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Biotechnology

STARCH AND CELLULOSE BASED BIO-POLYETHYLENE: A REVIEW

KEY WORDS: Starch, Biopolyethylene, Vegetables, Fruits, Food packaging

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ABSTRACT

In current year, there has been enhancing in the usage of plastic and disposal of waste coming from daily life. Various type of plastics are using for our daily needs, in order to reduce the impact of petroleum based plastics and other waste to be considered and focused on green plastics. Polyethylene are the polymers which can be produced by repeating the single units. It is one of the bittersweet coinage for human's better life. But this coinage causes a lot problems to the environment. To overcome this problem an alternative and eco-friendly Polyethylene is introduced called bio-polyethylene. Bio-polyethylene are from natural biomass sources. Among various source, starch has chief role in production of bio-polyethylene due to its low cost and nature abundance with plasticizers such as glycerol and vinegar. Starch based bio-polyethylene has good mechanical properties, tensile strength and biodegradability. Cellulose also used as source in development of biodegradable polyethylene. In this review starch and cellulose has led to their great innovative uses in food packaging and to improved biodegradation and mechanical properties.

INTRODUCTION

In current year, there has been enhancing in the usage of plastic and disposal of waste coming from daily life. Various type of plastics are using for our daily needs, in order to reduce the impact of petroleum based plastics and other waste to be considered and focused on green plastics (Valentina Siracusa and Ignazio Blanco, 2020). Polyethylene is a group of synthetic and semi synthetic organic polymer including carbon and hydrogen. Ancient Greek derives polyethylene as "Plastikos" means "FIT FOR MOULDING". Word itself tells us that Polyethylene can be molded into any solid shapes or form using heat and pressure. The first fully synthetic come onto the scene in 1907 called Bakelite, derived from fossil fuel (Nur Fahana Fadzil and Siti Amira Othman, 2021; Bakar, F, and Othman, 2019). Polyethylene materials are currently considered very important materials due to their properties and performance over materials such as metal and wood (Ezgi Bezirhan Arikan and Havva Duygu Ozsoy, 2015 and Alvarez-Chavez, et al., 2011). Polyethylene are the cheap production process, this suitability, combined with a variety of beneficial properties such as light, durability and flexibility has led to widespread use in today's society. As far as product packaging is concerned, packaging plays a role in product holdings and food safety for the food industry (Nor Izaida Ibrahim, et al., 2021).

Polyethylene are used to make products such as water bottles, coffee cups, forks, knives, Polyethylene bags to carry groceries. Etc. Petrochemical based polyethylene are not eco-friendly because of their high content of carbon footprint (Manali Shah, et al., 2021; Booniteewanich, et al., 2014). Polyethylene was thought to be a boon however it ended up to be a curse. Polyethylene is terribly harmful because it produces harmful gases once it is burnt. It is harmful to the soil, non-bio degradable or takes many years to degrade (Vrinda Lamba and Ranjana Singh, 2021; Jimenez- Rosado, et al., 2020; Bayer, et al., 2014). It is estimated that about 34 million tons of Polyethylene are produced per year by humans (Manali Shah, et al., 2021).

Out of that only 7% is recycled and that remaining 93% is dumped into the landfills, oceans and sea (Manali Shah, et al., 2021; Sushmitha, et al., 2016). In 2015, more than 300 million tons of Polyethylene was used in the world [Mekonnen, et al., 2013; Manali Shah, et al., 2021] and in India it has estimated as 1.7 million metric tons. Polyethylene waste generation data

gathered from 60 major cities showed 25940 tons of Polyethylene waste per day (TPD) is generated in India. However, the extended high development paces of GDP and proceeding with quick urbanization propose that India's direction of Polyethylene utilization and Polyethylene waste is probably going to have a huge increment (Vrinda Lamba and Ranjana Singh, 2021; Mekonnen, et al., 2013; Tsang, et al., 2014). Indeed, in nature, the degradation of Polyethylene takes a long time until hundreds or even thousands of years to break the carbon chains (Nandiyanto, et al., 2020; Wu, 2009; Ezeoha and Ezenwanne, 2013).

Polyethylene based products causes many problems like accumulation of waste in land area, in natural habitats like sea, oceans (Manali Shah, et al., 2021; Thompson, et al., 2009). The environmental threats of Polyethylene include landfills (Philip R Ritchie and Guy, 2013), Polyethylene waste in the oceans (Law, 2010) release of toxic gases (Metz, et al., 2007) and non-biodegradability (Jahnavi Dalmia and Gayatri Wadiye 2020). One of the best solutions to overcome this situation is by reducing the synthetic polymer and promoting new version of Polyethylene, biopolymer (Nur Fahana Fadzil and Siti Amira Othman, 2021).

Bio-Polyethylene are the bio based and biodegradable polyethylene which are derived from micro-organisms or plants (Chozhavendhan, et al., 2020). Bio-polyethylene are one of the most innovative materials that are biobased and biodegradable which is made from waste, biomass and renewable sources such as jackfruit (Lothfy, et al., 2018), waste banana peels (Mohapatra, 2015), organic waste (Goswami, et al., 2015), agriculture waste (Zulkafli, 2014), newspaper waste (Joshi, et al., 2015), oil palm empty fruit bunch (Isroi, et al., 2017), sugar cane (Khosravi-Darani and Bucci, 2015), corn starch (Keziah, et al., 2018), rice straw (Agustin, et al., 2014), rapeseed oil (Delgado, et al., 2018), vegetable oil, cellulose from plants, starch, cotton, bacteria (Shamsuddin, et al., 2017) and sometimes from several Nano particles like carbohydrate chains (polysaccharides) (Jabeen, et al., 2015; Izathul Shafina Sidek, et al., 2019). Biodegradable polyethylene may decompose into carbon dioxide and water in 20-45 days if there is enough humidity, oxygen and an appropriate number of micro-organisms, which can be found in natural landfills or manure (Taofeeq D Moshod, et al., 2022). These are currently used as packaging materials, but in future these will be used in forming various products such as electronics and vehicles parts (Chen, et al., 2014).

Starch is a bountiful and naturally available material made out of two types of glucose polymers, amylose and amylopectin (Vrinda Lamba and Ranjana Singh, 2021). The Bio-polyethylene produced from starch has a high biodegradability in the soil (Nandiyanto, et al., 2020). Thermoplastic starch is the most commonly used bio-Polyethylene, making up about 50% of the bio-polyethylene in the market (Kokila muniyandi, et al., 2020). Starch and derivatives are produced from various raw materials such as potato and its peel, corn, wheat, pea. Starch based Bio-Polyethylene are used for packaging materials and for producing food utensils such as cups, bowls, bottles, cutlery, egg cartons, and straws (Ghada atiweh, et al., 2021). The high amylose starch films showed better mechanical properties such as high modulus, high tensile strength and much higher impact strength (Hooman molave, et al., 2015). The aim of the project is to produce Bio-polyethylene from rice washed water, wheat barley, Avocado pit waste, Tapioca roots and cellulose based peanut shell waste. Starch alone cannot form films with satisfactory mechanical properties (high percentage elongation, tensile and flexural strength) unless it is Plasticized, blended with other materials, chemically modified, or modified with a combination of treatments (Waham ashaier laftah (2017). Common Plasticizers used include glycerol, vinegar and other low-molecular-weight poly hydroxy compounds, polyether and urea. Rice starch is a natural polymeric carbohydrate and it contains 35% of amylose and 65% of amylopectin (Nandiyanto, et al., 2020). The quality of wheat barley products is much determined by its starch properties. The avocado pit is encased in a shell and comprises 13-18% of the size of the whole fruit. Though avocado seed also has potency as source of starch and pigment. The seed contains high starch. This content of starch that becomes raw material for manufacturing bio-polyethylene (Hendra S Ginting, et al., 2015). Tapioca is a cassava starch which has been widely used as raw material of biodegradable polyethylene and showed a promising result (Syaubari, et al., 2018).

Cellulose is a homopolymer synthesized by the polymerization of glucose residues from a substrate as UDP-glucose linked by beta-1,4-Glycosidic bonds forming a beta-1,4-D-glucan (Maricarmen Iniguez-Moreno, et al., 2021). It is found in the wall of all major plants and green algae and all the membrane of most fungi (Selvamurugan and Sivakumar (2019). Cellulose is extracted from crystalline state in microfibrils, it is soluble in solvent as N-methylmorpholine N-oxide. It is very difficult to use in packaging because of it is hydrophilic and crystalline in nature possessing poor mechanical properties in its raw form. Therefore it must be treated with chemicals like NaOH, H₂SO₄, CS₂, etc to produce cellophane having excellent mechanical characteristics (Rukhsana Rahman, et al., 2019; Majid, et al., 2018). As a result, research has been focused on cellulose derivatives for packaging purpose. In our study, peanut shell waste is used to prepare a bio-polyethylene mold to increase the water absorption. Peanut shell is a waste product obtained after harvesting peanut seed from its pod (Mohammed Awwalu Usman, et al., 2020). It is the abundant Agro-industrial waste product which has a very slow degradation rate under natural conditions. One method for improving the quality of moisture barrier can be performed by addition of hydrophobic compounds such as fatty acids into cellulose matrix to develop the composite films (Manali Shah, et al., 2021).

Materials and methodology

Preparation of bio-polyethylene film from starch

The bio-polyethylene film is prepared according to the following procedure: Firstly take 8gm of starch into the beaker. Add around 50ml of water, a tea spoon of vinegar and Glycerol to form a solution. Heat the solution gently for two minutes. While heating, make sure to stir the solution continuously. After heating a jelly like liquid solution is formed. Keep stirring till the gelatinous substance turns

transparent. Stop the heating, keep the viscous liquid into a mold and dry it for a day under the sun (Pramode Ghale, et al., 2018).

Preparation of bio-polyethylene mold from cellulose

The bio-polyethylene mold is prepared by following procedure: Take the 100gr of peanut shell waste and put it in a grinder to grind. Now transfer the peanut shell waste powder to the beaker. Add 100gr of starch, 130ml of water, 24ml of glycerol and 38ml of vinegar. Mix all the ingredients, Heat the mixture for 10min and then put the mixture on desired mold. Now keep the mold in oven at 15°C for minutes. Remove the bio-polyethylene mold (<https://youtu.be/xylnlv6xWCc>).

Tapioca roots

The study Bio-polyethylene using cassava flour, tapioca starch and tapioca starch nanoparticle with the presence of glycerol as plasticizer. The result showed that cassava flour and tapioca starch bio-plastics were potent to be applied as sweet soy sauce and vegetable oil packaging, but not as water and chili sauce packaging. The resulted bio-plastics absorbed UV-A so that the products inside could be protected from photooxidative degradation (Noryawati Mulyono, et al., 2015). The study on synthesis of biodegradable plastic from tpioca with N-Isopropylacrylamid and chitosan using glycerol as plasticizer. Polyethylene with the addition of NIPAm and 1ml glycerol has the highest tensile value about 31.1MPa. Polyethylene with poly (NIPAm) with 4ml glycerol produces the highest elongation value about 153.7%. Polyethylene with Chitosan-graft-poly (NIPAm) with 1ml glycerol has the longest biodegradation because of the small mass-loss for six weeks which is about 6.6% (Syaubari, et al., 2018).

Avocado pit waste

A Bio-Polyethylene development study from avocado seed starch has been carried out by adding chitosan and glycerol. Avocado seed starch used had a moisture content of 1.087%, an ash content of 1.007%, the starch content of 67.6950%, amylose content of 32.4739%, amylopectin content of 35.3212%, the protein content of 10.440%, the fat content of 1.860%, peak viscosity is 3847 by setting the gelatinization temperature at 85.17°C. The best bio-plastic conditions of avocado seeds were obtained at 90°C with a ratio of starch: chitosan (w/w)=7:3 and glycerol 0.2 ml/g with a tensile strength of 5.096MPa, breaking extension 14.016%, Modulus young 36.359 MPa. Bio-polyethylene with chitosan as fillers and glycerol as plasticizers have a smooth and soft fracture surface and few cavities (Ginting-Lopez, 1999; Ramadhan and Handayani, 2020).

The ratio of bio-plastic avocado seeds with the addition of microcrystalline to have the best mechanical properties is 7:3. It possesses 2.74MPa tensile strength and an extension of 3.61%. The Bioplastic surface morphology is uneven and hollow (Triawan, et al., 2020; Lubis, 2018).

Two types of biofilms were produced: T1 (starch) and T2 (starch and glycerol). The color was measured, and the films were characterized by mechanical tests. A yield of 11.38% was obtained for the isolated starch. Glycerol addition significantly influenced the tensile strength being T1 higher than T2. In elongation percentage, T1 was lower than T2. The results indicate that it is possible to use Hass avocado seed waste for starch extraction and produce biofilms to give it added value. This biofilm can be used in moisture food coatings (Rosalia JIMENEZ, et al., 2018).

The mechanical properties of bio-plastic prepared from avocado seed starch and microcrystalline cellulose from sugar palm fibers as filler using Schweizer reagent as solvent is studied. Morphological analysis showed that the isolated microcrystalline cellulose from sugar palm fibers are rodlike shape with diameter of 5.55-9.44 micro meter and crystalline size of 25.08nm. Mechanical properties of bioplastic showed

that the best condition of bioplastics obtained at comparison of mass starch-microcrystalline cellulose 7:3 and the addition of glycerol 0.1(v/w) for tensile strength 20.874MPa and elongation at break of 6.22% (Mora sartika, et al., 2018).

Bio-polyethylene were analyzed physical and chemical properties. From the analysis, best condition of bioplastics obtained at temperature 90°C with comparison of mass starch- chitosan 7:3 for tensile strength 5.096MPa, elongation at break 14.016% and modulus young 36.359Mpa. The results of mechanical properties were supported by scanning electron microscopy showed the bioplastic with chitosan as filler and plasticizer glycerol have the fracture surfaces were smooth and soft and a little cavity (Hendra S Ginting, et al., 2015).

Peanut shell waste

The Physical and mechanical properties such as its tensile strength (TS), Young's modulus (YM) and elongation(E) at break were found to be increased on loading with cellulose Microfiber (CMF). Ts is found to be increased from 38.8±3.2 to 49.4±4.3 and there is about 6% increase in the elongation at break. Similarly, CMF films exhibit 54±0.2% water solubility, 1.304 opacity and 1.71±0.07 GPa, respectively. Also, the bio-nanocomposite film showed less swelling property and an increase in soil degradation rate, enabling it to be applied in food packaging (Ananda Raj, et al., 2019).

The study on DIY Bio-polyethylene development from Peanut Hulls waste in a Starch-Milk Based Matrix. Its limits were clarified by thermal characterization in terms of not being able to exceed temperatures of around 80°C and suffering non uniform deformation, especially in the case its thickness does not go beyond a few millimeters. Coloration tests proved effective. Of course, the material is in search of full mechanical characterization though it proved suitable to be punched and did not suffer fragmentation or excessive porosity (Troiano, et al., 2017).

Wheat barley

This review summarizes the recent developments in our understanding of the isolation, chemical composition, granular structure, chemical structure of starch components, physicochemical properties, and various modifications of barley starch. The structure–function relationships of starch are discussed (Fan Zhu, 2017).

Rice starch water

The bio-plastics developed from corn and rice starch, with different compositions of corn and rice starch, glycerol, citric acid, and gelatin. The tensile properties were improved after adding rice starch. However, water absorption and water solubility were reduced. The results show the suitability of rice and corn based thermoplastic starch for packaging applications (Marichelvam, et al., 2019).

Characterization of bio-Polyethylene

Biodegradability Test

The specimen was cut into pieces of 4.0cm². Found near the roots of plants which are rich in nitrogenous bacteria, 500g of soil (having slight moisture content) was collected and stored in a container. One sample was buried inside the soil at a depth of 2cm and another buried at a depth of 3cm for 15days under the condition of the room. The weight of the specimen was measured before and after the testing (Marichelvam, et al., 2019).

Tensile strength test

To measure the tensile test, the sample was sliced into 5x5mm. It is tested out on a control universal testing machine at a cross head speed of 2 kg/min with the load cell of 50kn. It is defined as the maximum force that can be held by the specimen when stretched or pulled before the material is broken (Nandiyanto, et al., 2020).

Water solubility test

The film samples were cut into square sections of 2.0cm², and the dry film mass was weighed accurately and recorded. The samples remained immersed in 100ml distilled water and fixed agitation at 180 rpm was carried out for 6h at 25°C. The lasting portions of the films were filtered after 6h. They were then dried in a hot air oven at 110°C until an ultimate fixed weight was found. Glycerol has a good water solubility range from 18% to 25%. The percentage of total soluble matter was calculated as (Marichelvam, et al., 2019).

$$WS (\%) = [(WO - Wf) / WO] \times 100.$$

Elongation test

Elongation test is done by taking bio Polyethylene samples by cutting 1cm x 5cm x 0.1cm. Then it was tested and obtained the maximum pull until the bio Polyethylene break up. For calculating elongation using a formula as follows:

$$\text{Elongation} = (b - a) / a \times 100\% . [64]$$

Advantages and Disadvantages of Bio- polyethylene

Potentially a far lower carbon footprint. Bio-Polyethylene products are produced from renewable resources and it contributes to reduction of greenhouse gases emission through reduced carbon footprint (Harsini Venkatachalam and Radha palaniswamy, 2020; Prasteen, et al., 2018). Eco-Safety. Bio-polyethylene also generates fewer greenhouse gases and contains no toxins. Lower energy costs in manufacturing. The bio-polyethylene can evade the environmental problems like uncontrolled dumping of wastes on land and disposal to sea, and the related emission of toxic substances. A bio- polyethylene can feel softer and more palpable. For applications like cosmetic packaging, this will be a serious perceived consumer benefit. Bio-polyethylene from plant resources can manage the plant waste or plant residues in an efficient manner.

Disadvantages

Bio-Polyethylene require favorable climatic conditions in easing their degradation process. Risk of contamination: Bio-Polyethylene should not be blended in with non-biodegradable polyethylene. High costs. It is acclaimed that bio-polyethylene costs two times more than conventional polyethylene (Sata, et al., 2004). Recycling problems. Bio-polyethylene material might actually contaminate the recycling process if not separated from conventional polyethylene. Raw materials reserves might reduce by bio-polyethylene production (Sartore, et al., 2016). Bio-polyethylene may release methane in landfills (Valentina, et al., 2020).

Conclusions

The bio-polyethylene have been developed to address environmental issues associated with conventional petroleum based polyethylene. Bio-polyethylene are not only the solution to the problem of pollution. The changes in consumer behavior in buying, consuming and disposing of bio-Polyethylene and spread public awareness of the bio-polyethylene are also essential. The starch is one of the promising source for the application of biofilms. Around 50% of the bio-Polyethylene are manufactured using starch as a main source. Because of its natural abundance, low cost it is considered for preparing bio-polyethylene. The research has been done to investigate various source of starch for the application of bio-polyethylene in packaging industry. Starch possess various properties like mechanical property, tensile strength, flexural property and biodegradability. Similarly, cellulose plays vital role in the preparation of bio-polyethylene. As it is difficult to produce bio-polyethylene because of its highly crystalline structure, poor flexibility and tensile strength. We use cellulose to blend with starch to produce bio-polyethylene and to perform good mechanical properties and biodegradability. By studies it shows that the

harms associated with bio-Polyethylene are still less severe when compared to conventional polyethylene.

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