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Special Issue “Natural Fiber Based Composites”

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In the last twenty years, the use of cellulosic and lignocellulosic agricultural by-products for composite applications has been of great interest, especially for reinforcing matrices. Fibers of renewable origin have many advantages. They are abundant and cheap, they have a reduced impact on the environment, and they are also independent from fossil resources. Their ability to mechanically reinforce thermoplastic matrices is well known, as is their natural heat insulation ability. The matrices can themselves be of renewable origin (e.g., proteins, starch, polylactic acid, polyhydroxyalkanoates, polyamides, etc.), thus contributing to the development of 100% bio-based composites with a controlled end of life.

This Special Issue’s objective is to give an inventory of the latest research in this area of composites reinforced with natural fibers, focusing in particular on the preparation and molding processes of such materials (e.g., extrusion, injection-molding, hot pressing, etc.) and their characterization. It contains one review and nineteen research reports authored by researchers from four continents and sixteen countries, namely, Brazil, China, France, Italy, Japan, Malaysia, Mexico, Pakistan, Poland, Qatar, Serbia, Slovenia, Spain, Sweden, Tunisia, and Vietnam.

An overview of the chemical treatments, manufacturing techniques, and potential lightweight engineering applications of natural fiber based biodegradable composites is provided in a comprehensive review paper [1]. Many natural fibers are presented, including jute and sisal. These have been utilized for ages in several applications, such as ropes, building materials, particle boards, automotive industry, etc., and the chemical treatments and surface modifications are presented as solutions to improve the quality of the natural fibers.

Some of the research articles in this Special Issue suggest the use of very different raw materials to obtain biobased materials that may find future applications in many fields, e.g., agriculture, ecological engineering, construction, load-bearing composites, and packaging.

- Especially, once denaturated in a twin-screw extruder, sunflower proteins can be used for their thermoplastic behavior to produce slow release fertilizers through injection-molding with urea and/or municipal biowastes acting as additional sources of nutrients for plants [2].
- Fibers from linseed flax straw may be mechanically extracted in a continuous mode before their use in geotextile applications [3].
- Constituting an interesting source of vegetable squalene in its seeds, the stems of the amaranth plant are presented as a new perspective for building applications with low-density insulation blocks obtained from their pith fraction and hardboards from the bark one [4].
- After their mechanical mixing, flax and polylactide fibers can be transformed into green sound-absorbing composite materials through hot pressing, and various structure and profiling can be obtained by using different multilayer structures of nonwovens and adjusting the pressing conditions [5].
- The same sound-absorption ability can be obtained from hot pressed composites made from polycaprolactone (PCL) and kapok fibers as thermoplastic matrix and mechanical



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reinforcement, respectively, their sound-absorption performance depending on their volume density, the mass fraction of kapok fibers inside the materials, and their thickness [6].

- A thermoplastic biocomposite based on hemp fibers and polyamide 11 (PA11) has also been developed through a three-step process, i.e., a wrapping operation to obtain 100% biosourced commingled yarns followed by their weaving to produce two different fabrics and then the molding through thermocompression, and the resulting composites, with stacking of two cross-ply, were characterized regarding mechanical strength [7].
- Electrospun films developed from wastewater treatment plant sludge (WTPS) are also presented as new, local, circular, renewable, and environmentally friendly packaging materials, their characteristics (i.e., tensile properties, contact angle, and surface properties) being largely influenced by both purification and treatment of WTPS to make it suitable for the electrospinning process [8].

Some other research articles propose various pre-treatments on natural fibers with the objectives to favor both the manufacturing and use properties of the resulting natural fiber-based composites or cellulosic textiles. Pre-treatments for plant fibers are very popular for increasing the fineness of bundles, promoting individualization of fibers, modifying the fiber-matrix interface inside composites, or even reducing their water uptake. For subsequent textile applications, the pre-treatments can also be proposed to make the cellulosic fibers more resistant to fungi, bacteria, fire, etc.

- In particular, flax-poly(lactic acid) (PLA) non-woven load-bearing composites were obtained after two different physical pre-treatments of flax tows (i.e., ultrasound or gamma irradiation) rather than the addition of chemicals, and this resulted in effective fiber individualization and a significant increase in the stress at break after the ultrasound pre-treatment [9].
- Hot water, green liquor, and sodium chlorite pre-treatments were also suggested to improve the thermal stability and UV resistance performance of films made from cellulose nanofibers (CNFs) from sugarcane bagasse and spruce [10].
- Another pre-treatment example consists in the surface modification on the nanoscale of cotton fabrics using electrospun sericin/PNIPAM, and this pre-treatment allowed the fast forming of cotton fabrics that also revealed promising antimicrobial activity for subsequent textile applications [11].
- Antibacterial efficiency of textile cellulosic fibers can also be enhanced by means of microencapsulation and green grafting strategies using the bactericidal activity of chemicals from natural essential oils [12].
- In the same way, the grafting of chitosan-essential oil microcapsules onto cellulosic fibers would make it possible to consider the manufacture of protective textile substrates such as antimicrobial masks, bacteriostatic fabrics and healthcare textiles [13].
- It is also possible to enhance the flame resistance of cellulosic fibers using an eco-friendly coating consisting in the grafting of acrylic acid onto the surface of cotton using plasma technology for improved attachment of acrylate phosphate monomer, and this opens up the possibility of producing flame-retardant cellulosic textiles [14].

Additionally, last but not least, olive pomace (OP) is presented as a potential fibrous reinforcement in polypropylene (PP) and polyethylene (PE) biocomposite materials [15]. More specifically, this contribution proposes a life cycle assessment (LCA) of these new construction materials (e.g., decking), and the latter confirmed their environmental friendliness. This constitutes a new perspective for the valorization of OP, an agricultural by-product widely present in southern Europe.

The plant world is also the source of natural substances other than fibers, which can also find applications in the field of composite materials, e.g., in the building industry. By way of example, one research article in this Special Issue evidences the interest to add mucilages extracted from five specific plants to improve the quality of lime-based mortars,

making these natural extracts promising additives for restoring and protecting historical buildings [16]. Vegetable raw materials can also be the source of natural dyes, possibly useful in the construction sector (case of the exterior wall paints [17]), but also for textiles, paper, etc. In particular, using natural dyes on textiles has many advantages, as they are healthier and more environmentally friendly than the synthetic ones. A natural dye extracted from the purple petals of an invasive plant has thus been successfully used for the screen printing of both cotton and polyester woven fabrics just as various papers [18].

Plants can also be the source of natural adsorbents. Another research report in this Special Issue thus presents the obtaining of efficient biobased carbon adsorbents [19]. Prepared from spruce bark residues, these will find application for efficient removal of reactive dyes and colors from synthetic effluents, and may be used for treating contaminated wastewater to remove pollutants.

In addition to the vegetable fibers and other chemicals from natural resources, the animal world is also the source of promising renewable biopolymers for material applications. In particular, chitin is one of the most abundant polysaccharides, known to possibly act as a structural material in biological systems. In [20], nanochitin/polystyrene composite particles are prepared by Pickering emulsion polymerization using scaled-down chitin nanofibers. Owing to both their biocompatibility and hydrophobicity, these are promising candidates for future medical uses, e.g., by being used as carriers to control the release of hydrophobic drugs over time.

This Special Issue thus presents a very wide range of topics. It provides an update on current research in the field of natural fiber based composite materials. I am convinced that all these contributions will be a source of inspiration for the development of new composites. Generally speaking, these new materials are environmentally friendly and will undoubtedly find numerous applications in the years to come in many sectors.

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