned properties – with an increasing percentage of water uptake.

Of the thermoplastics, the following can be applied in combination with jute fibers polypropylene (PP), polyethene (PE), polyamide (also known as nylon), etc.:

- **PP:** The study by Rahaman *et al.* [21] compares the properties of composites with jute fiber and PP / LLDPE (linear low-density polyethene) matrices produced by compression molding techniques. The first alternative of composites with PP matrix exhibits better mechanical properties. The study by Bledzki and Jaszkiewicz [22] compares the mechanical properties of 2 different types of matrices, i.e. PLA / PP combined with natural fibers (including jute fibers). PLA + jute fibers exhibit the best mechanical properties; on the other hand, PP + jute fibers exhibit better impact strength performance (components manufacturing technology: injection molding);
- **PLA:** YU *et al.* [23] suggested the application of jute fibers composites and the PLA matrix in a max ratio of 30:70. The values of mechanical properties increase after 30 % fiber share (stress is expected to transfer from the matrix to the fiber) and subsequently decrease. A similar conclusion is valid for the evaluation of the composites' thermal stability;
- **PE / HDPE:** studies by Mohanty *et al.* [24], [25] evaluate the application of jute fibers in HDPE. When 30% of jute fibers were applied (with maleic anhydride grafted polyethene agent), the "optimum" mechanical strength was achieved (47.3 % increase in the tensile test / 26.4 % in the flexural test and 28.1 % in impact strength). The damping properties of the treated/untreated composites → decreased in comparison to the virgin matrix;
- **PES:** an earlier dated study by Semsarzadeh [26] concludes that the mechanical properties of the resulting composites depend not only on fiber strength and fiber-matrix interbonding but also on secondary chemical bonding between the matrix chain and the polar surfaces of jute.

Table 2. Mechanical properties of composite materials with jute fiber + different matrices [22], [27]

Matrix / fiber	*Rm* [MPa]	*E* [GPa]	*A* [%]
PP / jute	47.9 ± 2.7	5.8 ± 0.47	1.4 ± 0.1
PLA / jute	81.9 ± 2.9	9.6 ± 0.36	1.8
PHBV / jute	35.2 ± 1.3	7.0 ± 0.26	0.8

Note: technology of production → injection molding.

3. Flax Fiber-Reinforced Polymer Composites

Flax fibers are often applied in automotive industry materials, one of the reasons for their application being significantly higher values of mechanical properties, e.g. in comparison with wood (tensile strength of pine wood = 100-200 MPa, the tensile modulus of pine wood = 12-27 / values of mechanical properties of flax fibers – shown in Table 1) [28].

The concept of technical flax fiber is explained in Fig. 6.

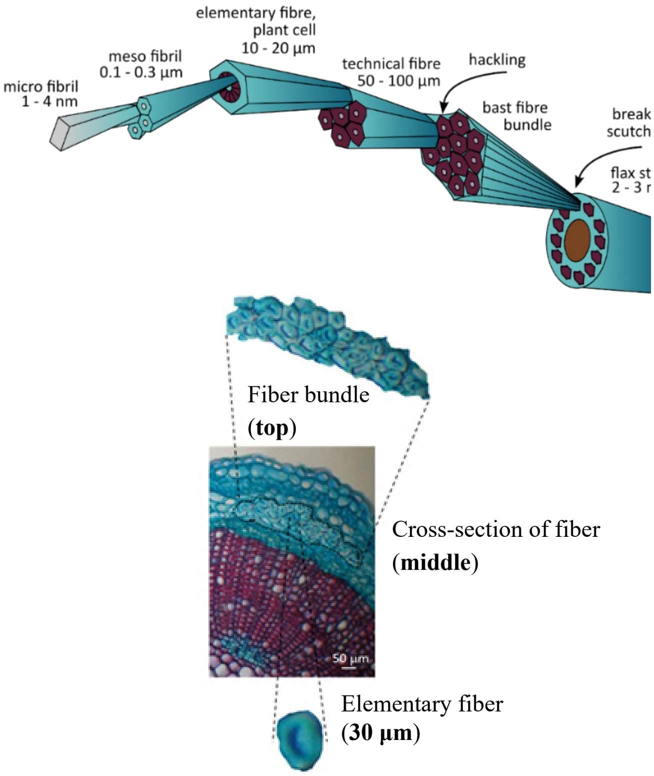

Figure 6. Schematic plot of technical fiber, illustration of flax fiber (light microscope image) [29], [30]

When combined with epoxy resin, the resulting composite has excellent sound absorption and vibration damping [31].

In conjunction with flax fibers, the matrices can also be applied as follows:

- **PP / PLA:** Oksman [32] compares the mechanical properties of composite material manufactured from short randomly arranged flax fibers (wt. % = 30 and 40) combined with a PP / PLA matrix, produced by compression molding technology. TS for flax-PP = 29 MPa; in the flax-PLA composite, the tensile strength was 1.8 times. Oksman also states that in the case of PLA matrix, the same manufacturing technologies can be used for production as for CM with PP matrix – extrusion or compression molding. A study published similar results and stated that an increase in impact strength occurs with an increasing proportion of natural fiber. The study by Van Den Oever and Van Kemenade [34] from the Agrotechnological Research Institute applies the wet laid process (flax fibers – randomly arranged, two types: hackled and scotched).
- The technology allows controlling the size of the fiber, which is necessary to compare the different data using the Cox-Krenchel model (for E-modulus) and the Kelly-Tyson model (for the strength of composites);
- **HDPE:** Singleton et al. [36] apply HDPE recyclate in the production of NFCs. They report that a volume fraction in the interval - 15-20 % flax fiber is optimum to maximize the mechanical properties measured. Adding flax and recycled ground tire rubber to the HDPE matrix provides a toughening effect [35].
- **PLA:** incorporating fibers into PLA matrix increases stiffness, impact energy, glass transition temperature and toughness. Several studies compare PP, epoxy, and PLA matrices, concluding that the best alternative for the resulting properties and biodegradability is the PLA matrix. Among the biodegradable matrices, cellulose acetate butyrate, in short CAB (with abaca, hemp, and sisal) can also be used with flax fibers [31], [37].

Figure 7. Bcomp products for composite manufacturers. Left side: unidirectional flax fabrics, right side: ± 45 bi-axial flax fabric [38]

Several studies evaluate the application of MAPP compatibilizers in short flax fiber polypropylene composites. The application of MAPP compatibilizer has a good effect on increasing tensile and flexural strength (about a one-third increase, compared to untreated flax fiber composites) and decreasing water uptake rate. Maleic-anhydride PP improves fiber wettability because of the compatibilizer's long chain, improving the adhesion to the matrix. On the contrary, adding a MAPP compatibilizer negatively impacts flow behaviour by raising viscosity or reducing the melt flow index. SEM images of the flax / PP + MAPP compatibilizer composite material – Fig. 8 [39], [40], [41].

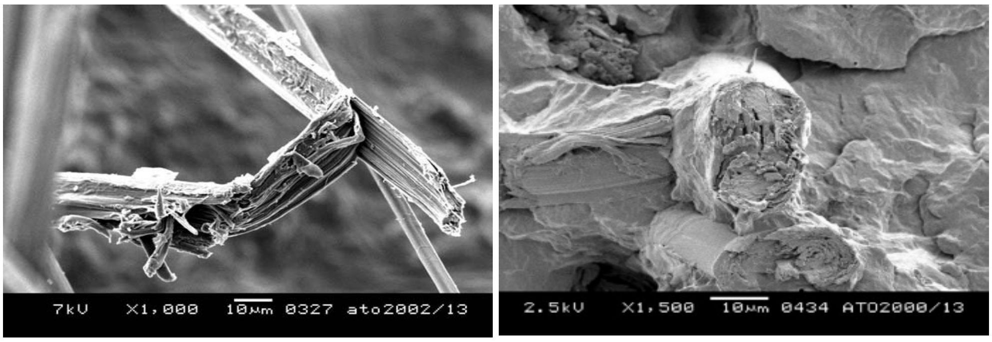

Figure 8. Left side: SEM micrograph of split elementary flax fiber, right side: composite material (30 wt. % flax / PP + MAPP) [33]

4. Conclusion

Natural fiber composites for technical and engineering applications have been the subject of significant investigation around the world. Various natural fibers in different designs (short fibers, yarn, nonwoven mats, yarn, bundle, etc.) and properties can be applied in composites. Concerning global production, several fibers are currently utilized: flax, jute, hemp, ramie, coir, etc., in conjunction with polyolefins, thermosets and phenolic resins. PLA, PHA, starch / oil-based resins are utilized to improve biodegradability. Studies reported in the research review conclude that fiber reinforcement improves mechanical performance in terms of tensile/flexural strength, impact resistance, and interfacial shear strength. The resulting properties of the NFC depend on the properties of their components and interfacial adhesion. The adhesion can be 'boosted' by modifying the fiber using physical and chemical methods or by adding suitable additives – modifiers.

Acknowledgements

Research was supported by project KEGA 024TUKE-4/2023 „Update and innovation of methodology for practical education of Automotive Production Technologies study programme in the field of heat treatment and surface finishing.“

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