

Recent Progress in Materials

Review

Review on Some Important Research Progresses in Biodegradable Plastics/Polymers

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Abstract

Biodegradable plastics/polymers may serve as a promising solution to the global problem of plastic waste accumulation in oceans and soil and may significantly reduce carbon emissions from the manufacturing process, since the materials used to make biodegradable polymers are carbon-based and emitted during the synthesis processes. This article systematically reviewed the existing and closely related scientific literature on materials, biomaterials, and biodegradable materials to find answers on how to effectively study and develop biodegradable polymers. This article reviewed and summarized the source classifications of the biodegradable plastics. Some of the major manufacturing techniques for making biodegradable polymer products were discussed, including micro-extrusion for biofibers, solvent casting method for thin films, 3D printing, injection and compression molding and extrusion processes, as well as the fabrication methods applied to some important biopolymers, such as cellulose, starch, bacterial concrete, packaging materials, and paper-based biodegradable materials. More importantly, experimental and computational methodologies applied for materials characterization and development that can be adopted



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to characterize the properties of biodegradable polymers and understand the physicochemical mechanisms of the materials were described in detail, including experimental methods (physical and chemical methods) and computational methods at different scales (from quantum mechanics at subatomic scale, molecular dynamics at atomic scale, to finite element analysis at micro or macro scale), and data analysis methods. The degradation mechanisms and factors affecting the biodegradability of the polymers were discussed. Finally, the future perspective of biodegradable polymers has been described. Properly adopting the effective state-of-the-art biomaterial research and characterization techniques (experimental and computational methods) and advanced data analysis methods discussed in this article will help advance the development of novel biodegradable polymers.

Keywords

Biodegradable plastics/polymers; sustainable; biomass; experimental characterization; numerical characterization; data analysis; biofibers; cellulose; starch; bacterial concrete; paper-based material

1. Introduction

Plastics or polymers are vital materials in our modern life. Due to their durability (resistance to physicochemical and biological degradation) and affordability (less cost), society mainly relies on them and their widespread consumption worldwide [1]. The most common uses of plastics are in packaging, construction, and transportation industries. In the medical industry, plastics are critical to contamination and infection control, such as syringes, pipettes, and gloves, but they are not reusable. While the overuse of plastic packaging is concerning, some forms of packaging are often required to preserve food hygiene or freshness or maintain product integrity during transportation. Small or travel-size toiletries and personal hygiene items are often considered wasteful but are critical to providing affordable hygiene options to some of our most vulnerable communities [2]. The two main factors that make plastics so widely used are their durability and cheapness, allowing them to be used in various applications. Unfortunately, the post-production of plastics is poorly managed. Plastics has become a major problem exacerbating its impact on the environment. Inefficient and ineffective waste management and intentional littering result in large amounts of plastic waste disposed into soil or floating in the oceans, causing increasing damage to the ecosystems. Annual global plastic production increased from 1.5 million tons in 1950 to 390.7 million tons in 2021 [3]. The current global daily production of disposable PPE (facemasks) is estimated at 1.6 million tons, which means approximately 3.4 billion disposable facemasks or face shields are being discarded daily due to the COVID-19 pandemic. The major countries that generate plastic waste are China, India, the United States, and Brazil [4]. However, the failure of these materials to degrade has become one of the biggest threats to the environment, human community, and health. The overuse of plastics by the evolving human society has resulted in various current or potential environmental and health issues [5-7]. Replacing plastics with other materials is neither simple nor straightforward, mainly because finding a replacement that combines all of the most desirable properties of plastics is a challenge. Biodegradable alternatives, such as specialty plastics,

paper, or cardboard, may have a higher greenhouse footprint due to the amount of water, electricity, or other resources consumed in their manufacturing [2]. This requires finding an alternative to the ubiquitous plastics, and bioplastics have emerged as a promising alternative. "Biodegradable plastics/polymers (bio-DPs)" are plastics/polymers derived from renewable biomass that are bio-based such as starch, cellulose, collagen, polylactic acid, and polyester-amides [8]. The use of bioplastics can reduce the dependency of limited quantities of fossil fuels. Unlike petroleum-based plastics, bio-DPs emit fewer greenhouse gases [9].

The term bioplastics or biopolymers does not necessarily mean that they are biodegradable or more environmentally friendly. Some bioplastics are not biodegradable or may take a long time to be degraded. Bio-polyethylene (bio-PE), bio-polypropylene (bio-PP), bio-polyethylene-terephthalate (bio-PET), and bio-polyamide (bio-PA) are typical non-biodegradable bioplastics [10]. Bio-DPs are defined as plastics/polymers that can degrade into carbon dioxide, water, methane, and other low-molecular-weight compounds [11]. However, when considering bioplastics instead of regular plastics, biodegradability is the most important consideration. Bioplastics from different sources exhibit various biodegradation mechanisms and rates, depending on many factors, such as material' composition and structure, environmental factors, humidity, and the presence of microorganisms. Several new technologies for making bioplastics through biomanufacturing have emerged in recent years.

There is growing interest in producing bio-DPs, which can be easily disposed of after the end of their use without causing harm to the environment. However, there are many factors that affect the changes in the chemical composition, properties, and structural aspects of producing bio-DPs. Recent advances in materials research and development based on experimental (physical and chemical) methods and computational methods in different scales (from subatomic, atomic, to continuum scales) offer great opportunities and possibilities for researchers and engineers to effectively research and develop bio-DPs.

This article aims to provide a comprehensive review of the common sources and classifications of the bio-DPs, the advanced technologies applied to make bioplastics, and to provide an in-depth understanding of the state-of-the-art characterization techniques can be adopted in the research and development of novel bio-DPs, specifically, the experimental methods (physical and chemical methods) and computational methods at various scales (from quantum mechanics at subatomic scale, molecular dynamics at atomic scale, to finite element analysis at micro or macro scale), and data analysis methods. The degradation mechanisms and some of the main factors affecting degradability of the bio-DPs will be discussed. In addition, the future perspective of bio-DPs will be summarized.

2. Sources and Classifications of Biodegradable Plastics

Strategic materials play an important role in many fields, including electronics, renewable energy systems, defense equipment, material recycling, CO₂ capture and utilization, sustainable biomaterials, agricultural production, etc. [12]. Today, multiple industries use many bio-DPs for a variety of applications. When discussing bio-DPs, there are many bio-based polymers that can be discussed that have a variety of unique attributes and applications. Bio-DPs include a broad and growing class of biomaterial-based plastics that have their own unique properties and applications. This list is constantly expanding as new inventions emerge in the field. Bio-DPs can be classified

differently according to different scales [7, 13, 14]. Based on their origin, the three main types of bio-DPs are traditionally divided into natural polymers, such as proteins or polysaccharides; biologically derived monomers used in synthetic polymers, such as polylactic acid; and biopolymers derived from microbial fermentation [15-17]. Speaking of microbial fermentation of biopolymers, since we have unlimited atmospheric nitrogen (N₂) and solar energy resources around us, research on bio-solar production of N-rich biopolymers through genetically engineered N₂-fixing cyanobacteria is beginning. For example, cyanobacteria use carbon dioxide (CO₂) to produce biomass through photosynthesis, as well as directly producing carbon- and nitrogen-based compounds that store solar energy. Therefore, the uniquely solar-powered N₂-fixation of cyanobacteria offers greater prospects to produce N-rich bioproducts than all other diazotrophic bacteria that cannot use solar energy and require anaerobic conditions unfavorable for to their practical applications. The classifications of biopolymers according to their origins are shown in Figure 1 [18].

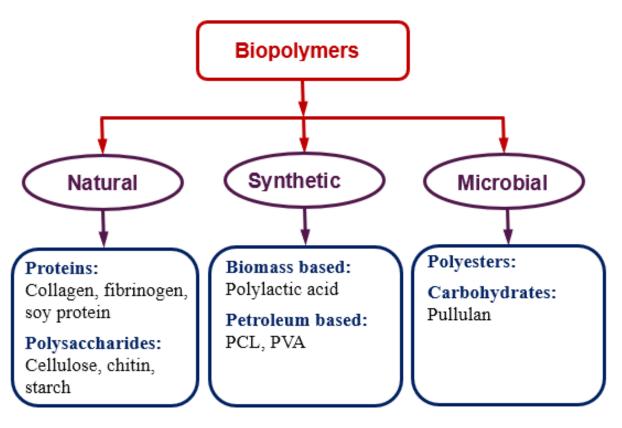


Figure 1 Classifications of biopolymers based on their origins.

Degradability can be one of the indicators to classify plastics. In fact, mainly two different classes can be defined, such as biodegradable and non-biodegradable polymers and alternatively, biobased and non-bio-based polymers. Furthermore, they can be classified into polymer backbones, so these groups can be found in, for example, polyesters, polysaccharides, polycarbonates, polyamides, and vinyl polymers. Depending on the types of monomers, three groups can be found, such as polysaccharide, protein, and nucleic acid. Another way to classify plastics based on their response to heat is elastomers, thermoplastics, and thermosets. These are different categories for classifying plastics, and each group can be classified into several subgroups, as shown in Figure 2 [18]. Recent Progress in Materials 2024; 6(2), doi:10.21926/rpm.2402015

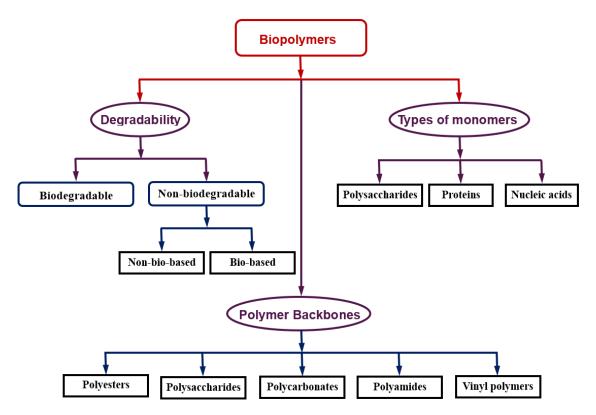


Figure 2 Classifications of biopolymers based on their biodegradability, types of monomers and polymer backbones.

However, the most common bio-based plastics can be divided into five categories:

2.1 Starch-based

This type of biopolymer uses corn starch, potato starch, etc., as raw materials, and is often blended with biodegradable polyesters to enhance the performance of the biopolymers. Starch is the most widely used raw material for making biopolymers. It is easy to make and can be a do-ityourself (DIY) experiment to try in your home kitchen. Starch-based biopolymers are used in the medical industry to produce capsules. Furthermore, they are often blended with other biopolyesters to create high-strength biopolymers that can be used in commercial applications. Starchbased biopolymers are also considered cost-effective because the raw material, i. e., starch, is cheap and abundant.

2.2 Cellulose-based

These biopolymers are produced using cellulose esters and cellulose derivatives. Cellulose is also added to the starch to create a biopolymer with enhanced mechanical properties that is highly water-resistant.

2.3 Protein-based

These biopolymers are produced using various protein sources such as wheat gluten, soy protein, casein, and milk.

2.4 Aliphatic Polyesters

A variety of bio-based polyester types exist, collectively known as Aliphatic polyesters, including: PHB (poly-3-hydroxybutyrate), PHA (polyhydroxyalkanoates), PHV (polyhydroxy valerate), PHH (polyhydroxy hexanoate), PLA (polylactic acid), and PA11 (polyamide 11), and all of them are susceptible to hydrolytic degradation and can be blended with other compounds.

2.5 Organic Polyethylene

Also known as bio-derived polyethylene (PE), they are typically produced through the fermentation of agricultural feedstocks such as sugar cane and corn rather than fossil fuels. Ethylene, the monomer used to make PE, can be extracted using ethanol and other agricultural fermented waste. The resulting bioplastics are chemically and physically similar to conventional PE.

3. Technologies to Make Biodegradable Plastics Products

The growing interest in and manufacture of plastics over the past few decades has endangered many human lives and pristine environments due to the accumulation and continued contamination of small, degraded small plastic particles known as microplastics. Plastic recycling, composing, incineration, and sanitary landfilling therefore provide answers to this troubling issue, but each comes with huge barriers and uncontrolled consequences, as well as a lack of environmental sustainability. Therefore, bio-DPs from renewable raw materials represent the best opportunity to achieve the Sustainable Development Goals universally launched by the United Nations in 2015 [19]. Bio-DPs are formed by the fermentation of sugar or canola oil to produce PLA or PHA, which are then converted to bio-DPs. The market can be segmented based on material, end-use, and regional prospects.

The functionality of a bio-DP in addition to its structure and composition, depends on several factors, such as the type and quantity of the solvent used, as well as the processing conditions and techniques used to fabricate the final product, which will determine the interactions of the materials. The selected processing techniques for producing bio-DPs from renewable sources are discussed below [20].

3.1 Biofiber Production Methods

Bioplastics are long-chain molecules made of repeating chemical blocks (monomers) originated from the renewable resources that can decay in the environment. Natural protein fibers, such as collagen fibers, hair, and silk, play an important role in different stages of the life cycle by supporting, stabilizing, and protecting cells, tissues, and organisms [21]. Interestingly, these types of protein fibers share a common structure with smaller fibril bundles along their long axis [22-26]. In the case of silk, a monofilament fiber is a bundle of fine fibrils arranged along its long axis, and the width of the fibrils ranges from nanometers to sub-micrometers [26-28], as shown in Figure 3. Adjacent fibrils have relatively weak interactions with each other, so they can be separated from silk fibers by simple processing [29]. Single fibrils can be thought of as domains of supramolecular structures/networks. The in vitro reproduction of the hierarchical fibrillar structure of natural fibers, such as silk, helps understand the in vivo assembly process and gaining insight into the design and development of advanced artificial biomaterials in the form of fibrils [30, 31].

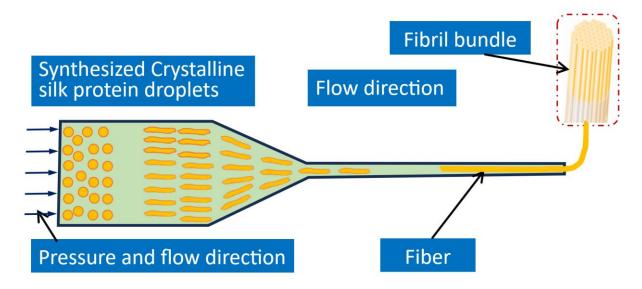


Figure 3 Schematic diagram of silk-like protein molecule droplets forming suprafibrillar fibers through micro-extrusion.

3.2 Solvent Casting Methods

Solvent casting method, also known as solution functionalization or wet processing, deals with producing aqueous or hydroalcoholic mixtures with biopolymers. This method uses solvents to suspend the polymer in the film-forming solution, then to evaporate the solvent and reform the polymer chains [32-34]. Alcohol, water, or other organic solvents are often used to dissolve polymers. Sometimes, the suspended polymer solution is heated, or the pH adjusted for best results. Then ,the mixture is poured into a mold, drum, or flat surface and allowed to dry there for a certain amount of time. Once the solvent has completely evaporated, a polymer matrix is formed and can be peeled off the model. Figure 4 shows the schematic diagram of the solvent casting process.

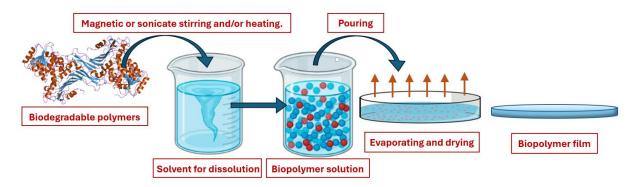


Figure 4 Schematic diagram of solvent casting process.

Casting is a simple method. However, there are several requirements that need to be considered. The most important point is to properly choose the right solvent that can properly dissolve the polymer. If the attractions between polymer molecules in solution is weaker than the attractions between polymer molecules and the solvent, the chain segments will stretch by diffusion of solvent molecules. This results in the polymer matrix being swollen by the solvent. However, it should be noted that the solubility capacity of polymers varies depending on the solvent [34].

The molecular weight of the polymer is another important point in the process. The molecular weight of the polymer affects the rate of solvent penetration. Higher molecular weight polymers dissolve more slowly than lower molecular weight polymers, and therefore, need greater permeability of the solvents. This is due to the slower rate of relaxation of the high molecular weight polymer chains as it has greater entanglement in the chains [35, 36]. Moreover, this biopolymer has sufficient cohesive strength and coalescing ability. Another important requirement is that the polymer must be dissolved in a volatile solvent or water. For best results, stable solutions with proper viscosity should be generated [34]. Environmental factors, such as temperature and humidity, are also important to the correct development of the process [37].

The major advantages of the casting method are that it is easy to develop and does not require special and expensive equipment. Since casting is a wet process, there is better inter-particle contact, resulting in more uniform particle packing and fewer and smaller defects [38, 39]. On the other hand, the disadvantage of this method is the limitation on the shape of the final product, which is usually a simple sheet and shape. Perhaps the greatest challenge is applying solvent methods on an industrial scale, as multiple variables such as environmental conditions may cause changes in the product quality [40, 41].

3.3 3D Printing Methods

3D printing techniques make it possible to create objects by layering materials through the print heads and nozzles. The printable material solution is applied on a substrate with a pre-designed specific geometry. Then, during the printing process, different layers of the chosen material are poured. Finally, the structure is removed from its supports. Depending on the type of printing, a curing stage may be required [42, 43]. Figure 5 shows the process performed by the 3D printer.

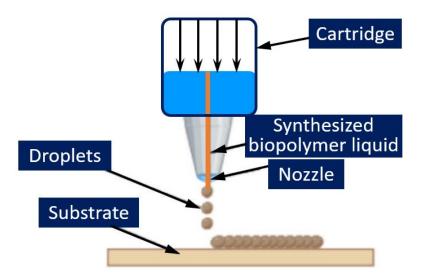


Figure 5 Schematic diagram of synthesized biopolymer making 3D bulk specimens by 3D printing.

In order to use equipment correctly during the 3D printing process, several basic requirements must be satisfied. The flow rate in the printing process depends on the viscosity of the material to be printed, which is a suitable material if the required volumetric flow rate is achieved at normal system pressures. The formation of beads with tabletop geometry is another critical consideration

depending on the surface energy of the system. However, it is normal for some residual stresses to occur within components formed layer by layer [44].

The 3D printing techniques have important applications and could transform medical production processes. Complex structures can be created using these techniques to create or repair worn bone and cartilage tissues. Its applications even extend to the manufacturing of customized food products, where cellulose-based packaging is developed for foods with low moisture content.

3.4 Other Important Methods

Injection modeling is one of the most efficient and extensively used techniques for manufacturing polymer-based products. Like the other methods, injection molding processes cause basic variations in the rheology and thermodynamics related properties of the products due to stress changes, experiencing high temperatures and cooling rates during the process. Therefore, it is important to carefully analyze the factors that may affect injection molding process and product performance before determining the manufacturing process for the product [45, 46].

Compression molding technique, known as press molding, involves placing the polymer (in the forms of pellets or sheets) between two molds that are heated to above the glass transition temperatures of the polymers. Then, the polymer attains its final shape after applying pressure to the mold, and then cools and removes the final matrix after solidification [47-50]. This technique is a secondary process because the polymers adopted in this process must be pre-processed. The compression molding process offers several advantages, such as the relatively high reproducibility, with model cycle times of just a few minutes, so it can be applied on an industrial scale. It also can create complex shapes with minimal material loss. However, compression molds are more expensive compared to other equipment used in other techniques.

Extrusion process is a method of forcing materials to flow through a certain-shaped orifice at a pre-determined rate under various conditions to obtain products. In extrusion process, shear deformation and thermal energies are applied to the polymers. This produces structural, physicochemical, and nutritional changes such as gelatinization and starch degradation, protein denaturation, and lipid oxidation. Extrusion is one of the commonly used methods in the polymer industry. The food and pharmaceutical industries are other application areas of this technique for modifying the microstructure or chemical properties of polymers [51].

3.5 Preparation for Some Important Biodegradable Materials

Based on processing methods, some additional descriptions are given here specifically for the preparation of biodegradable plastics from several important biomass sources [52].

3.5.1 Cellulose

As one of the most widely sourced natural polymers, cellulose has many outstanding advantages such as renewable, biodegradable, inexpensive, and environmentally friendly. Cellulose is the most abundant polymer found on Earth [53, 54]. It was discovered in 1838 by French chemist Anselme Payen, who isolated it from a plant and determined its chemical formula to be $(C_6H_{10}O_5)_n$ [55-57]. A schematic diagram of its molecular structure is shown in Figure 6. The Hyatt Company produced the first successful thermoplastic polymer, celluloid, in 1870 using cellulose. This renewable and

biodegradable material has received significant attention due to its different physical and chemical properties, which are different from those of synthetic polymers. Cellulose can be obtained from a variety of sources, such as plants, bacteria, and algae. Cellulose obtained from bacteria is purer than cellulose from plant-derived celluloses, as the latter also contains hemicellulose [58]. Cellulose was one of the key research areas during the emergence of polymer chemistry, and its structure was first demonstrated by pioneers in polymer science.

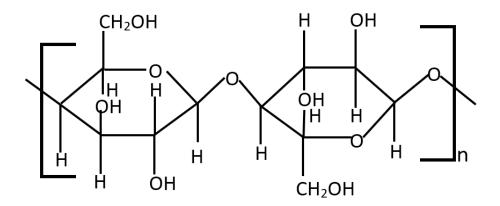


Figure 6 Schematic diagram of cellulose molecular structure (C₆H₁₀O₅)_n.

In fact, cellulose exists in various forms, such as α -cellulose, β -cellulose, γ -cellulose, microcrystalline cellulose, hydroxypropyl cellulose, hydroxyethyl cellulose, carboxymethyl cellulose, etc., each variety is distinguished according to its origin and nature, and applications to meet industry-specific needs. Among the different types of cellulose, α -cellulose is arguably the most common. It has long, uniform cellulose chains that contribute to the strength and durability of the finished product. α -cellulose is produced from wood pulp as raw material through a series of pulping and bleaching processes. It is the basis to produce the production of high-end paper products. In addition, α -cellulose also plays an important role in textile production – it is processed into cellulosic fibers used in production of fabrics such as rayon and viscose and can be used as fiber-reinforced composites due to its similar properties to engineered fibers, being another option for biocomposites and polymer composites. α -cellulose is also an important component in the production of specialty chemicals such as hydroxypropyl methylcellulose and ethyl cellulose, which are widely used in construction, pharmaceutical, food service and other industries. In addition, the fibers may also contain hemicellulose and lignin, and different percentages of these components can change the mechanical properties of the fibers [59-61]. A new method to produce cellulose nanofibers via the interfacial molecular film method at the air/water interface was developed, in which the formation of a hydrogen bonding network in a 2D plane promotes the formation of the fibrous nanocellulose monolayers. In addition, insoluble starch is dissolved, creating a spreading solvent for the monomolecular film on the water surface [62]. Furthermore, from a review perspective, the performance and processing of biodegradable foams can be enhanced through the incorporation of cellulose nanoparticles, which can promote the foaming of the polymers, thus improving the rheological properties and crystallization behavior. Upon incorporation of these nanoparticles at very low concentrations (1-5 wt.%), the mechanical, thermal, and dynamic properties are improved [53]. Furthermore, development of cellulose/ZnO based biopolymers can achieve enhanced gas barrier, UV-shielding effect and antibacterial activity [63].

3.5.2 Starch

An analog of cellulose, a mixture of α -glucose polymers and the main raw material of rice. Starch is a cheap, biodegradable biopolymer that is in abundant supply and naturally available. Nanofibers and microfibers can be added to the polymer matrix to increase the mechanical properties of starch, thereby improving elasticity and strength. Without the fibers, starch has poor mechanical properties due to its sensitivity to moisture. Starch is biodegradable and renewable and can be used in many applications, including plastics and pharmaceutical tablets. Starch-based materials show great potential, especially as more and more countries pass regulations banning disposable conventional plastics. In addition, starch comes from a wide range of sources, including corn, potato, and cassava starch produced on a commercial scale, and is the most explored starch source in plastic production [64, 65]. The starch industry extracts and refines starch from crops through wet grinding, washing, sieving, and drying. Today, the major commercially refined starches are corn starch, tapioca starch, arrowroot starch, wheat starch, rice starch, and potato starch. To a lesser extent, sources of refined starch are sweet potato, sago, and mung bean. To date, starch has been extracted from more than 50 species of plants. Crude starch is processed on an industrial scale into maltodextrin, glucose syrup, and fructose syrup. These massive transformations are mediated by a variety of enzymes that break down the starch to varying degrees. Breakdown here involves hydrolysis, which is the breaking down of bonds between sugar subunits by adding water. Some sugars are isomerized. The process is described as occurring in two stages: liquefaction and saccharification. Liquefaction converts starch into dextrin. Amylase is the key enzyme in the production of dextrin. The saccharification converts dextrin into maltose and glucose. The second stage uses a variety of enzymes, including pullulans and other amylases enzymes. Starch and polylactic acid (PLA) are commercially available and biodegradable, making them common choices for packaging. However, their barrier properties (either moisture-barrier or gas-barrier properties) and thermal properties are not ideal [66].

3.5.3 Bacterial Concrete

Concrete has been the world's most used building material since the invention of industrially produced Portland cement in the 19th century. However, cracks in concrete structures can significantly shorten their service life. Furthermore, due to the large amount of CO₂ emissions generated during cement manufacturing and concrete maintenance and repair costs, sustainably and economically repairing the cracked concrete and improving concrete durability has become a topic of interest. In order to solve these problems, self-healing techniques have been proposed and developed, including biomineralization-based self-healing (i.e., bacterial self-healing), which uses bacteria trigger microorganism-induced CaCO₃ precipitation to repair cracks in concrete, thereby improving the structure's durability. Although the mechanism is complex and developing a successful process is a challenge. The bacterial self-healing mechanism involves (1) First, the selection of bacteria species: In order to trigger the chemical reaction and biological activity of microorganisms and cause CaCO₃ precipitation. It is necessary to select appropriate bacteria, such as cyanobacteria, sulfate-reducing bacteria, denitrifying and urease-producing bacteria which can cause CaCO₃ precipitation. (2) Bacterial treatment method: such as surface treatment of building materials to reinforce the protection of construction materials, and self-healing bacteria are introduced into the cement-based composite materials during the casting process. When cracks form, water and oxygen penetrate through the cracks and interact with the cement-based composite material. Bacterial CaCO₃ will automatically fill the fissure through the controlling of pathway of the bacteria concentration, temperature, and pH value, and encapsulation. (3) Biological grouting: In addition to the positive results of the microbial self-heling processes in concrete crack repair, there are certain limitations to the commercialization of this technique. One of these is the use of laboratory-grade nutrient supply and bacterial transport materials in field applications. The exterior treatment of cracks in concrete structure is another disadvantage. Injectable biological grouting may be a potential engineering solution. (4) Nutrient types: The production of natural CaCO₃ production is limited by the calcium content in cement. Therefore, nutrients need to be added to provide additional calcium as a calcium source for bio-concrete. (5) Morphology of CaCO₃ precipitation: As an important aspect of CaCO₃ precipitation, three different anhydrous crystalline morphs, such as calcite, aragonite, and vaterite, are the most suitable. However, there are many factors that can affect the morphology, such as bacterial species, microbial excretions, and solution composition. Figure 7 schematically shows the mechanism of bacterial self-healing of cracked concrete [67-71].

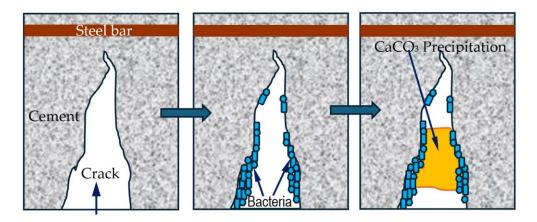


Figure 7 Schematic diagram of the mechanism for bacterial self-healing cracked concrete.

3.5.4 Packaging Materials

The packaging industry has been recognized as an important industrial sector in the food supply chain. Plastic food packaging materials account for more than 40% of the world's total plastic waste, and the vast majority of synthetic polymers used in food packaging are derived from petroleumbased polymers, such as the poly (vinyl alcohol) polymer. In order to combat the impact of the petroleum-derived plastics production and to renew and innovate the packaging materials from renewable sources with biodegradability and comparable performance, natural polymers have been intensively studied to improve their properties for replacing food packaging materials in recent years. Among them, natural polymer compounds such as starch, chitosan, cellulose, and gelatin have low-cost, abundant resources, good biocompatibility, and good biodegradability. One of the notable developments is the development of chitosan-based nanocomposite films using simple solution casting and solvent evaporation techniques with potential for food packaging applications. The preparation process includes: first preparing/synthesizing graphene oxide (GO), and cerium oxide (GeO₂), where the GO nanomaterial can be used to improve the overall performance of the film matrix and the GeO₂ nanoparticles can be used to the chitosan matrix to form chitosan-based composite blend films; finally, a variety of the properties of the film were physically characterized accordingly [72-75].

3.5.5 Paper-based Materials

Some cellulosic applications are based on the dissolution of cellulose, and several solvent systems have been developed. However, partial dissolution of cellulose offers another potential application. By partially dissolving cellulose and then undergoing a regeneration process, the cellulose fibers can maintain their fiber shape and be covered with regenerated fibrils. Many hydrogen bonds can be formed between fibers. In this way, paper-based biodegradable plastics can be produced by partially dissolving the cellulose. Paper and other pulp-based products can be considered alternatives to single-use plastics in food packaging applications. Paper is made from pulp materials. The raw pulp contains approximately 96% water and requires multiple steps of processing to obtain paper and other molded products. Various types of paper are used for food packaging in the form of paper bags, composite cans, fiber drums, multi-layer paper sacks, rigid boxes, folding cartons, corrugated fiberboards, paper bottles, etc. In addition, paper-based batteries have also attracted attention. A lot of research has been done over the past few years to find possible solutions to the need for environmentally friendly, portable, and biodegradable energy storage devices. These batteries use paper substrates to create flexible, lightweight energy storge devices that can also generate energy. Paper has been around the world for centuries, but it seems that just a few years ago it was rediscovered as a valuable substrate for sensors. We could easily list some of the countless advantages of this simple cellulosic substrate. Those characteristics make paper a preferred substrate for disposable sensors and integrated sensing platforms. Today, many examples of paperbased sensor-related applications are proposed in the literature, including optical and electrochemical sensor applications [76-81].

4. Methods for Characterizing the Biodegradable Plastics

The development of new products or materials is both exciting and difficult. From initial conception/design, through research testing and evaluation of the initial design (either through trial-and-error experimental approaches or computational simulation approaches) to final commercialization, no two material/product development pathways are exactly the same. However, there are six basic steps that can help get the process started, as shown in Figure 8. One of the most challenging tasks in the process is the "Test/Evaluate the Model". During the development procedures, research on material property characterization is necessary to test and evaluate the designed materials/products to determine if an initially designed material or product meets performance criteria set by the designer based on the market/customer's needs. There are many methods that can be used to test and evaluate the designed material systems, such as experimental methods (by physical and chemical methods) and computational methods at different scales (such as at subatomic, atomic, and continuum scales). The properties and characteristics of the materials to be tested or evaluated include physical, chemical, mechanical, thermal, structural, molecular, compositional properties, etc. If the process does not employ optimization algorithms, the new solution will only be a satisfactory solution, not an optimal solution. The new material/product development process has evolved in recent years, allowing for better organization of the process due to the scientific advances in materials research and development.

Recent Progress in Materials 2024; 6(2), doi:10.21926/rpm.2402015

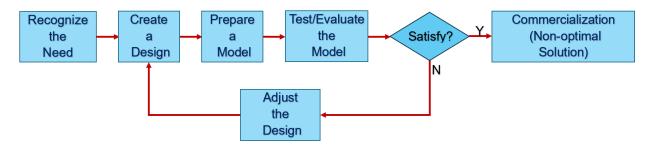


Figure 8 Schematic flowchart for six basic steps involving material/product development processes.

Biopolymers are long-chain molecules made up of repeating chemical blocks (monomers) obtained from renewable resources that may decay in the environment. It is becoming increasingly popular that the use of bio-DPs as a means of reducing the use of non-degradable and non-renewable resources for reducing environmental pollution [82]. The physiochemical, mechanical, and other catalogic properties of bio-DPs have led to a growing need for comprehensive characterization of biopolymers. With the advancement of analytical product development, various analysis/characterization methods based on experimental instruments and computer simulations at various scales, and data analysis and mathematical algorithms have been promoted as practical and preferred bioanalytical technology methods, as shown in Figure 9, enabling the understanding of the complex composition, structure, and performance of biopolymers to optimize the design and improvement of biopolymer applications [83, 84].

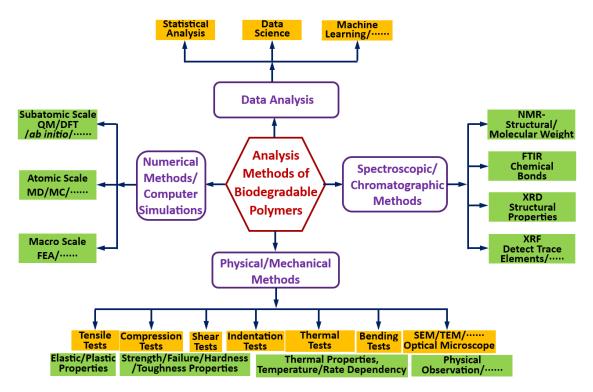


Figure 9 Schematic diagram of the characterization methods for biodegradable polymers.

The composition, structural and morphological information, etc. of the biopolymers often provide useful clues for predicting biodegradability, physicochemical properties, and other

properties of the polymers. Both qualitative and quantitative experimental and computational analyses of bio-DPs are available.

4.1 Experimental Methods

Experimental methods include physical observations of biopolymers and their surface micromorphology, elastic/nonelastic/strength of mechanical properties, and molecular structure, chemical bonds, weight loss, etc.

4.1.1 Physical Observations

Currently, many characterization techniques are available for analyzing and characterizing biopolymers, which help identify the material's end-use by providing a comprehensive understanding of the structure and property relationships. Various microscopy techniques occupy a unique position in physical observation and analysis of many characteristics, such as topology, composition, microstructure, morphology, and interfacial properties, etc.

Optical microscopy is possible the oldest and simplest microscopy technique. It is a 2D imaging system. Since its inception, research has been continuously upgraded to meet modern needs [85]. Optical microscopy has long been a basic device of materials research for sample analysis due to its simplicity and easy preparation of the samples. Optical microscopies help observe various characteristics, such surface morphology, size, shape, uniformity of the void content, failure analysis, and quality control. Conventional light, the source of light in optical microscopies, can magnify images thousands of times. Laser scanning confocal microscopy is an advanced optical microscopy that offers certain advantages over traditional widefield optical microscopy due to its outstanding capability of minimizing or eliminating background noise in the focal plane and its ability to scan relative thicker samples. It uses spatial filtering techniques to eliminate any out-of-focus light in specimens thicker than the direct focus plane. Laser microscopies can provide higher quality/resolution images than traditional fluorescence microscopies do, and its magnification can reach 4 or 5 thousand times of the original images, which greatly improves physical observation, but still cannot provide nanometer-scale resolution [86, 87].

Electron microscopy, such as scanning electron microscopy (SEM), can observe surface morphology and degradation of biopolymers [88], analyze surface morphology, and can observe porous and rough surfaces, indicating fast degradation and resulting in low crystallinity of HHx [89]. Transmission electron microscopy (TEM) is usually used to observe ultrathin cross-sections of samples [90]. For example, PHA depolymerase induces PHA biodegradation or bioerosion on the polymer surface. This qualitative examination allows for visible physical observation of the porosity, asperity, and pore structure of the polymer, which promotes bacterial attachment and results in the secretion of PHA depolymerase [89, 91]. The presence of large porous surface areas due to the reaction of microorganisms can therefore be explained by the extent of biodegradation [92].

4.1.2 Mechanical and Thermal Property Tests

Mechanical testing is an essential method for characterizing materials' mechanical properties, which requires material samples to be subjected to different loading cases and evaluates their responses to determine their mechanical characteristics. The mechanical properties of bio-DPs and

composites are crucial to the performance of devices or structures in which they are applied. Mechanical test methods are diverse, and testing of mechanical property degradation can be performed under a variety of conditions [93]. The most measured mechanical properties in bio-DPs' degradation experiments are stiffness, strength, and elongation-at-break. These mechanical properties are affected by various factors, such as chemical composition, production technique, processing characteristics, aging process, and application conditions [94]. Perhaps the most important of these are molecular weight and crystallinity [95]. Tensile testing, compression testing, bending testing, nanoindentation testing, and shear testing are some of the popular mechanical testing methods for biopolymers, as shown in Figure 9 [96]. Commercial mechanical testing systems are available for testing the mechanical properties of materials.

Bulk tensile testing is commonly used to measure the elastic modulus, yield strength, ultimate strength, and fracture properties of a material. While bulk testing provides useful quantitative information about the overall mechanical properties of the sample. However, among all the mechanical tests, micro- and nano-tensile testing can be one of the challenging tests because of the need to prepare small scale thin-film testing biopolymer samples and the need for proper sample clamping to prevent the sample from slipping from grips or breaking at the grips. Due to the small size of the samples, traditional mechanical clamps are not suitable for holding both ends of the thin films. Tensile testing evaluates a material's response to tensile stress, while compression testing evaluates its response to the compressive force. The bending testing evaluates the biopolymer's resistance to bending stress, the nanoindentation evaluates the elasticity and the resistance/hardness response to the impressive deformation, and the shear testing evaluates its response to forces applied parallel to its surface. These mechanical tests provide useful information about the strength, stiffness, elasticity, and other mechanical properties of biopolymers, helping to characterize and understand their possible uses [97].

The use of mechanical tests to evaluate the mechanical properties and to analyze the thermal properties of the biopolymers helps understand their thermal stability and degradation behavior. For example, differential scanning calorimetry (DSC) is a thermoanalytic technique in which the difference in heat required to raise a sample and a reference temperature is measured as a function of temperature, allowing the heat flow of a sample to be measured over a temperature range. Differential thermal analysis (DTA) which is a thermoanalytic technique like DSC in which the material under investigation and an inert reference material are subjected to the same thermal cycles, (i.e., the same cooling or heating procedure) while the temperature of any sample is recorded and the difference between the sample and the reference. This temperature difference is then plotted against time or temperature. Therefore, DTA curves provide data on structural changes, such as glass transition, crystallization, melting and sublimation. The area under the DTA peak is not affected by the heat capacity of the sample, instead it is the enthalpy change of the sample. Thermogravimetric analysis (TGA) is another thermal analysis method by which the mass of a sample is recorded as a function of time and temperature. This measurement provides quantified information on physical phenomena, such as phase changes, absorption, and desorption, as well as chemical phenomena, such as chemisorption, thermal decomposition, and solid-fluid-gas reactions (e.g., oxidation or reduction in %) of the samples over a range of temperatures [97].

4.1.3 Spectroscopic/Chromatographic Methods

Fourier transform infrared spectroscopy (FTIR) is a commonly used method to examine the molecular vibrations and functional groups present in biopolymers, providing information about their chemical composition. Using FTIR, it is possible to analyze the chemical interactions within a biopolymer or bio-composites, the molecular structure and functional groups of the materials, providing insight into their properties and behavior. FTIR can also be used to identify the root cause of material failure, such as cracks, delamination, or degradation. Biodegradability testing determines how the environmental conditions or other factors affect the materials' degradation processes. Assessment for the product life cycle makes it possible to evaluate the ecological footprint [98].

The crystal structure of biopolymers can be investigated using X-ray diffraction (XRD), providing information about their molecular structure and packing order [99]. Rheology and SEM are applied to further examine the structural and physical characteristics of the materials [100].

Nuclear magnetic resonance spectroscopy (NMR), also known as NMR spectroscopy or magnetic resonance spectroscopy (MRS), is a spectroscopic analytical technique based on the re-orientation of atomic nuclei with non-zero nuclear spin in an external magnetic field. In recent years, NMR spectroscopy has developed into a powerful tool for probing the structures of biopolymers in solutions and solid state. NMR provides a way to study the dynamics of polymers in solutions and examine the effects of solutes, solvents, and other factors on polymer performance. With the advances in 2D and 3D NMR spectroscopies, now it is possible to evaluate the solution conformation of small proteins, oligonucleotides, and oligosaccharides [101].

X-ray fluorescence (XRF) spectroscopy is an accurate, but fast and non-destructive analytical technique used to detect the elemental composition of materials. It is a compatible device applicable to solid, liquid, or powered samples and requires less or no sample preparation. Even the samples need not to be placed in a vacuum chamber similar to dealing with the energy-dispersive spectroscopies. XRF analyzers determine the chemical properties of a sample by measuring the fluorescent (or secondary) X-ray it emits when excited by a primary X-ray source [102].

4.2 Numerical Methods

To improve the predictability of the functionality of the biopolymers, multiple design factors need to be considered throughout the entire biopolymer fabrication processes. In biopolymer synthesis and processing, a coupled design method in which experimental and bottom-up computational approaches are crucial because it is important to be able to gain insights at different time- and length-scales [103]. A white paper on "Material Genome Initiative (MGI)" published by the U.S. National Science and Technology Council in 2011 aims to accelerate the discovery, design, and deployment of new materials, at lower cost, by harnessing the power of data and computational tools combining with experiments. Scientists and engineers from academia, industry, and government have made significant progress in expanding understanding and building the foundation required for models, computational and experimental tools, and data [104]. The ability to quickly share materials knowledge among scientists, engineers, and manufacturers can accelerate the discovery and fabrication of more capable materials, tools for better designing devices and structures, and more efficient manufacturing. Therefore, computational approaches are critical to speed up the design and development processes of novel materials. In the study of

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biomaterials, hierarchical architectures have attracted attention [105]. Figure 10 shows the hierarchical diagram for simulation approaches based on the length-scale and timescale. Therefore. computational models should incorporate all critical lengths and time scales. It is currently impossible to study materials such as biopolymers simultaneously at all scales with a single computational approach. For the understanding and prediction of structure-process-property relationships, bottom-up multiscale computational modeling approaches can provide a rigorous basis for the reliable and effective design and development of novel materials. Depending on the problem's interests and the available spatial and time scales, several viable modeling techniques at different scales can be adopted to investigate the structure and behavior of materials, ranging from quantum mechanics (QM – subatomic modeling) to finite element analysis (FEA – continuum simulation).

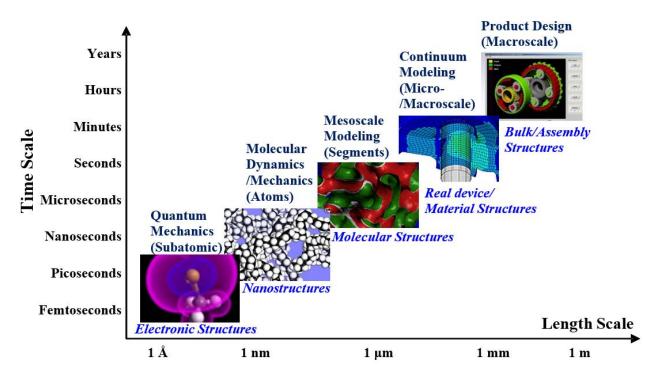


Figure 10 Hierarchical diagram for simulation approaches based on length-scale and timescale.

4.2.1 Quantum Mechanics, Density Functional Theory, Ab Initio

Quantum mechanics (QM) simulations are ubiquitous computational methods to study various features of atomic/molecular systems. One of the general applications of QM simulations is the study of reaction mechanisms in biological systems or condensed-phase systems. A significant application of the QM methods was the study of enzymatic reactions, which was proposed in the mid-1970s. Since then, the QM simulations have been extensively applied to investigate a variety of chemical and biochemical systems. The QM methods have been aimed to exploit the advantages of advanced quantum and classical approaches by combining two levels of theories. The quantum models focus on the regional or local details of the system. For instance, if a process consists of a reaction at the active site of an enzyme, the residual molecules (or fragments) involved in the reaction would be specified as the QM subsystem, while the rest of the molecular system including

enzyme, solvent, counterions, etc. would be specified by the classical molecular mechanics subsystem [106]. Coupling these two different level approaches to establish the entire systems has some great challenges, such as the way to combine these two quite different levels of theories across the covalent boundaries, the way to handle the long-range effects in the context of QM subsystems, the way to deal with the quantum couplings and classical Hamiltonians, and the way to deal with the explicit couplings, etc. [107].

Another computational method based on the QM fundamentals is the density functional theory (DFT) widely applied in chemistry, physics, and materials science. It is used to simulate the electronic structure of atoms, molecules, and solids [108-111]. It has been widely applied in computational solid-state physics since the 1970s. However, it was not until the 1990s that improvements in the method brought it to acceptable accuracy for quantum chemistry applications. Its actual advantage is its good price-performance ratio compared to the methods based on electron-correlated wave function, such as coupled cluster theory or Møller-Plesset perturbation theory. Accordingly, current DFT methods can be used to study the larger (and often more corresponding) molecular systems with sufficient accuracy, thus expanding the predictive capability inherent in electronic structural theory. Therefore, the DFT method is by far one of most popular methods applied in calculation of the electronic structures of the molecular systems and materials. The immense importance of the DFT method in the research of physics and chemistry was demonstrated in 1998 when Walter Kohn was awarded the Noble Prize "for his development of the density-functional theory" [112].

Ab initio is a Latin term meaning "from the beginning", implying a solid foundation of the method. Ab initio methods can calculate the essential characteristics of the materials based on the theories of QM. The foundation is the disintegration of the Schrödinger equations for the many-body systems posing several dozens to several thousands of atoms and the relevant electrons. Since the computational complexity increases dramatically with the number of atoms/electrons, various solutions to the problem have emerged. For example, the Born-Oppenheimer approximation based on non-relativity postulates that electrons move much faster than atomic nuclei because they are thousands of times lighter than atomic nuclei. Therefore, the motions of the electrons and nuclei can be decoupled. A clear fundamental of this approach is based on the wave functions. One of the simplified approaches is the so-called Hartree-Fock method, which can yield reasonable accurate results for various essential characteristics of the systems via the variational theorem to obtain approximation solutions. However, it ignores electron correlations in electronic structure calculations, meaning each electron treats the rest of electrons as an averaged field. A farther method to the Hartree-Fock method is the DFT based on the Hohenberg-Kohn theorem, which assumes the ground-state electronic energy being particularly determined according to the electron density [113].

4.2.2 Molecular Dynamics/Monte Carlo Method

Molecular dynamics (MD) simulations are atomic-scale simulations that describe the interatomic interactions based on the interatomic potentials. In the MD methods, the electron connective relationship estimation is based on the Born-Oppenheimer theory and hance the effects of the electrons are averaged, and the changes in positions and velocities of the atoms with time can be calculated based on Newton's second law, i.e., Newton's equations of motion, which requires that the MD time step size or time step number used to describe the atomic motions are sufficient small

Recent Progress in Materials 2024; 6(2), doi:10.21926/rpm.2402015

or large enough for electrons to effectively reach their ground stable states compared to the nucleus due to mass differences. Interatomic force fields or potentials are generated experimentally or based on the first principles to describe the interactions between atoms, in the form of reproducible forces, such as repulsive or attractive forces. The accuracy or reliability of the interatomic force fields or potentials determines the accuracy or effectiveness of the MD simulations, reflecting the ability to bridge the mesoscale methods [52, 114-119]. As the advancement of MD, MD has evolved as a ubiquitous, versatile, and powerful computational method for scientific fundamental research in biology, chemistry, biomedicine, and physics [120]. In recent decades, driven by the rapid development of supercomputing technology, MD has evolved as a power tool used as a first principles prediction method for investigation of the properties of materials and biomaterials and the physiochemical processes, and has even entered the engineering field as a design tool for developing novel materials. These significant developments have great impacts and are described in many recent articles focusing on the applications of MD in energy systems (such gas/liquid/solid fuel oxidations, catalytic combustion), chemical reaction processes (such as pyrolysis, electrochemistry, nanoparticle synthesis), thermal and fluid mechanics (such as heat transfer, phase change). Due to the great significance and potential of the MD simulations for studying the largescale reactive chemical systems, the reactive force field (ReaxFF) systems based on MD simulation framework were proposed and applied successfully to the biomass or biomaterials systems [120-131]. The ReaxFF uses the regular relationships between chemical bond distance and bond order to represent atomic interactions. The angular, torsional, or other valence terms were combined into the force field functions via the same bond orders so that all the bond orders (potentials) smoothly go to zero when the bond breaks. In addition, the ReaxFF has secondary or physical bond potentials (such as van der Waals bonds) in terms of Coulomb and Morse potentials to represent non-chemical bond interactions between all atoms. These non-chemical bonded interactions are taken effect over short distances. The coefficients/parameters in the ReaxFF are derived from QM according to the molecules/atoms studied, and heat of formation and geometric data for some stable hydrocarbons [123].

The Monte Carlo (MC) methods, or MC experiments, are a large family of computational algorithms that rely on a large number of repeated random samplings, such as clusters of atoms in the molecular systems, to obtain numerical results. The basic concept is to use stochasticity to solve problems that may in principle be deterministic, such as the minimization of the total energy in a system. The MC methods are mainly used in three different problem classes: optimization, numerical integration, and generating plots from probability distributions. They can also be used to model phenomena with significant uncertainty in the input. The MC methods are often implemented using computer simulations, which can provide approximate solutions to problems that are difficult to analyze mathematically or are too complex. In MD, the trajectories of a group of atoms are simulated according to Newton's equations of motion, as opposed to the MC methods which finds the equilibrium of the system based on the generation of random states. All-atom simulations are often used to sample the energy landscape and predict the structure/system of the sequence. The MC methods are widely used in various fields, such as physics, chemistry, materials, biology, statistics, artificial intelligence, finance, and cryptography. They are also applied in social sciences such as sociology, psychology, and political science. The MC methods are considered one of the most important and influential ideas of the 20th century and have achieved many scientific and technological breakthroughs [103, 132-141].

4.2.3 Finite Element Analysis/Finite Element Method

Finite element analysis (FEA) or finite element method (FEM) is a powerful computational tool for numerically solving differential equations that arise in engineering, scientifical, and mathematical modeling. The representative problem areas of interest can involve the traditional fields of solid/structural mechanics, thermal systems/heat transfer and mass transport/fluid mechanics, and acoustics and electromagnetic fields, often at micro to macro scales.

To solve a problem via FEA method, a large and complex system/problem domain is divided into smaller and simpler parts called finite elements, and the adjacent elements are connected by nodes. This is achieved by a specific spatial discretization in the spatial dimensions. This discretization process is called meshing of the object, resulting in the numerical domain for solutions, which has a finite number of points (nodes) and elements. The FEA formulation of boundary value problems ultimately yields a systematic assembly/matrix of algebraic equations for modeling the entire problem. This method approximates unknown functions over the domain via a shape function over each element or the total potential over the entire domain via variational calculations.

The FEA can be used to analyze problems on complex domains (such as cars and oil pipelines) when the domain changes (such as the system/problem domain responses to the loading and/or moving boundaries), when the desired accuracy varies across the entire domain, or when the solution are lacking smoothness. The FEA simulations provide a valuable resource because they eliminate instances of creating and testing complex prototypes for various high-fidelity situations. For example, in a frontal crash simulation of a vehicle, it is possible to increase the prediction accuracy for "important" areas such as the front of the car and reduce the prediction accuracy for the rear by changing the mesh size or density (thereby reducing simulation costs). Another example is numerical weather prediction, where accurate predictions of developing highly nonlinear phenomena (such as tropical cyclones in the atmosphere or eddies in the ocean) are more important than in relatively calm regions. Since its introduction in the 1940s, the FEA has been widely used in biomaterials and biocomposites, mainly focusing on the fields of structural design and optimization or mechanical behavior of the materials [142-147]. Since the synthesis, fabrication and behavior of the bio-DPs strongly involve chemical reactions and biochemical mechanisms essentially at the atomic scale. There will be great challenges and efforts to apply FEA to the design and prediction of a broader aspect of bio-DPs.

4.3 Data Analysis

In recent years, data analysis has become a powerful tool in the science and engineering fields such as materials, biomaterials, polymers, and biopolymers. With exponential growth in the availability of data obtained through experimental and computational methods, and the advancement of experimental and computational techniques, researchers can now extract valuable insights from large and complex material data sets. Data analysis enables the exploration of patterns, relationships, and trends in these data sets, ultimately improving understanding of biopolymer behavior and promoting informed decision-making [148-150]. A variety of analysis methods are available in this field, which involve artificial intelligence (AI), statistics, machine learning (ML), deep learning (DL), neural networks, data mining techniques, etc. Each of which provides a unique way to extract meaningful information from the material data sets [151-154]. Figure 11 shows the

schematic flowchart of the data analysis, in which it shows the roles of the data analysis in the new material/product development process.

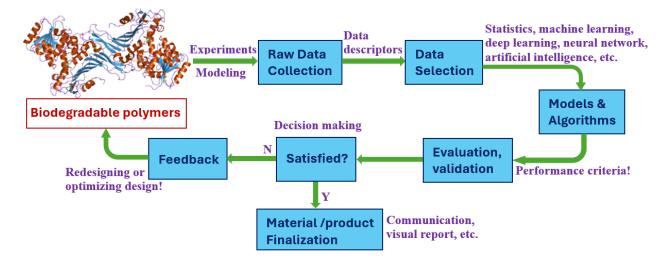


Figure 11 Schematic flowchart of data analysis.

The common data analysis approaches contain three main components [155]. The first component consists of original data sets containing information about the material structure, measurements directly related to these structures, and physical properties relevant to the material development goals. The second part regards data representation, i.e., quantitatively describing the data instances in the first part, collecting raw descriptions of the materials for identification and analogical reasoning. The final component is a summarizing part that utilizes the data analysis methods or algorithms aforementioned (either a single method or a combination of methods) to extract knowledge and relationships from the materials data sets for a specific purpose, such as predicting properties, providing guidance for material processing or synthisis, or designing and optimizing new material compositions and structures for optimal performance. The raw data for the first component of the data analysis comes from the experimental and/or computational methods. Details of the second and third components of the data analysis are discussed below.

4.3.1 Data Analysis Foundations – Statistical Analysis

Data analysis foundations – Statistical analysis plays a fundamental role in data analysis in materials science, allowing researchers to quantify the relationships between materials composition, processing conditions, and materials properties and performance. Statistical analysis helps establish structure-property relationships, providing valuable insights into materials design and development [156]. Many researchers have tried to find descriptors that intuitively predict material properties from combinations of basic parameters [155, 157]. Techniques can identify key factors/descriptors that influence material behavior and help optimize materials, such as Symbol Regress [158], LASSO algorithm [159], and SISSO algorithm [160]. Descriptor-based prediction of materials properties is becoming a new approach in materials science. Although these methods can achieve good results, they require many conditions, such as big data, appropriate algorithms, and sufficient features [161].

4.3.2 Raw Data Mathematic Conversion

Unlike traditional trial-and-error approaches, data analysis based on material informatics methods involve converting the raw data descriptions into appropriate representations and relationships that can be used for mathematical reasoning and inference. These methods are targeted on qualitative and quantitative interactions between materials based on these transformed representations and relationships, allowing researchers to discover potential patterns in the material data and use these potential patters for further applications. The development of material representation and relationships (i.e., the identification of material descriptors or methods applied for learning material representations and relationships from raw material data) plays a crucial role in data analysis methods. This is because the effectiveness of data analysis algorithms highly depends on the material representation, as it directly affects the performance of the algorithm and contributes to the explanation and interpretation of the inference process and prediction results. Recent advancements in automated experiments and high-performance computing facilities facilitate the acquisition of large amounts of experimental and computational data. Therefore, there is an increasing need to develop the ability of interpretable and explainable data analysis methods to enhance our understanding of physiochemical phenomena for materials and biomaterials research [162, 163].

4.3.3 Quantitative Analysis

A core challenge in the field of materials science and engineering is to discover the relationships between strucure, processing, and performance of the materials based on the experience and available theories, which needs to quantitatively understand the compositions and structures of materials with specific properties and subsequently verify them experimentally. Unfortunately, material research and development in this way is time-consuming and costing, and sometimes is impossible endeavor that often relies on serendipity. Data analysis based on materials informatics has emerged as a rapidly growing interdisciplinary field aimed at addressing these challenges. It uses a data-driven approach to extract practical knowledge about materials and their associated physicochemical phenomena from experimental and computational data, ultimately accelerating the discovery and development of novel materials [164-167].

4.3.4 Advanced Data Analysis Methods

Data analysis using mining and learning from large amounts of data in the materials science can solve the problems of slow development of novel materials and high experimental costs of traditional trial-and-error methods, which is an important part of next generation material discovery. A new era of rapid advances in information technology offers a viable approach: using machine learning (ML) to help discover, design, and optimize new materials. Some novel applications include generating drug candidates, studying chemical phenomena, and aiding theoretical calculations. One of the most important tasks for applying ML learning is the prediction of physical or chemical properties. In this direction, quantitative structure-property relationships are specially used to predict the biological activity of compounds based on their structural information. Likewise, materials scientists use a similar technique called quantitative structure-property relationships to predict various properties of materials based their 2D or 3D structural data. In these methods, the

prediction task relies heavily on a set of descriptors, which serve as numerical representations of structural information. Typically, these descriptors are digital objects obtained by converting raw molecular structural information through some predefined procedures. These descriptors can be as simple as a list of molecular compositions and structures or as complex as matrices, fingerprints, etc. [168].

The ML methods are good at discovering and establishing connections/relationships among data points and have powerful data processing capabilities and are often used to analyze fuzzy data and nonlinear data [169, 170]. ML is an interdisciplinary subject within the topics of artificial intelligence and between computer science and statistics. It is at the core of artificial intelligence (AI) and data science. It promotes the availability of online data and low-cost computing through algorithmic learning. ML is a rapidly innovating technology, and the importance of applied ML relies on the capability of finding reliable models or relationships of structure, property, and processing, which will never go out of style. There are various types of complex factors in material research affecting the results, such as types of materials, processing and environmental conditions, and applications. Therefore, the data relationships need to be clear, and the ML methods are good at discovering and establishing connections and relationships among numerous data sets.

The key procedures of ML can be interpreted as data preparation, descriptor/feature selection, algorithm/model selection or generation, relationship prediction and application [171]. By applying these procedures to the material discovery and design, i.e., from experimental process data collection or computational data collection to performance prediction and finally to experimental validation, a full data analysis cycle by ML method is completed. Recently, material design and development using ML have received more attention and have been greatly improved in the perspectives of time efficiency and prediction accuracy. A variety of deep learning (DL) methods have been proposed and applied to address the challenges related to the structure-processing-performance relationships of a material and the materials' characteristic predictivity based on the big data [148, 163, 167].

5. Degradability of Bioplastics

Almost all compounds and materials biodegrade, and the key factor is time. The biodegradability standard used by the European Union is that more than 90 percent of the original material must be converted into carbon dioxide (CO₂), water (H₂O), and minerals through biological processes within a half year, while the International Union of Pure and Applied Chemistry (IUPAC) organization defines biodegradation as "degradation caused by enzymatic process resulting from the action of cells" and states that this definition has been modified to exclude abiotic enzymatic processes [172]. Biodegradability refers to the ability of a material to decompose when interacting with biological elements, or it is defined as the chemical decomposition of a substance, accomplished through the enzymatic action of microorganisms, resulting in changes in chemical composition, mechanical and structural properties, and forming metabolites that are environmentally friendly materials such as methane, water, biomass, and carbon dioxide [173]. The rate of this degradation varies significantly. Biodegradation can take decades, while some industrial processes can completely break down polymers within hours.

The biodegradation mechanism of polymers can be described as the physical and chemical properties of polymers deteriorate and the molecular weight decreases under the influence of

microorganisms in both aerobic and anaerobic environments with the assistance of abiotic chemical reactions such as photodegradation, oxidation, and hydrolysis, and low molecular weight products such as CO_2 , H_2O , CH_4 generated. The entire process can be divided into three stages: (1) stage I Biodeterioration – at the end of the polymer's service life, the polymer begins to lose its physical and structural properties. The rate of degradation depends on many factors, such as environmental factors (temperature, UV radiation, humidity, etc.), enzyme activity (bacteria, atmospheric pollutants or agrochemicals, thermal oxidation, etc.), and polymer characteristics (polymer chain length, crystallinity, molecular weight, size, topology, and morphology, etc.), as well as physical forces (compressive force, tension, shear forces such as air and water turbulence, snow pressure, and animal tearing, etc.). Stage II Bio-fragmentation - Once the polymer is broken down into shorter chains, it becomes more susceptible to enzymatic, i.e., biological, "attack". The rate of this breakdown is related to the properties of the polymers, for example linear non-reactive segments will be more difficult for enzymes to reach and destroy. Stage III Microbial Assimilation and Mineralization – It can be thought of as the microorganism eating and digesting the polymers for their own development and energy needs, producing cellular biomass as well as CO₂, H₂O, CH₄, depending on oxygen availability [174-181].

The biodegradation process is affected by many factors, including molecular structure, polymer morphology, chemical composition, processing conditions, and environmental conditions. In general, the effects of heat, light, air, and moisture are the most important factors in the biopolymer degradation processes. The main chemical changes are oxidation and chain scission, resulting in a reduction in % in the molecular weight and depolymerization of the polymer. These changes can affect physiochemical properties such as strength, ductility, melt flow index, appearance, and color. The changes in performance are often referred to as "aging".

5.1 Degradation During Processing

Thermal polymers (whether virgin or recycled) must be heated until molted to form their final shapes, with processing temperature ranging from 150-320°C, depending on the polymers. Even in the absence of air, polymers can be oxidized by heating them to temperatures high enough to cause thermal degradation and damaging chemical changes. The molten polymers also experience significant shear stress/strain during modeling or extrusion process, which is sufficient to cause the polymer chains to break. Distinct from many other types of degradation, the melt-processing affects the degradation process of the entire polymer system, not just on the surface layers of the polymer system. If degradation starts, the chemical weaknesses in the polymer system initiate, which become excitation spots for further degradation during the life of the object [182].

5.2 Microbial Biodegradation

The term "biodegradation" is used to describe the mechanisms that lead to widespread damage to biomaterials and is often associated with terms such as ecology, waste management, environment, and plastics due to their long service life. Another term related to them is biomineralization, which relates to the conversion of organic substances into minerals. Microorganisms (mainly bacteria and fungi) often produce extracellular enzymes that help degrade various types of bio- and fossil-based plastics. Bacteria and fungi can degrade these polymers or decay the organic substances into simpler matters such as CO₂ and H₂O via different metabolic and

enzymatic mechanisms. Therefore, different microbial enzymes degrade different types of polymers or biopolymers, and consequently, different microbial enzymes degrade different types of polymers or biopolymers with different biodegradation rate due to the multiple factors, such as the material structure, environment, etc. Both biodegradable and non-biodegradable polymers, such as PHA, PLA, PET, PHB, PVC, PCL, and PBS, have been reported to attach to various microorganisms and their enzymes [183, 184].

5.3 Environmental Biodegradation

Generally speaking, any biological process depends on the existence of water, or more precisely moisture. Since water/moisture is an essential condition for developing and reproducing microorganisms, moisture can affect the biodegradation of polymers in different ways. Therefore, the degradation rate of polymers increases in the presence of sufficient moisture due to rapid microbial action. Furthermore, rich-moisture conditions help the hydrolysis process via producing more chain scission reactions [184-186].

When talking about the biodegradation in soil, one must consider that two possible situations exist: biodegradable polymers that are disposed of through composting processes (littering) and biodegradable polymers that are intended to biodegrade directly into the soil, e.g., the materials used in agriculture (e.g., mulch, irrigation pipes, flowerpots, etc.). Since soil is used to produce food for humans and animals, much attention must be paid to assessing any negative and persistent impacts of on-site plastic disposal. Another possibility must also be considered: biodegradable polymers could be used to produce compost if intentionally disposed into the soil [184]. In addition, soil microbes play an important role in degrading harmful effects of biodegradable microplastics on plants [187].

The pH number can alter the hydrolysis reaction rate by varying the acidic or alkaline conditions. Degradation outcomes from different polymers can change the pH conditions, thereby altering the rate of the degradation processes and microbial growth. Likewise, enzymatic degradability is significantly affected by the softening temperature of the polymer. Polyesters with higher melting points are less likely to biodegrade. Potential enzymatic degradability decreases with increasing temperature [188, 189].

The conditions to which materials are exposed or experienced, such as weather, ageing, sunlight, soil burial, water, etc. can affect the degradation processes. Therefore, polymers experience thermal, chemical, mechanical, and photo degradation, becoming synergistic factors in accelerating the biodegradation processes [190].

5.4 Biodegradation Due to Polymer Characteristics

From a biodegradability perspective, molecular structure and weight plays an important role in determining the polymers' characteristics. Degradability decreases with increasing molecular weight. Higher molecular weights degrade more slowly. It is easier for microbial enzymes to attack low molecular weight substances [188].

Furthermore, the shape and size of the polymers also play a critical role in the degradation processes. The polymers having large surface areas can be degraded rapidly compared to the polymers with small surface areas. There is a standard criterion for the biodegradation of various plastics in terms of shape and size [191].

In addition, non-polymeric contaminants, such as fillers or dyes, can affect degradability. In fact, when the cellulosic filler is increased in the polymer materials, the thermal stability decreases and the ash content increases. The dispersion and interfacial adhesion between the cellulosic fillers and the thermoplastic polymers are the main factors affecting the thermal stability of the composite systems. Likewise, metals act as good co-oxidants in the manufacture of polyolefin for polymers that are susceptible to thermos-oxidative degradation [192].

Biosurfactants are amphiphilic compounds produced primarily on biological surfaces. Due to their low toxicity and high biodegradability, the addition of biosurfactants can enhance the biodegradability of polymers. Biosurfactants facilitate the biodegradation processes due to the presence of specific functional groups, therefore, they allow activity under extreme conditions of temperature, pH, and salinity [193-195].

5.5 Assimilation and Mineralization

The final phase in the degradation processes, i.e., assimilation, is the most problematic because of the lack of proven techniques to provide legal justifications for the environmental impact of the materials, especially for those newly developed materials. However, verification of the assimilation processes, such as mineralization, is an essential way to identify if the material is so called "environmentally friendly". During this step, direct interactions between the polymer fragments and the microbial cells are evident. Microorganisms survive by recovering energy from the decomposition of polymers and use carbon, phosphorus, oxygen, sulfur as nutrients for their cell structures. The mineral substances released by microorganisms do not indicate a toxicological risk to the environment, while the microbial organic substances from the polymer decomposition may possess certain level of real hazard [184].

6. Future Perspectives and Conclusions

In September 2015, the General Assembly of The United Nations approved the 2030 vision agenda, which contains 17 Sustainable Development Goals (SDGs) based on the principle of "leaving no one behind. The new agenda emphasizes a holistic approach to sustainable development for all (with full inclusion of persons with disabilities). The 17 SDGs reflect a comprehensive approach to achieving healthy and sustainable societies. Most of the SDGs are closely related to and achievable by the plastics industry and polymers. Materials science and engineering contributes to goals such as SDG 3: Good Health And Well-Being, SDG 6: Clean Water And Sanitation, SDG 7: Affordable And Clean Energy, SDG 8: Decent Work And Economic Growth, SDG 9: Industry, Innovation, And Infrastructure, SDG 11: Sustainable Cities And Communities, SDG 12: Responsible Consumption And Production, SDG 13: Climate Action, SDG 14: Life Below Water, SDG 15: Life On Land, and SDG 17: Partnership For The Goals [196]. Therefore, the innovation, manufacturing, marketing, consumption, recycling, and other characteristics of bio-DPs ensure that the commodity is commercially, socially, and ecologically acceptable. As a result, the lives of everyone improve and the entire community benefits. Using sustainable materials and energy, waste disposal and operating costs may all be reduced. BioDPs will open new possibilities in the effort to create a better environment free of hazardous substances and products [197, 198].

The need to develop biodegradable alternatives to petroleum-based polymers has led to much research. Due to their unique physicochemical, biological, and degradation features, bio-DPs are

also attractive materials for biomedical applications. The medical industry will benefit greatly as biopolymer-based bio-implants and drug delivery agents are currently being developed. Additional advances in the future might lead to a revolution in the economics of medical implants [173, 199, 200].

It is worth noting that researchers and engineers should pay more attention to the practical aspects of bioplastics. In fact, it is expected that examples of the current use of bioplastics or their potential for applications in daily life will become the focus, while a deeper understanding of the changes in the structure and properties of the biomass resources after different treatments will provide better ways to utilize the resource [201-203].

Science is rigorous process involving good hypotheses coupled with reliable and effective methods, such as experimental approaches, computational approaches, or a combination of experimental and computational approaches, to advance and enrich the knowledge, followed by data analysis to reveal findings and pathways to new material development. For the development of bio-DPs, it will follow the same path to advance the fundamental science of the target materials. Among them, the first basic information, and probably the most important one, is polymer chemistry, which should provide various information such as molecular weight, degree of polymerization, and distribution of molecular weight. In addition, microbiological science should be enhanced to deepen the mechanistic understanding of enrichment, series transferring, and medium composition, etc. Verification of polymer degradation should be properly controlled to produce high quality research results [204].

Therefore, in this paper, various perspectives on biodegradable polymers were systematically reviewed and discussed in detail based on the existing and closely related scientific literature on materials, biomaterials, and biodegradable materials to find the answers on how to effectively study and develop biodegradable polymers. First, the source classifications were summarized from different perspectives. Some major preparation methods of biodegradable polymers were discussed, such as micro-extrusion of biofibers, solvent casting of thin-films, 3D printing, injection and compression molding and extrusion processes, especially those applied in the preparation and applications of some important biopolymers, such as cellulose, starch, bacterial concrete, packaging materials, and paper-based materials. More importantly, research and characterization methods applied for materials characterization and development that can be employed in biodegradable polymer research and development were particularly discussed, including experimental (physical and chemical) and computational approaches at various scales (such as quantum mechanics at subatomic scale, molecular dynamics at atomic scale, and finite element analysis at continuum scale), as well as data analysis methods, were described in detail. The biodegradation mechanism of polymers and factors affecting their biodegradability were discussed. Finally, the future perspective of the biodegradable polymers was also discussed. Based on the review, the proper and effective adoption of the state-of-the-art biomaterial research and characterization techniques, including experimental, computational, or a combination of both methods, as well as advanced data analysis methods will help advance the development of novel biodegradable polymers [205].

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Author Contributions

Conceptualization, Z.H. and R.Z.; methodology, Z.H.; investigation, Z.H.; resources, Z.H. and R.Z.; data curation, Z.H.; writing—original draft preparation, Z.H.; writing—review and editing, Z.H. and R.Z.; visualization, Z.H.; project administration, Z.H. and R.Z. All authors have read and agreed to the published version of the manuscript.

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