ANALYTICAL HIERARCHICAL PROCESS OF EDIBLE BIOMASS AS PRIMARY FEEDSTOCKS IN BIODEGRADABLE PLASTIC PRODUCTION

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ABSTRACT

Plastic is one of the world's most versatile materials due to its durability, stability, and low production costs. Plastics take an extremely long time to degrade and due to their widespread use, large amounts of plastic waste are released into the environment worldwide, which contributes to the current problem of white pollution. The waste plastics that accumulate in the environment can be further degraded by weathering into smaller pieces, such as microplastics and nanoplastics, but these small pieces can cause more harm to the environment than larger plastics. How to solve the environmental problems caused by plastics is a global topic and challenge. The production of biodegradable plastics, a countermeasure to the root cause, has become a hot topic of research in recent years. At present, biodegradable plastics are not an alternative to conventional plastics, as many different factors such as production costs, production processes, product properties, and the handling of biodegradable plastics are hindering the development of biodegradable plastics. Therefore, the selection of suitable raw materials is of paramount importance for the development and widespread use of biodegradable plastics. This study will begin with the selection of natural biomass resources for use as feedstock in the production of biodegradable plastics through an extensive literature search. Then various criteria influencing the selection of natural biomass were rated with the assistance of literature review. Finally, the most viable natural biomass was selected as a composite feedstock in the production of biodegradable plastics by using the Analytical Hierarchy Process (AHP). The results of the study showed that maize is the most favorable natural biomass for the production of biodegradable plastic.

Keywords: biodegradable plastic, feedstock, natural biomass, AHP

KAEDAH PEMILIHAN KUANTITATIF BIOJISIM SEMULA JADI SEBAGAI BAHAN MENTAH KOMPOSIT UNTUK MENGHASILKAN PLASTIK TERDEGRADASI

ABSTRAK

Plastik adalah salah satu bahan yang paling popular dan memiliki pelbagai fungsi di dunia kerana kebaikannya seperti ketahanan, kestabilan dan kos pengeluaran yang rendah. Bahan-bahan plastik perlu mengambil masa yang sangat lama untuk terdegradasi, dan kerana penggunaannya yang meluas, sejumlah besar sisa plastik sudah dilepaskan ke alam sekitar di seluruh dunia, dan juga menyebabkan masalah pencemaran putih kepada alam semula jadi di seluruh dunia. Sisa plastik yang terkumpul di alam sekitar boleh terus terdegradasi kepada serpihan yang lebih kecil dengan tindakan luluhawa, seperti mikroplastik dan nanoplastik, serta serpihan kecil ini boleh menyebabkan pencemaran yang lebih berbahaya kepada alam sekitar daripada plastik yang lebih besar. Pencarian cara untuk menyelesaikan masalah alam sekitar yang disebabkan oleh plastik adalah topik dan cabaran yang besar bagi seluruh dunia. Oleh itu, pengeluaran plastik terbiodegradasi yang boleh menjadi satu penyelesaian kepada punca utama, telah menjadi tumpuan penyelidikan yang pertama dalam beberapa tahun kebelakangan ini. Pada masa ini, plastik biodegradasi memang bukanlah pengganti plastik tradisional, kerana terdapat banyak faktor halangan pembangunan plastik terbiodegradasi, seperti kos pengeluaran produk, proses pengeluaran, fungsi produk dan pelupusan plastik terbiodegradasi. Oleh sebab itu, pemilihan bahan mentah yang sesuai adalah sangat penting untuk pembangunan dan penggunaan plastik terbiodegradasi dengan meluas. Kajian ini akan dimulakan dengan pemilihan sumber biojisim semula jadi sebagai bahan mentah untuk menghasilkan plastik terbiodegradasi dengan cara kajian ilmiah yang meluas. Seterusnya, kriteria-kriteria yang mempengaruhi pemilihan bahan-bahan biojisim semula jadi akan dinilaikan dengan bantuan daripada kajian ilmiah. Akhirnya, biojisim semula jadi yang paling berdaya maju akan dipilih sebagai bahan komposit untuk menghasilkan plastik terbiodegradasi dengan penilaian melalui kaedah Proses Hierarki Analitik (AHP). Penyelidikan menunjukkan bahawa jagung adalah biojisim semula jadi yang paling bersesuaian bagi pengeluaran plastik terdegradasi.

Kata kunci: plastik terbiodegradasi, bahan mentah, biojisim semula jadi, proses hierarki analitik

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TABLE OF CONTENTS

LIST OF FUGURES

LIST OF TABLES

LIST OF TABLES

LIST OF SYMBOLS AND ABBREVIATIONS

AHP	:	Analytic Hierarchy Process
BPs	:	Biodegradable plastics
CI	:	Consistency Index
CR	:	Consistency Ratio
PE	:	Polyethylene
PHA	:	Polyhydroxyalkanoate
PHB	:	polyhydroxybutyrate
PLA	:	Polylactic Acid
PS	:	Polystyrene
PU	:	Polyurethane
RI	:	Random Consistency Index

CHAPTER 1: INTRODUCTION

1.1 Research Background

Plastic has been used extensively in various fields because of its lightweight, water resistance, durability, mature production technology and low cost. Plastic has brought great convenience to human life and its use shows an increasing trend with each passing year. According to the survey, global plastic production in 2019 totaled 368 million tons. It is estimated that production in 2020 decreases by about 0.3% to 367 million tons due to the impact of COVID-19 (Tiseo, 2021). However, improper disposal of conventional plastic waste leads to its entry into the ecosystem and failure to degrade for a long period time. According to a survey, in 2015, 55% of the plastic waste worldwide was discarded, 25% was incinerated, and only 20% was recycled (Ritchie & Roser, 2018). This has caused long-term as well as extremely damaging effects on the ecological environment.

When plastic waste is mixed into the soil, it may interfere with the absorption of nutrients and water by crops, resulting in reduced crop yields and even contaminating groundwater, endangering the surrounding environment. When plastic waste is abandoned on land or in water, it may be accidentally consumed by animals, resulting in their death. Moreover, invisible plastics that spread with the food chain threaten food safety and human health (Rochman & Hoellein, 2020). The release of carbon dioxide in the atmosphere from burning plastic waste also contributes to climate change. Effective recycling of plastic waste is an effective solution to plastic pollution, but it is difficult to do under current economic conditions because the cost of recycling is currently much higher than the cost of direct production. As a result, researchers are now increasingly focusing on biodegradable plastics, a countermeasure that tackles plastic pollution at its roots.

Biodegradable plastics (BPs) are plastics that can be decomposed by living organisms into harmless substances such as water, carbon dioxide, methane, and biomass and are generally produced from renewable raw materials, microorganisms, petrochemicals, or a combination of them (Ammala et al., 2011). The ideal BPs have the basic properties of ordinary plastics, but after disposal can be completely decomposed by environmental microorganisms and eventually inorganized to become a component of the carbon cycle in nature (Haider et al., 2019).

Biodegradable plastics can be classified into bio-based biodegradable plastics and fossil fuel-based biodegradable plastics according to the source of raw materials. Bio-based biodegradable plastics mainly include polylactic acid (PLA) and polyhydroxyalkanoate (PHA) (Paula et al., 2018), while fossil fuel-based biodegradable plastics mainly include polybutylene succinate (PBS), polycaprolactone (PCL) and polybutyrate adipate terephthalate (PBAT) (Gerard, 2016; Xu & Guo, 2010). PLA and PBAT are currently the two types of BPs with the largest production capacity, accounting for 18.7% and 13.5% of global bioplastics production respectively, followed by PBS and PHA with 4.1% and 1.7%, respectively (Buchholz, 2020).

Fossil fuel-based BPs such as PBAT and PBS are commercially available at high levels, with melting points and mechanical properties comparable to those of conventional plastics, and have the ability to cover the use of conventional plastics in the disposable products industry (Camani et al., 2021; Su, Kopitzky, Tolga, & Kabasci, 2019). But they come from fossil fuels, which are facing depletion and are not renewable. Therefore, bio-based BPs from renewable biomass is of increasing interest considering the environmental impact and the conservation of non-renewable resources.

Bio-based BPs such as PLA and PHA are derived from renewable materials, especially plants, commonly sugar cane, maize, cassava and cotton, etc. (Poletto, 2016). They have a higher melting point and strength than traditional plastics, although the tensile toughness is lower, they are promising to replace traditional plastics after improvement (Farah, et al., 2016; Tarrahi et al., 2020). Their good biodegradability and biocompatibility allow them to be completely degraded under certain conditions, producing CO_2 and H_2O that are inherently present in nature. It significantly reduces the burden on non-renewable resources as well as on the environment. But it is difficult to argue that BPs have a demonstrable advantage over conventional plastics.

PBAT and PBS are promising to replace the traditional plastics market because of their superior performance and high commercialization level (Narancic, et al., 2020). But fossil fuel-based BPs are derived from the same non-renewable energy sources that are in severe shortage as conventional plastics. In contrast, bio-based BPs use natural biomass as a substrate, which is widely available and does not burden the environment and resources (Meereboer et al., 2020). Therefore, bio-based BPs are one of the effective means to solve plastic pollution and are expected to have a promising development in the future.

However, there are many challenges for the large-scale commercial production of bio-based BPs. PHA has outstanding advantages, such as high strength, good gas barrier properties and high melting point. However, the high production cost makes it impossible to commercialize (Meereboer et al., 2020). The choice of raw materials is closely related to the cost and performance etc. of BPs. Accordingly, the selection of optimal natural biomass as a feedstock for the production of BPs has become a hot research topic.

As we all know, the production process of ordinary plastics is quite mature and the production costs are very low. It has been estimated that the current cost of producing BPs is about 3 to 10 times that of conventional plastics (Luyt & Malik, 2019). Cost, product properties such as plasticity and strength, biodegradability, application, and environmental impact should all be taken into account in the commercial production of BPs. And the choice of raw materials is closely related to these factors.

To produce BPs that can replace conventional plastics on a large scale, it is extremely important to select the most suitable raw materials. The natural biomass for bio-based BPs comes from a wide range of sources, which can be various terrestrial plants or agricultural by-products or even microalgae (Beckstrom et al., 2020; Jõgi & Bhat, 2020; Mojibayo et al., 2020). The selection of the most suitable natural biomass as a feedstock for the production of BPs can effectively increase the environmental and economic benefits of BPs, thus contributing to the solution of the plastic pollution problem and eventually replacing conventional plastics.

The tremendous amount of conventional plastics used and discarded has led to increasing environmental pollution. Therefore, degradable plastic that can replace traditional plastics without causing environmental pollution is needed. This study will analyze various nature biomass used as feedstock in BPs production and the criteria influencing the selection of the most viable natural biomass through the application of AHP (Analytic Hierarchy Process).

1.2 Problem Statement

One of the reasons why BPs are currently not competitive in the market and cannot replace traditional plastics is the cost issue. The production cost of BPs is about 9.4 to 18.8/kg, which is more than double that of conventional plastics (8.03 to 9.4 MYR/kg) (Rujnić-Sokele & Pilipović, 2017). Raw materials are an important factor in determining the cost of manufactured BPs.

The performance of the product also depends to some extent on the choice of raw materials. Only when the performance of BPs equals or even surpasses that of conventional plastics, can BPs hope to completely replace conventional plastics. Current bio-based feedstocks are still mainly agricultural products. But the large-scale production of BPs from these agricultural products may also pose some problems. The questions such as whether BPs' production will compete with local food supplies and whether their agricultural activities will generate more CO_2 emissions all need to be considered (Filiciotto & Rothenberg, 2021; Kubowicz & Booth, 2017).

In order to produce marketable BPs and improve the environmental pollution problems caused by conventional plastics, selecting the most suitable raw materials on a worldwide scale, reducing production costs while maintaining product performance, and ensuring the sustainability of BPs are the focus of current research.

1.3 Research Aim and Objectives

The aim of this study was to select the optimal natural edible biomass to be used as a feedstock for the production of biodegradable plastics using analytic hierarchy process (AHP) to alleviate the current global environmental problems caused by conventional plastics.

Objectives of this study are as follows:

1. To identify suitable natural edible biomass resources for the production of biodegradable plastics (BPs) by using Analytical Hierachical Process.
2. To determine crucial criteria for selecting feedstocks for the production of BPs by using Analytical Hierachical Process.
3. To calculate the favoured natural biomass resources based on Analytic Hierarchy Process (AHP) results.

1.4 Scope of Work

This study will focus on the selection of natural edible biomasses that are used as feedstocks for the production of BPs. A multi-program or multi-objective decision making method, AHP will be applied in the selection of natural biomass.

The criteria for evaluating these natural edible biomasses will also be selected by using AHP.

The study will also use AHP to analyze various natural biomasses and calculate their priority as feedstocks for BPs production. In order to make the obtained prioritization results more reliable, this study will also test the consistency of the results using AHP. And this will also be helpful in decision-making in BPs commercial production.

1.5 Dissertation Structure

The dissertation includes five chapters namely introduction, literature review, materials and method, result and discussion and conclusion and recommendation.

Chapter 1 provides an overview of the research background, problems, research aims and objectives, and scope of work of this paper.

Chapter 2 is a literature review, focusing on a review of previously conducted research on BPs standard and specification, types of BPs, raw materials for the production of BPs, the applications and challenges of BPs.

Chapter 3 describes the selection method of natural biomass, the selection of criteria, and the steps for selecting the most feasible feedstock of natural biomass for BPs production using AHP.

Chapter 4 presents all the obtained results and a discussion of the results and shortcomings.

Chapter 5 is the conclusion, which summarizes the results of each research objective, as well as the findings of this study and some recommendations for future work.

CHAPTER 2: Literature Review

2.1 Introductions

Plastic has caused great harm not only to humans, but also to animals, plants and the entire ecosystem. First, traditional plastics consume high energy in the manufacturing process and produce waste materials, which cause a great burden to the environment. Secondly, plastic incineration will produce harmful gases, such as hydrogen chloride, dioxins, etc (Shen et al., 2020). These gases will spread to the air, soil and water, affecting nearby plants and animals, human inhalation can cause respiratory health problems. In addition, plastic landfills take up a lot of land, resulting in soil pollution, long-term recovery. The degradation time of plastic bags thrown into the ocean is 200-1000 years, and over 100,000 marine animals die every year due to being entangled in plastic or accidentally eating it (Vegter et al., 2014). According to marine conservation organizations, plastic has been found in over 60% of seabirds and 100% of sea turtles (Isangedighi et al., 2020). Finally, microplastics are also a risk factor that cannot be ignored, as they may be passed along the food chain and accumulate in the human body, where they are difficult to digest and break down, thus causing irreversible damage to the human body (Tong et al., 2022).

Since there is no established definition of "biodegradable", there are still some misconceptions among the public about BPs, such as equating bio-based plastics with BPs, equating BPs with fully BPs, and equating industrially compostable plastics with plastics that can biodegrade in the natural environment. The confusion of consumers about these plastics and the inability to properly dispose of them can lead to ineffective recycling of BPs and even more serious environmental pollution. Therefore, accurately distinguishing bioplastics and establishing a standard system for bioplastics play a crucial role in environmental protection and sustainable development of the bioplastics industry.

As defined by the European Bioplastics, biobased plastics are polymeric materials whose raw materials are partially or fully derived from renewable biomass (plants). When bio-based plastics are burned after use, the biomass-derived carbon in them becomes CO_2, which is reconverted to biomass through photosynthesis (Mülhaupt, 2013). It is clear from the definition that bio-based plastics do not require consideration for biodegradability. Therefore, not all bio-based plastics are biodegradable or compostable. And since petrochemical raw materials do not contain ^{14}C due to their longevity, ^{14}C can be used to determine the carbon content of bio-based plastics according to ASTM D6866 standard to evaluate their environmental value.

Unlike bio-based plastics, which are defined and classified based on the origin of the material, BPs are classified in terms of the end-of-life of the material. BPs are plastics that are degraded under natural conditions (e.g. soil, sand, seawater, etc.), or under specific conditions (e.g. composting conditions, anaerobic digestion conditions or aqueous cultures, etc.), caused by microbial action (e.g. bacteria, molds, fungi and algae, etc.) and eventually turned into CO_2, H_2O, CH_4 and biomass. Figure 2.1 demonstrates the life cycle of BPs.

Figure 2.1: Biodegradable plastics life cycle (Adapted from European Bioplastics, 2020).

According to its working definition, BPs can be bio-based or non-bio-based. Currently, BPs can be divided into two categories based on their raw material sources: bio-based BPs and petroleum-based BPs. Bio-based BPs include plastics obtained by direct processing of natural materials (e.g. starch-based BPs, cellulose-based BPs), polymers obtained by a combination of microbial fermentation and chemical synthesis (e.g. PLA) and polymers directly synthesized by microorganisms (e.g. PHA), etc. (Ahmed et al., 2018). Petroleum-based BPs are usually produced by the polymerization of petrochemical monomers through chemical synthesis (e.g. PBS and PBAT) (Ahmed et al., 2018).

In 2020, bioplastics production is dominated by Asia and Europe. The main producers of bioplastics are Asia and Europe, which produce 956kt and 551kt respectively, accounting for 46.9% and 27.3% of global production which are followed by North and South America (Moshood et al., 2022). And The share of production capacity in Europe is expected to increase in the next five years (Moshood et al., 2022). Figure 2.2 displays the global production capacities of bioplastics in 2020.

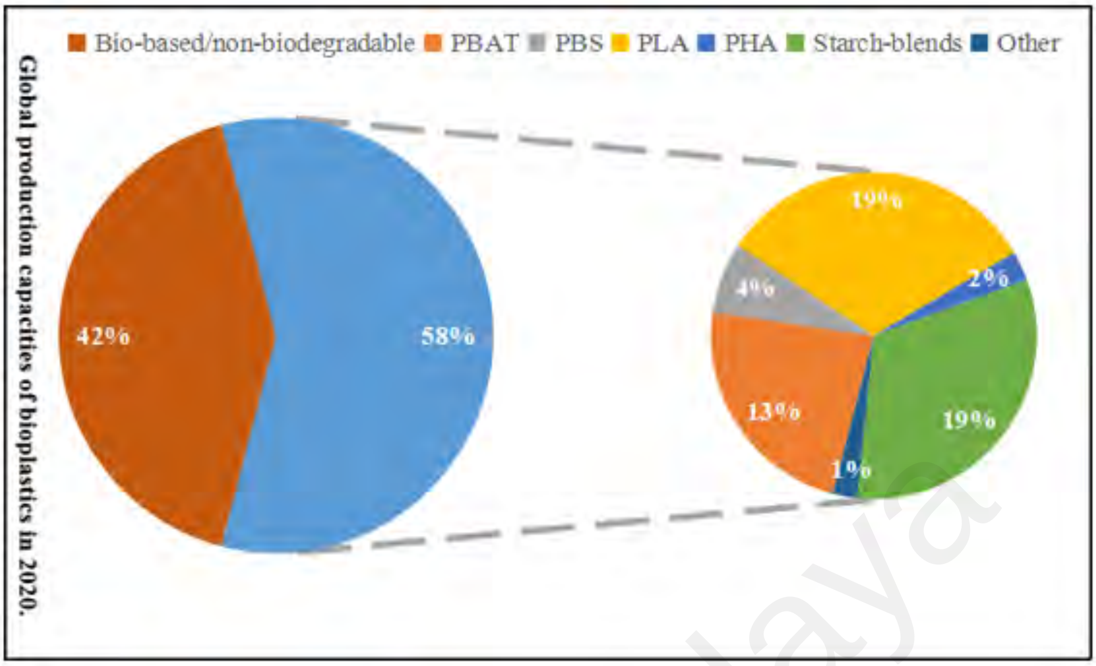

Figure 2.2: Global production capacities of bioplastics in 2020 (Adapted from European Bioplastics, 2020).

Compostable plastics are biodegradable plastics, a subset of biodegradable plastics, which can be converted to CO_2, H_2O and biomass under composting conditions within a certain period of time (Kale et al., 2007). And the final compost should comply with the relevant standards and pass tests such as heavy metal content and toxicity. Currently, compostable plastics on the market are basically biodegradable plastics under industrial composting conditions. Therefore, if they are left unattended in the natural environment (e.g., soil and seawater), these plastics degrade very slowly and cause essentially the same amount of pollution to the environment as conventional plastics.

In this study, the working definition for biodegradable is the ability of things that can be degraded under natural conditions (e.g. soil, sand, seawater, etc.), or under specific conditions (e.g. composting conditions, anaerobic digestion conditions or aqueous cultures, etc.), caused by microbial action (e.g. bacteria, molds, fungi and algae, etc.) and eventually turned into CO_2, H_2O, CH_4 and biomass. Biodegradable does not restrict the origin of things, so things can be both bio-based and non-bio-based. However, biodegradable things have end-of-life constraints, therefore photodegradable, thermodegradable, oxygen-degradable, and non-biodegradable things cannot be included. Therefore, the term "biodegradable plastic" in this study will only cover bio-based biodegradable plastics (e.g. PBAT, PBS and PCL) and fossil fuel-based biodegradable plastics (e.g. PLA, PHA, starch-based BPs and cellulose-based BPs).

2.2 Bio-based BPs

Compared to petroleum-based BPs, bio-based BPs are made from natural biomass and offer superior environmental benefits. In the following part of this paper, researches related to bio-based BPs will be elaborated.

With the increasing awareness of environmental protection and the implementation of national policies, BPs are taking up a growing share of the market and their applications are becoming increasingly widespread.

Table 2.1 compares the performance indexes of different BPs products. Petroleum-based BPs perform better than bio-based BPs in terms of glass transition temperature and Elongation at break. However, bio-based BPs exhibit better biodegradability. It can be seen that the performance of BPs must be improved in order to achieve wide application of BP.

Table 2.1: Comparison of performance indicators of different BPs products.

Name of BPs products	Glass Transition Temperature / ℃	Melting Point / ℃	Tensile Strength /MPa	Elongation at break / %	Density / ($g\cdot cm^{-3}$)	Vapor Barrier Property	Oxygen Barrier Property	Degradation Rate	Market Application Level	References
polybutylene Adipate Terephthalate (PBAT)	-30	110~120	20~30	477~458	1.25	Poor	Poor	Medium	High	(Deng, Yu, Wongwiwattana, & Thomas, 2018)
Polybutylene Succinate (PBS)	-32	114	33~40	400	1.26	/	/	Fast	High	(L. Wang, Zhang, Lawson, Kanwal, & Miao, 2019)
Polycaprolactone (PCL)	-60	60	10.5~ 16.1	800~1000	1.15	General	/	Low	Low	(Leonés et al., 2020)
Polylactic Acid (PLA)	60	177~180	45~60	8~12	1.21	General	General	Medium	High	(H. T. H. Nguyen, Qi, Rostagno, Feteha, & Miller, 2018)
Polyhydroxyalkanoate (PHA)	2~8	150~175	15~40	1~2	1.25	Good	Good	Fast	Medium	(Bugnicourt, Cinelli, Lazzeri, & Alvarez, 2014)

2.2.1 Polylactic Acid (PLA)

PLA is a biodegradable and environmentally friendly aliphatic polyester. Among the many biodegradable polymer materials, PLA has performance advantages comparable to some engineering plastics (polyethylene and polypropylene), such as high mechanical strength, heat resistance, biodegradability, biocompatibility, bioabsorbability, gloss, transparency, low toxicity and easy processability. It can be processed and molded by extrusion, spinning, uniaxial and biaxial stretching, injection molding, blow molding, etc. (Li et al., 2016; H.-A. Lim, Raku, & Tokiwa, 2005; Zhou, Zhao, & Jiang, 2016). PLA is made from starch extracted from various biomass resources including maize, wheat, cassava, potato, sugar beet and sugar cane (John et al., 2008; 2007; Vink & Davies, 2015) which is converted into glucose, then fermented into lactic acid (LA), and finally condensed or polymerized from LA. PLA can be finally decomposed into H_2O and CO_2 through industrial composting or artificial treatment such as incineration, and the decomposition products are re-involved in the ecological cycle of nature.

The molecular formula of LA is $C_3H_6O_3$, and the molecule contains hydroxyl and carboxyl groups. Due to the chiral form of LA, LA has two optical isomers called D-LA and L-LA (Saeidlou et al., 2012), and therefore also has a variety of types of prepared PLA : 1. Direct condensation of D-LA and or L-LA; 2. Azeotropic dehydration polymerization; 3. Propylene cross ester ring-opening polymerization. Different types of PLA (poly-L-lactide, poly-D-lactide, poly-DL-lactide) will be produced depending on different processes (Monticelli et al., 2011; Saeidlou et al., 2012).

PLA is recognized as the most marketable biodegradable polymer in the world and has a wide range of applications in various fields such as food packaging, construction, agriculture, forestry, paper and medical due to its excellent properties. PLA has good biodegradability and biocompatibility, but in the early stage, it could only be used in the medical field due to its low yield. For example, absorbable sutures, surgical bone nails and other surgical appliances (Castro-Aguirre et al., 2016; Hamad et al., 2015; Langer & Tirrell, 2004). As the production process continues to mature and progress, PLA is gradually becoming more familiar in a wider range of fields, especially in the packaging industry. The high strength, breathability and high elasticity of PLA fibers allow them to be used as textile fibers for quilts, garments, etc. (Mushtaq et al., 2020; Z. Raza et al., 2019; Yang et al., 2019). Its good transparency allows it to be used in agricultural applications such as agricultural land films (J. Li et al., 2018; Thompson et al., 2019). The heat-resistant modified PLA material can also be used on food containers such as plates or cups (Musa et al., 2019; Z. Raza et al., 2019). In addition, PLA films have excellent anti-bacterial and anti-mold properties (Turalija et al., 2016), which allow them to be used as materials for epidemic prevention and control (especially COVID-19, which is currently a global concern), such as disposable masks, medical materials, gloves, protective clothing, etc. To further expand the application and scope of PLA, it is necessary to improve the mechanical properties and functions of PLA such as heat resistance, toughness, hydrolysis resistance, barrier properties, antibacterial properties and bacterial inhibition by using physical and/or chemical modifications.

PLA decomposes in the presence of oxygen and the end products are CO_2 and H_2O, therefore PLA is considered sustainable under aerobic conditions (Hottle, Agüero, Bilec, & Landis, 2016; Krause & Townsend, 2016). But if PLA degrades in a landfill without oxygen, the methane gas it produces is 20 times more harmful to the environment than CO_2 emissions (Wright & Kelly, 2017). PLA does not compost easily in the backyard and is difficult to hydrolyze in the marine environment. Its degradation rates under industrial composting conditions (>60°C, in the presence of O_2 and moisture) are considerable (Chamas et al., 2020). In recent years, a variety of microorganisms (mostly *Actinomycetes*) isolated and purified from soil or water have been found to be effective in degrading PLA. For example, *Amycolatopsis* strain K104-1(Nakamura, Tomita, Abe, & Kamio, 2001), *Amycolatopsis* strain SCM_MK2-4 (Penkhrue et al., 2015), *Pseudonocardia sp.* RM423 (Apinya et al., 2015) and *Trichoderma viride* (Lipsa et al., 2016), etc.

Currently, commercial production of PLA has been achieved. In 2020, the global production of PLA has reached about 395,000 tons, which is 18.7% of the global production of bioplastics. Its production is expected to account for an increasingly high share of about 19.5% of global bioplastics by 2025 (Tiseo, 2021).

2.2.2 Polyhydroxyalkanoate (PHA)

PHA is a generic term for a class of polymeric polyesters synthesized entirely by microorganisms, which have physicochemical properties similar to those of synthetic plastics. PHAs have many excellent properties such as biodegradability and biocompatibility that traditional synthetic plastics do not have (G. Q. Chen & Patel, 2012). Many microorganisms such as *Cupriavidus*, *Pseudomonas*, *Alcaligenes*, *Bacillus* and *Aeromonas*, etc. (Koller, 2018) have been shown to synthesize PHAs and store them in the body when there is an excess of carbon sources but other growth factors are limited (lack of nitrogen, phosphorus, low oxygen, UV radiation, etc.). PHAs are present in microbial cells as spherical particles (100 to 800nm) with a hydrophobic inner core composed of molecular chains of PHAs and an outer covering of hydrophilic enzymes and proteins (Sudesh & Doi, 2000). PHAs are structurally diverse and at least 150 different monomeric structures of PHAs have been identified to date (Choi et al., 2020).

PHAs can be broadly classified into two categories according to the number of carbon atoms of the monomer: one is the short-chain-length (SCL) PHAs composed of 3-5 carbon atoms, such as polyhydroxybutyrate (PHB), polyhydroxyvalerate (PHV), etc.; the other is the medium-chain-length (MCL) PHAs composed of 6-14 carbon atoms, such as polyhydroxyhexanoate (PHHx), polyhydroxyoctanoate (PHO), etc (L. Ge et al., 2016). Among them, SCL PHAs are hard and fragile, while MCL PHAs have good toughness. When they are mixed in different proportions the resulting copolymers will exhibit different hardness and toughness. Because PHAs polymerase generally selects only one of scl-hydroxyacyl coenzyme A or mcl-hydroxyacyl coenzyme A as substrate, a microorganism usually synthesizes only one of these types of PHAs. However, some PHAs polymerases have been found to synthesize scl-mcl PHA using both hydroxyacyl coenzymes A as substrates (Sagong et al., 2018). Microbial synthesis of PHAs is mainly based on the conversion of carbon sources into various hydroxyacyl coenzymes A through a variety of carbon source metabolic pathways, followed by the synthesis of PHAs catalyzed by PHAs polymerase (D.-C. Meng et al., 2014). In addition, PHAs can also be produced through naturally occurring or artificially constructed biosynthetic pathways such as the glycolytic pathway (EMP) and the TCA cycle (Z.-J. Li et al., 2010; D. Tan, Wu, Chen, & Chen, 2014).

Besides excellent biodegradability and biocompatibility, PHAs also have gas-phase isolation, biohistocompatibility, and anti-coagulability, which make them have broad applications in biomedical, agricultural, and food packaging fields (Gadgil et al., 2017). The histological response to PHAs implanted in the body is extremely weak, and they do not adhere to the surrounding tissues within the tissue and are easily removed (Rai, Keshavarz et al., 2011). The modified PHAs material has the property of supporting cell growth, providing an environment for cell growth in a variety of tissues and organs, and most of its degradation products are present in animals without carcinogenicity. Therefore, PHAs are widely used in the fields of cardiovascular, bone, cartilage, nerve conduit, esophagus and skin tissue engineering (Gadgil et al., 2017), controlled drug carrier materials (Peer et al., 2007), and healthcare (Zhang et al., 2018). In agriculture, agricultural films made of PHAs can protect water to improve soil quality and fertility (Hassan et al., 2006), and when PHAs are used as carriers of herbicides and released in the soil in a controlled manner, they can effectively suppress the growth of weeds (Prudnikova et al., 2013), and PHAs also help to improve the resistance of bacteria added to the soil and enhance nitrogen fixation. In the packaging industry, PHAs are mainly used in the production of packaging films for bags, boxes, and paper. In addition, PHAs are also used for the packaging of diapers, feminine hygiene products, shampoo bottles and cosmetics, etc. (Muhammadi et al., 2015).

PHAs can be fully composted in an environment rich in microorganisms and fungi, especially soil. it takes two months for PHAs to decompose in backyards (Meereboer et al., 2020), but their decomposition rate in seawater is much slower, with less than 50% decomposition after six months (Dilkes-Hoffman et al., 2019).

Currently, the production of PHA is far from meeting the market demand and its high production cost (10.34 to 23.5 MYR/kg) greatly limits its application as a green material. In 2020, PHAs will account for only 1.7% of global bioplastics production capacity. However, with the improvement of the fermentation technology of PHAs and the optimization of genetic and metabolic engineering of synthetic bacteria, there should be a breakthrough in the productivity of PHAs in the next 5 years.

2.2.3 Starch-based

As a natural polymer from a wide range of sources, starch is considered to be one of the most promising alternatives to traditional petroleum molecules because of its biodegradable, inexpensive and renewable properties. The main sources of starch are maize, wheat, potato and cassava. The low cost of starch raw materials and the ability to use conventional plastic processing equipment provides good prospects for the industrialization of starch-based BPs.

The size of starch granules ranges from 3 ~ 130 μm, which can be used as a filler to prepare degradable plastics as well as modified to prepare degradable plastics (H. Liu et al., 2009). Starch can be divided into two categories: straight-chain starch and branched-chain starch. Straight-chain starches are mainly linked by α-1,4 glycosidic bonds to form a linear structure; Branched starch is a highly branched structure with straight chains connected by α-1,4 glycosidic bonds and branched chains connected by α-1,6 glycosidic bonds (P. Chen et al., 2009; P. Liu, Chen, Corrigan, Yu, & Liu, 2008). Most natural starches show a semi-crystalline structure consisting of ordered crystalline regions and disordered amorphous regions, with a degree of crystallinity of about 20% to 45% (H.

Liu, Yu, et al., 2009). Branched starch in starch granules is thought to be the key component leading to the crystalline structure (P. Chen et al., 2011; H. Liu, Xie, et al., 2009).

Starch-based BPs have a wide range of applications in food storage, agricultural production, and the medical industry. When starch granules are subjected to both heat and force, the flowability is extremely poor, processing and molding is difficult and the starch polymer itself is brittle, so it is often mixed with other polymers, additives and nano-fillers (Hu et al., 2015). Starch-based films of different origins can act as a barrier to external bacteria and air while regulating the internal gaseous and microbial environment. In addition, it can also slow down the evaporation of water to maintain the quality of the food, thus enabling longer storage times (Syafiq et al., 2020; Vásconez, Flores, Campos, Alvarado, & Gerschenson, 2009). Starch-based agricultural films modified by cross-linking, plasticizing and hydrophobization can effectively control soil moisture and temperature, reduce water and nutrient loss, prevent weed growth and promote early crop maturity (Merino et al., 2018; Otey et al., 1974). The safe, non-toxic and biodegradable properties of starch-based materials allow them to be widely used in the pharmaceutical industry, including as excipients and binders for tablets and in the preparation of starch capsules as well (Vilivalam et al., 2000).

Although all starches are biodegradable, not all composites, additives as well as plasticizers, etc. are. Therefore, the biodegradability of these modifiers determines the biodegradability of the starch blends. Biodegradable starch blends include starch-PLA and starch-PCL blend etc. Under aerobic conditions (composting), the biodegradation rate of the starch-PCL mixture was about 88% in 44 days, while under anaerobic conditions (buried in a landfill), it reached a biodegradation rate of 83% in 139 days (Cho et al., 2011).

The capacity of starch blends currently remains at the forefront of biobased plastics. In 2020, the production capacity of starch blends is the same as PLA, accounting for 18.7% of the global production capacity of bio-based plastics. However, due to the biodegradability of some of the starch-based blends, their production capacity will probably decrease in the future.

2.2.4 Cellulose-based

Cellulose is a non-toxic, biodegradable and renewable natural polymer material widely derived from wood, cotton and leaves, etc. Unlike other bio-based BPs, its raw materials do not compete with the food supply. Therefore, it has great research value and development potential. Cellulose is the main component of plant fiber. Plant fiber has the advantages of low density, high specific strength, renewable, good ecological compatibility and low harm to the human body (Rana et al., 1998).

Cellulose is a linear polymer formed by β-D-glucose subunits linked by B-1,4 glycosidic bonds with a degree of polymerization ranging from roughly 7,000 to 15,000 (Klemm et al., 2005). There are three free hydroxyl groups in the cellulose structure, which have strong reactivity and control the degree of substitution of functional groups reacting with the hydroxyl groups (Klemm et al., 2005). The introduction of different kinds of functional groups into the cellulose structure can produce a variety of materials with excellent properties.

Currently, the more researched cellulose-based BPs is cellulose acetate (CA). Forming materials are the most important applications for cellulosic plastics, such as extruded films, eyeglass frames, electronics, and cigarette filters, etc. (Bilo et al., 2018). With the continuous progress in modification and production process, cellulose and its derivatives will occupy a more important role in various fields such as pharmaceuticals, medical devices, construction materials and apparel (Mostafa et al., 2012).

For the biodegradability of cellulose-based BPs, according to the study, Sponge cloth (70% cellulose, 30% cotton) can biodegrade close to 100% at 84 days under commercial-scale composting conditions (1 m depth and 15.7°C average outdoor temperature) (Adamcová et al., 2017). CA made from fibrous flax achieved a biodegradation rate of 44% in 14 days under municipal solid waste composting conditions (Mostafa et al., 2018).

2.3 Feedstocks for Bio-based BPs

Bio-based BPs are available from a wide range of sources, and their feedstocks can usually be divided into three generations (Wellenreuther & Wolf, 2020). First-generation feedstock mainly refers to edible nature biomass, such as maize, wheat, rice, sugar cane, sugar beet and potato, etc. Second-generation feedstock mainly consists of lignocellulose-based feedstock, including mostly inedible biomass (e.g., wood, wheat straw, sugar cane bagasse, maize stover and palm fruit bunches, etc.), such as by-products obtained from non-food crops or from growing food crops, and municipal waste. Research on third-generation feedstock has focused on microorganisms, such as algae and modified microbial polymers, etc.

Currently, most bio-based BPs are made from first-generation feedstocks and have been partially produced commercially on a large scale. Tapioca starch can be used to create smooth, flexible and strong bioplastics (ÖZDAMAR & Murat, 2018). Bioplastics can also be produced by bacterial sugar assimilation using sugarcane as a substrate (Pohare, Bhor, & Patil, 2017). The production of BPs using first-generation feedstock has the advantages of lower production costs and a more mature production process, but it also has some drawbacks, such as possible competition with the food supply and the potential for more carbon emissions and more pesticides, fertilizers, land and water use from the process of growing and producing raw materials.

Cellulose-based production is significantly lower in terms of greenhouse gas emissions and energy consumption compared to the first generation. The biodegradable composites prepared by blending cellulose and biodegradable resin have the advantages of good biocompatibility, biodegradability, low energy consumption and no pollution of degradation products to the environment, and have great economic potential. However, at this stage, the preparation and cost control of cellulose-based BPs are still the biggest technical challenges in this field. In general, the application of second-generation feedstock can achieve effective sharing with agricultural production and turn waste into treasure, so it still has obvious advantages and broad development prospects.

Microalgae is a very promising alternative source for the production of BPs because of the high growth rate and the fact that they do not compete with food. Currently, the most researched microalgae are *Chlorella* and *Spirulina*. *Chlorella* has a better bioplastic behavior, while *Spirulina* shows better mixing properties (Zeller et al., 2013). In addition to microalgae, polysaccharide-rich aquatic plants such as macroalgae or seaweeds are also potential feedstocks (Thiruchelvi et al., 2021). The biggest advantage of third-generation feedstock is that it greatly improves land use and shortens the growth cycle. However, there are still large research gaps and the production process needs to be further improved.

2.4 Modifications of Bio-based BPs

BPs have been attracting widespread attention due to their environmentally friendly properties such as excellent biodegradability, but they are unable to completely replace conventional plastics in the market due to some functional and performance deficiencies and high costs.

Compared with traditional plastics, BPs do not have an advantage in performance, so the modification of BPs is particularly important. The most widely used methods are physical and chemical modifications. Biological modification is commonly used in the modification of PHAs. Table 2.2 demonstrates the disadvantages as well as the modification methods of some major BPs.

Table 2.2: Application challenges and modification methods of BPs.

Name of products	Application Challenges	Main Modification Methods	References
polybutylene Adipate Terephthalate (PBAT)	Poor thermal stability, mechanical properties and melt crystallinity，insufficient water vapor barrier	Compound modification with other degradable materials (e.g. PLA), nano-fillers (e.g. cellulose nanocrystals) and (e.g. starch) natural polymers	(Pinheiro et al., 2017) (Wei, Wang, Xiao, Zheng, & Yang, 2015)
Polybutylene Succinate (PBS)	Rapid degradation rate, poor mechanical properties, high brittleness	Modified by filling talc, blending with PBAT	(Platnieks et al., 2020) (Su et al., 2019)
Polylactic Acid (PLA)	Poor toughness, processability of blown film and gas barrier, low heat deflection temperature,	Blending with plasticizers (e.g. PEG), nucleating agents, inorganic fillers (montmorillonite), cellulose, other biodegradable materials (e.g. PCL) Co-polymerization, chain expansion, grafting and cross-linking modification	(Puthumana, Santhana Gopala Krishnan, & Nayak, 2020) (Elsawy, Kim, Park, & Deep, 2017)
Polyhydroxyalkanoate (PHA)	Difficult to process, poor thermal stability, easy to hydrolyze; slow crystallization speed, long production cycle; general toughness, poor mechanical properties	Blending with cellulose derivatives, lignin and PLA. Carboxylation, halogenation, hydroxylation, epoxidation and grafting. Co-feeding substrates in the culture medium of bacteria.	(Sharma, Sehgal, & Gupta, 2021) (Z. A. Raza, Riaz, & Banat, 2018)
Starch-Based	Strong brittleness, poor mechanical properties, easy to absorb water	Thermoplastic modification, plasticizer, compounding with cellulose, lignin, chitosan and PLA, etc.	(Jiang, Duan, Zhu, Liu, & Yu, 2020)

As for bio-based BPs, PLA is considered to be a very promising BP due to its excellent performance. However, at the same time, PLA has the disadvantages of high brittleness, poor heat resistance, low impact resistance, low crystallization rate, lack of reactive functional groups and long degradation cycle, etc. (Bastioli, 2001; Bishai et al., 2014). PHAs have good mechanical properties and strength but have the disadvantage of poor thermal stability, strong brittleness and high costs (Bugnicourt et al., 2014). Starch polymers are widely available and affordable, but are inherently brittle at room temperature. Cellulose, a natural polymer, is widely available from natural biomass such as wood, it is non-toxic and biodegradable, but extremely expensive. Therefore, the improvement of processing temperature, crystallization rate, surface structure mechanical properties and degradation properties of various bio-based BPs through different techniques has become a hot topic of current research.

There are three main types of modification methods for bio-based BPs: chemical modification, biological modification and physical modification (Bhatnagar et al., 2013). Chemical modification refers to the generation of graft or block polymers through chemical reactions to improve the physical and chemical properties of the material by changing the interfacial tension between the components to form a compatible system. Copolymerization modification is the most dominant method. It can improve a variety of different properties of polymers such as pore size, hydrophilicity, degradability, crystallinity, mechanical properties, and biocompatibility through the design of the molecular structure and molecular weight size (Li et al., 2016). Most synthesis methods for copolymerization modifications are similar to those for homopolymers and do not require changes in the equipment and apparatus used for production (S. Wang et al., 2005).

Biomodification refers to the introduction of other hydroxyalkanoic acid units in the fermentation process to modify the target product in a targeted manner to produce polymers with different chain compositions (Li, Yang, et al., 2016), but this approach has some limitations in strain selection, carbon source control and synthesis mechanism and is costly. This method is frequently used for the modification of PHAs.

Physical modification refers to the improvement of the thermomechanical properties of the blends by selecting different blending components, adjusting the ratio between components, and making effective blends by solution or melting methods (Li, Yang, et al., 2016). Physical-mechanical blending is a simple, economical and practical modification method, and therefore has received extensive research and attention. Blending modification is one of the most widely used methods. Theoretically, a variety of composites can be produced by blending polymers with different properties with bio-based polymers. However, most of the time, polymers and bio-based polymers are not compatible with each other. Therefore, improving the compatibility and blend performance of incompatible polymers through a compatibilizer is the key to producing blends with excellent performance. Secondly, composite modification is also one of the common physical modification methods. The addition of various composite materials can greatly improve the performance of the material for various applications.

For PLA, the modification is primarily done by physical and chemical methods. copolymerization of lactic acid with glycolic acid monomer allows improvement of properties such as low crystallinity of PLA, making it suitable for medical applications such as surgical sutures and tissue engineering scaffolds (Ugartemendia et al., 1993).

PLA-PEG diblock copolymers modify the hydrophobicity of PLA and contribute to the delivery of hydrophilic drugs (Perinelli et al., 2019). Rathi et al. (2012) synthesized ABA triblock copolymers (PDLA-PLLA-PDLA) based on the difference in stereochemical properties of PLLA and Poly (D-lactide) PDLA, improving the toughness and flexibility of PLA. Commonly used co-blended modifiers for PLA include chitosan (Cs), heparin, hydroxyapatite, polyethylene and thermoplastic biodegradable plastics. Grande et al. (2015) used PLA and polyvinyl alcohol (PVA) as bulking agents and used glycerol to plasticize the whole system, then the PVA/Cs copolymer made by solvent blending method was melt blended with PLA to make ternary melt blends of PLA/PVA-Cs. The addition of a certain amount of small molecular weight substances with high boiling points and low volatility in the process of PLA miscibility can improve the mechanical properties and processing properties of PLA, and the commonly used PLA plasticizers include citrate plasticizers, PEG and LA oligomers, etc. Greco et al. (2016) used cashew phenol acetate extracted from cashew nuts to plasticize and modify PLA to prepare polymers with good miscibility, stability and ductility.

Chemical, biological and physical modifications are all used as common methods for PHAs. PHAs can be modified by adding chemical groups with various properties to the PHA structure. The crystallinity of PHB can be improved by grafting methyl methacrylate monomer onto the main chain of PHB (S. Nguyen & Marchessault, 2004). A multi-block polyurethane based on PHBHHx and PEG was synthesized by melt polymerization using diisocyanate as a linker which improved the hydrophilicity of the material (Z. Li et al., 2009). When *Halomonas bluephagenesis* cell is supplied with carbon sources such as glucose and propionic acid under nitrogen-limited conditions, it produces PHBV copolymer depending on its concentration (Mitra et al., 2020). A variety of $P^{3/4}HB$ with ^{4}HB content can be biosynthesized by different strains and the addition of appropriate carbon sources, and the thermal and mechanical properties of $P^{3/4}HB$ copolymer change greatly with the increase of ^{4}HB content, and the processing properties are also greatly improved (X. Wang et al., & Xu, 2010). As for physical modification, blending modification between PHA and PLA is a relatively common modification means. Burzic et al. (2019) added a 10% to 20% mass fraction of PHA to the PLA base material and greatly improved the impact resistance of the blends. In addition, natural cellulose-based and low-molecular substances have been used for the modification of PHAs. Don et al. (2010) blended starch modified by polyvinyl acetate (PVAC) with PHB to obtain composites, which greatly improved their thermal stability and mechanical strength.

While starch-based materials have many advantages, they also have disadvantages such as poor mechanical properties and high sensitivity to moisture. Modification of starch-based materials to produce starch-based blends or composites can effectively ameliorate these disadvantages and greatly enhance their application range. In order to maintain the advantages of biodegradable starch-based materials, they are blended with biodegradable polyesters and natural reinforcements such as natural fibers and montmorillonite. Compared with inorganic fillers, natural fiber composites offer many advantages, including renewable, low cost and high strength. Its addition can greatly enhance the mechanical properties of starch-based materials and prevent the transmission of water molecules to improve their moisture sensitivity. Wollerdorfer and Bader (1998) compounded cellulose with a wheat starch thermoplastic material to increase its strength by a factor of 4 over the starch material without fibers. Chen et al. (2019) formed starch-protein-nori fiber composites by adding nori, which improved the air permeability as well as the tensile strength of the starch film. By blending or compounding with other natural polymers, starch can not only improve processing properties but also reduce the cost of other natural polymers. Blends of hydrogels are a hot research topic in this area. For example, hydroxypropyl methylcellulose (HPMC) and hydroxypropyl starch (HPS) have good processing properties, mechanical properties as well as good barrier properties to oxygen and water vapor (Ali et al., 2019). Nanomaterials are also a widely popular composite material. Cellulose nanocrystals and starch nanocrystals can be used to improve tensile strength (X. Li et al., 2015). Surface coating and cross-linking are often used to improve the sensitivity of starch-based materials to water. Ge et al. used acrylic acid epoxidized soybean oil (AESO)-based coatings to reduce moisture sensitivity and improve barrier properties of starch-based films (X. Ge et al., 2019).

The main problem faced in the preparation of cellulose-based BPs is the high hydrophilicity of cellulose (Canche-Escamilla et al., 2002). This can reduce the mechanical properties of the BPs and limit their applications. Chemical, physical and biological methods are often used to modify the surface of cellulose to improve its properties. Commonly used chemical modification methods include alkali treatment, maleic anhydride treatment, acetylation, benzoylation and acrylonitrile grafting et al., 2014). Manalo et al. (2015) prepared bamboo fiber polyester composites by alkali treatment of bamboo fibers. Their tensile strength, compressive strength, and stiffness were improved compared to the composites prepared from untreated bamboo fibers. Cold plasma treatment and corona treatment are commonly used methods to physically modify cellulose. The unevenness and roughness of the fiber surface increased after the cold plasma treatment, which facilitated its better dispersion in the matrix. The corona treatment is simple and non-polluting and the surface of the treated cellulose can be modified (Faruk et al., 2012). Enzyme treatment is one of the commonly used methods for biological modification. The selectivity and specificity of the enzyme can effectively remove impurities from cellulose without damaging other molecules.

2.5 Processing of Bio-based BPs

Currently, the market volume of bio-based BPs is still relatively low. The processing of conventional plastics is applicable but not fully applicable to bio-based BPs. In order to successfully commercialize bio-based BPs in various areas of the market from natural biomass feedstocks, and to increase the market penetration of bio-based BPs, it is crucial to improve different procedures in the processing of bio-based BPs. Due to some defects in the properties of bio-based materials, their processing properties need to be improved before they can be processed and molded into bio-based BPs for various applications. Therefore, various additives, fillers, compatibilizers and plasticizers are often used in the modification of bio-based materials. There are several technologies commonly used in the processing and molding of bio-based BPs, such as extrusion, injection molding, blow molding, thermoforming and 3D printing.

2.5.1 Extrusion

Extrusion is an efficient, continuous, low-cost, and widely adaptable molding process. It mainly compresses polymer materials together to form a uniform melt by the rotation of a helical screw (single screw or twin screw). The extrusion process mainly includes material addition, melting and plasticizing, extrusion molding, shaping and cooling, etc.

(Repka et al., 2012). Single and twin screws have their advantages and their use depends on the different materials to be processed and the products to be manufactured. Single screws are commonly used for general polymers, while twin screws are commonly used for compounds and blends (Sakai, 2013).

Extrusion can be used to produce continuous products in various forms such as sheets and plates, as well as for mixing, plasticizing, granulating, coloring and compounding of polymer materials. For bio-based BPs that are easily hydrolyzed, such as PLA and PHB, it is necessary to pre-dry them before processing to avoid the reduction of molecular weight. The polar nature of PLA allows it to generate strong intermolecular forces at low shear rates and has a high activation energy(C. Lee et al., 2020). Therefore, PLA resins can be processed with conventional extruders and general-purpose screws with lower shear rates, as well as extruder screws dedicated to PET polymers. The heaters are usually set at 200 to 210 °C to achieve optimal melt viscosity while ensuring the complete dissolution of the PLA crystals (C. Lee et al., 2020).

2.5.2 Injection Molding

The advantages of the injection molding method are high production speed, high efficiency, automated operation, accurate product size and the ability to form complex-shaped parts, making injection molding suitable for mass production and the production of complex-shaped products. The injection molding process can be roughly divided into six stages: mold closing, glue injection, holding pressure, cooling, mold opening, and product removal. The above processes can be repeated to produce products in the batch cycle (Heim, 2015).

The hygroscopicity and heat sensitivity of fibers can cause a decrease in their mechanical properties and surface carbonization. Therefore, during the injection molding process of natural fibers, it should be avoided that they are heavily exposed to the plasticizing unit of the injection molding machine and adjust the processing parameters to minimize the circulation and residence time of the material (Feldmann & Fuchs, 2016). Gunning et al. (2013) used a twin-screw extruder to make composites of natural fibers and PHB, which were further investigated for their mechanical properties as well as degradability by injection molding. In the experiments, fiber agglomerates impeded the melt flow and resulted in low melt flow velocity, which affected the performance of the composites in the injection molding process. Therefore, the construction of the injection molding machine must be adjusted to the high viscosity of the natural fiber-reinforced composite.

2.5.3 Blow Molding

Blow molding is a method of forming hollow products by blowing hot molten blanks closed in a mold with the pressure of gas. Blow molding is widely used in the production of containers for packaging food, beverages, cosmetics, pharmaceuticals and daily necessities. All plastic bottles are made from thermoplastic materials by blow molding. Extrusion blow molding is mainly used to process unsupported blanks; Injection blow molding is mainly used for the processing of blanks supported by metal cores; Stretch blow molding can process biaxially oriented products, greatly reducing production costs and improving product performance.

Preheating is required for the blow molding of polymers before they enter the stretch molding process. Subjecting to shear and high temperatures in the extruder may result in low melt strength of PLA and PHA. Therefore, processing the polymers at low extrusion temperatures with melt strength enhancers can be used to improve this process. Studies have shown that PLA blow molding processing under the above conditions can lead to high melt viscosity and stable blown film production (Mallegni et al., 2018).

2.5.4 Thermoforming

Thermoforming is a plastic processing method in which thermoplastic sheets are processed into various products. The sheet is clamped to a frame and heated to its glass transition temperature, and under the action of an external force, it is pressed against the mold to obtain a shape similar to that of the mold. After cooling and shaping, the product is formed by trimming. Thermoformed products include cups, plates, food trays, and automotive parts, etc.

The standard method for producing PLA containers such as cups and food trays is thermoforming. The thermoforming temperature of PLA sheets is similar to that of aluminum, around 80-110 °C (Castro-Aguirre et al., 2016). Therefore, aluminum molds can be used for the thermoforming of PLA. Compared to the original PLA sheet, the thermoformed PLA has more toughness and better drop impact performance (Castro-Aguirre et al., 2016).

2.5.5 Other Processes

In addition to the above processes, 3D Printing and foaming are often used in the production of BPs. Speed of production is a major advantage of 3D printing, which allows a complex design to be uploaded through a model and printed out in a short time. 3D printing technology can be used in combinatorial engineering. Du et al. grafted maleic anhydride (MA) to PHA and compounded it with coupling agent-treated palm fiber (TPF) to make a PHA-g-MA/TPF composite, which can be used as a 3D printed filament for tissue scaffold structures (Du, Fu, & Zhu, 2018).

Foaming is the process of creating microporous structures in plastics. Almost all thermoset and thermoplastic plastics can be made into foam. Environmentally friendly starch-based foam materials can replace Replacing traditional plastic foaming materials such as PS, PE and PU, etc. Meng et al. (2019) investigated the relationship between phase transition and foaming properties of starch-based materials. The higher water content enhances the melt strength, and a large amount of water in the evaporation process reduces the temperature, resulting in a closed pore structure. The closed pore structure prevents the evaporation of water in the pore during the foaming process, which makes the negative pressure in the pore when the foaming material is cooled, resulting in the shrinkage of the foaming material and reducing the foaming rate. In order to reduce transportation costs, the foam is usually produced using a two-step foaming process. The starch masterbatch is first made for easy delivery and then foamed to increase the volume using a simple single-screw extruder (Duan et al., 2019).

2.6 Sustainability

In theory, the raw materials for BPs are renewable, the manufacturing process is more environmentally friendly and energy-efficient than petroleum-based plastics and the products can be composted or recycled. However, the BPs that have been commercially produced are currently struggling to reach full sustainability. Multiple factors combine to influence the sustainability of BPs. Cost, product performance, biodegradability, environmental and economic benefits, and land use are all very important determinants.

The lifetime of BPs products depends on their performance as well as their functionality. Most of the products made of bio-based raw materials have disadvantages such as poor mechanical properties and poor heat resistance. In order to maintain the renewable nature of raw materials, natural polymers such as starch, cellulose, lignin and chitin are used as composites and additives for bio-based BPs, which greatly increase the durability and application range of BPs.

Although the raw materials for bio-based BPs are derived from renewable resources, currently the feedstocks are mostly land-grown crops such as maize, sugarcane, wheat and potatoes. This may lead to competition with the food supply. According to data provided by Bioplastics Europe (2020), land use for bioplastics production is currently minimal and there is no competition with food. But with the growth of the world's population and the growing demand for biobased plastics, land use remains an issue that needs to be considered. In addition, the process of growing agricultural products may generate a lot of water use and may produce more carbon emissions. The use of fertilizers or pesticides may also accelerate soil degradation. As a result, second and third-generation feedstocks such as agricultural by-products, lignocellulose and microalgae, as mentioned above, are beginning to attract attention (Jain & Tiwari, 2015). They reduce production costs and competition with land for food crops, increase waste utilization, and have good prospects for development.

The disposal of BPs products at the end of their useful life (e.g., recycling and composting) is critical to ensuring the sustainability of BPs. Improper handling may lead to more serious contamination. For example, PLA and PET are very similar in appearance and difficult to distinguish, and the mixing of PLA into the PET recycling stream can significantly reduce the quality and yield of PET recycling (Papong et al., 2014). Secondly, the biodegradation rate, as well as compostable conditions vary among BPs. If they are not sorted but mixed together, BPs may not degrade effectively and may even produce harmful gases such as CH_4. Therefore, it is essential to provide consumers with identifiable labels, explore efficient plastic separation methods (e.g., near-infrared spectroscopy), and establish robust waste disposal systems. At present, the research on the biodegradability of BPs, especially some blends, is still incomplete and needs to be further explored in order to explain the biodegradability of different BPs products to consumers and to achieve effective classification and recycling of BPs.

In conclusion, the sustainability of BPs needs further research and examination. How to increase the environmental benefits of BPs and approach full sustainability should become critical points to be considered in the current production of BPs products.

2.7 Summary

Various types of BPs have different characteristics. The most important types of bio-based BPs such as PLA and PHA already have various applications in the market. They differ in terms of parameters and applications. The key parameters and recent study of the bio-based BPs are summarized in Table 2.3 and 2.4. It can be seen that PLA and PHA have the relatively better performance. Starch-based BPs have a significant advantage in terms of price. On the contrary cellulose-based BPs are too expensive and therefore are generally used only in medical applications.

Table 2.3: Summary of the main parameters of bio-based BPs.

Bio-Based BPs	Classification	Feedstocks	Monomer/Sub-Unit	Production Technology	End of Life	Performance Characteristics	Applications	References
Polylactic Acid (PLA)	Aliphatic polyester	Maize, cassava, sugarcane, sugar beet	Lactic acid (L- and D-Isomers)	Ring opening polymerisation technique, polycondensation	Industrial composting, mechanical and chemical recycling	Good transparency, low heat deflection temperature, high brittleness, easy deformation, good biocompatibility	Film, medical product packaging	(Auras, Lim, Selke, & Tsuji, 2011) (Cheng, Deng, Chen, & Ruan, 2009) (Garlotta, 2001)
Polyhydroxyalkanoate (PHA)	Aliphatic polyester	Maize, sugar, vegetable oil	Depending on the sub type	Microbial fermentation	Industrial and home composting; biogas installation	High heat deflection temperature, poor thermal stability; high brittleness	Disposables, bags, medical sutures	(Zhao, Deng, Chen, & Chen, 2003) (Zinn, Witholt, & Egli, 2001) (S. Y. Lee, 1996)
Starch-Based	Polysaccharide	Maize, Wheat, Potatoes, Cassava	D-glucose	Naturally occurring	Industrial or home compost	Low cost, poor mechanical properties, high viscosity, high hydrophilicity	Disposable bags, cutlery	(Xiong, Tang, Tang, & Zou, 2008) (Lörcks, 1998) (Shogren, Fanta, & Doane, 1993)
Cellulose-Based	Polysaccharide	Wood, cotton, leaves	β-D-glucose	Naturally occurring	Home composting, industrial composting, anaerobic digestion	Low density, high specific strength, high hydrophilicity, high elasticity	Cellophane, cigarette filters, coatings	(Pandey et al., 2005) (Bledzki & Gassan, 1999)

Table 2.4: Recent study on biodegradable plastic production and its applications.

Name of product	Global Capacity (2020)/(MT)	Estimated Global Capacity (2025)/(MT)	Applications	References
polybutylene Adipate Terephthalate (PBAT)	0.28	0.40	Packaging films, shopping bags, agriculture mulch films and sheets, plant pots, hygiene products	(European Bioplastics, 2020) (Ferreira, Cividanes, Gouveia, & Lona, 2019)
Polybutylene Succinate (PBS)	0.09	0.09	Waste bags, agricultural mulch films, packaging (wrapping) films, disposable packaging and tableware	(European Bioplastics, 2020) (Rujnić-Sokele & Pilipović, 2017)
Polylactic Acid (PLA)	0.39	0.55	packaging (cups and bowls, foils), textiles (T-shirts and furniture textiles), nappies, foils for agriculture and cutlery, injection stretch blow molded bottles and jars, 3D printing	(European Bioplastics, 2020) (Di Lorenzo & Androsch, 2018)
Polyhydroxyalkanoate (PHA)	0.04	0.33	packaging of films, blow molded bottles, coating on paper, medical applications (medical sutures)	(European Bioplastics, 2020) (Kalia, Ray, Patel, Singh, & Singh, 2019)
Starch Blends	0.39	0.40	loose-fill packaging, shopping bags, refuse sacks, thermoformed trays, cosmetics products	(European Bioplastics, 2020) (Gadhave, Das, Mahanwar, & Gadekar, 2018)
Other (biodegradable)	0.03	0.03	Cellulose acetate (CA) can be used to form molded solid plastics, cigarette filters, coatings, photographic-films and filters	(Tu, Zhu, Duan, & Zhang, 2021) (European Bioplastics, 2020)

CHAPTER 3: MATERIALS AND METHODS

3.1 Introduction

Based on a search and review of the literature in Google Scholar, NCBI, Science Direct, and Web of Science. six natural biomasses were chosen to be the alternatives for the production of BPs, they are maize, rice, cassava, potato, sweet potato and wheat. In this study, literature review was also used to select the criteria to evaluate the priority of natural biomass feedstocks for the production of BPs.

Among the multi-criteria decision-making methods, the Analysis Hierarchy Process (AHP) is widely used because of its advantages of system, flexibility and simplicity. In this study, the AHP was used to determine the weighting of the criteria for the production of biodegradable plastics and the prioritization of natural biomass to ultimately arrive at the most suitable natural biomass substrate for the production of biodegradable plastics. The approximate process is shown in the Figure 3.1.

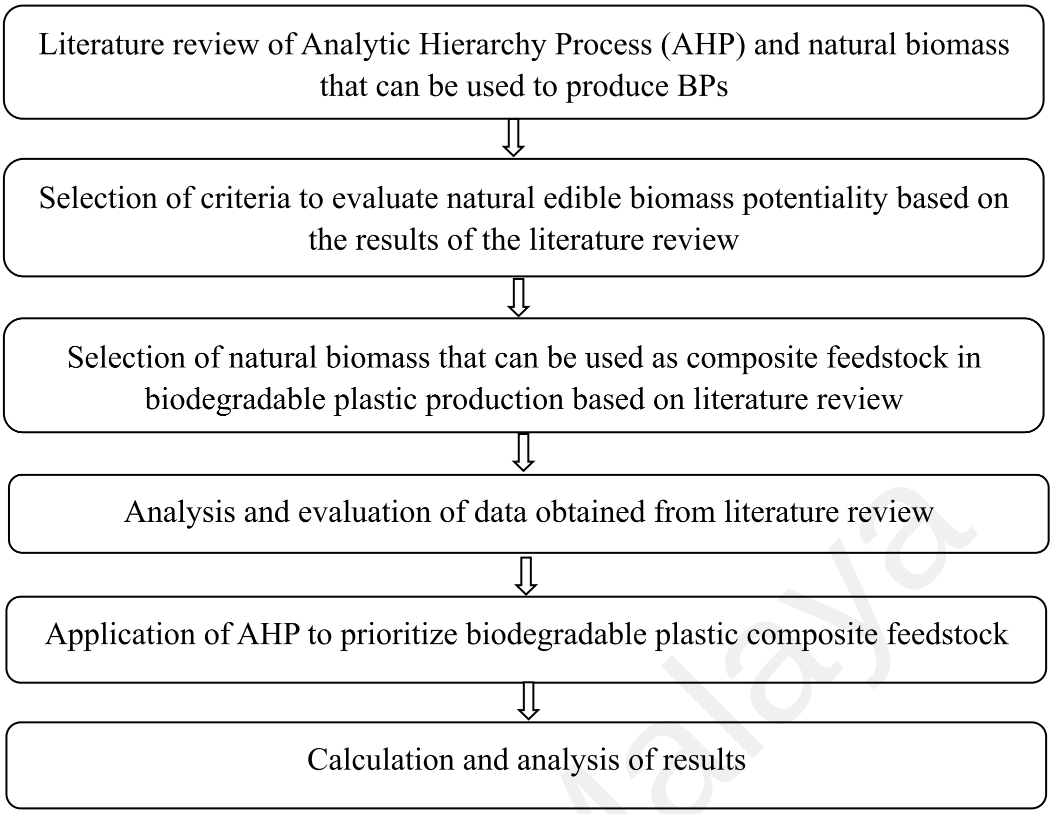

Figure 3.1: Overall flow of methodology

3.2 Methodology

The selection of criteria and natural biomass in this study, as well as their scoring, are based on a review of 129 papers. A literature review exhibits a comprehensive description of a large body of literature, which synthesizes developments and situations over a wide spatial and temporal range, with both vertical descriptions and horizontal coverage (Snyder, 2019).

The literature in this paper was searched from Google Scholar, NCBI's Pubmed database, Science Direct and Web of Science. Use Boolean search method and search by the keyword "natural biomass and biodegradable plastics" to find suitable feedstocks for the production of BPs. Search by keywords "criteria, biodegradable plastics, production impact factors, application impact factors" to find suitable criteria for evaluating feedstocks for BPs production. The time frame for determining the literature on criteria and feedstocks is within 10 years. Subsequently, a search of the literature within 5 years was conducted. A search with each selected natural biomass and each corresponding criterion as a keyword was used as input to the AHP data.

Judgments and scores made using actual data collected from the literature review will not be obviously biased, and will be based on facts and data, and will strive to be objective rather than imaginative.

3.3 Criteria of Natural Biomass

Bio-based BPs can be made by extracting starch, cellulose, oil, lignin, protein and polysaccharides from renewable natural biomass. Most of the bio-based BPs (PLA, PHA, starch-based BPs) in the world are currently made from starch through various processing processes. Therefore, the criteria for judging the natural biomass of starch will be selected in this study. And six criteria were selected to judge the priority of biomass as feedstock in the production of biodegradable plastics in terms of economic, application and environmental considerations. The six criteria are: the cost of producing biodegradable plastics from the natural biomass, the availability of natural biomass for production, the shelf life of natural biomass, the amylose content of starch, the gelatinization temperature of starch, and the water absorption capacity of starch.

3.3.1 Production Cost

The issue of cost is an important criterion for evaluating a product. The extremely low production cost of traditional plastics has resulted in biodegradable plastics currently being far less competitive in the market than traditional plastics. Most businesses will choose non-biodegradable plastics because of the difficulty in accepting the cost of biodegradable plastics. Therefore, the production cost of raw materials and the processing cost of making biodegradable plastics influence the choice of raw materials.

3.3.2 Availability per Hectare

Bio-based biodegradable plastics are generally derived from renewable resources, but the current raw materials are generally land grown crops, and there may be competition for food in the future. Therefore, the availability of natural biomass feedstock affects the sustainability and production efficiency of biodegradable bioplastics

3.3.3 Shelf Life

The procedures for harvesting, transporting, making and processing of natural biomass take time. If the shelf life of the natural biomass is too short, the economic benefits of the BPs produced are greatly reduced. Tuber and root crops, for example, are easily damaged, and their respiration generates heat and reduces the available starch content. Therefore, they have a relatively short shelf life. On the contrary, natural biomass of cereals such as wheat can be stored well and have a longer shelf life after sufficient drying.

3.3.4 Amylose Content

The straight chain starch content affects the size of the starch granules, morphological structure, etc. This affects the thermal properties of the starch and thus the quality of the produced BPs (Jane et al., 1999). For example, the larger the granules of starch, the thicker the BPs produced and the lower the strength, elongation and tensile strength (S.T. Lim et al., 2008). Conversely, the strength, toughness and other properties of the produced BPs are much better.

3.3.5 Crystallinity

The crystallinity of natural starch granules is generally 14%~45% (Singh, Ali et al., 2006). Starch is usually given three crystal structures: type A, type B and type C starch (Zobel, 1988). During gelatinization, the starch loses some of its crystallinity, which is very important for the formation of BPs (García et al., 1998). The lower the crystallinity, the greater the flexibility, impact resistance, filler compatibility and thermal sealing properties of the produced BPs. The difference in the crystallinity of the polymers allows for a wide range of material properties. Therefore, it can be used as a criterion for judging alternatives.

3.3.6 Water Absorption Index

Because starch is a relatively simple polysaccharide, it contains a large number of hydroxyl groups, which are a hydrophilic group and therefore have an absorbent nature. The water absorption capacity is the main limitation for its production of BPs. The higher the water absorption, the worse the product performance. And the reduction of water absorption can increase the shelf life of BPs use (Marichelvam et al., 2019).

3.4 Natural Biomass

3.4.1 Selection of Natural Biomass

According to Analytic Hierarchy Process (AHP), a hierarchical model is built first. The top level is the objective, which is the selection of the optimal natural biomass as a feedstock for the production of biodegradable plastics. The middle level is the criteria for evaluation, and six criteria are included in this study, which are production cost, availability of biomass, product properties, biodegradability of BPs, applications of BPs, and recycling and composting of BPs. The bottom layer is the alternatives (natural biomass). Figure 3.2 depicts the natural biomass selection hierarchy.

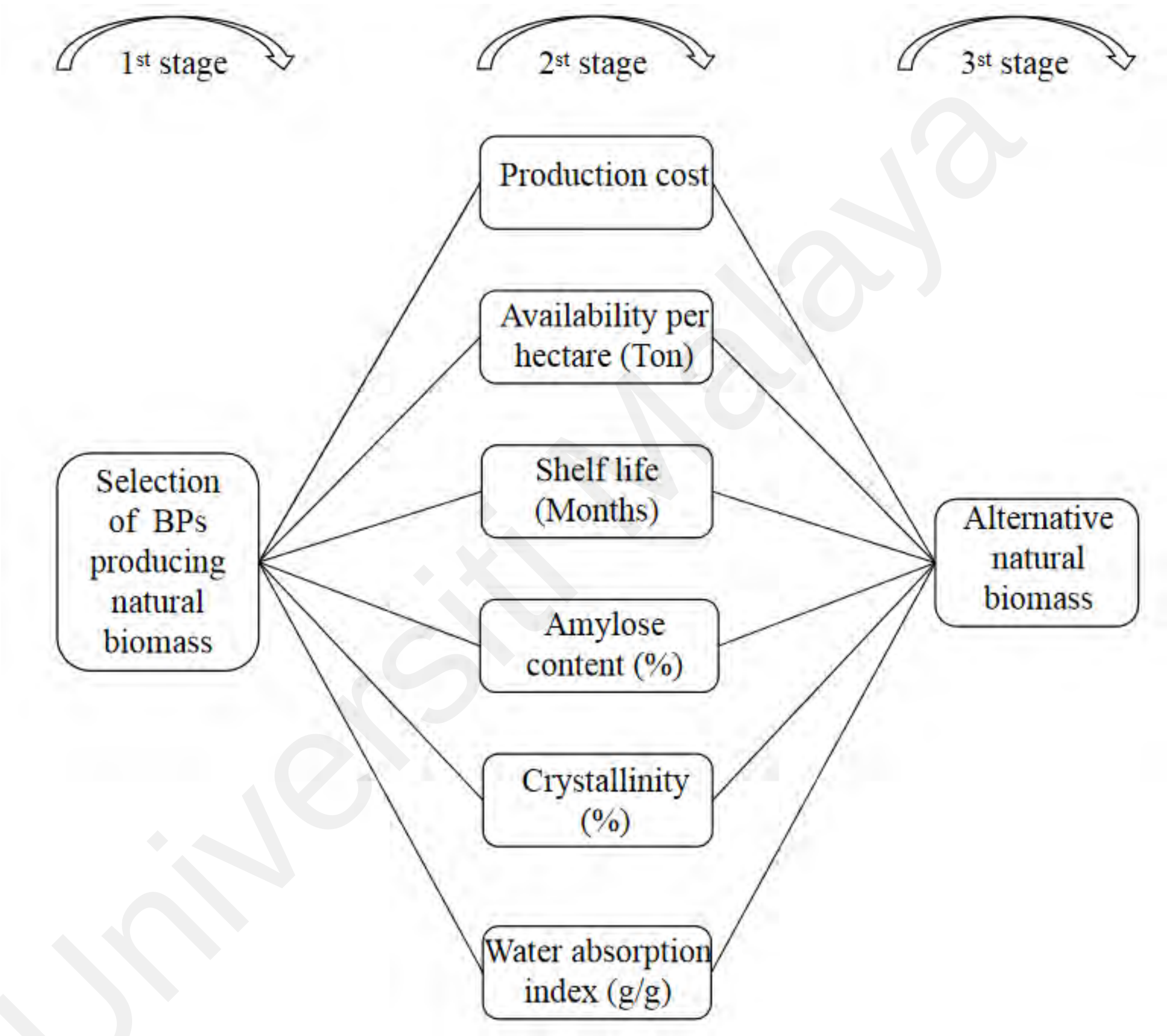

Figure 3.2: Alternative natural biomass selection hierarchy

3.4.2 Natural Biomass for BPs Production (Alternatives)

Natural sources of biomass for the production of BPs are abundant, such as starch, lignocellulose, chitin, algae, etc. However, if different categories of natural biomass are selected, it will be difficult to standardize and establish the criteria. For example, one of the important criteria for starchy natural biomass, straight-chain starch content, cannot be used as a criterion for judging lignin-based natural biomass. In the meanwhile, starch is capable of generating a wider variety of BPs; therefore, the natural biomasses that appear more frequently in the literature review and are starch-based were selected for this study: maize, rice, cassava, potato, sweet potato, and wheat. In the screening of natural biomass 100 literatures were reviewed and the 10 natural biomasses with the highest number of occurrences in the literature are shown in Table 3.1. And the properties of each natural biomass are presented in Table 3.2.

Table 3.1: Top 10 natural biomasses appeared in the literature review

Rank	1	2	3	4	5	6	7	8	9	10
Natural Biomasses	Potato	Rice	Cassava	Maize	Sweet potato	Wheat	Sugar cane	Wood	Cotton	Micro algae
Number	29	22	19	16	16	15	11	8	8	5

Table 3.2: Natural biomasses with properties

Natural biomasses	Plant	Fruit/Stem
Maize		
Rice		
Cassava		
Potato		
Sweet potato		
Wheat		

3.4.2.1 Maize

Maize, commonly known as corn, is an annual monoecious and dioecious pollinated plant with tall, strong stems, and is an important food and fodder crop, as well as the world's most productive crop, second only to rice and wheat in terms of area planted and total production (Dowswell et al., 2019). Maize is rich in protein, fat, vitamins, trace elements, fiber and other nutrients, and has great potential for developing highly nutritious and biologically functional foods (Kumar & Jhariya, 2013). Maize is native to Central and South America. Nowadays it is cultivated all over the world. It is mainly distributed between latitudes of 30° - 50°. The most cultivated areas are in the United States, China, Brazil, Mexico, South Africa, India and Romania (Smith et al., 2004).

The low cost required to produce BPs from maize as a raw material. The Availability of maize per hectare is 8.63 Mt/ha (Langemeier, 2021), the shelf life of maize is 9-12 months (Mushtaq et al., 2020), the amylose content of maize starch is 24-28% (Bertoft, 2017; Nawaz & Arêas, 2013), the crystallinity is 14-39% (Luchese, Spada, & Tessaro, 2017) and the water absorption index is 0.72 g/g (gerçekaslan, 2020).

3.4.2.2 Rice

Rice, also known as paddy, is a food made from rice after the processes of cleaning, hulling, milling and finishing, and is a staple food for more than half of the world's population. Rice contains about 75% carbohydrates, 7%-8% protein, 1.3%-1. 8% fat, and is rich in B vitamins, etc. (Paine et al., 2005). Rice is grown in almost most places except Antarctica. The largest country in the world that grows rice is India, with nearly 50 million hectares under cultivation (Crawford & Shen, 1998).

The cost for the production of BPs from rice starch is high. The Availability of rice per hectare is 3 Mt/ha (Oben et al., 2015; B. T. Tan et al., 2021), the shelf life of rice is 6-8 months (Bandonill et al., 2003), the amylose content of rice starch is 15-35% (Zakaria et al., 2017), the crystallinity is ~38% (Dome et al., 2020) and the water absorption index is 1.51 g/g (Han et al., 2012).

3.4.2.3 Cassava

Cassava is drought and infertile resistant, widely grown in more than 100 countries or regions in Africa, America and Asia, is the main staple food in developing countries, the third largest food crop in the tropics and the sixth largest food crop in the world, known as the "king of starch", and is the ration of nearly 600 million people in the world (Allem, 2002). Edible cassava tubers are rich in starch, which is an important raw material for many pharmaceutical and food industries, and is also an important raw material for the development of biomass sources (Morgan & Choct, 2016).

The cost for the production of BPs from cassava starch is low. The Availability of cassava per hectare is 22 Mt/ha (Ikuemonisan et al., 2020; Peuo et al., 2021), the shelf life of cassava is 3 months (Lestari et al., 2019), the amylose content of cassava starch is 19-22% (Zakaria et al., 2017), the crystallinity is ~13% (Luchese et al., 2017) and the water absorption index is 1.82 g/g (Elisa, 2011).

3.4.2.4 Potato

Potatoes are one of the world's major food crops. It is second only to rice, wheat and maize. The main producers of potatoes in the world are Russia, Poland, China, the United

States, and Mexico (Bradshaw & Ramsay, 2009). Potatoes contain a lot of starch as well as protein, B vitamins, and vitamin C (Lutaladio & Castaldi, 2009). After the potatoes are harvested, they can be stored until the following autumn. Generally, they should be covered with straw, kept away from light, and stored in cool, dry conditions, protected from freezing in winter and from sprouting in spring (Eltawil et al., 2006).

The high cost required to produce BPs from potato as a raw material. The Availability of potato per hectare is 41.95 Mt/ha (Sweden, 2021), the shelf life of potato is 6-8 months (Misra & Kulshrestha, 2003), the amylose content of potato starch is 17-24% (Zakaria et al., 2017), the crystallinity is 23-25% (Dome et al., 2020) and the water absorption index is 2.7 g/g (Lin et al., 2017).

3.4.2.5 Sweet potato

Sweet potato is a short-day crop, not cold-tolerant, with a well-developed root system and relatively drought-resistant. The main production areas of sweet potato in the world are located south of 40°N latitude (Loebenstein, 2009). The cultivated area is the most in Asia, followed by Africa, and the third in America. Sweet potato is rich in nutrition, rich in starch, sugar, protein, vitamins, fiber and various amino acids, is a very good nutritional food (Bovell - Benjamin, 2007). The industries using sweet potato as raw material have spread to more than ten industrial categories such as food, chemical, medical, and paper making, and more than 400 kinds of products made from sweet potato (Loebenstein, 2009).

The cost for the production of BPs from sweet potato starch is low. The Availability of sweet potato per hectare is 6.4-14.89 Mt/Ha (Adeyeye et al., 2021; Ezin et al., 2018), the shelf life of sweet potato is 3 months (Amajor et al., 2014; Jemziya & Mahendran, 2014), the amylose content of sweet potato starch is 18.9% (Nawaz & Arêas, 2013), the crystallinity is 33-34% (Cavalcanti et al., 2019; Kim, 2013) and the water absorption index is 1.82 g/g (Babu, 2014).

3.4.2.6 Wheat

Wheat is a cereal crop that is widely grown around the world. The caryopsis of wheat is one of the staple foods for human beings and is ground into flour to make bread, buns, cookies, noodles and other foods, and fermented to make beer, alcohol, liquor or biomass fuel (Asseng et al., 2015). Wheat is rich in starch, protein, fat, minerals, calcium, iron, thiamin, riboflavin, niacin and vitamin A (Gutierrez-Alamo et al., 2008).

The moderate cost required to produce BPs from wheat as a natural biomass feedstock. The Availability of wheat per hectare is 8.15 metric tons, the shelf life of potato is 4-6 months (Doblado-Maldonado et al., 2012), the amylose content of wheat starch is 20-25% (Zakaria et al., 2017), the crystallinity is 27-36% (Alcázar-Alay & Meireles, 2015) and the water absorption index is 1.02g/g (Han et al., 2012).

3.5 AHP (Analytic Hierarchy Process)

Multi-criteria decision making (MCDM) is a decision to choose among sets of conflicting, non-commensurable options. MCDM allows multiple projects to be judged, ranked and selected for merit. In the decision-making process, each influence factor is given a certain value and a series of information processing is performed to assign a weight to the importance of each factor. Such a decision-making method allows for a quick response to the opinions of decision participants and facilitates the formation of a consistent view.

Depending on the purpose, the MCDM has been extended by multiple methods such as the Analytic hierarchy process (AHP), the Base-criterion method (BCM) (Haseli, Sheikh, & Sana, 2019), Characteristic Objects Method (COMET) (Sałabun, 2015), Simple Multi-Attribute Rating Technique (SMART), Technique for the Order of Prioritization by Similarity to Ideal Solution (TOPSIS), VIKOR method (Opricovic & Tzeng, 2007), Weighted sum model (WSM) and so on.

Analytic Hierarchy Process (AHP) is a decision-making method that decomposes the elements always related to decision making into levels such as objectives, criteria and alternatives, and then performs qualitative and quantitative analysis on top of that, which was proposed by American operations researcher Satie in the early 1970s (Albayrak & Erensal, 2004).

Analytic Hierarchy Process (AHP) has been widely used by decision makers and researchers in various fields since its introduction, and is one of the most widely used methods of multi-quasi-measurement decision making. This approach combines qualitative and quantitative methods to decompose complex systems, mathematizing and systematizing the participants' thinking processes without requiring advanced mathematical knowledge. The degree of influence of each factor in each level on the outcome is quantified, and simple mathematical operations can be used to obtain clear and acceptable results.

The crucial and basic steps of AHP are shown in the figure 3.3.

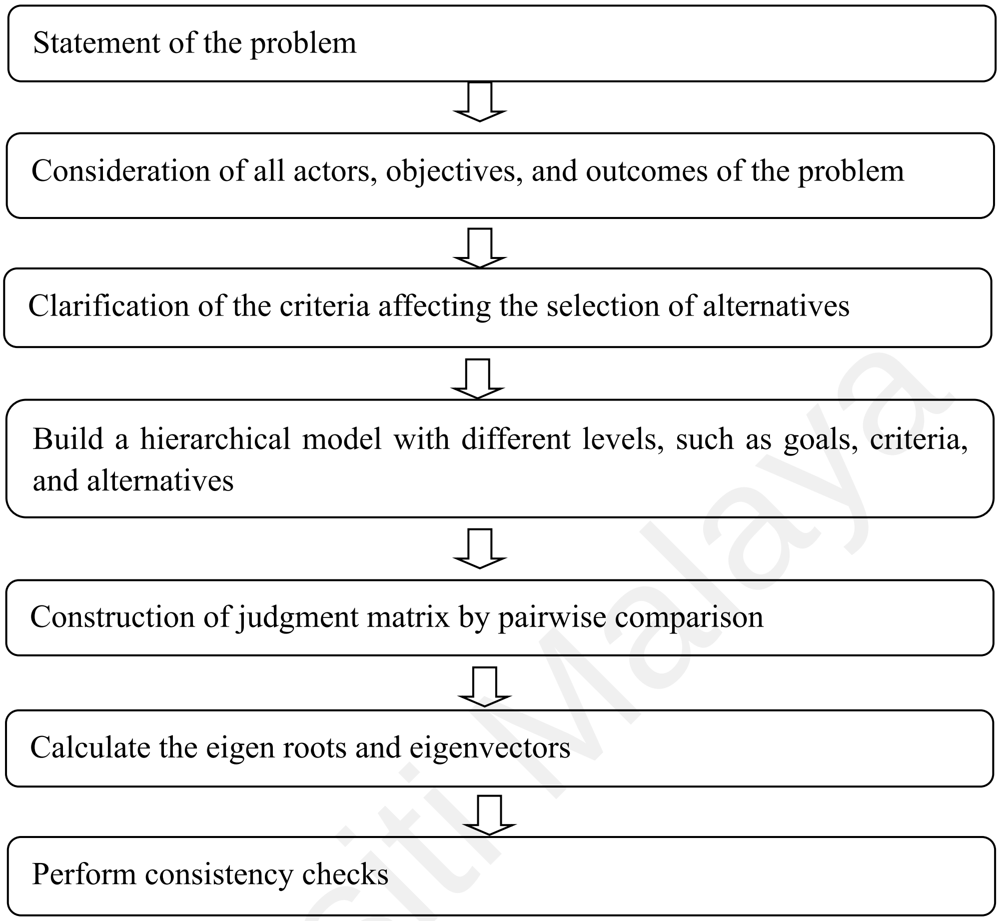

Figure 3.3: Crucial and basic steps of AHP (Vaidya & Kumar, 2006)

Analytic Hierarchy Process (AHP) is widely used by people with the advantages of simplicity and practicality, but also has some disadvantages. First, since AHP selects the optimal solution from the existing alternatives which leads to its lack of innovation and cannot generate new solutions. Secondly, due to the difficulty of fully modeling and mathematically describing the process of human brain consideration, AHP has a more qualitative rather than quantitative component. Furthermore, the order of the matrix gradually increases as the number of indicators or factors increases, which leads to more difficult calculations and inaccuracy of the weights (Macharis et al. 2004; Okudan & Tauhid, 2008).

Analytic Hierarchy Process (AHP) provides a basis for the selection of the best solution by comparing the importance of the factors at each level of the structure in pairs. The process of using AHP is generally summarized in the following steps: 1. Modelling the problem as a hierarchy containing decision objectives, 2. Making a series of judgements to determine priorities between elements of the hierarchy by comparing them in pairs, 3. Calculating the weights of the elements of the structure with respect to the overall goal and establishing priorities, and 4. Checking for consistency and making a final decision based on the above results (Teknomo, 2006).

3.5.1 Building a hierarchy model

The first and critical step of the AHP, which makes a significant impression on the results, is the structure of a hierarchy. When applying AHP to analyze decision problems, the problem needs to be analyzed, hierarchized and constructed into a hierarchical structure. These levels can generally be divided into three categories: the top level (building goals), the middle level (containing criteria on which lower level elements depend), and the bottom level (alternatives). The principle to be followed in the construction of the hierarchy is that the elements in the next higher level need to be used as criteria for comparing elements in lower levels (Thomas L Saaty, 1990). The number of levels in the structure is generally not limited and it relates to the complexity of the problem, etc.

3.5.2 Constructing pairwise comparison judgment matrix

After constructing the structural hierarchy, it is necessary to analyze the relationships among the factors in the system and to construct pairwise comparisons of the importance of the elements of the same level with respect to a criterion in the previous level and using the 1-9 comparison scale shown in Table 3.3 to construct a pairwise comparison array (Thomas L Saaty, 2008).

In order to make scientifically based decisions, the weight vector of the judgment matrix needs to be calculated. The characteristic roots and eigenvectors are calculated by the square root method or sum-product method, and then normalized as weights (Albayrak & Erensal, 2004).

Table 3.3: Standard scale of AHP (Albayrak & Erensal, 2004; Thomas L Saaty, 2008)

Intensity of importance	Description
1	Both elements have the same importance compared to each other.
3	Compared to the two elements, the former is slightly more important than the latter.
5	Compared to the two elements, the former is significantly more important than the latter.
7	Compared to the two elements, the former is strongly more important than the latter.
9	Compared to the two elements, the former is extremely more important than the latter.
2, 4, 6, 8	The intermediate value of the above adjacent judgments.
Reciprocals	If the ratio of the importance of element i to element j is a_{ij}, then the ratio of the importance of element j to element i is $a_{ji} = 1/a_{ij}$.

The size of the paired comparison matrix is determined by the number of criteria. According to the six criteria and six alternatives in this study, six 6/6 pairwise comparison matrices were constructed. The values in the matrices were decided through literature review.

3.5.3 Synthesis of the Priorities

According to the nine importance degrees and their assigned values given by Saaty, a judgment matrix is formed according to the results of pairwise comparison. For example, when comparing the importance of the i element with the j element with respect to the certain element in the upper level, a quantitative relative weight a_{ij} is used to describe it. Let there be n elements involved in the comparison, then $A=(a_{ij})_{nxn}$ is called the pairwise comparison matrix A.

$$A = \begin{bmatrix} a_{11} & a_{12} & \cdots & a_{1n} \\ a_{21} & a_{22} & \cdots & a_{2n} \\ \vdots & \vdots & \cdots & \vdots \\ a_{n1} & a_{n2} & \cdots & a_{nn} \end{bmatrix}$$

Here, $a_{ij} > 0$, $a_{ij}= \frac{1}{aji}$, $a_{ij}=a_{ji}=1$ (i=j).

Based on the judgment matrix, the normalized weight vector (w) corresponding to the largest eigen root λmax is found (Albayrak & Erensal, 2004). The equations (Eq. 1,2,3) are as follows :

$$Aw = \lambda max\ w \qquad (1)$$

$$w = (w_1, w_2, w_3, \ldots, w_n) \qquad (2)$$

$$\lambda max = \sum_{j=1}^{n} \left(a_{ij} \frac{w_j}{w_i} \right) \qquad (3)$$

Theoretically, if A is a perfectly consistent pairwise comparison matrix, the following regularity is obtained:

$$a_{ij}a_{jk}=aik, \ 1 \leq i, j, k \leq n \qquad (4)$$

AHP determines the priority of each alternative by determining the Overall Priority Vector (OPV), and thus selects the optimal alternative. The total score for each alternative is the weighted average of that alternative's scores under each criterion (Albayrak & Erensal, 2004).

3.5.4 Consistency Check

In practice, it is not possible to form perfectly consistent pairwise comparison matrices. Thus, it is only necessary to satisfy that the pairwise comparison matrix has some consistency, which means that a certain degree of inconsistency can exist. This determines the quality as well as the accuracy of the analysis results.

In order to test whether the weight assignments derived from the pairwise comparison matrix are reasonable, the following equation (Eq. 5) is required for consistency testing.

$$CR = \frac{CI}{RI} \quad (5)$$

In the equation, CR is the stochastic Consistency Ratio of the pairwise comparison matrix; CI is the Consistency Index of the pairwise comparison matrix, which is given by the following equation (Eq. 6):

$$CI = \frac{(\lambda max - n)}{(n-1)} \quad (6)$$

RI is the average Random Consistency Index of the pairwise comparison matrix, and the RI values of the judgment matrix of order 1 to 9 are shown in Table 3.4.

When CR < 0.1, the pairwise comparison matrix is judged to have satisfactory consistency, which means that the degree of inconsistency is acceptable; otherwise, the evaluation procedure is repeated and the pairwise comparison matrix is adjusted until satisfactory consistency is achieved (Albayrak & Erensal, 2004).

Table 3.4: Random Consistency Index (RI) (Thomas L. Saaty, 1982)

Matrix size	1	2	3	4	5	6	7	8	9	10
RI	0.00	0.00	0.58	0.90	1.12	1.24	1.32	1.41	1.45	1.49

CHAPTER 4: RESULT AND DISCUSSION

In AHP, the judgment matrix is formed according to pairwise comparisons. Then the eigenvectors corresponding to the largest eigenvalues are found and then normalized as weights. In this study, six criteria and six feedstocks were selected and a three-level hierarchy consisting of objectives, criteria, and alternatives was constructed (Figure 4.1). According to the results of literature review, six natural edible biomasses are maize, rice, cassava, potato, sweet potato and wheat. And six criteria were selected for this study, which were production cost, availability per hectare, shelf life, amylose content, crystallinity and water absorption index.

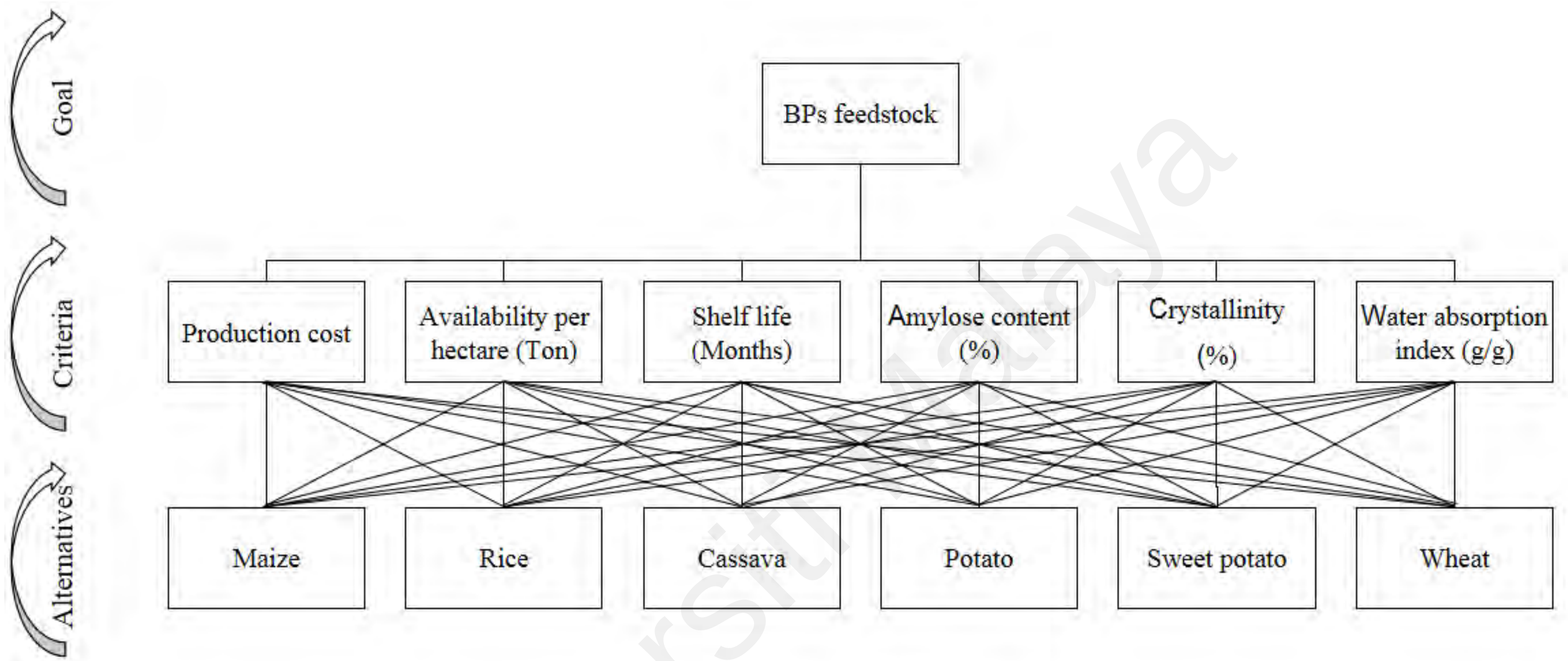

Figure 4.1: AHP Hierarchy

Based on a previous literature review, the properties of different natural biomass corresponding to different criteria were compiled. The decision matric of the natural biomass alternatives is presented as Table 4.1.

Table 4.1: Decision matric of natural biomass

	Criteria					
Alternatives	Production cost	Availability per hectare (Metric Tons)	Shelf life (Months)	Amylose content (%)	Crystallinity (%)	Water absorption index (g/g)
Maize	Low	8.63	9-12	24-28	14-39	0.72
Rice	High	3	6-8	15-35	~38	1.51
Cassava	Low	22	3	19-22	~13	1.82
Potato	High	41.95	6-8	17-24	23-25	2.7
Sweet potato	Very high	6.4-14.89	3	18.9	33-34	1.82
Wheat	Moderate	8.15	4-6	20-25	27-36	1.02

The values in the paired comparison matrix are taken from the literature review. A 6/6 pairwise comparison matrix (Table 4.2) was constructed to evaluate the importance of each criterion and calculate their weight values. The results of the calculation and ranking are shown in Table 4.3 and Figure 4.2.

Table 4.2: Pairwise comparison matrix of criteria

	Production cost	Availability per hectare (Ton)	Shelf life (Months)	Amylose content (%)	Crystallinity (%)	Water absorption index (g/g)
Production cost	1	3	4	4	6	8
Availability per hectare (Ton)	1/3	1	2	2	4	6
Shelf life (Months)	1/4	1/2	1	1	3	5
Amylose content (%)	1/4	1/2	1	1	3	5
Crystallinity (%)	1/6	1/4	1/3	1/3	1	3
Water absorption index (g/g)	1/8	1/6	1/5	1/5	1/3	1

Table 4.3: Results obtained from AHP computations for criteria

Criteria	Criteria Weight	λmax, CI, RI	CR
Production cost	0.4335		
Availability per hectare (Ton)	0.2126		
Shelf life (Months)	0.1325	λmax = 6.1913	
Amylose content (%)	0.1325	CI = 0.0383	0.0309
Crystallinity (%)	0.0585	RI = 1.24	
Water absorption index (g/g)	0.0305		

The key to screening these natural biomasses is to ensure sustainable sourcing of the feedstock. These natural biomasses are widely sourced and produced in large quantities, which have a significant advantage in sustainability compared to other natural biomasses that are produced in smaller quantities. And these selected natural biomasses are more in line with the original purpose of this study to solve the environmental pollution problems caused by traditional plastics and to achieve sustainable development.

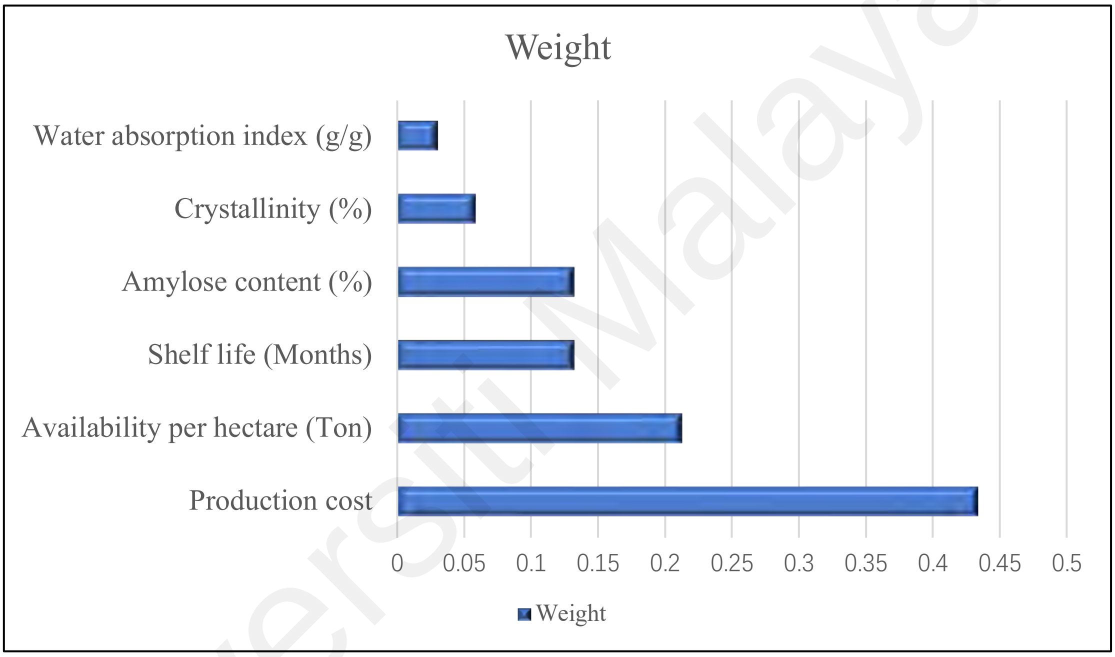

Figure 4.2: Weight of the criteria

The weighting calculations showed that production cost was the most influential criterion in determining the priority of the criteria referred to the selection of natural biomass for the production of BPs. And the water absorption index is the least influential criterion. The calculated weights of each criterion are ranked from highest to lowest as follows: Production cost (0.4335), Availability per hectare (0.2126), Shelf life (0.1325), Amylose content (0.1325), Crystallinity (0.0585) and Water absorption index (0.0305). The consistency index (CR) of the criteria pairwise comparison matrix is 0.0309. The consistency ratio (CR) of the pairwise comparison matrix of the criteria is 0.0309, which is less than 0.1 (standard ratio), so the judgment of the priority of the criteria passes the consistency test.

The pairwise comparison matrices of alternatives (natural biomass) for different criteria and their calculations are shown in Tables 4.4-4.9.

Table 4.4: Pairwise comparison matrix of BPs producing natural biomass for production cost

	Maize	Rice	Cassava	Potato	Sweet potato	Wheat	Priority vector
Maize	1	6	1	5	8	3	0.3401
Rice	1/6	1	1/6	1/2	3	1/4	0.0531
Cassava	1	6	1	5	8	3	0.3401
Potato	1/5	2	1/5	1	4	1/3	0.0782
Sweet potato	1/8	1/3	1/8	1/4	1	1/6	0.0278
Wheat	1/3	4	1/3	3	6	1	0.1607

(λmax = 6.2185, CI = 0.0437, RI =1.24, CR = 0.035 ≤ 0.10 OK)

For production costs, maize (0.3401) and cassava (0.3401) are the best choices, which isfollowed by wheat (0.1607), potato (0.0782), rice (0.0531) and sweet potato (0.0531).

Table 4.5: Pairwise comparison matrix of BPs producing natural biomass for availability per hectare

	Maize	Rice	Cassava	Potato	Sweet potato	Wheat	Priority vector
Maize	1	2	1/4	1/6	1/3	1	0.0626
Rice	1/2	1	1/7	1/9	1/6	1/2	0.0336
Cassava	4	7	1	1/3	2	4	0.2335
Potato	6	9	3	1	4	6	0.4512
Sweet potato	3	6	1/2	1/4	1	3	0.1564
Wheat	1	2	1/4	1/6	1/3	1	0.0626

(λmax = 6.1287, CI = 0.0257, RI =1.24, CR = 0.0207 $\leq$ 0.10 OK)

For availability per hectare, potato (0.4512) is the best choice, which is followed by cassava (0.2335), sweet potato (0.1564), maize (0.0626), wheat (0.0626) and rice(0.0336).

Table 4.6: Pairwise comparison matrix of BPs producing natural biomass for shelf life

	Maize	Rice	Cassava	Potato	Sweet potato	Wheat	Priority vector
Maize	1	3	6	3	6	4	0.415
Rice	1/3	1	4	1	4	2	0.1865
Cassava	1/6	1/4	1	1/4	1	1/3	0.0489
Potato	1/3	1	4	1	4	2	0.1865
Sweet potato	1/6	1/4	1	1/4	1	1/3	0.0489
Wheat	1/4	1/2	3	1/2	3	1	0.1142

(λmax = 6.1083, CI = 0.0217, RI =1.24, CR = 0.0175 $\leq$ 0.10 OK)

For shelf life, maize (0.415) is the best choices, which is followed by rice (0.1865), potato (0.1865), wheat (0.1142), cassava (0.0489) and sweet potato (0.0489).

Table 4.7: Pairwise comparison matrix of BPs producing natural biomass for amylose content

	Maize	Rice	Cassava	Potato	Sweet potato	Wheat	Priority vector
Maize	1	1/2	3	2	4	2	0.2118
Rice	2	1	6	5	7	5	0.4463
Cassava	1/3	1/6	1	1/2	2	1/2	0.0686
Potato	1/2	1/5	2	1	3	1	0.1145
Sweet potato	1/4	1/7	1/2	1/3	1	1/3	0.0442
Wheat	1/2	1/5	2	1	3	1	0.1145

(λmax = 6.0815, CI = 0.0163, RI =1.24, CR = 0.0131 $\leq$ 0.10 OK)

For amylose content, rice (0.4463) is the best choices, which is followed by maize (0.2118), potato (0.1145), wheat (0.1145), cassava (0.1145) and sweet potato (0.0442).

Table 4.8: Pairwise comparison matrix of BPs producing natural biomass for crystallinity

	Maize	Rice	Cassava	Potato	Sweet potato	Wheat	Priority vector
Maize	1	5	1/3	1/2	3	4	0.1597
Rice	1/5	1	1/9	1/8	1/3	1/2	0.0303
Cassava	3	9	1	2	7	8	0.4136
Potato	2	8	1/2	1	6	7	0.2868
Sweet potato	1/3	3	1/7	1/6	1	2	0.0655
Wheat	1/4	2	1/8	1/7	1/2	1	0.0441

(λmax = 6.1404, CI = 0.0281, RI =1.24, CR = 0.0227≤ 0.10 OK)

For crystallinity, cassava (0.4136) is the best choices, which is followed by potato (0.2868), maize (0.1597), sweet potato (0.0655), wheat (0.0441) and rice (0.0303).

Table 4.9: Pairwise comparison matrix of BPs producing natural biomass for water absorption index

	Maize	Rice	Cassava	Potato	Sweet potato	Wheat	Priority vector
Maize	1	4	5	9	5	2	0.4037
Rice	1/4	1	2	6	2	1/3	0.1299
Cassava	1/5	1/2	1	5	1	1/4	0.0819
Potato	1/9	1/6	1/5	1	1/5	1/8	0.0246
Sweet potato	1/5	1/2	1	5	1	1/4	0.0819
Wheat	1/2	3	4	8	4	1	0.278

(λmax = 6.2148, CI = 0.043, RI =1.24, CR = 0.035≤ 0.10 OK)

For water absorption index, maize (0.4037) is the best choices, which is followed by wheat (0.278), rice (0.1299), cassava (0.0819), sweet potato (0.0819) and potato (0.0246).

Table 4.10: Result of priority determination calculation of biofuel producing plants

Alternatives	**Criteria**						
	Production cost (0.4335)	Availability per hectare (Ton) (0.2126)	Shelf life (Months) (0.1325)	Amylose content (%) (0.1325)	Crystallinity (%) (0.0585)	Water absorption index (g/g) (0.0305)	Overall priority Vector (OPV)
Maize	0.3401	0.0626	0.415	0.2118	0.1597	0.4037	0.2654
Rice	0.0531	0.0336	0.1865	0.4463	0.0303	0.1299	0.1197
Cassava	0.3401	0.2335	0.0489	0.0686	0.4136	0.0819	0.2393
Potato	0.0782	0.4512	0.1865	0.1145	0.2868	0.0246	0.1872
Sweet potato	0.0278	0.1564	0.0489	0.0442	0.0655	0.0819	0.064
Wheat	0.1607	0.0626	0.1142	0.1145	0.0441	0.278	0.1243

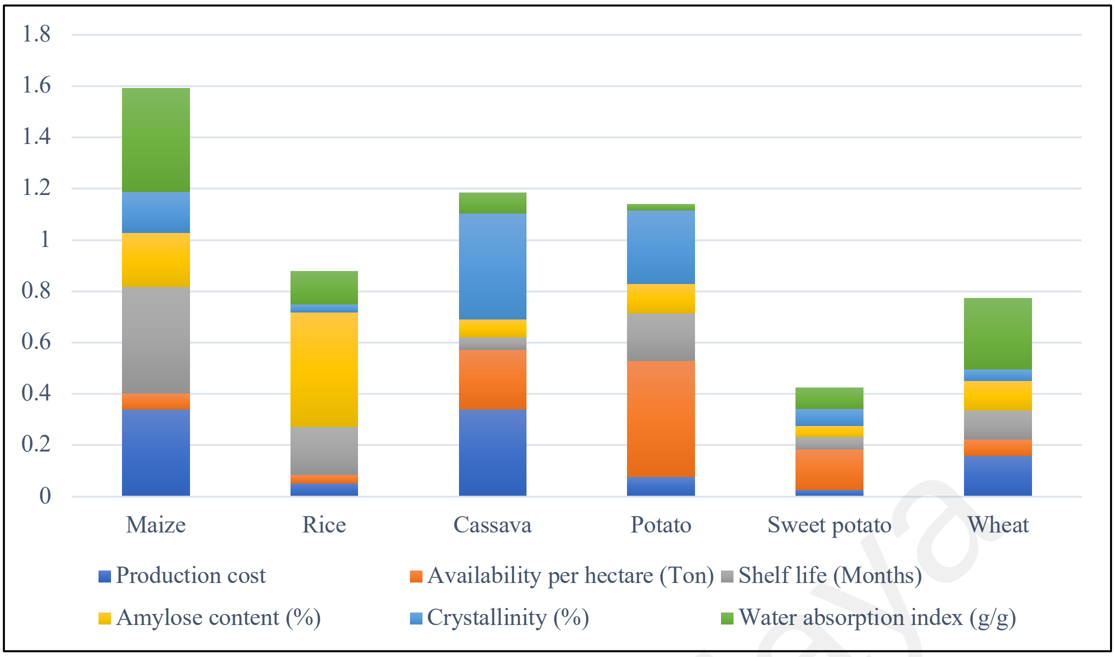

Figure 4.3: Alternatives choice values of natural biomass for biodegradable plastic production

Based on the overall priority vector (OPV) obtained in Table 4.10 it can be concluded that maize is the best natural biomass feedstock for the production of BPs. The alternatives are ranked according to their overall weight size from highest to lowest: maize (0.2654), cassava (0.2393), potato (0.1872), rice (0.1197), wheat (0.1243) and sweet potato (0.064). Among them, the difference in weights between rice and wheat is not significant, so these two natural biomasses can be interchanged.

According to the AHP results, the most suitable nature edible biomass as feedstock for the production of BPs is maize among those selected alternatives. Maize is grown on a wide area, has a high yield, has a clear advantage in terms of production cost, shelf life and water absorption index, and has a medium weighting under other criteria. Amalia (2020) et al. made a BP sample with a tensile strength of 11.7164 MPa using maize starch as the base material and maize husk as the base filler. This sample degraded 70% to 100% in 21 days. Marichelvam (2019) et al. modified maize BPs by adding rice starch to maize starch to increase its biodegradability and mechanical properties. de Azevedo et al. (2020)

et al. made BPs from extracted potato starch and maize starch and compared their performance. The results showed that the BPs prepared from maize starch had better physical, mechanical and thermal properties. Currently, maize plays a very important role in the production of BPs.

CHAPTER 5: CONCLUsion and recommendation

5.1 Conclusion

AHP is a good method for determining weights. It can divide and organize the factors in a complex problem into an ordered hierarchy of correlations. This study used AHP to calculate the favored natural biomass resources. For the first objectives, maize, rice, cassava, potato, sweet potato and wheat were identified as alternatives for the production of BPs. For the second objectives, production cost, availability per hectare, shelf life, amylose content, crystallinity and water absorption index were determined as crucial criteria for selecting feedstocks for the production of BPs. And for the third objectives, according to the AHP calculations, maize was considered the favored natural biomass resource as a feedstock to produce BPs.

Since starch is currently the main raw material used for BPs production and the key factors in the production of BPs from different raw materials are difficult to be harmonized, this study was conducted to screen the alternatives from starch raw materials and the remaining sources such as cellulose, lignin, chitin and microalgae were not sufficiently studied. Suitable natural biomass for the production of BPs can be selected according to different sources in future studies and finally in integrated comparisons.

5.2 Suggestions for Future Studies

This study was unable to conduct FGD due to COVID-19 and lacked the process of experts debating and convincing each other about this study. Therefore, this study will increase the number of literature reviews, find studies from different countries on related aspects, and screen high-quality literature to make the data as objective, accurate, and comprehensive as possible.

This study is not comprehensive enough in the selection of natural biomass. Only natural biomass containing starch was selected in this study because the criteria could not be standardized. In future studies, the natural biomass can be classified first, and each category can be selected quantitatively by AHP method, and finally each category can be combined to select the optimal natural biomass again by using AHP again.

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