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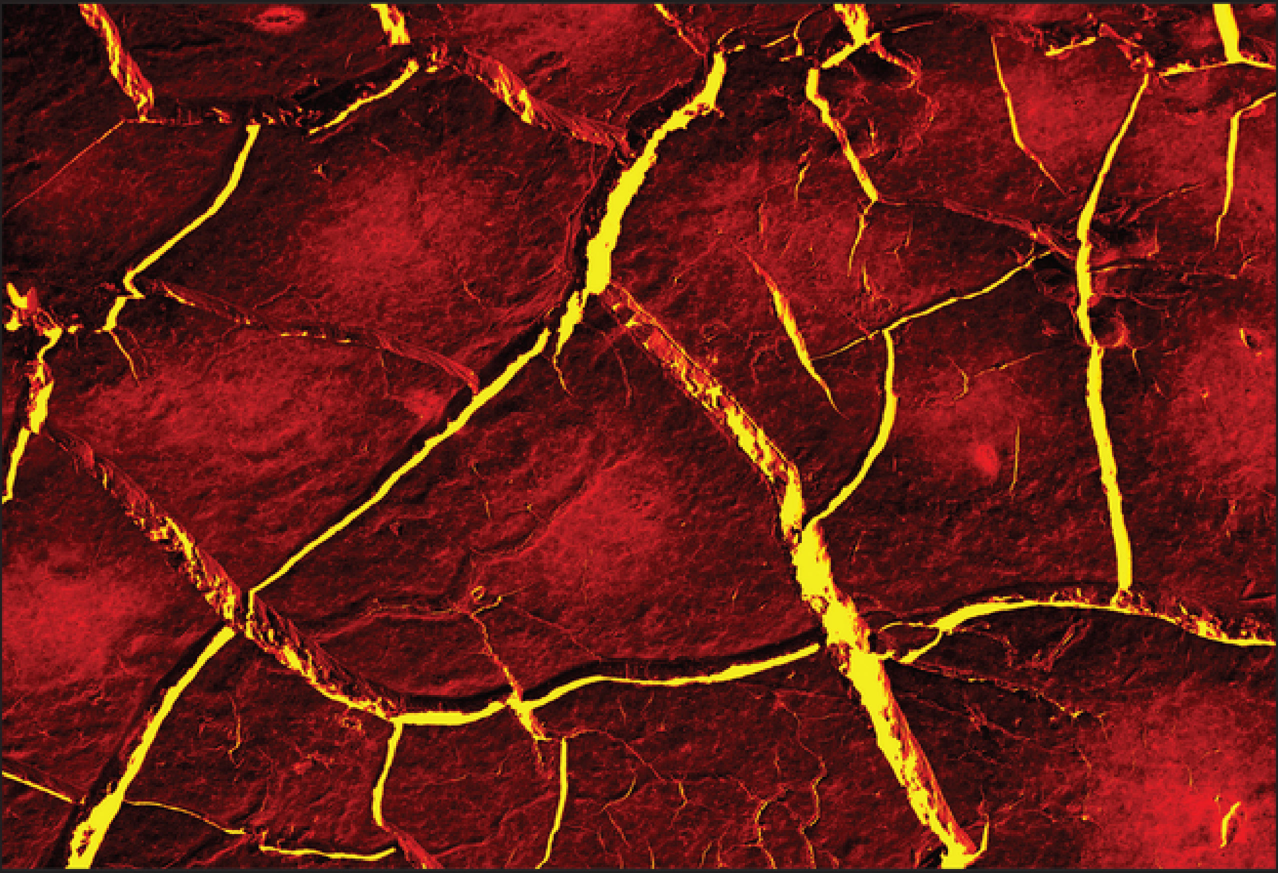
BIO MONOMERS FOR GREEN POLYMERIC COMPOSITES MATERIALS



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Preface

This book summarizes many of the recent research accomplishments in the area of biomonomers for green polymeric composites and their nanocomposites. It includes an introduction to biomonomers for green polymers, the current status of these compounds, new challenges, opportunities, and processing methods for bionanocomposites, biopolymeric material-based blends, the preparation, characterization, and applications of biomonomers and their nanocomposites, applications of biopolymeric gels in medical biotechnology, an introduction to green polymeric membranes, properties and applications of gelatins, pectins, and carrageenans gels, the biodegradation of green polymeric composite materials, applications of green polymeric composites materials, hydrogels used for biomedical applications, and natural aerogels as thermal insulations.

This book is a valuable reference source for university and college faculties, professionals, post-doctoral research fellows, senior graduate students, and researchers from R&D laboratories working in the area of biomonomers and green polymeric composites materials. The chapters are contributed by prominent researchers from industry, academia, and government/private research laboratories across the globe and present an up-to-date record of the major findings and observations in the field of biomonomers and green polymeric composite materials.

The first chapter discusses the state of art and new challenges of biomonomers and green polymeric composite materials.

The second chapter covers several topics, including classification of nanobiocomposites, general processing methods, and properties. The author also includes many subtopics, such as polysaccharide nanocomposites, animal protein-based nanocomposites, plant protein-based nanocomposites, metal nanocomposites, and inorganic nanocomposites. General processing methods such as pressure extrusion, solid-state shear pulverization, electrospinning, solution casting, evaporation, melt intercalation, *in situ* polymerization, drying techniques and polymer grafting are also discussed in this chapter.

Various topics on biopolymeric material-based blends, such as preparation, characterization, and applications, are addressed in Chapter 3. This chapter presents a comprehensive study of biopolymeric material-based blends, including general preparative methods, aqueous blending technology, their hydrophilic or hydrophobic nature, degradation problems, thermodynamics of miscibility, and their opportunities or challenges. Biopolymeric blends have attracted academic, research, and industrial scientists' attention as their properties are desirable for various applications. The challenges due to the unique structure, preparative methods, and resultant properties of blends of natural polymeric materials are discussed, and examples are drawn from the scientific literature. The various forms of natural polymers, i.e. polysaccharides, proteins, lipids, natural rubber, chitosan, starch, and silk-based blends, are reviewed with respect to preparative techniques, characterization methods, and various applications.

The fourth chapter of this book discusses the applications of biopolymeric gels in medical biotechnology. The primary objective of this chapter is to review the literature regarding the classification of the properties of hydrogels and their biomedical applications. The composition and structure of hydrogels, especially their use in biological fields, makes them ideal candidates for biopharmaceutical implementation. Innovations in recent manufacturing and world-wide resources of hydrogels are also reported.

The fifth chapter introduces green polymeric membranes, covering types of green polymeric membranes, their physicochemical properties, and their potential applications. The application of these materials in various industries was facilitated by their tremendous and significant physicochemical properties. However, despite these advances, there are still some drawbacks which prevent the wider commercialization of green polymeric membranes in many applications. This chapter reviews the current trend of research involving green polymeric membranes that focuses on the fabrication method, processing, and surface and structure modification. In addition, the long-term stability and durability of green polymeric membranes for specific applications has become a challenge to researchers all around the world. The introduction of nanostructure fillers (e.g. graphene oxide, metal oxides, carbon nanotubes, nano-clay, etc.) and the blending with other polymers, or the making of new copolymers, has significantly improved their overall properties and performance. These improvements are generally attained at low filler content, and this nano-reinforcement is a very attractive route to generate new functional green polymeric membranes for various applications. It should be noted that the development of green polymeric membranes with specific physicochemical properties for specific functionalities is crucial for practical applications in industry. Green polymeric membranes with various physicochemical properties have a promising contribution to make in various applications.

Chapter 6 discusses the properties and applications of gelatins, pectins, and carrageenans gels. For each of these substances the authors cover various subtopics, such as structural units, molecular structure, properties, thickening ability, gelling ability, film-forming properties, microbiological properties, food applications, cosmetics applications, and pharmaceutical applications. In the chapter on biodegradation of green polymeric composite materials, the authors consider a wide range of review studies on this subject, including biomechanical pathways for the degradation of green polymers and green polymer composites. Several studies have been carried out to design polymers with biodegradable properties to help keep the environment safe and clean.

In Chapter 8, on applications of green polymeric composites materials, the authors discuss several different topics, including a series of interesting green polymer composites developed from thermoplastic starch and its blends, poly(lactic acids) and its modifications, cellulose, gelatin, and chitosan. The authors also describe how natural fibers have more environmentally friendly properties than synthetic fibers synthesized from agricultural sources such as jute, banana, bamboo, and coconut coir, etc. There is thus a wide range of possible applications of nanocomposites from agriculture to automobiles. However, problems of poor adhesion of matrix and fiber, difficulty with fiber orientation, achieving nanoscale sizes, and the evolution of truly green polymers that are environmentally friendly and renewable must first be solved.

In Chapter 9, constituents, fabrication, crosslinking and clinical applications of hydrogels are described. Hydrogels are extensively found in everyday products although their potential has so far not been thoroughly investigated. The authors review the fabrication and composition of hydrogels along with their different properties, and the natural and synthetic polymers used for the development of hydrogels in the presence of different crosslinking agents. The major characteristics of hydrogels related to clinical, pharmaceutical, and biomedical applications are also identified, particularly for applications of hydrogels in contact lenses, oral drug delivery, wound healing, tissue engineering matrices, and gene delivery.

The final chapter examines the use of natural aerogels as thermal insulation and in other applications. A further review of natural aerogel-based composites and nanocomposites is also provided.

The editors would like to express their sincere gratitude to all the contributors to this book, whose excellent support led to the successful completion of this venture. We are grateful to them for the commitment and the sincerity they have shown toward their contribution to the book. Without their enthusiasm and support, the compilation of this book would have not been possible. We would like to thank all the reviewers who have given their valuable time to make critical comments on each chapter. We also thank Wiley for recognizing the demand for such a book, and for realizing the increasing importance of the area of bio monomers for green polymeric composites materials and supporting this project.

Tomsk, Russia, March 2019

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1

Biomonomers for Green Polymers: Introduction

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1.1 Processing Methods for Bionanocomposites

The new generation of hybrid nanostructured materials has two crucial properties: biocompatibility and biodegradability [1, 2]. Exploitation of various biopolymers such as proteins, nucleic acids, polysaccharides, etc. for preparation of nanocomposites has been done in last few decades [3]. Processing methods for matrix and filler are sometimes the same. However, some matrices are prepared using combinations of techniques to achieve the desired quality of bionanocomposites, therefore we will discuss the processing methods for bionanocomposites with suitable examples. Bionanocomposites of polysaccharide matrices are mainly prepared by solvent intercalation or melt processing and not through in situ polymerization where nature of the polysaccharide directly influences the route of preparation. Some polysaccharides with nanostructure fillers are discussed as examples. Most of the cellulose whiskers-reinforced poly(lactic acid) (PLA) nanocomposites are prepared by melt extrusion to avoid agglomeration and aggregation during drying [4]. Porous networks and thickened cellulose ribbons in gelatin/nanocellulose composites are prepared using an enzymatically modified form of gelatin [5]. Cellulose nanocomposites based on nanoparticles, such as clay [6–14], carbon nanotubes (CNTs) [15], graphene, layered double hydroxide (LDH) [16], and silica [17] have been prepared.

Starch is another abundant, inexpensive, naturally renewable and biodegradable polysaccharide, produced by most green plants as an energy store. It is the most common carbohydrate in human diets and animal feeds. Starch nanocomposites are mixtures of starch-based biopolymers with nanofillers (solid layered clays, synthetic polymer nanofibers, cellulose nanowhiskers, CNTs, and other metal nanostructures). Environmentally friendly starch nanocomposites exhibit significant improvements in mechanical properties, dimensional stability, transparency, improved processability, and solvent or gas resistance. Chitosan (CS)/chitin, the second most abundant natural biopolymer, also can be integrated with clay, graphene, and carbon nanostructures to prepare bionanocomposites [18–21]. Due to its high content of amino ($-NH_2$) and hydroxyl ($-OH$) groups, chitosan and its derivatives are excellent adsorbents for the removal of heavy metal ions, fluoride, and organic dyes. Films of spin-coated chitosan–alginate nanocomposite have potential uses in bioapplications. Lignin-based

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nanocomposite films have been prepared using CNCs (carbon nanocomposites) and used in various applications such as medical, biological, optical and sensors, and electronic [22]. They are also used as adhesives, stabilizing agents, and precursors for many aromatic chemicals. Modified lignins, such as lignosulfates, kraft lignin, and acetylated lignin, contain CNCs or commercial derivatives or nanocellulosic polysaccharides. Polyethylene terephthalate (PET) film coated with graphene oxide (GO)/pullulan nanocomposite can be used in food/pharmaceutical applications [23]. Bionanocomposites with enriched properties based on two microbial polysaccharides, pullulan and bacterial cellulose (BC), were prepared by Trovatti et al. for possible application in organic electronics, dry food packaging and the biomedical field [24]. Pullulan composites with many materials, including chitosan [25], caseinate [26], starch nanocrystals [27], collagen [28], poly (vinyl alcohol) [29], and hydrogel with methacrylate [30], have excellent compatibility.

Their biodegradability, low cost, and surfaced modification with active functional groups for catching targeting molecules make these matrices feasible candidates for applications in the pharmaceutical industry [31]. Electrospun collagen-chitosan nanofibers were stabilized by glutaraldehyde vapor via crosslinking, which afforded a biomimetic extracellular matrix (ECM) for cell growth [32]. Collagen is regarded as one of the most useful biomaterials, exhibiting a number of biological advantages. The outstanding performance and biomedical application of this protein biomaterial have induced researcher interests in synthetic composite material fabrication. Soy protein isolate (SPI) has been extensively studied for bioderived packaging materials. Several recent studies have investigated the improvement of mechanical and barrier properties of nanocomposite films after incorporating nanoclays such as montmorillonite (MMT) [33–41]. Further, these nanocomposite films have also been reported for decreased water vapor and oxygen permeability, and increased elastic modulus and tensile strength, which makes them suitable for packaging industry. Recent studies have also reported that the SPI-based nanocomposite bioplastics with highly exfoliated MMT have significantly improved mechanical strength and thermal stability [42]. Thus, bio-based polycaprolactone–SPI is not only ecofriendly but intercalated nanocomposites with enhanced tensile and dynamic mechanical properties when produced by the melt compounding method [43].

In the case of biocomposites, the properties of the composites produced are dependent on the inter-phase interaction of the reinforced material and matrix. Filler is also a value-added material, but wise selection of processing methodology, optimum conditions, and compatible phase components is needed. Polymer/metal nanocomposites consisting of polymer as matrix and metal nanoparticles as nanofiller commonly exhibit several attractive advantages, such as electrical, mechanical, and optical characteristics [44]. Metal nanocomposites with protein, nucleic acid, and polysaccharides have shown potential applications in drug delivery, tissue engineering, bioimaging, wound healing, biomedicine, energy production and storage, and electronic devices such as biosensors, affinity materials, etc. [45]. Bottom-up methods are found to be promising for controlling the properties and specific orientation of nanomaterials. Thermal evaporation and sputtering techniques have been considered as facile, simple, low-cost, and high-yield methods for synthesis of high-quality nanomaterials/nanostructures [46, 47]. Various immobilization methods, including entrapment, adsorption, crosslinking, electro-polymerization, and encapsulation, have been used for capturing biological moieties in the matrix. This is one of the main processes employed in the manufacturing of nanobiocomposites (NBCs) [48]. There are two main types of extrusion: reactive extrusion and extrusion cooking. Reactive extrusion uses chemical modification via crosslinking [49]. Generally, extrusion technology used in the food industry is referred to as extrusion cooking and results in different physical and chemical properties of the extrudates depending on the raw materials and extrusion conditions used [50]. Various starch nanocomposite varieties have been prepared and reported by many researchers for biodegradable packaging applications in food industry. Moigne et al. developed a continuous CO_2 assisted extrusion process to prepare poly(3-hydroxybutyrate-co-3-hydroxyvalerate)/clays NBC foams with better homogeneity and high porosity [51]. Inventor Torkelson has successfully produced a well-dispersed graphite–polymer nanocomposite [52]. Taking advantage of near-ambient-temperature processing, solid-state shear pulverization (SSSP) was recently used to produce biodegradable polymer matrix composites with starch [53], rice husk ash [54], and eggshell filler [55, 56]. This technique that has proven to effectively disperse nanoscale structural entities to achieve compatibilized polymer blends and exfoliated polymer nanocomposites.

This physical method uses extrusion of the polymer solution with reinforcement of nanomaterials and biological entity for the preparation of NBCs. Polymers, molten at high temperature, can also be made into nanofibers by electrically charging a suspended droplet of polymer melt or solution [57–63]. Instead of a solution, the polymer melt is introduced into the capillary tube. The major difference is that a compound spinneret with two (or more) components can be fed through different coaxial capillary channels [64]. Wet-dry electrospinning and wet-wet electrospinning techniques are used for volatile and non-volatile solvents respectively. Both techniques offer the possibility of producing nanofibers with controlled fiber diameter to make film or membrane or an oriental controlled fiber. Such fibrous scaffolds are ideal for the purpose of tissue regeneration because their dimensions are similar to the components of the extracellular matrix and mimic its fibrillar structure, providing essential signals for cellular assembly and proliferation. Core–shell structured nanofibers where collagen as the shell and poly(ε-caprolactone) (PCL) as core were prepared by co-axial electrospinning and show the advantage of controlled shell thickness and manipulative mechanical strength and degradation properties of the resulting composite nanofibers, without affecting biocompatibility [65]. Thus, such core–shell structured composite nanofibers have potential uses in drug or growth factor encapsulation and the development of highly sensitive sensors and tissue engineering applications [66].

The solution casting method is based on a solvent system in which the polymer or pre-polymer is soluble. The polymer is usually dissolved in a suitable solvent while the nanoparticles are dispersed in the same or a different solvent before the two are mixed. For example, during the preparation of bionanocomposites based on clays, the solvent is normally used to pre-swell the clays [67]. In case of non-water-soluble polymers, additional steps are required before processing. Solvent exchange, use of surfactant, freeze-drying, and chemical modification can be used for this purpose. Melt intercalation is a mechano-chemical process that is highly preferred in case of clay/silicate biocomposites, and this processing method is compatible with extrusion and injection molding. This method is also fast and clean, ecofriendly, and can alter the lifecycle analysis [68]. In this technique nanomaterials and/or biomaterials are mixed with the polymer in the molten state. The process involves mixing the particles with the polymer and heating the mixture above the softening point of the polymer, statically or under shear.

Like other techniques, proper dispersion of the nanoparticles is always a goal during processing. During melt processing, a number of factors are important to achieve homogenous dispersion of reinforced nanomaterial into the polymer matrix, including enthalpic interaction between the polymer matrix, biocomponent, and nanoparticle. Multi-walled CNT-poly methyl methacrylate (MWNT/PMMA) nanocomposite has been prepared by in situ polymerization of MMA dispersed with MWNTs with fairly good dispersion stability [69]. Grafting of long chains can also be used to transform the nanofiller into a co-continuous material by increasing apolar character through grafting agents bearing a reactive end group and a long "compatibilizing" tail [70].

1.2 Biopolymeric Material-based Blends: Preparation, Characterization, and Applications

Preparation of biopolymeric blends is encouraged due to their biocompatible and biodegradable properties [71, 72]. The reason for the increased research in the preparation of versatile biopolymer blends is their broad application, e.g. biomedical applications [73]. The biopolymer blends in this regard have emerged as promising materials with suitable thermal, biocompatible, and mechanical properties for use in the intended applications [74, 75]. The main biopolymers used in the preparation of blends for various applications include collagen, chitin, chitosan, keratin, silk, and elastin, all natural polymers derived from animals [76, 77]. Property of biopolymers is useful in blends formation with the other soluble polymers. The polymers with little solvent affinity, e.g. elastin, silk, or keratin, have a problem during blend formation [78, 79]. The environmentally friendly nature of the biopolymers due to their biodegradable properties is also advantageous [80]. It is also notable that polymers have dominant hydrophobic properties, degrade after use, have mechanical properties, and behavior toward aqueous environments [81–84]. Biopolymeric blends are non-biodegradable in nature [85] and therefore these do not have the natural advantage of environmental friendly properties.

The melt process in making biopolymeric blends is advantageous to overcome various shortcomings associated with the basic physicochemical properties of biopolymers. The new techniques were developed to make biopolymeric blends, e.g. reaction extrusion technology to make starch–cellulose–acetate blends. In this technology, a number of materials were used during the blend formation process. The biodegradable hydrogels were prepared by biopolymeric blends formed by the combination of starch derived from corn starch and cellulose acetate. The blending reaction proceeds through free radical mechanization after reaction between methacrylate or acrylic acid monomers. The free radical reaction was initiated through a redox system consisting of 4-dimethylaminobenzyl alcohols and benzyl peroxide at ambient temperature. The addition of hydroxyapatite content in these blends provided a biocompatible character in the blended materials along with osteoconductive or oleophilic properties. The effect of polyol on the thermal stability, mechanical strength, and water or gas permeability was monitored to establish the usability of these blends. The study findings suggest that the polyol or water contents have a positive impact on the mechanical properties or stability of the biopolymeric blends [85]. The use of acetic acid promoted the formation of sheet-like structures in the blended materials. The characterization results of the biopolymeric blends showed improved thermal stability and degradation stability. In another study, Lazaridou and Biliaderis studied the effect of blended materials on thermal and mechanical properties [86].

Blends formed by the injection molding method show higher tensile strengths, lower water absorption/adsorption, and longer elongation values. The impacts of the chemical or physical properties of the starch on the biopolymeric blend properties were studied by Park and Im [87]. The starch was gelatinized by the addition of a mixture of water and glycerol in a twin-screw mixer. The microstructural properties of the starch blends with PLA, polyethylene, and vernonia oil were prepared by the melt processing technique and acid hydrolysis. Surface fractures were observed during scanning electron microscopic (SEM) examination of the blends. Polyvinyl alcohols (PVOHs) were also used as compatibilizers in PLA-starch-based biopolymeric blends [88]. This study reported that the use of PVOHs enables preparation of blends with better compatibility and improved mechanical strengths.

1.3 Applications of Biopolymeric Gels in Medical Biotechnology

Based on natural and synthetic polymers, hydrogels can be utilized in research into cell encapsulation and in particular have facilitated the establishment of a novel field in tissue engineering. Hydrogels are a significant class of biomaterials in medical biotechnology. These biomaterials categories may include tissue engineering and regeneration, diagnostics, cellular immobilization, cellular biomolecular separation, and utilization of barrier materials for biological adhesion for regulation. Hydrogels are invaluable in their three-dimensional network capacity to capture and release active compounds and biomolecules. Hydrogels have the sponge-like capacity to absorb water due to their hydrophilic functional groups. The water absorbed into hydrogels permits diffusion of certain molecules while the polymer component of the hydrogel acts as a matrix for holding water molecules together. Hydrogels have the ability to imitate the physical, chemical, biological, and electrical features of many tissues. These features earn them the ability to serve as potential candidates for biomaterials. These novel approaches involve super porous hydrogels [89], comb-like grafted hydrogels [90–92], self-assembling hydrogels [93, 94], and recombinant triblock copolymers [95–97]. To construct hydrogel systems with well defined chemical characteristics, information regarding polymer chemistry and synthesis, features of the materials to be utilized, parameters of mode of interaction, material release capability, and delivery systems need to be taken in to consideration. In order to construct hydrogel systems with well-defined chemical characteristics, information regarding polymer chemistry and synthesis, the features of the materials to be utilized, the parameters of the mode of interaction, material release capability and delivery systems need to be taken into consideration. Crosslinking is the most versatile method to facilitate biopolymeric deficiencies [98]. Mechanical properties and the stability of biomaterials can be ameliorated by crosslinking agents. However, limitations exist due to reduced degradability and lack of functional groups and potential cytotoxicity introduced by crosslinking agents [99, 100]. The chemical and physical nature of hydrogels depends on the concentration