Materials from Renewable Resources

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Guest Editors

Abstract

The drive for greater use of renewable materials is one that has recently gained momentum due to the need to rely less heavily on petroleum. These renewable materials are defined as such since they are derived from plant-based sources. Some renewable materials also offer properties that conventional materials cannot provide: hierarchical structure, environmental compatibility, low thermal expansion, and the ability to be modified chemically to suit custom-made applications. Nature's materials, particularly from plant- and animal-based polysaccharides and proteins, have hierarchical structures, and these structures can be utilized for conventional applications via biomimetic approaches. This issue begins with an article covering renewable polymers or plastics that can be used to generate block copolymers (where two polymers with specific functions are combined) as an alternative to conventional materials. Applications of renewable polymers, such as cellulose from plants, bacteria, and animal sources, are also covered. Also presented are the use of bacterial cellulose and other plant-based nanofibers for transparent electronic display screens and, in a wider sense, the use of cellulose nanofibers for composite materials, where renewable resources are required to generate larger amounts of material. Finally, this issue shows the use of biomimetic approaches to take the multifunctional properties of renewable materials and use these concepts, or the materials themselves, in conventional materials applications.

Introduction

Recently, a large emphasis has been placed on the need to better utilize resources such as oil, mostly for energy purposes, but also for the generation of plastics. At present, the human race is thought to be at or near peak oil consumption, depending on which theory is used.[1] After peak oil consumption, it is predicted that the use of this resource will decline dramatically as a result of the dwindling supply. At the same time, the price of oil is volatile, often increasing dramatically at times of economic and/or supply uncertainty. It is envisaged that as supplies decrease, the cost will increase. This places an onus on the human race to utilize other resources, even if we just want to maintain our productivity and consumption at current levels. As the worldwide population continues to grow, more strain will be placed on declining oil resources due to increased consumption.

Oil is derived from plant material, which is formed under high pressure over many millions of years, and so, in a sense, oil is renewable when viewed over a long period of time. This large time-disparity between the renewability of this resource and the rapid industrial expansion and consumption of oil has led to its further depletion. Energy is the most consumptive use of oil. The International Energy Agency has warned that the demand for energy will increase by 45% between now and 2030, and this will put further pressure on this resource.

Typical levels of oil usage for chemicals in the mid-1990s only reached about 7% of the total usage.[2] Other sources of chemicals for materials from plants are available that have renewable timescales more in-tune with our consumption. These materials can be derived from natural oils present in plants and the cellulosic material that they use structurally. Nature also has its own consumption and renewable materials cycle, which would continue without the intervention of humankind. These cycles appear to take place with relatively less energy consumption than most industrial processes. For instance, one example of a natural low-energy process is sequestration of carbon dioxide and its subsequent formation into long chain sugars, which form the structure of plants; this process is fueled by the capture of light. The plant matter produced also can degrade and provide nutrients for successive plant life in a cycle that appears to consume little in the way of resources—neglecting usage of land, modern farm machinery, and, more recently, water. Annual planting and harvesting of crops could provide a significant proportion of the total biomass required for fuel/energy (10–20%) and materials consumption,[3] although we have not yet developed this to its full extent.

Figure 1 summarizes the two process routes to materials derived from natural and oil-based resources, showing the presence of significant energy inputs. Energy inputs into the production of oil are thought to be higher than from harvesting natural feedstocks, although significant costs are associated with the latter. The processing of natural and oil-based feedstocks could use similar technology, and both require a significant energy input at this stage. Any natural process that generates material without the use of significant energy input is likely to be competitive to either of these processing routes. One energy input not explicitly considered in this summary is transportation. Any energy cost analysis on the relative benefits of using a renewable resource would have to take this into consideration. It is, however, worth pointing out that the sequestration of carbon dioxide during the plant growth process could potentially make a significant contribution to climate change. Some examples of products are given in the lower part of Figure 1, where often the competition between the usage of these materials is presently governed on the basis of price. However, this competition is likely to become less significant with a dwindling oil supply. There is a potential for both renewable and oil-based materials to be recycled, with an additional option of degrading the former in landfills. The presence of oil-based materials in landfills, although posing immense problems, could be viewed as a recycling resource if separation can be achieved.

There is a range of renewable resources that has been extensively researched, particularly for use as polymers and composites.[4] The materials that have been investigated include naturally occur-

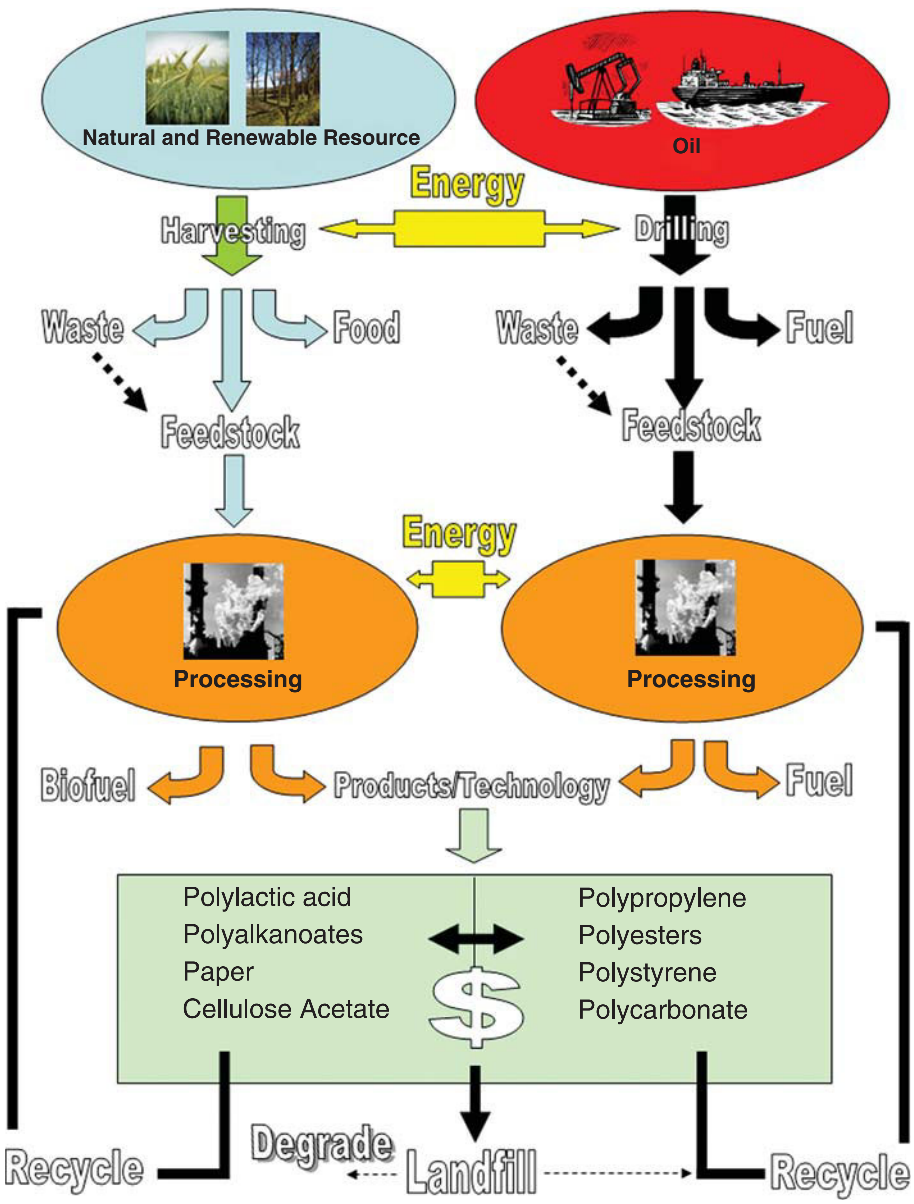

Figure 1. Summary of renewable materials versus oil production indicating stages of significant energy input and some examples of products with their competition on price.

ring polymers such as cellulose, chitin, starch, and numerous proteins (collagen, keratin, silk).

The first of these materials (cellulose) is the most utilized, with traditional applications as a building material in the form of wood, in textiles from cotton, for packaging, and print media in the form of paper. Cellulose also is widely used for films and as a coating material, as a polyelectrolyte material, as an emulsifier in food, and as a drug-delivery medium. The first-ever plastic was formed from a derivatized cellulose (cellulose nitrate).[2] Subsequent to this, the development of cellulose acetate, particularly for filter tow for cigarettes, has dominated the worldwide markets in terms of production of cellulosics. In 2003, this production stood at 600,000 metric tons.[5] Cellulose acetate also has been utilized for photographic film, although with the advent of digital technology, this market is currently diminishing. Recently, the demands for cellulose acetate have been for LCD flat screens.[5] Other cellulose derivatives, such as hydroxypropyl cellulose, are used in the pharmaceutical industry. Non-derivatized grades, such as microcrystalline cellulose, however, also are used, constituting the major component of solid dosage tableting. Cellulose is widely used for bandaging and wound repair materials, as a possible scaffold for tissue engineering, and as a severe burn repair material.[6]

Chitin is mainly used in a derivatized form as chitosan, in the biomedical industry, and as a dietary supplement material. Starch is mostly used in the food industry, although a major development in the area of bioplastics has been the production of polylactic acid from starch. Polylactic acid is derived via bacterial fermentation of corn starch or sugarcane. Global production of this polymer has recently been reported to exceed 250,000 tons per year.[7] Although initially very expensive, the price of polylactic acid is coming down. Particular examples of recent commercial ventures using polylactic acid have been the Cargill Natureworks' Ingeo bioplastic (sole U.S. manufacturer), although Toyota also produces the polymer in Japan, as does PURAC in the Netherlands. Polylactic acid–derived polymers promise applications in biopackaging, with degradation properties that are not deemed to be harmful to the environment.

Two forms of what can be termed "bacterial polymers" have recently gained prominence, mainly in research laboratories: bacterial cellulose and poly(hydroxyalkanoate)s, such as poly(hydroxybutyrate). Bacterial cellulose is produced under culture conditions by aerobic bacteria and has many potential applications, albeit currently in specialized markets such as speaker diaphragms and in the biomedical sector.[6,8] Applications using poly(hydroxyalkanoate)s are heavily restricted on the basis of price, although it is envisaged that the cost will decrease as the drive to use polymers from renewable sources increases.

Silk still has its traditional use in the textiles industry, but recent interest has focused on using it for tissue engineering applications.[9] Silk, in essence, is a renewable material, as proteins are essentially an abundant natural resource. Spiders recycle their silk, and given the aqueous processing of the material, it presents itself as a potentially "green" material. The volume production of silk is sufficient for the textile industry, but if structural applications are to be realized, then this needs to increase significantly. In order to do this, efforts have been made to produce artificial silks. Efforts to mimic silk fibers, however, have not been that successful. An example of one approach has been the seeding of human cells with proteins found in silk in order to produce filaments.[10] Most, if not all, attempts to produce artificial silk have failed, mostly due to the fact that this artificial material's properties did not match that of the naturally derived material. Nevertheless, much progress has been made on understanding structure-property relationships of silk in spinning and how processing could be the route for better silk fibers for engineering applications.[11] There are many challenges ahead for industry and the scientific community when designing technological applications that use materials from renewable resources. The next section addresses some of these challenges.

Challenges for the Use of Materials from Renewable Resources

It is almost certain that industrial use of renewable materials will only take place if there is an economically good reason to do so. The expected rise in oil prices is a strong economic driver and will continue to direct industrial policy into renewable technology. The technology used to generate conventional polymers from petroleum is less than 100 years old. Nevertheless, considerable progress has been made on the development of high-performance polymers, and therefore considerable investment in the production infrastructure (plants, knowledge base) has already taken place. One of the first challenges of the 21st century is to use non-petroleum feedstocks where traditionally petroleum has been used, and this may come at an additional cost to the manufacturer. New processes to control variability and mixed stream feedstocks from plant material will require additional investment from industry. A rapid learning process also will have to take place if these materials are to replace traditional resources. The use of a "biorefinery concept" is now required, as has been explicitly outlined recently,[12] where a continuous loop of biofuels and materials can be developed without depletion of natural resources. This also will have to come with the technological means to generate advanced materials.

Renewable materials have to match up to the properties of existing materials and add something in order to make them viable. In the first instance, the additional property will be their renewability, but it is vital to ensure that this is truly exploited. Some of the initial materials in this area have promise, but actual renewability must be addressed in order to meet our environmental targets. Additional challenges lie ahead in being able to exploit other properties of natural and renewable materials, such as a hierarchical structure. Controlling the inherent variability in natural and renewable feedstocks presents another challenge to industrialists and academia in the generation of a reliable product.

There are many technical challenges that we will face over the use of renewable materials, but this is not a complete picture. The challenge of using renewable feedstocks also brings nonscientific problems, including the social and economic consequences of the use of food crops for biofuels and plastics. Recent rises in the price of corn, maize, and other staple food stuffs, because of their potential use in biofuels (and therefore bioplastics), has been a cause for concern, which could be remediated through the use of nonfood crops. The challenge, therefore, is to use nonfood crops where possible.

Efforts to investigate the production of petroleum precursors and liquid fuels from biomass are reported in the literature,[13] and presumably a percentage of this could be returned to polymeric materials, using the same model that has worked for oil. The challenge is not to fall into the same trap where consumption and demand exceed supply, where the renewable aspects of the materials may be lost.

It has been recently recognized that technology solves problems through the use of "energy," whereas biology tends to use "information" and "structure."[14] This is true for a number of renewable materials when they remain in a biological cycle. The self-assembly of plant materials and proteins is less dependent on raw energy *per se* and more dependent on the use of entropy (order and disorder). Conversely, large amounts of energy are expended to extract oil, from which polymers can then be synthesized with a relatively lower energy expense. Minimal use of energy as a whole to generate renewable materials and chemical feedstocks needs to be addressed in order to avoid high costs. Some methods of extraction of biomass for natural materials use large amounts of energy, which is something that is largely ignored but will play a key role in decision-making when technological approaches are adopted. The question is, are there alternative methods for renewable materials production that are less energy intensive? Perhaps the use of biomimetic methods of materials production is a way forward, relying more on self-assembly, structure, and, in a wider context, information. Enzymatic and bacterial processes of materials production are two options that can, if carefully controlled, generate biomass in a more energy conservative way. The following section briefly introduces the five articles selected for this issue.

Content of This Issue

Each of the five articles reports on the use of renewable materials in technological applications. The articles highlight the renewability of the materials and the benefits in terms of properties that they bring to the particular application. At the same time, they represent the state-of-the art research that is taking place in this field and offer insight into how we might better utilize renewable materials.

The first article by Robertson et al. discusses the production and use of renewable block copolymers, which are hybrid macromolecules composed of two or more covalently connected segments. These fascinating self-assembled materials are used in applications ranging from footwear to bitumen modification to microelectronics. The number of technologies that utilize or could benefit from block copolymers is expanding at a rapid rate. This growth is due to the development of simple scalable synthetic technologies, a deeper understanding of their structure-property relationships, and their effectiveness as low-level additives. As industrial uses of block copolymer-based materials become more prevalent, there will be an increased focus on alternative preparative approaches that do not rely on petroleum feedstocks. Therefore, the development of biorenewable block copolymers is an important research endeavor. The article explores the synthesis, self-assembly, and properties of renewable block copolymers that contain aliphatic polyesters, as well as segmented polyurethanes from bio-sources. These two classes of block copolymers are the most promising and practical candidates for implementation in the next generation of sustainable materials.

The second article by Berglund and Peijs concerns the challenges and opportunities raised in using cellulose in biocomposite materials. Cellulose fibers are the key reinforcement phase in the composite structure of plant cell walls, which are present in the form of nanofibrils. These nanofibrils can be exploited in a variety of materials applications, and some of these are discussed. The problems associated with their extraction and interfacing with other materials also are covered. A vast range of sizes of fibers is available from micron-sized fibers to nanowhiskers and nanofibrils. These reinforcements can be arranged in the form of weaves, continuous fibers, and co-mingled matrix fibers, all of which are discussed in this article. Developments in modifying the fiber/matrix interface using novel grafting agents are discussed, as well as the relatively recent work on all-cellulose composites, which have the potential to be fully biodegradable and/or recyclable. In recent times, cellulose nanofibers have gained widespread interest. Nanofibrils of this type have been produced in a cost-effective manner using enzymatic or chemical pretreatment followed by mechanical disintegration from plant cells. The resulting nanocomposites show much-improved properties in terms of mechanical strength, Young's modulus, toughness, reduced thermal expansion, and optical transparency. All of these aspects are covered as well as theoretical modeling studies, which demonstrate the molecular level mechanisms for such observations. The properties of specific materials, such as aerogels, where gels of cellulose nanofibers are freeze-dried to form very low density networks, also are presented. These aerogels are characterized by mechanical robustness, low thermal conductivity, and high acoustic damping. The use of these materials as templates for inorganic hybrids, such as magnetic nanoparticle composites, also is discussed. By combining cellulose with starch foams, materials with properties similar to conventional polymers can be obtained. This means that foams can be generated from renewable materials (starch and cellulose), with properties compatible with the materials that are currently preferred by industry. The future for cellulose fibers deserves more than a simple blending with conventional oil-based thermoplastics, and this is demonstrated with specific recent developments in the area.

The third article by Gatenholm and Klemm looks at cellulose from a bacterial source for a new and exciting application. Nanofibers of bacterial cellulose are an emerging biomaterial with great potential for wound and burn dressings and for scaffolds for tissue regeneration. Bacterial cellulose has remarkable mechanical properties despite the fact that it is comprised almost entirely of water. The water-retaining capacity of bacterial cellulose is the most probable explanation for the fact that implants made of this material do not elicit any foreign body reactions. Moreover, the nanostructure and morphological resemblance with collagen makes bacterial cellulose attractive for cell immobilization and support. The architecture of bacterial cellulose can be engineered at different length scales, from micro to nano, which makes this material an ideal scaffold for tissue engineering. The fact that bacterial cellulose can be generated readily in the laboratory using benign processing makes the material infinitely renewable. This article covers all the recent developments in this area.

The fourth article by Nakagaito et al. reports on another use of cellulose nanofibers. Flexibility is an essential characteristic not only for future electronic devices such as flexible displays and solar cells, but also for materials suitable for forthcoming roll-to-roll production processes. These processes enable the continuous deposition of functional materials on a roll of optically transparent flexible plastic. However, most plastics have large coefficients of thermal expansion (CTE). Therefore the functional materials that are deposited on the plastic substrates could be broken or damaged by the temperatures involved in the assembly and mounting processes due to the mismatch of the CTEs. Reinforcement of transparent plastics with nano-sized cellulose fibers can overcome these difficulties, as reinforcing elements with diameters less than one-tenth of the wavelength of visible light are free from light scattering, enabling the formation of optically transparent composites. The advantages of nano-sized reinforcements have been experimentally demonstrated using bundles of cellulose nanofibrils. The resulting nanocomposites are not only highly transparent but also exhibit a low CTE (due to the low CTE of cellulose) comparable to that of glass; they also have mechanical strengths comparable to mild steel. It is also shown how an electroluminescent layer can be placed on these transparent cellulose nanocomposites, such that they can be used for display devices.

The final article by Paris et al. deals with biomimetics and biotemplating of natural materials. Natural materials have developed a wealth of structures to fulfill many functions. Hierarchical structuring is key to multifunctionality and allows adaptation to the varying needs of the organism. As a consequence, our natural environment represents not only a direct and renewable source of useful materials, such as wood, plant fibers, or even proteins of pharmaceutical importance, but also an enormous "database" of structures with exceptional mechanical, optical, and magnetic properties. Rather than using the natural materials directly, the approach discussed in this article extracts the structural principles encoded in them and implements these in the manufacture of artificial materials of entirely different types and chemical compositions. The required information may be extracted in two ways, either by direct copying or templating—for example using chemical or thermal transformation of the natural material—or by a virtual extraction of the materials concepts and their application to the fabrication of engineering materials. The first route often is described as biotemplating and the second as biomimetic materials research. This final article briefly describes the principles of these two approaches and gives some illustrative examples.

Each article gives a tutorial overview of an area of materials from renewable resources. To further support the very premise of this theme, the technologies are presented along with an explanation of how the materials are renewable and how they will contribute in a meaningful way to current and new applications. For instance, the use of plant-based materials (e.g., starch, furans) to generate a wide range of polymers, in particular block co-polymers, is given. It is also shown how cellulose, derived from plants, can be used in a number of technological applications.

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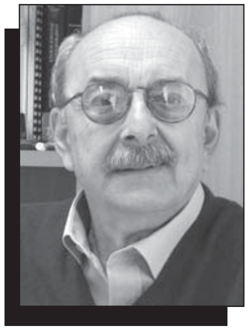

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