Composites from renewable and sustainable resources: Challenges and innovations

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Interest in constructing composite materials from biosourced, recycled materials; waste resources; and their combinations is growing. Biocomposites have attracted the attention of automakers for the design of lightweight parts. Hybrid biocomposites made of petrochemical-based and biosourced materials have led to technological advances in manufacturing. Greener biocomposites from plant-derived fiber and crop-derived plastics with higher biobased content are continuously being developed. Biodegradable composites have shown potential for major uses in sustainable packaging. Recycled plastic materials originally destined for landfills can be redirected and repurposed for blending in composite applications, thus leading to reduced dependence on virgin petro-based materials. Studies on compatibility of recycled and waste materials with other components in composite structure for improved interface and better mechanical performance pose major scientific challenges. This research holds the promise of advancing a key global sustainability goal.

The era of natural fiber composites currently known as biocomposites dates back to 1908 with the introduction of cellulose fiber-reinforced phenolic composites. This innovation was followed by synthetic glass fiber-reinforced polyester composites, which obtained commodity status in the 1940s. The use of biobased green polymers to manufacture auto parts began in 1941, when Henry Ford made fenders and deck lids from soy protein–based bioplastic. The use of composite materials, made with renewable and sustainable resources, has become one of the vital components of the next generation of industrial practice. Their expanding use is driven by a variety of factors, including the need for sustainable growth, energy security, lower carbon footprint, and effective resource management, while functional properties of the materials are simultaneously being improved. Innovative sustainable resources such as biosourced materials, as well as wastes, coproducts, and recycled materials, can be used as both the matrix and reinforcement in composites to minimize the use of nonrenewable resources and to make better use of waste streams.

Composite materials find a wide range of potential applications in construction and auto-parts structures, electronic components, civil structures, and biomedical implants. Traditionally, industrial sectors that require materials with superior mechanical properties use composites made from glass, aramid, and carbon fibers to reinforce thermoplastics such as polyamide (PA), polypropylene (PP), and poly(vinyl chloride) (PVC), as well as thermoset resins such as unsaturated polyester (UPE) and epoxy resin. In addition to fiber, mineral fillers such as talc, clay, and calcium carbonate are being used in composite manufacturing. Such hybrids of fiber and mineral fillers play a major role in industrial automotive, housing, and even packaging applications. Carbon black plays a vital role as a reinforcement, especially in rubber-based composites. The key environmental concern with regard to composite materials is the difficulty of removing individual components from their structures to enable recycling at the end of a material’s service life. At this point, most composite materials are either sent to a landfill or incinerated. Wood and other natural fibers (e.g., flax, jute, sisal, and cotton), collectively called “biofibers,” can be used to reinforce fossil fuel–based plastic, thus resulting in biocomposite materials. Synthetic glass fiber–reinforced biobased plastics such as polylactides (PLAs) are a type of bio-composite. Biofiber-PP and biofiber-UPE composites have reached commodity status in many auto parts, as well as decking, furniture, and housing applications. Hybrid biocomposites of natural and synthetic fibers as well as mixed matrix systems also represent a key strategy in engineering new classes of biobased composites. As part of feedstock selection, a wide range of renewable products that includes agricultural and forestry residues, wheat straw, rice straw, and waste wood, as well as undervalued industrial coproducts including biofuel coproducts such as lignin, bagasse, and clean municipal solid wastes, is currently being explored to derive chemicals and materials. Recent advancements in biorefinery concepts create new opportunities with side-stream product feedstock that can be valorized in the fabrication of a diverse array of biocomposites.

Materials scientists can help in advancing sustainable alternatives by quantifying the environmental burden of a material through its product life-cycle analysis (1, 2). The exponential growth of population and modernization of our society will lead to a threefold increase in the demand for global resources if the current resource-intensive path is continued (3). According to the United Nations, a truckload of plastic waste is poured into the sea every minute. By 2050, at current rates, the amount of plastic in the ocean will exceed the number of fish. The benefit of diverting plastic packaging material is estimated at around $80 billion to $120 billion, which is currently lost to the economy (4). If diverted for composite use, the recycled and waste plastic currently destined for landfills and incineration would be used for sustainable development, thereby reducing dependence on nonrenewable resources such as petroleum. Postindustrial food processing wastes are being explored as biofiller in biodegradable plastics for the development of compostable biocomposites. Low-value biomass and waste resources can be pyrolyzed to provide biocarbon (biochar) as sustainable filler for biocomposite uses. The increased sustainability in composite industries requires basic and transformative research toward the design of entirely green composites. Renewable resource–based sustainable polymers and bioplastics, as well as advanced green fibers such as lignin-based carbon fiber and nanocellulose, have great potential for sustainable composites. Biobased nonbiodegradable composites show promise in sustainable packaging. This comprehensive Review on composites from sustainable and renewable resources aims to summarize their

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current status, constraints on wider adoption, and future opportunities. In keeping with the broad focus of this article, we analyze the current development of such composites and discuss various fibers and fillers for reinforcements, current trends in polymer matrix systems, and integration of recycled and waste coproducts into composite systems to outline future research trends.

Fibers and fillers from renewable and sustainable resources

In polymer composites, plastic resins remain as continuous phase, whereas fibers and fillers stay in discontinuous phase to provide reinforcement effects. The composite performance is governed through the interface between the fibers and the polymer matrix. In composite science, the key target is the interface and related interfacial bonding as the stress transfer between the fiber and polymer matrix dictates the overall mechanical performance. When deciding on the appropriate fiber and filler system for sustainable composite use in industrial sectors, it is necessary to compare the cost and availability, consistency of properties, and environmental advantages of sustainable fibers with those of their traditional synthetic counterparts. Figure 1 depicts examples of different types of fibers and fillers for biocomposites.

Lignocellulosic plant fibers

This category, well known as natural fiber or biofiber, is broadly classified as wood and nonwood fibers. These include various types such as bast, leaf or fruit, straw, grass, and wood. The mechanical performance of various plant fibers depends on their cell wall structure, composition, and morphology. Cellulose content, lumen size, and microfibrillar angle are other key factors governing the stiffness of plant fiber reinforcements. The advantages of biofibers over traditional glass fibers and mineral fillers are their lower density, reduced cost, eco-friendly nature, and enhanced performance in certain applications. Among the available natural fillers that can be used for composite applications, wood is the most commonly used. Cotton fibers are also widely available. Other agricultural natural fibers such as flax, jute, kenaf, industrial hemp, and sisal are used as well. Because of their desirable structural properties, the construction field is the major arena for the use of natural fiber composites. Unconventional natural fibers such as agro-residues (e.g., wheat and rice straws, ground coconut shells) and grasses (e.g., miscanthus, switch grass, and bamboo) find application in biocomposites.

Biofuel coproducts, food processing wastes, and other postindustrial wastes

Value-added uses of coproducts and by-products from biorefinery and biomass conversion processes are beneficial for sustainable development. Distillers’ dried grains with solubles (DDGS) from the corn ethanol industry, lignin coproduct from lignocellulosic ethanol industry as well as the pulp and paper industry, and bagasse from the sugarcane ethanol industry are being used in biocomposites. Lignin, which is polyphenolic in composition, has found value-added uses in sustainable biobased composite materials. Lignin, with its many functional surface -OH groups as well as with surface modification, has shown improved reinforcing effect on resulting biocomposite performance. Additionally, fruit and vegetable pomace, coffee chaff, and grain hulls have been explored in composites. Most of these biofillers are used for waste valorization, whereas the resulting biocomposites can be used for nonstructural applications. Chicken feathers, currently treated as waste in the global poultry industry, can find application as renewable fiber reinforcement in lightweight biocomposites. Recycled carbon fiber from the aerospace and sporting goods industries is another sustainable source that can be used to generate hybrid composites with biofibers.

Biocarbon, a new sustainable filler and functional material

Biocarbon, also known as biochar, has emerged as a new sustainable material for many applications. Biochar is not limited to filler and reinforcement uses for biocomposites; it is also beneficial for the development of next-generation functional carbon materials for potential applications in energy storage and filtration devices. The thermochemical conversion of biomass when oxygen is absent or in limited supply (also known as pyrolysis) generates liquid bio-oil, solid biochar, and syngas. Depending on temperature, time, and the nature of biomass in the pyrolysis process, the amount of oil, char, and gas may vary. Biochar or biocarbon (BioC) is an amorphous carbon-rich material that can be tunable in terms of chemical structure, porosity, size, and intrinsic modulus. Two other amorphous carbon-based materials are activated carbon (AC) and carbon black (CB). The main distinctions among such carbon-rich materials are based on their origin, formation process, and structure. The carbon content of BioC varies from 40 to 90%, as compared with 80 to 95% for AC and >95% for CB. Regarding origin of formation, BioC is generated from biomass; AC from asphalt, coal, and biomass; and CB from petroleum and coal tar.

Matrix polymer from renewable and sustainable resources

The majority of plastic resins in biocomposites are predominantly concentrated among petro-based...
The development of polymers from sustainable renewable feedstocks poses key scientific challenges from a combined societal, economic, environmental, and human health perspective. The sustainability measurement is difficult to quantify. Although more energy is required to produce bio-based low-density polyethylene (bio-LDPE) in comparison with its petroleum-based counterpart, its overall global warming potential from the life-cycle assessment perspective is relatively lower (16).

**Renewable and sustainable resource-based biocomposites: Scientific challenges**

Fiber-matrix adhesion, matrix and fiber modification, hybrid strategy, and the desired processing approach are key factors in making high-performance biocomposites for specific end-use applications. Across the wide spectrum of possible matrix and fiber/filler systems, hybrid synergistic assembly for improved compatibility is a key scientific challenge. Biofibers have been developed to tackle the biofiber drawbacks and potential for packaging applications. Across the wide spectrum of possible matrix and fiber/filler systems, hybrid synergistic assembly for improved compatibility is a key scientific challenge. Biofibers have been developed to tackle the biofiber drawbacks and potential for packaging applications.
A Matrix compatibilization strategy

i. Physical compatibilization

PE/iPP blends
Block copolymer design

LHI

PE-block-iPP-block-PE-block-iPP

ii. Physical & Chemical compatibilization

Branching and crosslinking
PLA/iPP blends

polyethylene-glycidyl methacrylate–methyl acrylate terpolymer (PEGMA)

B Matrix & Fiber modification

Grafting strategy
Acrylation
Maleation
Acetylation
Plasma
Oligomers
Polymers...

Treatment for natural fibers
Physical
Milling, sizing
Pattern fabric...

Chemical
Alkaline
Silane chemistry...

Fiber with enhanced functionality

Adhesion mechanism by trapped entanglements

C Hybrid synergistic assembly in sustainable composites

Hybrid Morphology

Compatibilization Reactions

Interphase phenomena

Fig. 3. Compatibilization strategies. The scheme in section i of (A) was redrawn from (18) with permission. The scheme in section ii of (A) was reprinted and adapted from (19) with permission; copyright (2015) American Chemical Society.
into three types: partially biobased, fully biobased, and hybrid biocomposites. In accordance with the selected polymer resin, the resulting composites may be thermoplastic or thermostet in nature. Figure 4 shows a schematic of biocomposite production from some representative raw materials and their processing in manufacturing, packaging, and consumer goods areas.

Natural fiber composites are more environmentally friendly, economic, and lightweight than traditional glass or aramid fibers and talc-filled composites in both thermoplastic and thermostet platforms. They have many industrial uses, including applications in construction, automotive parts, and sporting goods. Biocomposites also have potential usage in electronics and specialty niche markets. A wide array of agro-forestry biofibers (predominantly wood, as well as flax, kenaf, and sisal) have been explored as natural reinforcement or fillers for composite fabrication. For auto manufacturers, one direct benefit of using sustainable composites is greater assurance of long-term price stability. Thus, many automakers continue to seek low-cost alternatives using nontraditional fibers and fillers and matrix systems including “waste” agro-forestry coproducts, waste rubber, cork, and recycling waste (20). Another example is the substitution of ground coconut shells (the major by-product of the coconut processing industry) for mineral-based talc. Natural fiber composites in auto parts have been thoroughly explored (21). Both reinforcement and matrices are being substituted with sustainable materials.

Plastics originating from renewable resources (e.g., PLA and Bio-PA) are being studied for automotive applications. More attention is now being devoted to the use of recycled content with virgin plastics for the fabrication of composite materials. However, the broader acceptance of these biocomposite materials in automotive applications depends on many factors, including class A finishing, moisture repellence, structural stability, and flame-retardant properties. Sustainable composites are used in auto parts such as trim panels, seat backs, packaging trays, spare wheel covers, headliners, dashboards, and air-baffle components. Apart from automotive components, sustainable composites also receive considerable attention in construction and packaging applications. Tables 1 and 2 summarize a few representative biocomposites with their key properties and industrial uses.

Needle-punch flax/PP mats on compression molding result in biocomposites with very high impact properties that may result from the aligned nature of the fibers, along with high fiber loading (50%) (22). The construction and municipal waste plastic-wood fiber-based biocomposites exhibited undesirable properties compared with virgin plastic LDPE-based wood-plastic composites (23). Multicomponent systems using such mixed plastic wastes (mostly PP and PE) deliver inferior performance due to incompatibility. Food wastes provide another potential sustainable resource for composite manufacture. The use of biofuel coproduct and food processing wastes as filler in biocomposites may help improve modulus, as compared with virgin plastic, while sacrificing toughness. The composite matrix can also be made from renewable resources. Bio-PU can be prepared from rubber seed oil monoglyceride reacting with diisocyanate (24). Combination of PLA and PBS, and, in a larger sense, biodegradable aliphatic polyesters with natural fibers, can lead to biodegradable formulations (25). Because of their woodlike aesthetic, hybrid biocomposite formulations containing natural resins with lignin and natural fibers were commercially successful when used in jewelry and musical instruments (26). Coffee chalk, a lignocellulosic waste from the coffee roasting industry, has been used to develop compostable biocomposites for a disposable food packaging application in coffee pods (27). Kiwifruit skin waste biomass and PLA, as well as grape pomace—biodegradable polyester-based biocomposites—resulted in compostable knives and clips, respectively, for industrial applications (28). DDGS, a corn ethanol coproduct, has been used as filler to increase the thermomechanical properties of PHA bioplastic and the biodegradation of the resulting composites, showing potential for disposable agricultural applications (29). Chicken feather fiber (CFF) waste from the poultry industry, with glass fiber and epoxy resin, resulted in lightweight hybrid biocomposites (30). As compared with glass fiber–epoxy composites, the CFF or CFF–glass fiber hybrid composites exhibited 30 to 40% density reduction because of internal void and lower aspect ratio of CFF as compared with glass fiber.

The biocarbon from pyrolyzed biomass and waste resources has created opportunities to advance sustainable composites, ranging from

![Fig. 4. Biocomposite product examples: From raw materials to manufacturing.](image-url)
their uses in commodity thermoplastic such as PP (31) to high-melting engineering plastic such as PET (32). Limitations of plant fiber in current biocomposite uses include unwanted odor, hydrophilicity, and low thermal stability. Because biofibers degrade around 200°C, their widespread use in engineering plastics is restricted. In the automotive industry, tires and polyurethane-based products contain CB filler up to 50 wt. %. Again, most of the interior auto parts are made in black with the help of petro-based CB. Along with these components, the electronic housing products are also made with CB as a colorant. The pyrolysis process also results in bio-oil and syngas, which have been explored as raw materials for various grades of fuels and chemicals. In comparison with traditional natural fibers, biocarbon is thermally stable and is particularly suitable to reinforce or fill thermoplastic engineering plastics, like PET, for biocomposite uses. Traditionally associated with high strength and stiffness, the definition of advanced composites is evolving toward expression of multifunctionality that may include a combination of sophisticated mechanical properties with high weight, light weight, and electrical and thermal conductivity. Appropriate functionalization of bioresin with the intended reinforcement can result in marked performance improvements. Highly functionalized resin derived from vegetable oil reinforced with fiberglass fabric resulted in advanced biocomposites (33). Lignin-based carbon fibers and nanocelluloses are considered the next generation of biobased reinforcements for sustainable composite applications. Carbon fiber is attracting attention for lightweight composite uses. Synthetic carbon fiber is made mostly from acrylonitrile. Lignin, being a renewable resource, has been heavily researched for the development of lignin-based carbon fibers (LCFs) (34). The diversity of lignin sources poses challenges in the manufacturing process. The pretreatment process and mechanical performance, among other aspects, must be further developed to bring more cost-competitive LCFs to commercialization. Nanocelluloses from agricultural and industrial waste have also attracted attention as sustainable materials (35). Nanofibrillated cellulose and cellulose nanocrystals are two key types of nanocelluloses. Polymer- and nanocellulose-based composites are under constant development (36). Melt processing of nanocellulose and thermoplastic has drawn more attention, and the major scientific challenges pertain to process development for improved dispersion of nanocellulose in the polymer matrix. Surface modification of nanocellulose is critical for improved fiber-matrix adhesion. Treatment of microfibrillated cellulose (MFC), a nanosized cellulose fiber, with silica changed its surface characteristics from hydrophilic to hydrophobic without affecting the crystalline structure (37), and such modification improved fiber-matrix adhesion in MFC-epoxy composites. Green composites have been developed using MFC-modified wax maize starch–based bioresin and modified liquid crystalline cellulose through hand lay-up and compression molding. Such advanced composites exhibit very high strength (~800 MPa), thus creating new possibilities for structural applications (38). 3D printing, also known as additive manufacturing, can be widely used in sustainable composite manufacturing, especially in biomedical, automotive, and construction industries. 3D printing enables the creation of complex structures that cannot be created by traditional composite manufacturing technologies. A 3D printing technique has been developed for processing unidirectional continuous fiber-reinforced PLA composites with very high modulus and strength (39).

**Challenges in adopting sustainable composites widely**

More eco-friendly composites with enhanced sustainability face challenges to their wide-scale application. Measuring the sustainability of plastic and reinforcement/fillers is a complex task affected by factors such as the nature of the feedstock, energy input during production, durability, health impacts, and after-life recycling or disposal (40). Biomass supply chains, which address types of biomass, harvesting and collecting strategies, transport and storage mechanisms, as well as processing methodologies, are complex in nature and often vary with biomass type. It is necessary to establish a unified protocol for the effective utilization of bioresources, including waste resources. The sustainable method of expanding purpose-grown biomass, for example, requires the use of marginal agricultural land. Such an approach is essential for meeting the emerging massive requirement for biomass in the future. Durability is a critical test for any biocomposite material proposed for replacing traditional synthetic composite materials. To achieve functionality, biocomposite materials for automotive, construction, and other structural applications need to deliver the required service life and long-term durability. Inclusion of bioplastic and recycled materials in sustainable composite uses poses major scientific challenges. Designing and engineering new classes of biocomposite materials that can exhibit high tolerance against various external factors is essential. The classification of biodegradable and nonbiodegradable

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**Table 1. Properties of representative biocomposites and their hybrids.** –, not determined; MAPE, maleic anhydride–grafted polyethylene.

<table>
<thead>
<tr>
<th>Resin</th>
<th>Filler</th>
<th>Impact strength</th>
<th>Tensile strength</th>
<th>Tensile modulus</th>
<th>Comments</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plastic waste (PE and PP)</td>
<td>Wood flour</td>
<td>2.9–6.2 kJ/m², Unnotch</td>
<td>6–13 MPa</td>
<td>2.3–3.9 GPa</td>
<td>MAPE compatibilization and lubricant utilization</td>
<td>(23)</td>
</tr>
<tr>
<td>PP</td>
<td>Wood, poultry litter biochar</td>
<td>8.1 kJ/m², Notch</td>
<td>27 MPa</td>
<td>4.3 GPa</td>
<td>Hybrid biocomposites–MAPP compatibilization</td>
<td>(31)</td>
</tr>
<tr>
<td>PP</td>
<td>Flax fiber</td>
<td>751 J/m, Unnotch</td>
<td>40 MPa</td>
<td>6.5 GPa</td>
<td>Needle-punch fiber mat composite</td>
<td>(22)</td>
</tr>
<tr>
<td>Waxy maize starch</td>
<td>Neat and modified liquid crystalline cellulose, microcrystalline cellulose</td>
<td>–</td>
<td>505–790 MPa</td>
<td>22–32 GPa</td>
<td>Starch/cellulose hybrid biocomposites</td>
<td>(38)</td>
</tr>
<tr>
<td>Epoxy/acrylate</td>
<td>Glass fiber</td>
<td>237 kJ/m², Notch</td>
<td>532 MPa</td>
<td>37 GPa</td>
<td>Methacrylated epoxidized sucrose soyaate resin/glass fiber</td>
<td>(33)</td>
</tr>
<tr>
<td>Bio-polyurethane (Bio-PU)</td>
<td>Sisal fiber</td>
<td>–</td>
<td>57–119 MPa</td>
<td>1.2–2.2 GPa</td>
<td>Rubber seed oil polyurethane</td>
<td>(24)</td>
</tr>
<tr>
<td>PBS/PLA</td>
<td>Flax fiber</td>
<td>9.1-178 kJ/m², Notch</td>
<td>39–55 MPa</td>
<td>3.6–7.4 GPa</td>
<td>Fully biodegradable composite</td>
<td>(25)</td>
</tr>
<tr>
<td>PLA</td>
<td>Carbon fibers, twisted yarns of jute fibers</td>
<td>–</td>
<td>57–185 MPa</td>
<td>5.1–19.5 GPa</td>
<td>Continuous fiber reinforcement probed by 3D printing</td>
<td>(39)</td>
</tr>
</tbody>
</table>
composites is also important from an application perspective. In addition to durable applications, certain biocomposites are targeted for short life cycles. These materials must adhere to international standards for biodegradability and compostability. One challenge is having the required composting facility at the disposal site of such materials.

**Outlook**

Fossil fuel–based traditional composite structures persist in the environment. Because they are minimally recycled, these materials often end up being incinerated or placed in a landfill. In addition to fiber-reinforced composites, minerals such as talc and calcium carbonate–filled polymer composites and/or their hybrids with fibers are being used widely in composite industries. Hybrid biocomposites with petro- and bio-based combinations, which are neither 100% fossil fuel–based nor 100% bio-based, have achieved some commercial success. Wood and other agricultural natural fibers (flax, jute, etc.) used with petro-based plastics (PP, PE, epoxy, etc.) are more eco-friendly than 100% fossil fuel–based composites and have found use in housing structures, decking industries with wood plastic composites, other natural fiber–based hybrid biocomposites in automotive parts, and consumer products. Biocomposites from recycled fibers and natural fibers have also entered into consumer product applications. Currently, all-green (i.e., 100% bio-based) composites exhibit limited success because of their cost and durability restrictions in automotive and/or housing structures. All-green composites and biodegradable plastics are gaining momentum in sustainable packaging. The use of sustainable biocarbon fillers derived from waste biomass, industrial waste, and food waste demonstrates enormous potential for lightweight sustainable composites in auto parts and other growing demands from the manufacturing sector. Achieving increased utilization of wastes and undervalued industrial coproducts depends on creating a strong value proposition across the entire value chain. The economic and functional merits of composites made from renewable and sustainable resources must be coupled with leadership from industry executives and senior government officials to drive global growth in this innovative class of materials for positive societal, environmental, and economic impacts.

**REFERENCES AND NOTES**


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