Development of Biodegradable Packaging Materials from Bio-Based Raw Materials

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Abstract: Seaweed, creatures and cellulose based packaging materials are biodegradable and promising natural polymer and their films can be prepared from bio-based raw materials. This article reviews the basic information and recent developments of both seaweed, creatures, cellulose and plant based biopolymer materials as well as analyses the feasible formation of seaweed/creatures/cellulose/plant based biodegradable packaging films which possesses excellent mechanical strength and water resistance properties. Moreover, bio-based packaging films can prolong a product's shelf life while maintaining its biodegradability. Additionally, the films show potential in contributing to the bio-economy. These type of bio-based materials exhibit interesting film-forming properties that can be used in biomedical application and for making composites for packaging. Bio-based films can be used for the large-scale applications in food packaging in place of synthetic petroleum based non-degradable packaging. Bio-based films have the potential to be used in textile and decoration paper industries also. Currently, bio packaging gains huge attention to the scientist and general people because this type packaging materials are environmental friendly products. Some of the viewpoints are highlighted for future developments and applications.

Keywords: Packaging films, Seaweed, Cellulose, Plant-based materials, Biodegradibility.

1. INTRODUCTION

Comestible and degradable polymers are a type of material made from renewable and comestible ingredients such as polysaccharides, proteins and lipids, as degrade more readily than non-renewable petroleum-based polymers. This polymer has been used in food and biomaterial products or as packaging films in food coatings or wrappings due to its degradable properties and preservative capabilities, which are of great benefit to the environment and health. Polysaccharide is promising edible polymer as, it is cheap, readily available, biocompatible and environmentally friendly. Cellulose, starch and chitosan are natural renewable sources of polysaccharide, which has been commonly applied as an edible film in agricultural products for the purpose of prolonging the shelf life of fresh fruits and vegetables, reducing the oil/fat absorption in fried food and hindering the loss of food flavor [1]. In the furtherance of modern medicine, the demand for biomaterials has increased dramatically since the last decade. Biomaterial can be defined as "Any component or combination of components natural or synthetic in basis, that can be used any time, as a part or whole of a system which treats or replaces any tissue, organ or function of the body and systematically engineered to act together with biological system to direct medical healing action" [2].

Seaweed based raw materials are also using the packaging properties. Alginate, carrageenan and agar are common seaweed-derived products that exhibit interesting film forming properties. Generally, seaweed is plant like organism mostly found in coastal areas. Seaweed refers to several species of marine plants and algae which may be microscopic, multicellular that grows in the ocean as well as in river, lakes and other water bodies. Since the mid-19th century, they are distinguished into four major groups based on pigmentation: Blue algae, green algae, brown algae or red algae and none are known to be poisonous [3].

In recent years, economic development of the medical wound dressing market has been observed. Many polymers have been extensively investigated as potential materials for wound covering, realizing that an ideal wound dressing should present several specific properties for the intended application, including capacity to absorb wound exudates, maintenance of the moisture, protection from secondary infection, provision of adequate gaseous exchange, provision of thermal insulation free from particulate or toxic contaminants, mechanical durability, flexibility, and

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non-toxicity. The use of natural polymers such as creatures based (chitosan, collagen and gelatin) in the construction of wound healing devices is quite attractive, mostly because of their biocompatibility, possibility of various chemical modifications, and relatively low cost of production. The functional properties of the novel materials indicate a potential for packaging applications [4].

Cellulose is a commercially important polysaccharide. Since cellulose is capable of forming hydrocolloids in a suitable solvent system, it is an excellent film-making material. Cellulose film has been reported to have higher water and microwave heating resistances. Moreover, cellulose has attracted significant interest as polymer matrix composites. In addition, it also blends with other hydrocolloids, which widens its applications. Past researches have shown that the addition of cellulose and/or its derivatives into a polymeric matrix can increase the tensile strength and rigidity of the film [5].

From the last few years, for packaging purpose and for the treatments of humankind, seaweed, creatures and cellulose has become very popular from laboratory to industrial scale. Since seaweed blends/ creature blends/ cellulose blends are a new research area, a literature review that covers the published reports of edible packaging films made of seaweed and cellulose is necessary. Hence, this paper reviews the fundamental knowledge and current state of research into seaweed and cellulose as edible polymeric materials. Furthermore, this review also analyses the compatibility of seaweed and cellulose based on the structure and properties of the composite film, as well as the environmental (biodegradable) and economic aspects of this packaging film. Future development of seaweed/creature/cellulose packaging film is discussed as well.

2. TYPE OF BIO-BASED RAW MATERIALS

Most of the world's packaging materials are petroleum-based plastics which are not biodegradable. The current global consumption of non-biodegradable plastics is more than 200 million tons, with an annual growth of approximately 5%, which represents one of the largest focus areas for crude oil. Furthermore, this situation demonstrates the increasing need to use alternative bio-based raw materials. Until now, petrochemical-based plastics such as polyethylene, polypropylene, polyethylene terephthalate, polyvinylchloride, polystyrene, and polyamide have been increasingly used as packaging materials because of their wide availability at relatively low cost and their good mechanical performance. Nowadays, their use has to be restricted because they are not biodegradable, and therefore they pose serious ecological problems. Plastic packaging materials are also often contaminated by foodstuffs and biological substances, making recycling of these materials impracticable and often uneconomical. This has prompted much research in recent years into the development of products such as bio-based polymers as could replace these non-biodegradable materials in the packaging industry. Many bio-based polymers such as cellulose, starch, alginate, and chitosan have been studied by many researchers [6]. The interest in using natural fibers such as different plant fibers has increased dramatically during last few years. With regard to the environmental aspect it would be very interesting if natural fibers like jute, banana, and coir could be used instead of artificial fibers as reinforcement in some structural applications. Investigations carried out in this field have shown that stiffness, hardness and dimensional stability of plastics have also been improved by incorporation of lignocellulosic fillers. Natural fibers have many advantages compared to synthetic fibers; for example they have low cost, low density, low abrasion, competitive specific mechanical properties, reduced energy consumption, they are biodegradable. In addition, they are renewable raw materials and have relatively high strength and stiffness and cause no skin irritation [7].

2.1. Seaweed Based

Seaweed based polymer has been broadly utilized as a part of bio packaging, food and biomedical applications in terms of their known biocompatibility, bio absorbability, biodegradability and nontoxicity. These polymers can be degradable without enzymatically. Commonly three seaweed-derived hydrocolloids polysaccharides are available in nature that has diversified application as bio polymeric film such as alginate, carrageenan and agar. However, there are others seaweed hydrocolloids polysaccharides, which are less significant like *mannitol*, *fucoidan* and *funoran.Gracilaria* and *Gelidium* can create gelatin like substances primarily known as agar. Red Algae *Kappaphycus* and *Betaphycus* are the important sources of carrageenan. The film forming and binding ability makes it suitable for the use of packaging material [8].

2.1.1. Alginate

Alginates, a naturally occurring polysaccharide, were obtained from marine brown algae (*Sargassum*). Algae are one kind of seaweed. Algae, like any other plant, require three magical ingredients: (i) light, (ii) water, and (iii) nutrients. Alginates are linear copolymers of b-(1-4)-linked D-mannuronic acid and a-(1-4)-linked L-glucuronic acid units, which exist widely in many species of brown seaweeds[9].

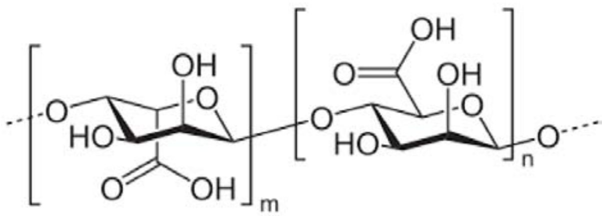

Figure 1: Chemical structure of Alginate.

Alginate has been used in a wide range of industries such as food, textile printing, paper and pharmaceuticals, and for many other novel end-uses. As a water-soluble polymer, alginate is an excellent gel-forming material capable of holding a large amount of water. Alginate has been utilized to develop biodegradable or edible films due to its unique colloidal properties such as thickening, stabilizing, suspending, film forming, gel producing, and emulsion stabilizing properties [10].

Alginate is the most widely used material for bio polymeric film. It is a natural polysaccharide derived from marine plants. Alginate is of interest as a potential biopolymer film component because of its unique colloidal properties, which include thickening, stabilizing, suspending, film forming, gel producing, and emulsion stabilizing. The main objective of this work was to develop a renewable and biodegradable alginate based Nano composite films by incorporating NCC for food-packaging applications [10].

2.1.2. Carrageenan

Carrageenan extracted from red algae specifically from the Rhodophyceae family. It's consisting of linear-sulfated polysaccharides of D-galactose and 3,6-anhydro-D-galactose (3,6-AG). This is specific type of seaweed is common in the Atlantic Ocean near Europe, North America and Britain. It is a complex mixture of five distinct polymers designated ι-, κ-, λ-, μ-, and ν- carrageenan. Among those, three major types lambda(λ), kappa(κ) and iota(ι). There sulfate contents are 41%, 33% and 20%. Classification of carrageenan was made based on its solubility in KCl. λ-, ι- and κ-carrageenan membranes reveal better mechanical properties [11-12].

In order to meet the new renewable resources for the production of edible and biodegradable material, carrageenan can be used as an interesting alternative to produce comestible films and coatings, food and beverage, bio packaging. This opaque or colored packaging is widely used in food containers, trays, cups, wraps and other packaging designed to preserve light or UV sensitive products. But pure carrageenan films limit its use for food packaging [13].

2.1.3. Agar

Agar (AG), a gelatinous polysaccharide that is extracted from marine red algae such as *Gelidium*and *Gracilaria spp*., is one of the most promising polysaccharide for developing biodegradable packaging films. It has been widely used for the preparation of bio basedfilms due to its high mechanical strength and moderate water resistant properties. Moreover, AG based films were found to be heat-sealable. Upon considering the best characteristics of AG separately, we speculated that their combination would lead to better films than those formed by each individual material alone. Until recently, AG were studied alone or in combination with other

Figure 2: Alginate based packaging materials.

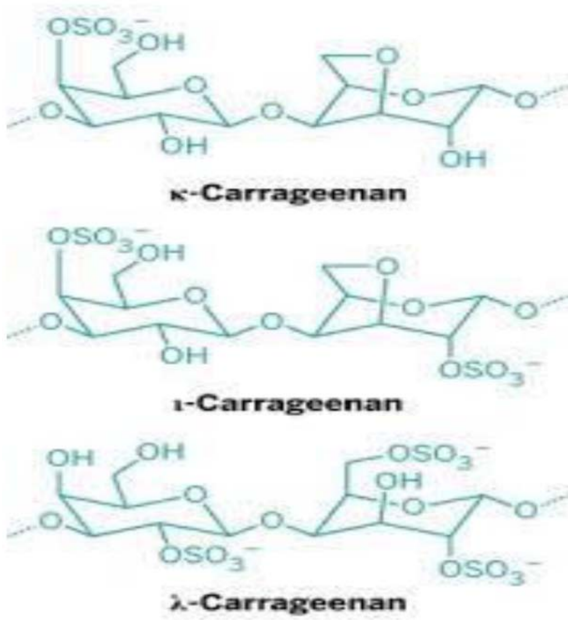

Figure 3: κ, ι, λ-Carrageenan structure.

biopolymers for edible films and coating applications, however, the fabrication of the bio composites from cold water fish skin gelatin and agar has not been elucidated. Hence, the objectives of the present work were to examine the effect of different ratios of AG on the functional characteristics of the blendfilms such as mechanical properties, water vapor permeability (WVP), film solubility, and transparency. The films were also characterized in their thermal stability (TGA) and morph structural (FTIR, SEM and AFM) features. AG bio composite films exhibited a high UV barrier property that implies the blend films have a high potential for being used to protect food products from UV light induced photo-chemical reactions. Meanwhile, TGA analysis indicated that the presence of AG improved thermal stability. The blending AG rendered films of homogeneous structure, due to the high compatibility of both polymers, as revealed by microscopy images. AG is a suitable approach to obtain environmentally friendly packaging films. However, further studies are needed to investigate potential performance improvement for industrialized use of the film [14].

2.2. Creatures Based

Proteins, polysaccharides and lipids or mixtures of these compounds may be used for production of biodegradable, edible films, and coatings. The properties and possibilities of applications of such materials have been reviewed. The edible films can be used not only as packaging materials but also as carriers of food additives, e.g., antioxidants, antimicrobials, flavoring agents, and pigments [15]. The main creatures based are collagen, gelatin and chitosan. Collagen is an enzymatically degradable polymer which accounts for about 20-30% of total mammalian body proteins. Gelatin is a nature biopolymer. Cattle bones, hides, pig skin and fish are the principal commercial sources of gelatin. It is relatively low-cost polymer and it is an edible protein where all essential amino acid except tryptophan are available. Chitosan are regarded as the second most abundant natural polymer after cellulose found in the exoskeletons of crustacean and insects as well as some bacterial and fungal cell walls [16].

2.2.1. Collagen

Biodegradable polymers are polymers which completely degrade by the action of biological organisms such as proteins (for example, collagen hydrolyzate) and carbohydrates. This type of biopolymer is completely biodegradable, consequently, it is effective for the volume reduction of synthetic polymers waste by partial degradation. However, another type of biodegradable polymers are not completely degrade, which is called the bio-disintegrable polymers. Industrial and technological applications reported for plastics are as follows: packaging (41%), construction of buildings (20%), electric insulation (9%), automobile parts (7%),

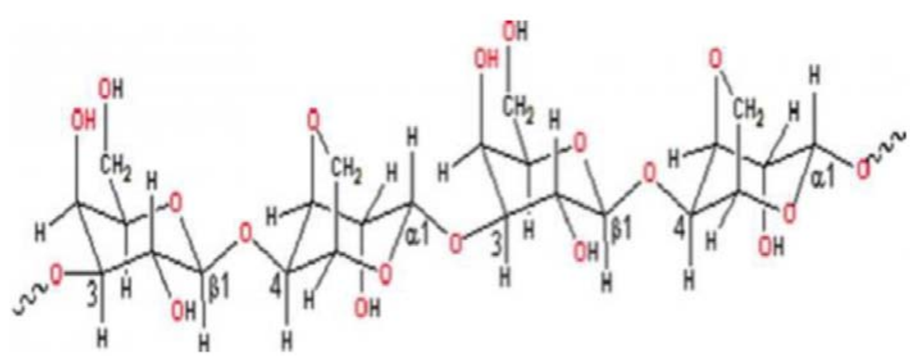

Figure 4: Agar structure.

agriculture (2%), and miscellaneous (21%). Thus, the increasing use of plastics as packaging materials and their nonbiodegradability has raised an environmental awareness for the development of low cost environmentally friendly biodegradable materials through polymer modification. One of the ways to reach this goal is by using renewable, natural, low cost, and easily available biopolymers such as starch,5,6 cellulose,7 protein (collagen). The majority of thermoplastics currently used as packaging materials are based on polyolefin, such as high density polyethylene (HDPE), low density polyethylene (LDPE), and polypropylene (PP). Moreover, for low value items such as shopping bags, agriculture mulch films, and food packaging (foils and thermoformed items) recycling would be neither practical nor economical. Blends of synthetic and natural polymers have been used in recent decades to develop new materials, which are called bio-artificial polymeric materials. Their capability of combining good physical and mechanical properties with biocompatibility characteristics was used for the purpose of making new materials for biomedical applications. Collagen hydrolyzates (CH) was previously prepared by alkaline and enzymatic hydrolysis of chrome leather waste which condensate with long-chain carboxylic acids and their chlorides at atmospheric pressure. A number of authors have reported on the chemical and enzymatic treatment of leather solid waste and the composition of leather industry waste are polymeric materials which are biodegradable [17].

Collagen is the most widely occurring collagen in connective tissue. Interstitial collagen molecules are composed of three a-chains intertwined in the so-called collagen triple-helix. This particular structure, mainly stabilized by intra- and inter-chain hydrogen bonding, is the product of an almost continuous repeating of the

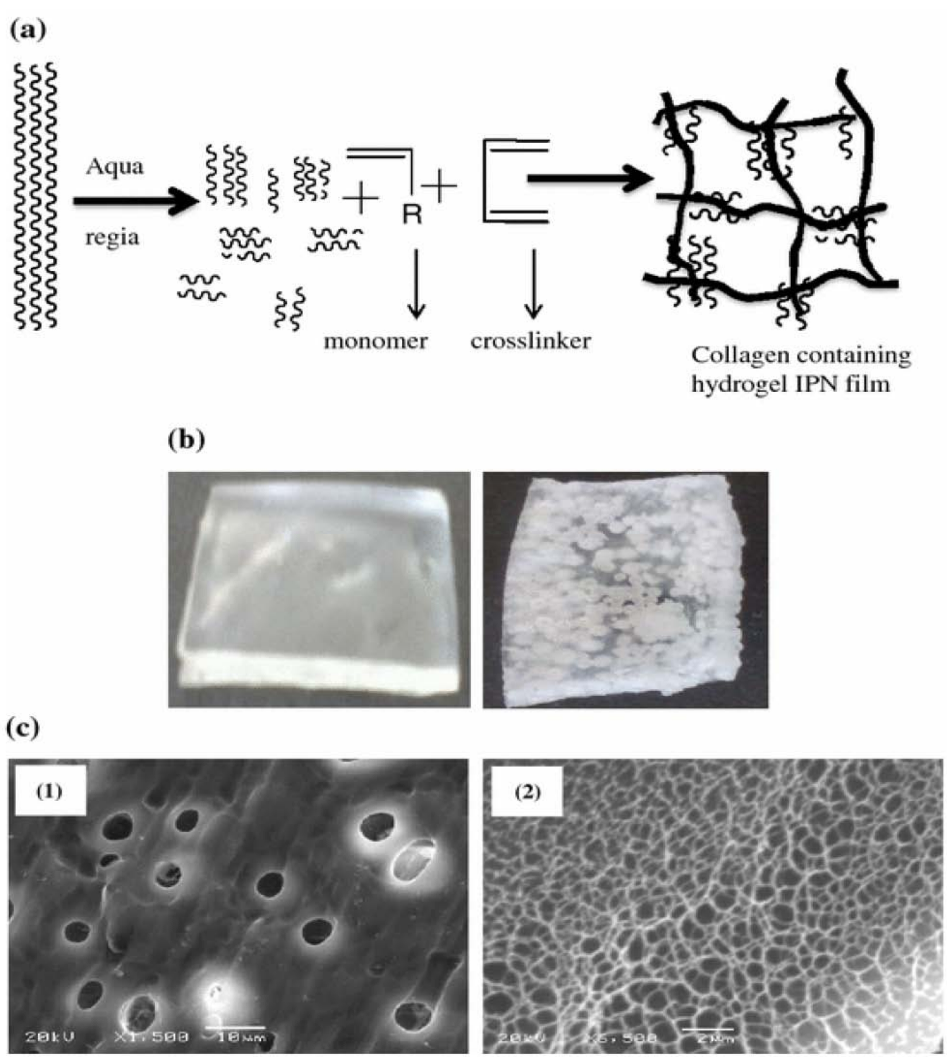

Figure 5: Collagen based packaging films.

Gly-X-Y- sequence, where X is mostly proline and Y is mostly hydroxiproline. Only the very short N- and C-terminal regions, called telopeptides (15e26 amino acid residues), do not form triple helical structures as they are largely made up of lysine and hydroxyl sine (Hyl) residues, as well as their aldehyde derivatives, in both intra- and inter-molecular covalent cross-links. Four to eight collagen molecules in cross-section are stabilized and reinforced by covalent bonds to constitute the basic unit of collagen fibrils. Thus, the typical strong, rigid nature of skins, tendons and bones is due to the basic structure formed by many of these cross-linked collagen fibrils. Collagen, a well-known febrile protein obtained from connective tissues of animals, has received considerable attention because of its abundance and many important biological functions such as tissue formation and cell adhesion with excellent biodegradability and biocompatibility. Collagen has been widely used as biomaterials in a variety of applications suchas wound dressing, scaffold materials for tissue engineering, drug delivery systems, orthopedics, and preparation of biopolymer composites. The main objective of the present study was to prepare collagen ternary blend hydrogel film by including antimicrobial additives such as silver nanoparticles (AgNPs) and grapefruit seed extract (GSE) to develop functional bio hydrogel films for food packaging application.

On the other hand, enzyme-hydrolyzed collagen plays an increasingly important role in various products and applications. Its different properties and functionalities benefit the end consumer now in ways which were not present ten years ago. Over the past decade, a large number of studies have investigated enzymatic hydrolysis of collagen for the production of bioactive peptides. Besides exploring diverse types of bioactivity, studies focused on the effect of oral intake in both animal and human models have revealed the excellent absorption and metabolism of Hyp-containing peptides. Already this biomaterial has seen significant use, so that outstanding impact of collagen as biomaterial will be far better than before [18-19].

2.2.2. Gelatin

Currently, the idea of replacing commonly used plastics prepared from synthetic polymers with biodegradable plastics prepared from natural polymers is increasingly interesting. Gelatin is one of the natural polymers, which are now widely studied. It is a high molecular weight polypeptide composing of amino acids mainly glycine (27%), hydroxyproline, and proline (25%). Because its molecules are tightly bound with hydrogen bonds, pure gelatin films are normally brittle. Commercially, gelatin is presented as a colorless or slightly yellow, transparent, brittle, practically odorless, tasteless sheets, flakes, or coarse powder. Their uses include not only food (confectionery, jellies, and ice cream) and pharmaceutical technology but also manufacturing of rubber substitutes, adhesives, photographic plates and films, matches and clarifying agent. Furthermore, the polar groups present in its structure cause a gelatin film to have high moisture absorption [20].

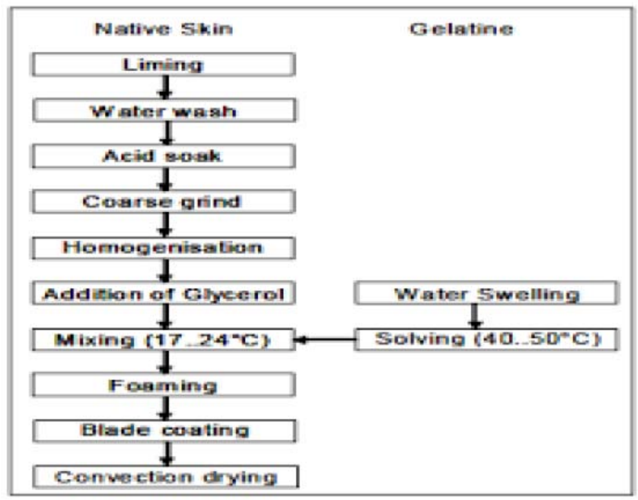

Figure 6: Gelatin absorption.

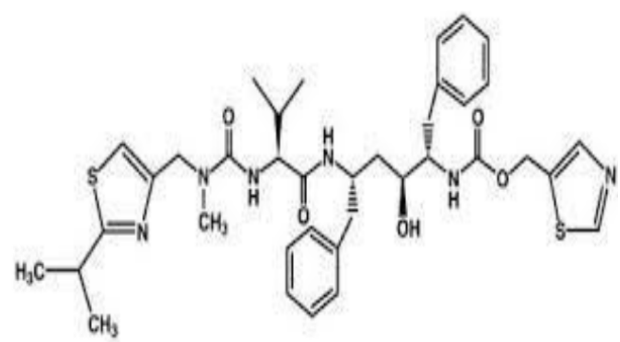

Figure 7: Gelatin structure.

Gelatin, a natural protein, is insoluble in water and is solubilized by hydrolysis. It is obtained by selective hydrolysis of collagen, which is a fibrous material that occurs in skin, bones and connective tissues of animals. This heterogeneous product which is a mixture of three extended polypeptide chains, super-coiled together to form a right-handed triple helix. The triple-helix structure is stabilized by the formation of inter-chain hydrogen bonds between C¼O and N-OH groups. Gelatin is biocompatible, biodegradable,

Figure 8: Gelatin based films.

edible, and soluble at the body temperature, which undergoes gelation at temperatures just above ambient. The properties of gelatin depend on the major protein constituent derived from the breakdown of collagen [21].

Gelatin possesses priority for various applications in medical sciences. Gelatin, a natural polymer is used as a tissue engineering scaffold. Gelatin based sponges composed of gelatin and polysaccharides have the potential for wound dressing materials. Cross linked gelatin sponges have also been investigated for their application as a component of artificial skin or tissue transplants to promote epithelialization and granulation tissue formation in wound. The swelling rate and the equilibrium swelling of gelatin by casting are also studied. Compression molded gelatin/starch (1:1 w/w) blend shows improved mechanical properties as compared to gelatin. Gelatin graft copolymer macromolecules are formed when methyl methacrylate is polymerized in aqueous medium in the presence of gelatin. Research has been previously done to explore the irradiation effects on gelatin. It is reported that, gamma radiation increased the crosslinking between protein chains which increase the mechanical properties of the film. Observed that gamma radiation enhance the tensile properties (TS) of the pectin and gelatin based films [22].

2.2.3. Chitosan

Chitosan, the second most abundant natural biopolymer after cellulose, composed mainly of b-(1,4)-linked 2-deoxy-2-amino-Dglucopyranose units. It is the major component of the shells of crustacean such as crab, shrimp, lobsters, and crawfish. Chitosan contains a large number of hydroxyl and amino groups. These two functional groups provide several possibilities for grafting of desirable bioactive groups. Chitosan could be used to prepare biodegradable packaging materials [23].

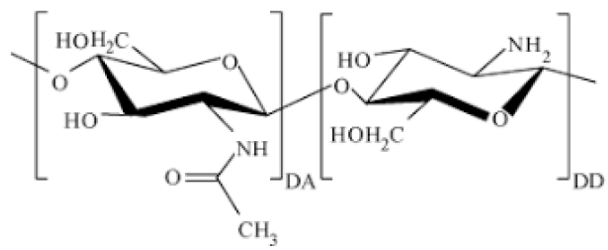

Figure 9: Chemical structure of Chitosan.

Chitosan is a linear bio-polyamino saccharide, which is obtained by alkaline deacetylation of chitin, similar in structure to cellulose. Both are made by linear b (1!4)-linked mono saccharaids. However, an important difference to cellulose is that chitosan is composed of 2-amino-2-deoxy-b-D-glucan combined with glycosidic linkages. Chitosan dissolved in weak organic acids such as acetic acid can be cast into membranes, fibers, and sponges. Chitosan and its oligomers are well known for their interesting biological properties, which have led to various applications. Chitosan membrane-based wound products have been investigated both in laboratory animals and humans, but are still at the early stage of development. Graft copolymerization has been studied for many years especially in cellulose using electromagnetic radiation

Figure 10: Chitosan based film.

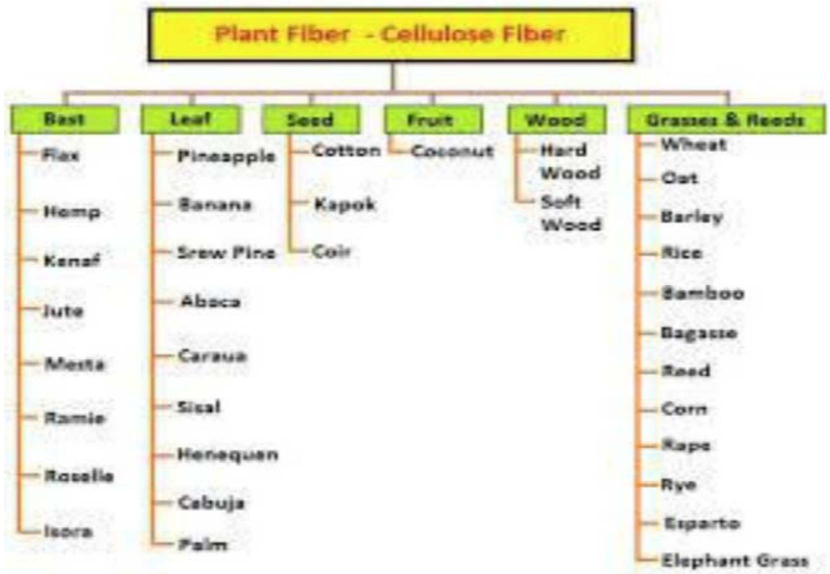

Figure 11: Cellulose obtain form plant fiber.

such as gamma ray, ultraviolet light and free radical initiators. In the present work UV radiation or photocuring is employed to improve the properties of chitosan/urea film [24].

Since chitosan is non-toxic and biocompatible with the human physiological system, it has been investigated as biomaterials in the fields such as biomedicine, pharmacology and biotechnology. Chitin and chitosan have already been used in agricultural, food, industrial and medical fields. It also acts as flocculent for the treatment of wastewater. Chitosan is non-toxic, biodegradable, bio functional, biocompatible and was reported by several researchers to have strong antimicrobial and antifungal activities. Chitosan films have been successfully used as a packaging material for the quality of preservation of foods [25-26].

2.3. Cellulose Based

Cellulose is an organic compound with the formula $(C_6H_{10}O_5)_n$, a polysaccharide consisting of a linear chain of several hundred to over 10000 β(1→4) linked D-glucose units. Cellulose is the most abundant organic polymer in the biosphere. It is the main constituent of plants; moreover, it is lightweight, biodegradable, and an available natural resource. The interest in using cellulosic materials as the main components in the manufacture of biodegradable packaging materials is increasing day by day [27].

2.3.1. Plant Cellulose

Cellulose is the most abundant organic polymer. It is the main constituent of plants. Cellulose is a homopolysaccharide, which is the main constituent of wood and is composed of b(1!4) linked glucopyranose units. Each repeating unit contains three hydroxyl (AOH) groups. These hydroxyl groups have the ability to form hydrogen bonds. Cellulosic materials have good mechanical properties. They are also light weight, biodegradable, and widely available. A major interest is going on to use cellulosic materials as main components in the manufacture of biodegradable packaging. Plant-derived cellulose is already being used extensively in the paper and textile industries. So plant cellulose based films related to applications in the packaging sectors. This is a new door to preparing more hydrophobic biodegradable films for packaging applications by grafting functional monomers and filling film matrices with cellulose Nano whiskers [28-29].

2.3.2. Bacterial Cellulose

Bacteria cellulose (BC) as a microbial cellulose. It is formed by aerobic bacteria such as acetic acid bacteria of the genus Gluconacetobactor. BC a purely cellulose polymer. The chemical treatment is not needed for cellulose isolation. It is interesting material because it has both excellent mechanical strength and degradability compared to green plant cellulose. It has ribbon-shaped fibrils. There are approximately 20-100 nm in diameter and are composed of much finer 2-4 nanofibrils. It's also commonly referred to as bacterial nanocellulose (BNC). They also possess a higher water holding capacity, a higher degree of polymerization (4000-6000) and a finer wed-link network than the cellulose form plants. BC has a wide range of applications form food to functional materials.

Such as electronic devices, wound dressing, additives in paper, membrane filters and more. So, it's also used by the biodegradable films [30].

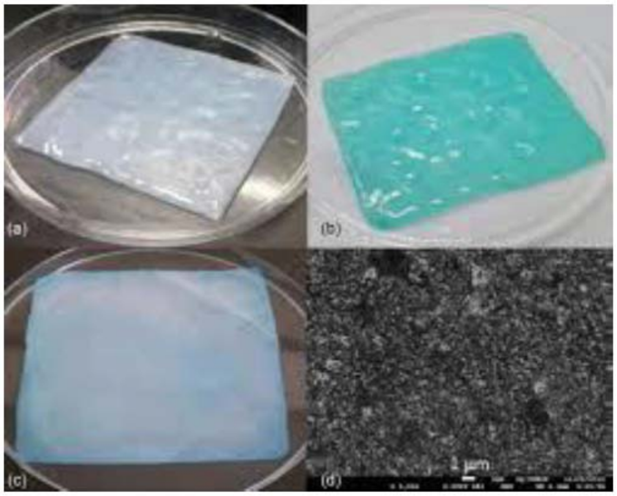

Figure 12: Biodegradable films by bacterial cellulose.

2.3.3. Micro/Nanoscale Cellulose

Nanocellulose (NC) is also a cellulose derivative composed of a nanosized fiber network, which determines the product properties and its functionality. NC fibers are very interesting nanomaterials for production of cheap, lightweight, and very strong nano-composites. NC fibers have nano sized diameters (2-20 nm), and lengths ranging from a few hundred nanometers to a few micrometers. NC is expected to show high stiffness because the Young's modulus of the cellulose crystal is as high as 134 GPa. A considerable amount of research has been done on the isolation of these nanofibers from plants for use as fillers in bio composites [31].

Figure 13: Nano cellulose by nature.

2.4. Plant Based

We are more concerned about our environment. As we are polluting our environment more than any other time, our existence is in danger. To lead a healthy and happy life, we should keep our environment pollution-free. The main reason of this pollution as most of them are non-biodegradable. To reduce the applications of non-biodegradable materials, we are trying to find new materials which are biodegradable. Now we are considering natural fiber as biodegradable materials for different applications. The well-known natural fibers are bagasse, jute, coir, banana, cotton, sisal, oil palm, and kenaf [32].

2.4.1. Bagasse

Whey protein isolate (WPI) is a valuable by-product of the cheese production and it consists of protein content >90%. It has been studied for film formation and coating application. Several authors have investigated the properties of whey protein isolate films as transparency, flexibility, odorless, excellent barrier to oxygen transmission and providing moderate mechanical properties. However, the limitation of whey protein isolate films to reach the expansive commercial applications was due to low mechanical and high water vapor permeability properties because of their hydrophilic nature. Therefore, incorporation of cellulose Nano crystals (CNCs) in a biopolymer to improve these properties may be useful because CNCs have strong hydrogen bonding and a high surface area. Thailand is the second largest sugar exporter in the world and>20 million tons of sugarcane bagasse (SCB) is created annually. Generally, SCB is used as boiler fuel to produce steam which in turn is used in the sugar production industry. Moreover, SCB is also utilized in ethanol and pulp production. SCB consists of approximately 40–50% cellulose. Therefore, SCB is an interesting source for CNC extraction. Acid hydrolysis is the most well-known process for CNCs extraction. It breaks and removes the disordered and amorphous regions of cellulosic fibers leaving well-defined crystals in the form of CNCs. SCB produced using acid hydrolysis and to investigate the effect of cellulose Nano crystals from sugar bagasse on the properties of Nano composite whey protein film to determine its further utilization as a food packaging material [33].

Sugarcane bagasse (SB) contain appreciable amount of cellulose and hemicellulose, which can be

Figure 14: Bagasse fiber and their chemical structure.

depolymerized by chemical or enzyme cocktails into simple sugar monomers (glucose, xylose, arabinose, mannose, galactose, etc.). Such sugar streams obtained from SB and SL can be converted into bioethanol and value-added products of commercial significance, which has joint economic important. Harnessing bagasse for industrial purposes could provide a sustainable and economic solution for the production of bio based, value-added products such as ethanol, xylitol, organic acids, industrial enzymes, and other products. The efficient utilization of sustainable resources will assist in improving the socioeconomic status of developing countries, creating employment opportunities and improving the environment. Among sugarcane-producing countries, Brazil is the top producer, with 625 million tons of sugarcane in 2011, followed by India and China (http://www.unica.com.br). Generally, 280 kg of humid bagasse is generated from 1 ton of sugarcane. Pandey *et al*. 4 reported that 50% of SB is used for energy generation within the plant and the rest remains unused in the environment. Therefore, the bioconversion of leftover bagasse into value-added products may have sustainable economic and strategic benefit. Summarizes the procedural steps involved in the application of SB for the production of various industrially important products of commercial significance (bio-products and bio refineries) along with its application for entrapment of microbial cells as an immobilized or growth support [34].

2.4.2. Jute Fiber

Efforts to exploit the possible use of lignocellulosic fibers like jute, sisal, coir, banana, and PALF1–15 as reinforcing composites have been extensive in recent years. Among these, jute fiber is most common because of its easy availability at low cost, especially in the eastern regions of India. For jute fiber to be an effective reinforcing constituent, it is essential that the fiber and the resin matrix have a good compatibility and bonding. To make them suitable reinforcing candidates with adequate bond characteristics for general applications, various chemical modifications of jute fibers have been attempted. Among the many surface treatments undertaken, the most economically viable one is alkali treatment [35]. Jute is an important natural fiber occupying second place in economic importance only after cotton. It is one of the most important fibers used for industrial applications. More importantly it is a commodity on which millions of households in some of the countries depend for their cash earnings. India, Bangladesh, China, Nepal and Thailand are the major producers of jute accounting for over 95% of the global output. The main traditional use of jute has been for the packaging market. Cloth, sacks and bags made of woven jute fabrics are widely used for transportation and storage of agricultural products, fertilizers, cement and some chemical products. The prime reasons of growing market of jute-nonwovens in technical applications are as following.

- High strength, modulus and dimension stability.
- Stiffness or moderate draping.
- Higher frictional properties.
- Coarseness.
- Easy rot / biodegradability and eco-friendliness.
- Higher moisture absorption characteristics.

- Good moisture absorption and breathability.
- Good bleach ability, dye ability and printability.
- Low cost.
- Jute is annually renewable and abundantly grown in India and neighboring countries.
- Utilization of mill wastes of jute i.e. use of short fiber for making jute based non-woven.

Figure 15: Jute Fiber based Bags.

It was found in study that jute is more environmentally sound and less costly to society than its competing synthetic material [36].

2.4.3. Coir Fiber

The natural fiber in the present study is coir, which is one of the most common well-known natural fibers on earth. It is the seed-hair fibers obtained from the outer shell or husk of the coconut, the fruit Cocus nucifera, a tropical plant of the Arecaceae (Palmae) family. It contains 36-43 % cellulose, and allied substance containing 41-45 % hemicellulose, 0.15-0.25 % lignin and 3-4 % pectin. The cell length is about 0.8 mm, spiral angle 41-45 o having tensile strength of 131-175 MPa, elongation at break of 15-40 % and Young's modulus 4-6 GPa is hydrophilic glucan polymer consisting of linear chain of 1,4-β bonded anhydroglucose unit, which contains alcoholic hydroxyl groups. The coir fiber is hydrophilic in nature and this is the most important disadvantage of this fiber [37].

Therefore, coir fiber is hydrophilic in nature which is the main disadvantage of this fiber. Attempts have been made continually to improve natural polymers both genetically and chemically. Thus, natural polymers are treated with different materials in different modes to

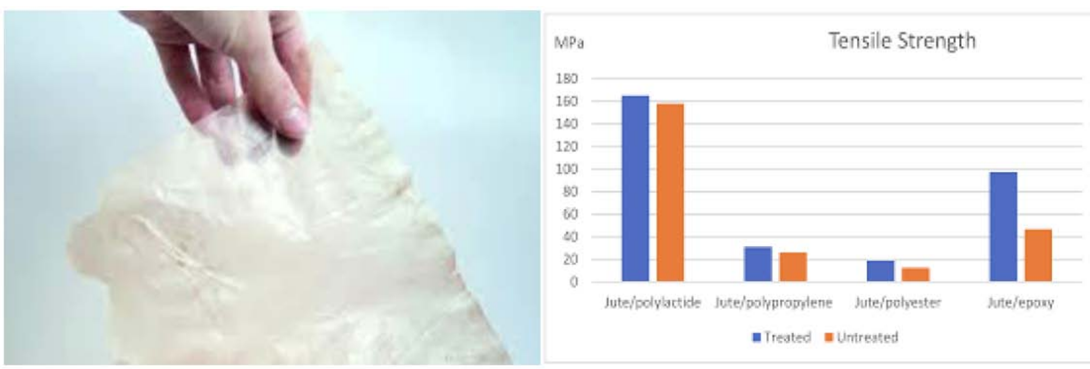

Figure 16: Biodegradable packaging film by jute fiber based and their tensile strength.

Figure 17: Coir/coconut fiber.

improve their strength, durability, and sustainability while possessing their inherent degradable character to maintain a pollution free environment. Various treatments have been used to improve the properties of natural fibers [38].

Figure 18: Coir based packaging film.

2.4.4. Banana Fiber

Banana fibers are a lignocellulic bast fiber that comes from the pseudo-stem of the banana plant. The fibers were harvested and processed in Sri Lanka by the Yaal foundation – a group associated with banana farmers. Banana, cooking banana and plantain (Musa spp.) are major starch staple crops of considerable importance in the developing world. They are consumed both as an energy yielding food and as a dessert. Most edible-fruited bananas, are seedless, belong to the species *Musa acuminata Colla*. *Musa balbisiana Colla* of southern Asia and the East Indies bears a seedy fruit but the plant is valued for its disease-resistance and therefore plays an important role as a “parent” in the breeding of edible bananas (Morton 1987). Bananas and plantains are today grown in every humid tropical region and constitute the 4th largest food crop of the world.

Farmers are made using waste from this industry using a semi-manual multifiber bundle decorticating machine that was donated by a German NGO. Natural fibers are not very abrasive to processing equipment and to the people handling them making them a perfect choice for our application. Characterizing the banana fibers’ mechanical properties can be difficult, as natural fibers tend to vary greatly depending on yield, locality, maturity of the plant, location of fibers within the plant, and the varying weather conditions such as rainfall and sun exposure. The individual fibers also present a number of physical characteristics such as variation in microfibril angles, variable cross-section and numerous flaws such as links, dislocations, nodes and slip planes. These can come from the plant’s growth process or from the fiber extraction method used (retting process vs. mechanical extraction). As the trees were not grown primarily for their fibers, there is not much that can be done about these flaws. In the present work, natural fiber composite panels using agricultural and plastic waste were made using compression molding. Also, different types of natural fibers available locally could be used instead of banana fibers produced from agricultural waste. For the fresh bananas to reach the consumer in the right condition, it must be marketed properly, bearing in mind the application of most suitable temperature and humidity as well as appropriate packaging and handling methods. Good handling during harvesting can minimize mechanical

Figure 19: Banana Fibers.

damage and reduce subsequent wastage due to microbial attack. Low temperature handling and storage are the most important physical method of postharvest management. The traditional packaging method for banana is nested packaging in which dried banana leaf and straw are used but the effectiveness of these packaging materials has not yet been investigated and reported.

It was shown that packaging materials can be successfully manufactured using local plastic agricultural waste [39-40].

Figure 20: Banana based packaging bags.

Figure 21: Banana based decoration paper.

3. BIODEGRADABLE PACKAGING FILMS

At the turn of the 20th century, most non-fuel industrial products like inks, dyes, paints, medicines, chemicals, clothing, and also plastics were made from biologically derived resources. However, 70 years later petroleum-derived chemicals to a major extent replaced these. Now, at the turn of the 21st century recent developments are raising the prospects that naturally derived resources again will be a major contributor to the production of industrial products. Currently, scientists and engineers successfully perform developments and technologies that will bring down costs and optimize the performance of bio-based products. At the same time, environmental concerns are intensifying the interest in agricultural and forestry resources as alternative feedstocks. A sustained growth of this industry depends on the development of new markets and cost and performance competitive bio-based products. A potential new market for these materials is food packaging, a highly competitive area with great demands for performance and costs. The polymers and materials used for food-packaging today consist of a variety of petroleum-derived plastic materials, metals, glass, paper and board, or combinations hereof. With the exception of paper and board, all of these packaging materials are actually based on non-renewable materials, implying that at some point, more alternative packaging materials based on renewable resources have to be found [41].

Biodegradable packaging is defined as packaging that contains raw materials originating from agricultural and marine sources. There are three such categories of biopolymers: (a) produced by chemical synthesis from bio-derived monomers; (b) produced by microorganisms; (c) extracted directly from natural raw materials, such as cellulose. Biodegradable packaging, made from entirely renewable natural polymers, could contribute to solving environmental pollution and creating new markets for agricultural products. Environmental problems can thus result from using non-renewable raw materials and accumulation of such non-biodegradable packaging. One solution is used of biodegradable materials made from polysaccharides, proteins, lipids, polyesters or a combination of these. Moreover, some additives such as colorants, antioxidants and antimicrobial agents can provide to packaging materials some functional properties that can prevent or delay microbial or chemical spoilage of food products [42].

Biodegradable polymeric materials have excellent and promising properties for the applications in packaging sectors. Besides the inherent biodegradability of these materials, there are other important properties like higher strength, transparency, low cost, and excellent film-forming properties *via* casting. It is, therefore, of industrial interest to increase the overall performance of these materials in order to generate value added product to counteract negative issues such as current pricing to enable their substitution for more well-established petroleum-based materials. Petroleum-based synthetic polymers are being using as packaging materials due to their excellent thermo-mechanical properties and low cost. But these materials are not biodegradable. A number of blends using bio-polymers can be the alternative of currently used synthetic polymeric materials. The most common and potential bio-polymers are starch, chitosan, alginate, gelatin etc.[43]. Bio-based packaging must serve a number of important functions, including containment and protection of food, maintaining its sensory quality and safety, and communicating information to consumers. A big effort to extend the shelf life and enhance food quality while reducing packaging waste has encouraged the exploration of new bio-based packaging materials, such as edible and biodegradable films from renewable resources. Biodegradable films can be used as a vehicle for the incorporation of food additives such as antioxidants and antimicrobial agents delivering them to the food surfaces where deterioration by microbial growth or oxidation often begins. The demands for high quality foods and opportunities to create new market outlets have contributed to increase the interest in the development of biodegradable packaging. In particular, the application of biodegradable edible films as effective barriers against gas, moisture and liquid migration has appeared to be the appropriate way for prolongation of the shelf-life of ready-to-eat food and for the increase of its quality. Films can be produced from natural polymers, such as polysaccharides, lipids and/or proteins, and are perfectly biodegradable and safe to the environment [44]. Fresh products are now the second leading cause of food-borne illness. Commonly, pathogens contained in fresh products include Escherichia coli, Salmonella sp., Listeria monocytogenes and Bacillus cereus. Several techniques have been used to extend the postharvest life and maintain the quality of fresh products including food irradiation, high pressure processing, antimicrobial packaging and essential oils (EOs). The EOs obtained from plant material has been used for centuries as antimicrobial agents, and now most of the studies are carried out on these natural extracted agents to directly preserve food products or to encapsulate them as bioactive agents in antimicrobial packaging. In this decade, words like biocompatibility, environment friendly, biodegradability are commonly in use. Materials contained in the packaging are preferred to be biodegradable. This finds an explanation due to the increasing demand of consumers for high quality foods and environment concerns on limited natural resources, the use of renewable resources to produce edible or biodegradable packaging materials has gained significant attention. Antimicrobial packaging is very attractive and has increased the attention of the food and packaging industries due to a minimal process and preservative free products. The incorporation of antimicrobial agents into packaging materials are useful to prevent the growth of microorganisms on the product surface and improve microbial safety of the final product [45].

4. RESENT DEVELOPMENT

Alginate: Bio-based packaging must serve a number of important functions, including containment and protection of food, maintaining its sensory quality and safety, and communicating information to consumers. A big effort to extend the shelf life and enhance food quality while reducing packaging waste has encouraged the exploration of new bio/alginate-based packaging materials, such as edible and biodegradable films from renewable resources. Biodegradable films can be used as a vehicle for the incorporation of food additives such as antioxidants and antimicrobial agents delivering them to the food surfaces where deterioration by microbial growth or oxidation often begins. Alginate is widely used in food, pharmaceutical and bioengineering industries for its gel- and film-forming properties [46].

Carrageenan: Carrageenan is a complex mixture of several polysaccharides. To date, there are only a limited number of studies addressing the use of carrageenan with muscle foods. Carrageenan-based coatings have been used to prolong the shelf life of a variety of muscle foods including poultry and fish. Additional studies have demonstrated that antioxidants, such as gallic or ascorbic acids or lecithin, salt or antibiotics, can be added to the coatings to improve the quality and microbiological stability of muscle foods. IT is a selective inhibitor of several enveloped viruses, such as human pathogens as human immunodeficiency virus, herpes simplex virus and others. It can be used in both topics. It has excellent thickening and binding property for which carrageenan is being used in prevents soild-liquid separation and packaging [16, 47].

Agar: Another seaweed-derived polysaccharide is agar. Used extensively in microbiological media to provide firmness, agar exhibits characteristics that make it useful for packaging and it can also purify human body as it has binding ability to heavy metals, carcinogens and pesticides and can remove form our body through its cell wall. It also used by biodegradable packaging industries and various biomedical industries [47, 48].

Gelatin: This is a unique polymer comprising multifunctionalities like gelling, thickening, water-binding, emulsifying stabilizing, foaming, film forming and fining characteristics. Besides, gelatin, similar to synthetic high polymers, shows a rather wide molecular weight distribution. It is soluble in water and in aqueous solutions of polyhydric alcohols such as glycerol and propylene glycol and also hydrogen bonding organic solvents like acetic acid, trifluoroethanol, and formamide. Gelatin is practically insoluble in less polar organic solvents such as acetone, carbon tetrachloride, di- methyl formamide and most other nonpolar organic sol- vents. In the present study we reported on simultaneous copolymerization of 2-hydroxyethyl methacrylate with gelatin using blending and casting method, where simultaneous evaporation at room temperature was driving force. Later the films were subjected to gamma irradiation. The mechanical and thermo-mechanical properties of the films were analyzed. The gelatin films were subjected to irradiation with different gamma doses (50 to 500 krad) at a dose rate of 350 krad/hr using Co60 gamma source. The relative humidity was around 78% and the temperature was 32°C. These samples were stored in a laminated poly ethylene bag until testing [49].

Collagen: Gelatin films have been used as a delivery system for applying antioxidants to poultry or applied directly to poultry meat surfaces or processed meats to prevent microbial growth, salt rust, grease bleeding, handling abuse, water transfer, moisture loss, and oil adsorption during frying. Despite these successes, gelatin lacks strength and requires a drying step to form more durable films. Currently, the meat industry currently uses collagen films during the processing of meat products. When heated, intact collagen films can form a ‘‘skin’’ or edible film that becomes an integral part of the meat product (Cutter & Miller, 2004). These commercially available collagen films have been purported to reduce shrink loss, increase permeability of smoke to the meat product, increase juiciness, allow for easy removal of nets after cooking or smoking, and absorb fluid exudates [50].

Chitosan: It was found as a good reinforcing and antibacterial agent. Structural changes were proved due to chitosan addition in the cellulosic films. The fabricated facial tissue paper had a good appearance, softness, antibacterial and biodegradable property. The collectivity of the above investigation indicates that prepared films can be performed as an antibacterial facial tissue paper. While the study demonstrated that the incorporation of PLA into chitosan improved water barrier properties and decreased water sensitivity of the chitosan films, tensile strength and other mechanical and thermal properties were not improved. In additional experiments, the authors demonstrated that a phase separation occurred, thereby proving the incompatibility of the two materials. Several additional studies have demonstrated the effect of PLA, alone or in combination with other antimicrobials to inhibit microorganisms on fresh or further processed meat products. In 2002, demonstrated the synergistic effect of 2% low molecular weight polylactic acid alone or in combination with lactic acid or nisin against Escherichia coli O157:H7 on raw beef during irradiation and during refrigerated storage.

While the authors demonstrated inhibition against the pathogen on beef using PLA or lactic acid, as well as combinations of PLA with lactic acid and nisin, the authors concluded that the antimicrobial effect of PLA was not significantly different than that of lactic acid alone. In another study it was examined the effect of PLA for reducing pathogens on raw meat. E. coli O157:H7, Listeria monocytogenes, S. typhimurium, or Yersinia enterocolitica associated with lean beef surfaces treated with PLA, lactic acid, or sterile water. PLA treatments at pH of 3.0 resulted in significant reductions of E. coli O157:H7; however, E. coli O157:H7 was not inhibited when PLA was applied at pH 5.0, 6.0, and 7.0. When applied to ground beef, ground pork or breakfast sausage inoculated with E. coli O157:H7 and subjected to long term refrigerated storage, PLA treatments did result in up to a 1.7 log10 reduction of the pathogen. In 2004, demonstrated that low dose PLA, in combination with low-dose irradiation (2.0 kGy), followed by long term refrigerated storage could effectively reduce populations of E. coli O157:H7 and S. typhimurium up to 5 log10 CFU/cm2 (99.999%) on beef surfaces. Subsequent experiments on PLA also demonstrated that irradiation did not affect the tensile strength of the packaging materials [47, 51].

Starch: It is a good raw material for the preparation of biodegradable cling film. This is determined by the characteristics of starch. First, starch as glucose molecules polymerization and high molecular compound, is vulnerable to microbial decomposition, a membrane component which can make film degradation rate greatly increase. So it is applied to one of the important factors of the preparation of biodegradable film. Second, starch, cheap, as a kind of renewable resources, is one of the most widely used food raw materials. So the starch has a wider range of applications in the production of plastic packaging. For example, Yan *et al.* used oxidative esterification starch as substrate, adding glycerin, sodium alginate, dehydrogenation sodium acetate to make antibacterial film that is used in cake packaging. Experiments show that the antibacterial film has higher security and can prolong the shelf life, being of great economic benefits [52].

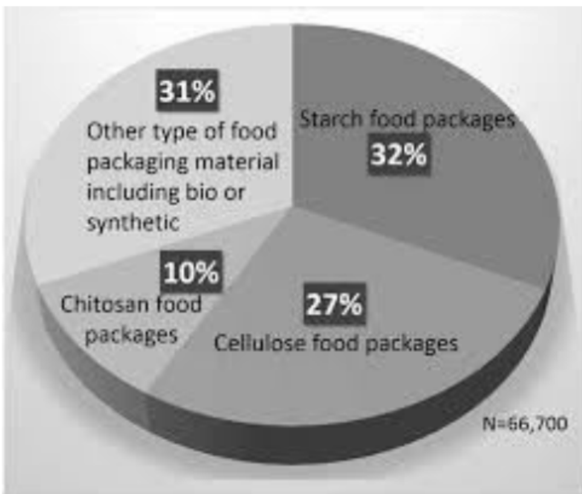

Figure 22: Percentage of packaging materials.

Cellulose: Cellulose is a non-digestible component of plant cell walls. In the manufacture of edible films, cellulose-based films tend to be water soluble, resistant to fats and oils, tough, and flexible. Cellulose based films applied to muscle foods can reduce oil uptake during frying, minimize run-off during cooking, and reduce moisture loss when applied as glazes for poultry and seafood. Cellulose based films have been commonly used to provide mechanical, oxygen barrier, and oil barrier properties for foods such as pizza and ice cream cones, whereas very little information exists for application to fresh or further processed muscle foods. The plant fiber can be pulped to produce paper and board products, or it can be treated by several processes such as pre-hydrolysis or pulping to obtain pure cellulose (>90% cellulose content). This material can be further chemically and/or mechanically processed into modified forms such as micro/ nanoscale cellulose and cellulose derivatives (ether and ester) for use in cellulose-based composites, thin polymer films that can be also used in packaging materials [53].

Bagasse: The bio-products generated during agro-industrial processing of sugarcane bagasse (SB), SB constitute potential sources of carbohydrates that can be used to generate valuable products of commercial interest. Bio-industrial applications appear to provide sustainable, economic, environmental, and strategic advantages, with breakthroughs in micro-biotechnology offering huge potential opportunities. SB has already been successfully converted into many value-added products such as ethanol, xylitol, organic acids, industrial enzymes, and other important specialty chemicals, as summarized. Scaling up to the pilot scale, however, is still a necessity. Such a step forward in the developing world would be highly rewarding. However, recent advancements in genetic engineering to improve microbial strains, media formulations, and product recovery have enabled more efficient conversion of SB into value-added products of commercial interest. For instance, it has already been used in bio packaging composite materials industries [54].

Jute: Environmental and economical concerns are stimulating research in the design of new materials for construction, furniture, packaging and automotive industries. Particularly attractive are the new materials in which a good part is based on natural renewable resources, preventing further stresses on the environment. Nevertheless is important to know that renewable resources depend of a balance, in which their harvests have to be lower than its growth, in this sense jute fibers need to be used based on a sustainable system to avoid the Amazon deforestation. Examples of such raw material sources are annual growth native crops/plants/fibers, which are abundantly available in tropical regions over the world. These plants/fibers (like jute and sisal) have been used for hundreds of years for many applications such as ropes, beds, bags, etc. [55].

Coir: Coir fibers were soaked in each type of monomer solutions containing different monomer concentrations (10–50 %) for 10 min and irradiated under UV radiation at different doses (5–30 UV passes) expressed by the number of passes. For HEMA solutions, irradiated coir fibers were extracted with hot benzene for 48 h to remove the homopolymers and then dried at 105 C to determine the exact amount of grafting of the polymer with fibers. For this solutions, irradiated coir fibers were then extracted with hot acetone for 48 and 45 min, respectively, and then dried at 105 C to determine the exact amount of grafting of the polymer with fibers. Strength as a measure of uniformity is a very useful test, since a change in any physical property or a change in the chemical composition of a textile material will nearly always result in a change in strength; even here, however, a warning must be voiced, as strength tends to give minimum rather than average results. The tensile properties such as tensile strength (TS) and tensile modulus (TM) of the untreated coir fiber were measured and the average values for 15 samples were: TS = 115.3 ± 2 MPa and TM = 533.7 ± 4 MPa. The TS and TM of each type of monomer (HEMA) treated coir fibers are graphically demonstrated against the number of UV passes as a function of optimized monomer concentration. For almost all cases, the TS and TM increases with increasing radiation dose up to certain limit and then decreases with the increase of UV passes. Simulating weathering studies demonstrated that the TS and TM of the treated sample could be lower than that of the untreated sample with respect to their degradation time. This is the significant improvement of coir fibers obtained by an easy, fast, and pollution-free photo grafting method. So it has great prospects for commercial uses, such as to make brushes, floor mates, heavy cord, coarse nets and bags etc. [56].

Banana: Waste Banana Fiber (WBF) contains very low amount of α –cellulose and high amount of ash. WBF is constituted by fibers, broken fibers and nonfibrous cells. At 120 min of cooking with 8% alkali charge, WBF is defibrated with pulp yield of 35.9%. Initial SR value of Extracted Banana Fiber (EBF) pulp is very high and consequently increases to a very high tensile strength. Tear index of EBF is also very high due to the longer fiber length. The papermaking properties of WBF are quite acceptable for handmade paper. But these properties are much lower as compared to EBF pulp. The waste generated in banana fibre extraction plant can be used in handmade paper which will mitigate the pollution problem along with creating employment opportunity in rural area [57].

5. CONCLUSIONS

This review shows that seaweed, creatures and cellulose are great potential film-forming to packaging with natural insurance and support of physical wellbeing. The concepts that a biomaterial can change with the radically new types of substance that we are using that many new ways, in packaging industries and its well go together in nature that means their biodegradability are so much strong. The study of seaweed, creatures and cellulose in regards to their biodegradability, compatibility and potential contribution to the bio-economy have verified that seaweed, creatures and cellulose are compatible for forming biodegradable and low cost effective blend films for a wide range of their packaging application. Numerous challenges still exist today across biomaterials, so there has been an exponential need to focus on more research and interest into making such a technology into reality. Hence, it is worth of further investigate this newly developed packaging material product to highly sustainable, functional and low cost effective products and many students are also research this topic for MS and PhD program.

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