



TALLINN UNIVERSITY OF TECHNOLOGY
SCHOOL OF ENGINEERING
Water and Environmental Engineering Research Group

ASSESSMENT OF THE BIODEGRADATION OF (PBS)-BASED PLASTIC UNDER CONTROLLED COMPOSTING CONDITIONS

(PBS)-PÕHISE PLASTI BIOLAGUNEMISE HINDAMINE KONTROLLITUD KOMPOSTIMISTINGIMUSTES

MASTER THESIS

Student: Femi Tosin Lawal
Student code: 213890EABM
Supervisor: Pavlo Lyshtva (PhD student)

Tallinn 2023

AUTHOR'S DECLARATION

Hereby I declare, that I have written this thesis independently.

No academic degree has been applied for based on this material. All works, major viewpoints and data of the other authors used in this thesis have been referenced.

"....." 20.....

Author: Femi Tosin Lawal

/signature /

This is in accordance with terms and requirements

"....." 20....

Supervisor:

/signature/

Accepted for defence

"....."20... .

Chairman of theses defence commission:

/name and signature/

Non-exclusive Licence for Publication and Reproduction of Graduation Thesis¹

I, Femi Tosin Lawal (date of birth: 17/05/1989) hereby

1. grant Tallinn University of Technology (TalTech) a non-exclusive license for my thesis Determination of the ultimate aerobic biodegradability of plastic material – PBS-based compound (Polybutylene Succinate) under controlled composting conditions.

supervised by Pavlo Lyshtva,

1.1 reproduced for the purposes of preservation and electronic publication, incl. to be entered in the digital collection of TalTech library until expiry of the term of copyright;

1.2 published via the web of TalTech, incl. to be entered in the digital collection of TalTech library until expiry of the term of copyright.

1.3 I am aware that the author also retains the rights specified in clause 1 of this license.

2. I confirm that granting the non-exclusive license does not infringe third persons' intellectual property rights, the rights arising from the Personal Data Protection Act or rights arising from other legislation.

¹ *Non-exclusive Licence for Publication and Reproduction of Graduation Thesis is not valid during the validity period of restriction on access, except the university`s right to reproduce the thesis only for preservation purposes.*

_____ (signature)

_____ (date)

Water and Environmental Engineering Research Group

THESIS TASK

Student: Femi Tosin Lawal, 213890EABM

Study programme, EABM03/18 - Environmental Engineering and Management

main speciality:

Supervisor(s): PhD student, Taltech. Pavlo Lyshtva, +37258055196,

pavlo.lyshtva@taltech.ee

Thesis topic:

(in English) Assessment of the biodegradation of (PBS)-based plastic under controlled composting conditions.

(in Estonian) (PBS)-põhise plasti biolagunemise hindamine kontrollitud kompostimistingimustes.

Thesis main objectives:

1. Literature review
2. Setup and maintenance of an aerobic biodegradability system for plastic materials under controlled composting conditions.
3. Determination of total dry solid, total volatile solid and total organic carbon of test materials.
4. Calculation of amount of carbon dioxide, percentage of biodegradability and loss in mass of plastic.
5. Analysis of data to show graphical representation of average daily CO₂ produced, cumulative CO₂ generated and degree of biodegradability of each test material and how they are related to each other.

Thesis tasks and time schedule:

No	Task description	Deadline
1.	Introduction, aim and literature review for thesis	12/07/2022
2.	Preparation of plastic materials for test	15/07/2022
3.	Apparatus installation and test commencement	19/07/2022
4.	Methodology, collection of data and results analysis	15/01/2023
5.	Discussion of results and general overview	12/05/2023

Language: English **Deadline for submission of thesis:** ".....".....2023

Student: Femi Tosin Lawal May 2023

/signature/

Supervisor: ".....".....20....a

/signature/

Consultant: ".....".....20....a

/signature/

Head of study programme: ".....".....20....a

/signature/

Terms of thesis closed defence and/or restricted access conditions to be formulated on the reverse side

Table of Contents

PREFACE.....	8
List of abbreviations and symbols	9
1. INTRODUCTION	10
2. LITERATURE REVIEW	12
2.1 Plastic material	12
2.1.1 Advantages and disadvantages of Plastic	12
2.2 Bioplastic overview	13
2.2.1 Polylactic Acid	15
2.2.2 Polyethylene Terephthalate	16
2.2.3 Hemp Plastic.....	16
2.2.4 PHA (Polyhydroxyalkanoates)	17
2.3 Biodegradation	17
2.3.1 Aerobic biodegradation	17
2.3.2 Anaerobic biodegradation	18
2.4 Biodegradable polyesters.....	19
2.4.1 Polybutylene Succinate.....	21
2.4.2 Biodegradable and biobased polymers.....	22
2.4.3 Compostable polymer properties and packaging applications	22
2.4.4 Production of PBS-based compound	25
2.4.5 Application of PBS-based compound.....	26
2.4.6 End-product of PBS-based compound.....	27
2.5 The comparison between compostable and biodegradable plastics.....	28
2.5.1 Recycling of biodegradable plastics	29
3. MATERIALS AND METHODS.....	30
3.1 Materials and their Preparation	30
3.2 Vermiculite Use.....	31
3.3 Instruments	33
3.4 Incubation Period	34
3.5 Final installation	35
3.6 Preparation of Mixtures	36
3.6.1 Indicator preparation	36
3.6.2 NaOH solution preparation	37
3.6.3 Determination of total dry solids and total volatile solids.....	37
3.7 Carbon dioxide determination	38
3.8 Computation of theoretical quantity of CO ₂ (Carbon dioxide)	40
3.9 Percentage biodegradation computation	40
4. RESULTS ANALYSIS.....	42

4.1 Average daily CO ₂ production.....	42
4.2 Cumulative CO ₂ generated	43
4.3 Calculation of Theoretical amount of CO ₂	44
4.4 Degree of biodegradability based on Maximum Reference Cellulose Material	46
4.5 Observation of PBS – based compound (granule and film) after 180 days.....	46
4.6 Comparison with other Studies.	48
5. CONCLUSIONS	51
SUMMARY	52
LIST OF REFERENCES.....	54

PREFACE

This MSc thesis is based on the analysis of plastic biodegradability was brought about by "Bio-plastic Europe" initiative. Its objective is to assess the biodegradation process of plastic derived from polybutylene succinate (PBS-based compound) in a controlled composting environment. The plastic material selected for this case study is PBS-based compound. The experimental testing was conducted at TalTech, the Estonian University of Technology.

Water(H₂O) and carbon dioxide (CO₂) are the decomposition products of PBS-based compound generally known as Polybutylene succinate when microorganisms in the soil act on it. PBS-based compound exhibits exceptional heat resistance compared to other biodegradable resins, and it also demonstrates a high level of compatibility with fibers. For this research, analysis will be done on the biodegradability properties of PBS-based compound the in the presence of Oxygen and also under controlled composting conditions.

This research work was supervised entirely by PhD student Pavlo Lyshtva.

Keywords: BIOPLASTIC, POLYBUTYLENE SUCCINATE (PBS), AEROBIC BIODEGRADABILITY, COMPOSTING, MASTER THESIS.

List of abbreviations and symbols

PBS	Polybutylene Succinate
AB	aerobic biodegradability
NaOH	Sodium hydroxide
HCl	Hydrochloric Acid
CO ₂	carbon dioxide
O ₂	Oxygen
N	Nitrogen
PA	Polyamide
PBAT	Polybutylene adipate terephthalate
PCL	Polycaprolactone
PE	Polyethylene
PET	Polvethylene terephthalate
PHA	Polyhydroxyalkanoate
PHVB	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PLA	Polylactic acid
PP	Polypropylene
PHB	Polyhydroxybutyrate
PEF	Polyethylene Furanoate
PTT	Polytrimethylene terephthalate
PES	Polyethylene Succinate
CF	Cotton Fiber
PGA	polyglycolic acid
LDPE	Low Density Polyethylene
HDPE	High Density Polyethylene

1. INTRODUCTION

Plastic has caused extensive damage to the environment, with vast quantities of it polluting the oceans, rivers, coastlines, and landfills. It is so prevalent that it can be found even in the air we breathe and spreading out the 9 billion metric tonnes of plastic produced to date would cover an area equivalent to that of Argentina or six times the size of California. Despite its widespread use, plastic has a lifespan that exceeds that of human generations, taking hundreds of years to decompose and breaking down only into micro particles. This ubiquitous material has infiltrated the food chain and can be found in animals worldwide [1].

Plastics are predominantly manufactured from fossil fuels and their mismanagement at the end of their useful life can lead to environmental accumulation, contributing to increased pollution and greenhouse gas emissions. However, more sustainable alternatives to traditional, non-biodegradable, fossil-based plastics have emerged, including biobased, biodegradable, and compostable plastics, which are considered promising solutions to plastic waste management problems. Nonetheless, these alternatives come with their own challenges, which need to be addressed [2].

The terminology surrounding bioplastics can lead to confusion among individuals. "Bioplastics" encompasses a diverse array of materials, while "bio-based" and "biodegradable" can be particularly confusing. Bio-based plastics, in contrast to the majority of conventional plastics derived from fossil fuels, are manufactured either wholly or partially using biological sources such as plant oils. However, it is important to note that the bio-based nature of these plastics does not inherently imply their ability to undergo decomposition or be composted [3].

The term "biodegradation" explains the way by which a substrate is enzymatically decomposed by bacteria and other organisms. These other organisms are widely distributed throughout the ecosystem and facilitate the breakdown of organic matter, resulting in the formation of water, carbon dioxide, and compost. However, it is crucial to acknowledge that the process of biodegradation is profoundly influenced by the specific environmental conditions in which it occurs [3].

Poly(1,4-butylene succinate) (PBS) is a type of thermoplastic polyester that is semi-crystalline in nature and has the ability to biodegrade. The synthesis of PBS involves polycondensation of two building blocks, namely succinic acid and 1-4-butanediol.

These components can be obtained from either fermentable sugars such as glucose and sucrose, or from feedstock derived from petroleum sources. [4].

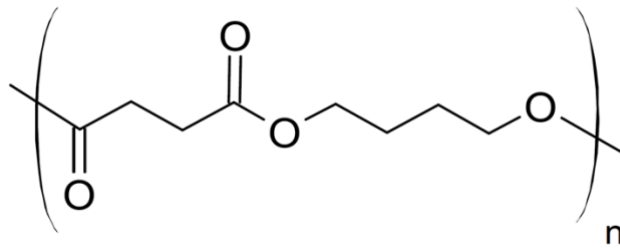


Figure 1.1 Polybutylene Succinate Structure (PBS-based compound) [4].

Polybutylene succinate (PBS) is a biopolymer that has demonstrated significant potential, primarily attributed to its remarkable mechanical properties that are comparable to high-density polyethylene (HDPE) and isotactic polypropylene (IPP). Unlike polylactic acid (PLA), PBS undergoes complete degradation into biomass, carbon dioxide (CO₂), and water (H₂O), enabling its disposal alongside other organic waste. Despite PBS having a lower melting point (115°C compared to 160°C for PLA), it remains preferable over PLA due to its absence of plasticizers.

When considering biopolymer options, Polybutylene succinate (PBS) emerges as the most economically advantageous selection in comparison to alternative choices such as Polybutylene adipate terephthalate (PBAT), Polylactic acid (PLA), and Polyhydroxybutyrate (PHB). PBS finds applications across a wide spectrum, including hygiene products, fishing lines and nets, food packaging, plant pots, and mulch films. It can be utilized as a matrix polymer on its own or in conjunction with other biopolymers like PLA [4].

The aim of this Master Thesis is to:

- Analyze the results of the ultimate aerobic biodegradation of PBS-based plastic under controlled composting conditions as reported in the literature.
- Observe and analyze the aerobic biodegradation of PBS-based plastic material in two different shapes, namely granules and film, under controlled composting conditions for a period of 184 days.
- Measure the amount of CO₂ production and biodegradability of these biopolymers and the results obtained from the test will be compared to those found in the literature.

2. LITERATURE REVIEW

2.1 Plastic material

Plastic is a type of polymer consisting of long, interconnected chains of molecules, which give it its unique properties of resilience, malleability, and durability. The production of synthetic plastics involves the use of refined natural gas or fossil fuels, and often involves the addition of chemical additives to enhance its properties. Natural plant matter can also be used as a raw material for plastic production. However, the major concern associated with fossil fuel-derived plastics is their persistence in the environment, as almost every mass-produced plastic item still exists today [5].

2.1.1 Advantages and disadvantages of Plastic

The profitability of the plastics industry stems from its utilization of byproducts derived from the refining of fossil fuels and natural gas. As long as these resources persist, the production of plastic will continue. In 2020, the global plastics market was valued at \$579 billion, with projections indicating its value will surpass \$750 billion by 2028. The worldwide production of plastic witnessed a significant increase, with 270 million metric tonnes produced in 2010 and a subsequent rise to 370 million metric tonnes in 2019 [5].

Despite the significant negative impacts of plastic pollution, the plastics industry continues to play a crucial role in the economy and supports a substantial workforce. Plastic products are everywhere due to their affordability and convenience. They find application in various everyday items, including toothbrushes, grocery bags, water and soda bottles, toys, takeaway containers, straws, and electronic device cases. However, the drawbacks associated with plastic, such as its persistent presence in the environment and the utilization of harmful chemicals in its production, outweigh its advantages by a considerable margin [5].

The presence of plastic waste poses a significant environmental challenge. Currently, approximately 150 million metric tonnes of plastic waste have accumulated in the world's oceans, with an additional 8 million tonnes being introduced each year. The ingestion of plastic fragments by marine organisms, including fish, poses a substantial risk to human health. The degradation process of plastic can take anywhere from

hundreds to millions of years, and the continued disposal of plastic exacerbates the issue rather than resolving it [5].

While bioplastics are touted as a solution to the plastic problem, their environmental impact is not yet fully understood. This review will examine whether bioplastics are a viable alternative to traditional plastics [5].

2.2 Bioplastic overview

Many people mistakenly use the words "Bio-based" and "biodegradable" interchangeably. However, it is important to note that these terms fall into distinct categories, which are outlined below and as clearly shown in Figure 2.1 below:

1. **Bio-based and biodegradable plastics:** Bioplastics encompass a range of materials derived from natural sources that possess the ability to undergo degradation over time, such as polyhydroxyalkanoates (PHA). Examples of bioplastics include TPS generally known as thermoplastic starch, cellulose acetate, and PHA known as Polyhydroxyalkanoate. Bioplastics can also be synthesised using techniques including microbial fermentation and also chemical polymerization, and examples include polylactic acid (PLA) and polybutylene succinate (PBS). [3].
2. **Biodegradable and Fossil-based plastics:** PCL commonly known as polycaprolactone and polybutylene adipate terephthalate (PBAT) are examples of biodegradable plastics; nevertheless, their production involves the use of resources derived from fossil fuels formed from decomposition organisms [3].
3. **Bio-based no biodegradable plastics:** PPC also known as Polypropylene carbonate, PEP also known as polyethylene plastics, Bio-PE known as bio polyethylene, Bio-PET known as bio polyethylene terephthalate, and Bio-PP known as bio polypropylene are recognized as significant bioplastics in the field. Among these, bio polyethylene terephthalate (Bio-PET) stands out as the most prevalent variant [3].

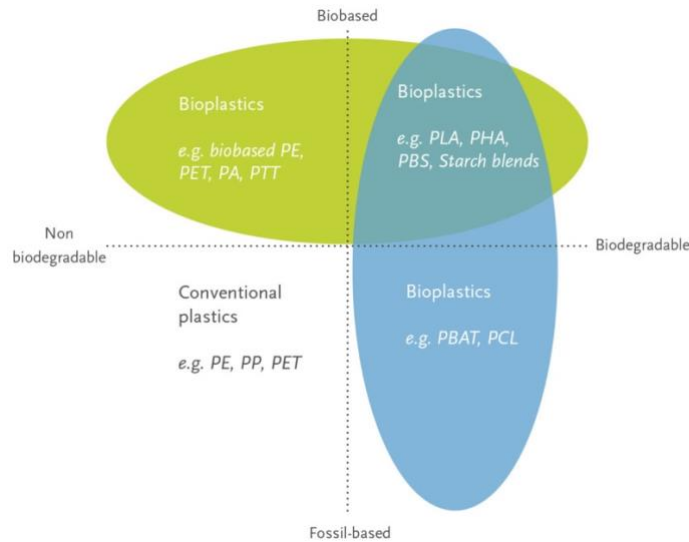


Figure 2.1 Classification of bioplastics [3].

Bioplastics exhibit a range of characteristics and not all possess complete or partial resistance to biodegradation. The term "bioplastics" encompasses both "biodegradable plastics" and other types of plastics. Biodegradable plastics undergo testing to ensure their natural breakdown, typically occurring within a period of 90 to 180 days. Bioplastics are derived from either synthetic or natural materials, and their production does not necessarily require organic sources. However, biodegradation is not universally guaranteed for all bioplastics, and certain biodegradable plastics may not degrade entirely or only under specific conditions. There is a risk of greenwashing, where some biodegradable plastic products may be misleadingly marketed. It is crucial to recognize that while bioplastics differ from conventional plastics, they cannot be considered a comprehensive solution to plastic pollution until they are both environmentally safe and biodegradable [3].

The provided Figure 2.2 depicts the global production capacity of bioplastics in 2018. The data reveals that a total of 2.11 million tonnes of bioplastics were manufactured during that year. Among these, 56.8% constituted bio-based/non-biodegradable plastics, while 43.2% comprised biodegradable plastics.

It is evident from these statistics that the annual global production capacity of bio-based/non-biodegradable plastics surpassed that of biodegradable plastics. However, there is optimism that these numbers will shift in favor of biodegradable plastics as ongoing research aims to enhance their quality and align them with the demands of everyday plastic use.

A closer examination reveals that out of the 56.8% annual production of bio-based/non-biodegradable plastics, the highest proportion (25.6%) is attributed to polyethylene terephthalate (PET). This is followed by polyamide or Nylon plastic (PA), polyethylene (PE), and polytrimethylene terephthalate (PTT) at 11.6%, 9.5%, and 9.2% respectively. Notably, polypropylene (PP) and polyethylene furanoate (PEF) are currently undergoing development and are expected to be commercially available in the near future [16].

Conversely, 43.2% of the global production in 2018 consisted of biodegradable plastics. This sector remains an area of extensive research and improvement and can be considered as the future of plastics. Among the biodegradable plastics produced, starch blends accounted for the highest proportion, approximately 18.2%. Other biodegradable plastics such as polylactic acid (PLA), polybutylene adipate terephthalate (PBAT), polybutylene succinate (PBS), and polyhydroxyalkanoate (PHA) collectively contributed around 25% to global production.

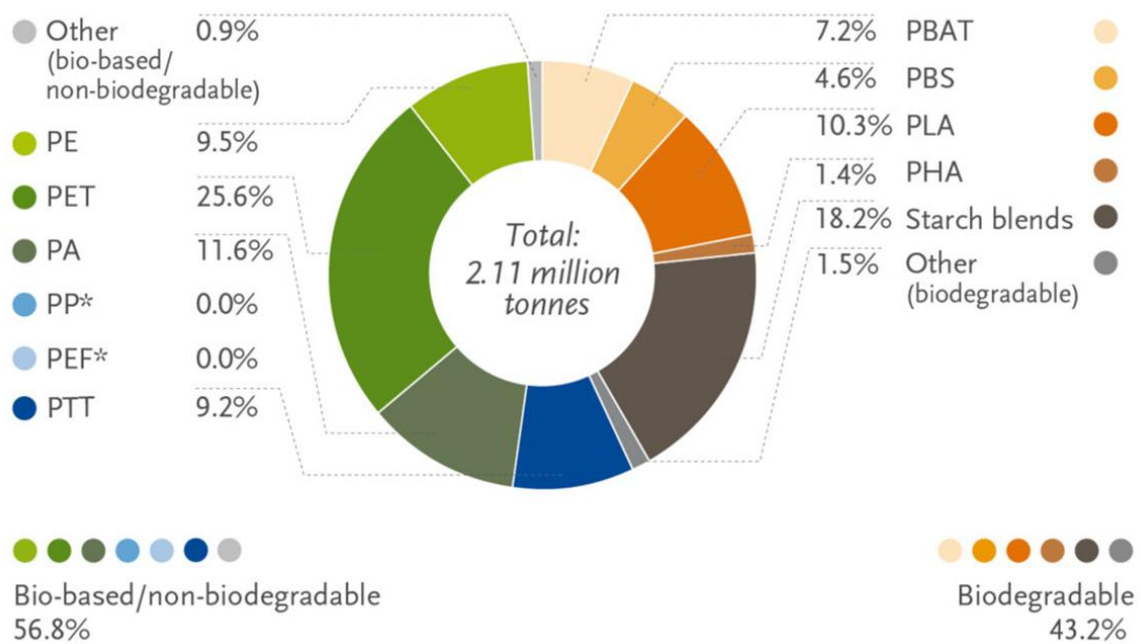


Figure 2.2 Global production capacities of bioplastics 2018 [16].

2.2.1 Polylactic Acid

Poly(lactic acid) (PLA) is a bioplastic that exhibits recyclability and natural degradation properties. It is derived from cellulose obtained from sources such as sugarcane, potatoes, or corn sugar. PLA finds application in the production of various items, including cups, 3D printing supplies, and reusable shopping bags, making it a preferable

alternative to traditional plastic. However, it is important to note that PLA biodegradation is contingent upon specific conditions. Specifically, it can only biodegrade in industrial composting facilities with temperatures exceeding 136 degrees Fahrenheit. Additionally, proper recycling facilities designed for PLA are required to ensure effective degradation. Under these controlled conditions, PLA can disintegrate within a few weeks. In contrast, PLA may take an extended period to break down in a landfill environment. Notably, PLA does not undergo degradation in saltwater, which diminishes its potential advantages over conventional plastic in marine environments [5].

2.2.2 Polyethylene Terephthalate

Polyethylene terephthalate (PET) is a type of plastic that is produced from a combination of natural and synthetic materials. The natural components may include substances derived from sugarcane and fossil fuel-based sources. PET is widely utilized in the manufacturing of water bottles. While it is derived from natural resources, this does not imply automatic degradation in the environment. The molecular polymer chain of PET remains consistent with that of synthetic plastics, thereby retaining similar characteristics. The duration required for PET to degrade in the environment remains uncertain. Despite the inclusion of plant-based constituents, PET may exhibit resistance to environmental factors over an extended period [5].

2.2.3 Hemp Plastic

Bioplastics, such as hemp plastic, offer complete biodegradability and represent a superior alternative to conventional plastics. The production of hemp plastic begins with the conversion of hemp leaves and fibers into pulp, enabling the extraction of cellulose plant matter present within the material. Subsequently, the hemp cellulose undergoes acid treatment, followed by water rinsing and drying, thereby removing the residual acid. It is then combined with other plant-based components and resin, and subjected to heat and pressure.

The resultant bioplastic exhibits remarkable attributes, including lighter weight compared to steel, enhanced durability, and increased strength. Hemp plastic possesses unparalleled biodegradability among plastic materials, typically breaking down within a timeframe of 90 to 180 days when exposed to the environment. Additionally, hemp plastic is capable of being recycled multiple times, further contributing to its

sustainability. Thus, hemp plastic stands out as a bioplastic that potentially surpasses conventional plastic in terms of environmental performance [5].

2.2.4 PHA (Polyhydroxyalkanoates)

PHA is a form of polyester that is produced and stored by bacteria during the fermentation process of sugar or fats. When employed in bioplastics, PHA can withstand temperatures that are greater than those of PLA.

PHAs are used in today's medical equipment production, modern food packaging and agricultural products among other things [5].

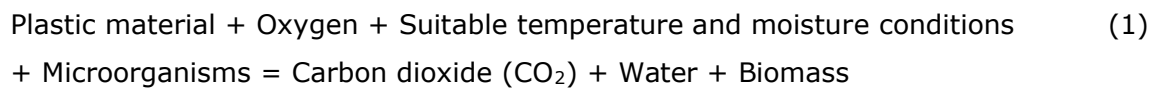
2.3 Biodegradation

Biodegradation refers to the enzymatic or metabolic processes employed by living microorganisms to convert complex organic molecules into simpler chemicals. This process can occur through various mechanisms, ultimately resulting in the production of carbon dioxide and methane. Biodegradation can take place in the presence or absence of oxygen, known as aerobic and anaerobic biodegradation, respectively. Organic materials subject to biodegradation can originate from natural sources such as plants and animals, or they can be artificially synthesized analogues that are metabolizable by bacteria. Hydrocarbons like cellulose, PCBs known as polychlorinated biphenyls, PAHs known as polyaromatic hydrocarbons, pharmaceuticals, radionuclides, and other compounds can all be broken down, transformed, or stored by specific microorganisms due to their unique metabolic pathways [6].

2.3.1 Aerobic biodegradation

This is said to occur when microorganisms degrade organic pollutants in the presence of oxygen. The presence of oxygen is vital for sustaining life, thereby establishing oxidative conditions within the system, environment, or organism. Aerobic bacteria, also referred to as "aerobes," exhibit the ability to efficiently degrade diverse organic pollutants within oxygen-rich environments. Aerobes represent the most prevalent type of bacteria, deriving their energy from cellular respiration, which involves the oxidative

breakdown of substrates like carbohydrates and lipids. Within the cytoplasm of aerobic organisms, glucose molecules undergo enzymatic breakdown in preparation for cellular respiration. The resulting fragmented molecules are subsequently transported into the mitochondria for further processing through aerobic metabolism. During this process, oxygen participates in chemical reactions, leading to the degradation of these smaller molecules into carbon dioxide and water, accompanied by the release of energy. Contrasting with anaerobic digestion, which can result in unpleasant odours, aerobic digestion facilitates more thorough waste breakdown, reducing its volume by over 50%. The aerobic process plays a crucial role in maintaining the health of humans and other organisms by reducing the population of harmful microorganisms in the surrounding environment [6, 24, 25]. The aerobic biodegradation process of plastic material can be further expressed by equation 1 below:



2.3.2 Anaerobic biodegradation

Anaerobic digestion is a microbial process wherein oxygen-independent microorganisms outcompete their oxygen-requiring counterparts. In the absence of oxygen, biodegradable waste undergoes anaerobic decomposition, resulting in slower degradation rates for substances like paper. Anaerobic digestion generates methane, a potent greenhouse gas with 21 times greater warming potential than carbon dioxide. However, adopting a "cradle to grave" perspective has prompted the capture of methane-rich biogas, which can be utilized for sustainable electricity generation. Throughout each stage of the process, bacteria facilitate the decomposition of organic matter without the presence of oxygen.

Anaerobic digestion is commonly employed for the treatment of biodegradable waste and wastewater sludge, leading to a significant reduction in both the quantity and size of input materials. The process mitigates the release of waste gases into the atmosphere, while the nutrient-rich residues remaining after digestion can be utilized for plant cultivation. Overall, anaerobic digestion holds promise as a renewable energy source that can complement or even replace fossil fuels [6, 26, 27]. The anaerobic biodegradation process of plastic material can be further expressed by equation 2 below:

Plastic material + Microorganisms + Suitable temperature and moisture conditions = Biogas (mainly methane and carbon dioxide) + Residual organic matter + Partial decomposition products (2)

2.4 Biodegradable polyesters

In recent years, there has been considerable research and development investment in biodegradable polymers. These can be broadly categorized into two types based on their manufacturing process:

- (a) biodegradable polymers obtained from renewable sources.
- (b) synthetic polymers derived primarily from mineral oil [7].

Among the biodegradable polymers, aliphatic polyesters made from renewable resources are particularly useful due to their high biodegradability and biocompatibility. Although several aliphatic biodegradable polyesters are available, only a few are currently on the market, including PLA known as polylactic acid, PGA known as polyglycolic acid, PCL known as poly-ε-caprolactone, PHB generally known as polyhydroxybutyrate, and poly(-caprolactone) [7].

Among all the listed biodegradable polyesters mentioned above, most researches are carried out on PHB and PLA. This is due to their potential as biodegradable thermoplastic polyesters. However, their fragility and short processing window have limited their commercial use, leading to the exploration of polymer blends.

Eliminating water from the reaction of a diacid or acid anhydride with a diol is a typical method for producing synthetic biodegradable polyesters such as PPS known as poly(propylene succinate), PBS-based compound known as poly(butylene succinate) and PES known as poly(ethylene succinate). These polyesters are all biodegradable and synthetic. These polyesters are categorized as both biodegradable and synthetic materials. At present, fossil fuels are predominantly used to manufacture aliphatic polyesters, although some can be made from sustainably sourced materials. Poly(butylene adipate-co-terephthalate) (PBAT) is an example of a copolymerized aromatic polyester produced commercially using adipic acid, terephthalic acid, and 1,4-butane diol [7].

Due to their hydrophilicity, aliphatic polyesters absorb moderate to large amounts of water when exposed to humidity, leading to reduced mechanical characteristics and stability. Therefore, some biodegradable polymers are combined with other polymers or copolymerized with aromatic building blocks to enhance their stability. The degradation time of biodegradable polyesters is a significant quality, as a quick degradation time can result in decreased corrosion resistance. The degradation time can range from a few months to a few years, depending on the polymer and environmental conditions [7].

Figure 2.3 below represents the properties and structures of various biodegradable polyesters available at the moment.

Polyester	Structure of Repeat Unit	Properties
Poly(lactic acid) or Polylactide (PLA)	$\text{H} \left[\text{O} - \underset{\text{O}}{\underset{\parallel}{\text{C}}} - \overset{\text{CH}_3}{\text{CH}} - \text{C} \right]_n \text{OH}$	$T_g \approx 45 - 60 \text{ }^\circ\text{C}$ $T_m \approx 150 - 160 \text{ }^\circ\text{C}$
Poly(L-lactic acid) or Poly(L-lactide) (L-PLA)	$\text{H} \left[\text{O} - \underset{\text{O}}{\underset{\parallel}{\text{C}}} - \overset{\text{CH}_3}{\text{CH}} - \text{C} \right]_n \text{OH}$	$T_g \approx 55 - 65 \text{ }^\circ\text{C}$ $T_m \approx 170 - 200 \text{ }^\circ\text{C}$
Poly(DL-lactic acid) or Poly(DL-lactide) (DL-PLA)	$\text{H} \left[\text{O} - \underset{\text{O}}{\underset{\parallel}{\text{C}}} - \overset{\text{CH}_3}{\text{CH}} - \text{C} \right]_n \text{OH}$	$T_g \approx 50 - 60 \text{ }^\circ\text{C}$ $T_m \approx \text{N/A (am.)}$
Poly(glycolic acid) or Polyglycolide (PGA)	$\text{H} \left[\text{O} - \text{CH}_2 - \underset{\text{O}}{\underset{\parallel}{\text{C}}} - \text{O} - \text{CH}_2 - \underset{\text{O}}{\underset{\parallel}{\text{C}}} \right]_n \text{OH}$	$T_g \approx 35 - 45 \text{ }^\circ\text{C}$ $T_m \approx 220 - 235 \text{ }^\circ\text{C}$
Poly(ϵ -caprolactone) (PCL)	$\text{H} \left[\text{O} - \left(\text{CH}_2 \right)_5 - \underset{\text{O}}{\underset{\parallel}{\text{C}}} \right]_n \text{OH}$	$T_g \approx -60 - (-65) \text{ }^\circ\text{C}$ $T_m \approx 58 - 65 \text{ }^\circ\text{C}$
Poly(hydroxybutyrate) (PHB)	$\text{H} \left[\text{O} - \underset{\text{O}}{\underset{\parallel}{\text{C}}} - \overset{\text{CH}_3}{\text{CH}} - \text{CH}_2 - \underset{\text{O}}{\underset{\parallel}{\text{C}}} \right]_n \text{OH}$	$T_g \approx 5 - 15 \text{ }^\circ\text{C}$ $T_m \approx 165 - 180 \text{ }^\circ\text{C}$

Figure 2.3 Properties of biodegradable polyesters [7].

2.4.1 Polybutylene Succinate

Polybutylene succinate (PBS) is a biodegradable material that undergoes microbial degradation in soil, resulting in the formation of water and carbon dioxide. Notably, PBS-based plastics exhibit a remarkable compatibility with fibers and demonstrate superior thermal stability compared to other recyclable resins. This advantageous thermal behavior, coupled with its favorable fiber interaction, enables PBS-based compounds to be effectively blended with various resins and materials, leading to the formation of high-performance composite materials. Such blends have shown enhanced properties and performance characteristics, surpassing those achieved by pure PBS-based compounds in isolation [8, 28].

In order to promote environmental sustainability in packaging, the manufacturing process and materials employed in packaging must both be considered. The use of bio-based polymers, such as PBS-based plastic, can offer a more eco-friendly packaging solution that also delivers superior performance. PBS-based compound, in particular, can offer a cleaner end-of-life solution and reduce costs associated with disposal when reuse or recycling is not possible [9].

Table 2.1. Properties of BIO PBS-based compound [8]

Properties	Test Method	Unit	BIO PBS-based compound		
			FZ71 [PM/PM]	FZ91[PM/PM]	FZ92[PM/PM]
Density	ISO1183	g/cm ³	1.26	1.26	1.24
MFR[190°C, 2.16kg]	ISO1133	g/10min	22	5	4
Melting Point	ISO3136	°C	115	115	84
Yield Stress	ISO527-2	MPa	40	40	17
Stress at Break	ISO527-2	MPa	30	36	24
Strain at Break	ISO527-2	%	170	210	380
Flexural Modulus	ISO178	MPa	630	650	250
Flexural Strenght	ISO178	MPa	40	40	18
Izod Impact Strenght [23°C]	ISO180	KJ/M ²	7	7	47
Heat Deflection Temperature[0.45MPa]	ISO75-2	°C	95	95	63
Rockwell hardness	ISO2039-2	R Scale	107	107	56

2.4.2 Biodegradable and biobased polymers

Polybutylene succinate (PBS), also known as a "PBS-based compound," is a prevalent biodegradable polyester synthesized through laboratory polycondensation of butanediol and succinic acid. The process involves two distinct steps. Initially, the diol and diacid undergo a reaction to form an ester, resulting in the formation of a PBS-based compound characterized by a high molecular weight. Subsequently, the polycondensation process takes place at elevated temperatures. Figure 2.4 illustrates the synthesis of a PBS-based molecule and its assembly.

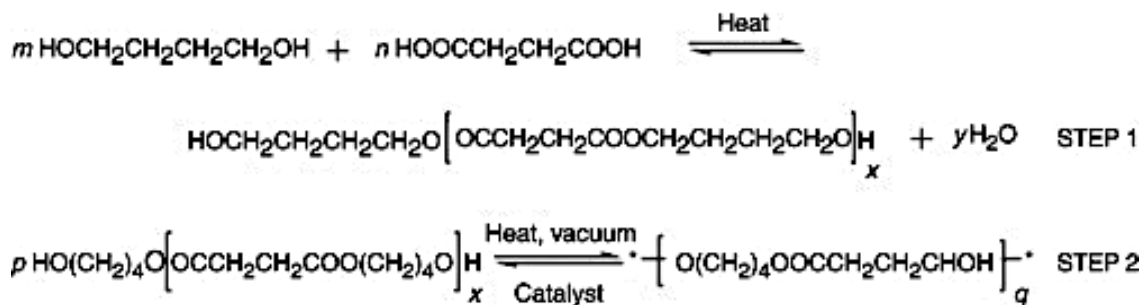


Figure 2.4 Synthesis and structure of PBS-based compound [10].

Showa Highpolymer, a Japanese company, has successfully developed aliphatic copolyesters utilizing succinate, adipate, ethylene glycol, and 1,4-butanediol. These copolyesters are commercialized under the brand name "vvBionolle." Similar to LDPE, HDPE, and PP, they exhibit comparable properties. With a melting temperature exceeding 100 degrees Celsius and a thermal degradation temperature surpassing 300 degrees Celsius, these copolyesters possess robust thermal characteristics. Furthermore, they can be processed into various forms using conventional techniques such as injection molding, extrusion, and film blowing. This versatility has sparked growing interest in utilizing these materials as potential alternatives to petrochemical-derived polyolefins, promoting sustainable solutions [10].

2.4.3 Compostable polymer properties and packaging applications

Poly(butylene succinate) (PBS), also known as PBS-based substance, is an aliphatic polyester readily available in the market. It possesses desirable properties including biodegradability, heat resistance, chemical resistance, and high melting point. The versatility of PBS-based compounds allows for their transformation into various fabric types such as melt-blown, multifilament, monofilament, flat, and split yarn.

Additionally, PBS-based compounds show promise for applications in the plastics industry, particularly in the production of injection-molded products. Showa Highpolymer, a Japanese company, has successfully developed aliphatic polyesters and copolyesters marketed under the brand name Bionolle. These thermoplastics are characterized by their white appearance, solid structure, and a density of approximately 1.25 g/cm³. Table 2.2 provides an overview of the key physical, chemical, and mechanical properties of different Bionolle series, including the 1000 series (PBS-based compounds), the 2000 and 3000 series (poly(butylene succinate adipate) or PBSA), and the 6000 series (poly(ethylene succinate)).

Table 2.2 Properties comparison for Bionolle, low density polyethylene, high density polyethylene and Polypropylene [10]

Properties	Bionolle(#1001)	Bionolle(#3001)	LDPE	HDPE	PP
Glass Transition (°C)	-32	-45	-120	-120	5
Melting point (°C)	114	94	110	129	163
HDT (°C)	97	69	88	110	145
Tensile strength (MPa)	57	47	35	39	44
Yield strength (MPa)	32	19	12	27	31
Flexural modulus (MPa)	656	323	176	1070	1370
Strain at break (%)	700	900	400	650	800
MFR at 190°C (g/10 min)	1.5	1.4	2	2	4

Bionolle, a commercial product developed by Showa Highpolymer in Japan, encompasses a range of aliphatic polyesters and co-polyesters distinguished by their white crystalline thermoplastic properties. These materials possess a density of approximately 1.25 g/cm³ and exhibit melting points that fall within the range of 90 to 120 degrees Celsius. The glass transition temperatures of these compounds exhibit

significant variation, ranging approximately from -45 degrees Celsius to -10 degrees Celsius. Table 2.3 provided below shows important properties of Bionolle grades 2000 and 3000 (PBS-based compounds) and likewise grade 6000 (PES-based compounds) can be found. [10].

Table: 2.3 Bionolle properties and different grades available in market [10].

Property	PBSU #1000	PBSU #2000	PBSU #3000	PESU #6000	LDPE F082	HDPE 5110	PP210
MFR 90°C(g/10min)	1.5	4.0	28	3.5	0.8	11	3.0 ^a
Density (g/cm ³)	1.26	1.25	1.23	1.32	0.92	0.95	0.90
Melting point (°C)	114	104	96	104	110	129	163
Glass transition temperature (°C)	-32	-39	-45	-10	-120	-120	-5
Yield strenght (kg/cm ²)	336	270	192	209	100	285	330
Elongation (%)	560	710	807	200	700	300	415
Stiffness 10 ³ (kg/cm ³)	5.6	4.2	3.3	5.9	1.8	12.0	13.5
Izod impact strength ^b (Kg- cm/cm20°C)	30	36	>40	10	>40	4	2
Combustion heat (cal/g)	5550	5640	5720	4490	>11,000	>11,000	>11,000

a MFR (Melt mass-flow rate) measurement was taken at 230°C

b Measurement of Izod impact strength was made using notched samples.

This study focuses on examining the influence of ethyl and n-octyl side chains on the properties of polybutylene succinate (PBS-based substance) and poly(ethylene adipate) (PEAd). The impact of increasing the number of branches in the polymer chain is investigated, specifically concerning glass transition temperature, melting temperature, melt viscosity, crystallinity, and spherulite growth rate. It is observed that as the number of branches increases, these properties decline.

Furthermore, the addition of n-octyl branches to the PBS-based compound enhances tensile strength, tear strength, and elongation, while the tensile strength and modulus remain unchanged. Additionally, the study examines the impact of polyester constituents on the mechanical and thermal properties of variety of aliphatic homopolyesters and copolyesters obtained from 1,4-butanediol and dimethyl esters of succinic and adipic acids [10].

The homopolymer based on polybutylene succinate (PBS) exhibits heat of fusion (Hf) of 68.4 J/g and melting temperature (Tm) of 114.1 degree celcius. In contrast, the poly(ethylene adipate) (PEAd), which has lower crystallinity, displays a Tm of 60.5 °C and an Hf of 52.8 J/g. The properties of copolyesters, depending on their composition, lie within this range. As the proportion of adipate units increases, the glass transition temperature (Tg) decreases from -31.3 °C to -60.7 °C. The tensile strength initially decreases with an increasing amount of adipate units, reaching a minimum around equimolar composition, before rising back to the value observed for poly(ethylene adipate). Conversely, the elongation at break increases when the adipate content per unit is in the range of 20–40 mol%. By increasing the chain length, the molecular weight of the copolyesters increases, leading to improved mechanical strength. While crystallinity and melting temperature decrease with chain length, Tg increases [10].

Moreover, polyesters derived from succinic acid and related aliphatic diols with 2-4 methylene groups demonstrated varying crystallization rates and degrees of crystallinity. Poly(propylene succinate) possesses the slowest rate of crystallization and also has lowest degree possible of crystallinity, while the equilibrium melting points were 114 °C for poly(ethylene succinate), 133.5 °C for PBS-based compound, and 58 °C for poly(propylene succinate), with calculated enthalpies of fusion of 180, 210, and 140 J/g, respectively [10].

2.4.4 Production of PBS-based compound

This polyester, which belongs to the class of aliphatic polymers and incorporates succinic acid as one of its monomers, can be manufactured utilizing either renewable energy sources or fossil fuels. The polymerization of succinic acid (also known as dimethyl succinate) and 1,4-butanediol (BDO) through condensation, mainly sourced from maleic anhydride, is the primary method of producing PBS-based compound. In essence, PBS-based compound is synthesized from maleic anhydride [10]. Figure 2.5 below shows the molecular structure of PBS-based compound.

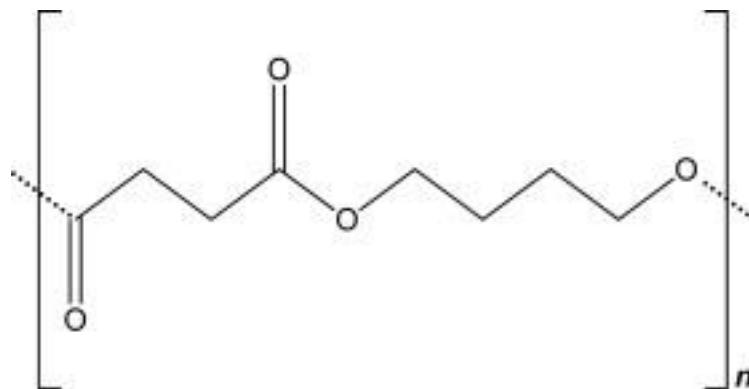


Figure 2.5 Molecular structure of PBS-based compound [10].

Polybutylene succinate (PBS-based compound) is an aliphatic polyester that can be commercially manufactured using succinic acid as one of its monomers. The production of succinic acid starts with the hydrogenation of maleic anhydride, which is then converted to succinic anhydride and hydrated to form succinic acid. This can be achieved through a variety of methods, including electrolysis or fermentation using microorganisms such as recombinant *E. coli*, *Mannheimia succiniciproducens*, and *Anaerobiospirillum succiniciproducens* [10].

The production of BDO, which is another monomer used in the synthesis of PBS-based compound, involves the use of petrochemical feedstocks and various industrial technologies. Large-scale industrial synthesis of both succinic acid and BDO is necessary to meet the requirements for the manufacturing of PBS-based compound [10].

The production of PBS-based compound involves two primary stages: esterification and poly-condensation. Stoichiometric amounts of BDO and succinic acid are first added to the reactor, followed by poly-condensation at high temperatures using catalysts such as tetraisopropyl titanate, lanthanide triflates, or SnCl_2 . The resulting PBS-based compound has a high molecular weight and can possess a range of physical and chemical properties that make it a promising material for various potential applications [10].

2.4.5 Application of PBS-based compound

The increasing concern about the negative impact of packaging on the environment has led to a growing interest in biodegradable packaging as an alternative to synthetic and non-biodegradable petroleum-based packaging. One promising material for this purpose is polybutylene succinate (PBS-based compound), which offers superior biodegradability and biocompatibility characteristics. Due to its transparent and rigid

characteristics, this material exhibits versatile applicability, encompassing a range of purposes including, but not limited to, mulching films, biodegradable bags, nonwoven sheets and textiles, food service utensils, and foams. Researchers have found that PBS-based compound is widely used in various industries, including agriculture, fishing, forestry, and civil engineering. In agriculture, PBS-based compound is also employed for the production of vegetation nets. Additionally, PBS-based compound has potential applications in the sectors of monofilament, injection molded items, tape, split yarn, and textiles [11, 29, 30].

2.4.6 End-product of PBS-based compound

The end-of-life scenario for PBS-based compound is dependent on its intended application. PBS-based compound can be utilized for various purposes, each of which may have a different end point. For example, in the medical industry, PBS-based compound is commonly used for implants and other applications that come in contact with the human body, and these are usually made of resorbable or absorbable polymer. Although the term "biodegradable" is used more commonly, PBS-based compound polymers utilized in biomedical applications are generally broken down in the body via non-enzymatic hydrolysis [12].

In contrast, PBS-based compound used for non-medical purposes is frequently discarded as waste in landfills, where it ultimately biodegrades into water and carbon dioxide, as illustrated in the figure below.

Figure 2.6 below shows an overview of the various degradation pathways involved in the breakdown of biodegradable PBS-based compounds. Polymer chain scission serves as the primary mechanism for the cleavage of chemical bonds within polymers. This process occurs through either the main chain or the side chains of the polymer molecules. The acceleration of this degradation process can be achieved through the application of heat, oxygen, light, or water.

During photo-degradation, polymers undergo fragmentation, resulting in the formation of molecules with reduced molecular weight. In contrast, physical and mechanical degradation occurs when external factors such as polymer processing, shear pressures, and other loads cause the breakdown of polymers into smaller fragments.

Oxidative degradation occurs due to the presence of atmospheric oxygen. This process involves the autocatalytic attack of hydrogen atoms, leading to the formation of hydroperoxides. This specific type of degradation is known as oxidative deterioration. Polymers with hetero backbone chains can undergo degradation through hydrolysis reactions, which may occur biotically (mediated by living cells or microbes) or abiotically (primarily influenced by pH), or through a combination of both. Such degradation is referred to as hydrolytic degradation[31, 32].

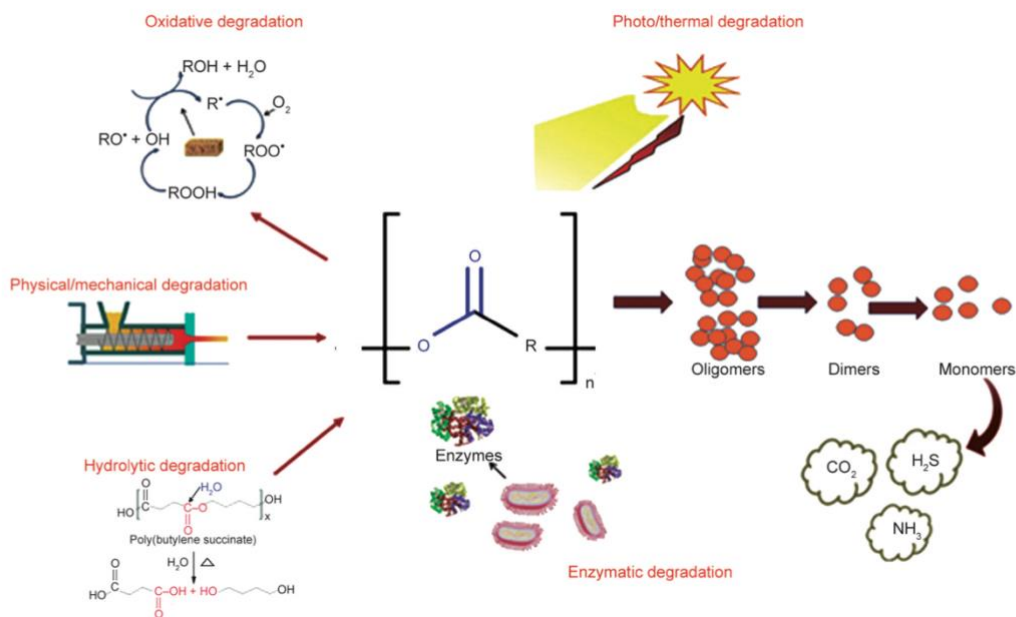


Figure 2.6 Modes of polymer degradation [12].

2.5 The comparison between compostable and biodegradable plastics

Biodegradability is the ability of microorganisms, such as bacteria, algae, or fungi, to break down a material. Considering compostable straws, they can be biodegraded by microorganisms. The time required for degradation relies on a couple of factors which include: temperature, humidity, and the composition of the material. Biodegradable bioplastics, which are derived from plants, animals, or minerals like hemp, do not always degrade completely into their initial constituents as residues might be left behind in some cases. If the proper conditions are met however, it may take only a few weeks for microorganisms to decompose biodegradable bioplastics into water and carbon

dioxide. Compostable materials, on the other hand, break down completely into natural substances such as water, carbon dioxide, and biomass in less than three months. They provide nutrients to plants as they decompose without leaving any undesirable substances behind [5, 23].

2.5.1 Recycling of biodegradable plastics

In many nations, bio-based plastics mostly pose great challenges in recycling due to complications in separating its various components. While bioplastics offer potential for recycling, recycling facilities do not currently accept them in large quantity, rendering them largely unrecyclable. The feasibility of bioplastics recycling rests on the availability of a significant volume of these materials to justify investments in recycling infrastructure [5, 18, 19].

Due to the low energy input required for the production of PBS and most bioplastics, it is essential to prioritize their reuse before considering recycling. However, complete reuse may not always be feasible in all situations. Therefore, it is advisable to mechanically recycle bioplastics multiple times until their quality degrades to a point where further recycling is no longer viable. In such cases, a chemical recycling method is employed to recover the monomers that have been lost due to degradation. [20, 21, 22]

3. MATERIALS AND METHODS

The assessment of the complete aerobic biodegradability of organic compound-based plastics entails conducting a controlled composting test under aerobic conditions. This test involves measuring the evolved carbon dioxide and the degree of plastic disintegration upon completion. The protocol for this evaluation is specified in EVS-EN ISO 14855-1:2012. The primary objective of this method is to simulate the aerobic composting process for the organic fraction of municipal solid waste (MSW). The test sample is inoculated with compost-derived microbial cultures to initiate the biodegradation process. Composting occurs in a well controlled atmosphere with regard to ventilation, temperature, and moisture content. It is anticipated that the test procedure will provide both the rate at which the carbon in the test material is converted to evolved carbon dioxide and the percentage at which this occurs.

As an alternative to employing fully developed compost, subclauses 8.6 and 8.7 of ISO 14855-1 detail the use of a mineral bed (vermiculite injected with thermophilic microorganisms obtained from compost with a particular activation phase). This variant is meant to provide information about how quickly the carbon in the test sample is being transformed to carbon dioxide.

3.1 Materials and their Preparation

The research (PBS)-based material is presented in two different shapes that has been tested in this study: PBS-based compound(soft peck)-granules, and PBS-based compound(soft peck)-Film. The aims to assess its biodegradability in a laboratory composting environment. PBS-based compound is a biodegradable polymer that soil microorganisms can break down into water and carbon dioxide. It exhibits superior heat resistance and compatibility with fibres compared to other biodegradable resins. These properties enable PBS-based compound to be blended with other resins and materials, resulting in high-performance composites [8].

To determine the biodegradability of the plastic, the total amount of carbon dioxide produced and the degree of material breakdown were evaluated under controlled aerobic conditions at the conclusion of the experiment. Daily degradation levels were measured as outlined in Chapter 4. Activated vermiculite was employed as a substitute

for traditional compost to simulate the conditions of composting the organic component of mixed municipal solid waste (MMSW). Optimal results were achieved by ensuring that the inoculum was applied in a composting environment that closely replicated temperature, ventilation, and moisture content conditions.

The rate and percentage of carbon dioxide production, along with the level of material degradation, were quantified using a two-step titration method applied to the sample mixture used in the test.

ISO 8245 was employed to measure the total organic carbon (TOC) content of both the test and control samples, and the data was expressed in milligrams of TOC per gram of dry material. The test materials were presented as granules and films, and the surface area of each sample was restricted to no more than 2X2 cm (Figure 3.1 a,b). To conform to the specified dimensions, the film material used in the experiment was cut into smaller pieces.

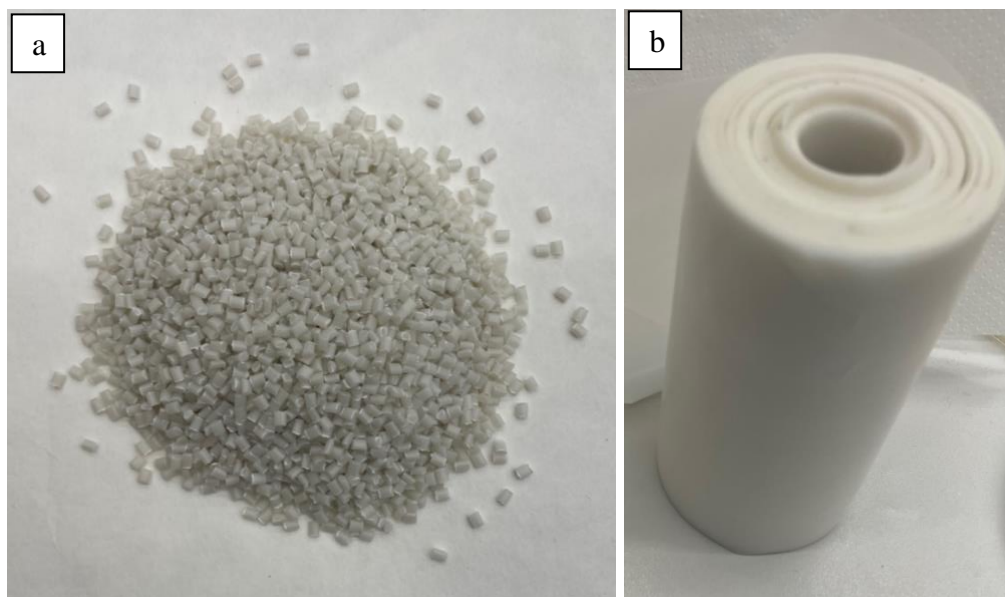


Figure 3.1 (a) PBS (Granule shape), (b) PBS (Film shape)

3.2 Vermiculite Use

The activated vermiculite was used as inoculum. The mineral part was inoculated with a blend of mature decomposition, organic and inorganic nutrients. This is achieved

by combining the vermiculite with an inoculum solution containing constituents listed in Tables 3.1, 3.2, and 3.3 in a mass-to-volume ratio of 1:3.

Table 3.1 1L of Composition of inoculum solution

Constituent	Mineral solution (table 3.3)	Suitable nutrient broth	Urea	Corn starch	Cellulose	Compost extract
Amount	500ml	13g	5.8g	20g	20g	500ml

Table 3.2 1L of Composition of mineral solution

Chemical	KH ₂ PO ₄	MgSO ₄	CaCl ₂	NaCl (10% solution)	Trace-element solution (table 3.4)
Amount	1g	0.5g	1ml	1ml	1ml

Table 3.3 1L of Composition of trace-element solution

Chemical	H ₂ BO ₃	KI	FeCl ₃	MnSO ₄	(NH ₄) ₆ MO ₇ O ₂₄	FeSO ₄
Amount	500mg	100mg	200mg	400mg	200mg	400mg

To prepare the inoculum compost extract, mature compost and deionized water were mixed at a mass-to-volume ratio of 20% and stirred for 30 minutes before being strained through a 1 mm hole size sieve.

The appropriate amount of vermiculite was combined with the inoculum solution and the resulting mixture was transferred into the bioreactors. The entire contents of each bioreactor were weighed and incubated at 50°C for three days.

The quantity of activated vermiculite and test material required is dependent on the volume of the composting containers which was 500ml, The test mixture occupied about half of the composting vessel, leaving enough headspace for manual shaking.

3.3 Instruments

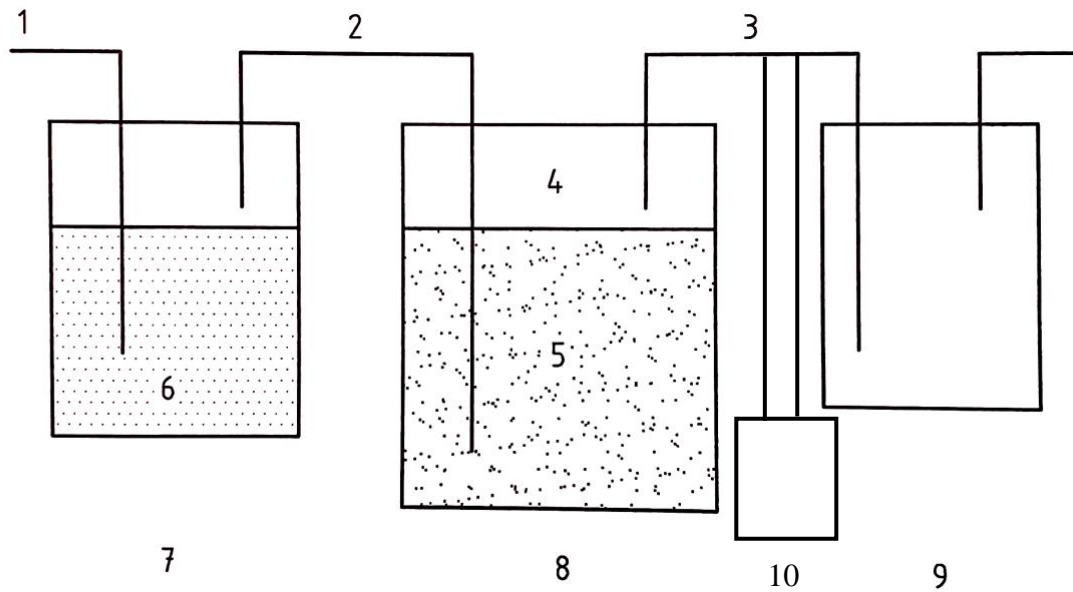
In this experiment, some of the instrument used include: glass wares, composting vessels, air-supply system, apparatus for the determination of carbon dioxide, gas-tight tubes, analytical equipment, balance, analytical equipment and heaters for activation of the vermiculite.

The composting vessel used consist of 500ml glass bottles that allow for uniform gas purge upward. Aquarium air pumps were used as air supply sustem to deliver carbon dioxide-free air (achieved by preliminary treatment with NaOH sollutions), at a constant flow rate sufficient to create true aerobic conditions for the duration of the test (as shown in the Figure 3.2 below).

In this experimental setup, an aquarium air pump supplies air through pipe 1 into vessel 6, which contains sodium hydroxide (NaOH). The purpose of this setup is twofold: first, to eliminate carbon dioxide (CO₂) from the incoming air, and second, to humidify the air due to the presence of water in NaOH. The result is the production of carbon dioxide-free air, which is then directed through pipe 2 to the bottom of vessel 5, where the sample of polybutylene succinate (PBS)-based compound is located. The compost in vessel 5 is aerated by this airflow.

The process within vessel 5 generates a mixture of CO₂, H₂O and biomass. This gas mixture is transferred through pipe 3 into vessel 10, which contains a solution of hydrochloric acid (HCl). The function of vessel 10 is to capture moisture from the gas. Finally, the gas is received in vessel 9, which contains a carbon dioxide trap solution consisting of NaOH.

During the experiment, the test sample in vessel 5, composed of the PBS-based compound, undergoes composting at an average temperature of 58°C ± 2°C.



Key:

- | | |
|------------------------------|--|
| 1. Air | 6. NaOH solution |
| 2. CO ₂ -free air | 7. CO ₂ -removal system |
| 3. Exhaust air | 8. Composting vessel |
| 4. Headspace | 9. CO ₂ -determination system |
| 5. Test mixture | 10. HCl Solution |

Figure 3.2 Layout of test system based on EVS-EN ISO 14855-1:2012

3.4 Incubation Period

During a biodegradability test, the ISO 8245 standard specifies a method for quantifying the cumulative quantity of carbon dioxide (CO₂) that develops as dissolved inorganic carbon (DIC). The method entails continuous measurements of the amount of CO₂ emitted from the exhaust air of each composting vessel. The measurement frequency is determined by the biodegradability test combination, the required accuracy of the biodegradation curve, and the measurement technique used. During the biodegradation phase, the CO₂ evolved was measured directly once every ten days. The composting receptacles were shaken weekly to ensure uniform attack of microorganisms on the test material and to prevent extensive channelling.

During the weekly agitation of the composting containers, observations of structure, moisture content, colour, fungal growth, exhaust air aroma, and test material decomposition are made and recorded at the conclusion of the test period.

3.5 Final installation

The Figure 3.3 below shows the final setup of the experiment. For this experiment, about 120 bottles were used. Some bottles were used for basic solution, while others the plastics under test for biodegradation, some bottles were used for acid solutions while the final set of bottles contains basic solutions used for titration.

Optimal conditions for incubation involved temperatures of 58°C (with a tolerance of $\pm 2^\circ\text{C}$) and the absence of vapours that could hinder microbial growth. The incubator's was performed in either complete darkness or diffused lighting, can facilitate such conditions.

In starting up the test, the following number of composting vessels were set up to start the test:

- 3 glass ware vessels for the PBS-based compound (film-shape) test material
- 3 glass ware vessels for the PBS-based compound (granule-shape) test material
- 3 glass ware vessels for the reference material
- 3 glass ware vessels for the blank material (Pure compost)

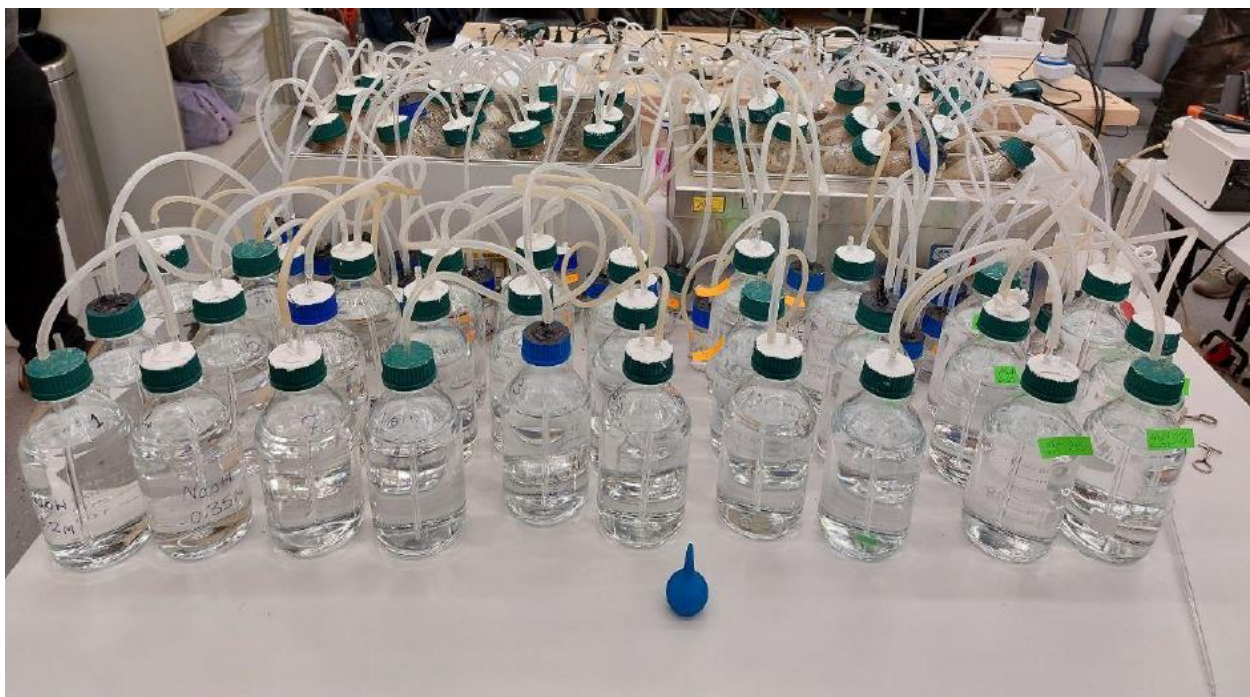


Figure 3.3 Final Setup of the experiment

3.6 Preparation of Mixtures

3.6.1 Indicator preparation

For this experiment, two step titration was carried out using Phenolphthalein and Methyl orange as indicators. Brief explanation about the preparation process will be enumerated below:

- **Phenolphthalein:** The main purpose for using phenolphthalein in this experiment was to serve as a PH indicator. Phenolphthalein is known to be fuchsia pink color when it's pH is between 8.3 - 10.5. Any PH value outside this range makes it become colorless. Phenolphthalein has properties such as low solubility in water but it is an organic compound that readily dissolves in Ethanol[13]

The process of preparing the phenolphthalein indicator for this experiment was quite straight forward. Phenolphthalein indicator solution of 0.5% has been prepared. To begin, 50 mL of 100-95% Ethanol is weighed, then 0.5 g of powdered Phenolphthalein

is added. The powder in the Ethanol must be thoroughly dissolved by stirring the solution. When these proces are completed, the solution is finally topped up with 100 mL of distilled water [14].

- **Methyl Orange:**

The process of preparing Methyl Orange indicator for this experiment was quite simple. Methyl orange indicator solution of 0.5% has been prepared. To begin, 50 mL of 100-95% Ethanol is weighed, then 0.5 g of Methyl orange chemical from the stock bottle is added. This chemical in the Ethanol must be thoroughly dissolved by stirring the solution. When these proces is completed, the solution is finally topped with 100 mL of distilled water [14].

3.6.2 NaOH solution preparation

The CO₂ absorption unit was constructed according to the prescribed procedure involving the mixing of NaOH with distilled water to produce solutions of varying concentrations (0.2M, 0.25M, and 0.3M). To obtain a final volume of 1 L, 24 g of pure NaOH was combined with 1 L of distilled water. Initially, the required quantity of NaOH was measured out and blended with approximately three-quarters of the total volume of distilled water. Considering the exothermic nature of NaOH dissolution, it is advisable to begin with a small additional amount of water and ensure thorough mixing before adding more. Subsequently, the remaining water was added gradually while maintaining appropriate mixing until complete dissolution of NaOH was achieved. Due to the emission of toxic fumes during the dissolution process, it is necessary to carry out NaOH handling in a well-ventilated environment [15].

3.6.3 Determination of total dry solids and total volatile solids.

Prior to commencing the experiment, a quantity of the inoculum was subjected to analysis to determine the total dry solids, estimate the volatile solids through combustion at 550 °C, and measure the total organic carbon according to the ISO 14855-1:2012 standard.

The moisture content of a substance is equivalent to its dry solids content. Therefore, the material was heated at 105°C for a duration of 9 hours (specific conditions may vary, such as temperature and duration) to evaporate the water without igniting the sample, resulting in the removal of moisture and attainment of dryness. The material

used in this project contained a specific quantity of total dry solids. In this experimental setup, 99.83% of the dry sample material remained in the PBS-based Compound (in granule form), 99.78% in the PBS-based Compound (in film form), and 95.6% in the reference cellulose. The respective weights of the test materials were as follows: vermiculite - 13.85g, PBS-based Compound (granule shape) - 13.86g, PBS-based Compound (film shape) - 13.86g, and cellulose - 14.47g per jar.

The combustible solid component was determined by subtracting the residual vermiculite after combustion at 550°C from the total dry solids of the same sample. By performing this calculation for each specimen, the total volatile solids can be determined. Moreover, this information provides insights into the organic content within each vessel.

See mathematical representation of TS(%) and VS(%) below:

TS - $\text{Solid(dry mass)/Solid(Wet mass)}$

VS - $[\text{Solid(dry mass)} - \text{oven deposit(dry mass)}] / \text{Solid(Wet mas)}$

3.7 Carbon dioxide determination

One of the major activity carried out during this test was titration. This was majorly to determine the practical amount of CO₂ evolved. The process of the tritration is enumerated below:

1. Using 20 ml pipet, three samples were drawn out of each vessel under consideration in order to be titrated.
2. The above samples were subjected to a two-step titration process using two different indicators (Phenolphthalein and Methyl orange).
3. Using a bourette, acidic solution is added to the basic solutions taken from the previous vessels. The basic solution turns into purple using two or three drops of Phenolphthalein.
4. By adding drops of acid, it could be determined at which volume of acid the mixture obtains equilibrium and gets clear again.
5. Then using two or three drops of Methyl orange, the solution turned light orange and by adding more acid, it could be determined at what added volume of acid the solution gets completely saturated by acid and the orange reddish colour of the solution does not change any more.

6. Both amounts are recorded for calculation of the amount of CO₂ emission and the range of biodegradability of the testing materials.
7. All the vessels which were used in the process are washed first with tap water and then with distilled water to be used for subsequent titrations.
8. The process was repeated for each titration round.

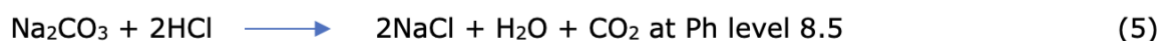
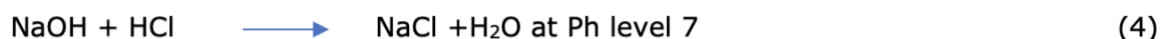
The evolved CO₂ reacts with NaOH in the following manner:



Na₂CO₃ Produced in the formula 3 above is soluble and does not precipitate.

The NaOH solution used for the purpose of CO₂ absorber will have both unreacted NaOH and Na₂CO₃ relating it to Formula 3.

During the titration, both chemical species reacted with HCl, as shown below:



As shown in Formula 6, the adsorbed CO₂ can be calculated by subtracting the H⁺ equivalents required to neutralise the original NaOH solution from the H⁺ equivalents required to neutralise the reactions represented by Formulae 4 and 5.

In practice:

$$\text{Mmol CO}_2 = (V_{\text{HCl}(5)} - V_{\text{HCl}(4/5)}) \times [\text{HCl}] \quad (6)$$

$$\text{mgCO}_2 = \text{mmol CO}_2 \times 44$$

Where:

Molecular weight (g/mol) of CO₂ is 44

V_{HCl(5)} is the volume of HCl (in ml) consumed in Formula 5;

V_{HCl(4/5)} is the volume of HCl (in ml) consumed in Formula 4 and 5

[HCl] is the concentration of the HCl (0.05 mol/l).

The precise quantity of carbon dioxide (CO₂) can be determined in milligrams (mg) using formula 6.

3.8 Computation of theoretical quantity of CO₂ (Carbon dioxide)

ThCO₂ produced by the test material was calculated in grammes per vessel using formula (7).

$$\text{ThCO}_2 = M_{\text{TOT}} * C_{\text{TOT}} * \frac{44}{12} \quad (7)$$

Where:

M_{TOT} - is total amount of dry solids in the test material, measured in grammes.

C_{TOT} - is total organic carbon content of the test material expressed as a percentage of the total dry solids in the sample, expressed in grammes per grammes.

44 - Is molecular mass of carbon dioxide (CO₂),

12 - Is atomic mass of carbon (C).

3.9 Percentage biodegradation computation

Formula (8) is used for calculating the percentage of biodegradation (D_t) of the test material during each measurement interval based on the total quantities of carbon dioxide released.

$$D_t = \frac{(\text{CO}_2)_T - (\text{CO}_2)_B}{\text{ThCO}_2} * 100 \quad (8)$$

Where:

(CO₂)_T - Is total amount of CO₂ produced in each composting vessel which contains test material (measured in grammes per vessel).

(CO₂)_B - Is mean cumulative quantity of carbon dioxide emitted per vessel.

ThCO₂ - Is theoretical amount of CO₂ that test material have potential to produce, in grammes per vessel.

4. RESULTS ANALYSIS

4.1 Average daily CO₂ production

The experiment spanned a duration of 180 days, with measurements of CO₂ emissions taken approximately every 10 days. Throughout this period, notable variations in CO₂ production were observed among the different materials investigated, as depicted in Figure 4.1.

Figure 4.1 illustrates that the highest level of CO₂ was generated by the Reference material on day 20, while the lowest CO₂ production was observed with the blank material on day 90.

Analysing each material used in the test, starting with the blank material (activated vermiculite used as compost), it exhibited consistently low daily CO₂ emissions, peaking at 1.526g on the 20th day and reaching a minimum of 0.276g on day 90.

As mentioned earlier, the Reference material displayed a peak average CO₂ generation of 4.871g on day 20 and a minimum of 0.403g on day 170. The PBS-based plastic in granules and film reached their respective peaks at 1.872g and 2.405g, with minimum emissions of 0.612g and 0.489g.

The blank material consistently demonstrated a desirable pattern of low CO₂ emission throughout the test, simulating an ideal representation of natural conditions. Comparing the CO₂ emissions of the PBS-based materials (granules and film) to the blank material, a similar trend is observed, indicating that PBS-based materials exhibit minimal CO₂ emissions when disposed of in the natural environment for biodegradation.

Conversely, the reference material mimicked the behaviour of conventional plastic, with consistently high CO₂ emissions throughout the test period.

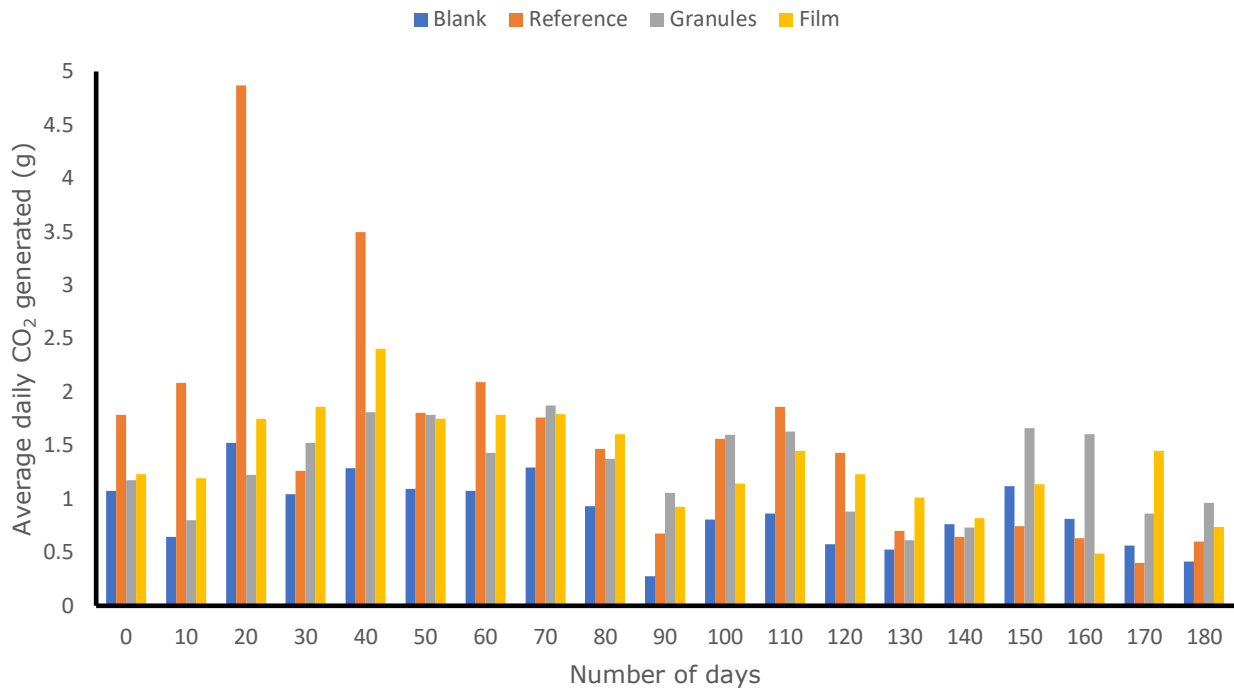


Figure 4.1 Average Daily CO₂ Production for blank, reference and PBS-based compounds (Granules and film)

4.2 Cumulative CO₂ generated

In this study, conducted over a 180-day period, the cumulative CO₂ generated by different materials was assessed. Figure 4.1 indicates that the reference material exhibited the highest average daily CO₂ generation throughout the test duration. Figure 4.2 provides insights into the accumulated mass of CO₂ generated by each material, including Blank, Reference, Granule, and Film. The accumulated CO₂ masses were recorded as follows: 16.693g for Blank, 29.902g for Reference, 24.604g for Granule, and 25.771g for Film. Notably, the CO₂ generation patterns of the PBS-based compounds (Granule and Film) closely resembled that of the Blank material. These findings highlight the favourable characteristics of PBS-based compound, emphasizing its environmental friendliness in comparison to conventional plastic materials represented by the reference material. The study suggests that PBS-based compound holds great promise as a potential substitute for traditional plastics and represents a significant advancement in the field of bioplastics.

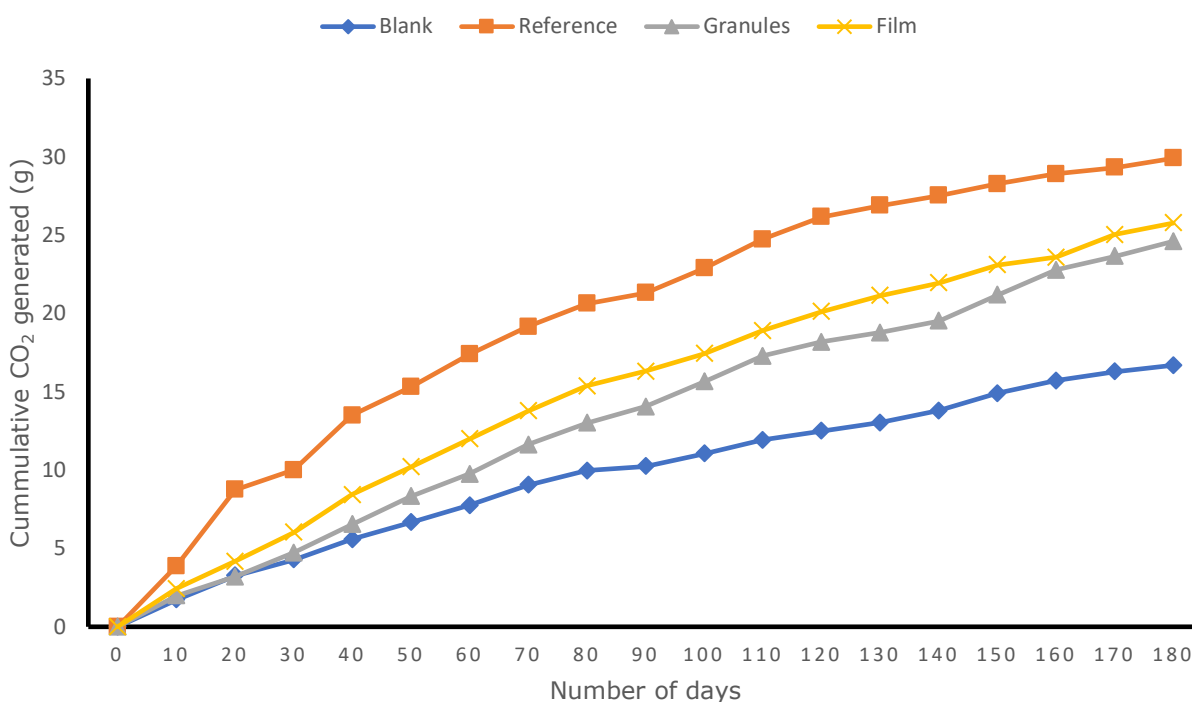


Figure 4.2 Cumulative CO₂ generated from blank, reference and PBS-based compounds (Granules and film)

4.3 Calculation of Theoretical amount of CO₂

To calculate the theoretical amount of CO₂, we need some parameter such as:

Molecular weight of CO₂, atomic Mass of C, total amount of dry solids (M_{TOT}), Total organic carbon (C_{TOT}) and Material weight per vessel(g).

The theoretical amount of CO₂ for PBS-based Compound (granule shape) will be calculated using the parameters in table 4.1.

Table 4.1 Parameters for the calculation of theoretical amount of CO₂.

Test Material	M _{TOT} (%)	C _{TOT} (%)	Material weight per vessel(g)
Granule shape	99.83	54	13.85
Film shape	99.78	53	13.86
Reference Cellulose	95.6	44	14.47

Where:

Molecular weight of CO₂ - 44g/mol

Atomic Mass of C - 12g/mol

$$\text{ThCO}_2 = M_{\text{TOT}} * C_{\text{TOT}} * \frac{44}{12} \quad (7)$$

- Theoretical amount of CO₂ for PBS-based Compound (granule shape):

$$\text{ThCO}_2 = ((99.83/100) * 13.85) * (54/100) * \frac{44}{12} = 27.376 \text{ g}$$

- Theoretical amount of CO₂ for PBS-based Compound (film shape):

$$\text{ThCO}_2 = ((99.78/100) * 13.86) * (53/100) * \frac{44}{12} = 26.875 \text{ g}$$

- Theoretical amount of CO₂ for Reference Material:

$$\text{ThCO}_2 = ((95.6/100) * 13.83) * (44/100) * \frac{44}{12} = 22.318 \text{ g}$$

Table 4.2 below shows the theoretical amount of CO₂ and the various parameters needed for calculations of the various test materials.

Table 4.2 Parameters for the calculation of theoretical amount of CO₂.

ThCO₂	M_{TOT} (%)	C_{TOT} (%)	Material weight per vessel(g)	Material
27.376	99.83	54	13.85	Granule shape
26.875	99.78	53	13.86	Film shape
22.318	95.6	44	14.47	Reference Cellulose

4.4 Degree of biodegradability based on Maximum Reference Cellulose Material

As shown in Figure 4.3, PBS-based compounds (Granules and film) degraded progressively all through the entire test period. The Film material has shown more biodegradability when compared to the granule material.

Figure 4.4 (a and b) below shows the outcome of PBS-based compound: granules and film after 180 days degradation. A close inspection at this material shows that these materials could break into flakes if squeezed light by hand.

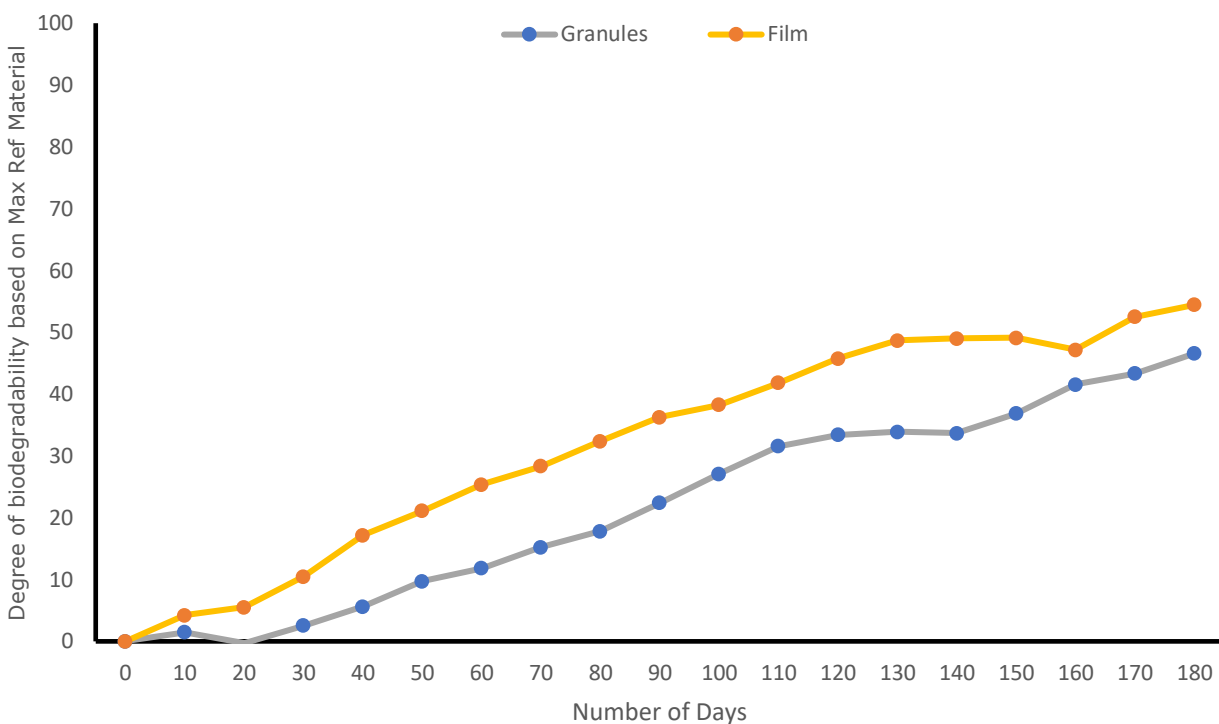


Figure 4.3 Degree of biodegradability of PBS-based compounds (Granules and film) based on Maximum Reference Cellulose Material

4.5 Observation of PBS – based compound (granule and film) after 180 days

Figure 3.1 (a and b) shows PBS (Granule shape) and PBS (Film shape) respectively before 180 days biodegradation test started. While on the other hand, Figure 4.4 (a and b) below shows the biodegraded test material.

Considering the degree of biodegradability of granule shape material as shown in Figure 4.3, it could be seen that this material did not biodegrade for the initial first 20 days of the test as the curve of biodegradation went in the downward direction giving degree of biodegradation of -0.292 on day 20 of the test. Eventually the curve took an upward turn from day 30 and progressed steadily in degradation till the end of the test hence ending in degree of biodegradation of 46.56%.

The degree of biodegradability for film shape material as shown in Figure 4.3 unlike granules took a steady upward curve from beginning of the test till the end. As can be seen, the film shape PBS-based material at the end of 180 days had a degree of biodegradation of 54.43%. This means that film shape PBS-based material has degraded more than the granule shape material within the circumstances of test condition. The figure 4.4 (a and b) below gives a pictorial view of the end product of both test materials after 180 days of biodegradation.

Considering the physical properties exhibited by the PBS-based compound material after 180 days biodegradation. It could be seen that the film material became fragile and can basically be crushed by the hand after biodegradation and the colour changed a little bit but can still be recognised in the compost.

However, the PBS-based granule material could almost not be recognized in the compost and did not degrade as much the film material. This illustrates that PBS-based granule material does not readily degrade as much as the PBS-based film material.

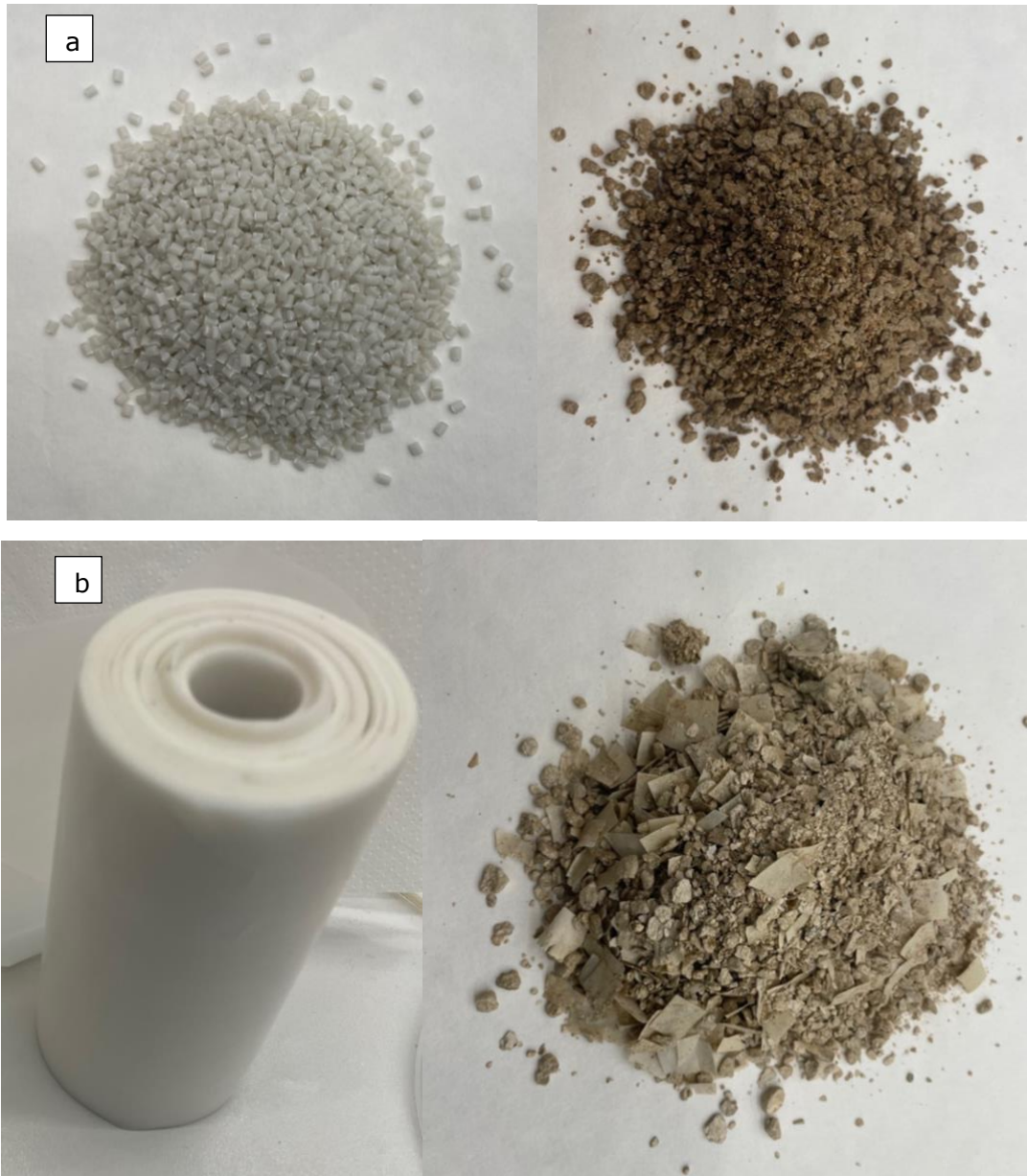


Figure 4.4 Observation of PBS-based compound after 180 days test (a) Granule shape, (b) Film shape.

4.6 Comparison with other Studies.

PBS-based compound was used in powder form for this comparative study carried out by B. P. Calabria et al., (2013). Degradation commenced on day 7. On day 60, the degree of biodegradation recorded was already 58%. On day 80, PBS-based compound (powder form) has moved to the plateau phase [17].

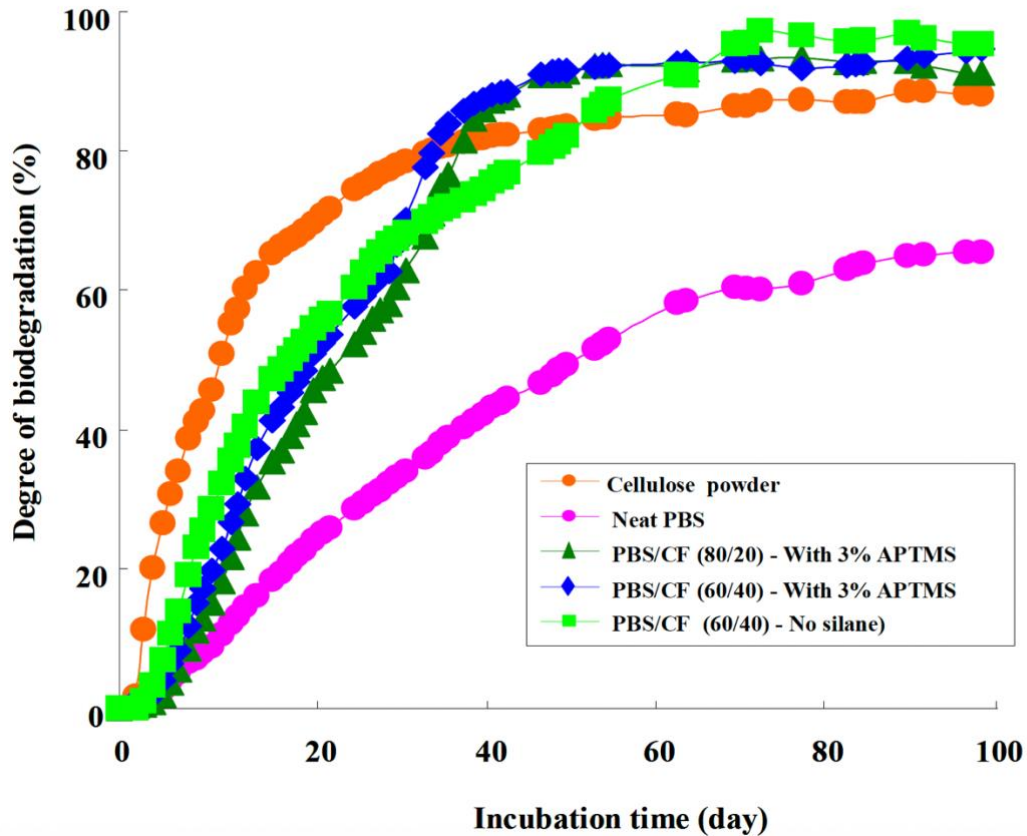


Figure 4.5 biodegradation rates of microcrystalline cellulose powder, pure polybutylene sulphide (PBS), and PBS/CF composite sheets (measuring 10 mm x 10 mm x 0.5 mm) in a controlled compost environment maintained at 58 °C [17].

This comparative study by B. P. Calabia et al., (2013) demonstrated that incorporating cotton fiber (CF) as a filler in polybutylene sulphide (PBS) yielded notable benefits, including enhanced tensile strength, and increased overall degradation rate of PBS/CF composites. The introduction of the silane coupling agent APTMS resulted in significant improvements in the tensile strength and crystallinity of the composites. While the addition of CF led to a decrease in the thermal stability of the composites, incorporating silane exhibited a minor enhancement in thermal stability compared to untreated composites due to the inherent thermal stability of silane.

Moreover, treating CF with APTMS led to a modest improvement in the interfacial adhesion between PBS and CF. Notably, the biodegradation rate of PBS/CF composites in a controlled compost environment at 58 degrees Celsius exceeded that of pure PBS, indicating that optimal outcomes can be achieved with PBS formulations incorporating both CF and APTMS [17].

Comparing the above to this project test result, PBS-based compound (Granule shape) has not been filled or treated with any reagent. Test samples has been used in its original state as bought from the manufacturer.

Regarding the material degradation, PBS-based compound (Granule shape) began to degrade after day 20 and it reached 46.562% degradation by 180 days. On the other hand, PBS-based compound (film shape) began to degrade after day 10 and reached 54.426% degradation by 180 days.

5. CONCLUSIONS

This research work was carried out within the scope and limitation of the ISO Standard ISO 14855 – 1 and the aim is to assess the biodegradation of (PBS)-based plastic under controlled composting conditions.

ISO 14855 – 1 recommends this experiment to last for a period of 6 months as is the case for this test to reach maximum degradation. In this test, some conclusions have been drawn as stated below:

1. During 180 days test under aerobic conditions, PBS-based plastic (film shape) degraded more than PBS-based plastic (granule shape).
2. It can be concluded that PBS-based compound has slow biodegradation rate since the standard recommends a 90% degradation in 6 months and the peak degradation recorded for this test was 54.426%. However, it can also be drawn that PBS-based compound (granule shape) has even slower biodegradation rate compared to PBS-based compound (film shape). Possible reason could be due to the larger surface area of the PBS-based compound (film shape) material.
3. Considering that this test was carried out under some standard conditions according to ISO 14855-1, there is a possibility of slight changes in test results when subjected to natural environment due to unstable temperature, uncontrolled oxygen flow, light and moisture.

This test was carried out within the specified period of 6 months as recommended by the standard which means maximum degradation possible was achieved. In the future, PBS-based compound could be subjected to varying conditions to determine the possibility of further degradation.

SUMMARY

Conventional plastics which consist of the greater number of plastics used all over the world today have many detrimental effects on the environment for various reasons. This has led to the presence of microplastics in the ocean today which poses a significant threat to the marine organisms as they can be ingested. This eventually introduces plastic into the food chain.

The proliferation of plastic in landfills leads to issues of land use and the generation of harmful leachate. It exhibits low recycling efficiency, contributing to waste accumulation and resource depletion. Incineration of it releases toxic chemicals into the atmosphere, posing risks to both human health and the environment.

To address these environmental concerns, biodegradable polymers are being advocated as a potential solution. Bioplastics can be classified into two categories: bio-based plastics, derived from renewable biomass sources, and biodegradable ones that break down in the environment, resulting in harmless compounds.

However, bioplastics do have certain drawbacks, including increased brittleness and lower melting temperatures. In order to overcome these limitations, enhancements are made to their properties, which can affect their biodegradability. Therefore, rigorous testing is crucial to evaluate the biodegradation process of these modified bioplastics.

Aerobic biodegradation within the scope of this experiment is the breakdown of plastic as much as possible under aerobic condition. The test materials used include PBS-based compound (granule shape and film shape), reference material (conventional plastic) and blank (compost).

To assess the ultimate aerobic biodegradability of plastic materials, several ISO standards have been developed. One commonly used method is ISO 14855-1, which employs continuous infrared analysis, gas chromatography, and titration techniques to measure the amount of carbon dioxide released during the degradation process.

As a result of this experiment, the following was achieved:

- The results of the ultimate aerobic biodegradation of PBS-based compound under controlled composting conditions was analyzed.
- The aerobic biodegradation of PBS-based plastic material in two different shapes, namely granules and film was observed and analyzed under controlled composting conditions for a period of 184 days.

- The CO₂ production and biodegradability of these biopolymers were measured, and the results obtained from the test was compared to those found in the literature.

The results for degree of biodegradation of PBS-based compounds (granule shape and film shape) obtained from this test shows that biodegradation has reached up to 46.562% for PBS-based compound (granule shape) and for PBS-based compound (film shape) up to 54.426% during 180 days. However, these results could have been influenced a little by gas leaks and human error.

LIST OF REFERENCES

- [1] Frontline. PBS. (2023) <https://www.pbs.org/wgbh/frontline/documentary/plastic-wars/>
- [2] European Commission – Environment. (18 January 2022). "Biobased, Biodegradable, and Compostable Plastics". https://environment.ec.europa.eu/topics/plastics/biobased-biodegradable-and-compostable-plastics_en
- [3] Mandala Project. (October 10, 2022). "What is the Difference Between Biobased and Biodegradable Plastics?". <https://mandalaproject.eu/what-is-the-difference-between-biobased-and-biodegradable-plastics/>
- [4] Polymer Database. (2022). "Polybutylene Succinate (PBS)". <https://polymerdatabase.com/Polymer%20Brands/PBS.html>
- [5] PlantSwitch. "Bioplastic vs Plastic: What's the Difference?". <https://www.plantswitch.com/bioplastic-vs-plastic/>
- [6] Jaime R Alvarado Bremer. OMICS International. (21-Apr-2022). Journal of Ecosystem & Ecography: "Aerobic and Anaerobic Biodegradation". <https://www.omicsonline.org/open-access/aerobic-and-anaerobic-biodegradation-119900.html#:~:text=Aerobic%20biodegradation%20is%20the%20breakdown,is%20characterized%20by%20oxidative%20conditions>
- [7] Polymer Database. (2022). BIODEGRADABLE POLYESTERS (BIOBASED) ALIPHATIC POLYESTERS. <https://polymerdatabase.com/polymer%20classes/Biodegradable%20Polyester%20type.html>
- [8] Mitsubishi Chemical Corporation. Biodegradable PolymerBioPBS. https://www.m-chemical.co.jp/en/products/departments/mcc/sustainable/product/1201025_7964.html
- [9] Roquette. (2023). "Polyesters (PBS)". <https://www.roquette.com/industries/performance-materials/polyesters-pbs>

- [10] ScienceDirect. (2023). "Polybutylene Succinate". <https://www.sciencedirect.com/topics/engineering/poly-butylene-succinate>
- [11] S. Ayu Rafiqah, Abdan Khalina, Ahmad Saffian Harmaen, Intan Amin Tawakkal, Khairul Zaman, M. Asim, M.N. Nurrazi, and Ching Hao Lee. PubMed Central (PMC). (29 Apr 2021). "A Review on Polybutylene Succinate (PBS): A Biodegradable Polymer for Sustainable Packaging Applications". <https://www.ncbi.nlm.nih.gov/pmc/articles/PMC8125033/>
- [12] Asanda Mtibe, Sudhakar Muniyasamy, Teboho Clement Mokhena, Osei Ofosu, Vincent Ojjo, and Maya John. Proquest. (Jan 2023). Express Polymer Letters: Recent insight into the biomedical applications of polybutylene succinate and polybutylene succinate-based materials. Vol. 17, no. 1. <https://www.proquest.com/openview/7bfdb1b9d474a3626b6be2b568571a37/1?pq-origsite=gscholar&cbl=2044900>.
- [13] Lee, H. J., Kim, D. G., Lee, J. H., & Cho, M. S. (2012). A Study for Carbonation Degree on Concrete using a Phenolphthalein Indicator and Fourier-Transform Infrared Spectroscopy. 152–158.
- [14] S. S., & DeCoste, D. J. (2012). Chemical principles. Cengage Learning.
- [15] Ensia. "Sorting out bioplastics: How bio-based and biodegradable plastics contribute to the circular economy". <https://ensia.com/features/bioplastics-bio-based-biodegradable-environment/>
- [16] Bioprocess Control. (2016). AMPTS II and AMPTS II Light: Automatic Methane Potential Test System.
- [17] B. P. Calabia et al., (2013). "Biodegradable poly (butylene succinate) composites reinforced by cotton fiber with silane coupling agent," vol. 5, no. 1, pp. 128-141.
- [18] Román-Ramírez LA, McKeown P, Jones MD, Wood J (2020) Kinetics of methyl lactate formation from the transesterification of polylactic acid catalyzed by

Zn(II) complexes. ACS Omega 5:5556–5564. <https://doi.org/10.1021/acsomega.0c00291>

[19] Al-Salem SM, Lettieri P, Baeyens J. (2009). Recycling and recovery routes of plastic solid waste (PSW): a review. Waste Management. 29:2625–2643. <https://doi.org/10.1016/j.wasman.2009.06.004>

[20] Kumar S, Panda AK, Singh RK (2011) A review on tertiary recycling of high-density polyethylene to fuel. Resour Conserv Recycl 55:893–910. <https://doi.org/10.1016/j.resconrec.2011.05.005>

[21] Grigore M (2017) Methods of recycling, properties and applications of recycled thermoplastic polymers. Recycling 2:24. <https://doi.org/10.3390/recycling2040024>

[22] Siddiqui J, Pandey G (2013) A review of plastic waste management strategies. Int Res J Environ Sci 2:84–88

[23] Singh N, Hui D, Singh R, et al (2017) Recycling of plastic solid waste: a state of art review and future applications. Compos Part B Eng 115:409–422. <https://doi.org/10.1016/j.compositesb.2016.09.013>

[24] Munasinghe M (2010) Addressing the sustainable development and climate change challenges together: applying the sustainomics framework. Procedia Social Behav Sci 2: 6634-6640.

[25] Kyte R (2014) Climate Change Is a Challenge for Sustainable Development. Gaidar Forum Moscow, Russian Federation.

[26] Princiotta FT, Loughlin DH (2014) Global climate change: the quantifiable sustainability challenge. J Air Waste Manag Assoc 64: 979-994.

[27] Martens P, Mcevoy D, Chang CT (2016) Climate change: responding to a major challenge for sustainable development. Sustainability Science.

[28] Ilatnieks O., Gaidukovs S., Barkane A., Sereda A., Gaidukova G., Grase L., Thakur V. K., Filipova I., Fridrihsone V., Skute M., Laka M. (2020). Bio-based poly (butylene succinate)/microcrystalline cellulose/nanofibrillated cellulose-based

sustainable polymer composites: Thermo-mechanical and biodegradation studies. *Polymers*, 12, 1472. <https://doi.org/10.3390/polym12071472>

[29] AL-Oqla F.M., Omari M.A. *Green Biocomposites*. Springer; Berlin/Heidelberg, Germany: 2017. Sustainable biocomposites: Challenges, potential and barriers for development; pp. 13–29.

[30] Bautista M., de Ilarduya A.M., Alla A., Vives M., Morató J., Muñoz-Guerra S. Cationic poly(butylene succinate) copolyesters. *Eur. Polym. J.* 2016;75:329–342.

[31] Su S., Kopitzky R., Tolga S., Kabasci S. (2019). Polylactide (PLA) and its blends with poly(butylene succinate) (PBS): A brief review. <https://doi.org/10.3390/polym11071193>.

[32] Gigli M., Fabbri M., Lotti N., Gamberini R., Rimini B., Munari A. (2016). Poly (butylene succinate)-based polyesters for biomedical applications: A review. *European Polymer Journal*, 75, 431-460. <https://doi.org/10.1016/j.eurpolymj.2016.01.016>