Impact of biobased packaging materials on quality of fully-baked frozen bread

Yoga Putranda

DIVISION OF PACKAGING LOGISTICS | DEPARTMENT OF DESIGN SCIENCES
FACULTY OF ENGINEERING LTH | LUND UNIVERSITY

2017

MASTER THESIS

FIPDes
Food Innovation & Product Design

This Master's thesis has been done within the Erasmus Mundus Master Course FIPDes, Food Innovation and Product Design.

Erasmus Mundus

FIPDes has been funded with support from the European Commission. This publication reflects the views only of the author, and the Commission cannot be held responsible for any use which may be made of the information contained therein.

Impact of biobased packaging materials on quality of fully-baked frozen bread

Yoga Putranda

LUND
UNIVERSITY

Impact of biobased packaging materials on quality of fully baked frozen bread

Published by
Division of Packaging Logistics
Department of Design Sciences
Faculty of Engineering LTH, Lund University
P.O. Box 118, SE-221 00 Lund, Sweden

Subject: Food Packaging Design (MTTM01)
Division: Packaging Logistics
Supervisor: Märit Beckeman
Co-supervisor: Karla Marie Batingan Paredes
Examiner: Daniel Hellström

This Master´s thesis has been done within the Erasmus Mundus Master Course FIPDes, Food Innovation and Product Design.

www.fipdes.eu

ISBN 978-91-7753-365-8

Abstract

The performance of five commercial available biobased packaging films for fully-baked frozen bread was studied in comparison to polyethylene (PE) film. The main purpose of this study was to understand how biobased packaging materials affect the quality of fully–baked frozen bread. The five biobased packaging films evaluated in this study were biobased polyethylene (bio-PE), Biodolomer®, polylactic acid (PLA), polybutylene succinate-co-adipate (PBSA), and paper laminated with biobased polyethylene terephthalate (paper/bio-PET). Two types of commercial fully-baked frozen bread were used i.e. hotdog buns and hamburger buns. The fully-baked buns were repacked manually in each packaging bag, sealed with a clip, and frozen for minimum 3 days. Weight loss, textural properties, water activity, and the appearance of the buns were evaluated 1, 3, and 7 days after thawing. Water vapor transmission rate (WVTR) of bio-PE was similar to the PE reference (6.37 g/m^{2}day), while Biodolomer®, PLA, PBSA, and paper/PET WVTR values were higher than PE reference, 71.1, 61.70, 140.12, and 25.09 g/m^{2}day respectively. The bio-PE could maintain all the investigated quality aspects of the packed buns along shelf life. A triangle test confirmed that there was no significant difference between hotdog buns packed in bio-PE and PE reference. Despite missing of transparent property, paper/bio-PET could maintain some of the bun quality aspects except the textural properties and weight loss (hotdog buns). Buns packed in Biodolomer®, PLA, and PBSA films showed quality loss i.e. higher weight loss already at day one after thawing and dry edge in the buns after 7 day-shelf life.

Keywords: biobased packaging material, buns, packaging, polyethylene, quality

Executive summary

Introduction

Global plastic production increases every year in which almost 40% of plastic demand in Europe was intended for packaging purposes (PlasticEurope, 2016). Reducing amount of fossil resources and environmental consideration from excavating it led many industries to look for more renewable resources to make plastic. Moreover, consumers, retails, and government regulation urge for a more sustainable packaging (McQuilken, 2016). Biobased packaging materials could be one of the solutions. However, suitability of biobased packaging application on specific food product needs to be evaluated to ensure the satisfactory of minimum product protection while improving the environmental impact of the packaging.

In this study, application of biobased packaging materials for fully-baked frozen bread was evaluated. Hotdog buns and hamburger buns were the types of bread chosen for the study provided by Lantmännen Unibake, Sweden. These breads undergo freezing in the package after baking and have 7-day shelf-life in retail shelves after thawing. It is an interest to understand the effect of the biobased packaging materials on the packed buns. The current packaging for these buns is petroleum-based polyethylene (PE).

Objectives

1. To identify the characteristics of different biobased packaging materials (film) that exist in the market.

2. To evaluate how biobased packaging material would maintain the quality of the hotdog buns and hamburger buns for 7-day shelf life after thawing in comparison with reference PE bag.

Materials and Methods

Total of six different packaging materials were used in this study. The five biobased packaging materials were biobased polyethylene (bio-PE), Biodolomer®, polylactic acid (PLA), polybutylene succinate-co-adipate (PBSA), and paper laminated with biobased polyethylene terephthalate (paper/bio-PET). Polyethylene (PE) is the reference packaging. Packaging water vapor transmission rate (WVTR) and puncture resistance were measured.

Fresh half-cut hotdog buns and hamburger bun were repacked manually and sealed with a clip. The buns were frozen at -18 °C for at least 72 hours. Thawing was done at room temperature for 24 h. Quality of bread in the package along shelf-life was evaluated i.e. weight loss, textural properties (crust hardness, crumb hardness, and crumb springiness), water activity, and appearance. Evaluations were performed day 1, 3, and 7 after the buns were thawed. Triangle test was performed to investigate if there is a significant difference between hotdog buns in bio-PE and hotdog buns in PE reference.

Results and discussion

Packaging evaluations

Transparency of the packaging is important for this product application. Bio-PE, PLA, and PBSA are transparent, providing good visibility of the product. On the other side, Biodolomer® is less transparent and paper/bio-PET is opaque. Less visibility of the product might affect how consumer perceive the overall product.

Water vapor transmission rate (WVTR) is an important parameter in packaging selection. WVTR value of bio-PE was not significantly different compared to both PE reference, 3.96-6.37 g/m^2day. Biodolomer®, PLA, PBSA, and paper/PET WVTR values were significantly higher than PE reference, 71.1, 61.70, 140.12, and 25.09 g/m^2day respectively.

Puncture resistance is a characteristic of flexible packaging materials that is related with packaging durability against impact from external penetration (Lange et. al., 2002). All the biobased packaging materials evaluated in this study had similar or higher puncture resistance compared to reference. Therefore, the performance against eternal penetration is expected to be similar or better.

Packed buns evaluations

Weight loss of hotdog buns and hamburger buns in bio-PE were not significantly different compared to PE reference day 7 after thawing. Paper/bio-PET could only maintain the weight loss of hamburger buns. Other packaging materials could not maintain weight loss of the bun even in the first day after thawing. Higher weight loss might be caused by the higher packaging WVTR value.

In term of textural properties, hotdog buns and hamburger buns packed with bio-PE had the most similar textural properties to hotdog buns in PE reference. These results correspond with the result of water activity and appearance of the buns (no dry edge observed). The triangle test confirmed that there was no significant difference between hotdog buns packed with bio-PE and hotdog buns packed with PE reference.

Paper/PET could limit the dry edge of the buns. However, it could not maintain crumb hardness (hotdog bun) and crumb springiness (hamburger bun). Dry edge was observed on the buns packed with Biodolomer®, PLA, and PBSA day 7 after thawing. In fact, these packaging could not maintain at least two out of three bun textural properties measured in this study.

Conclusions

Biobased packaging materials used in this study have different characteristics and barrier properties. Bio-PE had similar characteristics with the PE reference. Other biobased packaging materials had higher WVTR values and some of them are not transparent i.e. Biodolomer® and paper/bio-PET.

Bio-PE could maintain the quality of hotdog buns and hamburger buns in 7-day shelf life after thawing. A triangle test confirmed that there was no significant difference between hotdog buns packed in bio-PE and PE reference. On the other side, Biodolomer®, PLA, and PBSA did not perform well for this type of application which showed in the higher weight loss and dry edge of the buns in day 7 after thawing. Paper/bio-PET is an interesting biobased packaging material since it could maintain some of the quality parameters.

Recommendations for further studies

It might be interesting to evaluate the performance of other commercial biobased packaging materials. Packaging material thickness might also influence its performance. Therefore, it might be an interest to evaluate how different thickness of biobased packaging materials affects the quality of the packed product.

For fully-baked frozen bread, it is important to evaluate the performance of the packaging materials along frozen storage. This needs to be studied further.

Environmental impacts of biobased packaging materials might be an interesting issue to compare. Together with the price analysis, it could help to find the best biobased packaging option.

It might be interesting to evaluate biobased packaging materials for other type of product such as crusty bread, such as pastries, which require a more breathable packaging. Packaging for frozen bread can be another application to be evaluated since the water loss during freezing is minimum.

Acknowledgments

This master's thesis project marks the end of my master's study in Food Innovation and Product Design (FIPDes) which would not be possible without support from Erasmus Mundus Scholarship. I would like to thank all FIPDes consortium members especially Barbara Rega, Daniel Hellström, and Erik Andersson who made it possible for me to pursue this master and specialization. Special thanks to Isabelle Laissy who has helped me so much throughout the master.

I would like to thank to my supervisors from Lund University, Märit Beckeman and Karla Marie B. Paredes. Both of you have guided me from the early stage of my thesis and provided me with many valuable inputs in every stage in my work. I do appreciate it. I would like to thank Daniel Hellström, the examiner of my thesis.

I would like to thank to Lantmännen for supporting the research in this study. I am very grateful for having Annelie Moldin and Emma Nordell as my supervisors from Lantmännen who supported and trusted me in carrying out this project. Thank you for your valuable input to my work. I learned a lot during the project. Special thanks to people at Lantmännen Unibake who helped me preparing bread sample for my experiment. I would like to thank Ingegerd Sjöholm who facilitated me to conduct the lab experiment at the Dept. of Food Technology, Engineering and Nutrition, Lund University.

Packaging material is one of the most crucial things in this project. My great gratitude goes to Amerplast, Flextrus, GAIA Biomaterials, Reverdia and Sidaplax for providing packaging material samples and information. I do appreciate your support to this project.

I also want to thank all my FIPDes friends who made this 2-year of master's journey very exciting and unforgettable. For FIPDes Lund friends, I will never regret to spend my last master's year with all of you.

Finally, I would like to thank my parents and my sister Anna for always being there for me in every stage in my life. Thank you, I made it to this point.

Lund, June 2017

Yoga Putranda

Table of contents

List of tables

List of figures

List of acronyms and abbreviations

AIB	American Institure of Baking (AIB)
a_W	water activity
bio-PBS	biobased polybutylene succinate
bio-PE	biobased polyethylene
bio-PET	biobased polyethylene terephthalate
bio-PP	biobased polypropylene
CO_2TR	carbon dioxide transmission rate
DMT	dimethyl terephthalate
ERH	equilibrium relative humidity
FBF	fully-baked frozen
GHS	greenhouse gases
HDPE	high density polyethylene
LCA	life-cycle assessment
LDPE	low density polyethylene
MEG	mono ethylene glycol
MFC	microfibrillated cellulose
MFSL	mold-free shelf life
MJ	megajoule
MT	million tonnes
NFC	nanofibrillated cellulose
OTR	oxygen transmission rate
PBS	polybutylene succinate
PBSA	polybutylene succinate-co-adipate
PE	polyethylene
PET	polyethylene terephthalate
PHA	polyhydroxy-alkanoat
PHBV	poly (hydroxybutyrate-co-hydroxyvalerate)
PLA	polylactic acid
PP	polypropylene
PTA	purified terephthalic acid
TPS	thermoplastic starch
WVTR	water vapor transpiration rate

1 Introduction

This thesis is about evaluating the performance of different biobased packaging materials for fully-baked frozen bread application in comparison to a petroleum-based polyethylene (PE). This first chapter explains the background, aims, and delimitation of the study. The company involved is Lantmännen in Sweden.

1.1 Background

Plastic production has grown significantly around the globe. The increased production of plastic results in more fossil resources being exploited. Figure 1 shows that the production of plastic reached 322 MT in 2015, increased by 3.5% from the previous year. In Europe, plastic grows stable with 39.9% of total 49 MT demand in 2015 came from packaging application (PlasticEurope, 2016).

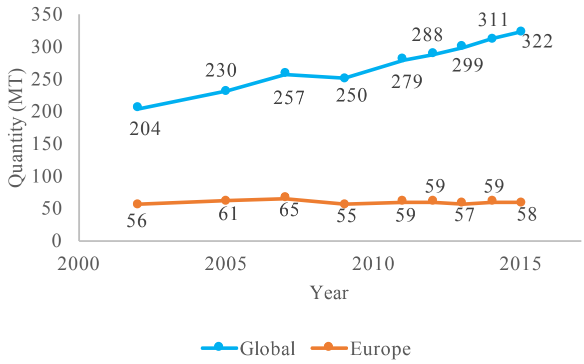


Figure 1 Plastic production globally and in Europe in 2002-2015, adapted from (PlasticEurope, 2015, 2016)

Reducing amount of finite fossil resource and environment consideration from excavating it lead many industries to look for possible renewable resources. Biobased material is one of the solutions for packaging that promise product protection, reduced environmental impacts, and better end-of-life option. Moreover, consumer concern on environmental impact creates a demand for more sustainable packaging. Not only affecting public perception, sustainable packaging becomes more of a requirement driven by retailers and government regulations (McQuilken, 2016).

While sustainability of packaging is one of major concerns, food packaging must perform its basic roles; protection and preservation, containment and food waste reduction, marketing and information (Marsh & Bugusu, 2007). Thus, selection of packaging for food needs to consider all those aspects.

Bread with one week shelf life is categorized as a short shelf life product that is mostly packed in polyethylene (PE) bag (Man, 2015). Global retail volume sales of bread accounts 85% of total baked goods, with retail volume 120 million tonnes in 2013 (see Figure 2). Packaged bread was one of the best performing baked good categories over 2008-2013, growing by 2 million tonnes (Euromonitor International, 2014). With this high number of packed bread volume, there is a consequence on the usage of packaging material. In fact, PE is number one of the most demand polymer type for packaging application in Europe in 2015 (PlasticEurope, 2016).

Figure 2 Global baked goods sales by category in 2013, adapted from (Euromonitor International, 2014)

Shifting the petroleum-based PE to biobased packaging could be one way to reduce the environmental impact from the packaging material. Excessive usage of packaging leads to negative impact to the environment while underpacking could lead to even higher negative impacts caused by product and packaging waste (Hellström, Olsson, & Nilsson, 2016b). Therefore, the suitability of biobased packaging application on specific product needs to be evaluated to ensure the satisfactory of minimum product protection while improving the environmental impact of the packaging.

A lot of research have been conducted on biobased packaging materials regarding the properties of the materials. This thesis contributes to fill the gap of knowledge in comparative study of biobased packaging materials for food product application.

1.1.1 Problem description

Many biobased packaging materials are being developed and commercialized. New materials and composite materials are coming to the market. Flooded information from packaging suppliers needs to be supported with the academic research. It is difficult for food industry as the customer to filter the different information about the performance of biobased packaging materials in different product application. This study aims to investigate the overview of the commercially available biobased packaging materials and its performance for food application in comparison with the reference fossil-based packaging material.

In this study, application of biobased packaging materials for a specific type of bread was evaluated. Fully-baked frozen bread i.e. hotdog buns and hamburger buns were the types of bread chosen for study provided by Lantmännen Unibake, Sweden. This type of bread undergoes freezing in the package after baking with short shelf life, 7 days in retail shelf after thawing. It is an interest to understand the effect of the biobased packaging materials on the packed buns. The current packaging for these buns is petroleum-based polyethylene (PE).

1.2 Objective

The research question of this thesis is *'how does biobased packaging material affect the quality of fully-baked frozen bread in comparison to reference packaging?'*

The objectives of the thesis include:

1. To identify the characteristics of different biobased packaging materials (film) that exist in the market.

2. To evaluate how biobased packaging material would maintain the quality of the hotdog buns and hamburger buns for 7-day shelf life after thawing in comparison with reference PE bag.

1.3 Focus and delimitations

The research focuses on the commercial available biobased packaging materials (flexible film) for food packaging application. In this study, biobased packaging materials supplier was limited to European suppliers. Biobased packaging materials used in the experiment were limited to the available sample from suppliers. Commercial hotdog buns and hamburger buns provided by Lantmännen Unibake, Sweden, were the bread used in the experiment. Evaluation of the buns focused on the quality aspects i.e. weight loss, textural properties, water activity, and the appearance along 7-day shelf life after thawing. Thawing was done at room temperature and humidity. Frozen shelf life quality of the bread was not investigated. Furthermore, material cost-related comparison is not considered in this study.

2 Theoretical framework

This chapter consists of literature reviews focus on two main topics that support this research. The first topic is about fully-baked frozen bread, bread quality changes during storage, and roles of packaging. The second part is about the overview of biobased packaging materials i.e. about the material, application, end-of-life characteristic, and commercial products.

2.1 Fully-baked frozen bread

Fully-baked frozen (FBF) bread is a type of bread produced with conventional bread making process and then it is frozen. The manufacturing process of FBF bread includes mixing-resting-dividing-shaping-fermentation-baking with addition of freezing step after the bread is fully-baked (Le Bail et al., 2006). Illustration of FBF bread making process can be seen in Figure 3.

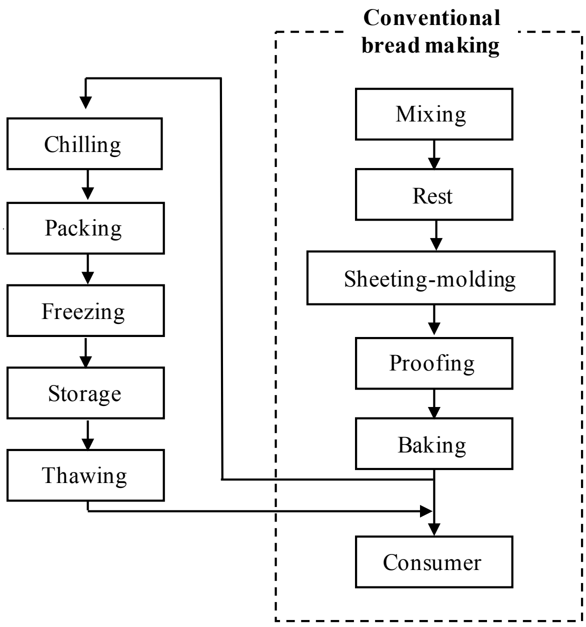


Figure 3 Production process flow diagram of fully-baked frozen bread, modified from ((Le Bail, Tzia, & Giannou, 2011)

Freezing application in bakery products has been developed since ninetieth century. This application becomes more popular because it provides extended shelf life, postpones the proofing-baking time, and allows production of fresh product (Le Bail et al., 2011). Freezing reduces the availability of free water and slows down chemical and enzymatic reactions (Sablani, 2011).

The first FBF bread was produce in 1970s (Le Bail et al., 2011). FBF bread benefits from long shelf life as well as the flexibility to produce the bread. Frozen storage makes it possible to store the bread longer prior to shipping. Moreover, it is possible to produce the bread months before the sales peak season.

Frozen bakery product plays an important part in the bread market share. In Europe, expansion of frozen bread is driven by (Le Bail et al., 2011):

1. Research on convenient products that can be easily prepared and promise 'fresh' quality for consumers.
2. Consumer demands on vast range of bakery products which are unprofitable to be prepared by retailers.
3. The urge of reducing products that are unsold and wasted.

2.2 Bread quality changes during storage

During the storage before consumption, significant changes continuously occur in bakery products. Organoleptic properties change and sometimes physical appearance change are two phenomena that affect the baked products quality (Cauvain & Young, 2009a). Some of the major bakery products characteristics that change during storage is listed in Table 1.

Bread staling, moisture transfer, and mold growing are three main bread spoilage mechanisms. Bread staling is considered as the result of various organoleptic changes in bread during storage. Loss of perceived freshness is a better descriptor than staleness since staleness covers diverse changes in bakery products (Cauvain & Young, 2009a; Man, 2015).

Table 1 Bakery products characteristics that change during storage, adopted from (Cauvain & Young, 2009a)

Crust or product crispiness
Crumb and crust moisture
Crumb firmness
Taste
Aroma

2.2.1 **Bread staling**

The four major quality factors in food include appearance, flavor, texture and nutrition. For bread, texture is important for the overall acceptability of the bread. This aspect has significant contribution to the total quality of the bread equivalently with flavor and appearance (Bourne, 2002).

Bread staling has been studied over centuries and it is responsible for a major economic loss in bakery industry. Staling in bread results in a firmer texture and dryer characteristic. This is caused by moisture migration from the crumb to the crust which lowers the crumb's moisture content. Factors that affect moisture migration include storage conditions, thickness of the crust, ratio of crust to crumb in the product and porosity of the crumb structure. Moisture migration leads to a soft leathery crust and increase in crust toughness. In fact, the textural changes in crumb are more important to consumer than changes in crust. The mechanism of crumb staling is very complex and less understood. Furthermore, availability of >20-30% water content in bread allows for starch retrogradation which contributes for the crust firming and decrease of springiness. The process is time and temperature dependent which maximum change occurs at 4 °C. At this temperature, maximum starch recrystallization happens (Cauvain & Young, 2009a; Cauvain & Young, 2010; Gray & Bemiller, 2003). Figure 4 illustrates the effect of storage time and temperature to bread firmness. As the bread is stored longer, the firmness value increases. Furthermore, as explained earlier, storing bread at 4 °C will result the fastest firming rate which decreases as the storage temperature rises.

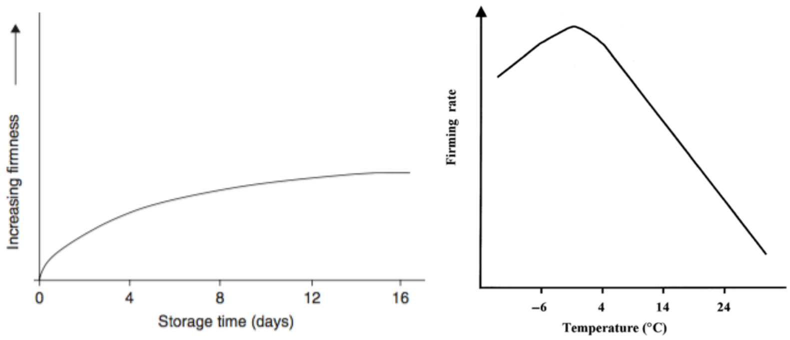


Figure 4 Effect of storage time and temperature to bread firmness, adopted from (Cauvain & Young, 2009a; Cauvain & Young, 2010)

Bread staling does not occur at temperature lower than -5 °C. Storage of bread under freezing condition under its glass transition temperature can maintain its quality if there is no significant moisture loss during storage. In fact, moisture transfer does not stop completely in sub-zero temperature. If there is a moisture loss during storage, a lower water content of the final thawed product will be achieved. This result a more 'stale' perception for the consumer. Furthermore, the freezing and thawing process of the bread is equivalent to 24-hour storage of the bread in ambient temperature (20 °C) (Cauvain, 2017b; Cauvain & Young, 2009a; Man, 2015).

2.2.2 **Moisture migration**

Water in bread is important because it has effect on the freshness perception of bread. Aspects related to the presence of water i.e. moisture content and water activity (a_W) are explained in *Section 2.3.1*. Moisture content could be a deciding aspect for consumers to say whether a product is freshly baked or not. The higher moisture content (within limit), the fresher is the product perceived by consumers. Water plays a role in lubricating while the product is eaten. In the point of purchase, soft characteristic with combination of a good recovery is usually evaluated by consumer by squeezing the bread (Cauvain & Young, 2009a)

There are two mechanism for moisture migration in bakery products, direct diffusion and vapor phase transfer. Direct diffusion occurs when two or more components with different water activities are in contact. The greater different of the water activities, the faster the rate of diffusion will be. In vapor phase transfer mechanism, moisture migrates from component with higher equilibrium relative humidity (ERH) to the one with lower ERH via surrounding atmosphere. This mechanism is most evident for wrapped product (Cauvain & Young, 2010).

Packaging influences the rate of moisture movement within and from the product. Moisture impermeable film will result in faster equilibrium. This leads to crust softening but little loss of moisture from the product overall. This situation is more suitable for pan bread. On the other side, crusty breads require moisture gradient throughout the product to maintain crust crispiness. A perforated film is suitable for crusty products. With this kind of packaging film, crumb moisture content of crusty product might fall rapidly (Cauvain & Young, 2010).

Total quantity of product moisture that move into the pack atmosphere depends on the product ERH and ratio of water mass in the air within the pack. Higher volume of air in the pack requires greater mass of water to saturate that atmosphere to reach equilibrium (Cauvain & Young, 2009b).

2.2.3 **Mold growing**

Microbial activity in bakery product is closely related with water activity. Spoilage risk of baked products depends on the water activity of the product. Bread with a_W 0.90-0.97 are prone to bacterial spoilage ('rope'), mold growth, and 'chalk molds'. Medium with lower water activity could inhibit the microbial growth. Lower ERH, for instance, can reduce the risk of microbial growth. However, some microorganism can be present in dormant condition in this environment and start to grow when the condition is more favorable. Moving wrapped products to a warmer condition might lead to water accumulation on the surface of the bread. This increases the water activity in the surface of the bread that consequently increases the risk of microbial growth. This issue often occurs for the packaged frozen product when defrosted (Cauvain & Young, 2010).

Mold growth is the main spoilage agent in bakery products. Spores of mold usually contaminate the surface of the bread right away after baking. Furthermore, product handling after baking at the manufacturer contributes to increase the load of microbial contamination. High enough ERH of the product can promote the growth of mold. Once the mold grows, it will reproduce and eventually could be visible as a mold colony. At this point, the product is already spoiled (Cauvain & Young, 2010).

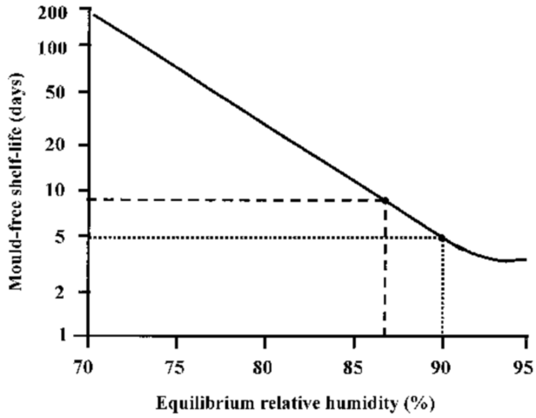


Figure 5 Relationship between product ERH and MFSL at 21 °C, adopted from (Cauvain & Young, 2009b)

Evaluating the time when the mold colony is visible is a way to decide mold-free shelf-life (MFSL) of bakery products. In a specific temperature, lower ERH will increase the MFSL. Figure 5 shows the relationship of ERH and MFSL at 21 °C where reducing ERH can increase MFSL. For bread, however, reducing ERH would be less effective to increase MFSL since the MFSL changes minimally for high ERH products. Moreover, reducing ERH of bread might influence the textural properties of the bread (Cauvain & Young, 2009b).

2.3 Bread quality measurements

2.3.1 **Moisture content and water activity**

In bakery product, there are two aspects related to the presence of water; moisture content and water activity (a_W). Moisture content related with the percentage of water in the product while a_W is related to potential on how water may behave within the product or between other components inside the product (Cauvain & Young, 2010).

Water activity (a_W) indicates the 'availability' of the water in a given solution. The value of a_W is temperature dependent (Roudaut & Debeaufort, 2010). It is calculated as vapor pressure of water above a sample (p) with vapor pressure of pure water (p_0) at that temperature. Equilibrium relative humidity (ERH) is the atmosphere condition in contact with the solution. When the atmosphere and the solution are in equilibrium, a_W equals to ERH. The ERH of the product is a unique humidity where there is no moisture gained or lost from the product (Cauvain & Young, 2010).

$$a_W = \frac{p}{p_0} = \frac{RH}{100};\ a_W = \frac{ERH}{100}$$

For each bakery product, there is a relationship between moisture content and water activity. In a specific product, an increase or decrease in water content will cause increase or decrease in the water activity accordingly (Cauvain & Young, 2009a).

2.3.2 **Texture analysis**

> *"The textural properties of a food are that group of physical characteristics that arise from the structural elements of the food, are sensed primarily by the feeling of touch, are related to the deformation, disintegration, and flow of the food under a force, and are measured objectively by functions of mass, time, and distance." (Bourne, 2002).*

Measurement of bread textural properties can be done using trained panelists or instrumental analysis. Instrumental analysis provides a more objective result. Firmness and resilience of the bread are two major textural properties that are used to measure the freshness of bread. Firmness related with the force to compress the crumb and resilience is related with crumb spring back characteristic when pressed. Spring back characteristic is less important than the softness of the bread (Cauvain, 2017a). Some key textural properties of bread are listed in the Table 2.

Table 2 Key textural properties of bread (Cauvain, 2004)

Textural properties	**Explanation**
Moistness	Directly related with the moisture content of the product. Bread crumb is expected to be moist, while bread crust is expected to be dry.
Firmness/hardness	Generally used to describe loss of softness in the crumb. It is considered as negative attribute for most bakery products.
Softness	Positive attributes for bread crumb.

Cohesiveness	Crumb of bread is expected to form a ball in the mouth and to require effort to chew. This attribute is related to moisture content. Loss of moisture results in crumbliness.
Springiness	Bread crumb is expected to be springy or to be recovered after compressing force.
Staleness	Collective change to bread texture that results in loss of its 'fresh baked' characteristic.

Compression/deformation test is one way to measure bread texture. The test is conducted by subjecting the force and distance compression to the sample using texture analyzer. Specific probe is used to do compression test. Another test to measure bread crust hardness is penetration test. This test is performed with needle-type probe or probe with small diameter (Cauvain, 2004).

Bread staling can be measured with texture analysis. Compression method with 25% compression (AACC Method 74-09) is the most effective method to determine the degree of firmness due to staling. Firmness of the bread correlates positively with the staleness measured by consumers (Baker, Walker, & Kemp, 1988; Gray & Bemiller, 2003). Texture profile analysis (TPA) with 40% compression shows a positive relationship between hardness and some sensorial parameters; "difficulty in swallowing", "crumbliness", "hardness" and "oral dryness". Decreasing of bread springiness over 20-day storage was noticed with higher TPA compression, 80% compression (Fiszman, Salvador, & Varela, 2005).

2.4 Current packaging

Petroleum-based low density polyethylene (LDPE) bag is a common packaging for frozen food including frozen bread (Cooksey & Krochta, 2011; Lee & Sun, 2011). PE can be made with two different polymerization process; 1) polymerization of ethylene at high temperature and pressure in absent of oxygen and 2) polymerization of ethylene at lower temperature and pressure in the presence of Ziegler-Natta catalyst. LDPE film has characteristics of clarity, easy processing by extrusion, and low permeability to water vapor but it is not to a good barrier to gasses, oils, or volatiles (Akelah, 2013). LDPE with a density around 910 kg/m^3 has physical characteristics of soft and stretchable (Lee & Sun, 2011). Moreover, it can withstand freezing temperature up to -70 °C (Kondo, 1990). High density polyethylene (HDPE) is highly used as food packaging due to its functional properties and relatively low price per unit area (Akelah, 2013).

2.5 Roles of packaging

Food packaging shall provide its basic function for containment, protection, communication, convenience, and reducing environmental impacts (Cooksey & Krochta, 2011). Different food requires different packaging characteristics due to its nature of product and processing. Thus, selection of packaging material must consider the balance between barrier properties of the materials with suitability of packaging form, method of preservation, and handling after purchase (Akelah, 2013).

2.5.1 **Containment**

Containment has been a fundamental function of the packaging since the early development of packaging concept (Cooksey & Krochta, 2011). Packaging should hold and keep the product or the surrounding safe. This function of packaging ensures that the product is collected in an assembled unit (Hellström, Olsson, & Nilsson, 2016a).

2.5.2 **Protection**

Packaging needs to provide sufficient protection to the contained product. Food packaging must protect the product from outside environment effect such as water, water vapor, gasses, odors, microorganism, dust, shock, vibration, compressive forces, etc. and protect the environment from the product (Robertson, 2016). Moreover, food packaging has a role in food safety, including preservation, limiting contamination, maintaining quality of the food, and extending shelf life (Akelah, 2013).

Frozen food products manufacturing process includes freezing process as a preservation method. Packaging of frozen food should be durable against the freezing temperature along shelf life. It must be able to be stored in the low temperature condition while still maintain its protection to the product. Furthermore, it has to protect the product from physical abuse during handling and to preserve the product from frozen quality problems (Cooksey & Krochta, 2011).

Fully-baked frozen bread is a unique type of product that requires to be stored in the freezing temperature and ambient temperature after thawing. Therefore, the packaging should not only perform well in the freezing condition, but also maintaining the product quality along the product shelf life after thawing. Fully-baked frozen bread packaging shall preserve the bread quality for 1 year shelf life under -18 °C storage and it should be sufficient to maintain the quality of the bread 7 days after thawing.

2.5.2.1 Packaging key properties

Key properties of packaging related to product shelf life that need to be considered are barrier properties; ratio of surface area and volume; and package closure and integrity.

1. Barrier properties

Nature of packaging material can give impact to product quality during storage. The choice of packaging material might affect the moisture content of the product which contribute to product textural properties and microbial shelf life. This is caused by the fact that polymer wall is usually permeable to some extend of water vapor and small molecules like gasses. Water vapor and oxygen permeability are two main barrier properties studied in food packaging (Robertson, 2016; Siracusa, 2012).

Water vapor transpiration rate (WVTR) is a parameter to be considered in selecting the packaging material for specific product, usually expressed as g/m^2 day. The lower WVTR value, the lower will be the gain or loss of water from product. Packaging WVTR, product moisture content, and a_W are important information to understand the change of wrapped product during storage. In most bakery product, packaging is needed to limit the excessive movement of water from product (Cauvain & Young, 2009a).

Beside water vapor permeability, oxygen permeability is also an important packaging property. Most of the time oxygen is responsible to product degradation from oxidation until changing the organoleptic properties of food products. Moreover, oxygen barrier property of packaging is important for fresh food product which contribute to the shelf life determination of the product. Packaging oxygen barrier is usually called as oxygen transmission rate (OTR). Another important gas barrier property of packaging is carbon dioxide barrier property, carbon dioxide transmission rate (CO_2TR). OTR and CO_2TR are usually expressed in cc/m^2 s or g/m^2 day (Siracusa, 2012).

2. Ratio of surface area and volume

Surface area of the packaging is a determining aspect for shelf life. Large surface increases the quantity of the moisture, oxygen, and other small molecules passing through the packaging. This causes many smaller pack of products to have shorter shelf life (Robertson, 2016).

3. Closure and integrity

Closure of packaging plays important role in achieving expected shelf life. Heat sealable films require application of heat to bond the films together. Some paper-based packaging materials are laminated with plastic film that improve its barrier properties and makes it possible to heat seal it (Robertson, 2016).

2.5.3 **Communication**

Packaging provides information for the consumers and packaging is a part of the product that interacts with consumers at the point of purchase (Marsh & Bugusu, 2007). Printability is important for packed bread packaging. Not only for marketing purposed, packaging is also carrying legal required information such as product identification, nutritional value, ingredient declaration, net weight, and manufacturer details. For product traceability, unique code is usually labeled in the packaging by the manufacturer that allows them to track the product along the distribution process (Marsh & Bugusu, 2007).

2.5.4 **Convenience**

Convenience is the most important aspect for the consumers. Some example of convenience features are easy to access, handle and dispose; product visibility; and reasealability (Marsh & Bugusu, 2007). Most of the bread flexible packaging has clip sealing format that allows easy opening and reasealability. For this format, packaging should be able to be sealed with a clip. This allows easy opening and resealability while maintaining the contained product quality.

2.6 Biobased and biodegradable materials

This section is dedicated as an opening before the review about biobased packaging materials. It is important to understand the different definition of biobased and biodegradable materials as well as the certifications on the materials.

Biobased and biodegradable characteristics of material are two different definitions. Biobased concept focuses on the origin of the material. Biobased materials means that the materials are derived from some amount of a renewable source (Greene, 2014a; Weber, 2000). On the other side, biodegradability is related with end-of-life distinction of the material. Biodegradable materials are defined as materials that can be broken down by microorganism into water and natural occurring gasses (carbon dioxide and methane) (Molenveld, Van Den Oever, & Bos, 2015). Biodegradable polymer is converted into biomass, CO_2, and water through thermochemical process in specific time limit and disposal environment (Greene, 2014a). Another term related to plastic polymer is bioplastics. Bioplastics are plastics that are biobased, biodegradable, or both (European Bioplastics, 2016a).

Biobased polymer can be non-biodegradable while fossil-based polymer can be fully biodegradable.

Certain certification is needed to declare the content of biobased in a material. ASTM D6866-16 (US) and EN 16785-1:2015 (EU) are standard methods that measure the biobased carbon content using radiocarbon analysis. DIN CERTCO (Germany) and Vinçotte (Belgium) are two certification bodies in Europe who give certification of biobased products. The certification is given based on the biobased carbon content in the product. The different certification labels can be seen in Figure 6.

Composting is more well-regulated compared to other path of biodegradation and non-biological degradation (Molenveld et al., 2015). Certification of compostable products is available from DIN CERTCO (DIN-Geprüft Industrial Compostable) and Vinçotte (OK compost) (see Figure 7). ‘Seedling’ logo will be given to product that pass the test according to EN 13432:2000 (packaging) and EN 14995:2006 (plastics). The logo indicates that the product will be fully degradable in industrial composting plant under temperature, moisture, and time frame (European Bioplastics, 2016c).

Figure 6 Biobased certification label from DIN CERTCO (top) and Vinçotte (bottom)

> *“The European standard EN 13432 requires at least 90% disintegration after twelve weeks, 90% biodegradation (CO_2 evolvement) in six months, and includes tests on ecotoxicity and heavy metal content. It is the standard for biodegradable packaging designed for treatment in industrial composting facilities and anaerobic digestion.” (European Bioplastics, 2016b).*

Figure 7 Logo of 'Seedling' (left), OK compost (middle), and DIN-Geprüft Industrial Compostable (right)

OK compost HOME and DIN-Geprüft Home Compostable certification (Figure 8) can be given to the product that can be composted in lower temperature, such as in the garden. It requires 90% degradation after 12 months at ambient temperature. Another biodegradable certificate from Vinçotte could be given to the product that can possibly degrade in marine, soil, and water.

Figure 8 Logo of OK compost HOME (left) and DIN-Geprüft Home Compostable

2.7 Biobased polymers

Biobased polymer can be categorized into 3 categories based on the origin and production process; polymer which is directly extracted from biomass, polymer produced by chemical synthesis from renewable biobased monomers, and polymers produced by microorganism (Molenveld et al., 2015; Weber, 2000). See Figure 9 (left) for schematic biobased classification.

2.7.1 **Polymer extracted from biomass**

Biodegradable polymers from agro-resources include polysaccharides, proteins, and lipids (Avérous, 2004). Polysaccharide-based films are commercially available as an alternative of fossil-based plastic. Protein-base films, on the other side, are still in development phase and limited by expensive process of solution casting. Many studies have been exploring packaging film from plant-based protein (rapeseed, wheat gluten, corn zein, soy protein, sorghum kafirin, oat avenin, rice brand protein, cottonseed protein, and peanut protein) and animal-based protein (whey protein, casein, albumin, keratin, collagen, and gelatin) (Gällstedt, Hedenqvist, & Ture, 2011). In this section, only polysaccharide-based polymers are discussed, i.e. starch-based material, cellulose-based material, and hemicellulose-based material.

2.7.1.1 Starch-based material

Starch is the most abundant form of polysaccharides in plant. It is renewable, relatively cheap, and may exhibit thermoplastic properties. Many attempts have been aimed to develop starch-based plastic. However, blending starch with additive is necessary due to starch lack stability on water absorption, aged-induced retrogradation, inferior mechanical properties, and poor processability. Characteristic of starch-based plastic is highly dependent on amylose/amylopectin ratio, humidity, type and content of plasticizer, processing method and final crystallinity (Vázquez, Foresti, & Cyras, 2011).

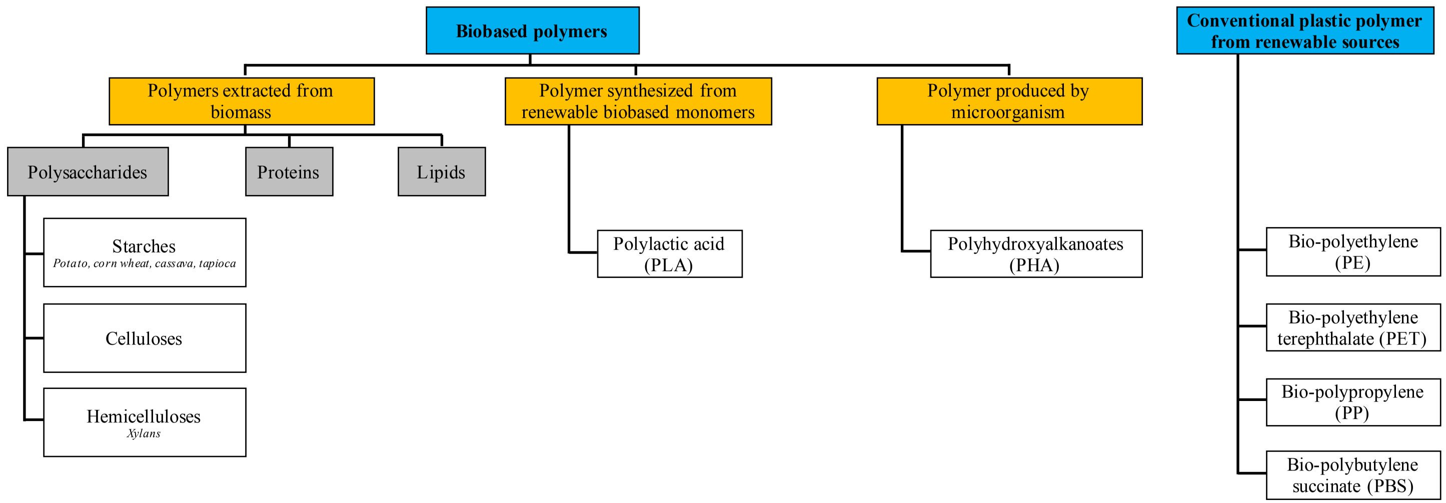


Figure 9 Classification of biobased polymers based on the origin and production process (left), modified from (Rastogi & Samyn, 2015) and conventional plastic polymer from renewable sources (right)

Starch-based polymer can be produced from potato, corn, wheat, cassava, or tapioca (Greene, 2014a). Thermoplastic starch (TPS) can be made from blend of starch, aliphatic polyester, glycerol, and water. Linear aliphatic polyester is added to starch to create compostable film, sheet, plastic bags, and liners (Vázquez et al., 2011). Not all starch-based materials can be used as food contact material. This is due to the migration of additives used in starch-based plastic into food that might exceed the limit in the regulation (Molenveld et al., 2015).

Application

The common application of starch-based plastic is as bin liner (Molenveld et al., 2015). Some plastic bags are made from starch-based plastics.

End-of-life

Starch-based polymer is compostable by its own. However, blending of starch-based materials with non-biodegradable fossil based polymer will result in non-biodegradable and non-recyclable materials (Greene, 2014a).

Commercial product in the market

Commercial starch blends are produced by Novamont Italy (Mater-Bi®), Biotec Germany (Bioplast), and Rodenburg The Netherlands (Solanyl®) (Molenveld et al., 2015).

2.7.1.2 Cellulose-based material

Cellulose constitutes a major part in paper. Another important application of cellulose is to make cellophane (film). Cellophane film has characteristic of transparent and 'dead fold'. Once the film is folded, it is irreversible. Unlike plastics, cellophane is not produced by thermoforming. It is not heat-sealable, therefore a sealant layer and/or a barrier layer is needed for its application as packaging. Cellophane can be laminated with starch-based or polylactic acid (PLA) sealant. Thin aluminium oxide layer is usually used for barrier layer while still maintaining the biodegradable properties (Molenveld et al., 2015).

Application

Cellophane is used for packaging material for confectionery and floral bouquets (Molenveld et al., 2015).

End-of-life

Cellophane is biodegradable in a diverse environment. There is no life cycle assessment (LCA) available for cellophane production. It is still debatable if production of cellophane is less harmful than many conventional plastics (Molenveld et al., 2015).

Commercial product in the market

Innovia is one of the producers of cellophane. However, in 2016 Innovia sold their Cellophane Division to Futamura Chemicals Co., Ltd (Embree, 2016). Futamura sells multilayer (containment layer, barrier layer, and sealant layer) biobased packaging material with various applications (Futamura brochure).

Development of nanocellulose

Currently, academic and industrial interest are towards developing microfibrillated cellulose (MFC) or nanofibrillated cellulose (NFC). These materials have been around since 1980s but commercial production required high amount of energy. Innventia, one of Swedish research organization is developing a reduced-energy production process of nanocellulose. This material has a potential to strengthen paper and to coat as barrier layer on food packaging (Innventia website).

2.7.1.3 Hemicellulose-based material

Xylans are the main hemicellulose in plant which is key structural component in the plant cell wall. Like other carbohydrate component, xylans can be obtained from *Gramineae* (grass and cereals), *Gymnosperms* (softwoods) and *Angiosperms* (hardwoods). Its chemical structure is characterized by having a xylopyranosyl backbone with primarily (1→4)-b-linkages (Albertsson, Edlund, & Varma, 2011). Representation of xylan structure can be seen in Figure 10.

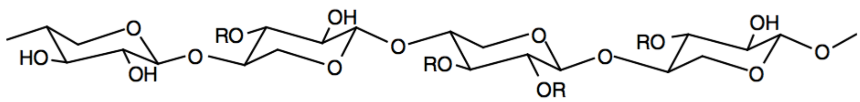


Figure 10 Structure of xylan (Albertsson et al., 2011)

Studies on oxygen permeability of xylan-based film shows good oxygen barrier properties, lower than 10 cm^3 mm/m^2 d kPa. Water vapor permeability of xylan-based films vary depending on the other components added (Mikkonen & Tenkanen, 2012). Arabinoxylan film from rye bran has a water vapor permeability 7.7 g mm/m^2 d kPa (RH 0/52%) (Sárossy, 2011) . Since polysaccharides is basically hydrophilic, it is expected to be a good barrier against grease (Mikkonen & Tenkanen, 2012).

Application

Xylans can be used for edible inner packaging for low moisture food, coating on fruit, cheese, or paper. It has comparable mechanical strength to some material for food packaging but it has low stretchability and resistance to water. The most potential application of this material would be oxygen barrier coating on food or packaging where the coated products provide mechanical support (Mikkonen & Tenkanen, 2012).

End-of-life

Xylan-based film has been tested as edible coating on fruits. It is also biodegradable (Mikkonen & Tenkanen, 2012).

Commercial product in the market

Large scale isolation of xylans is still limited even though the raw material is abundant (Mikkonen & Tenkanen, 2012). Seelution AB, a Swedish company produces xylan-based barrier coating material (Skalax®) against oxygen, grease and mineral oil for food packaging.

2.7.2 Polymer synthesized from renewable biobased monomers

2.7.2.1 Polylactic acid (PLA)

Polylactic acid or polylactide is 100% biobased polymer formed by lactic acid monomers. Since lactic acid has 2 stereoisomers, PLA can be found in the form of L-(poly(L-lactide)) or D-(poly(D-lactide), or combination of both (Södergård & Inkinen, 2011). Chemical structure of PLA can be seen in Figure 11.

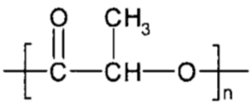


Figure 11 Chemical structure of PLA (Avérous, 2004)

Lactic acid can be produced by chemical reaction and fermentation. In the industrial production, microbial carbohydrate fermentation is more common. Carbohydrate source for the fermentation can come from whey, molasses, starch waste, beet, or sugar cane (Södergård & Inkinen, 2011).

PLA is industrial compostable and has been approved as food contact material. PLA is also transparent and breathable material (Molenveld et al., 2015). Barrier properties of PLA varies depending on the grade and processing history. Moreover, PLA can be printed with commercial printing lines without surface pre-treatment (Södergård & Inkinen, 2011).

Applications

PLA applications include tray, film, coating, fiber, and medical application (Molenveld et al., 2015; Södergård & Inkinen, 2011). It is also possible to combined PLA with paper to make paper cups or plates. PLA is used as window in bread bag due to breathable property. For liquid application, PLA might not be suitable due to its water permeable property (Molenveld et al., 2015).

End-of-life

PLA complies with the EN13432 standard for compostable products (Molenveld et al., 2015). It degrades by hydrolysis after several months exposing moisture. Temperature around 50-60 °C and high humidity will degrade PLA easily. PLA burns with blue flames through the incineration without any poisonous or corrosive gasses. PLA is also possible to be recycled by hydrolyzing with hot water (Auras, Harte, & Selke, 2004).

Commercial product in the market

NatureWorks LCC is the biggest manufacturer of PLA with their PLA trademark Ingeo™ (Molenveld et al., 2015).

Challenge

The challenge of PLA is regarding the price and its physical properties. Brittleness, poor thermal resistance, and limited gas barrier properties are three main unwanted properties of PLA for packaging application (Madhavan Nampoothiri, Nair, & John, 2010).

2.7.3 **Polymer produced by microorganism**

2.7.3.1 Polyhydroxyalkanoats (PHAs)

PHA (polyhydroxyalkanoat) is a biopolymer obtained from bacteria cell. PHA is produced as intracellular food and energy reserve for bacteria. PHA can be produced by fermentation of renewable sources. It is biodegradable and has highly biocompatibility with living tissue for medical application (Pollet & Avérous, 2011).

Industrial production of PHA is significantly lower than PLA. Around 24 companies are known to be engaged in PHA production and application. However, most of them are still in the pilot scale production. Furthermore, PHA is relatively more expensive than conventional plastics (Pollet & Avérous, 2011).

In PHA family, polyhydroxybutyrate homopolymer (PHB) is the main type of polymer. Other poly(hydroxybutyrate-co-hydroxyalkanoate) copolyesters exist. Chemical structure of PHA can be seen in Figure 12.

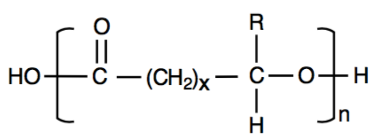


Figure 12 Chemical structure of PHA (Pollet & Avérous, 2011)

PHA is produced by bacterial biosynthesis. Recent studies aimed to produce PHA by biosynthesis in genetically modified plants. Starch, sugar, and ethanol are used for the substrates which is usually combined with various fatty acids and nutrients that depend on the bacteria strain used. More efficient technology is under development to produce cheap PHA from cheap carbon sources. In other way, synthesis PHA in genetically modified plant is an attractive route to produce cheap PHA in a large scale. A less sustainable chemical synthesis of PHA is also possible from lactones (Pollet & Avérous, 2011).

PHB has almost similar physical properties with polypropylene. It has low water vapor permeability which is very interesting for packaging application. However, it has low deformation at break because of low film toughness and unacceptable rigidity and brittleness (Pollet & Avérous, 2011). Main disadvantage of PHAs for packaging application is that they are not transparent (Molenveld et al., 2015).

<u>Application</u>

One of PHA family, PHBV poly (hydroxybutyrate-co-hydroxyvalerate), is used for flexible film application. Current PHAs application is biodegradable carrier bag and mulch film. PHAs are usually combined with Ecoflex® (fossil-based copolyester) and PLA (Molenveld et al., 2015).

<u>End-of-life</u>

PHA can degrade in soil, sludge, fresh water, seawater, and compost. Microorganism such as fungi and bacteria can use the degradation products as their carbon source. Since PHA is basically a polyester, hydrolysis can rapidly happen after disposal (Pollet & Avérous, 2011).

<u>Commercial product in the market</u>

In Europe, Biomer (Germany) is a company that produce PHB sold with Biomer® trademark. Biomer® is used for termocoating paper and carton. However, it is too crystalline to be formed as film. This information was obtained from the founder of Biomer.

2.8 Conventional plastic polymers from renewable sources

Polyethylene (PE), polyethylene tereftalate (PET), and polypropylene (PP) are originally made from natural gas or petroleum products. These plastics are possibly made from plant-based sources which then called biobased polyethylene (bio-PE), biobased polyethylene tereftalate (bio-PET), and biobased polypropylene (bio-PP). Some literatures define it as drop-in plastics (Molenveld et al., 2015). In Figure 9, it shows the position of the materials in this group compared to biobased polymer. Bio-PE and bio-PET are available commercially in the market. In this section, four different drop-in plastics are explained.

2.8.1 **Biobased polyethylene (bio-PE)**

Bio-PE can be produced from sugar cane or other agricultural materials. Ethanol from fermentation of sugar cane, corn, potato, or other agricultural products are the main ingredients for polyethylene. It is converted to ethene which then undergoes polymeric reaction resulting in polyethylene (Greene, 2014b). Bio-PE is 100% biobased. The molecular structure of ethanol, ethene, and polyethylene can be seen in Figure 13.

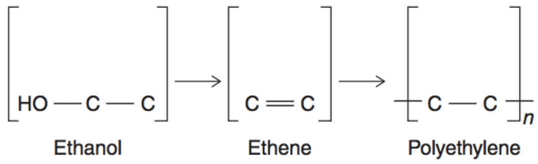


Figure 13 Molecular structure of ethanol, ethene, and polyethylene (Greene, 2014b)

Composition and mechanical properties of bio-PE are similar with conventional fossil based PE. Bio-PE can be used to produce many type of products, such as blown film, sheet, bottles, packaging products, fibers, etc. (Greene, 2014b).

<u>End-of-life and LCA analysis</u>

Bio-PE can be recycled through current PE recycling process. “Cradle to gate” LCA analysis shows that every 1000 kg bio-PE production requires 10 MJ energy and emits -2000 ton CO_2eq. This is lower than the fossil-based PE, 65 MJ and 1800 ton CO_2eq (Greene, 2014b). Another LCA analysis shows that bio-HDPE resulted -0.75 kg $CO_2eq/kg_{polyethylene}$ with approximately 65% lower non-renewable energy use (Tsiropoulos et al., 2015).

<u>Commercial product in the market</u>

Braskem and DOW Chemical are two main suppliers for bio-PE resin (Greene, 2014b). Bio-PE from Braskem is marketed with I’m Green™ polyethylene.

2.8.2 **Biobased polyethylene terephthalate (bio-PET)**

Polyethylene terephthalate (PET) is usually produced from terephthalic acid and mono ethylene glycol (MEG) (see Figure 14 for the molecular structure). MEG can be produces from biobased material such as sugarcane, corn, and soy. This means that PET only can be 30% biobased since 70% of its composition (terephthalic acid) is coming from fossil-based source (Greene, 2014b). However, 100% biobased Coca-Cola PET bottle prototype has been possibly manufactured and was shown in 2015 World Expo in Milan. Paraxylene which is raw material to produce purified terephthalic acid (PTA) and dimethyl terephthalate (DMT) was successfully produced by Virent, Inc. (US company) from beet. This is possible because of production of BioFormPX® (paraxylene) from sugar beet (Plastics Technology, 2015). The properties of PET from biobased material should be the same to fossil-based PET.

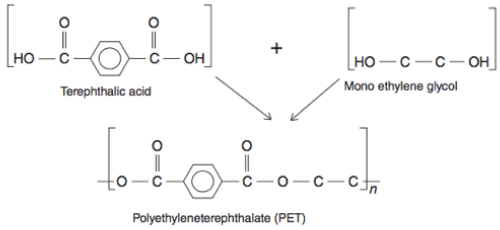


Figure 14 Molecular structures of terephthalic acid, MEG, and PET (Greene, 2014b)

End-of-life and LCA analysis

One of the advantage of drop-in plastics like bio-PET is that it does not give effect to the current PET recycling process. LCA analysis shows that bio-PET (31% MEG per repeating unit of PET) production resulted 3-11% saving of non-renewable energy usage but limited saving of greenhouse gases (GHG) emission (Tsiropoulos et al., 2015). This is because only MEG is biobased which account least part in the PET.

Commercial product in the market

Indorama is the biggest supplier of bio-PET under RAMAPET® brand (Molenveld et al., 2015).

2.8.3 Biobased polypropylene (bio-PP)

Bio-PP can be produces from corn, biomass, bio diesel, and vegetable oil. The two main technologies to produce polypropylene are biochemical and thermo chemical process. In the biochemical process, corn, sugar cane, beet or other source of raw material are fermented to produce ethanol. The ethanol then undergoes the process to convert them in to ethylene and butane. Polypropylene is obtained from metathesis of ethylene and butane (Venkataraman, 2012).

In the thermo chemical process, biomass is used to produce syngas through gasification. The syngas undergoes further process to produce methanol. Methanol is then used for polypropylene synthesis. There is a development of combination of biochemical process and thermo chemical process to produce bio-PP (Venkataraman, 2012).

The properties of PP made from biobased source is similar with fossil-based PP. The polymer can be used for different kind of product applications.

Commercial product in the market

Braskem and DOW Chemical are two companies that are continuously investigating technology route to produce bio-PP (Venkataraman, 2012).

2.8.4 **Biobased polybutylene succinate (bio-PBS)**

Polybutylene succinate (PBS) is a biodegradable aliphatic polyester which originally produced from petrochemical origin. PBS is synthesized from 1,4-butanediol (1,4 BDO) and succinic acid (Greene, 2014a). Chemical structure of PBS can be seen in Figure 15. Biobased succinic acid is possibly produced in large scale, therefore 50% biobased PBS can be obtained. Moreover, development of biobased 1,4 BDO will later make it possible to produce 100% biobased PBS (Molenveld et al., 2015).

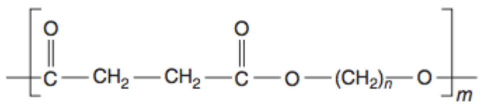


Figure 15 Chemical structure of PBS (Greene, 2014a)

PBS has promising properties such as biodegradability, melt processability, and thermal and chemical resistance. However, due to properties like softness and low gas barrier properties, this material need to be improved for wider application (Sinha Ray & Bousmina, 2005).

Application

PBS can be used as biodegradable sheets, film, bottles, and molded products (Greene, 2014a). PBS is applied in carrier bags in combination with other biopolymers such as starch blends (Molenveld et al., 2015).

End-of-life

PBS is biodegradable. PBS degrading microorganism are widely distributed in the environment (Tokiwa, Calabia, Ugwu, & Aiba, 2009).

Commercial product in the market

Reverdia, a joint venture of DSM (Netherlands) and Roquette (France) is one of bio succinic acid (Biosuccinium®) producer who is actively promoting its application. Biobased polybutylene succinate, BioPBS™, is planned to be commercially produced by PTT MCC Biochem Thailand (joint venture between PTT Public Company Limited and Mitsubishi Chemical Corporation) in 2017.

2.9 Summary of biobased polymers and conventional plastic polymers from renewable sources

There are many biobased materials to be developed and some of them have already available in the market. Figure 16 shows the summary of materials that have been discussed in the previous chapter and the mapping based on its renewable content and biodegradability.

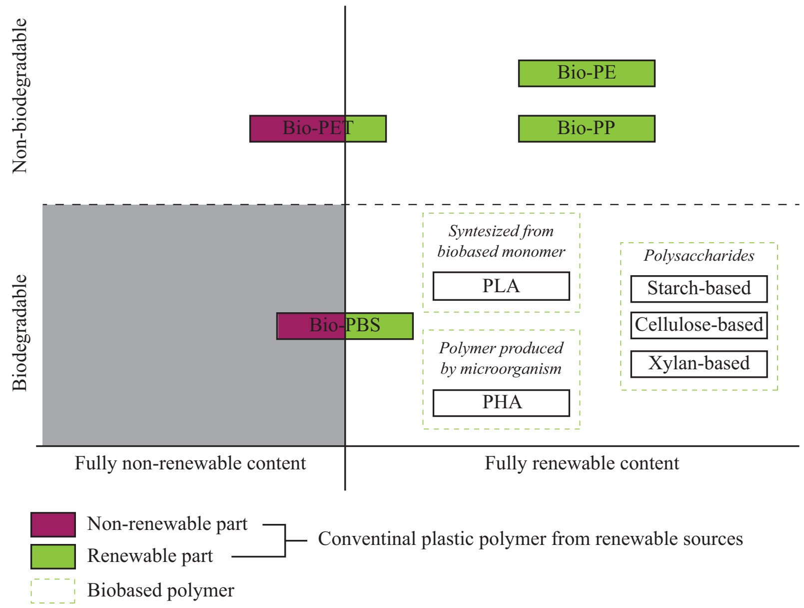


Figure 16 Grouping of the biobased materials

Biobased material usually combined with other materials to improve its properties for certain application. The combination could result as blends, composite, or multi-layer materials with improved properties. Blending polymer is also an attempt to decrease cost and tailoring degradation rates (Tokiwa et al., 2009). Many studies had been conducted for starch-based polymer blends. Properties of the final blends is highly depends on the property of individual component (Vázquez et al., 2011).

Comparison of water vapor permeability and oxygen transmission rate of some biobased and fossil-based packaging film can be seen in Table 3. However, since the measurement of these two characteristics of flexible film is usually done under different conditions, it is not easy to compare one another. Furthermore, in many published literature, details of the polymer materials are not stated which made it impossible to replicate the experiment (Robertson, 2016).

Table 3 Comparison between some biobased packaging films and fossil-based packaging films

Film	Based	Water vapor permeability	Oxygen transmission rate	Source
LDPE 25 μm	Petroleum	16-31 g/m^2 day; 38 °C, 90% RH	7750 ml/m^2 atm 25 °C, 0% RH	(Lee & Sun, 2011)
PP 25 μm	Petroleum	6 g/m^2 day; 38 °C, 90% RH	1550-2480 ml/m^2 atm 25 °C, 0% RH	(Lee & Sun, 2011)
PET 25 μm	Petroleum	16-23 g/m^2 day; 38 °C, 90% RH	50-90 ml/m^2 atm 25 °C, 0% RH	(Lee & Sun, 2011)
PBS 144.67 μm	Petroleum	330* g/m^2 day; 25 °C *Bionolle from Showa Denco	2866,66 cm^3/m^2 day bar 23 °C, 0% RH	(Shogren, 1997; Siracusa, Lotti, Munari, & Dalla Rosa, 2015)
PHB-6 (6% valerate)	Bio	13 g/m^2 day; 25 °C	-	(Shogren, 1997)
PLA	Bio	172 g/m^2 day; 25 °C	1.94 x 10^{-18} m^3 m/m^2 s Pa	(Ducruet et al., 2011; Shogren, 1997)
Xylan-based film (Arabinoxylan)	Bio	2.6–7.7 g mm/m^2 d kPa (RH 0/50–52%)	<10 cm^3 $\mu m/m^2$ day kPa	(Mikkonen & Tenkanen, 2012)
Starch-based (potato)	Bio	6.25x10^{-10} g/m s Pa	-	(Rejak, Wójtowicz, Oniszczuk, Niemczuk,

				& Nowacka, 2014)
Cellulose acetate	Bio	2920 g/m^2 day; 25 °C	3.7x10^{-12} cm^3 m/m^2 s Pa	(Paunonen, 2013; Shogren, 1997)
Paper/bio-PET laminate	Bio	17.4 g/m^2 day 25 °C, 75% RH	29.4 cm^3/m^2 day 23 °C, 75% RH	(Persson, 2016)

3 Methodology

This chapter aims to explain the research approach in this thesis and the experimental design. The experimental design focuses on the evaluation of biobased packaging materials and the packed buns. The methods of evaluation for 5 selected packaging materials with reference and methods of evaluation for packed hotdog buns and hamburger buns are explained in detail.

3.1 Overall research approach

Overall research approach has been developed in order to answer the research question *'how does biobased packaging material affect the quality of fully-baked frozen bread in comparison to reference packaging?'*. The strategy in this study can be categorized into three parts; biobased packaging material search and literature study, development of experimental design, and quantitative primary research.

3.1.1 **Biobased material search and literature study**

Biobased packaging material search and literature study on biobased packaging materials were done in parallel. Biobased material search focused to list down biobased packaging materials that are available in the market and their characteristics. Whereas, literature study aimed to understand about different sources, applications, and end-of-life characteristics of biobased packaging materials.

Biobased material search was narrowed down to the following criteria:

1. Focus only on biobased packaging materials in the form of film or bag.
2. Limiting the supplier of biobased packaging material in Europe.
3. Packaging materials that are available for industrial production or will be available soon.
4. Taking into consideration partially biobased packaging materials and non-transparent materials.

Selection of biobased packaging materials used in the experiment was done by evaluating the material list and discussion with Lantmännen. Packaging materials that are available in the market and represent different type of biobased materials were chosen for the experiment.

3.1.2 **Development of experimental design**

Plant visit to Lantmännen Unibake, Örebro, Sweden, was arranged in order to understand the manufacturing and packing process of fully-baked frozen bread. Process parameters after packing of the bread was also identified, including freezing time, freezing temperature, storage and thawing conditions. Moreover, quality parameters were discussed with the experts from the company.

Bread type was suggested from the company. The chosen bread i.e. hotdog buns and burger buns, represent classic and premium category. Characteristics of these two buns are also different because of different formulation and processing in the production. Polyethylene (PE) is the current packaging of these buns.

Experimental design was developed to mimic the parameters at all stages; bread packing, freezing, minimum frozen storage, distribution, and thawing condition. The design of the experiment is discussed in detail in the method part of primary research (*Section 3.2.2.1*).

3.1.3 **Quantitative primary research**

Primary research was aimed to evaluate the performance of selected biobased packaging materials for fully-baked frozen bread packaging application. The research focused on both the packaging materials and the packed bread. Packaging material barrier properties and quality changes of the bread along shelf life were compared. From the primary research, it is expected to better understand which biobased packaging materials that provide sufficient protection for the bread during shelf life in comparison with reference. The current packaging material of the bread was included in the experiment as a reference. A triangle test on the buns was performed by panelists to evaluate any differences between the buns packed in bio-PE and buns packed in the reference PE.

3.2 Primary Research

3.2.1 **Material**

3.2.1.1 The buns

All buns for the experiment were supplied from Lantmännen Unibake, Örebro, Sweden. The buns were produced in industrial scale. Only buns from the same batch were used to minimize the variability between buns. Two different type of buns were used; hotdog buns and hamburger buns. The weight of hotdog buns and hamburger buns were in average 27 g and 70 g respectively.

3.2.1.2 Packaging materials

Total of six different packaging materials were supplied from different packaging suppliers. The detail of five selected packaging materials and reference (PE) can be seen in Table 4. Some packaging materials were received in film form (packaging material number 3-6). Vacuum heat sealer Multivac model A 300/16 was used to make the bag from these materials. All the materials were easy to seal except Biodolomer®. The seal strength for Biodolomer® was weak, therefore it required to be treated gentler after sealing. All packaging bags were checked for its complete sealing prior to the experiment.

Table 4 Packaging materials used in the experiment

No	Packaging material	Thickness (μm)	Bag size (mm)	Supplied by
1.	Polyethylene (PE) – reference for hotdog buns	35	200 x 360 + 40	Lantmänen Unibake
	Polyethylene (PE) – reference for hamburger buns	40	200 x 360 + 50	Lantmänen Unibake
2.	Biobased polyethylene (bio-PE)	35	200 x 420	Amerplast Ltd., Finland
3.	Biodolomer® (compound of biodegradable co-polyester, calcium carbonate, and polylactic acid (PLA))	17	200 x 360	GAIA BioMaterials AB, Sweden
4.	Polylactic acid (PLA)	25	200 x 360	Sidaplax, Belgium

5.	Polybutylene succinate-co-adipate (PBSA)	35	200 x 360	Reverdia, Netherlands*
6.	Paper laminated with biobased polyethylene terephthalate (paper/bio-PET)	22.2 PET layer	200 x 360	Flextrus AB, Sweden

*PBSA grade was supplied from PTT-MCC Biochem Company Ltd., Thailand.

3.2.2 **Method**

3.2.2.1 Experimental design

The packing of the buns was done at Lantmännen Unibake, Örebro. Fresh half-cut buns from the production were repacked manually from the commercial packaging to packaging materials used for the experiment. For hotdog bun, ten buns were packed in one bag while for hamburger bun, four buns were packed in one bag. The buns were frozen at -18 °C for at least 72 hours and transported with frozen truck to Lund for evaluation. The design of the experiment can be seen in Figure 17.

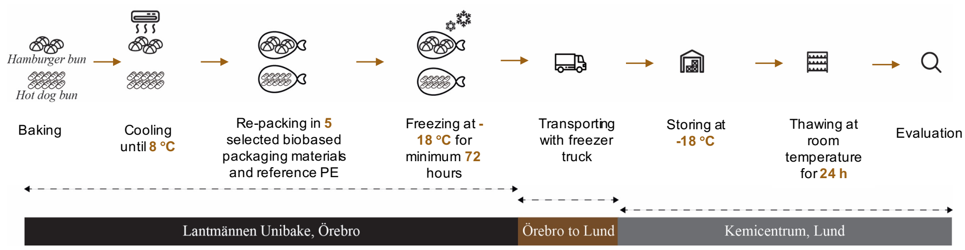


Figure 17 Illustration of experimental design

3.2.2.2 Storage and sample preparation

All packed bun samples were stored at -18 °C in the freezer. Prior to testing, the samples were thawed for 24 hours. All bag samples of each packaging materials were thawed at the same time for evaluation day 1, day 3, and day 7 after thawing. Data logger was used to record storage condition i.e. temperature and humidity.

3.2.2.3 Replication

Three bag replicate for every observation day (day 1, day 3, and day 7 after thawing) was applied for each packaging material. Total of 9 bags of each packaging material were evaluated, except for paper laminated with bio-PET (only 3 bags). Detailed illustration of sample and measurement replications can be seen in Figure 18.

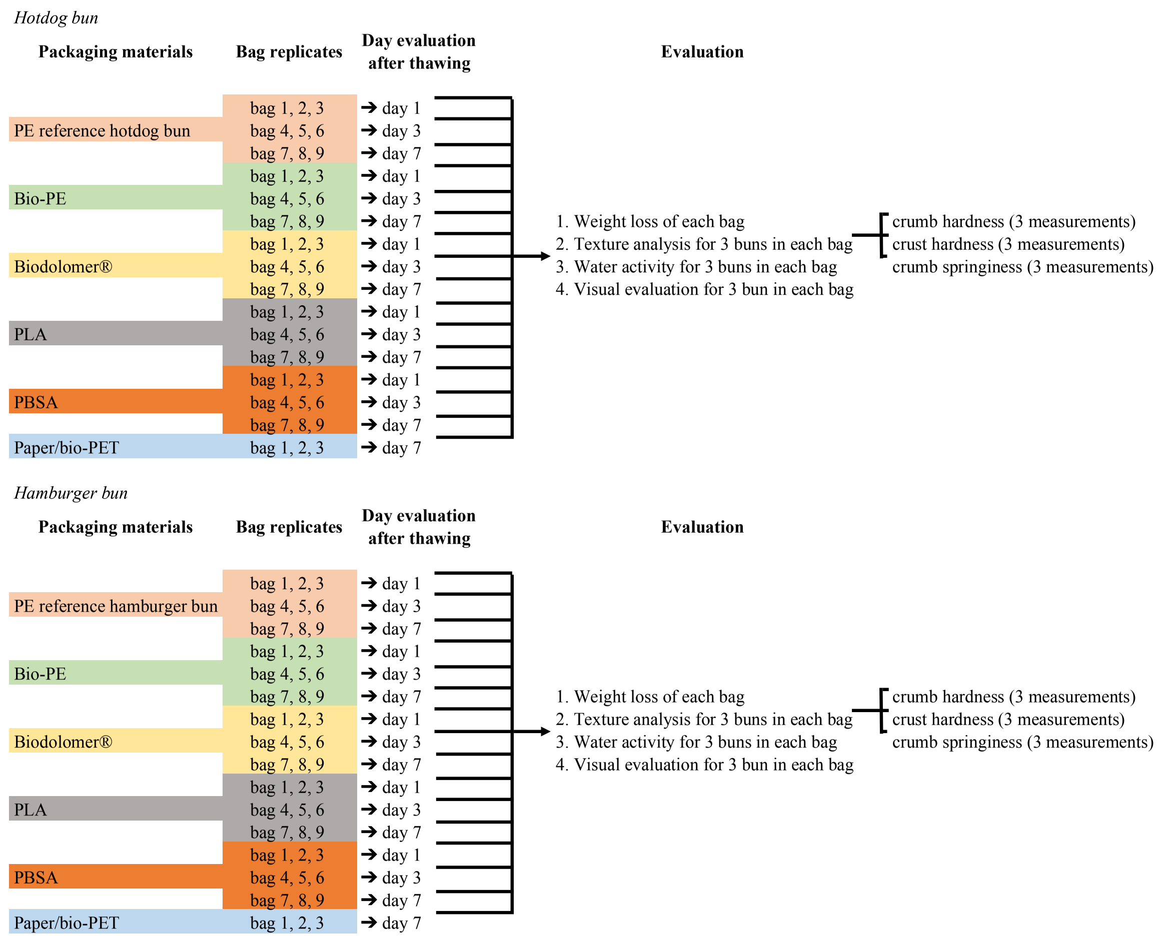


Figure 18 Illustration of sample and measurement replications in the experiment

3.2.2.4 Evaluation of packaging material

3.2.2.4.1 Water Vapor Transmission Rate (WVTR)

WVTR was measured following the method used by (Hu, Topolkaraev, Hiltner, & Baer, 2001) which is a modification of ASTM E 96-95 wet cup method. Window of 25 cm^2 packaging film was glued on aluminum foil. Epoxy glue was used for this experiment. The packaging film glued on aluminum foil was then covered in a plastic petri dish and glued. Using pipet, 30 mL of distilled water was filled into the petri dish by making small hole on the aluminum foil. The hole was then covered with aluminum foil and glued. The schematic representation of the test can be seen in Figure 19.

Three replicates of petri dishes were placed in the fume hood with air circulation around 0.5 m/s. The weight of the petri dish was measured every day for 4 days. Temperature and humidity of the fume hood were recorded by temperature-humidity data logger. WVTR is calculated with equation (1).

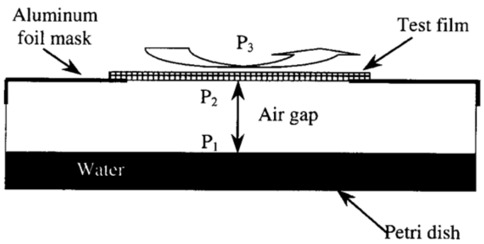


Figure 19 Schematic representation of the wet cup test (Hu et. al., 2000)

$$WVTR = \frac{mass\ H_2O\ lost}{area\ x\ time} = \frac{flux}{area} = \frac{gram}{m^2\ day} \qquad (1)$$

3.2.2.4.2 Puncture resistance

Puncture resistance is an important characteristics of flexible packaging material, especially laminates. It shows the resistance of packaging against damage by penetration. Understanding puncture resistance allows prevention of damage caused penetration that can lead to loss of barrier properties, packaging integrity and product quality. Puncture resistance is typically expressed as the load at puncture or as the energy (Lange, Mokdad, & Wysery, 2002). In this study, puncture resistance test of the packaging was done with Perten TexVol TVT-300XP instrument and TexCalc Software Version 4.0.4.67. The test was done for the packaging before it was used. The parameters put on the software for the testing can be seen in Table 5.

3.2.2.5 Quality evaluation of packed buns

3.2.2.5.1 Appearance

This evaluation was focused on the appearance of the buns (crust and crumb). The edge part of the buns was evaluated for its dryness visually. Pictures were taken in every observation.

3.2.2.5.2 Weight loss

Buns in the bag were weighed before thawing and after thawing (day 1, day 3, and day 7). Weight loss was calculated by deducting the weight of bag in the observation with the initial weight of the bag before thawing.

3.2.2.5.3 Water activity

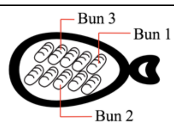
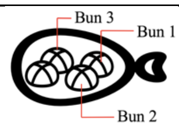
Water activity for crumb was measured by water activity meter AquaLab® Series 3 TE. The crumb was taken from the middle part of the buns. The measurement was done for 3 different buns in every bag.

3.2.2.5.4 Texture analysis

Texture analysis was performed with Perten TexVol TVT-300XP Texture Analyzer and TexCalc Software Version 4.0.4.67. Crust firmness was measured with a penetration test while crumb hardness and crumb springiness were measured by compression test. Test parameters for crumb hardness and springiness were chosen based on Perten TVT Method 01-04.03. This method is similar to AACC 74-09.01 Standard Method with two parameters obtained (hardness and springiness) in single compression. The parameters of the test can be seen in Table 5.

Table 5 Parameters in the texture analyzer

Analysis	**Hotdog bun and hamburger bun crust firmness**	**Hotdog bun crumb firmness and springiness**	**Hamburger bun crumb firmness and springiness**	**Packaging puncture resistance**
Standard procedure	American Institute of Baking (AIB) with modification	Perten TVT Method 01-04.03 with modification	Perten TVT Method 01-04.03 with modification	-
Load cell	5 kg	5 kg	5 kg	5 kg
Probe diameter	5 mm	25 mm	36 mm	needle

Type of test	Single cycle compression	Hold until time compression	Hold until time compression	Single cycle compression
Compression (mm)	5	7	7	8
Custom force distance (cm)	-	6.25	6.25	-
Hold time (s)	-	32	32	-
Initial speed (mm/s)	2	1	1	1
Test speed (mm/s)	1.7	1.7	1.7	1
Number of test	3 points each bun	3 points each bun	3 points each bun	3 point each bag
Sampling of the buns				
Achieved data	Crust firmness (g)	Crumb firmness (g) Crumb springiness (%)	Crumb firmness (g) Crumb springiness (%)	Maximum force to puncture the material (g)

3.2.2.6 Sensory analysis

Triangle test method (ASTM E1885 – 04) was used to identify if the hotdog buns packed with bio-PE was the same with hotdog buns packed with reference packaging in the last day shelf-life (day 7 after thawing). Twenty panelists were used in the test. All panelists were familiar with the test (the format, the task, and the procedure). The sensitivity of the test is a function of three values α-risk, β-risk, and maximum allowable proportion of distinguisher (p_d) which is listed in Table 6.

Table 6 Sensitivity of triangle test

	20 panelist
α	0.10
β	0.05
p_d	50%
Minimum number of correct respond needed for significance	10

Every panelist evaluated a single triad of whole piece of hotdog bun coded with a 3-digit different number on each bun. This was done two times (1 replicate). High degree of confidence in result can be obtained from 20 experienced panelists with a single replication (Stone & Sidel, 2004). Two samples were similar and one sample was different. There were six sequences of products that were distributed at random among the panelist.

The panelists were asked to touch the crust of the bun, to open the bun and touch the crumb, and to taste the bun. The panelists were then asked to name the sample that differed from the other two. Appearance of the bun was not considered in differentiating the bun. The evaluation form can be seen in Appendix 5.

3.2.2.7 Statistical analysis

The data collected was analyzed with Analysis of Variance (ANOVA) with 95% confident interval using IBM® SPSS Statistics® Version 24. For the result of packaging evaluations, a Tukey HSD (honestly significant differences) test was followed when there was a significant difference between the samples. For the result of bun evaluations, a Dunnet's test was performed when there was a significant difference between the samples. Dunnet's test was used to compare each treatment (different packaging materials) with control (reference packaging).

4 Results and discussion

This chapter presents the result in 2 parts. The first part explains the result of evaluation on 5 selected packaging materials characteristics and reference (PE). The second part explains the findings from the evaluation on packed buns (hotdog buns and hamburger buns) in different biobased packaging materials in comparison with buns in the reference packaging. Discussion are based on the summary of the result.

4.1 Evaluation on packaging materials

The first objective of this study is *to identify the characteristics of different biobased packaging materials (film) that exist in the market*. Five different biobased packaging materials that are available in the market were chosen to be compared with reference (PE). This first part of result presents the different characteristics of the materials i.e. transparency characteristic, biobased content, and biodegradability. The result of barrier properties (water vapour transmission rate) and material durability (puncture resistant) are presented later in the same section.

Both PE reference for hotdog buns and hamburger buns are transparent and allow printing. Characteristic of clarity is one of the advantage of HDPE film (Akelah, 2013). Bio-PE, PLA, and PBSA are also transparent, providing good visibility of the product. In contrast, Biodolomer® is less transparent which affect the visibility of the packed buns. Different from other materials, paper/bio-PET is opaque which does not allow the visibility of the packed buns. Product visibility is one of the convenience features that is important for the consumers (Marsh & Bugusu, 2007). Therefore, less visibility of the product might affect how consumer perceive the overall product. The transparency of the packaging on text and packed buns in different tested packaging materials can be seen in Table 7.

All tested biobased packaging materials are flexible and were easy to be seal by clip except paper/bio-PET. This because paper/bio-PET is quite stiff which made it difficult to seal it with clip. In this kind of sealing application, paper/bio-PET does not perform the best. This material might be better for other application which requires complete heat sealing in every edge. This material was developed for frozen pizza packages (Persson, 2016).

Table 7 The characteristics of packaging materials used

Packaging material	Picture on text	Picture of packed buns	Biobased content*	Biodegra-dability
PE reference hotdog buns			0%	Non-bio-degradable
PE reference hamburger buns			0%	Non-bio-degradable
Bio-PE			50-85%	Non-bio-degradable
Biodolomer ®			50-85%	Home compostable
PLA			>85%	Industrial compostable
PBSA			20-50%	Home compostable
Paper/bio-PET			50-85%	PET part is non-bio-degradable

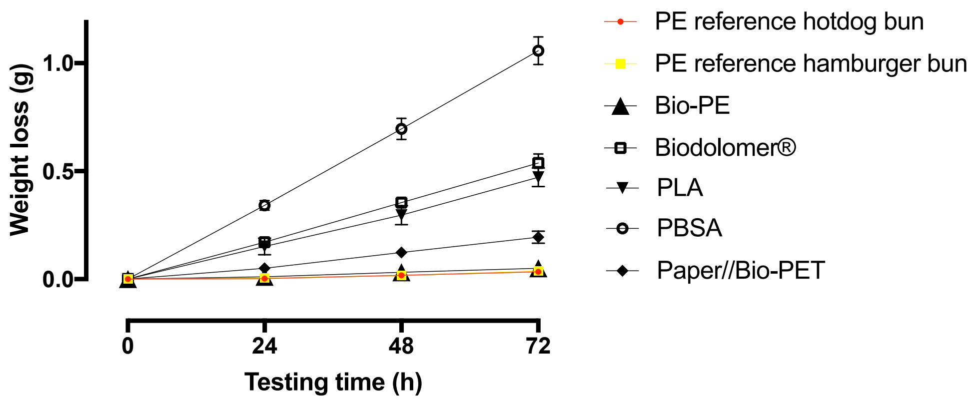
**Grouping of biobased content is based on DIN CERTCO certification. The information about exact percentage of biobased content of each materials can be seen in Appendix 1.*

Comparing the biobased content, PLA has the highest biobased content (>85% biobased content). Bio-PE, Biodolomer®, and paper/bio-PET have lower biobased content which is between 50-85%. From all the tested biobased packaging materials, PBSA has the lowest biobased content since only part of its precursor is biobased, i.e. succinic acid.

In term of biodegradability, Biodolomer® and PBSA is home compostable. Home compostable product shall be able to decompose in lower temperature (garden condition) or shall be 90% degraded after a year at ambient temperature (based on Vinçotte and DIN CERTO home compostable certification). PLA, even it has the highest biobased content, requires an industrial composting facility. PLA requires temperature around 50-60 °C and high humidity to be easily degraded (Auras et al., 2004). On the other hand, bio-PE is non-biodegradable. It can be recycled with petroleum-based PE in current PE recycling process (Tsiropoulos et al., 2015). Biodegradability is an interesting end-of-life characteristic of biobased packaging materials, however many other aspects need to be considered such as availability of recycling/composting facility and risk of plastic litter on the ground.

4.1.1 **Water Vapor Transmission Rate (WVTR)**

WVTR was measured to compare the barrier properties of the tested biobased packaging materials. The wet cup method modified by (Hu et al., 2001) was developed for highly permeable film. Therefore, it might not the best method to quantify the WVTR of PE film since PE has low permeability to water vapor (Akelah, 2013). The result of WVTR can show the comparison of water vapor barrier property of selected biobased packaging materials with PE film in ambient condition (19.4±0.3 °C and RH 31.4±4.8%).



Values represent the mean of three replicate samples with standard error of means.

Figure 20 Weight loss as a function of time in the wet cup test.

Figure 20 shows the weight loss of petri dish over 72 hours of different biobased packaging materials and PE reference. Minimum weight loss of the petri dish was observed on both PE reference and bio-PE which is shown by overlapping line in the graph. Petri dish with PBSA had the highest rate of water loss over testing time.

By calculating the flux from regression linear of the weight loss graph per unit time, WVTR was obtained with Formula (1) explained in *Section 3.2.2.4.1.* The result in Table 8 shows that bio-PE WVTR value was not significantly different compared to both PE reference. Both PE references and bio-PE had WVTR values around 3.96-6.37 g/m^{2}day. Paper/bio-PET had a higher WVTR value compared to both PE reference, 25.09 g/m^{2}day. Other sample biobased packaging materials had much higher WVTR values with PBSA had the highest WVTR value of 140.12 g/m^{2}day. This high water vapor barrier properties is one of the drawbacks of biobased packaging material characteristics for food application which can affect the moisture gain/loss of food product (Holm, 2009).

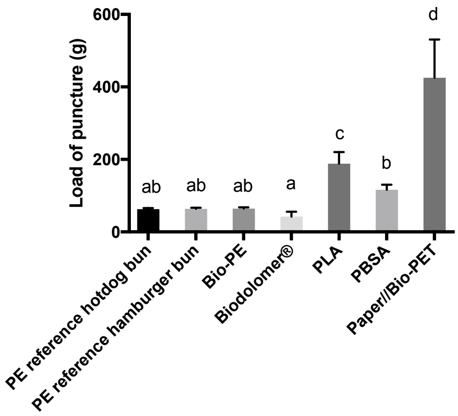
Table 8 WVTR of packaging materials

Packaging material	**Thickness (µm)**	**WVTR (g/m^2 day)**	**Standard Deviation (g/m^2 day)**
PE reference for hotdog buns	35	3.96a	0.28
PE reference for hamburger buns	40	4.41a	0.76
Bio-PE	35	6.37a	2.00
Biodolomer®	17	71.16c	5.47
PLA	25	61.70c	7.15
PBSA	35	140.12d	8.88
Paper/Bio-PET	22.2 PET layer	25.09b	3.34

Experiment was done in ambient condition (19.4±0.3 °C, RH 31.4±4.8%). Values followed by different letter (a-c) were significantly different at p≤0.05, according to Tukey's test.

4.1.2 **Puncture resistance**

Puncture resistance is a characteristic of flexible packaging materials that is related with packaging durability against impact from external penetration (Lange et al., 2002). The results in Figure 21 shows that bio-PE, Biodolomer®, and PBSA had similar load of puncture compared to both PE references. On the other side, PLA and paper/bio-PET had a significant higher load compared to the reference. This shows that PLA and paper/bio-PET had higher resistance to damage by penetration.



Values followed by different letter (a-c) were significantly different at $p \leq 0.05$, according to Tukey's test.

Figure 21 Puncture resistance of biobased packaging materials and PE reference.

Higher value in load of puncture means that the materials could resist more penetration impact from outside, for example during handling. The damage caused by penetration might lead to barrier properties and package integrity which could affect product quality (Lange et al., 2002). The packaging should have minimum load of puncture as the reference PE. Looking only from this aspect, all selected biobased packaging material had similar or higher performance compared to PE reference.

Summary

Bio-PE film had close characteristics to PE reference in term of transparency, WVTR, and puncture resistance. Other selected biobased packaging materials had similar or even higher puncture resistance but had significantly higher WVTR compared to PE reference. This might be the key difference that could affect the product quality. The summary of the characteristics of biobased packaging material can be seen in Table 9.

Table 9 Biobased packaging material characteristics in comparison with PE reference

Packaging material	Transpa-rent film	Biobased content	Bio-degradable	WVTR	Puncture resistance
Bio-PE	Yes	50-85%	No	⇔	⇔
Biodolomer®	No	50-85%	Yes	⇑⇑	⇔
PLA	Yes	>85%	Yes	⇑⇑	⇑⇑
PBSA	Yes	20-50%	Yes	⇑⇑⇑	⇔
Paper/bio-PET	No	50-85%	No	⇑	⇑⇑

⇑ means the value is significantly higher than PE reference; ⇔ means the value is not significantly different than PE reference; green color means favorable characteristic; red color means not favorable characteristic.

4.2 Evaluation on packed buns

The second purpose of this thesis is *to evaluate how biobased packaging material would maintain the quality of the hotdog buns and hamburger buns for 7-day shelf life after thawing in comparison with reference PE bag*. In this section, quality changes of hotdog buns and hamburger buns packed in different biobased packaging materials are presented. The evaluation of quality changes focused on weight loss of packed buns, textural properties of the buns, and water activity as well as changes in appearance of the buns during 7-day shelf life after thawing.

4.2.1 **Weight loss of packed buns**

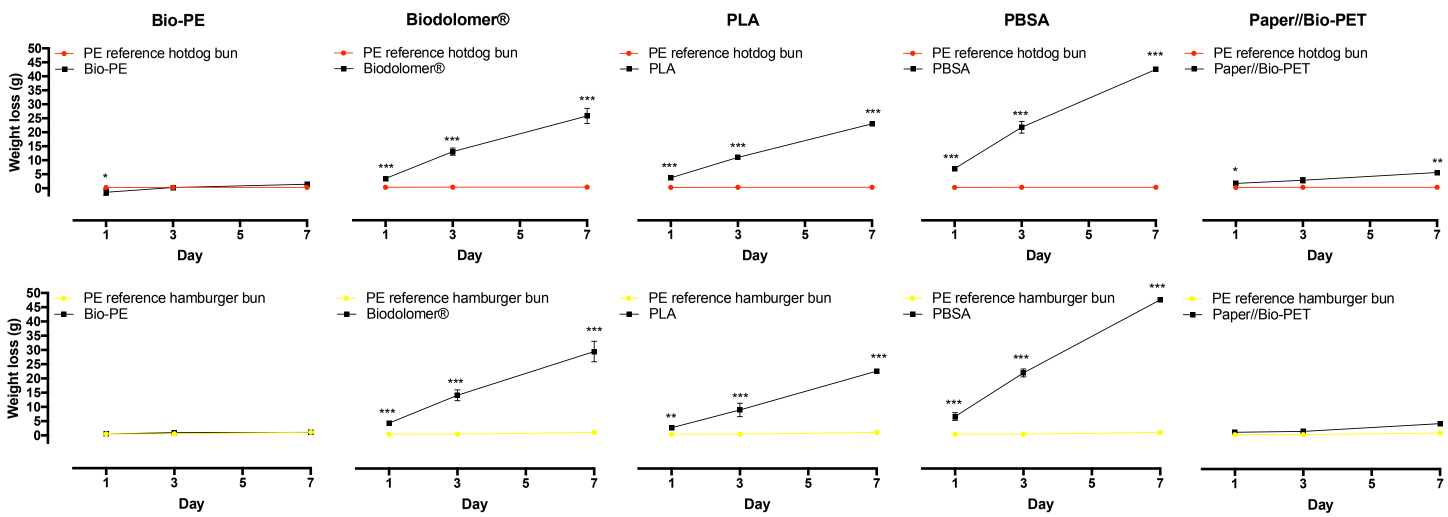
Weight loss is related with the moisture movement from the buns. Moisture movement from the product is affected by packaging material especially water permeability properties (Cauvain & Young, 2009c; Cauvain & Young, 2010). From the result in Figure 22, it shows that bio-PE have similar performance in limiting the weight loss compared to PE reference in both hotdog buns and hamburger buns along 7-day shelf life after thawing.

For hamburger buns, paper/bio-PET could maintain the buns weight loss along 7-day storage. Hamburger buns packed in paper/bio-PET showed non-significant weight loss along shelf life. This was not the case for hotdog buns because in day 7 the weight loss was significantly different from buns packed in PE reference.

In contrast, buns packed with Biodolomer®, PLA, and PBSA had significant weight loss compared to buns packed in PE reference, even in the first day after thawing. The trend of weight loss in these packaging materials was increasing along 7-day shelf life.

4.2.2 **Textural change of the buns**

The textural changes of buns in different packaging materials were evaluated by measuring the bun's textural properties. Texture analysis was performed to measure crust hardness, crumb hardness, and crumb springiness. These parameters were selected because both firmness and springiness are two major textural properties to measure the freshness of bread (Cauvain, 2017a). Three buns in each bag was evaluated. Bun samples in the same packaging were compared to observe the variability due to the position of the bun in the bag. The bun sample further from the opening was then chosen to compare to the effect of different packaging materials to the packed buns.



Storage condition: 19.7±0.5 ∘C, RH 35.2±5.9%; *=significant p≤0.05; **=significant p≤0.01; ***=significant p≤0.001.

Figure 22 Weight loss of buns in different biobased packaging materials

4.2.2.1 Variability of the buns in the bag

Three different sample buns were evaluated for texture analysis in each packaging bag. From the data, it shows that position of bun in the bag affected most of the textural parameter (see Table 10 and 11). Star in the table means a significant different textural parameter values of buns sample taken from 3 positions in the bag. Crust hardness and crumb hardness were two parameters that showed mostly significant different values depending on the position of the bun sample.

Table 10 ANOVA of textural parameters of 3 sample hotdog buns

	Crust hardness			Crumb hardness			Crumb springiness		
	Day 1	Day 3	Day 7	Day 1	Day 3	Day 7	Day 1	Day 3	Day 7
PE refernce hotdog bun	ns	*	*	ns	ns	*	ns	ns	**
Bio-PE	ns	ns	*	ns	***	**	ns	**	ns
Biodolomer	***	***	***	**	**	***	ns	ns	**
PLA	***	***	***	**	**	***	**	**	*
PBSA	***	***	***	**	**	NA	ns	**	NA
Paper//PET	-	-	ns	-		ns	-	-	ns

Table 11 ANOVA of textural parameters of 3 sample hamburger buns

	Crust hardness			Crumb hardness			Crumb springiness		
	Day 1	Day 3	Day 7	Day 1	Day 3	Day 7	Day 1	Day 3	Day 7
PE refernce hotdog bun	*	*	ns	ns	***	*	ns	ns	ns
Bio-PE	*	**	***	ns	**	*	ns	ns	*
Biodolomer	***	***	***	***	**	**	***	ns	ns
PLA	***	***	***	**	ns	ns	ns	ns	ns
PBSA	***	***	***	**	**	ns	***	ns	***
Paper//PET	-	-	ns	-	-	***	-	-	***

NA = not tested because the value exceeded the instrument maximum measured value; ns = not significant p>0.05; *= significant p<0.05; **= significant p<0.01; ***= significant p<0.001; star means significant different value of bun samples taken from 3 different positions in a bag.

Buns in the package were stacked in 2 layers in the bag (see illustration in Figure 23). Sample bun 2 which was in the middle of the lower row had a lower crust hardness and crumb hardness. In this position, the buns had limited exposure to the atmosphere inside the bag because it was in contact with other buns from its sides and from the higher stack.

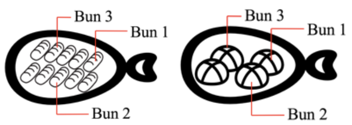


Figure 23 Illustration of bun samples position in the package

On the other side, sample bun 1 and bun 3 had higher crust hardness and crumb hardness. This happened because the surface of the buns in the upper stack were in direct contact to the atmosphere inside the bag.

Furthermore, sample bun 1 (near the opening) had higher hardness value indicating imperfect clip sealing. It showed that clip sealing still allows some leakage that increased the moisture movement of buns near the opening. Comparison of crust hardness, crumb hardness, and crumb springiness of bun 1, bun 2, and bun 3 sample of hotdog buns and hamburger buns can be seen in Appendix 2 and 3.

For the following data analysis on textural properties, only data from bun 3 (the furthest bun from opening) was considered to minimize the effect of imperfect clip sealing.

4.2.2.2 Textural properties of buns in different biobased packaging materials

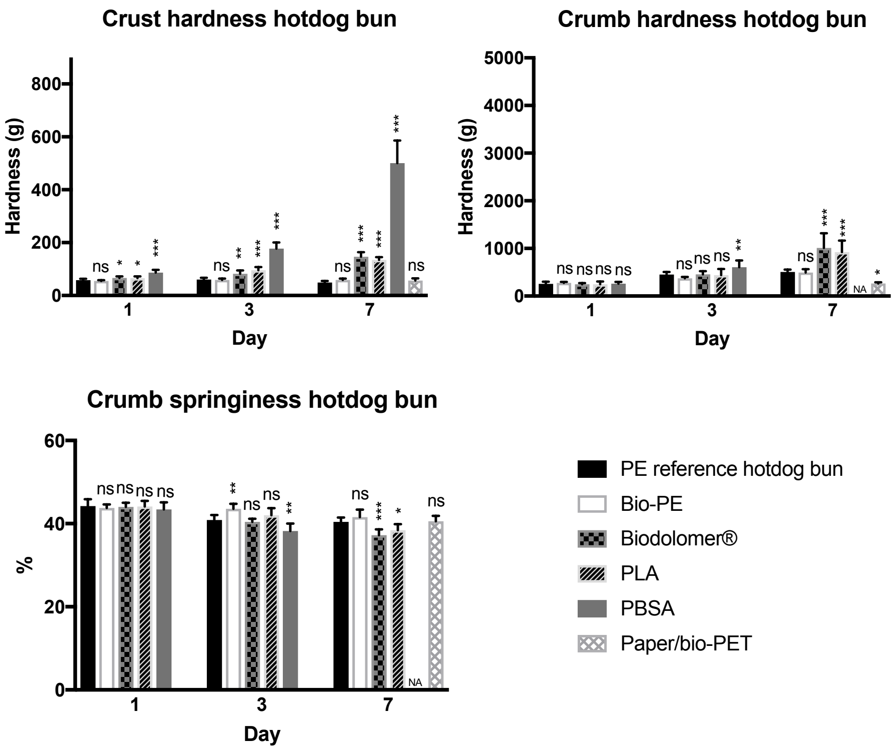
4.2.2.2.1 Hotdog buns

Crust hardness was one of parameters measured in the texture analysis. Increasing value of this parameter describes the loss of softness which is a negative attribute for bread (Cauvain, 2004). Figure 23 shows the comparison of crust hardness of hotdog buns packed in different biobased packaging materials and PE reference. Hotdog buns packed with bio-PE had similar crust hardness compared with hotdog buns in PE reference along the 7-day shelf life. Hotdog buns packed in paper/bio-PET, which was only evaluated in day 7 after thawing, showed the similar hardness to the PE reference. On the other side, significant different hardness of the hotdog buns was observed from the buns packed with Biodolomer®, PLA, and PBSA. In these biobased packaging materials, the crust softness lost in day 1 after thawing.

Crumb hardness was measured to evaluate the loss of softness in the crumb. From the same graph Figure 23, it shows that hotdog buns crumb hardness values between different packaging were not significantly different in the first day of shelf life after thawing. However, in day 7 after thawing, crumb hardness values of hotdog buns in Biodolomer®, PLA, and paper/bio-PET were significantly different than the buns in the PE reference. In fact, hot dog buns in PBSA were dried out and too hard to be measured.

Crumb springiness shows the crumb ability to recover after being compressed. Bread crumb is expected to be springy (Cauvain, 2004). Similar with crumb hardness, crumb springiness values of buns in day 1 were not significantly different among hotdog buns in different packaging materials (see Figure 23). In the last day of shelf life, only buns in bio-PE and paper/bio-PET had the same springiness compared to hot dog buns in PE reference.

In summary, hotdog buns packed with bio-PE and paper/bio-PET had the most similar textural properties to hotdog buns in PE reference. Biodolomer®, PLA, and PBSA seemed to maintain hotdog buns crumb hardness and springiness only in the first day after thawing but were unable to maintain the crust hardness of the hotdog buns.



NA = not tested because the value exceeded the instrument maximum measured value; ns = not significant p>0.05; *= significant p<0.05; **= significant p<0.01; ***= significant p<0.001; star means significant different value compared to PE reference in the same day of measurement.

Figure 24 Comparison of textural parameters of hotdog buns in different biobased packaging materials and PE reference

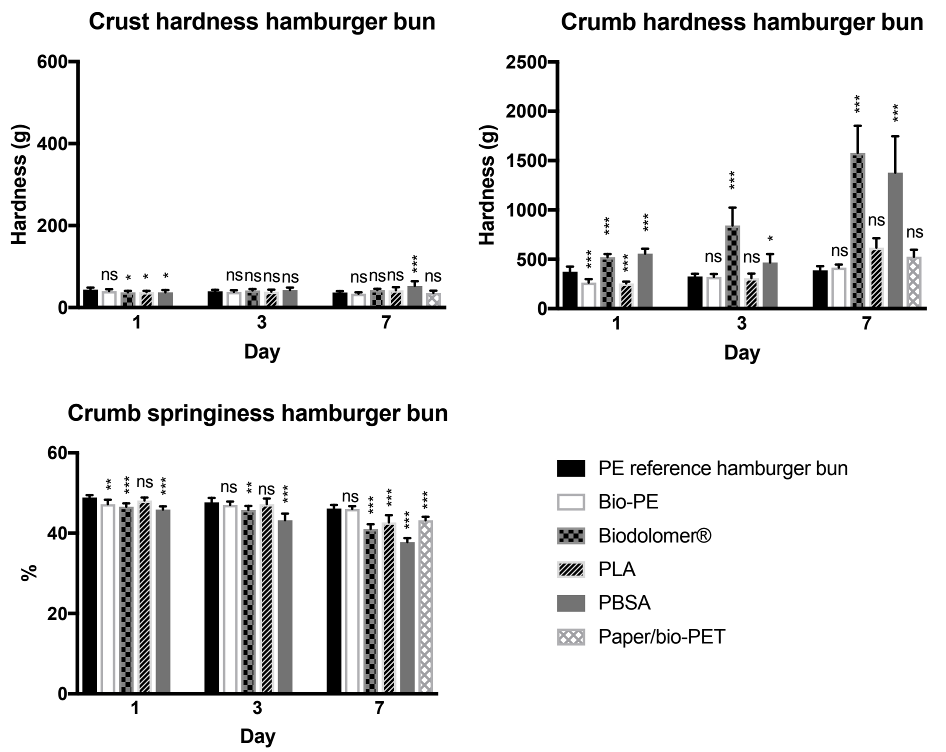
4.2.2.2.2 Hamburger buns

Crust hardness values of hamburger buns in bio-PE, Biodolomer®, PLA, and paper/bio-PET were not significantly different in the end of shelf life after thawing (see Figure 24). On the contrary, buns packed with PBSA had higher crust hardness compared to the PE reference.

Crumb hardness values of hamburger buns in bio-PE and PLA were significantly lower than the buns in the PE reference in day 1 after thawing (see Figure 24). However, in the day 7 after thawing, the crumb hardness values of hamburger buns in these packaging were not significantly different to buns in PE reference. The similar result in day 7 also observed from buns in paper/bio-PET. Crumb hardness values of buns in Biodolomer® and PBSA were significantly higher compared to PE reference along shelf life.

Crumb springiness values were decreasing in trend for buns in all packaging materials. In the day 7 after thawing, only buns in bio-PE had the same crumb springiness compared to buns in PE reference. Loss of springiness in hamburger bun seemed to be more prominent compared to loss of softness in crust and crumb.

In summary, only hamburger buns in bio-PE had the similar crust hardness, crumb hardness, and crumb springiness compared to buns in PE reference after 7-day shelf life. While crust hardness could be maintained by most of tested packaging materials after 7-day shelf life, crumb springiness showed a very significant lower result for buns in Biodolomer®, PLA, PBSA, and paper/bio-PET.



ns = not significant p>0.05; *= significant p<0.05; **= significant p<0.01; ***= significant p<0.001; star means significant different value compared to PE reference in the same day of measurement.

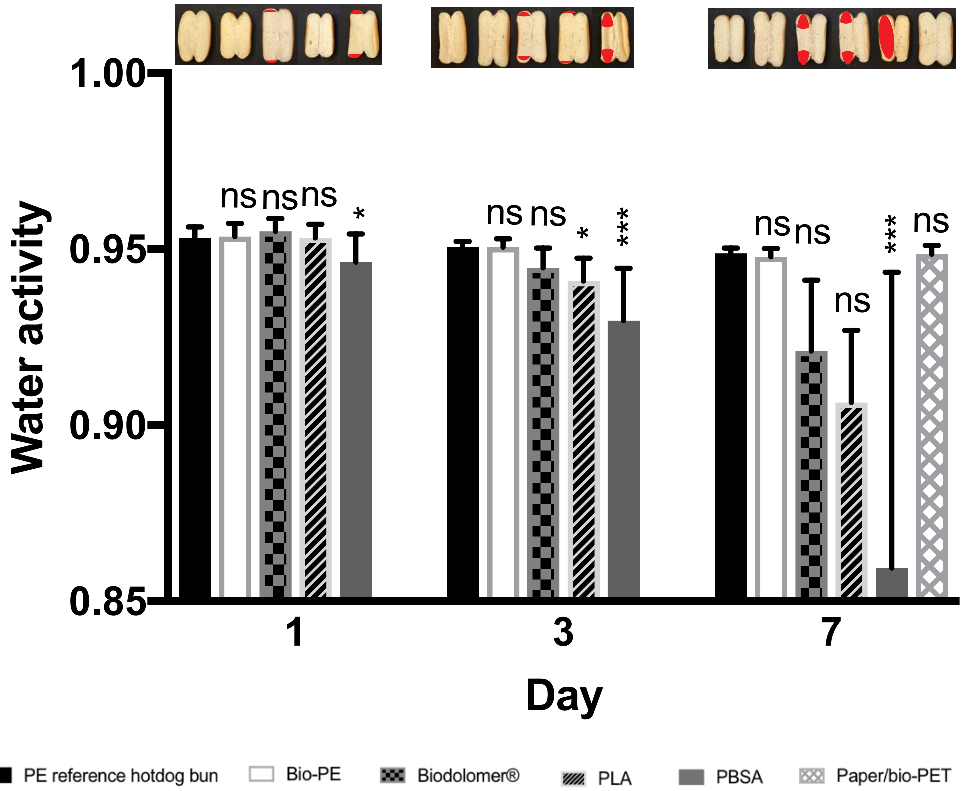
Figure 25 Comparison of textural parameters of hamburger buns in different biobased packaging materials and PE reference

4.2.3 Water activity and appearance of the buns

4.2.3.1 Hotdog buns

Water activity is closely related with available water in the buns that contribute to freshness perception as well as available water for biochemical reaction and microbial activity (Cauvain & Young, 2009a; Cauvain & Young, 2010). The water activity result is presented in complement with visual observation (appearance) of the buns.

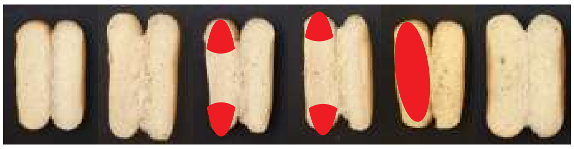
Figure 25 shows that the water activity values of buns in in bio-PE, Biodolomer®, and PLA were not significantly different compared to buns in PE reference. In day 7, it also showed that buns in all biobased packaging materials had the same water activity, except buns in the PBSA.



Marked area shows the dry area in the bun (see Appendix 2 for complete observation data on the appearance of the buns); ns = not significant p>0.05; *= significant p<0.05; **= significant p<0.01; ***= significant p<0.001; star means significant different value compared to PE reference in the same day of measurement.

Figure 26 Water activity of hotdog buns in different biobased packaging materials and PE reference

The visual observation of the hotdog buns showed that buns in bio-PE and paper/bio-PET had similar appearance with buns in PE reference. This corresponded with the non-significant water activity change compared to buns in PE reference. On the contrary, the edge of the buns packed with Biodolomer® and PBSA dried quite fast. Hotdog buns packed with Biodomomer® and PBSA showed a noticeable dry edge day 1 after thawing (see Figure 25). In the end of shelf-life, the buns in PBSA were entirely hard. Hotdog buns packed with PLA showed a dry edge which was noticed in day 3 but became more noticeable after 7-day after thawing. In Figure 26, marked area in the hotdog bun shows the dry part.



PE reference | Bio-PE | Bio-dolomer® | PLA | PBSA | Paper/bio-PET

Figure 27 Picture of hotdog buns day 7 after thawing

Water activity was measured only for the crumb in the middle of the hotdog buns which in this area the water activity seemed to change very minimally. Water activity values of buns in PLA and Biodolomer® were not significantly different in day 7 compared to water activity of buns in PE reference. However, observation of the appearance of the buns showed noticeable dry edge. It shows that moisture migration is more common to happen from outer part of the bun and less change in the very center of bun.

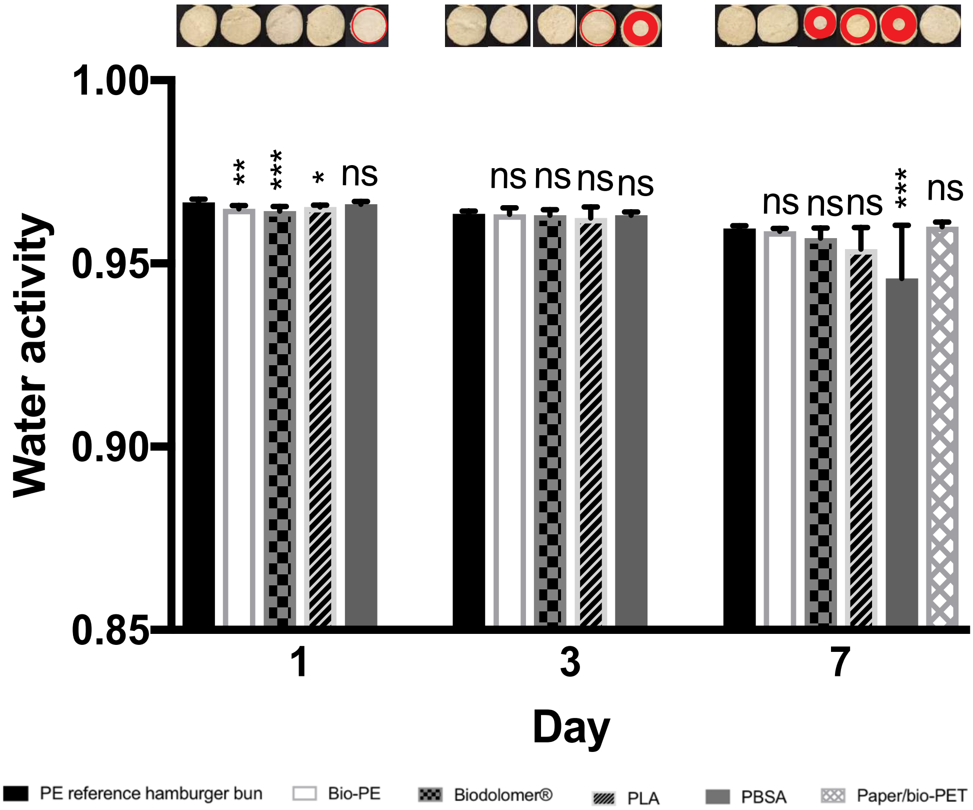
Comparing the water activity result and the appearance of the buns, only buns packed with bio-PE and paper/bio-PET had the similar characteristic with buns in PE reference.

4.2.3.2 Hamburger buns

Figure 27 shows that the water activity of hamburger buns in bio-PE, Biodolomer®, and PLA were lower compared to reference PE in day 1 after thawing. In the end of shelf life, however, the water activity values of the buns in these packaging were not significantly different compared to buns in the PE reference. On the contrary, buns in PBSA had significantly lower water activity in day 7 after thawing.

From the appearance, buns in PBSA were dry from day 1 after thawing especially in the edge of the buns. While water activity in the center was still maintained, the outer part of buns dried out. The edge of the buns was very hard in the end of shelf life. This also happened with buns in Biodolomer® and PLA which were more noticeable in day 7 after thawing (see Figure 28).

Only hamburger buns in bio-PE and paper/bio-PET had non-significant water activity and no dry edge after 7-day shelf life.



Marked area shows the dry area in the bun (see Appendix 3 for complete observation data on the appearance of the buns); ns = not significant p>0.05; *= significant p<0.05; **= significant p<0.01; ***= significant p<0.001; star means significant different value compared to PE reference in the same day of measurement.

Figure 28 Water activity of hamburger buns in different biobased packaging materials and PE reference

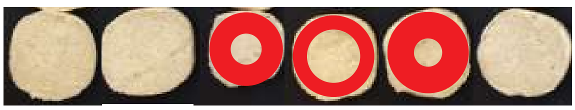


Figure 29 Picture of hamburger buns day 7 after thawing

4.3 Sensory analysis

Triangle test was performed to investigate if there were any sensory differences between the hotdog buns packed in bio-PE and the reference (PE) 7 days after thawing. Total of 20 panelists were asked to choose the sample that was different compared to the other two samples. The judgement was based on textural parameters and taste. The panelists were asked to evaluate the crust and crumb by touching. They were also asked to taste the buns to judge if there was any difference in the perceived texture in the mouth and flavor.

Table 12 Number of correct answer from triangle test

	Correct	Incorrect	Total
Triangle test 1	7[a]	13	20
Triangle test 2	4[b]	16	20

[a] correct answer in triangle test 1

[b] correct answer in triangle test 2

Triangle test (Table 12) shows that neither a and b is >10, which is the value required for statistical significance for 20 panelists. It means that no more than p_d=50% of the population were able to detect the different between hotdog buns in bio-PE and hotdog buns in PE reference with 95% certainty (β=0.05). Hotdog buns packed in Bio-PE and reference PE were similar until the end of shelf life (day 7).

Triangle test was not performed for hamburger bun. This due to the time constraint since hamburger buns are not produced as often as hotdog bun. Therefore, it was not possible to prepare hamburger bun sample for the sensory test. Triangle test is suggested to confirm if there is a significant different between hamburger buns in bio-PE and hamburger buns in PE reference.

4.4 Discussion

Five biobased packaging materials were evaluated for hotdog buns and hamburger buns packaging. The evaluation was designed to answer the research question in this study, *how does biobased packaging material affect the quality of fully-baked frozen bread in comparison to reference packaging?*

Table 13 shows the summary of evaluation of the buns the last day of shelf life (day 7). Evaluation results from bun 3 (the furthest sample from the opening) was chosen for the summary. Quality aspects for the evaluation include weight loss, crust hardness, crumb hardness, crumb springiness, a_W, and appearance of the buns.

Table 13 Summary of the evaluations on the buns in day 7 after thawing in comparison to PE reference

Hotdog bun

	Weight loss	Crust hardness	Crumb hardness	Crumb springi-ness	a_W	Visual observation (dry edge)
Bio-PE	⇔	⇔	⇔	⇔	⇔	No
Biodolomer®	⇑⇑⇑	⇑⇑⇑	⇑⇑⇑	⇓⇓⇓	⇔	Yes
PLA	⇑⇑⇑	⇑⇑⇑	⇑⇑⇑	⇓⇓⇓	⇔	Yes
PBSA	⇑⇑⇑	⇑⇑⇑	NA	NA	⇓⇓⇓	Yes
Paper/bio-PET	⇑⇑	⇔	⇓	⇔	⇔	No

Hamburger bun

	Weight loss	Crust hardness	Crumb hardness	Crumb springi-ness	a_W	Visual observation (dry edge)
Bio-PE	⇔	⇔	⇔	⇔	⇔	No
Biodolomer®	⇑⇑⇑	⇔	⇑⇑⇑	⇓⇓⇓	⇔	Yes
PLA	⇑⇑⇑	⇔	⇔	⇓⇓⇓	⇔	Yes
PBSA	⇑⇑⇑	⇑⇑⇑	⇑⇑⇑	⇓⇓⇓	⇓⇓⇓	Yes
Paper/bio-PET	⇔	⇔	⇔	⇓⇓⇓	⇔	No

⇑ means the value is significantly higher than PE reference; ⇓ means significantly lower than PE reference; ⇔ means the value is not significantly different than PE reference; green color means favorable characteristic; red color means not favorable characteristic.

From the result in this study, bio-PE seems to be a promising biobased packaging material. Buns in this packaging had similar quality compared to buns packed in PE reference in all investigated quality aspects. The same result was shown for both hotdog buns and hamburger buns. Moreover, the triangle test confirmed that there was no difference between hotdog buns packed with bio-PE and hotdog buns packed with PE reference perceived by the panelist. Changing the packaging to bio-PE will result the similar quality of hotdog buns until the end of shelf life after thawing (7 days).

Thinner bio-PE film (35 µm) compared to PE reference (40 µm) for hamburger buns did not give impact to the quality of the buns. In fact, the WVTR values of bio-PE were similar to PE reference for hamburger buns. However, sensorial aspect perceived by the consumer of the hamburger buns should be considered. This aspect was not evaluated in this study.

Paper/bio-PET could maintain buns quality aspects except weight loss (hotdog buns), crumb hardness (hotdog buns), and crumb springiness (hamburger bun). Higher weight loss might be caused by the higher packaging WVTR value. The lower WVTR value, the lower will be the gain or loss of water from product

(Cauvain & Young, 2009a). Paper/bio-PET had higher WVTR value, about 6 times higher than PE reference. Despite the high WVTR of this material, the observation on the appearance of the buns showed no dry edge was observed from the buns in this packaging. It might be interesting to evaluate this material further. However, this packaging does not allow product visibility. Moreover, sealing with clip might be difficult to apply to this material.

Biodolomer®, PLA, and PBSA could not maintain the quality of packed buns until the end of shelf life (7 days). Dry edge and significant weight loss from the packed buns were observed in all buns packed in these materials in day 7 after thawing. These materials had much higher WVTR values compared to PE reference. This explained the weight loss of bun packed with these materials was significantly higher than buns in PE reference.

WVTR of most biobased packaging materials used in this study were higher than PE reference. This characteristic seemed to affect the packed buns quality. Lower permeability to water vapour is important for preserving the buns quality after thawing. Bio-PE with similar WVTR value to reference could maintain the buns along shelf life. Paper/PET with WVTR value 25.09 g/m^2day could maintain some of the quality aspects such as the weight loss and minimizing the dry edge in hamburger buns. However, other quality aspects were not maintained, such as the crumb hardiness and springiness. Lower WVTR value of packaging is necessary for this type of bread.

5 Conclusions and future research recommendations

This section consists the conclusion of the findings in the evaluation of 5 biobased packaging material for hotdog buns and hamburger buns packaging application. The research question is answered and recommendations for further studies are suggested.

This study focuses on the evaluation of biobased packaging materials that are available in the market for two different type of commercial buns i.e. hotdog buns and hamburger buns. Five biobased packaging materials and PE reference were evaluated. The evaluations were performed for both the packaging materials and the quality of the buns. Quality aspects of buns include the weight loss along 7-day shelf life, textural properties, water activity, and the appearance of the buns.

Research question: How does biobased packaging material affect the quality of fully-baked frozen bread in comparison to reference packaging?

The conclusions drawn from the primary research in this study for each objective:

1. To identify the characteristics of different biobased packaging materials (film) that exist in the market.

Biobased packaging materials used in this study have different characteristics in term of transparency, biobased content, biodegradability, and barrier properties. Transparency is a major consideration for product visibility. This characteristic is missing in Biodolomer® and paper/bio-PET. All selected packaging materials had the similar or higher puncture resistance compared to PE reference. In term of barrier properties against water vapor, Biodolomer®, PLA, PBSA, and paper/bio-PET had higher WVTR value compared to PE reference. Only bio-PE had similar WVTR value to PE reference.

2. To evaluate how biobased packaging material would maintain the quality of the hotdog buns and hamburger buns for 7-day shelf life after thawing in comparison with reference PE bag.

Bio-PE could maintain the quality of hotdog buns and hamburger buns in 7-day shelf life after thawing. A triangle test confirmed that there was no significant difference between hotdog buns packed in bio-PE and PE reference. On the other side, Biodolomer®, PLA, and PBSA did not perform well for this type of application which showed in the higher weight loss and dry edge of the buns in day 7 after thawing. Paper/bio-PET is an interesting biobased packaging material since it could maintain some of the quality parameters except the textural properties and weight loss (hotdog buns).

Future research recommendations

The biobased packaging materials used in this study were limited to the availability of the sample from the suppliers. Many biobased packaging materials in the market such as starch-based materials, cellulose-based materials, or compound materials are also interesting to be evaluated for future research. Furthermore, thickness of the packaging material might influence its performance. Therefore, it might be an interest to evaluate how different thickness of biobased packaging materials affects the quality of the packed product.

The experiment in this study was limited to 7-day shelf life of the fully-baked frozen bread after thawing. Since this product should be able to be stored in frozen condition for 6-12 months prior to thawing, it is important to evaluate the performance of the packaging materials along frozen storage. This needs to be studied further.

Study on the environmental impacts of the biobased packaging materials might be an interesting issue to be studied. It is difficult to say which biobased packaging material is more environmentally friendly without any assessment of the environmental impacts. Moreover, price comparison might be an interest for the industry to decide the best biobased packaging material option.

Biobased packaging materials used in this study have significantly higher WVTR values compared to PE reference. For future packaging selection for this product, WVTR around the WVTR PE reference or below WVTR of paper/bio-PET shall be considered. However, it might be interesting to evaluate biobased packaging materials for other type of product such as crusty bread, such as pastries, which require a more breathable packaging. Packaging for frozen bread can be another application to be evaluated since the water loss during freezing is minimum.

6 References

Akelah, A. (2013). Polymers in Food Packaging and Protection. In A. Akelah (Ed.), *Functionalized Polymeric Materials in Agriculture and the Food Industry* (pp. 293-347). Boston, MA: Springer US.

Albertsson, A.-C., Edlund, U., & Varma, I. K. (2011). Synthesis, Chemistry and Properties of Hemicelluloses *Biopolymers – New Materials for Sustainable Films and Coatings* (pp. 133-150): John Wiley & Sons, Ltd.

Auras, R., Harte, B., & Selke, S. (2004). An Overview of Polylactides as Packaging Materials. *Macromolecular Bioscience, 4*(9), 835-864. doi:10.1002/mabi.200400043

Avérous, L. (2004). Biodegradable Multiphase Systems Based on Plasticized Starch: A Review. *Journal of Macromolecular Science: Polymer Reviews, 44*(3), 231-274. doi:10.1081/MC-200029326

Baker, A., Walker, C., & Kemp, K. (1988). An optimum compression depth for measuring bread crumb firmness. *Cereal Chem., 65(4)*, 302-307.

Bourne, M. C. (2002). Chapter 1 - Texture, Viscosity, and Food *Food Texture and Viscosity (Second Edition)* (pp. 1-32). London: Academic Press.

Cauvain, S. P. (2004). 18 - Improving the texture of bread A2 - Kilcast, David *Texture in Food* (pp. 432-450): Woodhead Publishing.

Cauvain, S. P. (2017a). Chapter 10 - Testing Methods *Baking Problems Solved (Second Edition)* (pp. 469-480): Woodhead Publishing.

Cauvain, S. P. (2017b). Chapter 11 - What? *Baking Problems Solved (Second Edition)* (pp. 481-500): Woodhead Publishing.

Cauvain, S. P., & Young, L. S. (2009a). Effects of Water on Product Textural Properties and Their Changes During Storage *Bakery Food Manufacture and Quality* (pp. 143-173): Wiley-Blackwell.

Cauvain, S. P., & Young, L. S. (2009b). Water Activity *Bakery Food Manufacture and Quality* (pp. 174-198): Wiley-Blackwell.

Cauvain, S. P., & Young, L. S. (2009c). Water and Its Roles in Baked Products *Bakery Food Manufacture and Quality* (pp. 1-31): Wiley-Blackwell.

Cauvain, S. P., & Young, L. S. (2010). 13 - Chemical and physical deterioration of bakery products *Chemical Deterioration and Physical Instability of Food and Beverages* (pp. 381-412): Woodhead Publishing.

Cooksey, K., & Krochta, J. (2011). Introduction to Frozen Food Packaging *Handbook of Frozen Food Processing and Packaging, Second Edition* (pp. 711-730): CRC Press.

Ducruet, V., Domenek, S., Guinault, A., Courgneau, C., Bernasconi, M., & Plessis, C. (2011). Barrier Properties of PLA towards Oxygen and Aroma Compounds. *Italian Journal of Food Science*, 59-62.

Embree, K. (2016). Innovia Groups sells Cellophane division to Futamura Chemicals. Retrieved May 12, 2017, from https://www.plasticstoday.com/packaging/innovia-groups-sells-cellophane-division-futamura-chemicals/6213795224461

Euromonitor International. (2014). *Bread, Cakes and Pastries: A Global Market Overview* from http://www.portal.euromonitor.com/portal/?8xlPvvKO7FgCz7581TtlfQ%3d%3d

European Bioplastics. (2016a). *Bioplastics - Facts and Figures*. Retrieved February 10, 2017, from http://docs.european-bioplastics.org/publications/EUBP_Facts_and_figures.pdf

European Bioplastics. (2016b). *Bioplastics – Industry standards & labels*. Retrieved February 9, 2017, from http://docs.european-bioplastics.org/2016/publications/fs/EUBP_fs_standards.pdf

European Bioplastics. (2016c). *Guidelines for the Use of Seedling Logo*. Retrieved February 9, 2017, from http://docs.european-bioplastics.org/2016/publications/EUBP_Guidelines_Seedling_logo.pdf

Fiszman, S. M., Salvador, A., & Varela, P. (2005). Methodological developments in bread staling assessment: application to enzyme-supplemented brown pan bread. *European Food Research and Technology, 221*(5), 616-623. doi:10.1007/s00217-005-0082-2

Futamura brochure. The ABC of Bio-laminates: Futamura.

Gällstedt, M., Hedenqvist, M. S., & Ture, H. (2011). Production, Chemistry and Properties of Proteins *Biopolymers – New Materials for Sustainable Films and Coatings* (pp. 107-132): John Wiley & Sons, Ltd.

Gray, J. A., & Bemiller, J. N. (2003). Bread Staling: Molecular Basis and Control. *Comprehensive Reviews in Food Science and Food Safety, 2*(1), 1.

Greene, J. P. (2014a). Biobased and Biodegradable Polymers *Sustainable Plastics* (pp. 71-106): John Wiley & Sons, Inc.

Greene, J. P. (2014b). Biobased and Recycled Petroleum-Based Plastics *Sustainable Plastics* (pp. 107-127): John Wiley & Sons, Inc.

Hellström, D., Olsson, A., & Nilsson, F. (2016a). Introduction to packaging *Managing Packaging Design for Sustainable Development* (pp. 3-15): John Wiley & Sons, Ltd.

Hellström, D., Olsson, A., & Nilsson, F. (2016b). Sustainable development and packaging *Managing Packaging Design for Sustainable Development* (pp. 17-33): John Wiley & Sons, Ltd.

Holm, V. (2009). Shelf Life of Foods in Biobased Packaging *Food Packaging and Shelf Life* (pp. 353-365): CRC Press.
Hu, Y., Topolkaraev, V., Hiltner, A., & Baer, E. (2001). Measurement of water vapor transmission rate in highly permeable films. *Journal of Applied Polymer Science, 81*(7), 1624-1633. doi:10.1002/app.1593
Innventia website. Nanocellulose. Retrieved March 13, 2017, from http://www.innventia.com/en/Our-Expertise/New-materials/Nanocellulose/
Kondo, K. (1990). Chapter 7 - Plastic Containers A2 - Kadoya, Takashi *Food Packaging* (pp. 117-145). San Diego: Academic Press.
Lange, J., Mokdad, H., & Wysery, Y. (2002). Understanding Puncture Resistance and Perforation Behavior of Packaging Laminates. *Journal of Plastic Film & Sheeting, 18*(4), 231-244. doi:doi:10.1177/8756087902033865
Le Bail, A., Hamdami, N., Jury, V., Monteau, J.-Y., Davenel, A., Lucas, T., & Ribotta, P. D. (2006, 11 August 2006). Examining crust problems of frozen bread. *New Food Magazine*.
Le Bail, A., Tzia, C., & Giannou, V. (2011). Quality and Safety of Frozen Bakery Products *Handbook of Frozen Food Processing and Packaging, Second Edition* (pp. 501-528): CRC Press.
Lee, K., & Sun, D.-W. (2011). Plastic Packaging of Frozen Foods *Handbook of Frozen Food Processing and Packaging, Second Edition* (pp. 731-742): CRC Press.
Madhavan Nampoothiri, K., Nair, N. R., & John, R. P. (2010). An overview of the recent developments in polylactide (PLA) research. *Bioresource Technology, 101*(22), 8493-8501. doi:http://dx.doi.org/10.1016/j.biortech.2010.05.092
Man, C. M. D. (2015). *Shelf life*: Hoboken, NJ : Wiley-Blackwell, 2015.
Marsh, K., & Bugusu, B. (2007). Food packaging - Roles, materials, and environmental issues. *JOURNAL OF FOOD SCIENCE, 72*(3), R39-R55.
McQuilken, K. (2016). SUSTAINABLE PACKAGING And Its Role in the Supply Chain. *Nutraceuticals World, 19*(7), 52-54.
Mikkonen, K. S., & Tenkanen, M. (2012). Sustainable food-packaging materials based on future biorefinery products: Xylans and mannans. *Trends in Food Science & Technology, 28*(2), 90-102. doi:http://dx.doi.org/10.1016/j.tifs.2012.06.012
Molenveld, K., Van Den Oever, M., & Bos, H. (2015). Biobased Packaging Catalogue. Wageningen: Wageningen UR Food & Biobased Research.
Paunonen, S. (2013). Strength and Barrier Enhancements of Cellophane and Cellulose Derivative Films: A Review. *BioResources, 8*(2), 3098-3121.
Persson, A. (2016). *The Performance of Bio-produced PE and PET in Flexible Packaging Materials : a Benchmarking Evaluation.* (Master), Lund University. Retrieved February 15, 2017, from https://lup.lub.lu.se/student-papers/search/publication/8627821

PlasticEurope. (2015). *Plastic – the Fact 2015: An analysis of European plastics production, demand, and waste data*. Retrieved December 1, 2016, from http://www.plasticseurope.org/Document/plastics-the-facts-2014.aspx
PlasticEurope. (2016). *Plastic – the Fact 2016: An analysis of European plastics production, demand, and waste data*. Retrieved March 30, 2017, from http://www.plasticseurope.org/Document/plastics---the-facts-2016-15787.aspx?FolID=2
Plastics Technology. (2015). Coca-Cola Debuts First 100% Biobased PET Bottle. Retrieved February 10, 2017, from http://www.ptonline.com/articles/coca-cola-debuts-first-100-biobased-pet-bottle(2)
Pollet, E., & Avérous, L. (2011). Production, Chemistry and Properties of Polyhydroxyalkanoates *Biopolymers – New Materials for Sustainable Films and Coatings* (pp. 65-86): John Wiley & Sons, Ltd.
Rastogi, K. V., & Samyn, P. (2015). Bio-Based Coatings for Paper Applications. *Coatings, 5*(4). doi:10.3390/coatings5040887
Rejak, A., Wójtowicz, A., Oniszczuk, T., Niemczuk, D., & Nowacka, M. (2014). Evaluation of Water Vapor Permeability of Biodegradable Starch-Based Films. *TEKA. COMMISSION OF MOTORIZATION AND ENERGETICS IN AGRICULTURE, Vol. 14, No.3*, 89-94.
Robertson, G. L. (2016). 3 - Packaging and Food and Beverage Shelf Life A2 - Subramaniam, Persis *The Stability and Shelf Life of Food (Second Edition)* (pp. 77-106): Woodhead Publishing.
Roudaut, G., & Debeaufort, F. (2010). 6 - Moisture loss, gain and migration in foods and its impact on food quality *Chemical Deterioration and Physical Instability of Food and Beverages* (pp. 143-185): Woodhead Publishing.
Sablani, S. (2011). Glass Transitions in Frozen Food Systems *Handbook of Frozen Food Processing and Packaging, Second Edition* (pp. 39-54): CRC Press.
Sárossy, Z. (2011). *Production and Utilization of Hemicelluloses from Renewable Resources for Sustainable Advanced Products.* (PhD), Technical University of Denmark. Retrieved February 13, 2017, from http://orbit.dtu.dk/files/7969594/Thesis_Zs_Sarossy.pdf
Shogren, R. (1997). Water vapor permeability of biodegradable polymers. *Journal of environmental polymer degradation, 5*(2), 91-95. doi:10.1007/BF02763592
Sinha Ray, S., & Bousmina, M. (2005). Biodegradable polymers and their layered silicate nanocomposites: In greening the 21st century materials world. *Progress in Materials Science, 50*(8), 962-1079. doi:http://dx.doi.org/10.1016/j.pmatsci.2005.05.002
Siracusa, V. (2012). Food Packaging Permeability Behaviour: A Report. *International Journal of Polymer Science, 2012*, 11. doi:10.1155/2012/302029
Siracusa, V., Lotti, N., Munari, A., & Dalla Rosa, M. (2015). Poly(butylene succinate) and poly(butylene succinate-co-adipate) for food packaging

applications: Gas barrier properties after stressed treatments. *Polymer Degradation and Stability, 119*, 35-45. doi:http://doi.org/10.1016/j.polymdegradstab.2015.04.026

Södergård, A., & Inkinen, S. (2011). Production, Chemistry and Properties of Polylactides *Biopolymers – New Materials for Sustainable Films and Coatings* (pp. 43-63): John Wiley & Sons, Ltd.

Stone, H., & Sidel, J. L. (2004). *Sensory Evaluation Practices*: Burlington : Academic Press, 2004, 3. ed.

Tokiwa, Y., Calabia, B. P., Ugwu, C. U., & Aiba, S. (2009). Biodegradability of Plastics. *International Journal of Molecular Sciences, 10*(9), 3722-3742. doi:10.3390/ijms10093722

Tsiropoulos, I., Faaij, A. P. C., Lundquist, L., Schenker, U., Briois, J. F., & Patel, M. K. (2015). Life cycle impact assessment of bio-based plastics from sugarcane ethanol. *Journal of Cleaner Production, 90*, 114-127. doi:http://dx.doi.org/10.1016/j.jclepro.2014.11.071

Vázquez, A., Foresti, M. L., & Cyras, V. (2011). Production, Chemistry and Degradation of Starch-Based Polymers *Biopolymers – New Materials for Sustainable Films and Coatings* (pp. 15-42): John Wiley & Sons, Ltd.

Venkataraman, V. (2012). *Propy-LENE Supply! Go Green! On-Purpose technologies for the future!* Retrieved February 10, 2017, from www.beroe-inc.com

Weber, C. J. (2000). *Packaging Materials for the Food Industry - Status and Perspective*. Retrieved February 9, 2017, from www.biodeg.net/fichiers/Book on biopolymers (Eng).pdf

7 Appendices

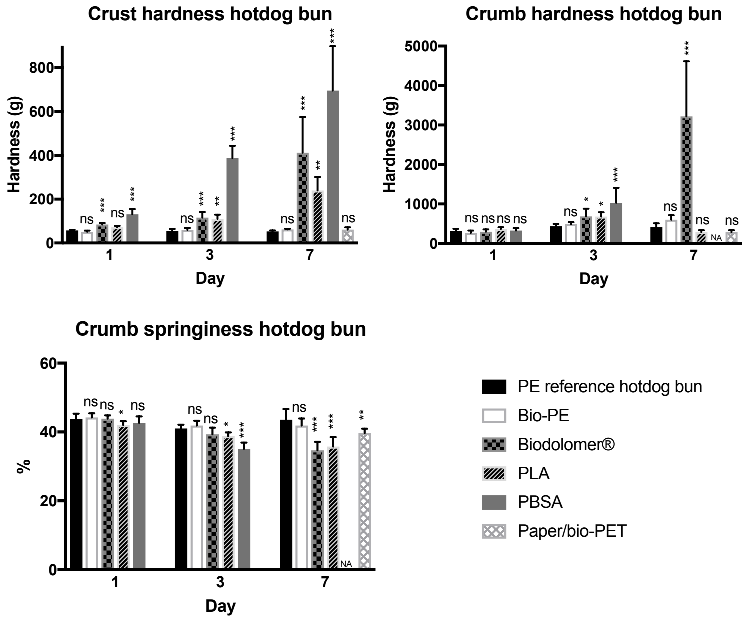
Appendix 1

Biobased content of the packaging materials

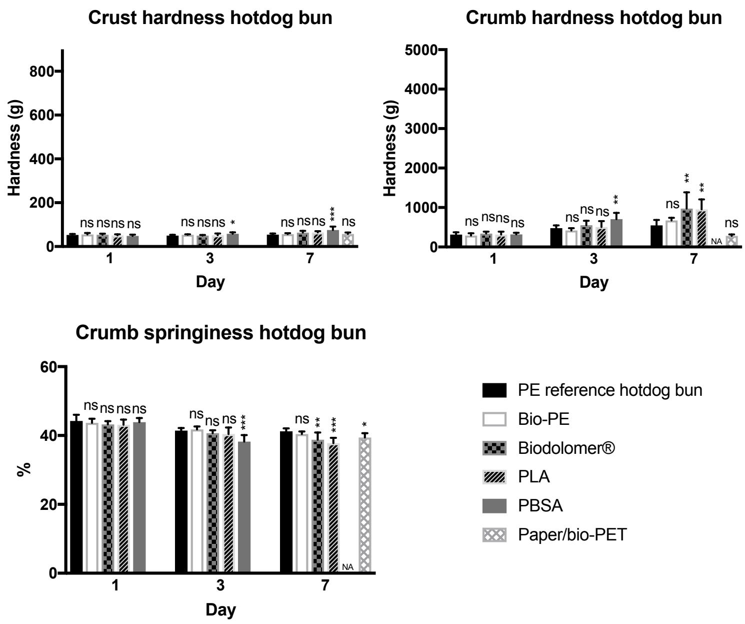
Packaging materials	Biobased content	Source	Contact
Bio-PE	78%	Amerplast	Ari-Pekka Pietilä
Biodolomer®	50-52%	GAIA Biomaterials	Åke Rosén
PLA	95%	Sidaplax	Mireille Lefever
PBSA	25%	Reverdia	Michel Hoogeveen
Paper/bio-PET	72%	(Persson, 2016)	Ronny Gimbe, Amanda Persson

Appendix 2

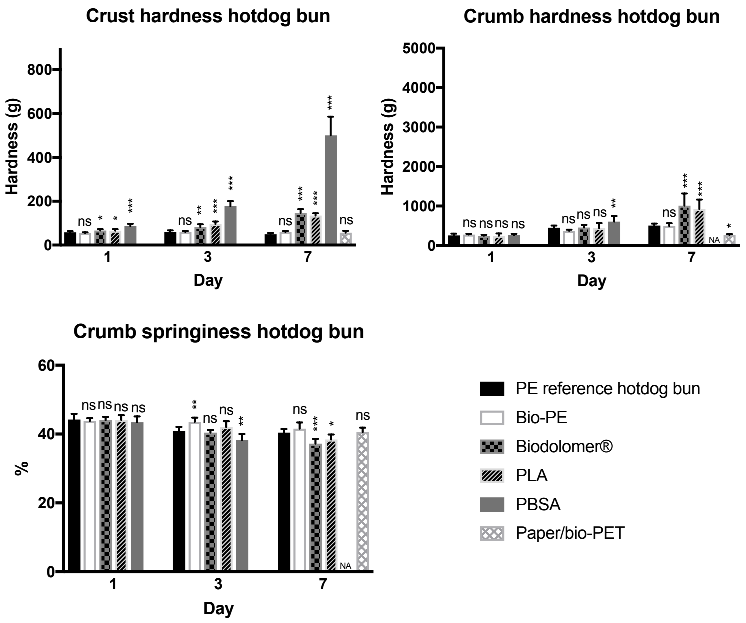
1. Textural measurement bun 1 - hotdog bun



2. Textural measurement bun 2 - hotdog bun

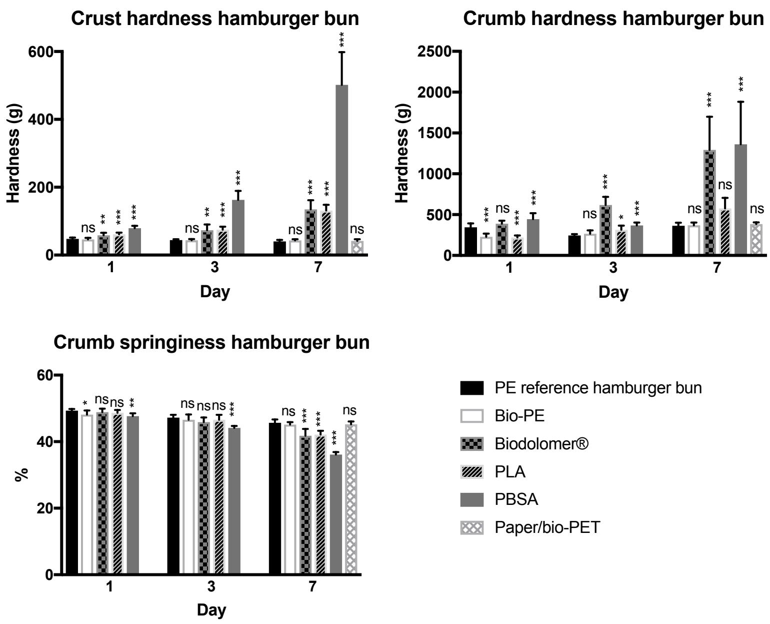


3. Textural measurement bun 3 - hotdog bun

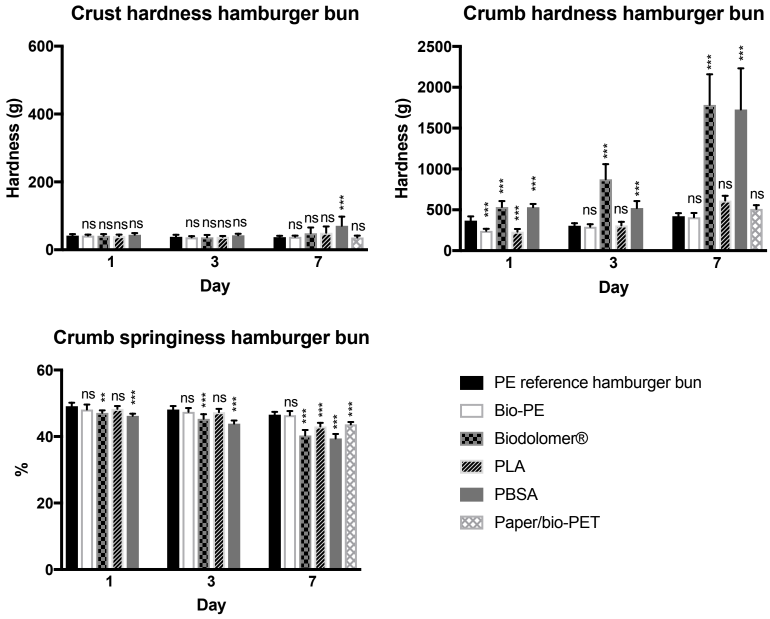


Appendix 3

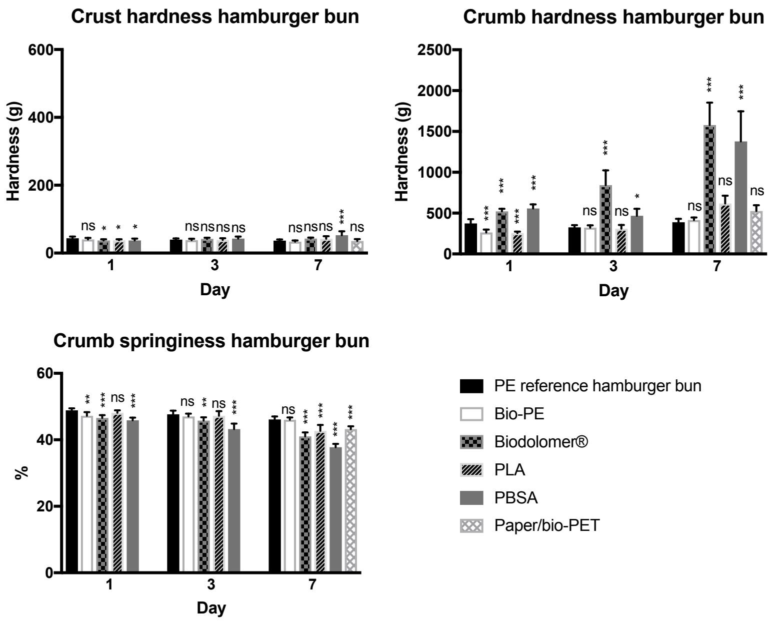
1. Textural measurement bun 1 - hamburger bun



2. Textural measurement bun 2 - hamburger bun

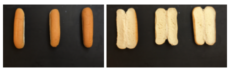
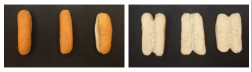
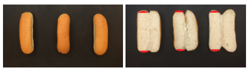


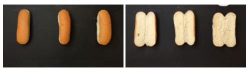
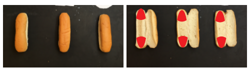
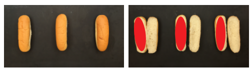
3. Textural measurement bun 3 - hamburger bun



Appendix 4

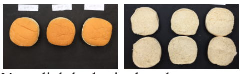
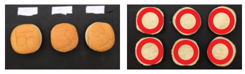
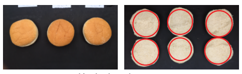
1. Observation on appearance of hotdog buns

	Day 1	Day 3	Day 7
PE reference	 No remarks	 No remarks	 No remarks
Bio-PE	 No remarks	 No remarks	 No remarks
Biodolomer®	 Slighlty hard edge (red area in the picture).	 Hard edge (red area in the picture).	 Hard edge (red area in the picture). Lock was still intack but very weak.

	Day 1	Day 3	Day 7
PLA	 Slightly dry in small area in the edge.	 Slighlty hard edge (red area in the picture).	 Wringkle and hard crust. Hard in the red area in the picture, center of the crumb is still moist. The seal was very weak.
PBSA	 Dry crust. Very hard edge (red area in the picture).	 Dry crust and crumb. Very hard edge (red area in the picture). Difficult to open.	 Wringkle crust surface Crust and crumb were completely hard The seal was torned after opening.
Paper/bio-PET			 No remarks

2. Observation on appearance of hamburger buns

	Day 1	Day 3	Day 7
PE reference	 No remarks	 No remarks	 No remarks
Bio-PE	 No remarks	 No remarks	 No remarks
Biodolomer®	 No remarks	 Crust was slighly dry. Slightly dry in the edge. Seal was slightly easy to open.	 Crust was very dry. Hard edge (red area in the picture). Ony center of the crumb was moist.

	Day 1	Day 3	Day 7
PLA	 Very slightly dry in the edge.	 Slightly dry crumb. Edge was slightly hard (red area in the picture).	 Crust was hard. Edge was hard (red area in the picture).
PBSA	 Crust was slightly dry. Edge was slightly hard (red area in the picture).	 Crumb is very hard. Dry edge (red area in the picture). Only middle part of crumb was moist.	 Wrinkle surface and shrinking size. Very hard crust and crumb (red area in the picture). Only middle part of crumb was moist.
Paper/bio-PET			 Very slightly hard in the edge but overall bun was still moist.

Appendix 5

ADULT CONSENT FORM

Starting Date	**16th May, 2017**
Venue	**Sensory Lab, Kemicentrum**
Project by	**Yoga Putranda**

Project Outline

A total of 20 volunteers will be asked to taste a sample of **commercial hotdog buns.** Further instructions will be given before starting the test. Participation in the study is strictly voluntary and participants can withdraw at any time, without giving a reason.

Please read the ingredient list carefully and make sure you are able and satisfied to consume the product/s being tested today.

If you have any allergies please make them known to the researcher at this point.

Participants must complete the consent form below before taking part in the taste test

		Please tick box
1.	I confirm that I have read and understand the above information and have had the opportunity to ask questions.	❒
2.	I understand that my participation is strictly voluntary and that. I am free to withdraw at any time, without giving reason	❒
3.	I have read the attached list of ingredients and confirm that I am able and satisfied to consume the products being tested.	❒
4.	I have **no allergies** to the tested product.	❒
5.	I agree to take part in the above study.	❒

List of Ingredients:
WHEAT FLOUR, water, invert sugar, rapeseed oil, yeast, **WHEAT GLUTEN**, fermented **WHEAT FLOUR**, salt, vegetable emulsifier (E471), **MALT FLOUR**, flour treatment agent E300.

ALLERGEN ALERT: **GLUTEN**, possible trace of **SESAME SEEDS**, lactose free

Triangle test

Tester No.: _____ Name: ____________ Date: 16 May 2017

Sample: hotdog bun

Instructions

Write down the code of the sample below. You will evaluate 2 sets of samples. Evaluate the samples from the left to right. Two samples are alike; one is different. Select the odd/different sample and identify it by placing X in the corresponding box.

Instruction for evaluation:

1. Touch the surface of the buns to evaluate dryness on the surface.
2. Open the bun and evaluate the crumb by pressing it with your finger.
3. Taste the bun by eating from the side of the bun (*see picture in the right*). Evaluate the texture in mouth and the flavor. Drink the water every time you move to the next sample.
4. Appearance is NOT a deciding parameter to select the odd sample.

Sample set 1

Samples code	Indicate odd sample	Remarks
____	☐	______________________________
____	☐	______________________________
____	☐	______________________________

Sample set 2

Samples code	Indicate odd sample	Remarks
____	☐	______________________________
____	☐	______________________________
____	☐	______________________________

If you wish to comment on the reason for your choice or on the characteristics of the sample, you may do so under Remarks.