Biodegradable Film Materials for Packaging

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In today's world, the problem of "white pollution" is becoming more and more serious, and many countries have paid special attention to this problem, and it has become one of the most important tasks to reduce polymer waste and to protect the environment. Due to the degradability, safety, economy and practicality of biodegradable packaging film materials, biodegradable packaging film materials have become a major trend in the packaging industry to replace traditional packaging film materials, provided that the packaging performance requirements are met. Degraded plastics are plastics that have been subjected to defined environmental conditions for a period of time and contain one or more steps that result in significant changes in the chemical structure of the material resulting in loss of certain properties (such as integrity, molecular mass, structure or mechanical strength) and/or fragmentation.

degradable | packaging film materials | degradation mechanism | modified

1. Introduction

Plastic was once hailed as one of the greatest inventions of the 20th century, because of its light weight, good processing performance, low price and many other advantages that make the global plastic industry has been rapid development [1]. According to statistics, the total global production of plastic products exceeds 300 million tons [2][3][4], with 13 million tons entering the water [5]. However, only 6–26% of plastic products are recycled, which means that at least 74% of plastic waste ends up in landfills or enters the environment every year [3][6], of which about 46% comes from the packaging industry, especially food packaging films, which are largely non-recyclable [7]. Since most plastics are now made from non-biodegradable materials, it often takes one to two hundred years to degrade these plastic products [8][9][10][11][12][13].

Plastic is the most commonly used packaging material [14][15], especially packaging film material. However, the packaging industry generates about 141 million tons of plastic waste each year [16], and most of the packaging film materials are composed of non-degradable materials, which obviously leads to many environmental problems, such as "white pollution" [17][18][19]. General purpose plastic packaging films such as polyethylene (PE), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC) [20][21] film materials undergo a long period of aging under the current common waste disposal method of sanitary landfill conditions. Under the action of abiotic factors (such as solar radiation, high temperature, wave impact, gravel abrasion) or biotic factors (such as ingestion, colonization, degradation) [22][23], physical or chemical property changes, molecular weight reduction and molecular weight distribution changes, but its decomposition is not complete, the majority of decomposition into microplastics (particle size < 5 mm) or nanosized-plastics (particle size < 0.1 μm) [24][25]. At present, microplastics have been widely detected in oceans [24][26], sediments [27], rivers [28][29][30], lakes [20], atmosphere [31][32][33], soil [34][35] and organisms [36], disrupting the normal metabolism and energy balance in organisms, thus affecting the normal growth and reproduction of organisms and causing potential harm to human health [37][38].

To solve these problems, it has become important for biodegradable packaging film materials to replace traditional packaging film materials [39][40]. However, biodegradable plastics currently account for less than 1% of total plastics production [41]. Compared with traditional packaging film materials, biodegradable packaging film materials are more expensive to produce and have poor mechanical properties and their barrier properties, which are the main reasons for their limited applications [42].

2. Degradation Mechanism of Degradable Packaging Film Materials

Degraded plastics are plastics that have been subjected to defined environmental conditions for a period of time and contain one or more steps that result in significant changes in the chemical structure of the material resulting in loss of certain properties (such as integrity, molecular mass, structure or mechanical strength) and/or fragmentation [43][44]. As shown in **Table 1**, the degradation degree can be divided into complete and incomplete degradation, and different degradation mechanisms can be divided into photodegradation, water degradation, thermal oxidative degradation and biodegradation [45].

Table 1. The classification and characteristic of degradable plastics.

Classification	Category	Features
By degradation principle	Biodegradable plastics	Similar performance to traditional plastics, good degradability, high safety
	Photodegradable plastics	Simple and low cost production process
	Thermal oxidative degradation plastics	Requires oxygen and heat
	Hydrodegradable plastics	Short degradation time, no trace, no pollution, low cost
By degradation characteristics	Fully degradable plastics	Completely disintegrates and leaves no trace
	Incomplete degradable plastics	Partial degradation

2.1. Photodegradation

Photodegradable materials are degraded to low molecular weight compounds that are relatively safe for the environment by photo-initiated fracture and free radical oxidative fracture reactions under the action of sunlight (mainly UV light) [46]. Photodegradable film materials can be mainly divided into photodegradable materials obtained by copolymerization and photodegradable materials with composite photosensitizers [47].

In sunlight, UV light with a wavelength of 290 nm–400 nm only accounts for about 5%, and it is the UV light that causes photodegradation of the film. **Figure 1** shows the photodegradation mechanism. The molecular chains react under certain conditions of oxygen, temperature and humidity, and the long molecular chains are decomposed into peroxides and eventually achieve photodegradation [48].

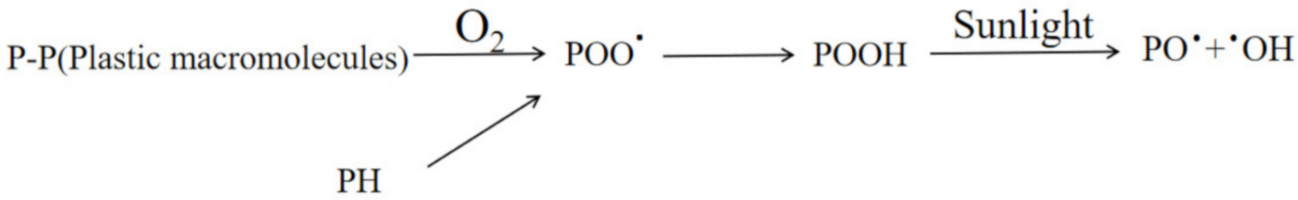

Figure 1. The mechanism of photodegradation.

Christensen et al. [49] investigated the photodegradation properties of polymers with a 1:1 mass ratio of polycaprolactone to polyvinyl chloride by monitoring CO_2 emissions during UV exposure. The results showed that the interaction of the two components in the polymer reduced the photodegradability. Najaf et al. [50] used polyaniline modified TiO_2 as a photocatalyst and then combined it with polyvinyl chloride to make photodegradable films. The results showed that the quality of polyaniline decreased by 67% when the molar ratio of polyaniline to TiO_2 was 10:1 under the condition of 30W UV lamp irradiation for 720 h, decreased by 12% compared with the pure polyvinyl chloride (PVC) film, and its photodegradation performance was greatly improved.

Photodegradable materials must be exposed to light and have a long degradation period, while most film materials are not exposed to natural light for a long time after disposal and it is difficult to ensure the degradation conditions required for photodegradable film materials, which greatly limits the large-scale application of photodegradable film materials.

2.2. Hydrodegradation

Hydrodegradable plastic is a kind of plastic that can self-degrade by hydrolysis. The essence is the presence of hydrolyzable covalent bonds in degradable plastics, such as esters, ethers, anhydrides, amides, carbamides or ester-amide groups [45], which can achieve dissolution when the plastic encounters water [51][52]. Water activity, temperature, pH and time are the key factors affecting the efficiency of hydrolysis [53].

Polyvinyl alcohol (PVA) is a water-soluble polymer with a carbon chain as the main chain and a large number of hydroxyl groups on the side chain [54][55]. It is non-toxic, easily processed, biodegradable, has good mechanical properties [56][57], and can be mixed with natural polymeric materials such as polysaccharides and proteins to improve its properties [58][59][60]. Mainly used in the packaging of water-soluble products, the buyer can do not touch the product in the process of using the product, safe and at the same time make the use of the product more convenient. However, the resistance of PVA film to water is very low, usually in a very short period of time can be completely dissolved [61]; therefore, if it is widely used in the field of packaging needs, it needs to be modified for water resistance.

Lv et al. [62] investigated the time-dependent hydrolysis behavior of polylactic acid (PLA) and starch/PLA composites. The results showed that the presence of starch may induce hydrolysis to occur at the interface between starch and PLA. In addition, starch can slightly slow down PLA hydrolysis without affecting the degree of PLA hydrolysis. **Table 2** shows the water degradation of several common biodegradable polyesters in different water environments.

Table 2. Hydrologic degradation of several typical biodegradable polyesters in different water environments. Data from [63].

Material	Conditions	Weight Loss %	Number-Average Molecular Weight (Mn)	Mechanical Properties
Polylactic acid (PLA)	Seawater	<2	96.60 × 10^3 to 83.85 × 10^3	No significant change
	Germicidal water	<2	96.60 × 10^3 to 67.98 × 10^3	
Poly (butyleneadipate-co-terephthalate) (PBAT)	Seawater	<2	46.67 × 10^3 to 20.31 × 10^3	Total loss
	Germicidal water	<2	46.67 × 10^3 to 16.02 × 10^3	
Poly (butylene succinate) (PBS)	Seawater	<2	41.56 × 10^3 to 30.11 × 10^3	Total loss
	Germicidal water	<2	41.56 × 10^3 to 18.63 × 10^3	
Polycaprolactone (PCL)	Seawater	32	77.79 × 10^3 to 77.09 × 10^3	Total loss
	Germicidal water	<2	77.79 × 10^3 to 14.82 × 10^3	

l contains in loss of certain properties (such as integrity, molecular mass, structure or mechanical strength) and/or fragmentation [64][65]. Heat can change the oxidation mechanism of plastics, and higher temperatures can improve the degradation of plastics [66][67]. **Figure 2** shows the mechanism of thermal oxidative degradation. Thermally oxygen degraded plastic is also very difficult to degrade completely in most cases due to the conditions.

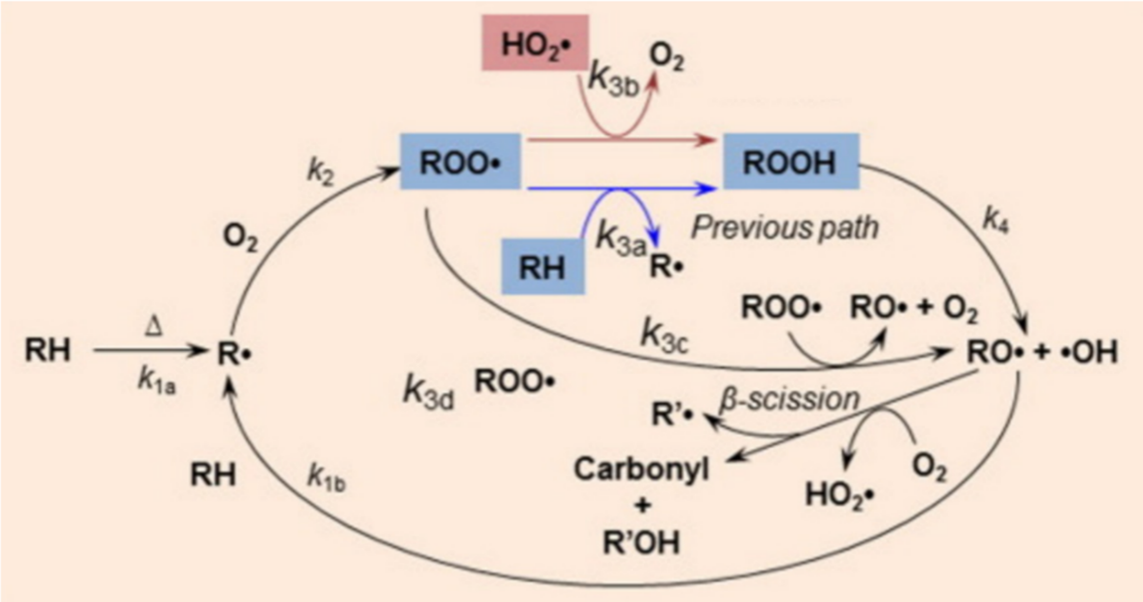

Figure 2. Auto-oxidation scheme of polymer. Reprinted from Ref. [68]. Copyright (2016), with permission from Elsevier.

Gaurav et al. [69] prepared two high-density polyethylene/polylactic acid blends with and without the addition of a compatibilizer and a pro-oxidant using a melt blending technique. The results showed that the addition of the compatibilizer led to a significant improvement in the mechanical properties of the blends and the addition of the pro-oxidant led to an improvement in their oxidative degradation properties.

2.4. Biodegradable

Biodegradable plastics are those degraded by naturally occurring microorganisms under natural conditions such as soil and/or sand, and/or specific conditions such as composting or anaerobic digestion or aqueous cultures, and ultimately degrade to environmentally benign biomass, CO_2, CH_4 and H_2O [70][71][72]. **Figure 3** shows the biodegradation mechanism. Biodegradable plastics have stable performance and can be completely degraded and returned to nature in a short period of time under composting conditions [73].

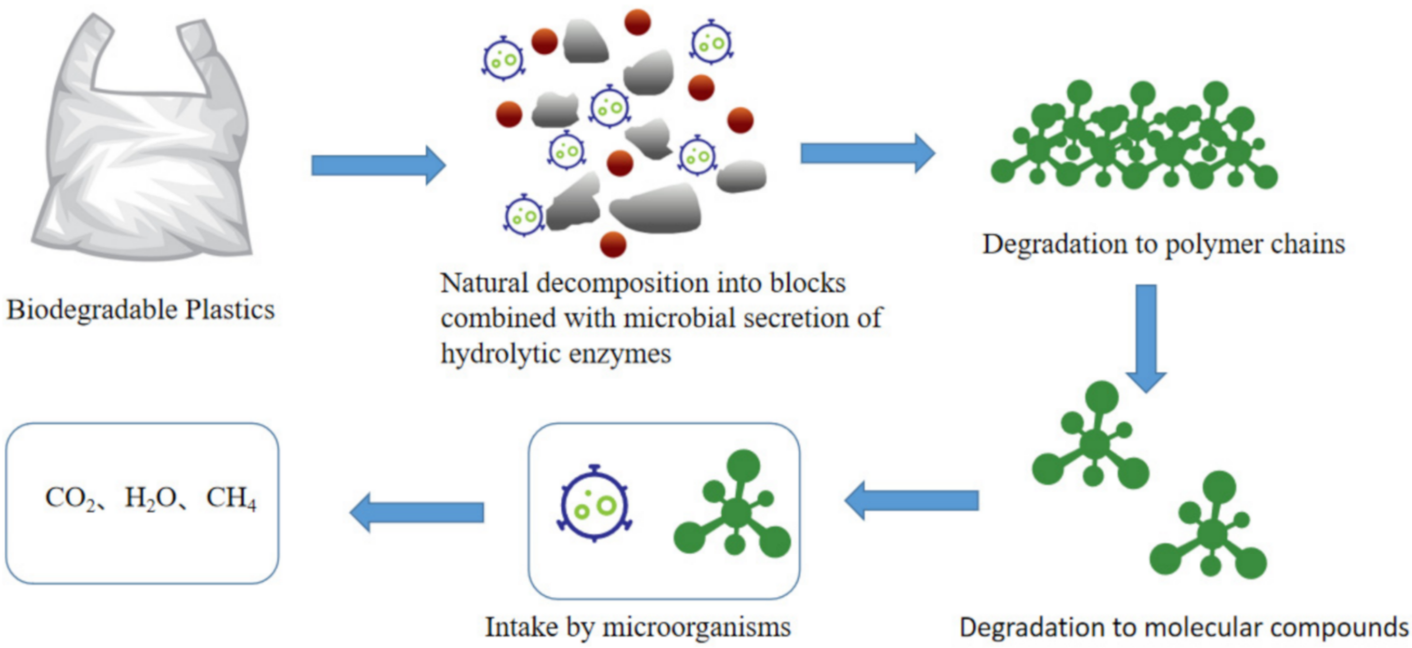

Figure 3. The mechanism of biodegradation.

Current research shows that animals, plants, microorganisms and enzymes all have some ability to degrade plastics [74][75]. **Table 3** shows the biodegradation of common plastics. Among the many ways to change the properties of plastics, biodegradation of plastics is one of the inevitable environmental processes for plastics to enter the environment, and it is also an in situ, green, relatively low-cost and low-technology way to treat plastic waste.

Table 3. Biodegradation of common plastics.

Material	Conditions	The Result of Degradation	References
Polyethylene	Degradation of high-density polyethylene with *Aspergillus flavus* PEDX3 strain for 28 days	Molecular weight reduction	[76]
Polypropylene	Degradation of polypropylene with microalgae *Spirulina* sp. for 112 days	Decrease in mechanical strength and relative molecular weight	[77]
Polystyrene	Degradation of polystyrene with *Achatina fulica* for 4 weeks	The mass loss was 30.7% on average, forming a functional group of oxidation intermediates	[78]
Polyethylene terephthalate	Degradation of polyethylene terephthalate with microalgae *Spirulina* sp. for 112 days	Decrease in mechanical strength	[77]
Polylactic acid	Degradation in accordance with ISO 17556	15% of Polylactic acid is degraded	[79]

Among various degradable mechanisms, biodegradation is more complete and faster than other degradation mechanisms, and the degradation products are harmless. Biodegradable plastics can be composted together with organic waste, thus eliminating the manual sorting step compared to general plastic waste, greatly facilitating waste collection and disposal, thus making composting and harmless disposal of organic waste into reality [80]. Biodegradable packaging film materials are green, environment-friendly and resource-saving compared with traditional film materials, thus gradually becoming a research hotspot in the packaging industry, the development of biodegradable packaging film is an effective way to fundamentally solve “white pollution”.

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