# CHARACTERISATION OF BIODEGRADABLE PACKAGING MATERIALS AND EVALUATION OF THEIR EFFECTIVENESS IN THE SHELF-LIFE EXTENSION OF ROUND AND CHERRY TOMATOES

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### DISCLAIMER

As the candidate's supervisor, I have approved this thesis for submission.

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### PREFACE

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#### ABSTRACT

Fresh fruit and vegetables are highly perishable agricultural products. The packaging of fresh agricultural produce helps to preserve its shelf-life during transportation and storage. Plasticbased films are commonly used for packaging fresh fruit and vegetables. These films, however, contain significant amounts of non-biodegradable materials and thus pose a substantial environmental risk. Various initiatives have been undertaken to resolve this issue; however, recent attention has focused on biopolymer-based films that are renewable and biodegradable. The biodegradable films are mechanically weaker and possess fewer barrier properties than their commercial counterparts. It has been found that the biodegradable film properties can be improved by adding various additives, such as plasticisers. Biodegradable, natural polymer-based packaging materials are gaining considerable momentum and attracting interest for mitigating the detrimental effects and the increasing challenges related to the accumulation and control of petroleum-based plastic waste in the environment. The study evaluated the combined effects of storage conditions and packaging materials on shelf life extension of round and cherry tomatoes.

This study characterised biodegradable packaging materials (Chapter 3) and investigated the effects of these materials vs. conventional packaging materials on the extended shelf-life of round and cherry tomatoes (Chapter 4). The tomato samples and packaging samples used in this study were obtained from the ZZ2 Farm in Limpopo and transported to the Pietermaritzburg Fresh Produce Market in South Africa. The experimental design was laid out as randomised complete block with packaging materials as main plots and storage condition as subplots. The experiment was carried out in winter season.

The biodegradable packaging materials were characterized by their physical, mechanical and water resistant and water permeability properties. They were stored under ambient (20°C, RH 60.06%) and cold (12°C, RH 79.55%) storage conditions for 28 days. The results revealed that the samples stored in a cold environment had a lower tensile strength and elasticity modulus, compared to the samples stored under ambient conditions. The cold storage conditions significantly (p<0.05) reduced the mechanical properties of the biodegradable materials, compared to those of the conventional packaging materials. The effects of storage on the mechanical properties of conventional packaging was not significant (p>0.05). The pulped paper tray for cherry tomatoes showed the lowest tensile strength ( $62.59\pm0.4719^{\text{ef}} - 48.50\pm0.2996^{\text{c}}$  mpa), compared to the other biodegradable packaging materials. Biodegradable

materials showed the highest water vapour permeability, with the pulped paper tray having the highest solubility and water uptake. The stamped and glued paper trays showed the ability to resist water and a low solubility percentage of 11.04% and 11.31%, respectively.

The effectiveness of biodegradable packaging materials for extending the shelf-life of tomatoes was investigated in Chapter 4. Tomatoes were stored at ambient (20°C) and cold (12°C) temperatures for 28 days and sampled on Days 0, 7, 14, 21 and 28. Changes in the quality of the produce (their colour, firmness, TSS, pH, marketability) were investigated by means of a subjective analysis.

The results revealed a highly significant difference (P<0.001) in the physiological weight loss and marketability percentage of cherry and round tomatoes packed in biodegradable and conventional packaging materials. Biodegradable packaging materials were the most effective for preserving the quality of tomatoes and recorded the highest marketability percentage (75%). Under cold storage, the weight loss was 2.725% for biodegradable materials and 3.642% for conventional materials. The weight loss of tomatoes kept under ambient storage conditions was 5.816% for biodegradable packaging and 7.119% for conventional packaging. The results revealed a highly significant difference (p<0.001) between the physiological weight loss of tomatoes and the packaging materials. The mean physiological weight loss was 4.3% and 5.4% for biodegradable and conventional packaging materials, respectively. The marketability percentage was also high for tomatoes stored in cold conditions, compared to ambient conditions. The firmness of the tomatoes decreased with the storage period across all the packaging materials. The average puncture force was 6.5 N and 6.1 N for biodegradable and conventional packaging materials, respectively. The mean compression force was also significantly high for the biodegradable materials (92 N) and conventional materials (86 N). For the shearing force, the mean values were 7.7 N.g<sup>-1</sup> and 7.9 N.g<sup>-1</sup> for biodegradable and conventional packaging materials, respectively. The hue values of fruit stored under cold conditions were slightly higher than those stored under ambient conditions. However, there was no significant difference (p>0.005) between the tomatoes in biodegradable and conventional packaging, in terms of their hue. The tomato pH showed varying trends, depending on the storage conditions and duration. The pH of cherry and round tomatoes was slightly higher under ambient storage conditions than the pH under cold storage conditions for both packaging groups (biodegradable and conventional packaging). The tomatoes stored in different packaging materials also varied significantly (p<0.001) with the storage conditions, with those under cold

storage conditions having lower soluble solids than those stored under ambient conditions. Biodegradable packaging materials were the most effective, in terms of minimising the physiological weight loss of tomatoes, and they had the highest marketability percentage at the end of the 28-day storage period. Therefore, these findings suggest that biodegradable packaging materials are the most effective for extending the shelf-life of tomatoes.

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# TABLE OF CONTENTS

ABS	STRA	СТ		III
ACI	KNOV	WLEDGI	EMENTS	VI
LIS	T OF	FIGURE	ES	X
LIS	T OF	TABLES	S	XII
LIS	T OF	ABBRE	VIATIONS AND SYMBOLS	XIII
1.	INT	RODUC	TION	1
2.			OF PROGRESS IN BIODEGRADABLE PACKAGING FILM : FINDINGS, APPLICATIONS AND GAPS	
	2.1	Overvie	ew of Biodegradable Packaging	4
	2.2	Applica	tion of Biodegradable Films for the Packaging of Fresh Produce	5
	2.3		nent of the Biodegradable and Compostable Packaging Landscape i	
	2.4	Econom	nic Importance of Packaging	10
	2.5	Biodegr	radation of biodegradable films	11
	2.6	Product	ion of Packaging Materials	14
		2.6.1	Casting	14
		2.6.2	Tape casting	14
		2.6.3	Extrusion	15
	2.7	Properti	ies of Packaging Materials	15
		2.7.1	Structural properties	15
		2.7.2	Mechanical properties	16
		2.7.3	Optical properties	19
		2.7.4	Thermal characteristics	19
		2.7.5	Solubility	20
		2.7.6	Physical/barrier properties	20

	2.8	Assessm	nent of Fruit Quality	. 22
		2.8.1	Physical properties	. 23
		2.8.2	Physiological properties	. 24
		2.8.3	Chemical properties	. 25
		2.8.4	Extrinsic factors affecting the quality of tomatoes	. 25
	2.9	Food Pa	ackaging Innovations	. 27
	2.10	Discuss	sion and Summary	. 27
	2.11	Referen	nces	. 30
3.	CHA	ARACTE	ERIZATION OF BIODEGRADABLE PACKAGING MATERIALS	. 43
	3.1	Introdu	ction	. 44
	3.2	Materia	ls and Methods	. 45
		3.2.1	Experimental site	. 45
		3.2.2	Packaging material samples	. 46
		3.2.3	Data collection	. 47
	3.3	Results	and Discussion	. 49
		3.3.1	Solubility properties and water uptake	. 49
		3.3.2	Water vapour permeability	. 50
		3.3.3	Morphology analysis	. 56
		3.3.4	Mechanical properties	.61
		3.3.5	Spectroscopic analysis	. 67
	3.4	Conclus	sion	. 69
	3.5	Referen	nces	.71
4.			ON OF THE EFFECTIVENESS OF BIODEGRADABLE VS ONAL PACKAGING MATERIALS IN SHELF-LIFE EXTENSION	
	OF	ROUND	AND CHERRY TOMATOES	. 74
	4.1	Introdu	ction	.76
	4.2	Materia	Is and Methods	.77

		4.2.1	Tomatoe	s samples	77
		4.2.2	Experime	ental design	77
		4.2.3	Data coll	ection	79
			4.2.3.1	Physiological weight loss	79
			4.2.3.2	Puncture test	79
			4.2.3.3	Kramer shear	79
			4.2.3.4	Compression	80
			4.2.3.5	Colour	80
			4.2.3.6	TSS	80
			4.2.3.7	РН	80
			4.2.3.8	Marketability percentage	80
		4.2.4	Data ana	lysis	
	4.3	Results	and Discu	ssion	
		4.3.1	Colour		
		4.3.2	Engineer	ing properties (Puncture, Kramer, Compression)	
		4.3.3	Physiolo	gical weight loss	
		4.3.4	рН		94
		4.3.5	Total Sol	uble Solids (TSS)	
		4.3.6	Subjectiv	ve analysis and marketability	
	4.4	Conclus	sion		
	4.5	Referen	ices		
5.	COI	NCLUSI	ON AND I	RECOMMENDATIONS	109
	5.1	Referen	ices		113
	5.2	Append	lix		114

## **LIST OF FIGURES**

Figure 2.1	ΓGA plot for pure poly (4-methyl, 2-pentyne) (PMP), fumed silica (FS), and	
	polyoctatrimethyl silsesquixane (POSS) nanoparticles and nanocomposite	
	membranes. POSS decomposed quickly, while FS had a low weight loss at	
	a specific temperature range (Hashemifard et al., 2020)	. 20
Figure 2.2	Comparison of traditional plastic and biodegradable packaging where (a) is	
	Meter-Bi, (b) Nature flex, (c) and (d) are flexible films made from starch	
	and cellulose	. 22
Figure 3.1 (	Complete randomized block. experimental design	. 46
Figure 3.2	Chemical structure of cellulose (de Cuadro et al., 2015)	. 46
Figure 3.3	Chemical structure of sorbitol (de Cuadro et al., 2015)	. 46
Figure 3.4	Solubility of biodegradable packaging materials	. 50
Figure 3.5	Water uptake of biodegradable packaging materials	. 50
Figure 3.6	Weight gained by the desiccant (CaCl <sub>2</sub> ) in the cups covered by packaging	
	films CV% = 1.4	. 53
Figure 3.7 V	WVP (a) and WVTR (b) of biodegradable packaging (stp tray, pulped pt, glued	
	pt), compared to conventional packaging (EPS tray, zipo pnt, ppl)	. 55
Figure 4.1	The experimental design layout	. 78
Figure 4.2(a	a) The effects of conventional materials, (b) biodegradable materials on the	
	colour hue angle. (c) Cherry packages and (d) round packages against	
	storage conditions.	. 83
Figure 4.3(a	a) Variations of cherry and round against storage days. (b)The comparison of	
	cherry and round tomatoes against storage conditions	. 84
Figure 4.5	The effect of the storage conditions on the firmness (Puncture) of cherry and	
	round tomatoes packed in biodegradable (SPT-F, SPT-PV, GPT-F, PPT-Z)	
	and conventional (PPPC, ZP-F, EPT-F, PPPR) packaging materials	. 87
Figure 4.6	The effect of the storage conditions on the firmness (compression) of cherry	
	and round tomatoes packed in biodegradable (SPT-F, SPT-PV, GPT-F,	

PPT-Z) and conventional (PPPC, ZP-F, EPT-F, PPPR) packaging materials.

Figure 4.7	The effect of the storage conditions on the firmness (Kramer shearing) of	
	cherry and round tomatoes packed in biodegradable (SPT-F, SPT-PV, GPT-	
	F, PPT-Z) and conventional (PPPC, ZP-F, EPT-F, PPPR) packaging	
	materials	89

# LIST OF TABLES

Table 2.1	A summary of research on the use of biodegradable films in the extension	
	of the shelf-life of fresh agricultural produce	7
Table 2.2	Examples of various biodegradable packaging materials used for packaging	
	freshly-cut fruit and vegetables	9
Table 2.3	Composting studies of biodegradable polymers under real conditions (after	
	Rudnik, 2019)	3
Table 2.4	Mechanical properties of different polymers	8
Table 3.1	Description of biodegradable packaging materials	5
Table 3.2	The weight gained (g) by CaCl <sub>2</sub> inside the containers covered by different	
	types of packaging materials over seven days	2
Table 3.3	Composition of the quantity of elements found in biodegradable packaging 60	C
Table 3.4	Tensile strength (mpa) of the packaging materials influenced by the ambient	
	and cold storage conditions during the 28-day period	3
Table 3.5	Modulus of elasticity (Nm <sup>-2</sup> ) of the packaging materials influenced by the	
	ambient and cold storage conditions during the 28-day period	4
Table 4.1	Variations of the mean pH of round and cherry tomatoes packed in	
	biodegradable and conventional materials90	6

### LIST OF ABBREVIATIONS AND SYMBOLS

The following is a list of abbreviations, acronyms and symbols that are used throughout this thesis. It gives the acronym, abbreviation or symbol, its meaning and the page where it is used for the first time.

Abbreviation or Symbol	Meaning	Page
PLA	Poly(lactide)	27
PHBV	Poly(hydroxybutyrate – co -hydroxyvalerate)	27
РНА	Polyhydroxyalkanoates	27
FTIR	Fourier transformed infrared	30
SEM	Scanning electron microscopy	30
ASTM	The American society of testing of testing and materials	30
РНВ	Polyhydroxybutyrate	32
LDPE	Low-Density Polythylene	32
PP	Polypropylene	32
TGA	Thermogravimetric Analysis	33
FS	Fumed Silica	34
WVP	Water vapour permeability	35
WVPC	Water vapour permeability coefficient	35
OTR	Oxygen transmission rate	36
O <sub>2</sub>	Oxygen	36
CO <sub>2</sub>	Carbon dioxide	36
TSS	Total Soluble Solids	39
WVTR	Water vapour transmission rate	58
NC	Nano-cellulose	61
RH	Relative humidity	62

EPS	Expanded polystyrene	66
Au	gold	71
ОН	Hydroxyl group	82
EPT-F	Expanded polystyrene covered with flow wrap	89
PWL	Physiological weight loss	94
L*	Lightness colour parameter	95
a*	Redness colour parameter	95
b*	Yellowness colour parameter	95
С	Chroma	95
SPT-P	Stamped paper tray covered with PVC cling wrap	96
SPT-F	Stamped paper tray covered with flow wrap	96
GPT	Glued paper tray covered with flow wrap	97
PPT	Pulped paper tray covered with Zipo PET lid	97
РРР	Polypropylene perforated Plastic	97
ZP	Zipo punnet covered with flow wrap	98
PPPR	Polypropylene perforated plastic Round	98
PPPC	Polypropylene perforated plastic Cherry	98

#### **1. INTRODUCTION**

Petroleum-derived plastic packaging films are widely used in the food industry to preserve and extend the shelf-life of foods. Nevertheless, material scientists and engineers have begun to focus their attention on recyclable and biodegradable food packaging materials, due to the negative environmental impact and excessive use of non-biodegradable plastics (Sanyang *et al.*, 2016). Researchers have been working intensively over the past decade to replace petroleum-based plastics with more environmentally-friendly alternatives for food packaging (Wu *et al.*, 2013; Atarés and Chiralt, 2016; Sanyang *et al.*, 2016). As a result, biopolymers have become an alternative solution for solving the environmental problems caused by waste from conventional packaging materials (Sanyang *et al.*, 2016).

Over the past few decades, the demand for plastic and synthetic polymers for both primary and secondary food packaging has increased rapidly (Zabihzadeh *et al.*, 2019). Recent studies have demonstrated that there are approximately 5 trillion plastic particles found in the surface waters of the earth (Sonam *et al.*, 2019). Waste generation has persistently existed in human life and it has become a crucial global issue (Tulashie *et al.*, 2020). A significant part of municipal solid waste is derived from non-biodegradable polymers and dumped packaging material, which is a major challenge for waste management (Zhong *et al.*, 2019). Some of the waste that is produced is recyclable and can be easily converted into other useful products (Tulashie *et al.*, 2020). Presently, the recycling of plastics from various sources, including disposable products and electronic waste (e-waste) is the focus of much research (Sahajwalla and Gaikwad, 2018).

Africa is one of the continents facing the destructive effects of plastic waste due to the increased consumption per capita, population growth, urbanization, as well as the lack of infrastructure in the management of waste generation (Jambeck *et al.*, 2018). The high percentage of plastic waste comes from the packaging of food products and only a small portion of it is finally recycled (Mistriotis *et al.*, 2016).

Due to the low biodegradability of plastics and them being derived from non-renewable natural resources, petrochemical-based packaging materials are placing considerable pressure on the environment (Chaturbhuj, 2015). This has contributed to a search for packaging polymers to fix the shortcomings of plastics. In an attempt to resolve this problem, sustainable and biodegradable films obtained from biopolymers have been subjected to further testing. Film preparation biopolymers are derived from biomass (gelatin, starch, cellulose, etc.), microbes (polyhydroxyalkanoates) and monomers (polylactic acid) (Khalil *et al.*, 2018). Polysaccharides,

proteins and lipids are biopolymers that form an appropriate alternative, due to their non-toxic and biodegradable characteristics and they are derived from sustainable natural resources (Chaturbhuj, 2015). However, their relatively low mechanical and barrier properties, compared to their non-biodegradable counterparts, are the main limitations in the use of biopolymers as packaging materials (Kang *et al.*, 2010; Chaturbhuj, 2015; Khalil *et al.*, 2018).

Post-harvest handling requirements differ according to the mechanical, thermal, physical and metabolic properties of different fruit (Mukama *et al.*, 2020). The packaging practice affects their precooling, the cold transportation processes, cold storage and the subsequent fruit quality (Mukama *et al.*, 2020). A recent study by Wu *et al.* (2013) revealed that the use of a ventilated packaging design and cold storage plays a key role in preserving the quality of fresh produce. In particular, the intention of packaging fresh fruit is to protect the fruit against mechanical damage and to avert the proliferation and spread of decay-causing micro-organisms (Mukama *et al.*, 2020). It is accepted that packaging is a major food processing operation that is used to contain, protect, preserve, store and distribute food (Fadiji *et al.*, 2019b). The importance of packaging during the entire life-cycle of the product, differs in terms of its environmental impacts. Therefore, companies that manufacture packaging materials have a responsibility to improve their environmental effects (Majid *et al.*, 2018b).

Fresh fruit packaging can be manufactured from various materials, such as wood, paper, glass and plastic. Corrugated fibre-board and reusable plastic containers are the most commonly-used packaging materials for shipping (Berry *et al.*, 2017; Mukama *et al.*, 2020). The primary packaging materials used for most commercial food products, including fresh produce, are paperboard and plastics (Teck Kim *et al.*, 2014). They are used frequently because they are cost effectiveness, light weight and biodegradable (except petroleum-based plastics), and they are also recyclable (Opara and Zou, 2007). In addition, the proper selection of packaging materials is essential for maintaining the quality and freshness of product during distribution and storage, while the shape and form of the packaging are also important, from a marketing perspective (Lalpuria *et al.*, 2012).

The increasing demand for renewable and biodegradable packaging materials has resulted from the extensive use of non-biodegradable and synthetic polymers (Zhong *et al.*, 2019). The additives used in the packaging material can also affect the packed food during the storage period (Zabihzadeh Khajavi *et al.*, 2019). However, there are management solutions for the

existing plastic waste and for reducing the impact of these materials on the environment (Jandas *et al.*, 2019).

A study conducted by Govindan (2018) revealed that the demand for food has increased drastically over the past fifty years and that the world has come to a point where consumption is 30% higher than the capacity of nature. The primary reason for this increase is the world's growing population. The sustainability of supply chains within the food industry has been dramatically affected by population growth and globalization. The way that food is produced, packaged, transported and consumed has an impact on whether its quality is maintained throughout the food supply chain. The latest innovation in the food packaging industry is the use of biodegradable polymers that are reinforced with nanofillers, due to their sustainable appeal, which matches the actual demands of consumers for more environmentally-friendly products (Abdollahi *et al.*, 2012; Zabihzadeh Khajavi *et al.*, 2019).

Bio-based materials are used primarily for packaging fresh agricultural products, such as fruit and vegetables, fresh juices, meat and fat-rich foodstuffs, as they are capable of improving the quality of the food. They have a beneficial impact on these foods, compared to conventional packaging. They are currently used to pack products with a short shelf-life and that require low barrier properties (Majid *et al.*, 2018b). The nutritional value of vegetables lies in their micronutrient, fibre and bioactive phytochemical content (Khalil *et al.*, 2018). Recent publications have examined the role of a biodegradable coatings in the preservation of minimally-processed fruits, strawberries and vegetables, which provide a barrier for the moisture and gases and improve the mechanical properties of the food (Meritaine da *et al.*, 2018).

It is essential for the environment that non-degradable conventional plastics, which are based on fossil fuels, are replaced by sustainable and biodegradable materials. Therefore, this study focuses on the identification and selection of biodegradable materials for the potential development of new and alternative packaging in South Africa. To also determine the physical, mechanical and water vapour permeability properties of innovative biodegradable packaging materials for cherry and round tomatoes.

# 2. A REVIEW OF PROGRESS IN BIODEGRADABLE PACKAGING FILM RESEARCH: FINDINGS, APPLICATIONS AND GAPS

#### 2.1 Overview of Biodegradable Packaging

Packaging plays an essential role in the quality of food products by protecting them from environmental, chemical and physical contaminants (Mekonnen, 2017). Protection can be achieved by providing a barrier against the movement of moisture, oxygen and carbon dioxide. The light-blocking properties of packaging materials prevent a light-catalysed oxidative reaction, which subsequently protects the nutrients and colour in a product from deteriorating (Jin *et al.*, 2016). In addition, the packaging can maintain the desired atmosphere surrounding the product (Risch, 2009).

Traditionally, plastics have been used as packaging materials, due to their good mechanical properties, their low cost, their low permeability to water vapour and their high compatibility with different foods (Assis *et al.*, 2017). Globally, corrugated paperboard packaging is commonly utilised to protect, store and transport products, including food, electronics and horticultural produce, such as fresh fruit and vegetables (Fadiji *et al.*, 2019a). The most extensively-used materials for packaging applications are paper and paperboard (Netramai *et al.*, 2016), and the most commonly-used material in the South African pome industry is corrugated paperboard (Mukama *et al.*, 2020). In the USA, over 90% of the packaging used for fresh fruit is corrugated paperboard (Little and Holmes, 2000).

The global market for biodegradable plastics is expanding, and there has been significant growth in North America, in particular. However, biodegradable plastics have become more expensive, compared to petroleum-based plastics. In the fresh fruit and vegetable packaging sector, there is an increasing demand to replace the petrochemical-based packaging films with more environmentally-friendly biodegradable materials (Tharanathan, 2003; Koide and Shi, 2007).

Although biodegradable films are more expensive than petrochemical materials, under aerobic conditions they will biodegrade to CO<sub>2</sub>, water and biomass, or methane and biomass, under anaerobic conditions (Avella *et al.*, 2005). Based on these features, biodegradable films can make an effective contribution to reducing environmental pollution (Koide and Shi, 2007). The industries that generate the most plastic waste, such as food packaging, are more interested in developing more environmentally-friendly products, like biodegradable films. The key reason

for this shift is to improve the environmental issues and the market demand for more sustainable products (Meritaine da *et al.*, 2018).

A study by Atarés and Chiralt (2019) showed that the application of essential oils to biodegradable food packaging in Spain resulted in natural bio-based products (with antioxidants and antimicrobial properties) providing health benefits. Formulating more lipid-rich essential oils can lower the water vapour permeability of hydrophilic materials, and can enhance their mechanical, optical and structural properties (Chisenga *et al.*, 2020).

Essential oils are an interesting ingredient for biodegradable food packaging, primarily due to their natural origin and functional (antioxidant/antimicrobial) properties, which enables active materials to be obtained, with the view to extending the shelf-life and adding value to the product. The film structure is usually weakened by the addition of oil, while the properties of the water barrier are strengthened and the optical engineering properties of the packaging materials are decreased. Essential oils may give antioxidant and/or antimicrobial properties to the films (Atarés and Chiralt, 2016).

Starches have been selected from among many biopolymers, for the preparation of biodegradable packaging films, for a variety of reasons. Films based on starch provide a number of advantages, including the fact that they are renewable, sustainable, biodegradable, easy to obtain and affordable. Furthermore, they exhibit similar physical properties to conventional packaging plastics, in terms of their optical properties, lack of odour and tastelessness (Xiong *et al.*, 2013; Atarés and Chiralt, 2016; Sanyang *et al.*, 2016). Starch and starch-based materials are being increasingly used, because they are readily available, biodegradable, cheap, have desirable properties and are viewed as a promising alternative for synthetic polymers (Wu *et al.*, 2013; Zhou *et al.*, 2019).

#### 2.2 Application of Biodegradable Films for the Packaging of Fresh Produce

A large number of micronutrients are present in vegetables, mostly vitamins and minerals, which are required by humans in small amounts to orchestrate a variety of physiological functions. Recent publications have examined the role of a biodegradable coating in the preservation of minimally-processed fruits, strawberries and vegetables, by providing a barrier against moisture and gases and improving the mechanical properties of the food (Meritaine da *et al.*, 2018).

Kantola and Helen (2001) conducted a three-week study on the quality changes of organic tomatoes stored in four biodegradable packages and in low-density polyethylene (LDPE) bags.

Organic tomatoes were stored at  $(11\pm^{\circ}C)$  and a RH of 75-85.5%. The analysis pertained to their weight loss, moisture content, colour, firmness and flavor. The results showed that the type of package had no significant influence on the sensory quality of tomatoes; however, the storage time significantly (p< 0.05) affected their colour and firmness. Tomatoes stored in biodegradable packages lost more weight than those stored in LDPE packages. In addition, the findings suggested that biodegradable films with a good permeability coefficient could be advantageous in the prevention of contamination from micro-organisms, compared to conventional packaging materials (Muratore *et al.*, 2005).

Sliced broccoli, tomatoes, sweetcorn and blueberries have also been found to be successfully stored in biodegradable pulp trays wrapped in caprolactone-poured foil packaging (Kumar *et al.*, 2020). There are several successful examples of biopolymer-based packaging films for extending the shelf-life of freshly-cut fruit and vegetables, as illustrated in Table 2.2. The results of a study conducted by Del Nobile *et al.* (2008) indicated that the barrier properties of the investigated films determine the oxygen concentration in the package headspace and control the rate of all the detrimental phenomena responsible for packed lettuce becoming unacceptable. Table 2.1 demonstrates the findings of the studies conducted by various researchers on the extension of the shelf-life of fresh fruit and vegetables, using biodegradable films.

Studies carried out in Finland developed and tested biodegradable packaging films for the preservation of tomatoes, which resulted in an extended shelf-life. Using starch edible coatings made from Colombian native potatoes, Andean blueberries (a wild fruit native to South America) were preserved for longer periods, with a 27% reduced respiration rate (Medina-Jaramillo *et al.*, 2019). Studies carried out by Ethiopian researchers tested a pectin-chitosan film on tomatoes and found that they had a shelf-life of 15-17 days, compared to the 10-day shelf-life of the control. In addition, there has also been huge interest in the production of biodegradable plastic film in Nigeria, by blending cassava starch and biodegradable polymer materials (Chisenga *et al.*, 2020). In eastern, central and southern African countries, the post-harvest losses of fresh tomatoes were reported to be between 9.50-10.04%, with Kenya, South Africa and Nigeria recording losses of 10.10-13.40%, respectively (Sibomana *et al.*, 2016). Nonetheless, commercial and emerging tomato farmers in the South Africa's supply chain have minimized their post-harvest losses by using recyclable cardboard boxes of various sizes, bulk bins, plastic crates and wooden crates (Chisenga *et al.*, 2020).

 Table 2.1
 A summary of research on the use of biodegradable films in the extension of the shelf-life of fresh agricultural produce

Biodegradable Film	Research findings	Reference
Master-Bi bag (based on starch)	The quality of tomatoes stored in low-density polyethylene bags for three weeks was similar to that of tomatoes packed in biodegradable bags.	(Kantola and Helen, 2001)
Yam starch and glycerol	Strawberry samples packed in starch film lasted 21 days and fresh strawberries 14 days.	(Mali <i>et al.</i> , 2003)
Banana starch and chitosan	Created a composite bag to protect Chinese cabbage, asparagus and baby corn from Staphylococcus aureus.	(Pitak <i>et al</i> ., 2011)
Chitosan, methyl cellulose and vanilin	On Day Six, the microbial population of freshly-cut pineapples decreased by four logs	(Sangsuwan <i>et al.</i> , 2008)
Carboxymethyl cellulose containing potassium sorbate	Fresh pistachios tested with sorbate showed strong activity against the Aspergillus species	(Sayanjali <i>et al.</i> , 2011)
Methylcellusoe and polycarprolactone/ alginate films incorporated with antimicrobial agents	On broccoli florets stored at 4°C, films inhibited Salmonella typhimurium growth for 12 days and L. monocytogenes and E.coli growth for four days	(Takala <i>et al.</i> , 2013)
Polylactic acid with Allium extract	It was reported that the developed films were effective for storing salads for up to five days at 4°C	(Llana-Ruiz- Cabello <i>et al.</i> , 2015)

Wheat gluten	Extend shelf-life of strawberries by	(Tanada-Palmu et
	12 days at 7–10°C	al., 2005)
Zein films plasticized with	Increase in the shelf-life of fresh	(Rakotonirainy et
oleic acid	broccoli florets by six days	al., 2001)
Apple puree with fatty acids,	During 12 days at 5°C, freshly-cut	(McHugh and
fatty alcohols, beeswax and	apples lost less moisture and	Senesi, 2000)
vegetable oil	browned less	

 
 Table 2.2
 Examples of various biodegradable packaging materials used for packaging
 freshly-cut fruit and vegetables

Biopolymers	Packed product image	References
Zein and polycaprolactone	Carrots	(Mensitieri et al., 2011)
Polyostor-based biodegradab	la Lettuce	(Del Nobile $at al 2008$ )

Polyester-based biodegradable Lettuce

films



(Del Nobile *et al.*, 2008)

Guar gum, beeswax, grape pomace and nano-clay	Pomegranate	(Chaturbhuj, 2015; Khalil <i>et al.</i> , 2018)
Oriented poly(lactide)	Mango	(Chonhenchob <i>et al.</i> , 2007;
		Khalil <i>et al.</i> , 2018)

## 2.3 Assessment of the Biodegradable and Compostable Packaging Landscape in South Africa

Various factors require consideration when incorporating biodegradable and compostable packaging into the South African consumer packaging waste landscape. Over 34% of all households do not have access to regular waste removal because the formal municipal waste collection is not effective in capturing all their post-consumer waste (Godfrey *et al.*, 2016).

Composting facilities, with different infrastructures and technologies, can be found all over South Africa. However, plastic pollution poses a significant problem for the majority of composters. It is important to note that composters, unlike recyclers, do not charge for the waste entering their facilities, except for a gate fee that is based on the composition of the material (Verster and Bouwman, 2020). Furthermore, like lightweight flexible and multi-layer items, informal pickers actually have no economic incentive to collect biodegradable or compostable packaging for processing, so they are likely to be ignored. Therefore, due to poor labelling and a lack of clear identification, these items, especially carrier bags, are being collected inadvertently, together with conventional plastic bags (Godfrey *et al.*, 2016).

The consumer demand for environmentally-friendly alternatives to conventional plastics has fueled the creation of biodegradable and compostable materials in South Africa. Consumers are currently not adequately educated about biodegradable and compostable plastics. In addition, most brand owners, retailers and consumers are unaware of the different types of materials and their properties, as well as the fact that some materials can only be certified to degrade or compost under specific, controlled conditions (O'Brien and Thondhlana, 2019b).

Biodegradable and compostable materials are being widely adopted in South Africa, primarily in response to the consumer demand for a better environmental performance, rather than using traditional plastics (Verster and Bouwman, 2020). At this stage, the applications include food and drink containers, utensils and carrier bags for niche markets. In spite of the relatively low volumes at present, the post-consumer management of these products needs to be considered proactively, as it is anticipated that this market will grow (Godfrey *et al.*, 2016; Verster and Bouwman, 2020).

#### 2.4 Economic Importance of Packaging

The packaging industry plays an active role in the world economy (Reichel et al., 2016). The idea of a circular economy is to counteract and decrease the impact of harmful packaging materials on the environment (Korhonen et al., 2018). This concept was developed in response

to the decline in ecosystems and the global environmental changes (Geueke et al., 2018). A circular economy promotes closing the loops in industrial systems, minimizing waste and reducing the raw materials and energy inputs (Reichel et al., 2016). Developing eco-design and waste prevention programs and prolonging the shelf-life of products are among the practical solutions of a circular economy (Stahel, 2016)). The United Nations Sustainable Development Goal 12 for responsible consumption and production, highlights that the increased resource efficiency decreases food wastage (Schmidt et al., 2019). The packaging industry represents 2% of the Gross Domestic Product (GDP) of South Africa. The growing demand for natural, minimally-processed and nutritious fresh foods and convenience products, and the globalisation of the food trade, has created major challenges for the food packaging industry (Tumwesigye et al., 2017). The continuous development of food packaging can be ascribed to changes in consumer demand and advances in science and technology (Mihindukulasuriya and Lim, 2014). Packaging is applied for the preservation of raw materials and for the final food products (Kadam et al., 2017). Besides, the increased awareness of a healthy diet and quality maintenance in the distribution chains has led to growth and advances in food-processing technologies. Thus, packaging is an indispensable technology for food processing, especially for the safe handling and supply of fresh products, like fruit and vegetables (Tumwesigye et al., 2017).

#### 2.5 Biodegradation of biodegradable films

Biodegradation refers to the mechanism by which micro-organisms work to break down organic matter into its essential raw materials (Rudnik, 2019). Micro-organisms in the soil can degrade biodegradable materials into natural compounds, such as water, carbon dioxide and methane, as well as monomers like amines, alcohols and carboxylates (Chisenga *et al.*, 2020). Biodegradability is influenced by the chemical composition, the nature of bonding and the availability of water (Bhatnagar *et al.*, 2018). Plant-derived metabolites, which are used as substrates by the microbial cells, promote saprophytic growth. Several amylases and cellulases are secreted by microbes, which break the starch and cellulose glycosidic bonds enzymatically and oxidatively. Esterase, cutinase and lipase are extracellular enzymes that hydrolyze the aliphatic ester links in plasticizing films (Tampau *et al.*, 2020). Currently research demonstrates that compostable containers can be composted in systems that manage waste from yards and manure, as well as in those that manage food waste. Therefore, more options for composting biodegradable polymers could be available if these composting facilities approved the

compostable polymers used in packaging applications (Rudnik, 2019). Table 2.3 summarises the methods for biodegradation of different polymers under real conditions.

Polymer	Type of Package	Composting conditions			Results
		Temperature/Moisture	Time	Compost source	
Starch-based	Bags	>60°C (first five days),	six	Green wastes	All the entire strips of plastics have completely disappeared, after one
(MaterBi)		63.1% moisture	weeks		week of monitoring.
Poly(lactide)	Containers	T>55°C, > 65% RH	30	Cow manure, wood	The degradation time of PLA containers was less than 30 days
(PLA)			days	shavings and waste	
				feed	
Poly(hydrox	Films	35–75°C (ISO 16929	12	Fresh apple	After 12 weeks, there were no residual PHBV film fragments found on
ybutyrate-co-		temperature variable) After	weeks	pericarp, wood,	the sieve (2 mm pore size) used to screen the compost. Therefore,
hydroxyvaler		three days, the temperature		scrap, rabbit	according to ISO16929, the degree of biodegradation of PHBV films was
ate) (PHBV)		reached 73°C, and then		feedstuff, cabbage,	100%.
		diminished to around 60°C		distilled water and	
		after four days. The duration		mature compost	
		during which the temperature			
		exceeded 40°C, was about 36			
		days			
Polyhydroxy	Bags	55-77°C	180	In-vessel food-waste	Material completely degraded after 180 days
alkanoates			days	Compost Facility	
(PHA) mirel					

# Table 2.3Composting studies of biodegradable polymers under real conditions (after Rudnik, 2019)

#### 2.6 Production of Packaging Materials

Food-packaging films can be produced by lamination, casting, injection molding, blow molding, thermoforming co-extrusion and coating processes from the raw plastic polymer, biopolymer and biodegradable materials (Gürler *et al.*, 2020). The food-packaging film is extracted from biopolymers, including gelatine, starch cellulose and bio-derived monomers, such as polylactic acid (Majid *et al.*, 2018b). The supplementation of different kinds of additives is recommended to improve the properties of biodegradable films.

#### 2.6.1 Casting

The casting process consists of spreading a film-forming solution or suspension on a small plexiglass or plate (e.g. petri dishes). The film-forming solution is then cast out from the surface onto a thin sheet, where it is dried and peeled (de Moraes *et al.*, 2013). The film thickness is determined by the mass of the suspension that is poured onto the plate (Tzia *et al.*, 2015; Meritaine da *et al.*, 2018). The casting method is widely used for laboratory-scale film making. The drying conditions in this method will vary from room temperature to forced air ovens at temperatures of 30-40°C.

Many of the studies on plastics films have focused on the polysaccharides and proteins that are used in the casting method (Rocha *et al.*, 2013; Meritaine da *et al.*, 2018). However, this method has two drawbacks, namely, the difficulty in scaling up the output to mass production, as well as the long drying times. These shortcomings render this technique unworkable on an industrial scale (Meritaine da *et al.*, 2018).

#### 2.6.2 Tape casting

Tape casting has been documented as an effective method for the manufacture of electronic equipment. This method can also be used to manufacture biodegradable films (Oliveira de Moraes *et al.*, 2015). The technique of tape-casting is also known as spread casting, or knife-coating, and it is well-known in the plastic, ceramic, paper and paint industries (Susarla *et al.*, 2013). This technique allows the spread of a film-forming suspension on broad supports and on continuous holding belts and allows the regulation of the film thickness by using an adjustable blade at the bottom of the spreading unit, called a doctor blade (Oliveira de Moraes *et al.*, 2015; Meritaine da *et al.*, 2018). The results indicated that both the thickness of the suspension and the drying temperature had a major effect on the drying rate and on the properties of the film. To obtain homogeneous films, the suspensions should be dispersed with doctor blade gaps of

3-4 mm. For films with acceptable properties, the optimum drying temperature is 60°C (Oliveira de Moraes *et al.*, 2015).

#### 2.6.3 Extrusion

Polymers that are derived naturally, such as starch, cellulose, proteins, etc., can produce biodegradable packaging materials (Khan *et al.*, 2017). Starch is an inexpensive, sustainable and abundantly-available biopolymer; however, due to intermolecular forces and hydrogen bonds, it cannot be easily extracted as a thermoplastic material (Khan *et al.*, 2017). Extrusion is a continuous process that often constitutes the first key step in plastics processing, due to its capacity to converting resins from a solid to a molten shape (Nesic *et al.*, 2020). Starch granules undergo numerous and complicated phase changes during extrusion processing, including starch swelling, fusion and solubilization (Jiang *et al.*, 2020).

Extrusion is one of the most popular techniques used to process polymeric materials, and it primarily involves melting-solidification (Jiang *et al.*, 2020). Although extruding machines were originally designed to be used in the traditional processing techniques for synthetic materials, it has been widely proven that they can also work for biomaterial- and biopolymer-processing (Nesic *et al.*, 2020). Recent packaging extrusion studies include blends of thermoplastic starch and low-density polyethylene, with a 40%-80% concentration of thermoplastic starch (TPS) (Mazerolles *et al.*, 2019; Nesic *et al.*, 2020). Blown-film extrusion is one of the most commonly-used approaches for producing films in industry (Nesic *et al.*, 2020).

#### 2.7 Properties of Packaging Materials

A knowledge of the properties of the packaging materials is essential, as they can predict the shelf-life of the product and packaging. Small molecules of vapour, gases, water and other liquids will usually be pushed in and out of the package wall, thus adversely affecting the consistency of the product and its shelf-life (Majid *et al.*, 2018b). Often, light penetrates the packaging materials and the oxidation reaction is accelerated, resulting in a reduction in the product's shelf-life (Majid *et al.*, 2018b). The most important properties of biopolymers are discussed, in detail, under the following subheadings:

#### 2.7.1 Structural properties

#### Spectroscopic analysis

Infrared spectroscopy is presently one of the most important analytical techniques available to scientists for polymer characterization. One of the greatest advantages of infrared spectroscopy is that virtually any sample, in any state, may be analysed (Hu *et al.*, 2018). For example, liquids, solutions, pastes, powders, films, fibres, gases, solids and surfaces can all be examined by using a judicious choice of sampling techniques (Smith, 2011). Fourier Transformed Infrared (FTIR) can provide researchers with further information on the super molecular structure and it can be used to determine the chemical composition of the native natural and modified natural fibres (Smith, 2011; Fan *et al.*, 2012; Hu *et al.*, 2018). In addition, it offers scientists an excellent range of solutions for understanding natural fibres and their related modification technologies and products, such as their chemical composition and microstructure (Fan *et al.*, 2012).

FTIR can examine the nature of molecular chains, their crystallinity and their correlation with various bonds. The chemical composition at microscopic level determines the ability to perform various functions that are useful when a package is made out of fibres. FTIR has been mostly successful in the accurate analysis of both (cellulose, hemicellulose and lignin) and the composition, interfaces and hence the properties of, natural fibres (Kruer-Zerhusen *et al.*, 2018). Change in the chemical composition, interface, and hence the properties of natural fibres and composites, could also be effectively identified by using FTIR (Hu *et al.*, 2018).

#### Surface morphology

Sahi *et al.* (2021) studied the properties of plasticised cornflour-filled low-density polyethylene composites for food-packaging applications. The morphological test was carried out by using a Scanning Electron Microscope (SEM), which is an indispensable tool for the characterization of materials, from a nanometer to a micrometer scale (Jin *et al.*, 2016; Goldstein *et al.*, 2017). It is one of the most versatile instruments available for the examination and analysis of the microstructure morphology and chemical composition (Goldstein *et al.*, 2017). An SEM image reflects the surface structure. Due to the very narrow electron beam, SEM micrographs have a great depth of field and can obtain a characteristic three-dimensional appearance, which is useful for understanding the surface structure of biopolymers and other packaging materials (Jin *et al.*, 2016).

#### 2.7.2 Mechanical properties

The mechanical properties of films represent their ability to maintain their integrity and endure external stress during the processing, transportation, handling and storage of packaged materials. Sufficient mechanical strength and extensibility are generally needed for their use in food-packaging applications (Zhou *et al.*, 2019). The mechanical properties of biopolymer films include their tensile strength, elongation, deformability and elastic modulus. They are extremely essential, since packaging materials have sufficient mechanical resistance to preserve the quality of packed foods during their handling, transportation, storage and marketing (Meritaine da *et al.*, 2018). The mechanical properties of packaging materials are comprised of the Young's modulus, tensile strength, puncture strength and split elongation, as shown in Table 2.2 (Ivonkovic *et al.*, 2017). The American Society of Testing and Materials (ASTM) D882-02 standard test method for the tensile properties of thin plastic sheeting is the most commonly-used technique for evaluating the mechanical characteristics of various forms of biodegradable films (Khalil *et al.*, 2018).

The greater the value of stress, or modulus, results in a greater resistance to the deformation or rigidity of a material (Kamdem *et al.*, 2019). Young's modulus is a good indicator of the rigidity of a material, while the strain or elongation is more related to flexibility (Kamdem *et al.*, 2019). The mechanical properties allow the prediction of film behaviour during the transport, handling and storage of packaged foods (Siracusa *et al.*, 2008).

 Table 2.4
 Mechanical properties of different polymers

Polymers	Melting	Glass	Youngs	Tensile	Elongation at break	References
	point	transition	Model of	strength	(%)	
	<i>Tm</i> °C	temperature	elasticity	(KN/ <i>mm</i> <sup>2</sup> )		
		<b>T</b> <sub><i>g</i></sub> °C	$(KN/mm^2)$			
Starch	110-115		0.6-0.85	35-80	580-820	(Ivonkovic et al., 2017)
PLA	130-180	40-70	3.5	48-53	3-25	(Ilyas <i>et al.</i> , 2020)
РНА	70-70	-30 to 10	0.7-1.8	18-24	3-25	(Ilyas <i>et al.</i> , 2020)
PHBV	145	1	1.2	20	50	(Siracusa <i>et al.</i> , 2008)
РНВ	140-180	0	3.5	43	5	(Ivonkovic et al., 2017)
LDPE	110	-30	0.2	10	620	(dos Santos <i>et al.</i> , 2013)
PP	176	0	1.7	3.8	400	(dos Santos et al., 2013)

Polylactide (PLA) is a polyester derived from a renewable biomass, such as fermented plant starch, Polyhydroxyalkanoate (PHA), a plant-based material produced by microbial fermentation of carbon-based feedstocks, Poly(hydroxybutyrate-co-valerate) (PHBV) is produced by microbial fermentation, Polyhydroxybutyrate (PHB) is produced by carbon sources, and Low-Density Polyethylene (LDPE) is made from the monomer ethylene, Polypropylene (PP).

#### 2.7.3 Optical properties

The term 'optical properties' refers to the response of food to electromagnetic radiation, and particularly, to visible light (Berk, 2013). Light energy in the ultraviolet and visible light regions plays a critical role in the overall food quality, which leads to various degradation and oxidation reactions. Food degradation and oxidation result in the destruction of nutrients and bioactive compounds, the formation of bad odours and flavours, the loss of food colour and the formation of toxic substances (Jafarzadeh *et al.*, 2021). Food compounds are sensitive to various light wavelengths (Duncan and Chang, 2012). The optical barrier property is a key attribute that influences the convenience, presentation and marketability of the films for different applications. Films with a lower UV light transmission value have a better UV penetration barrier through the film. Opacity measures how much an object can absorb light (Suderman *et al.*, 2018).

Optical properties, like transparency and gloss, primarily affect the aesthetics of a product. If the product has an attractive appearance and its light exposure is stable, the use of a very clear packaging material may be ideal (Berk, 2013). Totally amorphous polymers are transparent, as the degree of crystallinity increases, and as the clarity diminishes, the materials become more hazy and, ultimately, opaque (Emblem, 2012).

#### 2.7.4 Thermal characteristics

A Thermogravimetric Analysis (TGA) is a thermal analysis that monitors the sample mass against time or temperature in a controlled environmental furnace. The sample can be analysed as a crescent or decreasing (differential) temperature at a constant rate, or as an isothermal temperature (Tomoda *et al.*, 2020). Thermogravimetry is a process for determining the material weight with respect to a combination of the temperature and time increase. The TGA is a commonly-used instrument that is based on this process to investigate the thermal characteristics of a substance, under various heating environments, and it is applicable for predicting the temperature tolerance of the structural integrity of the packaging materials that are produced. A TGA analysis is a method in which the mass of a sample is measured over time, with changes in temperature in a specified trend. This measurement provides information about the physical phenomena, such as the mass changes, temperature stability, oxidation/reduction behaviour, decomposition, corrosion studies and compositional analysis (Hashemifard *et al.*, 2020). Hashemifred *et al.* (2020) used a TGA analysis to investigate the thermal

degradation of a membrane occurs at the single stage and that polyoctatrimethyl silsesquixane (POSS) decomposed quickly, while Fumed Silica (FS) had a low weight loss at a specific temperature range. Therefore, FS had better thermal stability effects, compared to those of POSS. The temperature of degradation is obtained via the breakthrough point of the curves. Figure 2.1 shows a TGA plot for the pure and nanocomposites of poly(4-methyl, 2-pentyne) (PMP).

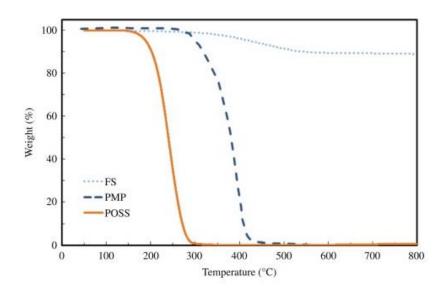


Figure 2.1 TGA plot for pure poly (4-methyl, 2-pentyne) (PMP), fumed silica (FS), and polyoctatrimethyl silsesquixane (POSS) nanoparticles and nanocomposite membranes. POSS decomposed quickly, while FS had a low weight loss at a specific temperature range (Hashemifard *et al.*, 2020)

#### 2.7.5 Solubility

Solubility is an essential property in the application of biodegradable and edible films. It guides the application of films for food packaging. A higher solubility means a lower water resistance. However, the high solubility of films can be an important factor when determining their biodegradability, when they are used as a packaging wrap material (Gürler *et al.*, 2020). The solubility of a film in water directly affects its moisture barrier properties, and the increased water content improves its elongation properties and decreases the tensile strength and elastic modulus (Meritaine da *et al.*, 2018)

#### 2.7.6 Physical/barrier properties

#### **Permeability properties**

An important parameter in food packaging materials is Water Vapour Permeability (WVP), which should be kept as low as possible. It is necessary to frequently restrict the moisture transfer between the food and the surrounding atmosphere (Gürler *et al.*, 2020). The moisture equilibrium is one of the major factors responsible for the physical and/or chemical deterioration and dehydration of packaged agricultural fresh produce. Thus, the characteristic water vapour barrier of food packaging films is of great importance for maintaining or extending the shelf-life of fruit and vegetables (Zhou *et al.*, 2019). The water vapour barrier is measured by the Water Vapour Coefficients (WVPC), which define the amount of water vapour that transmits per unit area of packaging material (kg mm<sup>-2</sup>s<sup>-1</sup>pa<sup>-1</sup>). The water vapour permeability of a biopolymer film is usually determined by the ASTM E 96-95 standard (Khalil *et al.*, 2018) and it is an important property in the application of films. Furthermore, the moisture content in food has a great effect on its quality, so it is necessary to select a packaging material with appropriate moisture permeability for maintaining the quality of food during storage (Nouraddini *et al.*, 2018).

#### **Gaseous permeability**

The poor barrier properties (especially humidity resistance) of the traditional and most widelyused biomaterials (paper, cellulose films and cellophane) are all known and, therefore, it is important to mix these materials with synthetic polymers, in order to achieve the required barrier properties for the packaging of many foodstuffs (Ivonkovic *et al.*, 2017). A specific environment is needed for the gas and oxygen barrier properties during storage, in order to preserve the quality and freshness of many food products. To this end, a certain gas mixture, consisting mainly of carbon dioxide (CO<sub>2</sub>), oxygen and nitrogen, or a combination of the three, is integrated into the packaging, in order to ensure optimal quality and safety (Majid *et al.*, 2018b). The studies in Figure 2.2 compare the barrier properties of biodegradable polymers that are derived from traditional mineral oil packaging materials.



Figure 2.2 Comparison of traditional plastic and biodegradable packaging where (a) is Meter-Bi, (b) Nature flex, (c) and (d) are flexible films made from starch and cellulose

Biopolymers have been observed to be fairly resistant to oxygen transmission and many efforts are currently being made to change the biopolymer barrier properties. The Oxygen Transmission Rate (OTR) in bio-based packaging is, however, higher than the traditionallyused polymers and can increase the oxidation reactions and reduce the product's shelf-life (Majid *et al.*, 2018b). Polymers are usually fairly permeable to small molecules, such as  $O_2$ ,  $CO_2$ , water and chemical vapours (Khalil *et al.*, 2018). Limited oxygen diffusion and migration within packaging films is desirable, because the presence of oxygen is usually associated with the oxidation of food, which leads to flavour changes, odour development and nutrient losses (Zhou *et al.*, 2019). The standard test method ASTM D3985-05 is done by using a coulometric sensor to establish the transmission rate of  $O_2$  through the plastic film and sheeting. The ASTM F2476-05 test method is used with an infrared detector to evaluate the rate of  $CO_2$  transmission through the barrier materials (Saurabh *et al.*, 2013; Khalil *et al.*, 2018).

### 2.8 Assessment of Fruit Quality

Vegetables and fruit are known as nutritious food products. However, because these products are considerably perishable, they cannot be consumed if they are not properly managed after harvesting. Moreover, fresh horticultural products have become critical in international trade, since the globalization of commerce. The high moisture content, nutrient richness and active metabolism of fresh fruit and vegetables render them vulnerable to dehydration, mechanical damage and environmental stress (Jafarzadeh *et al.*, 2021), which result in them losing a

substantial amount of shelf-life after harvest. The quality of tomatoes is evaluated by using a variety of parameters, including their physical, chemical, biochemical, microbiological and sensory properties. Tomatoes of high quality should have an appealing appearance (colour and shape), a desirable texture and an adequate aroma and flavor. Fruit firmness is an important quality indicator that directly affects the post-harvest quality of tomatoes, while soluble solids and acidity influence their flavour (Huang *et al.*, 2018; Liu *et al.*, 2019). Food quality attributes play a major role in influencing the consumer preference for a specific product. It is vital to consider several factors, in order to determine whether tomatoes are of a good or bad quality.

### 2.8.1 Physical properties

### **Textural mechanical properties**

One of the most important internal quality attributes of tomatoes for consumers is their texture. For the genetic improvement, quality control and post-harvest handling and processing of tomatoes, it is necessary to measure these textural properties (Sirisomboon *et al.*, 2012b). From freshly-harvested to overripe tomatoes, the firmness value decreases and the market acceptable level decreases (Batu, 2004). The two main forces encountered during tomato handling after harvest are compression and puncture. Generally-speaking, a compressed fruit experiences a compression force, whereas a punctured fruit experiences a puncture force. Bruising and damage are caused by excessive compression and shearing (Sirisomboon *et al.*, 2012b).

The firmness of a fruit is one of the most important quality attributes that influences consumer acceptance (Zhang *et al.*, 2010). It is usually used as a measure or gauge of its eating quality, its storability and its reaction resistance during post-harvest handling, storage and marketing (Liu *et al.*, 2019). Consumers evaluate its firmness by using a finger test. The firmness of tomatoes is determined by pectin, which is the cementing layer in most fruit and vegetables (Sirisomboon *et al.*, 2012b). Tomatoes lose their firmness due to tissue softening, which is caused by weight loss and enzymatic activity. Pinheiro *et al.* (2013) and Cherono *et al.* (2018) have stated that the kinetics of tomato firmness follows the Arrhenius fractional conversion kinetic model. The reduction in firmness in those stored at lower temperature conditions showed exponential, first-order kinetics. The Kramer shear test is used to define product quality and acceptability. Physical and Mechanical properties are needed for texture analysis and better understanding of product quality. A mechanical property (Kramer shear, puncture and compression) of horticulture products is frequently used to determine their maturity and

ripeness, which is important in handling, storing and processing procedures (Zhang *et al.*, 2010) . Several enzymes, most notably pectin methylesterase and polygalacturonase, are involved in degrading the pectin. As a result of this degradation, the tomato tissue exhibits an evident softening (Vu *et al.*, 2004).

Firmness has been used as an indicator of the quality of tomatoes, and it may be the final indicator by which consumers determine whether to buy a certain batch of tomatoes or not. A study conducted by Batu (2004) showed that the acceptable firmness of all 100% marketable tomatoes should have firmness values of above 1.45 N; however, for home use, the Instron value of tomatoes should be greater than 1.28 N. Mekonnen (2017) conducted a study by using four different packaging materials (an open box, an open market bag, an Xtend bag and a sealed box) stored under cold and ambient temperature conditions. The findings showed that the tomatoes packed in the Xtender and open market bags (stored at 4°C) were firmer (42.67 N), compared to the samples packed in other packaging, which ranged between 32.56-40.50 N. The results also showed that the firmness of tomatoes occurred more slowly at 4°C than at 17°C over a 10-day storage period.

# Colour

The colour of tomatoes is another quality parameter that consumers consider when they buy the fruit; it is used as a measure of their total quality. Thus, their colour must be maintained, in order to prevent losing consumer confidence in the product. Tomatoes accumulate carotenoids during ripening, due to the breakdown of chlorophyll. During the lag phase preceding maturation, chloroplasts are transformed into chromoplasts (Pataro *et al.*, 2015). Instrumental methods, such as the Tristimulus colourimeter, as well as subjective methods and colour charts, can be used to evaluate their colour. Instrumental methods are more commonly used since they are more accurate (Mekonnen, 2017). Tomatoes undergo three main colour changes during development, namely: green, orange and red. The green colour (with a high chlorophyll content) is degraded to accumulate carotenoids, principally beta-carotene (orange colour) and lycopene (red colour). Fresh tomatoes are mostly judged by their appearance (their colour, visual aspects, size and shape), as well as their firmness, taste and nutritional value (Pinheiro *et al.*, 2013).

# 2.8.2 Physiological properties

### Weight loss

Weight loss is generally considered to be a physiological phenomenon that is associated with post-harvest shriveling and it results in the softening of the fruit tissue (Pinheiro *et al.*, 2013).

It has been found that the weight loss of fruit is influenced by pre- and post-harvest factors, such as the date of harvest and the temperature during storage (Alia-Tejacal *et al.*, 2007; Pinheiro *et al.*, 2013). In order to prevent tomatoes from dehydration, weight loss and mechanical damage, it is essential to provide them with good shade during the harvest and proper packaging during storage (Mekonnen, 2017). As fruit continue to lose water through transpiration, they soften, shrink and fade, which contributes to the loss in their physiological mass (Pinheiro *et al.*, 2013).

# 2.8.3 Chemical properties

# **Total soluble solids**

Tomatoes contain various compounds that are soluble in water, including sugars, acids, vitamin C, amino acids and some pectin (Mekonnen, 2017). The soluble solids of fruit are comprised of these soluble compounds. Sugar is the main component of soluble solids in most ripe fruits, including tomatoes (Shezi, 2016). In the screening of new tomato cultivars, Total Soluble Solids (TSS) is a key post-harvest quality parameter (Majidi *et al.*, 2011). For the most common round tomatoes varieties, the TSS ranges from 3.2 to 5.9° Brix (Flores *et al.*, 2017). The soluble solids content of fruit can be used as an index of its maturity or ripeness, since the TSS or sugar increases as it ripens. Refractometers are used to measure the total soluble solid content of fruit (Mekonnen, 2017).

# PH

The pH of tomatoes is primarily defined by the amount of acid present in the fruit. Moreover, the acidity of the fruit contributes to the essential flavor attributes of tomato products (Anthon *et al.*, 2011). They are known to be one of the most highly-acidic fruits, with a pH range of 4 - 4.5 (Cheema and Sommerhalter, 2015). A pH meter is used to measure both the citric acid and other acids, such as malic acid in tomatoes. Citric acid is measured as the total acidity by a pH meter, and these results are reliable, since it is the dominant organic acid in tomatoes (Shahnawaz *et al.*, 2011; Shezi, 2016).

### **2.8.4** Extrinsic factors affecting the quality of tomatoes

Several factors affect the quality of tomatoes after harvest, a number of which have been extensively discussed in various studies (Risse *et al.*, 1985; Pataro *et al.*, 2015; Shezi, 2016). This section presents how the packaging and storage temperature affect their quality.

# Packaging

Packaging has been used to extend the storage life of many fresh fruit and vegetables by the inhibiting their physiological deterioration and reducing their weight loss (Risse et al., 1985). The effectiveness of the packaging material in extending the shelf-life of food will depend on the properties of the packaging (Dandago et al., 2017; Haile and Safawo, 2018). Shahnawaz et al. (2011) investigated the quality attributes of tomatoes stored in different wrapping materials (polyethylene, grease free paper and newspaper), while unwrapped tomatoes served as the control. The samples were stored at an ambient temperature of  $32 \pm 2^{\circ}$ C. The study reported that all the packaging materials prevented a significant loss of weight, compared to that of unwrapped tomatoes. Furthermore, tomatoes packaged in polyethylene bags were reported to be of a better quality and to have a longer shelf-life after 28 days, compared to the other wrapping materials (Shahnawaz et al., 2011). Other tomato studies also found that the weight loss of wrapped tomatoes was significantly decreased and that they were firmer than unwrapped tomatoes (Mekonnen, 2017). A study was conducted by Haile and Safawo (2018) to evaluate the effect of the packaging material on the shelf-life and quality of tomatoes at Samara, in northeastern Ethiopia. The findings revealed that the packaging had a significant effect on their physiological weight loss, decay percentage, colour, overall acceptability and marketability. Different packaging materials are used in the retail marketing of fresh tomatoes, and the properties of these packaging materials need to be evaluated to determine their suitability for individual tomato cultivars (Mekonnen, 2017). Packaging materials controlling the rate at which small molecular weight compounds permeate into, or out of, the package are able to extend the shelf-life of packed food (Muratore et al., 2005; Mekonnen, 2017). In a study by Muratore et al. (2005), which investigated the influence of biodegradable film on the quality and decay of plum tomatoes, it was reported that the use of packaging films with high barrier properties accelerates the quality kinetic decay of the tomatoes. It was also observed that the use of biodegradable films with an appropriate permeability coefficient was an effective method for preventing the contamination of tomatoes from both micro-organisms and insects, without reducing their shelf-life.

#### **Storage temperature**

Fresh produce spoilage can be influenced by several factors, including the temperature conditions in the storage environment. In order to meet the consumers' demand for high-quality produce, fresh produce is generally stored at low temperatures (Tanaka, 2005). Studies have determined that the best way to delay the ripening of tomatoes is to store them at a temperature of 12.7°C. The fresh produce temperature should be kept as low as possible, from harvest until

consumption, with the exception of produce that is susceptible to chilling injury (Parsons *et al.*, 1970; Mekonnen, 2017). However, when the fruit is ripe, lower temperatures of up to 9°C may be necessary. The type of packaging and the pre-treatments applied prior to packaging, as well as the appropriate temperature for storing freshly-harvested tomatoes, will vary, depending on the tomato cultivar. Therefore, it is vital to investigate the impact of retail packaging on the quality of tomatoes (Mekonnen, 2017)

#### 2.9 Food Packaging Innovations

Novel food packaging technologies have become necessary, due to the consumers' increased preference for convenient, ready-to-eat and minimally-processed food products that are of a high quality and that have a prolonged shelf-life (Majid *et al.*, 2018a). The recent lifestyle changes of consumers have resulted in them having a limited time to prepare food, which has created a major challenge for the food packaging industry, which is now seeking to establish new and innovative food packaging solutions (Majid *et al.*, 2018a). Novel technologies for food packaging are designed for the protection of mildly-processed and fresh food against spoilage agents (Lin *et al.*, 2019).

Moreover, the modern trends in retail practices and lifestyle changes are the incentive for the evolution of innovative packaging techniques, without compromising the safety and quality of the food (Dainelli *et al.*, 2008). The packaging also provides the necessary product details for consumers, in order to make the advertisement and promotion of the product easier (Mihindukulasuriya and Lim, 2014). Another important reason for innovative food packaging is the rising issue of food-borne microbial outbreaks, which demands that the packaging must have antimicrobial properties in order to retain the quality of the food (Appendini and Hotchkiss, 2002).

### 2.10 Discussion and Summary

The negative impact of non-degradable, petroleum-based packaging films on the environment has resulted in an increasing number of research studies on biodegradable packaging films. Biopolymers are eco-friendly and biodegradable, which makes them a potential source for biobased plastics. Biodegradable materials are renewable and bio-based, therefore they can protect the contents from the environment and preserve their quality parameters during storage (Majid *et al.*, 2018b). Nevertheless, biopolymers do exhibit performance constraints, such as limited mechanical and barrier properties (Majid *et al.*, 2018b). An alternative way of enhancing the mechanical and physical properties of these films is, therefore, to combine polysaccharides (e.g.

starches, alginates, cellulose and chitosan) with proteins (e.g. milk proteins, soy protein, collagen and gelatin) (Meritaine da *et al.*, 2018). Compared to the commercially-available packaging materials, the widespread use of bio-based films depends on their mechanical and barrier attributes. The mechanical properties determine the capacity of the film to protect food from physical damage.

The equilibrium moisture content is responsible for the physical and chemical deterioration and dehydration of packed fresh agricultural produce (Khalil *et al.*, 2018). The water vapour barrier characteristic of food packaging films is therefore of great importance for the preservation of tomatoes and the extension of their shelf-life. Petroleum-based films are commonly used for fresh tomatoes; however, non-biodegradable packaging that is derived from non-renewable materials can contribute to ecological degradation (Khalil *et al.*, 2018). Research efforts are presently focusing primarily on the development of biodegradable films derived from biopolymers.

The use of biodegradable packaging materials is increasing in the South African fresh food supply chain. Recent developments show the intensive application of biodegradable packaging materials in the post-harvest handling of tomatoes. However, there is limited information on the physical and mechanical properties of the packaging and the role that these properties play in maintaining the quality attributes of packaged tomatoes. In addition, there is a need for further information on the interaction of the mechanical properties with the firmness and colour attributes of tomatoes, under different storage conditions. The firmness (compression and puncture) of tomatoes can be determined by using the Instron Universal Testing Machine and Texture Analyzer, and the colour parameters (redness, lightness) can be evaluated by using a Minolta chromameter. This chapter reviewed and discussed the different biodegradable packaging materials and their potential for replacing plastic. The most crucial issue with biodegradable materials is that they possess weak mechanical properties, compared to plastics. Starch-based films have demonstrated several advantages, such as their recyclability, sustainability, biodegradability, availability and affordability. They also exhibit physical characteristics that are similar to conventional plastic packaging, in terms of being transparent, odorless and tasteless. Starch-based products have inherent drawbacks, such as their high brittleness and moisture sensitivity, as well as them being a poor water vapour barrier, which, in turn, affects their mechanical behaviour. Reports in the reviewed literature have demonstrated that the food industry is currently exploring alternatives to replace the petroleumbased packaging materials with biodegradable and environmentally-friendly packaging materials. Much research has been dedicated to the improved performance of packaging materials, to making available a wide range of materials for any purpose, and to be able to respond to the specific requirements of different food products. Research has shown that the utilisation of synthetic plastic material in food packaging has had an adverse effect on the environment due to its non-biodegradability. Studies have reported that starch and polylactic acid have the potential to replace synthetic plastics. However, starch alone cannot produce films with enough mechanical strength; therefore, it is often mixed with plasticizers to improve its mechanical strength. Polysaccharides, proteins and lipids are biopolymers that also form an appropriate alternative, because they are non-toxic and biodegradable and are derived from sustainable natural resources. Due to their relatively low mechanical and barrier properties, compared to their non-biodegradable counterparts, biodegradable films have limitations in their use as packaging materials.

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# 3. CHARACTERIZATION OF BIODEGRADABLE PACKAGING MATERIALS

### Abstract

Biodegradable polymers have emerged as a subject of enormous scientific and industrial interest due to their environmentally friendly composability. For the benefit of the market economy and reoccurring environmental hazards, biodegradable materials should play a more critical role in packaging materials. Biodegradable packaging materials (the stamped paper tray, glued paper tray and pulped paper tray) and conventional packaging materials (expanded polystyrene, polypropylene plastic and Zipo punnet) were characterized for their physical, mechanical and water barrier properties. The mechanical properties of biodegradable packaging materials were compared to conventional packaging in this study. The packaging was stored under two different storage conditions (cold 12°C, 80% RH, and ambient 20°C) for 28 days. The experimental design layout constituted of a complete randomized block, with packaging materials as main block and storage condition as subplot with random allocation of treatments. The results showed that the mechanical properties of biodegradable materials were significantly (P<0.01) affected by cold environmental conditions. Under cold storage conditions, the tensile strength decreased by 3.5%, compared to that under ambient storage conditions. The tensile strength of conventional packaging remained the same under both storage conditions, with the expanded paper tray in the range of 54.29-54.31 MPa, the polypropylene tray in the range of 93.56-95.56 MPa and the Zipo punnet in the range of 111.57-111.59 MPa. The glued paper tray showed the highest tensile strength for both the cold and ambient storage conditions, compared to the stamped paper tray and pulped paper tray. The modulus of elasticity was observed to be higher in conventional packaging, compared to biodegradable packaging. The Fourier Transformed Infrared (FTIR) showed a similar spectrum for biodegradable packaging material, with a broad absorption band ranging from 3338,98–3200 cm<sup>-1</sup>, which was attributed to the O– H stretching vibration. Among the main absorption peaks was a strong-intensity absorption peak that was centred around 1025.09-1001 cm-1, which can be attributed to the C-O bond stretching of the C–O–C groups in the anhydro-glucose ring. The FTIR results concurred with the EDX results, which revealed that the elementary composition of the biodegradable packaging materials contained the highest carbon atomic 60.5% and weight 39.01%. The highest WVTR (Water Vapour Transmission Rate was observed in the stamped paper tray (264.73 gm<sup>-2</sup> day<sup>-1</sup>), due to the plasticized NC-sorbitol coated substrate with three layers. The cross-linking in the glued paper tray (254.31 gm<sup>-2</sup> day<sup>-1</sup>) decreased the WVTR, when compared to the stamped paper tray. The pulped paper tray (coated with nanocellulose) showed a lower WVTR of 250.28 (gm<sup>-2</sup> day<sup>-1</sup>). Biodegradable packaging materials have the highest water vapour permeability (stamped paper tray =  $9.3 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>, glued paper tray =  $8.9 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> and pulped paper tray =  $11.05 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>), which can be attributed to their more porous structure and irregular surface from scanning electron microscopy. The conventional packaging had the lowest WVP values (zipo punnet =  $6.04 \times 10^{-4}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>, expanded polystyrene = $3.54 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> and polypropylene = $2.13 \times 10^{-5}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>). The pulped paper tray had the highest solubility % due to its hydrophobic nature. The incorporation of a sorbitol plasticizer and cross-linking improved the mechanical properties and decreased the solubility, which proved to be a good material for water stability.

# 3.1 Introduction

Packaging films developed from petroleum-based plastics are widely used in the food industry to preserve and extend the shelf-life of foodstuffs (Tumwesigye *et al.*, 2017). However, the negative environmental impacts that emanate from the excessive use of non-biodegradable plastics has drawn the attention of material scientists and engineers and made them focus on renewable and biodegradable food packaging materials (Sanyang *et al.*, 2016). Over the past few decades, intensive research efforts have been dedicated to substituting petroleum-based plastics with more environmentally-friendly materials for food-packaging applications (Tampau *et al.*, 2020). Hence, biopolymers have emerged strongly as an alternative replacement for resolving the environmental problems caused by conventional packaging plastic waste. To solve the problems generated by plastic waste, many research efforts have been made to produce an environmentally-friendly material (Sahi *et al.*, 2021).

Most of the research focuses on substituting petrochemical-based plastics with biodegradable materials that have similar properties and that are cheap (Meritaine da *et al.*, 2018). Significant efforts that have been made to extend the shelf-life and enhance the food quality, while reducing packaging waste, and this has encouraged food and packaging industries to explore new biobased packaging materials, in order to reduce the waste-disposal problems (Tumwesigye *et al.*, 2017). Biopolymers derived from natural resources have attracted a great deal of attention in recent years and are considered to be a potential substitute for traditional non-biodegradable plastic films, due to their low cost and availability from reproducible resources, as well as their biodegradability (Zhong *et al.*, 2020). This study therefore aims to investigate and characterise the properties of biodegradable packaging materials.

# 3.2 Materials and Methods

# **3.2.1** Experimental site

The packaging materials were supplied by the ZZ2 Farm in the Limpopo Province, South Africa. The experiments were conducted at the Food Science Laboratory of the University of KwaZulu-Natal, Scottsville, Pietermaritzburg.

The experimental design consisted of a complete randomized block with packaging materials as main block. The arrangement of the factors is demonstrated in Figure 3.1 below:

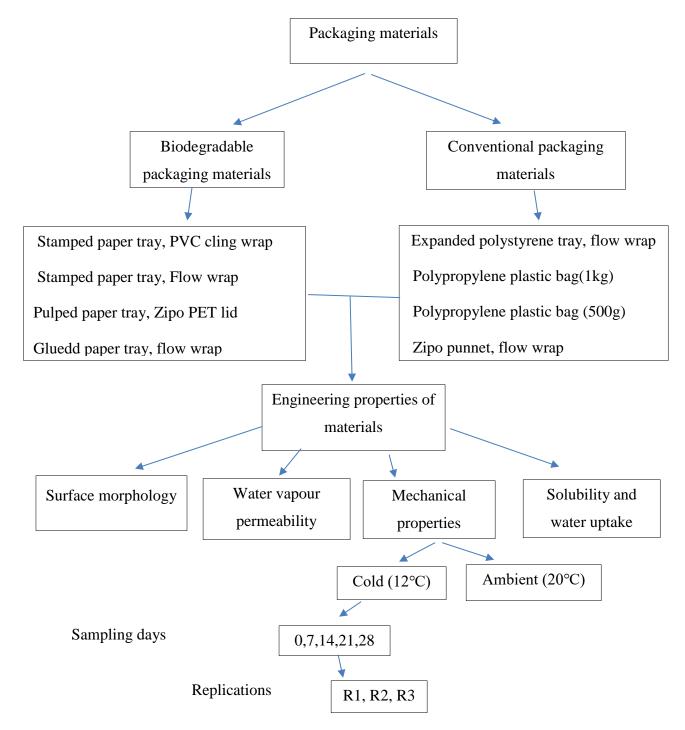


Figure 3.1 Complete randomized block experimental design.

# 3.2.2 Packaging material samples

Six different packaging materials were used in this study. Three packaging materials were biodegradable (the stamped paper tray, glued paper tray and pulped paper tray), while the other three were conventional packaging materials (polypropylene plastic, zipo punnet and expanded polystyrene). Biodegradable packaging materials are made up of wood pulp, which consists of nanocellulose fibres. Citric acid anhydrous was used as a cross-linking agent and D-sorbitol (98%) was used as a plasticizer. The chemical structure of cellulose is demonstrated in Figure 3.2 below. Sodium hypophosphite monohydrate was used as the catalyst for the cross-linking reaction and sulphate mixture was added to separate the cellulose fibre. To enhance the strength of this packaging against moisture, cationic starch was added to a wet pulp during the manufacturing process. Sorbitol was used as a plasticizer (its chemical structure is demonstrated in Figure 3.3 below). Layer-by-layer coatings were done on the porous substrate. The description of the materials was obtained from the manufacturer, as shown in Table 3.1 below.

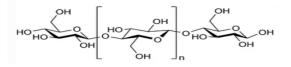


Figure 3.2 Chemical structure of cellulose (de Cuadro et al., 2015).

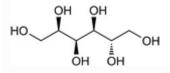


Figure 3.3 Chemical structure of sorbitol (de Cuadro et al., 2015).

 Table 3.1
 Description of biodegradable packaging materials

Sample names	Description of the materials	
Stamped paper tray	Plasticized NC-sorbitol coated substrate, three layers	

Glued paper tray	Cross -linked and plasticized NC-sorbitol coated substrate, six layers
Pulped paper tray	NC coating substrate, three layers

NC -(nanocellulose)

### **3.2.3** Data collection

The physical properties (solubility, water uptake, water vapour permeability, morphology) were measured and analysed. The mechanical properties were sampled from Day 0 and after 7, 14, 21 and 28 days. The assessment of these quality parameters was carried out as follows:

# 3.2.3.1 Solubility in water

The solubility of biodegradable films in water was measured by using the method of Tajik *et al.* (2013). The film samples were dried in a laboratory oven at 110°C for 24 h and then weighed, to determine their initial solid content. The pre-weighed film samples (1 cm  $\times$  3 cm) were immersed under constant agitation in 50 ml of distilled water for 6 h at 25°C. Thereafter, the remaining pieces of films were filtered and dried at 110°C to a constant weight (the final dry weight). The water solubility (%) of the film was calculated according to Equation 3.1 below:

WS (%) = 
$$\frac{W_i - W_f}{W_i} \times 100$$
 (3.1)

Where:

 $W_i$  is the initial weight of the film expressed as dry matter and  $W_f$  is the weight of the desiccated undissolved film.

# 3.2.3.2 Water uptake

The water uptake capacity of each film was determined by immersing a film sample in distilled water at room temperature  $(23\pm2^{\circ}C)$  for about three minutes. The water uptake was determined by using Equation 3.2 below:

Water uptake (%) = 
$$\frac{M_0 - M_i}{M_o} \times 100$$
 (3.2)

# 3.2.3.3 Water vapour permeability

Water Vapour Permeability (WVP) tests were conducted by using the standard ASTM method E96 (Astm) by Gürler *et al.* (2020), with slight modifications. Special blow-twin cups, with an average diameter of 3.5 cm and a depth of 2 cm, were utilized to determine the WVP of the films. The films were cut into discs with a diameter slightly larger than the diameter of the cup. After placing 10 g of the anhydrous CaCl<sub>2</sub> (RH = 0%) in each cup, they were covered with the

different films. Each cup was placed in a desiccator that contained a saturated potassium nitrate (KNO<sub>3</sub>) solution in a small beaker at the bottom. A small amount of solid KNO<sub>3</sub> was left at the bottom of the saturated solution to ensure that it remained saturated at all times. The molarity of KNO<sub>3</sub> is 2.532 mol/L. The saturated KNO<sub>3</sub> solution provides a constant RH of 97% at 25°C in the desiccator. Air was continuously circulated throughout the chamber at a velocity that was sufficient for maintaining uniform conditions. The desiccator was kept in an incubator at 38.0 ± 0.1°C. Cups were weighed every 24 h for seven days and the water vapour transport was determined by the weight gain of the cup. Changes in the weight of the cup were recorded as a function of time. The slopes were calculated by linear regression (weight change vs. time), and the water vapour transmission rate (WVTR) was defined as the slope (g/day) divided by the transfer area (m<sup>2</sup>). The WVP (g m<sup>-1</sup> day<sup>-1</sup> Pa<sup>-1</sup>) was calculated, as in Equation 3.3 below:

$$WVP = \frac{WVTR}{P(R_1 - R_2)} x$$
(3.3)

Where:

*P* is the saturation vapour pressure of water (Pa) at the test temperature (25°C),  $R_1$  is the RH in the desiccators (0.97),  $R_2$ , is the RH in the cup (0) and *X* is the film thickness (m). Under these conditions, the driving force [ $P(R_1-R_2)$ ] is 3073.93 Pa. All measurements were performed in triplicate. Three samples were prepared from three individual films.

### 3.2.3.4 Morphology analysis

The morphology of the film was tested by using Scanning Electron Microscopy (SEM). The tools that were used to prepare the samples were a pair of scissors, carbon tape and sample stubs. The samples were cut into 0.4 mm x 0.4 mm squares. The samples were mounted into a stub by sticking them onto a double stick carbon tape. The samples were then coated with a conductive coating, which was gold, to prevent charging problems. The samples were then placed into a sputter coder to deposit a thin layer of gold under vacuum (Zhou *et al.*, 2019). Three different areas of each sample were imaged at different magnifications and the composition of elements was determined by using EDX.

# 3.2.3.5 Mechanical properties

The mechanical properties of the measured samples were stored under ambient (20°C, 65% RH) and cold (12°C, 80% RH) conditions, using a Instron Universal Testing Machine (Model 3345). The samples were stored for 28 days. Tests were performed at seven-day intervals. The films were cut into 50 x 15 mm pieces and the initial gauge length and crosshead speed were

fixed at 30 mm.min<sup>-1</sup> and 10 mm.min<sup>-1</sup>, respectively. Tensile measurements were carried out by using a 25kg load cell at  $(23\pm1^{\circ}C)$  and a 50± 2% relative humidity. After the strip break, the computer software recorded the modulus of elasticity, the yield point and the stress at break. The tensile strength (N/mm<sup>2</sup>) was calculated from the maximum force by dividing it by the area of the cross-section. The reported values are the averages of the three samples.

### 3.2.3.6 FTIR

The film samples were crushed in liquid  $N_2$  and the analysis was carried out on a Fourier Transform Infrared (FTIR) spectrophotometer (PerkinElmer precisely spectrum 100 FT-IR Spectrometer). The FTIR spectra of the samples were scanned in a wave range of 4000-500  $cm^{-1}$  with a resolution of 4.0  $cm^{-1}$  at 25 (Zhou *et al.*, 2019).

### **3.2.4** Data analysis

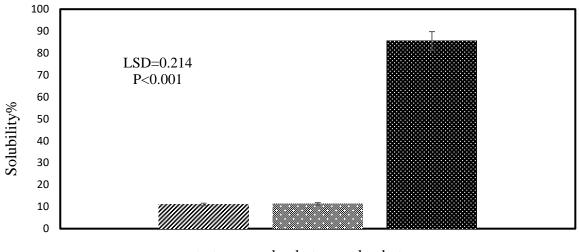
The data were analysed by using the Genstat 18th edition A VSNI. The data were analysed with the Analysis of Variance (ANOVA) at a 95% confidence level, and the differences at P<0.05 were considered to be statistically significant. Graphical representations were made by using Excel.

# **3.3 Results and Discussion**

# 3.3.1 Solubility properties and water uptake

It is vital that the packaging films have a high-water resistance for the preservation of foodstuffs. In this study, the pulped paper tray showed the highest water uptake, due to its hydrophilic nature. There was a significant (P<0.001) decrease of 8.33% water uptake and 74% solubility in the glued paper tray and stamped paper tray, which might be due to cross-linking and the addition of a sorbitol plasticizer. The pulped paper tray and glued paper tray, which highest film solubility (85%), compared to the stamped paper tray and glued paper tray, which had a film solubility value of 11.04% and 11.31%, respectively. The stamped and glued paper tray showed the lowest water uptake value, as demonstrated in Figure 3.5 compared to the pulped paper tray, which proved to be a good water stability material. It is evident that the pure coating with NC (nanocellulose) is responsible for the water absorption, whereas the cross-linked and NC-sorbitol strongly resisted water uptake, owing to hydrophobic nature. This observation agrees with the findings of Herrera *et al.* (2017), who also reported that the addition of

sorbitol enhances the water-resistant properties of the films, which is a relevant feature for food packaging materials, in order to improve the shelf-life of food products.



% stp tray  $\blacksquare$  glued pt  $\blacksquare$  pulped pt

Figure 3.4 Solubility of biodegradable packaging materials

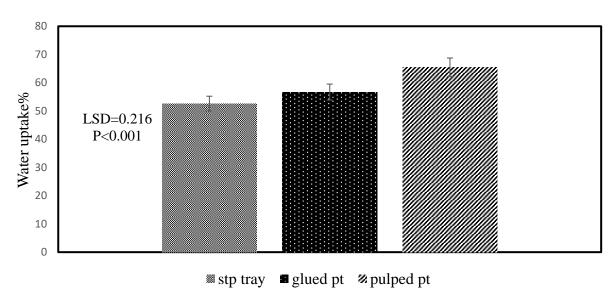


Figure 3.5 Water uptake of biodegradable packaging materials

# 3.3.2 Water vapour permeability

Water Vapour Permeability (WVP) is an essential parameter in food packaging materials, and it should be as low as possible in food packaging films, since it is frequently necessary to limit the transfer of moisture between the food and the surrounding atmosphere (Seligra *et al.*, 2016). It is important to emphasize that a low water vapour permeability is a desirable property for the use of biodegradable films in food packaging, especially for tomatoes. There are lots of factors (Intrinsic and extrinisic) that will contribute to low water activity and inherent contamination, more so many fruits, and vegetable requires gas exchange within and outside the package to minimize the accumulation of ethylene, and balance the oxygen and carbon dioxide concentrations, and hence high permeable or porous materials may be an advantage. The WVP was determined by using the cup method; anhydrous calcium chloride (CaCl<sub>2</sub>) was used as a desiccant because it effectively absorbs moisture from the surrounding air. The weight gain values of packaging materials are presented in Table 3.2. The results revealed a highly-significant (p<0.001) increase in the weight gained by the cups covered by different packaging films over time (days). The periodic weighing was used to measure the water vapour transmission rate through the films into the desiccant. Figure 3.6 demonstrates the exponential increase in weight gained over the seven days. Biodegradable packages (the stamped paper tray, glued paper tray and pulped paper tray) gained more weight over seven days than the conventional packaging (the Zipo punnet, the polypropylene plastic bag and the EPS polystyrene tray). The weight gained by the cups covered with biodegradable films after Day 7 ranged between  $15.74\pm0.120$  g to  $17.10\pm0.529$  g, whereas the weight gained by cups covered with conventional packaging ranged between  $10.73\pm0.056$  g to  $10.82\pm0.015$  g.

Packaging	Storage period (days) in an incubator at $30.0 \pm 0.1$ °C								
Material	1	2	3	4	5	6	7		
Stp tray	11.23±0.550 <sup>f</sup>	12.37±0.057 <sup>h</sup>	13.43±0.152 <sup>j</sup>	14.53±0.208 <sup>k</sup>	15.87±0.057 <sup>1</sup>	16.43±0.115 <sup>m</sup>	17.10±0.529 <sup>h</sup>		
EPS tray	10.30±0.095 <sup>ab</sup>	10.40±0.005 <sup>bcde</sup>	10.47±0.049 <sup>f</sup>	10.52±0.005 <sup>g</sup>	10.61±0.005 <sup>gh</sup>	10.72±0.015 <sup>i</sup>	10.82±0.015 <sup>j</sup>		
Ppl bag	10.20±0.000ª	10.31±0.010 <sup>ab</sup>	10.32±0.006 <sup>ab</sup>	10.41±0.006 <sup>abcde</sup>	10.46±0.006	10.67±0.029 <sup>abc</sup>	10.77±0.010 <sup>de</sup>		
Zipo punnet	10.10±0.100ª	10.30±0.000b	10.32±0.005 <sup>b</sup>	10.40±0.005 <sup>bc</sup>	10.50±0.002 <sup>cd</sup>	10.60±0.006 <sup>def</sup>	10.73±0.056 <sup>fg</sup>		
Glued pt	10.71±0.010 <sup>efg</sup>	11.61±0.092 <sup>k</sup>	12.86±0.015 <sup>1</sup>	13.69±0.254 <sup>m</sup>	14.50±0.070°	15.59±0.335°	16.53±0.068 <sup>q</sup>		
Pulp pt	10.31±0.011 <sup>b</sup>	10.52±0.015 <sup>cd</sup>	11.74±0.017 <sup>k</sup>	12.88±0.011 <sup>1</sup>	13.94±0.063°	14.59±0.080°	15.74±0.120 <sup>p</sup>		
Significance	level								
Packaging Treatment (		(A) <0.001,							
Days (B) <0.00		< 0.001	< 0.001						
A*B,		= 0.011							

Table 3.2 The weight gained (g) by CaCl<sub>2</sub> inside the containers covered by different types of packaging materials over seven days

STP tray - stamped paper tray, EPS tray - Expanded Polystyrene tray, PPL - Poly propylene plastic bag, PT - paper tray. LSD = 0.2724, %CV = 1.4; s.e = 0.2006. Means followed by the same letter(s) are not significant: Duncan's multiple range test (P < 0.05)

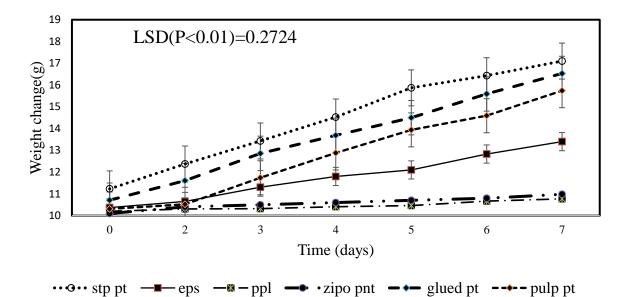


Figure 3.6 Weight gained by the desiccant (CaCl<sub>2</sub>) in the cups covered by packaging films CV% = 1.4.

The Water Vapour Permeability (WVP) and Water Vapour Transmission Rate (WVTR) of the samples were measured and are graphically illustrated in Figure 3.7. The highest WVTR was observed in the stamped paper tray (264.73 gm<sup>-2</sup> day<sup>-1</sup>) due to its plasticized NC-sorbitol-coated substrate with three layers. The cross-linking in the glued paper tray (254.31 gm<sup>-2</sup> day<sup>-1</sup>) decreased the WVTR, when compared to the stamped paper tray. The pulped paper tray (coated with nanocellulose) showed less WVTR of 250.28 (gm<sup>-2</sup> day<sup>-1</sup>). When comparing the values obtained in this study with the WVTR of pure micro-fibrillated cellulose films (234 gm<sup>-2</sup> day<sup>-</sup> <sup>1</sup>) reported by Rodionova *et al.* (2011), all three WVTR values obtained in this study are higher than this value. The conventional packaging showed the lowest WVTR, compared to the biodegradable packaging. In order to consider the differences in the thickness of the coatings, the WVP was studied. The three-layer coatings had the highest WVP, compared to the thicker coating with six layers. Biodegradable packaging materials have the highest WVP (the stamped paper tray =  $9.3 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>, glued paper tray =  $8.9 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> and pulped paper tray =  $11.05 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>), which is attributed to their more porous structures and their irregular surfaces for the Scanning Electron Microscopy. Similar results were reported by Nouraddini et al. (2018), who found that biodegradable packaging (egg-flour plant) had the highest level of WVP ( $27 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>), due to its low density and more porous structure, while conventional packaging had the lowest values of WVP (zibo punnet =  $6.04 \times 10^{-4}$  gm<sup>-1</sup>day<sup>-1</sup> <sup>1</sup>Pa<sup>-1</sup>, expanded polystyrene= $3.54 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> and polypropylene= $2.13 \times 10^{-5}$  gm<sup>-1</sup>day<sup>-1</sup>

<sup>1</sup>Pa<sup>-1</sup>). Biodegradable packaging often has a greater permeability to water vapour, when compared to conventional packaging, such as poly(ethylene), and it is affected by the temperature and relative humidity of the environment and by the structure of the polymers that comprise them (Stoll *et al.*, 2016).

It is necessary to frequently restrict the moisture transfer between the food and the surrounding atmosphere. Water vapour permeability is an important property of films and their application, because of the deteriorative reactions of water. Furthermore, the moisture content has a great effect on the quality of foods, so it is necessary to select a packaging material that has the appropriate moisture permeability for protecting the food quality during storage (Majzoobi *et al.*, 2015).

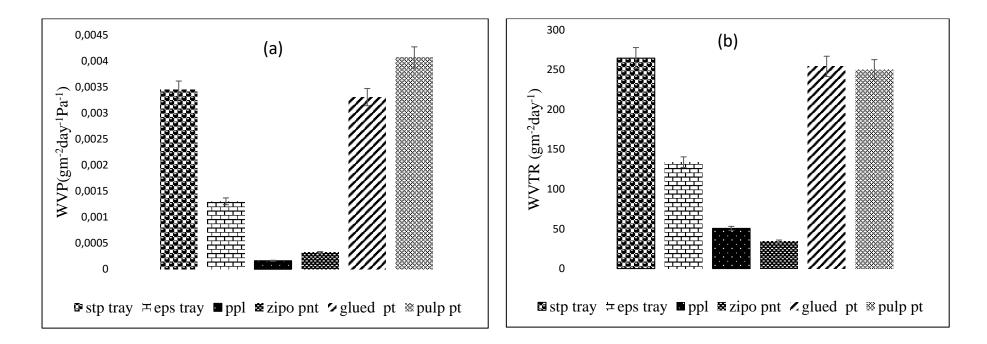


Figure 3.7 WVP (a) and WVTR (b) of biodegradable packaging (stp tray, pulped pt, glued pt), compared to conventional packaging (EPS tray, zipo pnt, ppl).

### 3.3.3 Morphology analysis

Scanning Electron Microscopy (SEM) has been widely used as a tool for the study and characterization of the microstructure of edible films (Tajik *et al.*, 2013). It was used to determine the surface morphology of biodegradable packaging materials. As shown in the Figures below, there were visible pores that resulted in a high-water vapour permeability rate. The SEM images showed the visible heterogenous surface cracks. Furthermore, the biodegradable materials did not show any intact starch granules, which indicates that the gelatinization was complete.

In order to determine the elemental composition of the packaging materials, an EDX analysis was performed. The EDX measurements focused on different areas, and the corresponding peaks are shown in Figure 3.8. The presence of Au (gold) and Ai (aluminium) peaks in the spectra is because the samples were coated by gold and mounted on aluminium stubs. The details of the three EDX spectra of the elemental compositions values of the biodegradable packaging are represented in Table 3.3. EDX shows the highest composition of carbon, because cellulose consists of more carbon atoms. The EDX spectra in Figure 3.9 contain mainly Ca (calcium), which is completely embedded by cellulose fibres, suggesting that it was present in the pulp during the manufacturing process. The Figures below all show a cluster of mineral materials containing mainly O (Oxygen) and C (Carbon), but also Au (gold). However, the fact that the minerals are embedded and partly covered by cellulose fibres suggests that their presence is associated with the preparation of the cellulose mixture.

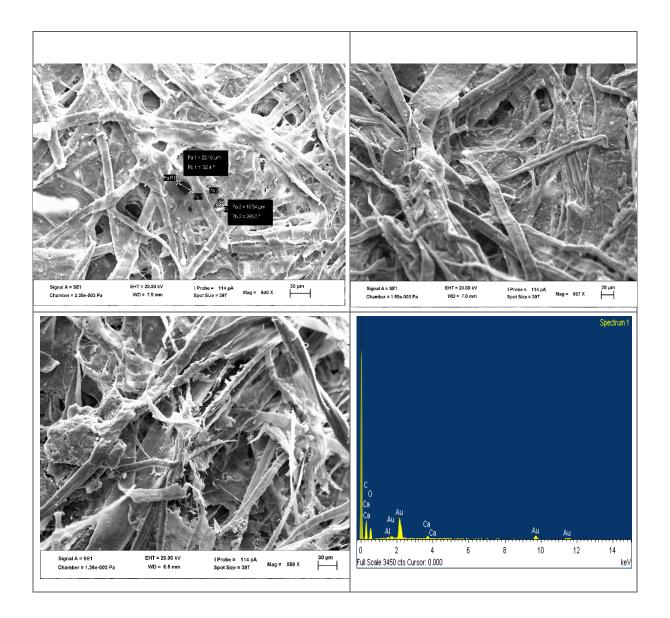


Figure 3.38 SEM images of the pulped paper tray at three random areas and EDX spectra.

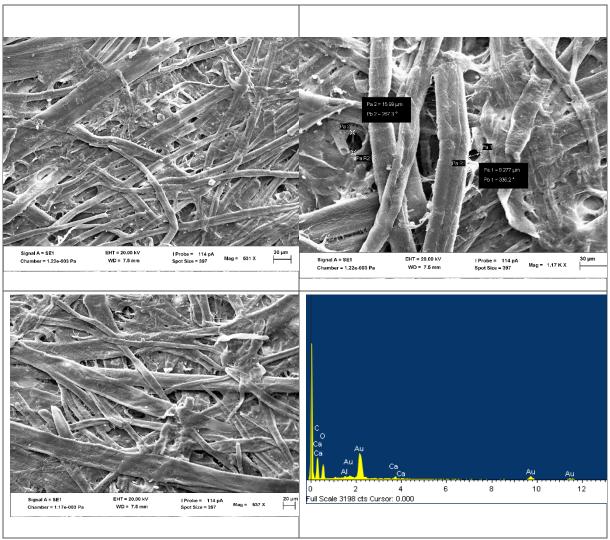


Figure 3.9 SEM images of the glued paper tray at three random areas and EDX spectra.

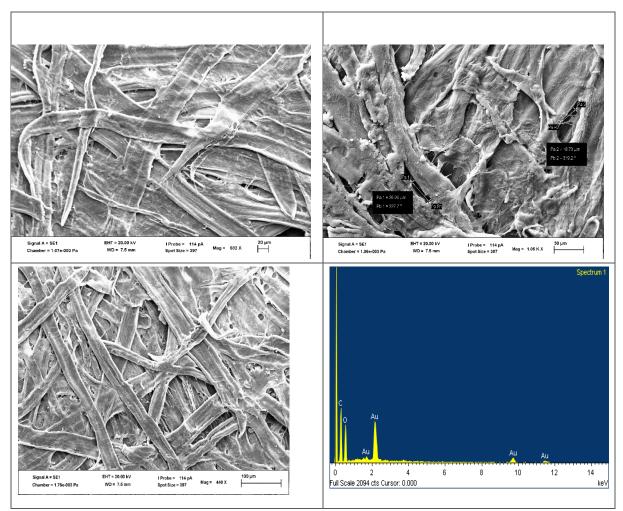


Figure 3. 10 SEM images of the stamped paper tray at three random areas and EDX spectra.

Packaging	C(carbon)		O(Oxygen)		Al (aluminium)		Au(gold)	
materials	Atomic%	Weight%	Atomic%	Weight%	Atomic%	Weight%	Atomic%	Weight%
Stamped paper tray	60.88±1.02 <sup>d</sup>	37.81±1.12	34.65±0.95°	28.65±0.92 <sup>cd</sup>	0.40±0.08ª	0.55±0.12ª	3.04±0.20 <sup>b</sup>	30.68±1.52 <sup>cde</sup>
Glued paper tray	59.42±1.11 <sup>d</sup>	38.33±1.39 <sup>ef</sup>	34.33±4.37°	32.83±2.30 <sup>cdef</sup>	0.41±0.04ª	0.52±0.16 <sup>b</sup>	2.73±0.8 <sup>ab</sup>	28.82±3.08 <sup>cd</sup>
Pulped paper tray	61.20±0.72 <sup>d</sup>	40.91±0.55 <sup>f</sup>	34.33±0.65°	32.11±0.57 <sup>cdef</sup>	0.27±0.01ª	0.41±0.12ª	2.41±0.04 <sup>ab</sup>	26.45±0.41°
Significant level								
Packaging Treatm	nent (A)	< 0.001						
Elements (B)		< 0.001						
A*B		=0.011						

Table 3.3	Composition of the quantit	y of elements found i	n biodegradable packaging

weans followed by the same letter(s) are not significant: Duncan's multiple range test (P < 0.05), LSD = 2.356, %CV = 5.7, s.e =1.398

#### **3.3.4** Mechanical properties

Biodegradable paperboard is prone and susceptible to water absorption from the environment, particularly during storage under at a high humidity, or when it comes into contact with food materials with a high moisture content, such as fresh horticultural produce (Fadiji *et al.*, 2019). Water absorption minimises the physical and mechanical strength of the biodegradable paperboard, which causes damage to the packed produce during its storage and distribution. The temperature and relative humidity under ambient storage conditions were 20°C and 80% RH, and for cold storage conditions they were kept at 12°C and 65% RH throughout the fourweek period (Figure 11). The results in Tables 3.4 and 3.5 revealed a highly-significant (P<0.001) difference between the cold and ambient storage conditions. The cold storage environment (12°C) significantly (P<0.001) reduced the tensile strength and modulus elasticity of the packaging materials. A similar study observed the influence of the standard conditions and refrigerated conditions, as the tensile strength of biodegradable paper reduced 38% at refrigerated conditions (Fadiji et al., 2017). This significant decrease is due to the high relative humidity of 80% under cold storage conditions. A high RH is associated with a high moisture content, and an increase in the moisture content reduces the fibre network strength, which decreases the tensile strength and elastic modulus (Table 3.5).

A decrease in the tensile strength can lead to the packed produce becoming susceptible to damage. The tensile strength and modulus of elasticity of the biodegradable packaging materials were significantly (P<0.001) affected by the cold storage conditions, compared to the conventional packaging stored under ambient conditions (Figure 3.12b). An elastic modulus is a quantity that measures the resistance of an object to being elastically deformed. The highest modulus of elasticity (1.925-1.482 Nm<sup>-2</sup>) was observed in the Zipo punnet and the polypropylene plastic bag. These results correlate positively with the findings of Herrera *et al.* (2017), which revealed a significant (P<0.001) decrease in the tensile strength of paper when the relative humidity was increased from 50% to 80%. The tensile strength and modulus of elasticity of biodegradable material in this present study continued to decrease from Day 0 to Day 28, compared to those of the conventional packaging materials (Figure 3.12a). The apparent difference between the cold and ambient conditions was because the relative humidity at ambient temperatures was kept at 65%, which as lower than the relative humidity of 80% at cold temperatures.

The equilibrium moisture content of the biodegradable paper was closely linked to the relative humidity. When the RH of paper increases, the paper fibres absorb moisture, or release it, to

the surrounding environment. Moreover, when paper material absorbs moisture, the water content increases significantly. The bonds of the cellulose fibre break in the paper material, which significantly affects its mechanical properties. The pulped paper tray was found to have the lowest tensile strength values, ranging from  $62.59\pm0.4719^{\text{ef}}$  -  $48.50\pm0.2996^{\circ}$  MPA. This may be due to the nanocellulose substrate coating with three layers. In general, the addition of a nanocellulose (NC) coating on the paper board is expected to increase its mechanical properties. However, in this study, the NC did not improve the tensile strength and elasticity modulus of the pulped paper tray. The addition of a sorbitol plasticizer positively affected the tensile strength and elasticity on the stamped paper tray. When comparing it to the cross-linked glued paper tray, it was observed to have the highest tensile strength after 28 days of storage, under both the cold and ambient storage conditions, with the averages being 61.23±0.5670<sup>gh</sup> for cold and 68.57±2.0838<sup>gh</sup> MPA for ambient storage conditions. The maximum tensile strength remained similar in conventional packaging under both storage conditions, as demonstrated by Figure 3.12b; it remained constant throughout the 28-day storage period. Thus, it can be concluded that the plasticization improved the mechanical properties of the paper. The effects of sorbitol on the mechanical properties of cellulose films was previously studied by Hansen et al. (2012) and Liu et al. (2013), who reported a general increase in the maximum strength values, when compared with the samples without sorbitol, which can be seen in the coatings in this study. The H-bonding (originating from an electrostatic and charge transfer) and stacking interactions (arising from van der Waals interactions and hydrophobic forces) determine the biomolecular structure of the cellulose layer (Hobza and Müller-Dethlefs, 2010)

Packaging	Storage	Storage days						
Materials	condition	0	7	14	21	28		
Stamped	Cold (12°C)	85.69±8.22 <sup>k</sup>	69.09±6.65 <sup>hi</sup>	64.64±0.21 <sup>ef</sup>	60.71±5.40 <sup>b</sup>	57.61±3.12 <sup>a</sup>		
-	× , ,							
paper tray	Ambient (20°C)	85.69±8.22 <sup>k</sup>	82.26±7.75 <sup>k</sup>	76.85±4.21 <sup>j</sup>	68.31±2.27 <sup>ef</sup>	65.80±1.29 <sup>cd</sup>		
Glued paper tray	Cold (12°C)	76.45±0.00 <sup>j</sup>	73.27±0.06 <sup>j</sup>	66.94±0.05 <sup>h</sup>	65.31±0.3359 <sup>gh</sup>	61.23±0.5670gh		
	Ambient (20°C)	76.45±0.0092 <sup>j</sup>	74.22±0.07 <sup>j</sup>	72.68±0.20 <sup>ij</sup>	70.54±0.01 <sup>h</sup>	68.57±2.08 <sup>gh</sup>		
Pulped paper	Cold (12°C)	62.59±0.47 <sup>fg</sup>	58.18±0.15 <sup>def</sup>	55.40±0.04 <sup>cde</sup>	51.03±0.35 <sup>cd</sup>	48.50±0.29°		
tray	Ambient (20°C)	62.59±0.47 <sup>fg</sup>	60.52± 0.03 <sup>ef</sup>	59.72±0.32 <sup>def</sup>	58.39±0.05 <sup>cdef</sup>	57.40±0.06 <sup>cde</sup>		
Expanded	Cold (12°C)	54.20±0.04°	54.25±0.05°	54.29±0.05°	54.33±0.05°	54.37±0.06°		
polystyrene	Ambient (20°C)	54.20±0.06°	54.23±0.05°	54.27±0.0065°	54.31±0.0065°	54.35±0.0065°		
Polypropylene	Cold (12°C)	95.55±0.06 <sup>1</sup>	94.55±0.00111	93.56±0.111	92.57±0.11 <sup>1</sup>	95.57±0.11 <sup>1</sup>		
plastic	Ambient (20°C)	95.55±0.06 <sup>1</sup>	95.55±0.011	95.56±0.01 <sup>1</sup>	95.56±0.011	95.57±0.01 <sup>1</sup>		
Zipo punnet	Cold (12°C)	111.50±0.01 <sup>m</sup>	111.53±0.05 <sup>m</sup>	111.59 ±0.06 <sup>m</sup>	111.63±0.05 <sup>m</sup>	111.67±0.05 <sup>m</sup>		
	Ambient (20°C)	111.50±0.03 <sup>m</sup>	111.55±0.05 <sup>m</sup>	111.57±0.05 <sup>m</sup>	111.61± 0.05 <sup>m</sup>	111.65±0.05 <sup>m</sup>		
Significant leve	1	1		1				
Packaging Treatment (A)		< 0.005						
Days (B)		< 0.001						
Storage period (c)		<.001						
B*C		<.001						
A*B*C		=0.070						
A*B		=0.011						
A*C		<.001						

Table 3.4Tensile strength (mpa) of the packaging materials influenced by the ambient and<br/>cold storage conditions during the 28-day period

Means followed by the same letter(s) are not significant: Duncan's multiple range test (P < 0.05), LSD = 3.668, %CV = 3, s.e = 2.69

Packaging	Storage	Storage days						
Materials	condition							
		0	7	14	21	28		
Stamped	Cold	0.385±0.14 <sup>kl</sup>	0.240±0.01 <sup>ij</sup>	0.149±0.02 <sup>defg</sup>	0.057±0.02 <sup>bcd</sup>	0.033±0.02 <sup>a</sup>		
paper tray	Ambient	0.385±0.15 <sup>kl</sup>	0.225±0.05 <sup>hij</sup>	0.191±0.52 <sup>ghi</sup>	0.106±0.02 <sup>bcd</sup>	0.137±0.02 <sup>cb</sup>		
Glued paper tray	Cold	0.430±0.00 <sup>lm</sup>	0.445±0.01 <sup>mn</sup>	0.399±0.02 <sup>lm</sup>	0.338±0.02 <sup>k</sup>	0.012±0.01 <sup>def</sup>		
	Ambient	0.430±0.00 <sup>lm</sup>	0.491±0.02 <sup>n</sup>	0.418±0.07 <sup>lm</sup>	$0.384{\pm}0.02^{kl}$	0.259±0.04 <sup>defg</sup>		
Pulped paper	Cold	0.198±0.00ghi	0.152±0.01 <sup>defg</sup>	0.130±0.06 <sup>def</sup>	0.101±0.02 <sup>bcd</sup>	0.054±0.00 <sup>ab</sup>		
tray	Ambient	0.1987±0.00 <sup>ghi</sup>	0.174±0.07 <sup>fgh</sup>	0.162±0.05 <sup>efg</sup>	0.124±0.02 <sup>def</sup>	0.110±0.07 <sup>cde</sup>		
Expanded	Cold	1.3604±0.52 <sup>v</sup>	1.268±0.01 <sup>tu</sup>	1.223±0.02 <sup>st</sup>	1.131±0.02 <sup>qr</sup>	1.040±0.02°		
polystyrene	Ambient	1.360±0.52 <sup>v</sup>	1.317±0.01 <sup>uv</sup>	1.177±0.02 <sup>rs</sup>	1.085±0.05 <sup>pq</sup>	0.994±0.02°		
Polypropylene	Cold	1.819±0.02 <sup>b</sup>	1.726±0.01 <sup>ab</sup>	1.619±0.02 <sup>yz</sup>	1.514±0.01 <sup>x</sup>	1.432±0.02 <sup>w</sup>		
plastic	Ambient	1.817±0.02 <sup>b</sup>	1.7722±0.02 <sup>ab</sup>	1.6654±0.01 <sup>yz</sup>	1.5739±0.01 <sup>y</sup>	1.4824±0.02 <sup>x</sup>		
Zipo punnet	Cold	$2.275 \pm 0.02^{j}$	2.183±0.52 <sup>hi</sup>	2.092±0.02 <sup>fg</sup>	2.009±0.012 <sup>de</sup>	1.925±0.04°		
	Ambient	$2.275 \pm 0.02^{j}$	2.229±0.02 <sup>ij</sup>	2.138±0.01 <sup>gh</sup>	2.047±0.02 <sup>ef</sup>	1.952±0.02 <sup>cd</sup>		
Significant leve Packaging Tre			<0.005					
Days (B)			<0.001					
Storage period(C)			<.001					
B*C		<.001						
A*B*C			= 0.070					
A*B			= 0.011					
A*C			<.001					

Table 3.5Modulus of elasticity (Nm-2) of the packaging materials influenced by the ambient<br/>and cold storage conditions during the 28-day period.

Means followed by the same letter(s) are not significant: Duncan's multiple range test (P < 0.05),

LSD = 0.1583, %CV = 13.5, s.e = 0.1266

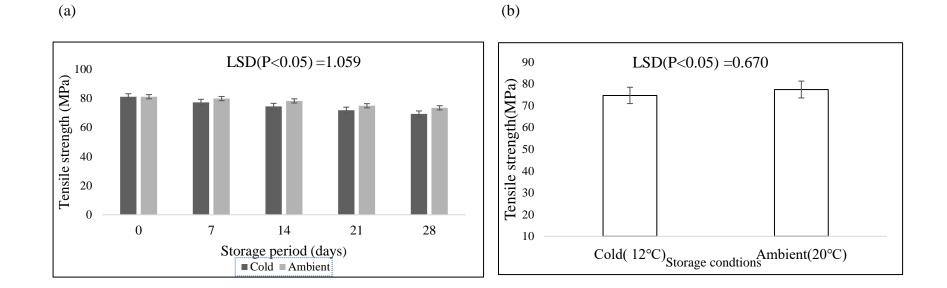


Figure 3.11 The interaction effect of ambient and cold storage on biodegradable and conventional packaging materials over the four-week storage period, (a) storage duration, (b) storage conditions.

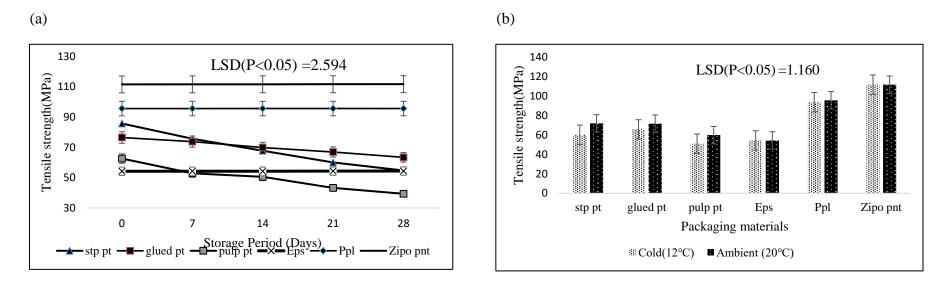


Figure 3.12 The interaction effect of ambient and cold storage on biodegradable and conventional packaging material over the four-week storage period. CV%=3, (a) packaging materials and storage duration, and (b) storage conditions and packaging materials on tensile strength.

#### **3.3.5** Spectroscopic analysis

FTIR spectroscopy was carried out to identify the functional groups and physical interactions that determine the properties of materials. The three different biodegradable materials demonstrated similar spectra. The FTIR spectra for biodegradable materials displayed three prominent characteristic absorption bands. The first was a broad absorption band, ranging from 3338,98–3200 cm<sup>-1</sup>, which is attributed to to the O–H stretching vibration. It is suggested that the intermolecular hydrogen bonding of organic material molecules is responsible for this O-H group band. The absorption peak centred at 2012,05 cm<sup>-1</sup> corresponded with the C-H stretching for the glued paper tray. The third main characteristic absorption band is attributed to the bond water molecules within the starch, which occurred at 1365.45-1316.52 cm<sup>-1</sup>, as illustrated in Figure 3.12. The last of the main absorption peaks is a strong-intensity absorption peak that centred around 1025.09-1001 cm-1, which can be attributed to the C-O bond stretching of the C–O–C groups in the anhydro-glucose ring. Similar bands that confirm the presence of C-O-C groups were reported by Sanyang et al. (2016). In Figure 3.13, the peak of approximately 3300 cm<sup>-1</sup> corresponds to that of the O-H groups. Figure 3.14 illustrates that the peak of approximately 3300 cm-1 corresponds to O-H groups; this broad absorption band  $(3650-3250 \text{ cm}^{-1})$  indicates a hydrogen bond, which confirms the existence of hydrate (H<sub>2</sub>O). At 2013.30 cm<sup>-1</sup>, the peak corresponds to that of the C–H groups, and at approximately 1025 cm-1, the peak corresponds to that of the C–O groups. These peaks correspond to the cellulose structure (de Cuadro et al., 2015; Herrera et al., 2017). No other peaks were observed between 3000 and 3200 cm<sup>-1</sup>, which shows that there is no aromatic structure. In the region of 2000-2500 cm<sup>-1</sup>, there is a band (2013.30 cm<sup>-1</sup>) that indicates C=C in the material. At 1159.04 cm<sup>-1</sup>, the peak indicates the hydrogen-bonded stretching mode of the C-OH groups. Usually, films made of hydrophilic polymers present a high barrier against oxygen at a low Relative Humidity (RH), because of the large number of hydroxyl groups (OH) in their structure. This occurs due to the low polarity of oxygen, which produces a weak interaction with the highly-polar hydroxyl groups of the polymer. When the relative humidity increases, the hydroxyl groups interact with the highly-polar water molecules, weakening the hydrogen bonds that hold the polymer chains together. The loosening of these bonds releases the structure of the polymer, which leads to an increased OP at a high RH (Tammelin and Vartiainen, 2014).

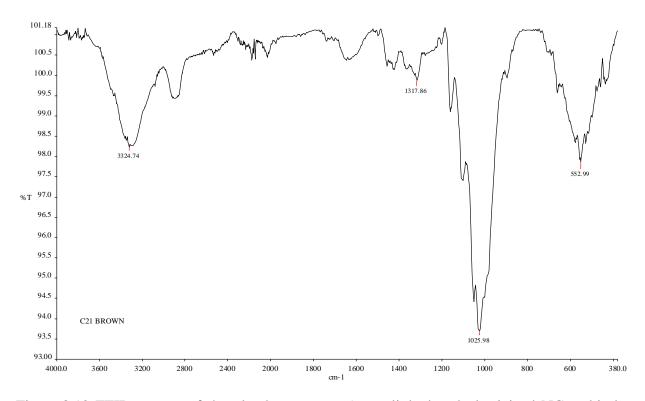


Figure 3.12 FTIR spectra of the glued paper tray (cross-linked and plasticized NC-sorbitol coated substrate, six layers).

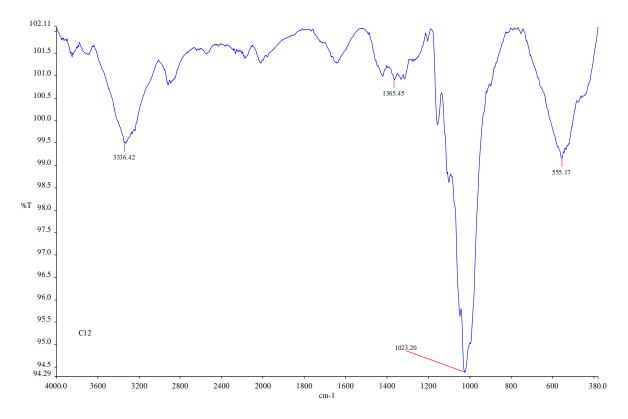


Figure 3.13 FTIR spectra of the pulped paper tray (NC (nanocellulose) coating substrate, three layers).

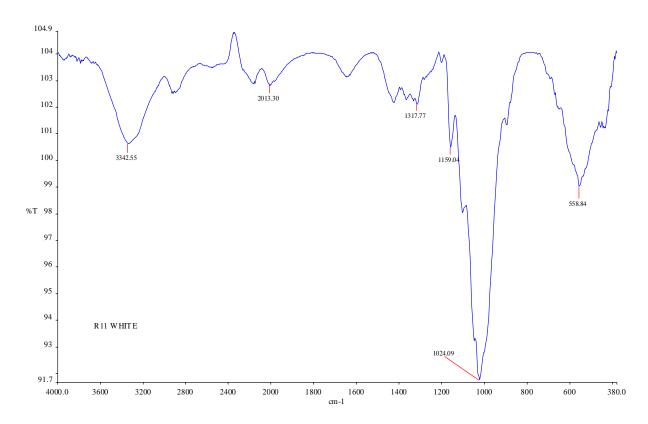


Figure 3.14 FTIR spectra of the stamped paper tray (plasticized NC-sorbitol coated substrate, three layers).

## 3.4 Conclusion

The mechanical properties of biodegradable packaging materials were compared to those of conventional packaging and it was shown that those of the conventional packaging materials were strong throughout the storage duration, while those of the biodegradable packaging materials were significantly affected by the storage conditions. The difference between the cold and ambient storage conditions for conventional packaging was not significant. The cold storage conditions significantly reduced the mechanical properties, such as the tensile strength and modulus of elasticity, compared to those under ambient storage conditions. The tensile strength was observed to be sensitive to the environmental conditions, with a reduction during cold storage conditions, compared to those under ambient storage conditions. The water uptake and solubility were generally lower for materials with cross-linking and sorbitol coating, which improved the mechanical properties of the stamped and glued paper trays. The pulped paper tray showed the highest solubility and water uptake, but decreased the tensile strength and modulus of elasticity throughout the storage duration. The incorporation of plasticizers improved the mechanical properties of the stamped and glued paper trays. The Edx was used in conjunction with scanning electron microscopy to determine the elemental/chemical

composition of biodegradable packaging material. The FTIR analysis confirmed the chemical composition, which showed similar spectra and strong carbon and oxygen bands in the material. The FTIR showed apparent stretching at 3000-2875 cm<sup>-1</sup> of C-H and absorption bands between 1450-1375, which originated from -C=O; these results concurred with the EDX findings. The water vapour permeability of biodegradable packaging materials was compared with those of conventional packaging materials, and the biodegradable packaging materials showed the highest water vapour permeability (stamped paper tray =  $9.3 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>, glued paper tray =  $8.9 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> and pulped paper tray =  $11.05 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>). The highest water vapour permeability was due to the heterogenous and porous structure confirmed by scanning electron microscopy. Overall, the addition of a plasticizer improved the mechanical properties and decreased the water uptake. These results show that the potential use of biodegradable packaging materials is a solution for the ecological problem of high plastic utilization.

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# 4. EVALUATION OF THE EFFECTIVENESS OF BIODEGRADABLE VS CONVENTIONAL PACKAGING MATERIALS IN SHELF-LIFE EXTENSION OF ROUND AND CHERRY TOMATOES

# Abstract

This study investigated the effects of biodegradable packaging and conventional packaging materials on the shelf-life extension of round and cherry tomatoes. The tomatoes were stored under cold (12°C) and ambient (20°C) conditions for 28 days. The quality attributes of tomatoes, such as their physiological weight loss, fruit firmness, Total Soluble Solids (TSS), pH, colour and marketability were assessed. Generally, the tomatoes stored under cold conditions were significantly (p<0.05) superior to those stored under ambient (20°C) conditions.

The results indicated that there was no significant difference (P>0.05) between the packaging groups with regard to the surface colour of the tomatoes. Biodegradable and conventional packaging materials generated a mean value hue angle of 45. The polypropylene perforated plastic bag gave the highest mean value of hue angles. The pulped paper tray (biodegradable), which was used for packaging the cherry tomatoes, gave the lowest Standard test methods for water vapor transmission of materials hue angle mean value of 36. The Kramer shearing force for round tomatoes was the highest for the stamped paper tray covered with PVC cling wrap (11 N.g<sup>-1</sup>), followed by the polypropylene perforated plastic bag (9 N.g<sup>-1</sup>), which is a conventional material. For cherry tomatoes, the glued paper tray gave the highest Kramer searing force of 8.5 N.g<sup>-1</sup>. The puncture test for the stamped paper tray covered with flow wrap gave the highest mean value of 7.8 N. The glued paper tray for cherry tomatoes gave the highest puncture force of 6.2 N, followed by the polypropylene perforated plastic bag, with 5.6 N. However, no significant difference (P>0.05) was observed between the conventional and biodegradable materials, in terms of firmness. The pH and TSS of tomatoes increased throughout the duration of the storage period. There was no significant difference (P>0.05) in the pH across the packaging materials. The Zipo punnets for cherry tomatoes gave the highest TSS of 4.8 Brix<sup>o</sup> under ambient conditions and 4.55 Brix<sup>o</sup> under cold storage conditions. For round tomatoes, the polypropylene perforated plastic bag gave the highest TSS of 5.1 Brix<sup>o</sup> under ambient storage conditions and 4.8 Brix<sup>o</sup> under cold conditions. The highest physiological weight loss (13.5%) was recorded by polypropylene plastic bags (15.64%), followed by EPT-F (expanded polystyrene covered with flow wrap) (14.39%).

The highest marketability was recorded for the stamped paper tray covered with flow wrap (82%) under cold storage conditions, and 60% under ambient conditions. Polypropylene perforated plastic bags had the lowest marketability of 10% under ambient storage conditions and 20.12% under cold conditions. For cherry tomatoes, the pulped paper tray had the highest marketability of 77% under cold conditions and 63% under ambient conditions. The Zipo punnet had the lowest marketability of 62.04% under cold conditions and 42.18% under ambient conditions. The overall analysis of the results shows that the biodegradable packaging and cold storage treatment extended the shelf-life of tomatoes. Tomatoes packed in biodegradable materials showed better physiochemical attributes, compared to those packed in conventional materials. The sections below show that the effects of biodegradable and conventional packages were only noticeable with respect to the physiological weight loss and marketability of tomatoes. For the other variables, these two packaging groups did not have any remarkable influence.

## 4.1 Introduction

The tomato is a perishable climacteric fruit that requires specific conditions for maintaining its freshness from the farm to the table (Azmai et al., 2019). Tomatoes are one of the most valuable and nutritious crops (Arah et al., 2015). Tomatoes are inherently perishable which make them deteriorate fast during postharvest value chain. As means of counteracting such losses tomatoes are harvested as early mature green, however, mature green tomatoes cannot be stored at temperatures less than 10 °C as this causes chilling injuries on the fruit. A peak in respiration, as well as the increased production of ethylene and CO2, is associated with their ripening. Their loss of quality is accelerated as a result of the physicochemical changes in respiration (Akbudak et al., 2007; Munhuewyi, 2012). In the fresh market tomato supply chain, post-harvest losses have been found to be similar across sub-Saharan African countries, with 9.50% in East Africa, 9.80% in central and southern Africa, and 10% in West Africa. Furthermore, the post-harvest losses of the individual countries were reported as being 10.10% in Kenya, 10.20% in South Africa and 13.40% in Nigeria (Sibomana et al., 2016). Round tomatoes are susceptible to mechanical damage and decay during post-harvest storage, transport and marketing. Therefore, they are subject to substantial post-harvest losses (PHL) in the tomato supply chain (Zeng et al., 2020). A cherry tomato salad is known as saint fruit is one of the top-four preferred fruits globally. Compared to the regular round tomatoes, cherry tomatoes contain 1.7 times more ascorbic acid and they are brightly coloured (Hu et al., 2012; Zeng et al., 2020). Cherry tomato also has a health-care effect of enhancing immunity, delaying aging, lowering blood pressure, lowering cholesterol and preventing cancer. Cherry tomato belongs to a kind of typical climacteric fruit, which has thin skin, soft and juicy texture, postharvest strong vitality and obvious post-ripening phenomenon, and then becomes soft and rotten after harvest (Zeng et al., 2020).

The food industry is currently searching for biodegradable and bio-friendly materials to replace the petroleum-based packaging materials. The use of synthetic plastics in food packaging can adversely affect the climate and the environment (Muller *et al.*, 2017). Hence, alternative ecofriendly packaging materials are now attracting attention. Muller *et al.* (2017) also reported that plastic food packaging materials can be replaced with polylactic acid and starch. There are a variety of challenges facing the food industry, including climate change, consumer safety concerns and government policies and legislation (Bader and Rahimifard, 2018). The ZZ2 Farm in South Africa has developed four different types of biodegradable packaging materials for cherry tomatoes and round tomatoes, in order to extend their shelf-life and maintain their quality.

The food industry is currently seeking to replace the petroleum-based packaging materials with biodegradable and bio-friendly materials. The utilisation of synthetic plastic materials in food packaging can have an adverse effect on the climate and the environment (Muller *et al.*, 2017); hence, alternative eco-friendly packaging materials are being given attention. Muller *et al.* (2017) also reported that polylactic acid and starch are the potential materials that will be used to replace synthetic polymer films i.e. plastic food packaging materials. The food industry is facing a range of challenges related to climate change, the increasing consumer safety demands and issues relating to government policies and legislative requirements (Bader and Rahimifard, 2018). The aim and objective of this study is to select the best suitable biodegradable packaging materials that can be utilized for cherry and round tomatoes in South Africa.

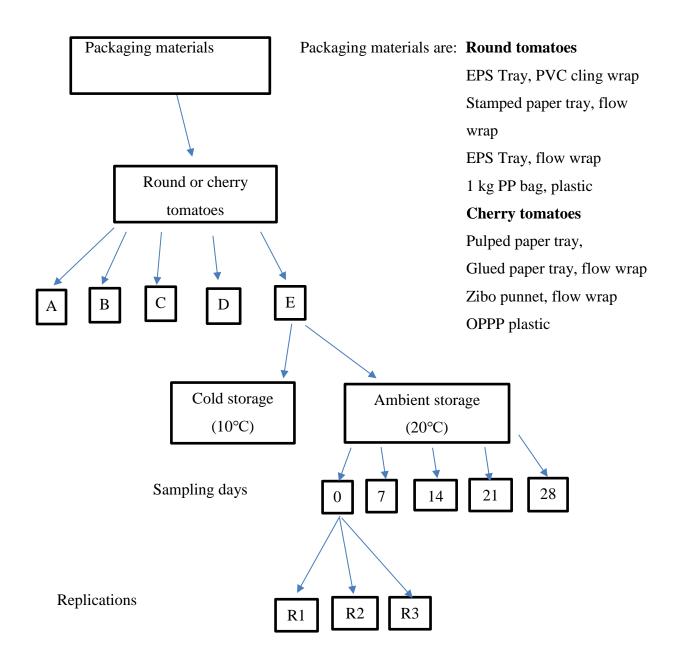
#### 4.2 Materials and Methods

## 4.2.1 Tomatoes samples

ZZ2 cherry and round tomatoes that were ready for market were packed at the Lanseria Pack House, Johannesburg. The tomatoes were supplied by the ZZ2 Farm in South Africa. A track truck with no cooling system, with an average temperature of 22.60°C and a relative humidity (RH) of 78.10%, was used to transport the tomatoes under ambient conditions to the Pietermaritzburg Fresh Produce Market, South Africa. On the other hand, the tomatoes under cold treatment were transported to the same Market by using a refrigerated truck that had an average temperature of 17.08°C (14.23-19.94°C) and an average RH of 73.77% (58.22-89.32%). The samples were then immediately transported from the Pietermaritzburg Fresh Produce Market to a laboratory at the University of KwaZulu-Natal (UKZN), Pietermaritzburg. The cold room was maintained at an average temperature of 12°C and a RH of 79.55%, while the average ambient temperature was 20°C and the average RH was 60.06%.

### 4.2.2 Experimental design

The experimental design was a factorial type of a randomized complete box, with the specified factors being packaging materials and the tomato types (round and cherry tomatoes), two storage conditions (ambient and cold storage), five sampling day and three replications. The quality parameters to be measured were arranged in the form of a split-plot design, with the tomato types and packaging materials being the main groups, storage condition as sub plot with random allocation of treatmets. The experimental design layout is shown in Figure 4.1.



Where: A= colour

B = engineering properties (Puncture, Kramer shear, compression)

C = physiological weight loss

- D = marketability
- E = PH and TSS

Figure 4.1 The experimental design layout

# 4.2.3 Data collection

The quality parameter assessment of tomatoes was carried out over a 28-day period, with sampling on Day 0, and after 7, 14, 21 and 28 days of storage. The on-site assessment of these quality parameters was conducted as follows:

# 4.2.3.1 Physiological weight loss

The Physiological Weight Loss (PWL) was determined gravimetrically. The change in weight of the samples was recorded every seven days of the storage period and converted to a percentage of the initial weight. The cumulative PWL (%) was expressed as a percentage, with respect to the storage period (Tefera *et al.*, 2007). This can be seen in Equation 4.1 below:

Physiological weight loss (PWL) = 
$$\frac{weight_{t=0} - weight_{t=t}}{weight_{t=0}} \times 100$$
 (4.1)

Where:

Physiological weight loss (PWL) = the percentage of weight loss of the sample tomato (%), Weight (t=0) = the initial weight of the sample tomato (kg), and Weight (t=t) = the weight of the sample tomato at time t (days of storage).

#### 4.2.3.2 Puncture test

The puncture test was measured by using a Texture Analyzer (Instron Universal Testing Machine (Model 3345), Buck, United Kingdom), as described by (Sirisomboon *et al.*, 2012a). The texture analyzer was fitted with a 2 mm probe and set at a speed of 3 mm.sec-1 and a 7.5 mm penetration depth for round tomatoes, and a speed of 1.5 mm.sec-1 and a 6 mm penetration depth for cherry tomatoes. The texture analyser was equipped with a 10 kg (100 N) load cell and all the data were automatically recorded by using the Easy-Match-QC software. The maximum force-deformation was recorded for both the round and cherry tomato samples.

# 4.2.3.3 Kramer shear

The Kramer shear was measured by using a Texture Analyzer (Instron Universal Testing Machine (Model 3345), Buck, United Kingdom). The tomatoes were cut into 10 mm round slices for each sample, using a Vanier caliper and a knife. The 10 mm slices were weighed and then positioned into a sample chamber where the shear press plate pressed the disk at a 10 mm.min-1 speed, with a shear press that was equipped with a 300 N load cell. The maximum

force applied was recorded and divided by the weight of the tomato sample disk, to accommodate for the difference in the area of the tissue cut by the plates (Harker *et al.*, 1997).

# 4.2.3.4 Compression

An Instron Universal Testing Machine (Model 3345, Instron, India), with a capacity of 5 KN set at a crosshead speed of 10 mm min-1, was used for the compression test. A 55 mm circular compression plate was used to compress the fruit. The tomatoes were laid out horizontally on a smooth surface, starting with the stem end and ending at the apex. Bluehill Instron data acquisition software was used to record the measurements (Sirisomboon *et al.*, 2012b).

## 4.2.3.5 Colour

The colour was measured, as described by Dominguez *et al.* (2012). The colour parameters L\*, a\* and b\* were measured by using a Minolta chromameter (Minolta CR-300, Ramsey, NJ, USA). Thereafter, the L\*, a\* and b\* were used to calculate the hue angle ( $h^\circ$ ) and chroma (C), based on the individual model formula (Domínguez *et al.*, 2012).

# 4.2.3.6 TSS

The TSS of the tomatoes was determined by using a digital refractometer (PAL-3 model, ATAGO, USA) after calibration, as well as distilled water. The tomatoes were cut with a sterile knife and the juice from the pulp was squeezed out by using gauze. The juice was placed on the prism of the refractometer and the value was read directly from the instrument (Mekonnen, 2017).

### 4.2.3.7 PH

An aliquot of juice was extracted by blending the tomatoes and transferring it into a beaker. The beakers were cleaned by using distilled water. The pH meter (PHS-3C model, Shanghai Puchun Measure Instrument Co. Ltd, China) was inserted into the liquid. Each of the tomato samples had its pH value determined by using a glass electrode pH meter that was calibrated before use with a 4.0 and 7 pH buffer (Domínguez *et al.*, 2012).

# 4.2.3.8 Marketability percentage

The marketable quantity of the tomato was subjectively assessed by a procedure suggested by Tigist *et al.* (2013). These descriptive quality attributes were determined subjectively by observing the visible level of mould growth, decay, shriveling, smoothness. A 1-5 rating scale was used to evaluate the tomato quality, with 1 = unusable, 2 = usable, 3 = fair, 4 = good, 5 =

excellent. Those receiving a rating of 3 and above were considered to be marketable. The number of marketable tomatoes were used as a measure for calculating the percentage of marketable tomatoes during storage. After subjectively assessing the product, this was calculated by using the Equation 4.2 below:

 $Percentage\ marketability = \frac{number\ of\ marketable\ tomato\ fruit}{total\ number\ of\ sampled\ tomato\ fruit} \times 100$  4.2

# 4.2.4 Data analysis

The data were analysed by using the Genstat 18th edition A VSNI. The data were treated with the one-way Analysis of Variance (ANOVA) at a 95% confidence level and with differences of P<0.05 considered to be statistically significant. Graphical representations were made by using Excel.

## 4.3 **Results and Discussion**

## 4.3.1 Colour

The tomato colour was measured by the hue angle on a  $360^{\circ}$  colour space. The  $90^{\circ}$  angle is assigned to a yellow hue, the  $180^{\circ}$  angle to a green hue, the  $270^{\circ}$  angle to a blue hue and the  $0^{\circ}$  angle to a red hue (Pinheiro *et al.*, 2013). With the progression of ripening, as the storage period progressed, the colour of the tomatoes changed from pink to red. There was a significant reduction (p<0.05) in the hue<sup>o</sup> between the successive sampling days over the storage duration Similar results were observed by Wang *et al.* (2011), where the tomato hue angle decreased continuously with the ripening of the tomatoes. The storage conditions made a significant (p<0.05) difference to the colour surface of the tomatoes, where those stored under cold storage conditions resulted in high hue<sup>o</sup> values (Figures 4.2b and c).

The groups that were compared were the biodegradable packaging material types in Figure 4.2b. The Stamped Paper Tray covered with PVC cling wrap (SPT-P) and the Stamped Paper Tray covered with Flow Wrap (SPT-F) made the surface colour of tomatoes yellower. On the other hand, the Glued Paper Tray covered with Flow Wrap (GPT) and the Pulped Paper Tray covered with a Zipo PET lid (PPT) made the surface colour of tomatoes redder. In Figure 4.2a, the groups being compared were the different types of conventional packaging materials. The Expanded Polystyrene Tray covered with Flow Wrap (EPT) and Polypropylene Perforated Plastic (PPP) altered the redness of the tomatoes. In contrast the Zipo Punnet covered with Flow Wrap (ZP) improved the surface colour of the tomatoes, tending them to look redder. Some packages (SPT-P and SPT-F) increase the hue angle of the tomatoes much more than

other packages (GPT and PPT). In Figure 4.2b, the GPT and SPT-P packaging materials had a hue angle of about 45.5 h° and 45.7h°, which generated a yellower surface. In contrast, SPT-F and PPT gave a hue angle about 43.5 h° and 44 h°, respectively. According to the RGB hue colour transformation scale, these values represent an orange-like aspect of the tomatoes. In Figure 4.2c and d, the cold storage conditions increased the hue<sup>o</sup> for both cherry and round tomatoes. Generally-speaking, the tomatoes stored in cold storage with conventional and biodegradable packaging materials had a lower hue<sup>o</sup> than those stored under ambient storage conditions. This means that the rate of ripening was slower in cold storage having an extended shelf-life. Similar results were reported by Tilahun (2010) showed that the rate of colour change was reduced significantly (p<0.05) by the cooling system. The difference between biodegradable and conventional packaging material was not significant (p>0.05). The hue angle of round tomatoes was generally high throughout the storage period (Figure 4.3b), because the surface area of round tomatoes is more extensive than that of cherry tomatoes.

One of the most critical quality attributes that buyers and consumers are interested in before buying them is their surface colour. The redness of tomatoes shows its ripeness or maturity (Wang *et al.*, 2011) and, for most people, colour is the only measure that shows their maturity. For marketing purposes, their redness can substantially influence the sales. However, this study has proved that the SPT-F and SPT-P packaging materials lowered the redness of tomatoes by increasing their yellowness. If this parameter alone could be sufficient for selecting which packaging material is the best, it would be advisable to use the GPT and PPT packaging materials, as they favor, or tend to have, a red-like colouration. The mean value of the hue angle for cherry tomatoes is around 37 h° (Figure 4.3b), while that of round tomatoes is around 51 h°. The hue angle of cherry and round tomatoes over the 28-day storage period is highly significant (p<0.01). This can be interpreted in the following way: the mean value of the hue colour of cherry tomatoes is statistically different from that of round tomato. Practically, this means that biodegradable packaging materials statistically decrease the hue colour of cherry tomatoes is in round tomatoes. These findings revealed that the storage condition has an influence on the shelf-life extension of tomatoes.

(a)

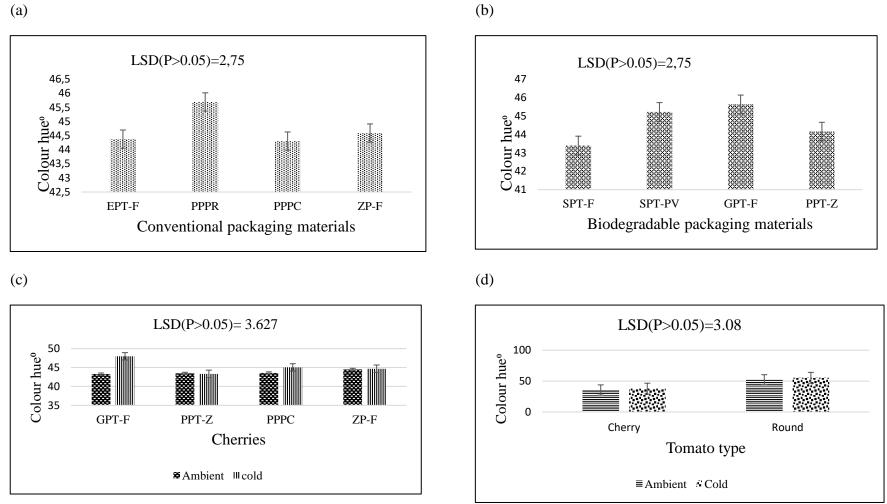


Figure 4.2(a) The effects of conventional materials, (b) biodegradable materials on the colour hue angle. (c) Cherry packages and (d) round packages against storage conditions.

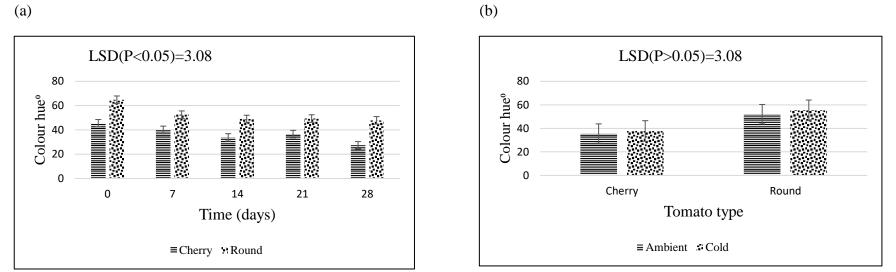


Figure 4.3(a) Variations of cherry and round against storage days. (b)The comparison of cherry and round tomatoes against storage conditions

#### **4.3.2** Engineering properties (Puncture, Kramer, Compression)

Over the storage period, the firmness of the tomatoes gradually decreased and they became significantly (p<0.05) softer with every successive sampling day. The rate of firmness reduction was comparatively higher for samples stored under ambient conditions, compared to those stored under cold storage conditions; as a result of the temperature difference between the two storage conditions, this is a physiological phenomenon (Figure 4.3). Similar results were reported by Tilahun (2010), where tomatoes stored in a cooling system remained firmer than those stored under ambient conditions. This suggests that the rate of firmness degradation in tomatoes depends on the temperature during storage and on the maturity stage. With regard to the engineering properties of biodegradable packaging materials, the SPT-P hardened the tomatoes, with a highest mean shearing force of about 11 N/g (Figure 4.3f). PPT packaging materials softened the tomatoes, with a lowest mean shearing force of about 4.5 N/g. As for the firmness of tomatoes, the puncture force was the highest in the SPT-F and SPT-P packaging materials under both storage conditions (Figure 4.3a and b). The PPT for cherry tomatoes had the lowest puncture force at the end of the 28-day storage period, compared to the GPT. Regarding the compression force, which is also a measure of tomato's firmness, the SPT-F and SPT-P materials remarkably increased the compression of tomatoes in cold storage (Figure 4.4d), compared to those in ambient storage (Figure 4.4c). This was unlike those packed in GPT and PPT, which sensibly lowered the compression force of the tomatoes. In general, cherry tomatoes had lower firmness values than round tomatoes, due to their smaller size. A higher firmness improves the quality of the fruit. This study revealed that it is paramount to also consider the various packaging material types, as they can affect the quality of the tomatoes. The compression and Kramer shearing forces showed a significant difference (p<0.05) between the packaging materials and the storage conditions. The choice of packaging is essential for improving or degrading the quality of tomatoes. The softening of tomatoes is a major problem because it may increase their susceptibility to damage, and the degree of firmness has long been considered as an indication of their quality. This may also be a criterion that consumers can use when purchasing a given set of tomatoes.

Globally, all tomatoes in conventional packaging (PPPR, PPPC, EPT-F and ZP) were observed to have the lowest firmness values at the end of the 28-day storage period, with the shearing force having very slight inter-group variations. The highest compression force was observed in the ZP tomatoes in cold storage (Figure 4.4d). Drastic difference shifts were observed in their compression properties. The PPPC and ZP conventional packaging materials lowered the compression of cherry tomatoes, while the EPT and PPPR materials increased the compression of round tomatoes (Figures 4.4c and d). The storage conditions also affected the Kramer shearing force of tomatoes, with cold conditions increasing the Kramer force, and ambient conditions decreasing it (Figure 4.5). The enzymatic disruption process of pectinesterase (PE) and polygalacturonase (PG) decreases the firmness of tomatoes (Tigist *et al.*, 2013).

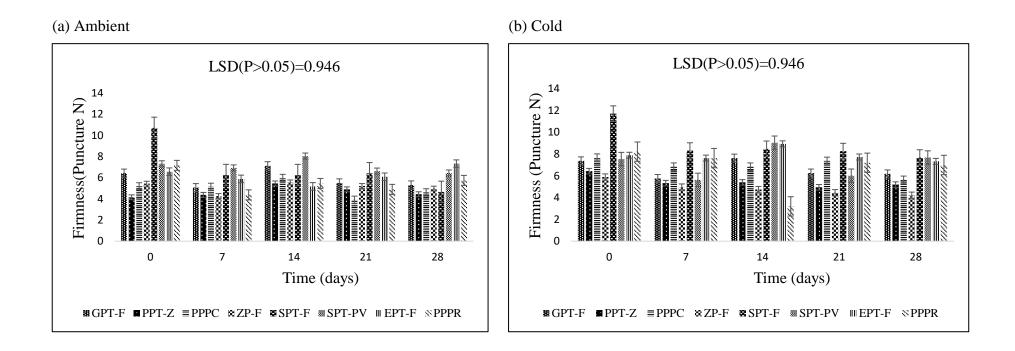


Figure 4.4 The effect of the storage conditions on the firmness (Puncture) of cherry and round tomatoes packed in biodegradable (SPT-F, SPT-PV, GPT-F, PPT-Z) and conventional (PPPC, ZP-F, EPT-F, PPPR) packaging materials

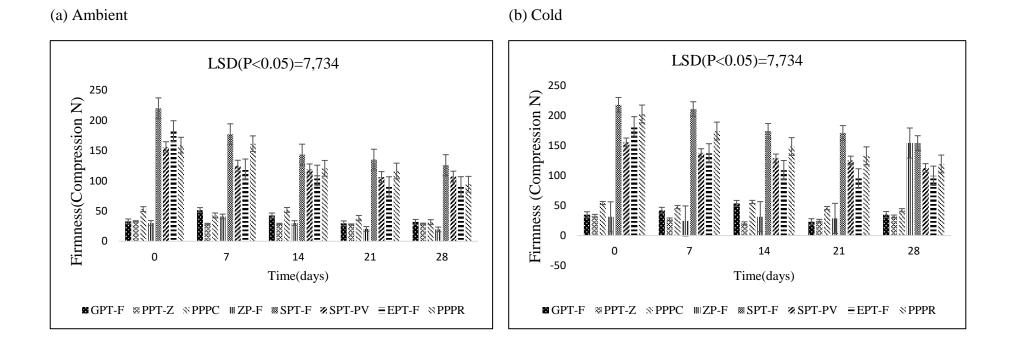


Figure 4.5 The effect of the storage conditions on the firmness (compression) of cherry and round tomatoes packed in biodegradable (SPT-F, SPT-PV, GPT-F, PPT-Z) and conventional (PPPC, ZP-F, EPT-F, PPPR) packaging materials.

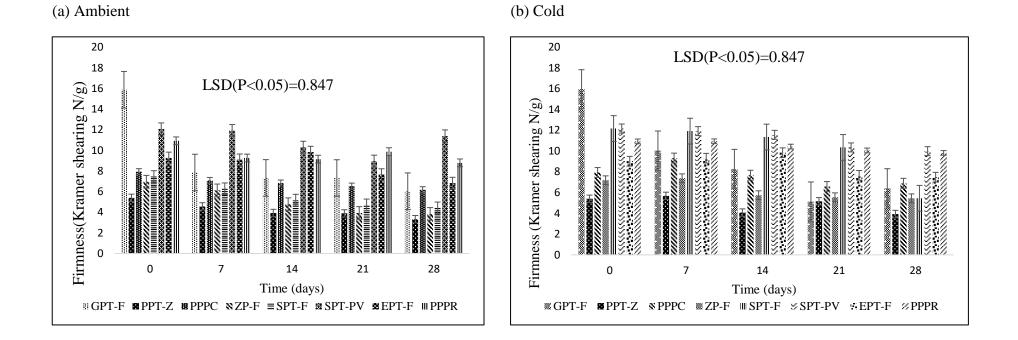


Figure 4.6 The effect of the storage conditions on the firmness (Kramer shearing) of cherry and round tomatoes packed in biodegradable (SPT-F, SPT-PV, GPT-F, PPT-Z) and conventional (PPPC, ZP-F, EPT-F, PPPR) packaging materials.

# 4.3.3 Physiological weight loss

There was a significant (p<0.05) and exponential increase in weight loss, between the successive sampling days, in tomatoes packed in both biodegradable and conventional packaging (Figure 4.6a). Similarly, the weight loss of the tomato samples stored under ambient conditions was significantly (p<0.01) higher, compared to those stored under cold conditions (Figure 4.8a). PWL increased progressively over the period of storage and the highest values were reached on the last day of observation. There was continuous loss of moisture over time due to transpiration from the tomatoes and respiration under ambient conditions. This is the reason was PWL increased with storage period as the tomato fruit continues to ripen. The PWL was more pronounced under ambient conditions implying that senescence may occur earlier and, therefore, result in a shorter shelf life. Weight loss and water loss in tomatoes are primarily determined by the storage temperatures, with higher storage temperatures causing more significant losses. The weight loss in tomatoes is primarily driven by evapotranspiration (Arah et al., 2015) and the rate of respiration. These processes are both RH and temperaturedependent. This study shows that samples stored under ambient conditions had a higher weight loss than those stored under cold conditions (Figures 4.8a and b). The type of packaging also had a significant (p<0.05) effect on their physiological weight loss (Figures 4.6a and b). This observation is consistent with findings of Haile and Safawo (2018), who compared the effects of different packaging materials on tomatoes. For round tomatoes in biodegradable packaging, the lowest physiological weight loss was recorded on the stamped paper tray cover with PVC cling wrap and the stamped paper tray covered with flow wrap (5.02%), see Figure 4.7. On the other hand, for cherry tomatoes in biodegradable packaging, the lowest physiological weight loss was recorded on the pulped paper tray (8%). The highest physiological weight loss was recorded in polypropylene plastic (15.64%), followed by EPT-F (expanded polystyrene covered with flow wrap (14.39%).

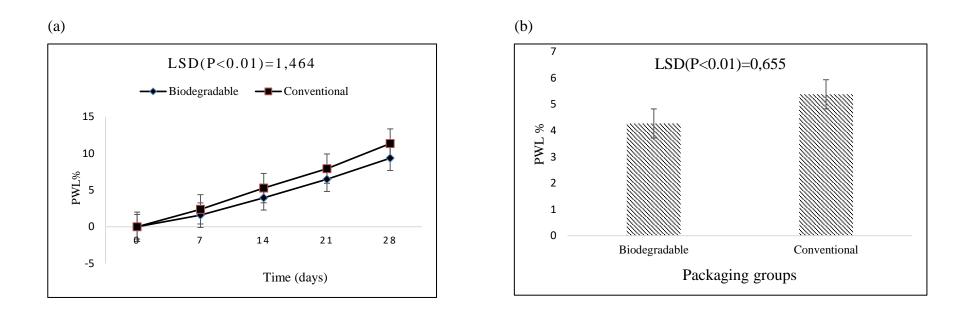


Figure 4.7(a) Weight loss of tomatoes packed in biodegradable and conventional packages over 28 days, (b)comparison of biodegradable vs conventional packaging

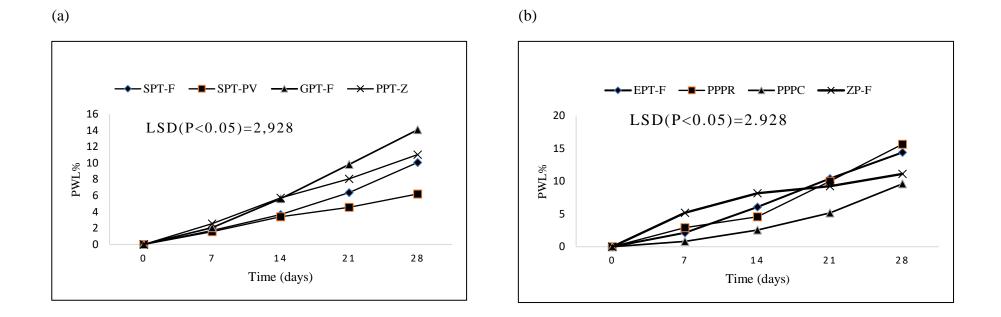


Figure 4.8(a) effects of packaging on weight loss of tomatoes during 28 days for biodegradable and (b) conventional materials.

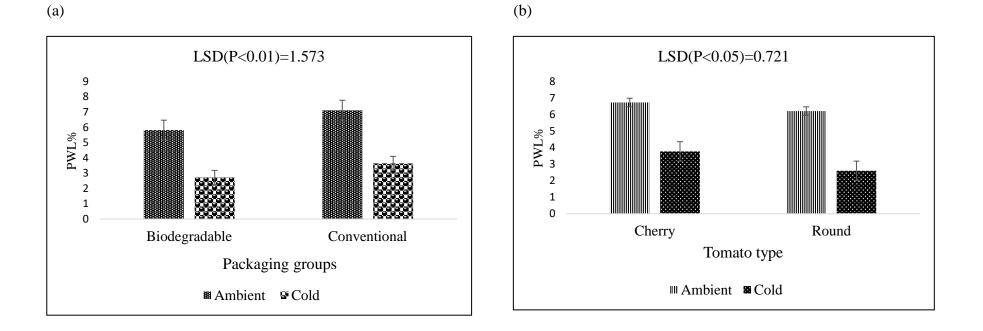


Figure 4.9 The effects of storage condition between (a) biodegradable vs conventional material and (b) cherry and round tomatoes.

The physiological weight loss of tomatoes was affected by the storage temperature and storage duration (p<0.001). An increasing trend in physiological weight loss was observed in cherry and round tomatoes that were stored at 12°C and 20°C, over a 28-day storage period. Tomatoes stored under ambient conditions (20°C 60.06%RH) lost weight quicker than those stored under cold conditions (12°C, 79.55% RH). The quantity of water in the product can also be decreased under low relative humidity at an ambient temperature, which accelerates the water evaporation process, hence the high weight loss under ambient conditions. Similar results were reported by Al-Dairi et al. (2021b). Moreover, the highest weight reduction was significantly higher in tomatoes stored under ambient conditions, in both biodegradable and conventional packaging, as shown by Figures 4.8a and b. At the end of the 28-day storage period, the round and cherry tomatoes stored at cold temperatures conditions experienced a weight loss of 2.60% and 3.77%, respectively, while those stored under ambient conditions experienced a weight loss of 6.2% and 6.72%, respectively. Tomato samples stored at an ambient temperature increased their weight loss, due to high relative humidity above the water activity. Respiration and withering resulted in water loss, which led to an increase in the physical barriers between the fresh produce and the surrounding air (Haile and Safawo, 2018). As a function of pressure, air temperature and relative humidity, transpiration occurs when the vapour pressure is deficient. In addition, respiration can lead to an increase in weight loss, as carbon (C) atoms are changed into atmospheric carbon dioxide (CO2) (Munhuewyi, 2012). An increase in the transpiration rate in tomatoes that are stored at higher temperatures, leads to shrivelling and wilting, thus reducing their consumer acceptability and market level (Al-Dairi et al., 2021a). This study found that low temperatures of 12°C increased the tomato weight, due to the increase in vapour pressure and water retention. Therefore, the PWL of tomatoes is greatly influenced by the storage temperature and relative humidity surrounding the produce (Tilahun, 2010). A high weight loss reduces the rate of soluble solids, water content and mineral components. It is therefore advisable to minimise the physiological weight loss of tomatoes as much as possible, in order to preserve their quality. In this regard, biodegradable packages are good, given that they caused an average loss of 4.3% in the global weight of tomatoes. This is satisfactory nowadays, because biodegradable packaging materials are being promoted for their ecologically-oriented benefits.

## 4.3.4 pH

Over the storage period, the pH of the tomatoes gradually increased, with significant changes (p<0.05) being observed between the successive sampling days. The ambient and cold storage conditions significantly (p<0.05) influenced the pH of tomatoes stored in biodegradable and

conventional packaging. The pH of the tomatoes increased with the storage period, under all treatment conditions (Table 4.1). In addition, the pH values of samples stored under ambient conditions were higher than those stored under cold conditions, which suggests that the pH is a valuable indicator of deterioration and that samples with a higher pH are expected to be nearing their senescence. Over the storage period, the pH increases, partly due to the fruit ripening, which causes the acid content to decrease, due to its conversion to sugar by means of gluconeogenesis (Cherono et al., 2018). The normal pH range for tomatoes is between 4.0 and 4.5 (Arah et al., 2015), and those packed in a polypropylene plastic bag (PPPR) had the highest pH of all the packaging materials (5.05), which is far beyond the acceptable range. This means that all the acidity of the product decreases while the sugar content increases through the process of glycolysis.. In addition, the cold conditions lowered the pH of the tomatoes to an average of 4. Other variables, such as the puncture, compression and total soluble solids of tomatoes, were not significant (p>0.05), which means that they are not affected by the storage conditions. The acid content of tomatoes influences their pH, which is an important quality parameter. They are a low pH fruit and this has an impact on their resistance to microbial attack and on their sensory qualities. In general, it is desirable to maintain the pH of tomatoes at an optimum level during storage (a pH of 4.25), as higher pH values alter their flavour (Macheka et al., 2018).

days	Storage conditions	Biodegradable materials (round tomatoes)		Conventional materials (round tomatoes)		Biodegradable materials (cherry tomatoes)		Conventional packaging (cherry tomatoes)			
		SPT-F	SPT-PV	EPT-F	PPPR	GPT-F	РРТ	ZP-F	PPPC		
0	Ambient	4.20±0.02 <sup>nopq</sup>	4.29±0.0200 <sup>nopq</sup>	4.21±0.02 <sup>nopqr</sup>	4.25±0.01 <sup>pqrst</sup>	3.42±0.02°	3.30±0.02 <sup>ab</sup>	3.23±0.01ª	3.28±0.03 <sup>ab</sup>		
	Cold	4.10±0.02 <sup>rstuv</sup>	4.29±0.02 <sup>rstu</sup>	4.23±0.01 nopqr	4.23±0.09 <sup>stuvw</sup>	3.42±0.02°	3.30±0.02 <sup>ab</sup>	3.23±0.01ª	3.28±0.03 <sup>ab</sup>		
7	Ambient	4.22±0.04 <sup>nopqr</sup>	4.32±0.11 <sup>lm</sup>	4.15±0.04 <sup>mn</sup>	4.28±0.03 <sup>mnop</sup>	3.81±0.03 <sup>f</sup>	3.84±0.03 <sup>d</sup>	3.84±0.01 <sup>fg</sup>	3.79±0.09 <sup>ef</sup>		
	Cold	4.15±0.00 <sup>jkl</sup>	4.30±0.01 <sup>nopq</sup>	4.36±0.13 <sup>bc</sup>	4.24±0.03 <sup>efg</sup>	3.77±0.04 <sup>ef</sup>	3.62±0.02 <sup>fh</sup>	3.72±0.03 <sup>e</sup>	3.83±0.09 <sup>ef</sup>		
14	Ambient	4.26±0.03 <sup>nopqr</sup>	4.40±0.11 <sup>mn</sup>	4.20±0.04 <sup>ij</sup>	4.30±0.05 <sup>jk</sup>	4.18±0.02 <sup>mnop</sup>	4.05±0.02 <sup>kl</sup>	4.15±0.02 <sup>mn</sup>	4.22±0.01 <sup>mno</sup>		
	Cold	4.19±0.06 <sup>ijk</sup>	4.32±0.06 <sup>gi</sup>	4.01±0.05 <sup>f</sup>	4.28±0.01 <sup>ef</sup>	4.10±0.03 <sup>lm</sup>	4.02±0.02 <sup>jkl</sup>	4.04±0.02 <sup>kl</sup>	4.16±0.01 <sup>nopqr</sup>		
21	Ambient	4.56±0.05 <sup>h</sup>	4.55±0.02 <sup>fg</sup>	4.28±0.01 <sup>zab</sup>	4.55±0.01 <sup>zabc</sup>	4.25±0.02 <sup>nopqr</sup>	4.19±0.03 <sup>mop</sup>	4.18±0.03 <sup>mnop</sup>	4.22±0.12 opqrs		
	Cold	4.29±0.03 <sup>def</sup>	4.44±0.01 <sup>cdef</sup>	4.26±0.09 <sup>zab</sup>	4.31±0.03 <sup>gh</sup>	4.24±0.01 <sup>nopqr</sup>	4.16±0.02 <sup>mno</sup>	4.16±0.04 <sup>mno</sup>	4.24±0.02 <sup>nopqr</sup>		
28	Ambient	4.66±0.03 <sup>bcde</sup>	4.67±0.05 <sup>xxyz</sup>	4.50±0.02 <sup>qrst</sup>	5.04±0.03 <sup>1</sup>	4.67±0.02 <sup>abc</sup>	4.39±0.03 <sup>uwx</sup>	4.46±0.03yza	4.56±0.00 <sup>abc</sup>		
	Cold	4.63±0.01 <sup>rstuv</sup>	4.60±0.01 <sup>bcd</sup>	4.49±0.02 <sup>pqrs</sup>	4.31±0.04 <sup>mn</sup>	4.53±0.01 <sup>abc</sup>	4.37±0.02 <sup>wxy</sup>	4.32±0.04 <sup>tuvw</sup>	4.53±0.01 <sup>bcde</sup>		
Signific	cant level	1			1	1					
Packag	ing Treatment (A	A)	< 0.005								
Days (B) <0.001											
Storage period(C) <.001			<.001								
B*C			<.001								
A*B*C			=0.070								
A*B			= 0.011								
A*C			<.001								

Table 4.1 Variations of the mean pH of round and cherry tomatoes packed in biodegradable and conventional materials.

Means followed by the same letter(s) are not significant: Duncan's multiple range test (P < 0.05), LSD = 0.131, %CV = 8.9, s.e = 0.094

#### 4.3.5 Total Soluble Solids (TSS)

Figures 4.7 and 4.8 show that the total soluble solids of tomatoes were also strongly influenced by the packaging type. The highest Brix<sup>o</sup> were observed for the GPT and PPT packaging materials. It is known that the Total Soluble Solids (TSS) of tomatoes are affected by the storage conditions (Shezi, 2016), this study revealed that the packaging type can also substantially affect their solid attributes. Figures 4.7a and b show that the GPT and PPT remarkably increased the value of the total soluble solids in tomatoes. The TSS indicates the accumulation of carbohydrates, organic acids, proteins, fats and minerals in the fruit, and it is desirable to have, and to preserve, a high number of these soluble solids. This study proved that the GPT and PPT indicated the ripening status of tomatoes, which substantially increased their TSS. On the other hand, SPT-F and SPT-P decreased the amount of TSS (Figure 4.7a) and deteriorated the quality of the tomatoes. This finding is essential for the quality control of tomatoes. In Figure 4.8c, the total soluble solids of tomatoes packed in conventional materials were slightly higher than those in biodegradable packaging materials, but still the variations between groups were slight, though significant (p<0.05). The conventional packaging materials showed the highest TSS, compared to the biodegradable packaging materials. Tomatoes packed at ambient temperatures had a higher TSS than those stored under cold conditions (Figure 4.8b and d). This is because tomatoes mature faster at high temperatures, and the soluble solids, the major components of which are sucrose, glucose and fructose, increase as fruit ripens (Majidi et al., 2011). A longer shelf-life is desirable, particularly where long-distance shipments or exports are involved, so reducing the onset of the physiological, biochemical and chemical processes can be advantageous. The major components of soluble solids are sucrose, glucose and fructose, which increase as the fruit ripens (Lira et al., 2016).

(a)

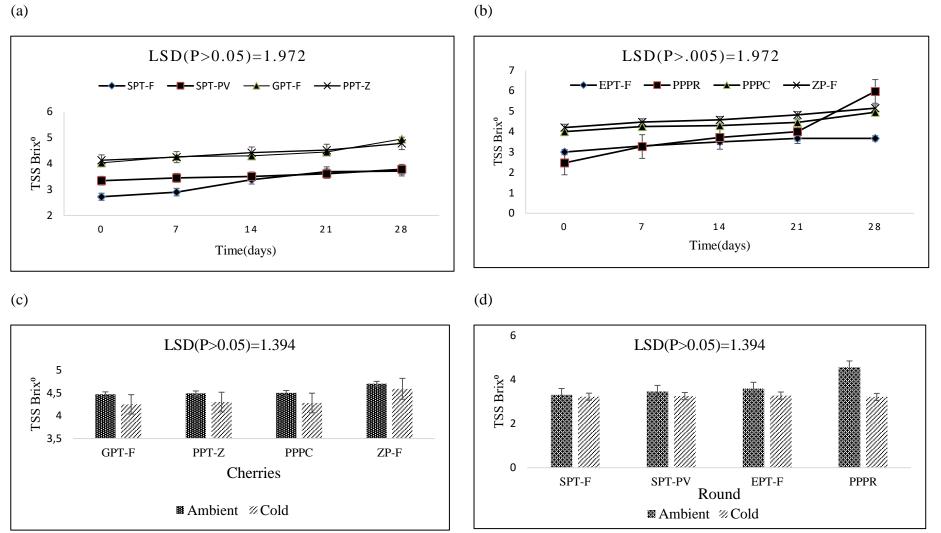


Figure 4.10 TSS comparison of individual packaging material against storage duration (a and b) and storage conditions (c and d).

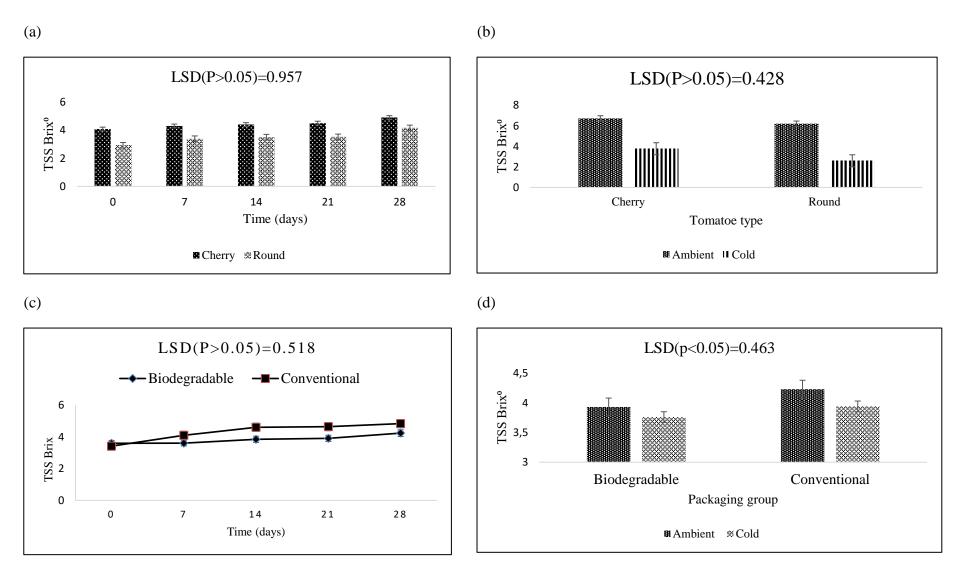


Figure 4.11 Comparison of the mean values of the TSS of biodegradable vs conventional packaging (c and d) for cherry and round tomatoes (a and b) against storage condition and storage duration

Tomatoes are currently among the most widely-grown plants worldwide (Haile and Safawo, 2018). From production to consumption, several parameters affect their chemical, engineering and dietetic properties. These parameters include the transportation conditions, the cold chain, the storage duration, their exposure to microbial agents, among others. In this study, it was revealed that, in addition to the currently-known parameters, the packaging type is also one of the most important parameters to consider.

## 4.3.6 Subjective analysis and marketability

A subjective analysis assessed the visibility tomatoes for moulds, wilting and bacterial growth. Tomatoes packed in biodegradable packaging showed minor defects Figure 4.13d, while those packed in convectional packaging appeared to have more defects. Figures 4.13a and b shows the bacterial growth in tomatoes packed in expanded polystyrene containers covered with flow wrap. On Day 14, some defects and mould started appearing in those packed in polypropylene plastic bags (Figure 4.13e). These were deemed to be unmarketable and were therefore discarded. At the end of the storage period, the tomatoes packed in biodegradable materials showed minimal defects, which resulted in their high marketability.

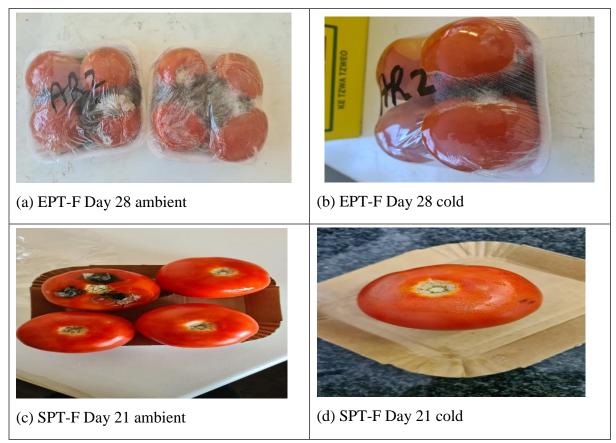




Figure 4.12 Photographs of representative tomato quality packed in biodegradable and conventional packaging under different storage conditions.

The marketability of tomatoes varied significantly (p<0.001) according to the packaging materials that were used. A decrease in the marketability percentage trends was observed for both cherry and round tomatoes in both biodegradable and conventional packaging over the storage period. Tomatoes packed in biodegradable packaging materials had the highest marketability percentage, with an average of 74.2%, while those packed with conventional packaging had a marketability percentage of 55%. The marketability percentage of tomatoes also varied significantly (P<0.05) with the storage conditions. Those that were stored under cold conditions retained a higher marketability percentage than those stored under ambient storage conditions. This could be caused by the efficiency of the cooling system, which reduced the metabolic activity rate taking place within the tomatoes and resulted in sustaining their quality, and thus their marketability (Tilahun, 2010). Similar results were reported by Cherono *et al.* (2018), where tomatoes stored under cold conditions retained a higher cold conditions retained a higher cold conditions retained a higher duality (Tilahun, 2010). Similar results were reported by Cherono *et al.* (2018), where tomatoes stored under cold conditions retained a higher duality than those stored under ambient conditions, thus making them highly marketable.

The differences between biodegradable and conventional packaging materials are demonstrated in Figure 4.14. The storage conditions made a significant difference (p<.005) on the marketability percentage of the tomatoes. At the end of the storage period, tomatoes packed in biodegradable materials (Figure 4.14a) showed minimal defects and this resulted in their high marketability. The round tomatoes in Figure 4.14a that were packed in the stamped paper tray covered with flow wrap and stored under cold conditions, had the highest marketability of 80.3%. In general, the round tomatoes packed in the stamped paper tray had the highest marketability, under both ambient and cold storage conditions, compared to the stamped paper tray covered with PVC cling wrap, while those packed in conventional packaging had the lowest marketability, as depicted in Figure 4.14b. Polypropylene plastic had the lowest marketability amongst all the packaging materials at the end of the storage period.

Similar results were also observed with cherry tomatoes (see Figure 4.15), where those packed in biodegradable packaging material (Figure 4.15a) showed the highest marketability percentage of 68%, while those packed in conventional packaging material (Figure 4.15b) had a marketability percentage of 56.89%. Polypropylene plastic also showed the lowest marketability percentage of 50% under ambient storage conditions and 64% under cold storage conditions. The Zipo punnet covered with flow wrap also showed the lowest marketability percentage of 51.52% under ambient storage conditions and 61.61% under cold storage conditions.

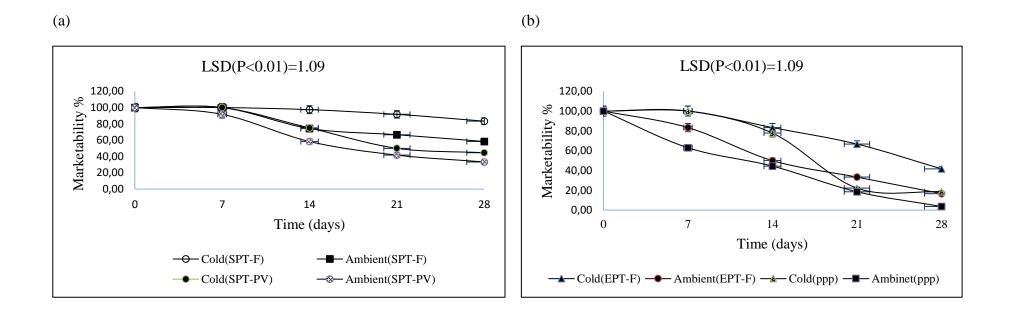


Figure 4.13 Marketability % of round tomatoes packed in (a) biodegradable and (b) conventional packaging materials that were stored at different environmental conditions (cold 12°C and ambient 20°C) for the 28-day period

(b) LSD(P<0.01)=1.09 LSD(P<0.01)=1.09 120,00 120,00 Marketability % 00'09 00'08 00,008 Marketability % 00'09 00'09 40,00 40,00 14 21 14 21 0 7 28 7 28 0 Time (days) Time (days)  $-\blacksquare-Cold(GPT-F) - \blacktriangle-Ambient(GPT-F) - \bullet-Cold(ppt) - \bullet-Ambient(ppt)$  $-\bullet$  Cold(ZP-F)  $-\bullet$  Ambient(ZP-F)  $-\bullet$  Cold(pppc)  $-\blacksquare$  Ambient(pppc)

Figure 4.14 Marketability % of cherry tomatoes packed in (a) biodegradable and (b) conventional packaging materials that were stored at different environmental conditions (cold 12°C and ambient 20°C) for the 28-day period.

(a)

### 4.4 Conclusion

This study investigated the effectiveness of biodegradable and conventional packaging materials on the extension of the shelf-life of cherry and round tomatoes. The results revealed that each category of packaging material had a significant influence on the extension of their shelf-life. For biodegradable packaging materials, GPT and PPT improved the redness of the surface colour of tomatoes, while SPT-P and SPT-F decreased it. In addition, SPT-F and SPT-P substantially improved their engineering properties, and the storage conditions strongly influenced the extension of their shelf-life.

With regard to conventional packaging materials, EPT and PPPR altered the redness of the tomatoes, while PPPC and ZP packaging improved it. While the PPPC and ZP packaging seemed to be good, they surprisingly decreased the firmness of tomatoes. The total soluble solids of tomatoes were high in conventional packaging materials.

The comparison between biodegradable and conventional packaging materials concluded that biodegradable packages are the best for minimizing the physiological weight loss of tomatoes. However, the suitability of biodegradable packages could only be observed in relation to their physiological weight loss, and neither of the two packaging groups showed conclusive performances with respect to the other response variables.

#### 4.5 References

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#### 5. CONCLUSION AND RECOMMENDATIONS

Tomatoes are one of the most commonly-cultivated crops in South Africa. They are grown for home consumption in almost every homestead in sub-Saharan Africa. They are a valuable source of vitamins and are an important cash crop for smallholders and medium-scale commercial farmers alike. Tomatoes are grown commercially wherever there are favourable agronomic conditions. They have become one of the most important crops in agriculture for smallholder farmers, both for processing and for the fresh market (Maliwichi *et al.*, 2014).

The objectives of this study were to investigate the effectiveness of biodegradable packaging materials on shelf life extension of tomatoes stored at different environmental condition, and to investigate the effect of storage temperature and time on physical and mechanical properties of biodegradable materials. In this study a randomized split plot experimental design was followed. Two variety of tomatoes (cherry and round) were used in this study, where eight quality parameters were determined and analyzed over 28 days storage period. The sampling was done on Days 0, 7, 14, 21 and 28. Tomatoes were stored under ambient condition ( $20^{\circ}$ C) and under cold condition ( $12^{\circ}$ C).

This study compared the effectiveness of biodegradable and conventional packaging materials on the extension of the shelf-life of tomatoes, stored under cold and ambient conditions. The biodegradable packaging materials were: Stamped paper tray covered with flow wrap, Stamped paper tray covered with PVC cling wrap, Glued paper tray covered with flow wrap and Pulped paper tray covered with zipo lid. The conventional packaging materials were: Expanded polystyrene tray covered with flow wrap, Polypropylene Perforated Plastics and Zipo punnet covered with flow wrap. The results indicated that there were no differences in the surface colour of tomatoes packaged in the biodegradable and conventional packaging materials.

Biodegradable packaging materials generated a mean value hue angle of 45 h°, and conventional packaging materials also generated a mean value of hue angle of 45 h°. The mean shearing force values were 7.7 N.g<sup>-1</sup> and 7.9 N.g<sup>-1</sup>, respectively, for the biodegradable and conventional packaging materials. A highly significant difference (p<0.01) was observed in the physiological weight loss and marketability of tomatoes. Unexpectedly, a highly significant (p<0.01) difference was observed with regard to their physiological weight loss and marketability, with the conventional materials causing a high physiological weight loss in tomatoes, while the biodegradable materials minimized it. A high loss in weight reduced the rate of soluble solids by 12% in conventional packaging and reduced by 5.1% in biodegradable

packaging (the water content and the mineral components). It is advisable to minimize the physiological weight loss of tomatoes as much as possible, in order to preserve their quality. In this regard, biodegradable packaging is good, given that it caused an average loss of 4.3% in the global weight of tomatoes. This is satisfactory because biodegradable packaging materials are currently being promoted for their ecologically-oriented benefits.

Chapter 4 showed that the effects of biodegradable and conventional packages were only perceptible with regard to the physiological weight loss and marketability of tomatoes. Biodegradable and conventional packaging groups did not exercise any remarkable influence on the other variables. As for the question of which packaging is good or bad, and which packaging can be recommended for use, it is essential to address this issue by analysing the individual effects of the packaging. The Polypropylene Perforated Plastics Round (PPPR) gave the highest mean value of the hue angles of 47°, which express the colouration of tomatoes. However, it is the smallest hue angle that depicts the redness of a tomato. Thus, the stamped paper tray covered with flow wrap (SPT-F) packaging material gave the slightest hue angle with a mean value of 43 h°. The Kramer force (11 N.g<sup>-1</sup>) was the highest in the stamped paper tray covered with PVC cling wrap (SPT-PV) packaging materials, which is a biodegradable material. PPPR gave 10 N.g<sup>-1</sup> as a mean shear force, which is not too different from the 11 N.g<sup>-1</sup> <sup>1</sup> of SPT-PV. In cherry tomatoes the highest Kramer force was recorded in glued paper tray covered with flow wrap (GPT-F). For Kramer shearing force the difference was not significant (p>0.05) in both cherry and round tomatoes. In terms of compression force the stamped paper tray covered with flow wrap (SPT-F) gave the highest force of 170 N. For cherry tomatoes the highest compression force of 44 N was recorded in polypropylene perforated plastic cherry (PPPC). The highest puncture force of 7.8 N was recorded in SPT-F for round tomatoes and 6.2 N in glued paper tray (GPT-F) for cherry tomatoes, however the difference was not significant (p>0.05) between biodegradable and conventional packaging materials. On average the puncture force for biodegrade material was 5.5 N compared to 4.95 of conventional materials. The packaging group did not have a significant influence (p>0.05) on the puncture. The pH of round and cherry tomatoes remained at the range of 3.9-4 and 4.1-4.4. The total soluble solids (TSS) of round tomatoes stored in biodegradable material (SPT-F and SPT-PV) had an average of 3.35 compared to 3.65 for tomatoes packed in conventional (EPT-F and PPPR) packaging materials. In cherry tomatoes TSS ranged between 4.4-.4.6, with Zipo punnet having a slightly high TSS of 4.6. For TSS the difference between biodegradable and conventional packaging materials was insignificant (p>0.05).

Biodegradable packaging materials creates a dry environment for tomatoes by absorbing the moisture from them, which results in a decreasing rate of condensation within the packaging material. This is because biodegradable packaging materials showed strong absorption bands in the FTIR spectra, as shown in Chapter 3. With stamped paper tray having the strongest absorption band at 3650-3250 cm<sup>-1</sup> followed by glued paper tray at 3338,98–3200 cm<sup>-1</sup>. Biodegradable packaging materials have the highest water vapour permeability (stamped paper tray =  $9.3 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>, glued paper tray =  $8.9 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> and pulped paper tray  $= 11.05 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>), which can be attributed to their more porous structure and irregular surface from scanning electron microscopy. The conventional packaging had the lowest WVP values (zipo punnet =  $6.04 \times 10^{-4}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>, expanded polystyrene =  $3.54 \times 10^{-3}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup> <sup>1</sup> and polypropylene = $2.13 \times 10^{-5}$  gm<sup>-1</sup>day<sup>-1</sup>Pa<sup>-1</sup>). The highest water vapour permeability was advantageous for controlling the condensation inside the packaging material, which resulted in the lowest physiological weight loss of the tomatoes packed in biodegradable packaging material. The cold storage environment had a positive impact on maintaining the quality of tomatoes for all the measured quality parameters; however, it was not the case for the mechanical properties. The cold storage conditions decreased the tensile strength of the materials over the storage period, compared to materials stored in an ambient environment. Even though biodegradable materials absorbed moisture to create a dry environment for the tomatoes, the mechanical properties were compromised.

The results showed that neither biodegradable nor conventional materials influenced the surface colour, the shearing force, puncture, compression, pH or the total soluble solids of tomatoes. To put it simply, none of the packaging materials was better than the other when it came to the surface colour, the shearing force, puncture, compression, pH or the total soluble solids of tomatoes. This study concludes that biodegradable packaging materials can be used to package tomatoes in small portions. The results revealed a highly significant difference (p<0.01) on the physiological weight loss and marketability percentages. The highest marketability percentage of 75.22% was recorded on stamped paper tray covered with flow wrap. On the other had for cherry tomatoes the highest marketability of 77.90% and 77.33% was recorded on glued paper tray covered with flow wrap and pulped paper tray covered with zipo lid.

The lowest physiological weight loss was recorded on stamped paper tray covered with PVC cling wrap (5.02%) and stamped paper tray covered with flow wrap 6.1%. For cherry tomatoes

the lowest physiological weight loss was recorded on pulped paper tray covered with Zipo lid (8%). The results revealed that biodegradable packaging and cold storage was the best treatment in extending the shelf life of tomatoes. It can be concluded that the stamped paper tray covered with PVC cling wrap for (round tomatoes) and the pulped paper tray covered with zipo lid (cherry tomatoes) resulted in a longer shelf-life. This study, therefore, recommends the use of Stamped paper tray covered with flow wrap and stamped paper tray covered with PVC cling wrap as they minimize the physiological weight loss. Although it was expected that the biodegradable materials would give promising results for all eight variables, unfortunately this was not the case.

#### Recommendation

It is recommended that a strong plasticizing agent should be added, to protect the material from softening and to improve the integrity of the packaging.

As the results of the present study did not identify a 'miraculous package', it therefore recommends that the 'suitability' of the packaging material should be treated individually.

While some individual packaging improved some of the quality aspects of tomatoes, at the same time, they decreased other quality aspects.

## 5.1 References

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# 5.2 Appendix

Table 5.1 The analysis of variance of tomato quality in response to different packaging material, storage period and days.

Parameters	Packaging	Days (B)	Storage (C)	B*C	A*B*C	A*B	A*C
	treatment (A)						
Color	ns	*	*	**	ns	ns	ns
Puncture	ns	**	*	***	ns	ns	ns
Kramer shear	*	***	**	**	**	**	*
Compression	*	**	**	*	*	*	*
TSS	*	ns	*	ns	ns	ns	**
PWL	***	***	***	***	***	***	***
Marketability	***	***	***	***	***	***	***

ns, \*, \*\*, \*\*\*,

ns: not significant,

\*: significant at P < 0.05,

\*\*: P < 0.01, and

\*\*\*: P < 0.001, respectively.