Synthesis, Characterization, and Applications of Hemicellulose Based Eco-friendly Polymer Composites

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1 Introduction

Generally, composites offer very promising solutions to the ongoing problems of the world[1–90]. Fuel cells [30–32, 90] materials for catalysis [79, 33–35, 80–86, 90–96], capacitors [20], solar cells [25, 26], thermopower applications [2], sensors [9, 10, 13, 42, 90, 94] are some examples of these solutions. Materials are also used corporately in combining their superior properties. Metal-metal combinations, polymer-metal combinations and their some hybrids with carbon-based materials are used for variety of nanomaterial applications [6, 7, 14–17, 23, 55, 78–84].

Composite materials are also widely used in developing appropriate materials for various applications as conductive polymer and other electronically active material composites. Graphene and graphene oxide [4, 27, 45–47], carbon nanotubes (CNTs) [14–17, 45–47, 79–85, 93, 95], activated carbon (AC) [14–17, 33], vulcanized carbon (VC) [30–32], carbon black [9, 10] and graphene derived materials which have different structure and morphologies as reduced graphene oxide [3, 30–32, 37, 45–47], graphene nanosheets, graphene nanoribbons, graphene nanoplatelets may thought as active materials for different applications. Also, a wide variety of nanoparticles are utilized in composite systems [34, 35, 73, 74, 85–89, 91–93, 95, 96]. Besides, biodegradable polymers or polymer composites (PCs) produced using renewable materials are referred to as eco-friendly polymer composites (EFPCs). Since they have a carbon-neutral lifecycle, their use as an alternative to petroleum-based materials can result in the reduction of carbon dioxide emissions. Having a net zero carbon footprint, EFPCs are also good materials for the envi-

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ronment and surrounding eco-systems [97–109]. It is an unavoidable fact that the world is currently facing serious challenges with regard to the environmental, social and political crisis. Another pressing issue that needs to be tackled by people is that of energy. Today, a large part of the world depends on fossil fuels to meet the demand for energy [110–115]. However, fossil-based energy sources are becoming exhausted at a great speed. Therefore, it is imperative that not only scientific organizations but also public and private sectors focus on the innovation of eco-friendly materials, also referred to as "green materials." This research area has recently witnessed important accomplishments thanks to scientists and engineers who have realized that making environmentally responsible decisions is the ultimate way to protect the environment and to prevent energy crises [108]. Figure 1 shows the world energy consumption according to energy sources.

Production of environmentally friendly materials requires different fields to participate in the process. These fields engage in sustainable chemistry and work on biodegradable and bio-based materials structurally, chemically, and physically [116–120]. They also pay close attention to make sure that environmentally hazardous substances are not included in any stage of the design and production process. The fields with which we are concerned are EFPCs, sustainable chemistry, natural resources and eco-friendly engineering processes, etc. Animal and plant products can be used to manufacture many natural polymers. These materials have various appealing features. They are ubiquitous, do not contain synthetic substances, can be recycled and consumed by microorganisms. In the last ten years, numerous studies have been carried out on the production of polymeric materials (blends, composites, and nanocomposites). These studies mostly conducted experiments polymers and fillers were mixed. When resin systems are integrated with reinforcing materials, EFPCs achieve excellent features. The combination of resin systems and reinforcing fiber/filler particles results in the composite material integrating the features of resin features and of fibers/fillers. In a composite material, the resin matrix distributes the load on the composite between fiber/filler particles.

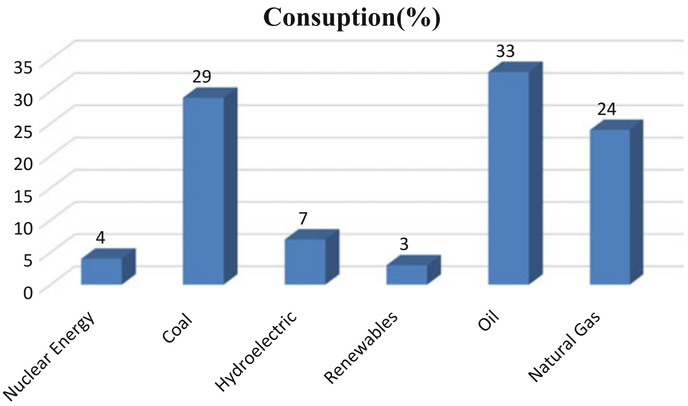

Fig. 1 The primary energy consumption in the world

In addition, it prevents fibers from being damaged due to abrasion and impact and enhances the load-bearing capacity of the composite. The resulting composite has high hardness and resistance and low density and is easy to use. Therefore, it is a better material than metals to be used in the manufacture of various products.

EFPCs have the high specific strength and specific stiffness, high fracture, abrasion, impact, corrosion and fatigue resistance, and low cost.

The greatest appeal of eco-friendly composites is that they are environmentally friendly, completely degradable and sustainable. Baillie addressed the construction and biorhythm evaluation of green composites in detail [8]. EFPCs might be employed effectively for various purposes such as short life-cycle goods manufactured in large quantities and disposable goods. Cellulose, NR, polysaccharides, starch, chitin, proteins, collagens/gelatin, and lignin are natural and biodegradable chemical compounds while polyamides, polyvinyl alcohol, polyvinyl acetate, polyglycolic acid, and polylactic acid are synthetic compounds. New fibre-reinforced materials are made from renewable resources and used in almost all areas [107].

Being superior to synthetic raw materials, EFPCs can play a significant role in the elimination of environmental problems, especially caused by plastic waste, and in the sustainable development of eco-friendly and cost-effective technologies [69]. Numerous studies are conducted on EFPCs to find innovative ways to manufacture eco-friendly materials from biodegradable polymers and renewable sources.

Eco-friendly Polymer Nanocomposites (EPNs) have also drawn enormous attention owing to their potential application in the field of agriculture such as product waste management [118]. EPNs can also be employed to improve the environmental compatibility of recycling processes.

There is the classification of six types of the most common EPN, including (i) EPN with Green Fillers and (ii) EPN with Green-Base-Composite such as—EPN from Cellulose,—EPN from Thermoplastic Starch,—EPN from Polylactic Acid,—EPN from Polymer Mixture,—EPN from Others (Gelatin, PHB, Chitosan).

2 Definitions About Hemicellulose and Derivatives

Of natural polymers, lignocellulosic is likely to be a very important material as it can be converted into biofuels and bioproducts. Mainly found in lignocellulose, hemicelluloses are the second most abundant renewable material in nature, easy to use, has film-forming features, good biocompatibility, and biodegradability [118, 119]. Hemicelluloses contain a very large quantity of free hydroxyl groups, which are perfect for chemical modification. Materials with exclusive features can be manufactured using chemically modified hemicelluloses in order to improve the biopolymers [39, 75, 101]. In recent years, hemicelluloses-based biocompatible films have been popular owing to reduced cost, oxygen barrier features and ease of access [18, 51].

Natural biopolymers are superior to synthetic ones in the sense that the former is more cost-effective, biocompatible, harmless and biodegradable. Being the non-cellulose cell-wall polysaccharides of agricultural and forest plants, hemicelluloses are regarded as unlimited and sustainable materials for the manufacturing of biopolymers and biomaterials [29, 70].

However, hemicelluloses contain a very large amount of hydroxyl groups distributed along the central and terminal region. In addition, these hydroxyl groups generate intermolecular and intramolecular hydrogen bonds [70, 77]. Hemicellulosic materials are non-hydrophobic, semi-crystalline and hygroscopic and have low mechanical features [36]. Due to these disadvantages, hemicelluloses films cannot be used widely. Plasticizers are generally used to enhance the mechanical features of films consist of hemicellulose [43, 44, 120].

As the main hemicelluloses in hardwood and annuals, xylan-type hemicelluloses are regarded as appropriate sources to achieve films. Emulsification and coating processes were employed to achieve arabinoxylan-based films [71, 72]. Lignin or glycerol was used to produce pure xylan, yet, self-supporting films [43, 44]. Table 1 shows the content of some lignocellulosic materials.

Plasticized films from aspen glucuronoxylan exhibit low oxygen permeability and therefore might be ideal for food packaging materials [49]. Up to 40% xylan in wheat gluten was used to produce composite films [57]. In addition, chemical modifications were performed in order to reduce the moisture sensitivity of

Table 1 Some lignocellulosic materials and their contents

Natural fibre	Cellulose (wt %)	Hemicellulose (wt %)	Lignin (wt %)	Total (wt %)
Cotton	89.7	1.0	2.7	93.4
Flax	80	13	2	95
Hemp	74.1	7.6	2.2	83.9
Sugar cane	51.8	27.6	10.7	90.1
Bamboo	54.6	11.4	21.7	87.7
Coconut	51.3	11.7	30.7	93.7
Wheat straw	38	36	22	96
	38.0	29.0	15.0	82
	33.2	24.0	15.1	72.3
	28.8	39.1	18.6	86.5
Rape straw	36	37	24	97
	37.6	31.4	21.3	90.3
Spruce + bark	42	27	26	95
	41.0	24.3	30.0	95.3
	50.8	21.2	27.5	99.5
Poplar wood	50	30	≤ 20	≤ 100

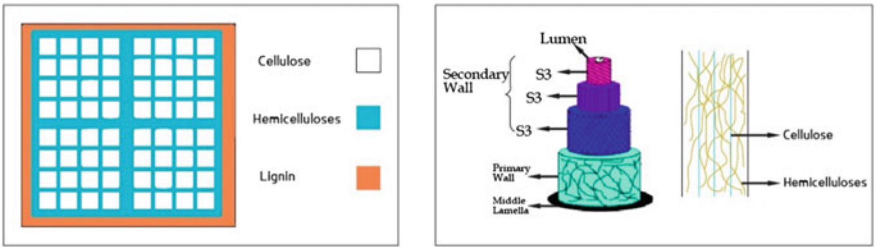

Fig. 2 The schematic representation of hierarchical structure of wood cell

hemicellulose-based films [50, 52]. The molecular structure of hemicelluloses had a large effect on film formation, structure, mechanical features, and moisture content. Side groups separated the xylan or mannan chains, and therefore, hemicellulose where the backbone was least substituted with arabinose or galactose was the most interactive one [29, 53, 66, 102].

Hemicelluloses can also be described in more detail and seen schematically in Fig. 2. They are the second most abundant polysaccharides in biomass. They are biosynthesized by trees and terrestrial plants. Despite the fact that they are in large quantity, hemicelluloses are little used by industries. Hemicelluloses are not completely degraded in most of the lignocellulosic refining processes, but they are removed or further used as feed raw material. Nevertheless, some methods have been developed to separate and isolate high molar mass hemicelluloses. Alkalines are used to extract hemicelluloses from plants. Water is also used to extract some parts of hemicelluloses. Forests and agricultural areas provide a large number of raw materials and studies have been and are being conducted to find potential application areas for it. Potential areas of use of hemicelluloses are paper, packaging, coatings, hydrogels, absorbents, and emulsifiers. As a group of polysaccharides, hemicelluloses have a positive effect on cell wall flexibility by cross-linking cellulose and lignin. They compose the hydrophilic component of the cell wall. They have categorized into different groups according to their structure: xylans, mannans, galactans, arabinans, and b-glucans. Xylans and mannans, the two most common ones, are used as components of polymer blends, PCs, and nanocomposites. As plant mannan gums have similar chemical features to that of hemicelluloses, they have also been categorized as hemicelluloses [73].

2.1 Properties

Most hemicelluloses support cell walls as do celluloses [100]. Thanks to the ability of hemicelluloses to absorb a large body of water, cell walls become less brittle and, more elastic and resistant to bending. Hemicelluloses also form absorbent molecular networks around cellulose crystallites, which enable the cell walls to transfer intermediate products and sources of nourishment [54]. In addition, it is reported that some fundamental properties of arabinoxylans allow intermolecular alignment between arabinoxylans and polysaccharides, which enables the composition of gels with several components in the wall matrix. Furthermore, ferulic acid residues on arabinoxylan chains allow for covalent interaction between polysaccharide–polysaccharide, polysaccharide-protein, or polysaccharide-lignin [40].

Having less degree of structural order and polymerization, hemicelluloses are chemically and thermally less stable than celluloses. Another difference between hemicelluloses and celluloses is that the former is soluble in alkalines, the feature of which is widely used to section various polysaccharides in samples which do not contain lignin. Nevertheless, this process leads to the removal of ester groups and decrease in solubility. Some hemicelluloses are able to be dissolved in water partially or completely [5]. Hemicelluloses gain hydrophilic property thanks to various free hydroxyl groups in them [38]. The more stable hemicelluloses are, the more they are dissolved in water and less tightly bind to cellulose. On the other hand, the more sporadic the side chains of molecules are, the less they are dissolved in water and more tightly bind to cellulose [60]. A reduction in polymerization generally leads to an increase in solubility [105]. Acetylated hemicelluloses can also be dissolved in dimethyl sulfoxide, formamide, and *N*,*N*-dimethylformamide. Almost all plants contain hemicelluloses, which are, therefore, found in diets. Cell-wall polysaccharides are important as dietary fibers and contribute to lipid metabolism. Therefore, their consumption helps feces pass through the digestive tract [5, 99, 108]. There is a classification of hemicelluloses such as (i) Softwood hemicelluloses;—Galactoglucomannan (Mannans),—Arabinoglucuronoxylan (Xylans),—Arabinogalactan,—Pectins (ii) Hardwood hemicelluloses such as—Glucuronxylan, -Glucomannan, (iii) Grasses like -Arabinoxylan-main.

3 Hemicellulose Based Composites and Their Applications

Environmentally friendly polymer composites (EFPCs) are used in the manufacture of packaging materials, films, sanitation products, bottles, fishing nets, cutlery, trays and various other products; sensing, adsorbing and many other applications [1, 19, 24, 28, 41, 61, 64, 99, 106, 109].

Expanding their areas of application, most eco-friendly polymers possess perfect features. Hemicelluloses contain a very large quantity of free hydroxyl groups distributed along the backbone and side chains, which are perfect for chemical modification. Materials with exclusive features can be manufactured using chemically modified hemicelluloses in order to improve the biopolymers [39, 101, 119]. In recent years, hemicellulose-based nanocomposite films (HBNCFs) have been popular owing to reduced cost, oxygen barrier features and ease of access [75]. In addition, these hydroxyl groups generate intermolecular and intramolecular hydrogen bonds [70, 77]. Hemicelluloses are hydrophilic and HBNCFs are semicrystalline and hygroscopic and have low mechanical features [36]. Due to these disadvantages, hemicelluloses films cannot be used widely. Plasticizers are generally used to improve the mechanical features of hemicellulose films [43, 44, 51, 120].

Owing to the fact that hemicellulose-based biomaterials are biocompatible, harmless and biodegradable, they have become of great interest for the manufacture of pharmaceutical, tissue engineering, and food packaging products. HBNCFs have attracted the greatest attention due to their oxygen barrier properties. However, they are not very good at forming films and have low performance. Plasticizers or hydrophobization is generally needed to turn hemicelluloses into appropriate materials for packaging products. Rather than concentrating on materials themselves, most studies on hemicelluloses address converting polymers into sugars [109]. Peng et al. suggested an efficient and feasible technique to manufacture superior HBNCFs. He combined cellulose nanofibers (CNFs) and xylan (XH) films as plasticizers were also included in the process. Sugar composition was xylose (89.38%), arabinose (5.75%), glucose (1.87%), galactose (0.66%) glucuronic acid (1.78%) and galacturonic acid (0.55%). Morphological tests indicated that XH film surface and CNF-reinforced NCF consisted mainly of nodules (10–70 nm) and that CNFs were embedded in the XH matrix. Aggregates started to develop in an interval of arabinose/xylose ranging from 0.23 to 0.31 in an aqueous solution and when the arabinose substituents were exhausted, unsubstituted xylan chains formed a steady connection, which shows that the intermolecular interaction could be adjusted by manipulating the substituted groups or by including a second component that establishes an interaction with the molecular chains of hemicelluloses. Film formation was better, and the tensile strength improved greatly due to high aspect ratio and strong interactions between CNF and the XH matrix. Fang et al. [39] employed the freeze-thaw method in order to use hemicelluloses from bamboo holocellulose, PVA, and chitin nanowhiskers to manufacture a hybrid hydrogel. FTIR and NMR results showed that the gelation process witnessed physical crosslinking instead of a chemical reaction. PVA functioned as a hydrogel scaffold while hemicelluloses were firmly hydrophilic and showed hydrogen-bonding features when chitin nanowhiskers functioned as a cross-linker. According to atomic force microscopy (AFM) images of chitin, whisker size was about 200 nm in length and 40 nm in width. The mechanical features of hydrogels were greatly enhanced by an increase in the rate of chitin nanowhiskers. The hydrogels are potential materials to be utilized in the manufacture of tissue engineering products. Laminated films, one layer of which is polyester and the other of which is made of carbon nanotubes and hemicellulose, have been patented in Japan [58]. Polymers were supplied by removing hemicellulose from *Abelmoschus manihot* or *Hydrangea paniculata.* The homogenous distribution of carbon nanotubes in hemicellulose films was leading to obtaining mechanically strong films [109].

3.1 Composite Formation and Characterization with Layered Silicates

For nanocomposite formation, polymer chains should be dispersed in the galleries between silicate layers in order to obtain two distinct classes of lamellar nanocomposites; intercalated and exfoliated as represented schematically in Fig. 3. Intercalation takes place with a little polymer permeating into the galleries, which leads to silicate layers expanding in a limited manner. This way, a systematic structure with multiple layers separated by several nanometers is obtained. Large-scale polymer diffusion causes silicate layers to exfoliate or delaminate. An exfoliated nanocomposite contains nanometer thick platelets that are uniformly dispersed all over the polymer matrix. On the contrary, when the polymer and silicate do not form a homogeneous mixture when mixed, the layers do not apart, and they function as agglomerates or tactoids.

Total diffusion of clay platelets in a polymer increases the number of usable reinforcing materials to carry an applied load and to avert the formation of cracks. The pairing of the enormous surface area of the clay and the polymer matrix enables stress to be shifted to the reinforcement stage, which leads to mechanical improvement. Furthermore, the impenetrable clay layers require a convoluted track for a permeant to travel across the nanocomposites as seen in Fig. 4. Thanks to obstructed diffusion tracks along nanocomposites, the barrier property and flame retardancy of polymer-clay nanocomposites are improved, and they are more resistant to chemicals and uptake less solvent [104].

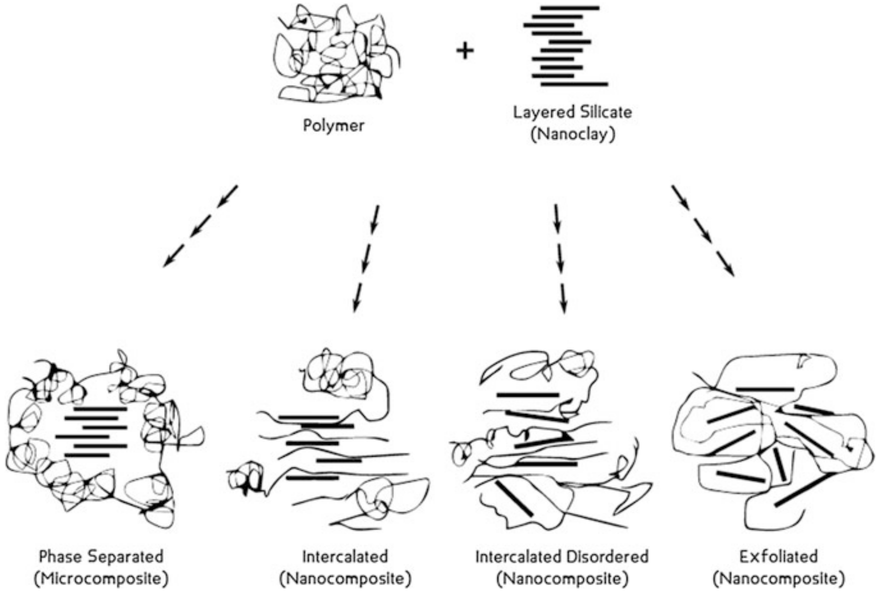

Fig. 3 Possible structures of nanocomposites

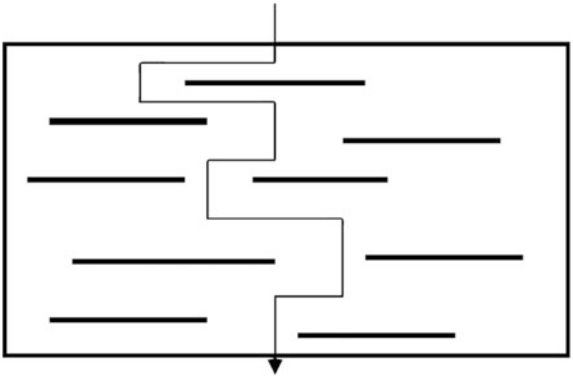

Fig. 4 Proposed model for the tortuous zigzag diffusion path in a polymer-clay nanocomposite when used as a gas barrier

3.2 Packaging Materials

The food producers have a wide range of natural recipes that are used in packaging applications and have a variety of needs that ensure the quality and safety of food. The first goal of this industry is assuring the protection of food products from the many factors related to the environment, gases, water etc. The most important factor in food packages is oxygen since oxygen has a very important material in the various chemical industries because oxygen mostly affects the expiration date of foods, fruits, vegetables, microbial growth etc. Nowadays, in order to cancel this effect of oxygen, Al plates and/or foil and some of the polymers such as polyethylene vinyl alcohol (EVOH), polyvinylidene chloride (PVDC) etc. are mainly used as packaging materials. Water, then again, is one of most important material, the catalyst that affects the chemical reactions and/or enzymatic changes and known as the principal food ingredient.

Biomass has become of greater interest for food packaging applications. The aim of most studies is to alter hemicelluloses and enhance the mechanical features of HBNCFs converted into a quaternary form. To this end, those studies focus on assuming an environmentally friendly and feasible approach. Chen et al. Integrated quaternized hemicelluloses, chitosan, and MMT to produce films. They analyzed the morphological structure and optical, physical, mechanical and thermal features of the films. Another aim of the study was to determine the ideal ratio in order to produce HBNCFs. The principal endeavour will present chitosan-based materials.

Peng et al. [70] reported that the molecular makeup of hemicelluloses had a great effect on film formation, mechanical features, and moisture content. The side groups made xylan or mannan chains separate, indicating that the hemicellulose which had the biggest interaction was the one in which arabinose or galactose substituted the backbone the least [53, 100, 102]. This was a pioneer study in the sense that it aimed to incorporate cellulose nanofibers into XH in order to manufacture mechanically improved nanocomposite films. SEM and AFM images pointed out that surface of XH and CNF-reinforced nanocomposite films consisted

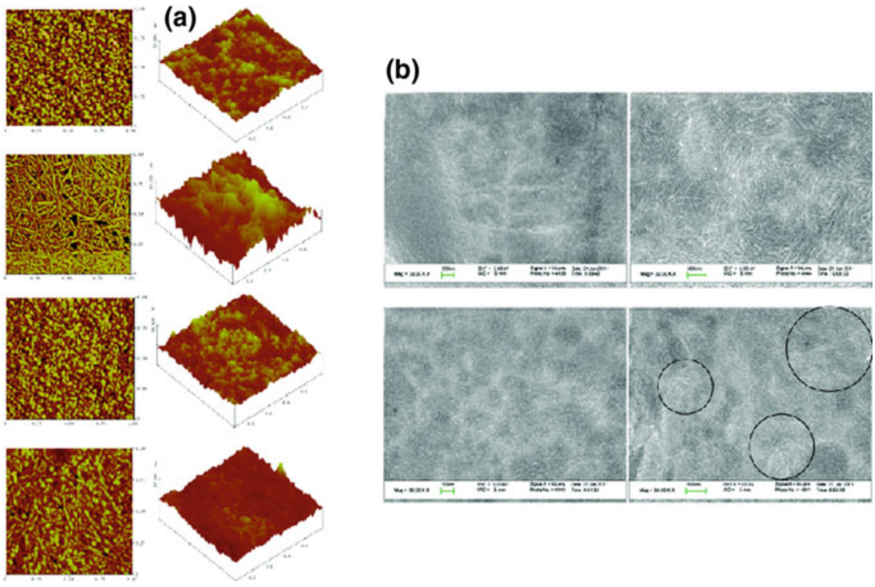

Fig. 5 Properties of CNF-reinforced nanocomposite films

mainly of nodules with a diameter of 10–70 nm and that CNFs were embedded in the XH matrix as shown in Fig. 5. The XH film had a peak at 2θ = ~18°, indicating that it was semicrystalline. The thermal stability of the nanocomposite film was better than that of the XH film. Film formation was better, and the tensile strength improved greatly due to high aspect ratio and strong interactions between CNF and the XH matrix, which points to an efficient and feasible technique for the manufacturing of superior HBNCFs [77].

3.3 *Other Applications*

Xylan is used in a wide variety of medical applications. Glyceronoxylene, xylitol or sorbitol-based films are widely used in the field of food packaging because their oxygen permeability is very low and there is a potential for further spread in the future [49]. Xylooligosaccharides (XOS) would hydrolysis results from claiming xylan-type hemicelluloses for guaranteeing possibility to a significant number of fields including chemical, sustenance and pharmaceutical commercial enterprises. For case, in the nourishment industry, XOS has an assortment of incredible physiological properties such as bringing down cholesterol levels, moving forward gastrointestinal function and the organic accessibility of calcium, and decreasing the risk of colon cancer [62, 76].

In order to make further improvements on the properties of the all-cellulose composites, it is recommended to use cellulose nanofibers that extracted from Napier grass fibers as reinforcement in cellulose matrix in further studies. According to the properties of composite films, it can be concluded that all these cellulose composites are suitable for using in warping, mulching and sorbent applications [97]. Another example of using natural fibers as reinforcement was seen in the study of Maniraman et al. [65]. Furcraea foetida (FF) plant was extracted and investigated. The structure of natural fibers generally longer, they have smaller diameter values and low spiral angle of the cellulose arrangement [56]. That quality of a bio-composite relies on the properties of the fiber and the resin and additionally on the quality from claiming their interfacial holding. So, to in front of fabricating bio-composites, additionally, it is a necessity to learn about those properties of fibers. There are many kinds of literature about natural fibers and their properties. They enlighten that celluloses, hemicelluloses and non-cellulosic components (lignin, wax, and pectin) are constructed in the natural fibers. But, the bio-fiber yielding plants grow only in particular regions based on environmental circumstances [11]. New assortments of fibers have appropriate properties for use as reinforcement are needed to be identified and developed. A lot of leaf fibers, such as those mined from Artisditahystrix, Sansevieria cylindrica, Tamarindus Indica L and sansevieria ehrenbergii have been used as an unsurpassed reinforcement for polymeric matrices [65].

Also, Kozlowski and Przybylak carried out a research and review to collect fire performance data for several types of eco-friendly polymer composites reinforced by cellulosic fibers [59].

Glucomannan type's materials can be seen as hemicellulosic materials which have film-forming properties. It is generally used in wooden as a "green" absorption material (gels, etc.) and it can be isolated from various method turbines or production fibreboard. Modification of new materials including non-hydrophilic and thermal character also can bring about the xylan which has high molecular weight displaying a stepped forward capability to form films. Film-forming is essential so as for xylan if you want to form self-supporting barrier films to be used in food packaging.

3.4 Basic Components of Composite Materials

The composites consist essentially of the matrix and reinforcing materials that give it physicochemical properties. The matrix part is the basic component of the composite and supports the physicochemical and biological properties of the reinforcing material. Matrixes are the main component to protect the reinforcements of the composites from abrasion and environmental influences. At the same time, it provides composites stability by allowing the matrix fibers and filler materials to coexist. A reaction with synergistic effect occurs by the matrix loading of the reinforcing materials. Thus, thanks to the reinforcement material and matrix, composite materials with the desired properties are prepared. Due to the physicomechanical properties of the polymer composites, they are of great interest because they have a much higher application area than the individual polymeric structures. The main materials reinforced with composite matrices are metallic oxides, clay, fiber constructions and carbonates. These reinforcing materials give physicomechanical and biological properties to the composites [12, 98].

The structure of lignocellulosic stocks forms polysaccharide and lignin. When lignocellulosic raw materials are introduced into biorefinery and subjected to a series of treatments, products such as cellulose, hemicellulose and lignin are formed. Cellulose, hemicellulose and lignin products are raw materials of lignocellulosic composites. These substances are the main components of animal feed and lumber resources. Cellulose is naturally present as a composition of lignin and hemicellulose fibers. Lignin protects carbohydrates from biological attack and is infectious by very few microorganisms [22, 68].

3.5 The Effect of Fibers on Composite Materials

Fibers play a very important role in the development of multifunctional composite materials. The shrinking the size of any composite material causes a reduction in material defects. The fibrous materials have a higher surface area, a larger surface area, compared to bulk materials. Many studies have shown that multifunctional composite properties depend on properties such as fiber length, volume, fiber type, fiber orientation, etc. These features can be briefly summarized as follows:

- Composite materials composed of long fiber structures have higher mechanical resistance than short fibers. Whereas short fiber composites are less costly and easier to process than long fibers [117].
- Increasing the amount of fiber loading to a certain amount of composite material, resulting in an increase in composite material properties. However, after this particular value, there is a decrease in the composite properties due to the interlocking between the matrix and fibers [63].
- Fiber loading method make a significant impact on composite material properties. The lowest value of Young's modulus was found to be 45° and 60° in the fibers and there have been seen discrepancies between theoretical and experimental values [48].
- Generally, the fibers are made of beeswax, gelatin starch and oily materials to improve adhesion and retention properties [67].

4 Conclusions

In this chapter; the synthesis and characterization of Eco-friendly polymer composites (EFPCs) and their usage in various applications such as food packaging, medical applications, gas barriers etc. have been examined in detail and as it can be seen these polymer composites are getting more and more attention to science and technology due to their superior properties such as biodegradability, renewability, high strength and stiffness, low cost and ecologically friendly formation. Eco-friendly polymer composites (EFPCs) from natural components provide a potential alternative to other commercial composite materials without compromising strength, stiffness and abrasion resistance properties to a variety of industrial and other applications.

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