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Sustainable Bio-Composites from Renewable Resources: Opportunities and Challenges in the Green Materials World

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Sustainability, industrial ecology, eco-efficiency, and green chemistry are guiding the development of the next generation of materials, products, and processes. Biodegradable plastics and bio-based polymer products based on annually renewable agricultural and biomass feedstock can form the basis for a portfolio of sustainable, eco-efficient products that can compete and capture markets currently dominated by products based exclusively on petroleum feedstock. Natural/Biofiber composites (Bio-Composites) are emerging as a viable alternative to glass fiber reinforced composites especially in automotive and building product applications. The combination of biofibers such as kenaf, hemp, flax, jute, henequen, pineapple leaf fiber, and sisal with polymer matrices from both nonrenewable and renewable resources to produce composite materials that are competitive with synthetic composites requires special attention, i.e., biofiber–matrix interface and novel processing. Natural fiber–reinforced polypropylene composites have attained commercial attraction in automotive industries. Natural fiber—polypropylene or natural fiber—polyester composites are not sufficiently eco-friendly because of the petroleum-based source and the nonbiodegradable nature of the polymer matrix. Using natural fibers with polymers based on renewable resources will allow many environmental issues to be solved. By embedding biofibers with renewable resource–based biopolymers such as cellulosic plastics; polylactides; starch plastics; polyhydroxyalkanoates (bacterial polyesters); and soy-based plastics, the so-called green bio-composites are continuously being developed.

KEY WORDS: Sustainable bio-composites; natural fiber; bioplastic; cellulosic plastic; polylactides; polyhydroxyalkanoates; soybean-based plastic; fiber-matrix interface.

INTRODUCTION AND BACKGROUND

There is a growing urgency to develop novel bio-based products and other innovative technologies that can unhook widespread dependence on fossil fuel. Simply stated, bio-based materials include industrial products, but not food or feed, made from renewable agricultural and forestry feed stocks, including wood, wood wastes and residues, grasses, crops, and crops by-products. Renewable, recyclable, sustainable, triggered biodegradable—all can make a difference in the environment today and tomorrow. The Technology Road Map for Plant/Crop-based Renewable Resources 2020, sponsored by the U.S. Department of Energy (DOE), has targeted to achieve 10% of basic chemical building blocks arising from plant-derived renewable sources by 2020, with development concepts in place by then to achieve a further increase to 50% by 2050. The U.S. agricultural, forestry, life sciences, and chemical communities have developed a strategic vision [1] for using crops, trees, and agricultural residues to manufacture industrial products, and have identified major barriers [2] to its implementation.

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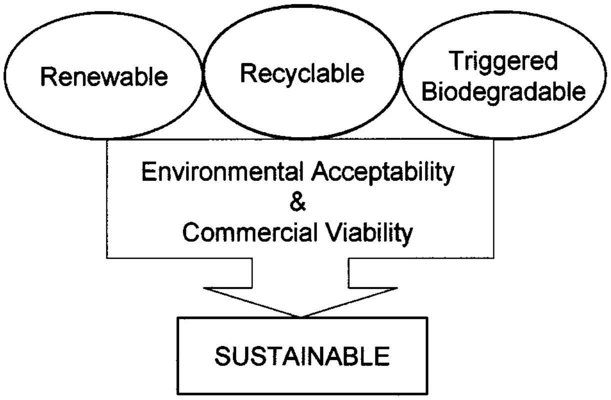


Fig. 1. Concept of "sustainable" bio-based product.

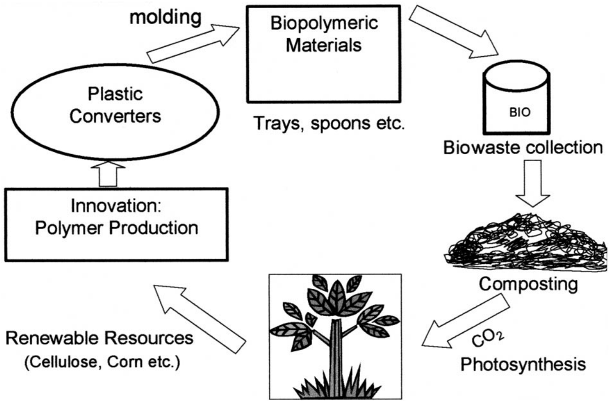


Fig. 2. Carbon dioxide sequestration.

Eco-friendly bio-composites from plant-derived fiber (natural/biofiber) and crop-derived plastics (bioplastic) are novel materials of the twenty-first century and would be of great importance to the materials world, not only as a solution to growing environmental threat but also as a solution to the uncertainty of petroleum supply [3, 4]. Biopolymers are now moving into mainstream use, and the polymers that are biodegradable or based on renewable "feedstock" may soon be competing with commodity plastics, as a result of the sales growth of more than 20–30% per year and improvement in the economics of sales [5]. The best examples of biopolymers based on renewable resources are: cellulosic plastics, polylactides (PLA), starch plastics, and soy-based plastics. Microbial synthesized biopolymers, i.e., polyhydroxy alkanoates (PHAs) polymers, have also attracted much attention recently. The use of materials from renewable resources is attaining increased importance, and the world's leading industries and manufacturers seek to replace dwindling petrochemical-based feedstock with composites derived from natural fibers and biopolymers.

CONCEPT OF SUSTAINABLE BIO-BASED PRODUCTS

A bio-based product derived from renewable resources having recycling capability and triggered biodegradability (i.e., stable in their intended lifetime but would biodegrade after disposal in composting conditions) with commercial viability and environmental acceptability is defined as a "sustainable" bio-based product (schematic representation in Fig. 1). Bio-composites, or more specifically the "green composites," consist of biofiber and bioplastic from renewable resources and thus are expected to be biodegradable. However, plastic derived from renewable resources may be nonbiodegradable, depending on the structure and curing nature of plastic during fabrication of bio-composites. Thermoplastic is having more environmental impact than thermosets because of its recyclibility. Again bio-based products obtained from renewable resources also maintain carbon dioxide neutrality (Fig. 2).

The "sustainability" issues of bioplastics, e.g., PLA, cellulosic plastics (cellulose esters), and PHA (bacterial polyesters) are undergoing considerable debate in scientific literature, providing divergent views [6–9]. The sustainability issue of each specific bioplastic is a complex problem, and several parameters must be considered, including the raw materials from which the bioplastic is generated, the energy consumed during bioplastic conversion, and its life cycle assessment analysis from production to ultimate disposal or recycle, with due recognition to the design and engineering of the bioplastic. In comparing the sustainability of a newly emerging bioplastic with a petroleum-based plastic, the analysis should take in to account the technology development time gap between petrochemicals (say ∼ 100 years old) and newly developing bioplastics (say from 5–10 years old or from now). The detailed descriptions of each of these factors are beyond the scope of this article. It is encouraging to derive cost-effective bio-based products or bio-composites from costly bioplastics through inexpensive natural/biofiber reinforcements. Most of the bioplastics cannot compete economically at their present state of technological development with the currently dominating petroleum-based plastics. The effective bio-composite formulations of such bioplastics from natural fiber reinforcements, through careful design and engineering, can result in new commercial attractions applications. The emergence of new applications of bio-composites would necessitate a large-scale demand of bioplastics, which would promote sustainability. A detailed understanding of natural/biofibers, bioplastic, and bio-composite formulations is necessary for the

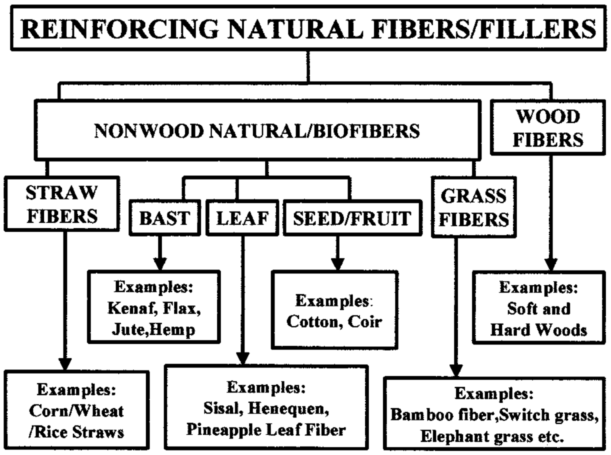


Fig. 3. Classification of natural/biofibers.

development of new and bio-based bio-composite materials. The following sections highlight issues in design and engineering of value-added bio-composite materials and the attainment of the sustainability of bio-based products to compete with petroleum-based products.

REINFORCING NATURAL/BIOFIBERS

After decades of high-tech development of artificial fibers such as carbon, aramid, and glass, it is remarkable that natural fibers such as kenaf, flax, jute, hemp, and sisal have attracted renewed interest, especially as a glass fiber substitute in the automotive industry. The advantages of natural fibers over synthetic or manmade fibers such as glass and carbon are low cost, low density, acceptable specific strength properties, ease of separation, carbon dioxide sequestration, and biodegradability.

Plastics are lighter but are not fit for load-bearing application because of the lack of sufficient strength, stiffness, and dimensional stability. However, fibers possess high strength and are sufficiently stiff but cannot be used for load-bearing applications because of their fibrous nature. In fiber-reinforced composites, the fibers serve as reinforcements by giving strength and stiffness to the composite structure. Natural/biofibers may be classified (Fig. 3) in two broad categories: nonwood fibers and wood fibers. In automotive applications, at the present level of technology, nonwood fibers such as hemp, kenaf, flax, and sisal have attained commercial success in the design of bio-composites from polypropylene. Replacement of polypropylene by biopolymers would result in more eco-friendly bio-composites for twenty-first–century green automotive parts applications. Grass fiber, another type of bio-fiber, is gaining the attention of scientists as a reinforcing fiber for automotive applications [10]. All the natural reinforcing fibers are lingno-cellulosic; the principal components are cellulose and lignin (Figs. 4–6). The contents of cellulose and lignin vary from one biofiber to another [4].

The exact chemical nature of the principal component of natural fiber, the lignin, still remains obscure. The main difficulty in lignin chemistry is that no method has so far been established by which it is possible to isolate the lignin in the native state from fiber. Although the exact structural formula of lignin in natural fiber has yet not been established, most of the functional groups and units that make up the molecule have been identified. Lignins are phenolic polymeric material formed from the phenolic precursors (Fig. 5) *p*-hydroxycinnamyl alcohols (I) such as *p*-coumaryl alcohol (II), coniferyl alcohol (III), and sinapyl alcohol (IV) through a metabolic pathway. A probable structure of lignin [11] is represented in Fig. 6.

It is well known that the tensile strength of natural fibers such as hemp, flax, jute, and sisal are lower than the tensile strength of E-glass fiber. The density of E-glass is much higher, i.e., about double the density of most of the natural fibers; thus the specific strength of some of the natural fibers are quite comparable to glass fibers. The E-modulus and specific modulus of some natural fibers are quite comparable or even superior [12, 13] to glass fibers (Table 1, Fig. 7).

BIOPOLYMERS: MATRIX POLYMERS

Biopolymers (biodegradable polymers; classification shown in Fig. 8) may be obtained from renewable resources, synthesized microbially, or synthesized from

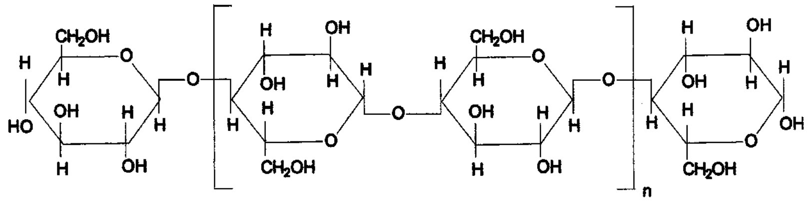


Fig. 4. Structure of cellulose.

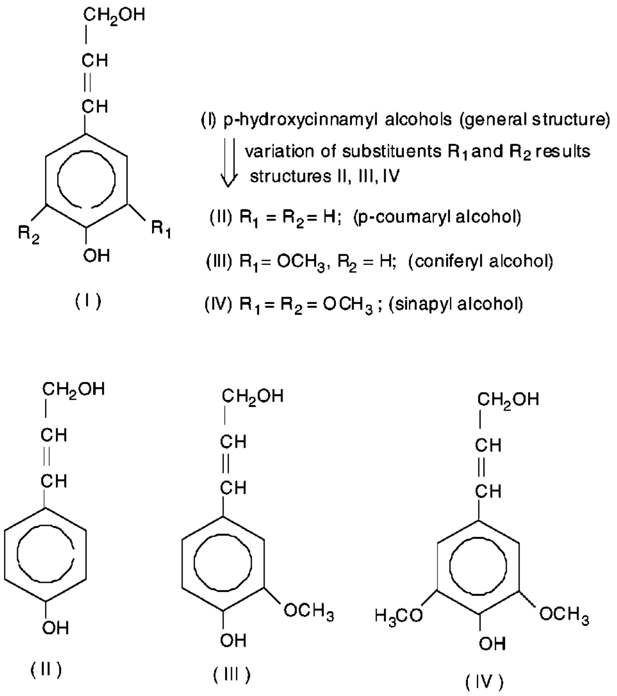


Fig. 5. Phenolic precursors that form the lignin.

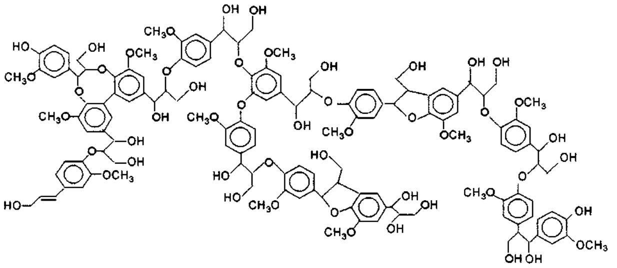


Fig. 6. Structure of lignin.

petroleum-based chemicals. Through blend of two or more biopolymers a new biopolymer may be designed for specific requirements. Thus biodegradability is not only a function of origin but also of chemical structure and degrading environment.

Table I. Modulus Comparison of E-Glass and Some Important Natural Fibers

Fiber Type	Density (g/cm^3)	E-modulus (GPa)	Specific Modulus (E-Modulus/Density)
E-glass	2.55	73	29
Hemp	1.48	70	47
Flax	1.4	60–80	43–57
Jute	1.46	10–30	7–21
Sisal	1.33	38	29
Coir	1.25	6	5
Cotton	1.51	12	8

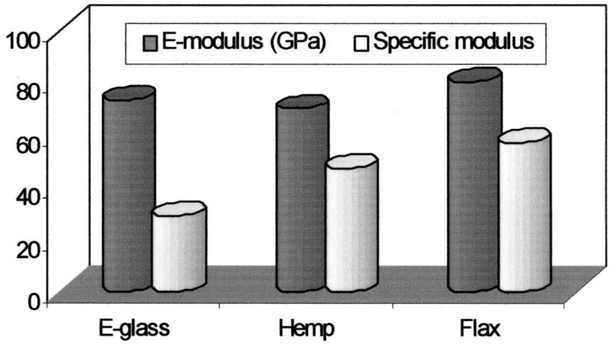


Fig. 7. Modulus and specific modules: E-glass versus some natural fibers.

Biodegradable polymers may be defined [3] as those that undergo microbially induced chain scission, leading to mineralization, photodegradation, oxidation, and hydrolysis, which can alter the polymer during the degradation process. Another definition states that biodegradable polymers are capable of undergoing decomposition, primarily through enzymatic action of microorganisms in to CO_2, methane, inorganic compounds, or biomass, in a specified period of time.

Biodegradable polymers need to be developed so as to make them suitable as matrix polymers for composite applications. Traditional plastics, such as polypropylene, polyethylene, polyester, and epoxy, have undergone considerable development and wide use in composite applications. Originally, biopolymers were intended to be used in packaging industries, farming, and other applications with minor strength requirements. Performance limitations and the high cost of biopolymers are major barriers for their widespread acceptance as substitute for traditional nonbiodegradable polymers. The high performance

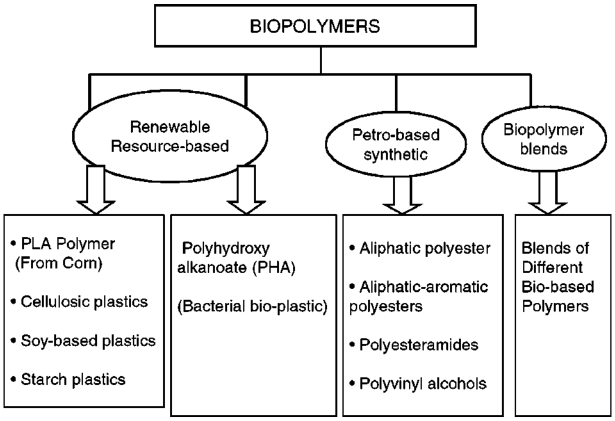


Fig. 8. Classification of biopolymers.

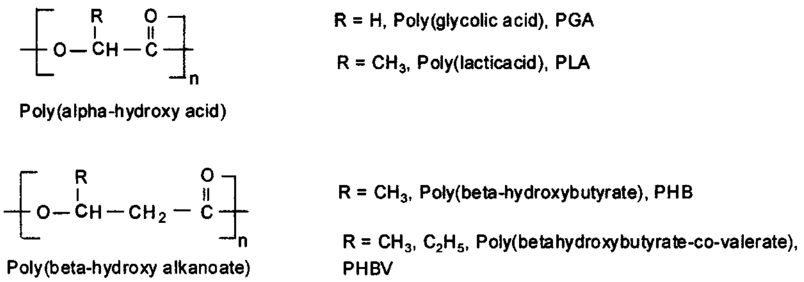
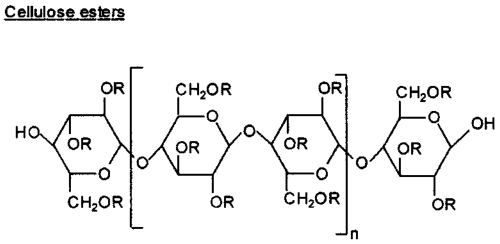


Fig. 9. Aliphatic polyesters—structure of some important biopolyesters.

of traditional plastic is the outcome of years of research; however, biopolymers are now of interest because of the current environmental threat and societal concern. The high cost of biopolymers compared to traditional plastics is not due to the raw material costs for biopolymer synthesis; rather it is mainly attributed to the low volume of production. New and emerging applications for biopolymers will result in increased production. The challenge for development of biodegradable polymers lies in the fact that such biopolymers should be stable during storage or usage and then degrade once disposed of after their intended lifetime. Biopolymers on reinforcement with biofibers can produce novel bio-composites to replace/substitute glass fiber–reinforced composites in various applications.

Biopolyesters (chemical structures of some biopolyesters are shown in Fig. 9) such as PLA and PHA are attracting much attention as promising biopolymers. PLA is a highly versatile biopolymer and is highlighted because it is derived from a renewable resource such as corn [14]. The use of products such as PLA as a cost-effective alternative to commodity petroleum-based plastic will increase the demand for agricultural products. The Cagill Dow plant in Nebraska is capable of producing 300 million pounds of renewable resource–based PLA per year (the plant became fully operational in January 2002) and uses up to 40,000 bushels of locally grown corn per day as the raw material for the manufacturing process. Bio-composites from natural fiber and PLA are attracting recent research interest [15]. The bacterial polyesters, e.g., PHA, have attracted recent attention as promising biopolymers in view of Metabolix's venture to make such biopolymers directly in plants [16]. Direct production of PHAs in plants would yield economics competitive with those of existing large-volume petrochemical polymers. In other words, the low costs achievable with plant-crop production of PHAs will allow polymers, materials, and chemicals derived from them to serve as realistic, cost-effective, sustainable alternatives to many of the largest-volume plastics and chemicals now made by the petrochemical industry. The first PHA discovered was polyhydroxybutyrate (PHB) [3]. Unlike conventional plastics, PHAs are produced from renewable resources—sugars in the case of the fermentation process and CO_2 and sunlight in the case of transgenic plants. The copolymer of PHA polymer, i.e., poly-β-hydroxybutyrate-co-alerate (PHBV), is successfully used as matrix polymer in designing jute fiber–based bio-composites [17]. Cellulose from trees and cotton plants is used as a substitute for petroleum feedstocks to make cellulosic plastic [18]. The natural polymer cellulose can undergo a reaction to produce a derivitized biopolymer. Cellulose esters are considered potentially useful biodegradable polymers. The structures of cellulose esters, including cellulose acetate (CA), cellulose acetate propionate (CAP), and cellulose acetate butyrate (CAB), are represented in Fig. 10. The cellulosic plastics such as CAB and CAP are now used in a variety of plastic applications. For example, premium toothbrush handles are typically made of CAP and screwdriver handles are often made from CAB. The production of cellulose esters from recycled paper and sugar cane has also been demonstrated [19]. Recently, cellulosic plastics are gaining importance in bio-composite formulations [20].

Research on applications of soybean for nonfood applications in plastics and composites is underway at various U.S. universities. Soybeans typically contain about 20% oil and 40% protein. Soy protein is available in three different forms, as soy flour, soy isolate, and soy concentrate. Both protein and oil from soybeans can be



n = 400-750; R = H (Cellulose), acetyl (Cellulose acetate), acetyl and propionyl (cellulose acetate propionate), or acetyl and butyryl (Cellulose acetate butyrate)

Fig. 10. Structure of cellulosic bioplastics.

converted to plastic/resin. Chemically, soy protein is an amino acid polymer or polypeptide and soy oil is a triglyceride. Through extrusion cooking and blending technology, soy protein polymers are converted to biodegradable plastics and their biocomposites [21], whereas through functionalization of soy oil [22], resin suitable for natural fiber composites is produced.

BIO-COMPOSITES: PRESENT TRENDS AND CHALLENGES FOR THE FUTURE

Cars made from grass may not sound like they would be sturdy, but plant-based cars are the wave of the future. Researchers at the Composite Materials and Structures Center at Michigan State University are working on developing bio-composite materials from plants such as hemp, kenaf, corn straw, and grass to replace plastic- and metal-based car components. Automakers now see strong promise in natural fiber composites [23]. Natural fiber composites are emerging as a realistic alternative to glass-reinforced composites. They can deliver the same performance for lower weight, and they can be 25–30% stronger for the same weight. Moreover, they exhibit a favorable nonbrittle fracture on impact, which is important requirement in the passenger compartment. In the United States, 10–11 million vehicles reach the end of their useful lives each year. A network of salvage and shredder facilities process about 96% of these old cars; about 25% of the vehicles by weight, including plastics, fibers, foams, glass, and rubber, remains as waste. A car made mostly of heated, treated, and molded biofiber would simply be buried at the end of its lifetime and consumed naturally by bacteria. Interior parts from natural fiber reinforced polypropylene composites and exterior parts from natural fiber reinforced polyester composites are already in use [24]. Ford Motor Company has a long history of R&D on new materials [25]. Henry Ford began experimenting with composites around 1941, initially using compressed soybeans to produce composite plastic-like components. During that period the petroleum-based chemicals were very cheap, and therefore soy-based plastic could not find economical importance. New environmental regulations, depletion, and uncertainty of petroleum sources have revived the interest of scientists in deriving a new generation of composite materials from soybean-based plastics and natural fibers. Johnson Controls, Inc., has started production [26] of door-trim panels from natural fiber and polypropylene. Auto companies are seeking materials with sound abatement capability and reduced weight for fuel efficiency. It is estimated that ~75% of a vehicle's energy consumption is directly related to factors associated with the vehicle's weight; thus there is a critical need to produce safe and cost-effective light-weight vehicles. Natural fibers possess excellent sound absorbtion efficiency, are more shatter resistant, and have better energy management characteristics than glass fiber in their respective composite structures. In automotive parts, compared to glass composites, the natural fiber composites reduce the mass of the component, lowering the energy needed for production [27] by 80%. It takes 6500 BTUs of energy to produce 1 pound of kenaf, whereas it takes almost four times that much of energy (~23,500 BTUs) to produce 1 pound of glass fiber [4]. To reduce vehicle weight, a shift away from steel alloys toward aluminum, plastics, and composites indicates that in the near future, polymer and polymer composites will comprise ~15% of a car's weight. Auto parts save mass by going from steel to glass fiber–reinforced plastic (GFRP). Our research has demonstrated that natural fiber composites show comparable or even superior mechanical properties over GFRP. Replacing glass by natural fiber would reduce the mass significantly. American market studies clearly identify the potential impact and opportunities for natural fiber composites [28]. In the year 2000, the North American market for natural fiber composites exceeded $150 million, and by 2005 this market is expected to reach nearly $1.4 billion in sales.

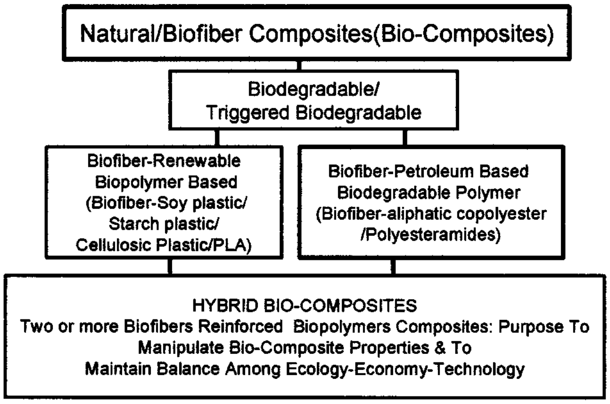


Fig. 11. Classification of bio-composites.

Natural fiber–traditional nonbiodegradable polymer–based composites are gaining market demand in light of increasingly strict environmental regulations, and eco-friendly bio-composites (Fig. 11) are attracting the attention of many industries. The challenge in replacing conventional glass-reinforced plastics with bio-composites is to design materials that exhibit structural and functional stability during storage and use, yet are susceptible to microbial and environmental degradation upon disposal, with no adverse environmental impact. A group of researchers at the Composite Materials and Structures

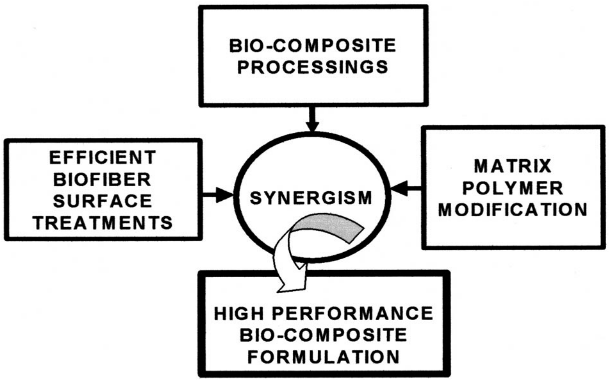


Fig. 12. Challenges to fabricate superior bio-composites.

Center at Michigan State University are applying their knowledge in composite technology in a new, broadened way to fabricate sustainable bio-composites from renewable resources. Besides using commercial green/bioplastics such as cellulose acetate, starch plastics, bacterial polyesters, and PLA, researchers are engaged in developing bioplastic from soy protein and vegetable oils through innovative technology. Our tri-corner approach in designing superior-strength (Fig. 12) bio-composites is:

- Efficient (low-cost but effective) biofiber treatment,
- Matrix modification (functionalization, blending), and
- Selection of efficient processing conditions.

The significant attraction of biofibers is their low cost; surface treatments that avoid using organic solvents i.e., water-based sizing, alkali treatment, and silane treatments, are logical approachs [29] to making reactive biofiber. Superior-strength bio-composites can be obtained through application of the reactive engineered natural fibers concept [30]. Design of engineered natural/Biofiber is schematically represented in Fig. 13. Engineered biofibers are defined as the suitable blend of surface-treated bast (e.g., kenaf, hemp) and a leaf fiber (e.g., pineapple leaf fiber [PALF]). Selection of blends of biofibers is based on the fact that the correct blend achieves an optimum balance in mechanical properties. The kenaf and/or hemp–based composites exhibit excellent tensile and flexural properties, while leaf fiber (PALF) composites have the best impact properties of the composites. The combination of bast and leaf fibers may be selected to achieve a balance of flexural and impact properties of the targeted bio-composites [30].

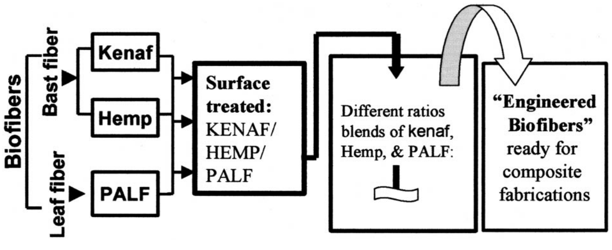


Fig. 13. Concept on design of engineered natural/biofiber.

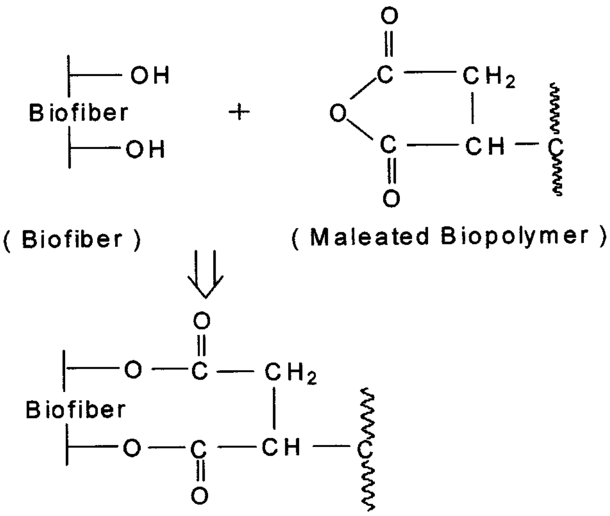


Fig. 14. Reacter of biofiber and maleated polymer—mechanism for compatibilization.

Maleic anhydride (MA) is used as a grafting monomer to functionalize biopolymer because of the higher reactivity of the anhydride group. MA-functionalized polymer is of considerable importance for application as a copolymer precursor in polymer blends and also as an adhesion promoter in bio-composite applications [31]. There are two ways by which the maleic anhydride compabilization chemistry can be applied during bio-composite fabrications: (1) Natural/biofibers are pretreated with maleated polymer, and such treated biofibers are then reinforced with the desired polymer matrix to obtain the bio-composites. (2) During extrusion processing chopped biofiber, polymer matrix, and maleic anhydride can be added with peroxide initiator in one-step processing to get the comptabilized bio-composite product for further compression molding/injection molding. During reactive extrusion processing, maleic anhydride reacts with the polymer matrix to form maleated biopolymer, which then reacts (see Fig. 14) with the biofiber. We target the cascade engineering approach in designing bio-composites for industrial attractions. Cascade engineering principles target a minimum or reduced number of steps to produce a final composite product. For example, the functionalized biopolymer (e.g., maleated biopolymer as the compatibilizer), the biofiber, and the polymer matrix in appropriate amounts can be fed simultaneously into the extruder in one step. The compatbilized extruded bio-composite granules on further injection molding would result in the final composite samples. Another novel approach is to adopt environmentally benign powder impregnation processing in fabricating the bio-composites [30]

CONCLUSIONS

Green materials are the wave of the future. There is immense opportunity in developing new bio-based products, but the real challenge is to design sustainable bio-based products. New environmental regulations and societal concern have triggered the search for new products and processes that are compatible with the environment. The incorporation of bio-resources in composite materials can reduce further dependency on petroleum reserves. The major limitations of present biodegradable polymers are their high cost. Renewable resource–based bioplastics are currently being developed, further research should overcome the performance limitations. Bio-composites can supplement and eventually replace petroleum-based composite materials in many applications, offering new agricultural, environmental, manufacturing, and consumer benefits. Several critical issues related to biofiber are surface treatment to make it more reactive, bioplastic modification to make it a suitable matrix for composite application, and development of appropriate processing techniques, depending on the type of fiber form (chopped, nonwoven/woven fabrics, yarn, silver, etc.), to meet the needs of commercial interests. Bio-composites are emerging as a realistic alternative to glass-reinforced composites. Because bio-composites are derived from renewable resources, materials costs can be markedly reduced with their large-scale usage. Recent advances in genetic engineering, natural fiber development, and composite science offer significant opportunities for improved value-added materials from renewable resources with enhanced support of global sustainability. Natural fibers are biodegradable, but renewable resource–based bioplastic can be designed to be either biodegradable or not according to the specific demands of a given application. Their unique balance of properties would open up new market development opportunities for bio-composites in the twenty-first–century green materials world.

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