

EFFECTS OF NANOCELLULOSE ON FILM PROPERTIES OF EDIBLE FILM

Submitted in partial fulfilment of the requirements for the award of Bachelor of
Engineering degree in B.E Mechanical Engineering

By

SANKARA NARAYANAN K (38150902)

KIRAN KUMAR A (38150050)



**DEPARTMENT OF MECHANICAL ENGINEERING
SCHOOL OF MECHANICAL ENGINEERING**

SATHYABAMA

**INSTITUTE OF SCIENCE AND TECHNOLOGY
(DEEMED TO BE UNIVERSITY)**

ACCREDITED WITH GRADE "A" BY NAAC

JEPPIAAR NAGAR, RAJIV GANDHI SALAI, CHENNAI – 600 119.

MAY - 2022



SATHYABAMA

INSTITUTE OF SCIENCE AND TECHNOLOGY
(DEEMED TO BE UNIVERSITY)

Accredited "A" Grade by NAAC | 12B Status by UGC | Approved by AICTE

www.sathyabama.ac.in

DEPARTMENT OF MECHANICAL ENGINEERING

BONAFIDE CERTIFICATE

This is to certify that this Project Report is the bonafide work of **SANKARA NARAYANAN K (38150902)** and **KIRAN KUMAR A (38150050)** who carried out the project entitled "EFFECTS OF NANOCELLULOSE ON FILM PROPERTIES OF EDIBLE FILM" under our supervision from **August 21 to March 22**.

Internal Guide

Dr. J. JEYA JEEVAHAN, M.E, Ph.D.,

Head of the Department

Dr. G. ARUNKUMAR, M.E, Ph.D.,

Submitted for Viva voce Examination held on _____

Internal Examiner

External Examiner

DECLARATION

We, **SANKARA NARAYANAN K (38150902)** and **KIRAN KUMAR A (38150050)**, hereby declare that the project Report entitled **“EFFECTS OF NANOCELLULOSE ON FILM PROPERTIES OF EDIBLE FILM”** done by us under the guidance of **Dr. J. JEYA JEEVAHAN, M.E, Ph.D.,** (Internal) at Sathyabama Institute of Science and Technology, Jeppiaar nagar, Chennai – 600 119 is submitted in partial fulfilment of the requirement for the award Bachelor of Engineering degree in Mechanical Engineering.

DATE: 21/04/2022

PLACE: CHENNAI

1.



2.



SIGNATURE OF THE CANDIDATES

ACKNOWLEDGEMENT

We are pleased to acknowledge my sincere thanks to Board of Management of **SATHYABAMA** for their kind encouragement in doing this project and for completing it successfully. We are grateful to them.

We convey our sincere thanks to **Dr. S. PRAKASH, M.E, Ph.D.**, Dean, School of Mechanical Engineering and **Dr. G. ARUN KUMAR, M.E, Ph.D.**, Head of the Department, Department of Mechanical Engineering for providing us necessary support and details at the right time during the progressive reviews.

We would like to express our sincere and deep sense of gratitude to our project guide **Dr. J. JEYA JEEVAHAN, M.E, Ph.D.**, for his valuable guidance, suggestions and constant encouragement paved way for the successful completion of our project work.

We wish to express our thanks to all Teaching and Non-teaching staff members of the Department, parents and friends of who were helpful in many ways for the completion of the project.

ABSTRACT

Edible packaging is a sort of packing that can be eaten alongside the food. The edible polymers, for example, polysaccharides, proteins and lipids, are utilized to make edible packing. Writing show that the mechanical strength and boundary properties to water fume and gases (oxygen and carbon dioxide, primarily) of unplasticized edible films produced using the local edible polymers are substandard than the oil inferred plastics. They are additionally weak prompting unfortunate adaptability. The expansion of plasticizer expanded the film adaptability of edible films fundamentally. Glycerol is the generally utilized plasticizers to make the consumable movies. The impacts of different plasticizers are least found. The expansion of nanomaterials is the latest thing of examination in making bionanocomposite edible films, where edible nanofiller is consolidated to work on the useful properties. While a large portion of the exploration works, found in writing, zeroed in because of a nanofiller expansion on a solitary wellspring of palatable polymer, there is no relative examination did in writing to comprehend the polymer-nanofiller communications on the film properties of edible films. . The effects of nanofillers, namely are nanocellulose.

TABLE OF CONTENTS

CHAPTER NO	TITLE	PAGE NO
	ABSTRACT	v
	LIST OF TABLES	viii
	LIST OF FIGURES	ix
1	INTRODUCTION	1
	1.1.INTROUDUCTION TO EDIBLE FLIMS AND COATINGS	2
	1.2. FORMATION OF EDIBLE FILMS AND COATING	3
	1.3. EDIBLE FILMS: CHALLENGES AND OPPORTUNITIES	4
	1.4. POPULAR EDIBLE PACKAGING MATERIALS	10
2	LITERATURE SURVEY	13
3	AIM AND SCOPE	16
	3.1. AIM	
	3.2. SCOPE	
4	MATERIALS AND METHODS	17
	4.1. MATERIALS	17
	4.2. EXTRACTION OF NANOFILLERS	18
	4.3. FORMATION OF EDIBLE FILMS	20
	4.4. TESTING OF FILM PROPERTIES	21
5	VARIOUS EFFECTS OF FILM FORMATION	27
	5.1. EFFECTS OF STARCH SOURCES	28

5.2. EFFECTS OF PLASTICIZERS	28
6 RESULT AND DISCUSSION	30
6.1. EFFECTS OF NANOFILLERS	31
6.2. EFFECT OF NANOFILLERS ON WVTR	31
6.3. EFFECT OF NANOFILLERS ON TENSILE PROPERTIES	34
6.4. EFFECT OF NANOFILLERS ON FILM COLOUR	41
7 CONCLUSION	47
REFERENCES	49

LIST OF TABLES

CHAPTER NO	TITLE	PAGE NO
5.1.	FILM COMPOSITION OF THE NANOCELLULOSE INCORPORATED STARCH EDIBLE FILMS	27
5.2.	FILM COMPOSTION OF THE PLATICIZED BIONANOCOMPOSTION FILMS	29
5.3.	PROPERTIES OF THE PLASTICIZERS	29
6.1	FILM COMPOSITIONS OF THE NANCELLULOSE INCORPORATED RICE STARCH EDIBLE FILMS	30
6.2	FILM COMPOSITIONS OF THE NANOSTARCH INCORPORATED RICE STARCH EDIBLE FILMS	31

LIST OF FIGURES

CHAPTER NO	TITLE	PAGE NO
1.1.	POTENTIAL MIGRANTS OF THE EDIBLE FILMS	2
1.2.	FILM FORMING METHODS	3
1.3.	LIST OF EDIBLE FILM FORMING POLYMERS	11
4.1.	MAKING OF EDIBLE FILMS USING SOLUTION CASTING METHOD	21
4.2.	MEASURING THE FILM THICKNESS OF A SAMPLE EDIBLE FILMS	21
4.3.	MEASURING WVTR OF THE SAMPLE EDIBLE FILMS	22
4.4.	MEASURING TENSILE PROPERTIES OF A SAMPLE EDIBLE FILMS	24
4.5.	MEASURING THE FILM COLOUR USING PHOTOGRAPHY	26
6.1.	WVTR OF NANOFILLER INCORPORATED RICE STARCH FILMS	32
6.2.	TS OF NANOFILLER INCORPORATED RICE STARCH FILMS	35
6.3.	YM OF NANOFILLER INCORPORATED RICE STARCH FILMS	36
6.4.	ELONGATION AT BREAK OF NANOFILLER INCORPORATED RICE STARCH FILMS	39
6.5.	'L' VALUES OF NANOFILLER INCORPORATED RICE STARCH FILMS	42
6.6.	'A' VALUES OF NANOFILLER INCORPORATED RICE STARCH FILMS	43
6.7.	'B' VALUES OF NANOFILLER INCORPORATED RICE STARCH FILMS	44
6.8.	'ΔE' VALUES OF NANOFILLER INCORPORATED RICE STARCH FILMS	45

LIST OF ABBREVIATIONS

ABBREVIATION	DESCRIPTION
WVTR	Water vapour transmission rate
TS	Tensile strength
YM	Young' modulus
EAB or E	Elongation at break
L	Lightness colour parameter
A	Greenness-Redness colour parameter
B	Yellow colour parameter
RH	Relative humidity
°C	Degree Celsius
MPa	Mega Pascal
NA	Not applicable
WVP	Water vapour polyethylene
LDPE	Low density polyethylene
IR	Infrared radiation
ΔE	Change in colour
RS	Rice starch
PS	Potato starch
CS	Corn starch
TS	Tapioca starch
WS	Wheat starch
GLY or G	Glycerol
MAL or M	Maltitol
SOR or S	Sorbitol
XYL or X	Xylitol
H ₂ O	Water
NS	Nanostarch
NC	Nanocellulose
GPa	Giga Pascal

CHAPTER 1

INTRODUCTION

Packaging is the method involved with wrapping or encasing the items for shielding them from harm during taking care of and transportation. Bundling is one of the unavoidable things in numerous applications like food also, drinks, purchaser products, drug applications, way of life things, beauty care products, clothing, medical services, and numerous others. The normal bundling materials are plastics, paper board, metals, glass, and different materials. With 42% of complete utilization of bundling materials, plastics is the biggest utilized packaging material, because of their simple handling, elite execution, lightweight, lower cost, and accommodation. Inside plastic packaging, food bundling is one of the biggest plastic devouring areas with around 54% of offer determined for the time of 2003-2009. This incorporates 40% in food also, 14% in refreshments areas separately. The significant elements of food bundling are to store the contained food things, shield them from harm during taking care of and transportation, or safeguards them from outer influences, and keep up with the food quality from the hour of bundling to the hour of utilization.

1.1. INTRODUCTION TO EDIBLE FILMS AND COATINGS

Edible packaging is a kind of packaging that can be eaten along with the food. As the edible packaging is made from edible polymers, it can be consumed along with the contained food. It is a thin layer formed on the food surface. It is formed either as a film or as a coating. When the thin layer is formed directly on the food surface, it is termed as "edible coating". If it is formed separately over another substrate (glass substrate, for example) and peeled off for the later use to wrap the food, it is termed as "edible film".

As the edible packaging materials are edible and obtained from natural resources, they can be eaten along with the contained food or beverages. As the packaging can also be eaten, nothing will be left out for disposal by virtue after the food consumption. Even if the packaging is not eaten, as they are produced from easily

digestible materials, they do not harm to the environment; they degrade more readily than both synthetic and biodegradable packages. Edible packaging is safe for human consumption, renewable, environmental friendly and does not require large landfill sites. As a result, they have high potential to fully or partially replace both synthetic and degradable plastics used in food packaging in the near future.

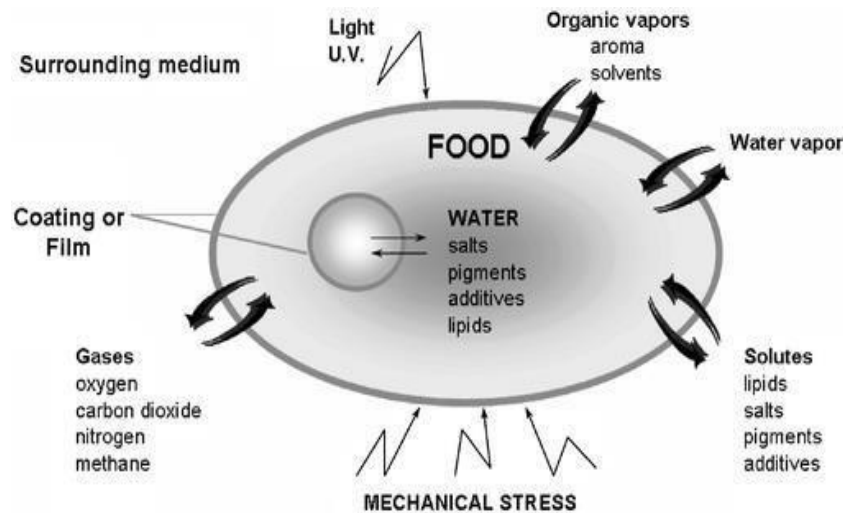


Fig: 1.1: Potential migrants of the edible films

Fig1.1. shows the potential migrants of the edible films between the contained food and the surrounding. The qualities of an ideal edible packaging material are listed as follows:

- It should not contain any toxic, allergic and non-digestible ingredients.
- It should provide sufficient structural stability to prevent mechanical damage during transportation and handling.
- It should display good adhesion with food surface for uniform coverage.
- It should control the migration of water vapors coming in and going out of it to maintain a desired moisture level.
- It should be semi-permeable to gases responsible for aerobic and anaerobic respiration for retarding senescence.
- It should not alter the aroma, flavor, nutritional, sensory and organoleptic characteristics, which are important for consumer acceptance.
- It should protect against contamination, pest infestation, microbe proliferation, and other types of decay.

- It should serve as carrier for desirable additives such as flavor, fragrance, coloring, nutrients, vitamins, antioxidants and antimicrobial agents.
- It should easily be processed and manufactured.
- It should be economically feasible.

1.2. FORMATION OF EDIBLE FILMS AND COATINGS

The ability of forming a continuous and cohesive structure of a packaging material determines whether or not an edible material can be used for making edible packaging

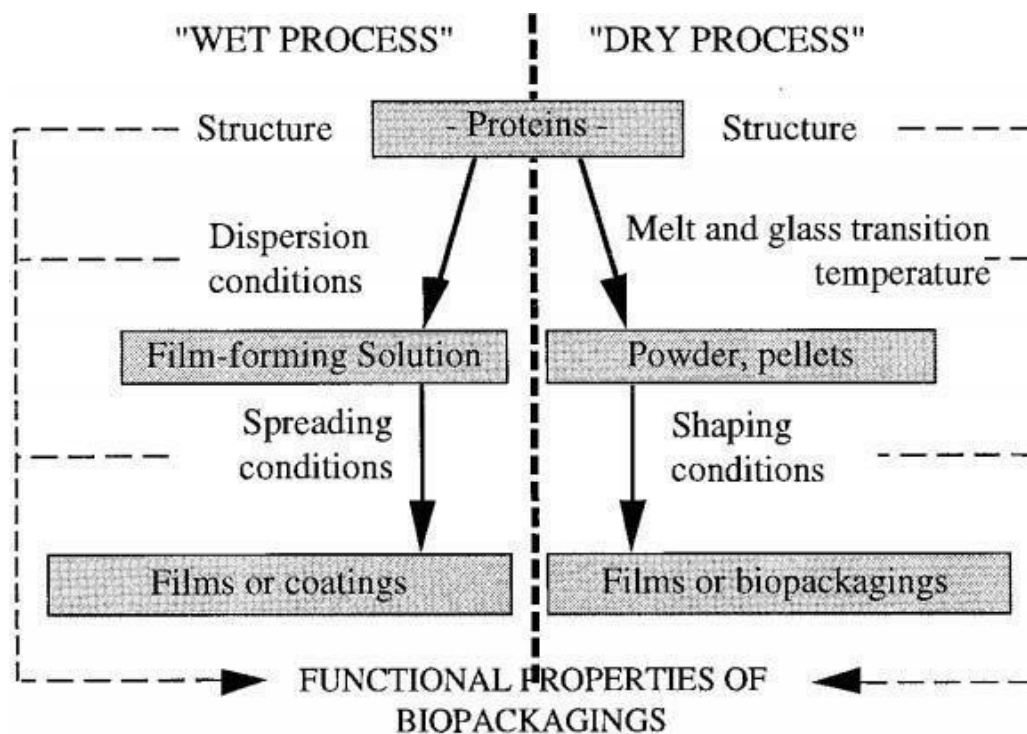


Fig: 1.2: Film forming methods

Two methods are commonly used for making edible films and coatings: (a) dry process and (b) wet process. In dry process, the films and coatings are prepared under low-moisture environment by thermal or thermomechanical treatments. In this method, thermoplastic raw materials are heated to a temperature above glass transition temperature to form soft and rubbery materials and are molded to form

the desired shape of the edible film and coatings. Cooling the mold to room temperature produces the desired edible films or coating in a more rigid form.

Two methods are commonly used for making edible films and coatings: (a) dry process and (b) wet process. In dry process, the films and coatings are prepared under low-moisture environment by thermal or thermomechanical treatments. In this method, thermoplastic raw materials are heated to a temperature above glass transition temperature to form soft and rubbery materials and are molded to form the desired shape of the edible film and coatings.

1.3. EDIBLE FILMS: CHALLENGES AND OPPORTUNITIES

Even though edible films seem to have high potential to replace synthetic and degradable plastics, the production of edible films is still in laboratory scale. There are many issues yet to be overcome for the industrial scale production of edible films. When compared to synthetic plastics, edible films exhibit some problems, such as poor mechanical strength (especially poor elongation), poor resistance to gases and liquids, lack of evidence on their edibility and biodegradability, processing difficulties, etc. This section reviews some of the major challenges and opportunities in the production of edible films.

1.3.1. Functional Properties

The functional properties of the edible films are the essential properties that describe the performance and suitability to a particular application. In this research work, three major functional properties, namely, (1) mechanical strength, (2) water vapor transmission rate (WVTR) and (3) film color, were considered. High mechanical strength is required to protect the contained food from the external loads. WVTR is the degree of resistance offered by the edible films to prevent migration of water vapor between the contained food and the environment. Film color is another important property that influences the consumer acceptability of edible films. The film properties of some edible films produced from native edible polymers. The table values show that the mechanical strength of neat edible films is lower than those of petroleum derived plastics. However, it should be noted that TS of most edible films is higher than 3.5 MPa, which is the

minimum requirement of most food packaging materials. On the other hand, EAB of edible films is very low as compared to petroleum derived plastics, which indicates that the edible films are highly brittle, leading to poor flexibility. As most of the edible polymers are hydrophilic in nature, the permeability of edible films is higher as compared to petroleum derived plastics. Edible films are therefore highly sensitive to water vapors, and allow them causing absorption of water molecules to swell the edible films and causing dimensional instability. From the film color parameters, it is learnt that most of the neat edible films produced from polysaccharides are clear and transparent, while the edible films produced from proteins, nonconventional sources and lipids might not be clear and transparent.

1.3.2. Plasticizers Effects

In order to improve the film flexibility of the edible films, a plasticizer is added to the film forming solution. Food-grade polyols, glycols, sugars, and lipids are the commonly used plasticizers. The short-chain plasticizers break the long-chain polymer-polymer interactions to make the shortchain polymer-plasticizer interactions. As a result, plasticized edible films become soft and flexible as compared to the unplasticized films. The effects of plasticizers on edible films. It is evident from the table that the addition of plasticizer significantly improved EAB of edible films indicating the improvement in film flexibility. As plasticizers are generally clear and odorless, they do not affect the film color significantly. However, the addition of plasticizer reduced TS and YM while increasing WVTR. Glycerol was the common plasticizer used in most edible films to make the edible films. However, the type of plasticizer can affect the film properties significantly. For example, sorbitol, glycols, sugars produced the stronger and more stretchable edible films than the glycerol plasticized films. While polyols produced soft, clear and smooth films with high mechanical strength, glycols produced hard, opaque and rough films with relatively lower mechanical strength. Sugars produced edible films of higher crystallinity, mechanical strength and elongation than both polyols and glycols, on the other hand, reported no significant differences in film properties due to plasticizer type. Therefore, the influence of the plasticizer type is one of the key research to be investigated to improve the film properties.

1.3.3. Multilayer/ Emulsion Composite Films

High mechanical strength of proteins and high water resistance of lipids may be incorporated in polysaccharide films having poor mechanical strength and water resistance. Thus, mixing of more than one edible polymer can produce the composite films with complementary benefits to each other. The effects of multilayer composite films on the film properties. Multi-layer composite films are formed as two or more layers of alternating edible polymers. Though multilayer composite films can improve the film properties of edible films, they require long production time, high energy consumption and high cost. They also tend to delaminate over the time, causing problems like pinholes, cracks, surface non-uniformity, and reduced cohesion. These problems make the multilayer edible films less popular. Table 1.4 shows the effects of emulsion composite films on the film properties. Emulsion composite films are formed by mixing a lipid dispersed in a hydrocolloid matrix so that the structural integrity of the hydrophilic matrix and hydrophobic behavior of the lipid could be complemented in order to improve the functional properties of the resulting films than the neat hydrocolloid films.

1.3.4. Film Making and Drying Methods

Casting is one of the most used film making methods for the laboratory scale production of edible films. In this method, the film forming solution is prepared first and heated to a gelatinization temperature in order to form a gel-like material. This gel-like material is then spread onto a surface and dried to form the films. These cast films contain high amount of water that is evaporated using the energy-consuming drying methods the effects of drying methods on the edible films. Most of these edible films are evaporated by hot air, which is considered to be a very slow process. It, sometimes, takes about 2-3 days for drying the edible films. There are other drying methods that could minimize the drying time. For example, vacuum drying, conduction, infrared drying and low-pressure superheated steam drying were used to dry the edible films in less than two hours. In the work of Kaya & Kaya, 2000, the microwave drying was used to dry the edible films in 5 min without significant difference in WVP. While most of the research works focused on the fabrication of edible films on lab-scale or batch size, the continuous formation of edible films is another problem with the edible films.

proposed a continuous fabrication method to produce the continuous edible films in which the film forming solution was spread in a moving conveyor and dried using both an IR pre-dryer and hot air drying. The results reported the total drying time as 7 min and productivity as 0.03 m² film/min. There is an immediate attention required in order to low cost, energy-efficient and high production for the industrial scale production of edible films.

1.3.5. Nanotechnology on Edible Films

Nanotechnology is one of the current trends in research in all fields of engineering and research. Realizing the unique properties offered by the nano-sized materials that could significantly enhance the film properties of the food packaging, the food and beverage industry has started investing on the use of nano-sized materials for food packaging (Jeevahan & Chandrasekaran, 2019a). Table 1.6 shows the effects of nanomaterials on the film properties of edible films. From the results, it is observed that the nanomaterials increased the mechanical strength while reducing the WVTR. While most research works used the nanomaterials as reinforcements with the macroscale edible polymers to produce the bionanocomposite edible films, only a few research was carried out with nano-sized matrix as well as reinforcement to produce the fully nano-sized edible films. For example, in the work of Taniguchi and Okamura (1998), the complete nanocellulose edible films were produced, which showed 2- 3 times higher mechanical strength than the synthetic LDPE films. In another work by Leitner et al. (2007), the complete nanocellulose edible films were produced with TS of 104 MPa and YM of 9 GPa, respectively. However, only limited research is carried out on the effects nano-scaled materials on the film properties of edible films.

1.3.6. Other aspects of edible films

While most research literature focused on the influence of biosources of edible polymers, concentration of the plasticizer addition and the relative humidity on the film properties of edible films, there are many factors that affects the film properties are yet to be investigated. For example, the effects of polymer structure, the orientation, composition, crystallinity index, degree of cohesion,

degree of adhesion, temperature, pressure, free volume, film thickness etc. are some of the basic factors are least investigated. Process optimization is another essential requirement for batch as well as continuous production of edible films. Multifunctionality is the key requirement of the future edible films and coatings. In addition to contain the food, the edible films should also serve many functions. For example, the edible films should be heat-resistant, water-resistant, air-resistant, corrosive resistant, oil-resistant, disease-resistant, and so on.

As far as mechanical strength is concerned, most literature focused on only tensile properties. But, other mechanical properties, such as, the puncture strength, sealing strength, tearing strength, resistance to scratch, light degradation, and so on, are also important. Also, the film properties should not change with respect to time. But, aging can change the film properties of the edible films. However, there are rare studies on the influence of aging on time dependant film properties of edible films. The permeability of edible films reported in most literature were observed at room temperature. But, the storage as well as processing temperatures can vary from 20°C to 60°C for food packaging in practice.

Thus, the permeability should also be observed at different storage as well as processing temperatures. Furthermore, most of the literature reported the permeability of water vapours and gases, but the migration of residual monomers, food additives, plasticizers, printed inks, etc. can also affect the film properties (such as, textural, nutritional and sensory properties) of the contained food. Hence, research on the permeability of oil, aroma, and other additives and food components, etc. should be evaluated for the suitability of edible films for the selected applications.

Edible films can also be used for making the beverage packaging. Only one research work, by Rodríguez-Castellanos et al. (2015), was carried out to develop the edible films for the beverages. The mixture of hydrolyzed corn starch–gelatin

pellets and cellulose were converted into the 500 ml bottles using extrusion and blow molding. The results showed that the internal surface of the blow-molded films was uniform, while the external surface was porous. Also, the films showed poor mechanical strength and poor barrier properties at high humidity. Future research is essential on the applicability of edible films for beverages packaging.

Another research area is the compatibility of edible films to the modern food packaging technologies, such as, active packaging, intelligent packaging and smart packaging. While the traditional packaging acts as the barrier between food and environment, the active packaging allows some interactions. With intelligent packaging, one can monitor and show the condition/ quality of the contained food . In addition to monitor the condition of the food, smart packaging tracks the 35 food and allows to control the environment. But, the research on this area needs much attention as placing the microsensors and integrating them with the communications systems are difficult and challenging.

Consumers acceptability is another key area to be investigated for the success of edible films. Researchers claim the edible films to be safe because they are made of materials that are generally regarded as safe. But, there is lack of evidence on their biodegradability as well as the edibility. Many factors, such as, the filmforming methods, the film-forming materials, plasticizer type, their composition, the added functional additives, the chemical and physical properties of these materials etc., can have a significant role on the digestibility as well as the digestion rates of the edible films. For example, a research conducted by Hernandez, Emaldi, & Tovar (2008) revealed that the edible films produced from three different starch sources had the significantly different digestibility rates from those of starches.

Hence, it is important to evaluate the biodegradability and edibility of the edible films to assure the safety. Not only consumers see the functional properties of the films, there are other factors that are important. The edible films should not affect the sensory properties OF the contained food (Han, 2014). There are certain people or religions which do not allow the animal derived food, and may be unwilling to use the

edible films as some of them are made from animal-derived materials. Furthermore, if the edible films may contain allergens that may cause allergic reactions to the consumers. Hence, proper labeling and guidelines should be developed and the food packaging producers should be emphasized to label the ingredients and the allergens. In addition, consumers generally do not look on the environmental or industrial benefits. From the consumers' point of view, the film properties of the edible films are highly important. Hence, the research is required on improving the film properties close to the synthetic plastics without the penalty on the cost, easiness etc. The edible films are at least 10-50 times costlier than the synthetic packaging. Future works should also focus on the production of low cost edible films with lower than or equal to the cost of plastics for the customers' attention towards the edible films.

1.4. POPULAR EDIBLE PACKAGING MATERIALS

Figure 1.3 shows three major classes of edible polymers used to make edible packaging. They are polysaccharides, proteins and lipids.

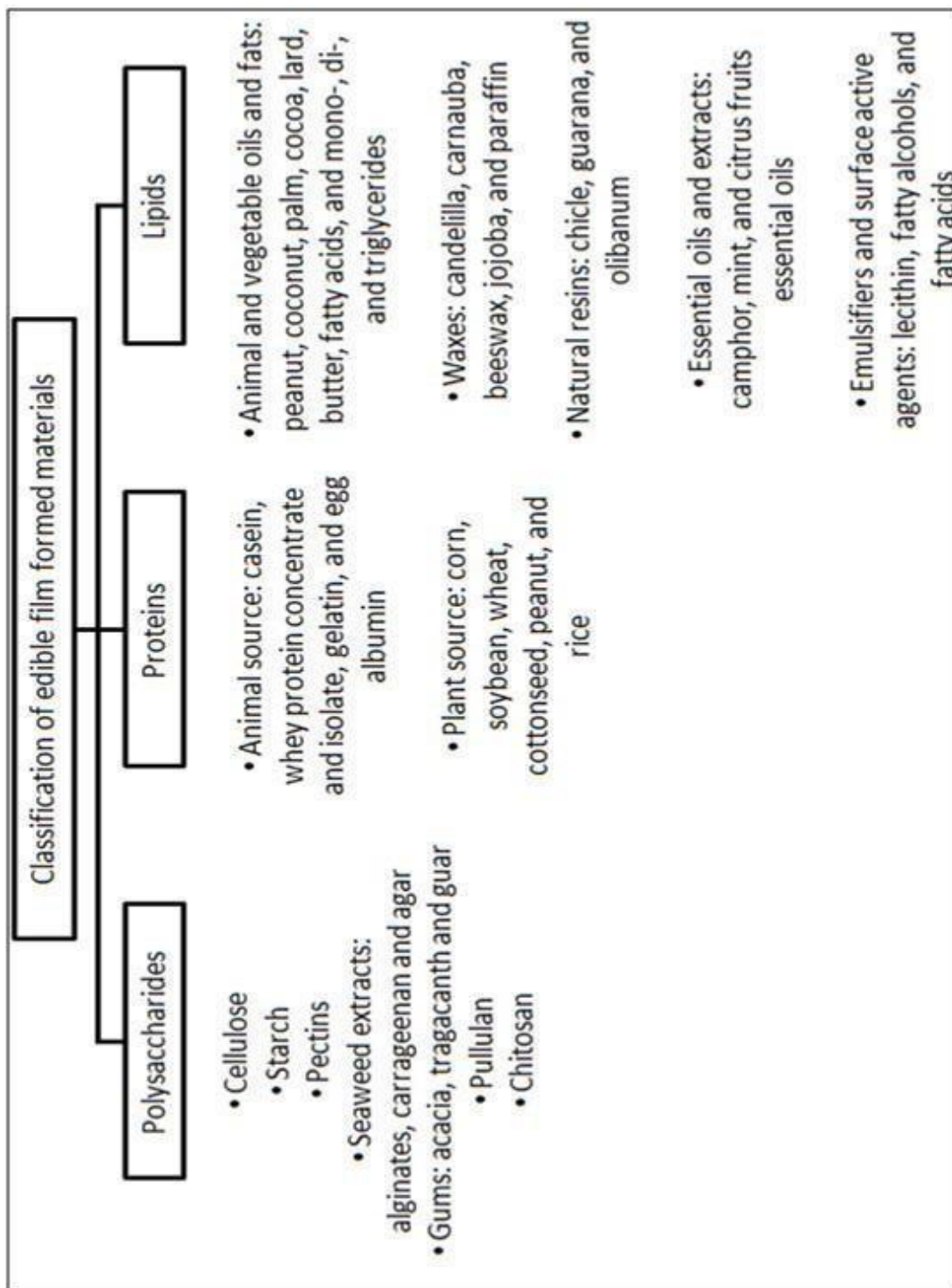


Fig: 1.3 : List of edible film forming polymers

Polysaccharides are homo-polymers consisting of a repeating units of monomer, such as glucose, while proteins are hetero-polymers consisting of more than at

least 20 amino acids. While edible films produced from polysaccharides exhibit poor mechanical strength, the proteins-based edible films offer good better mechanical strength due to their complex nature of amino structure. As both polysaccharides and proteins are hydrophilic in nature, the edible films made from them exhibit poor water resistance. Lipids, on the other hand, are hydrophobic in nature and they exhibit good resistance to water transmission. But, lipids are not capable of forming the self-supporting film structures. Hence, lipids can be used as a edible coating or as an additive along with polysaccharides and/ or proteins in order to form the composite films (Janjarasskul & Krocha, 2010; Bourtoom, 2008).

CHAPTER 2

LITERATURE SURVEY

Guilbert et al.1995 The ability of forming a continuous and cohesive structure of a packaging material determines whether or not an edible material can be used for making edible packaging.

Pavlath & Orts, 2009; Debeaufort & Voilley, 1994; Debeaufort et al., 1998; Siracusa, 2013; Otoni et al., 2017the potential migrants of the edible films between the contained food and the surrounding. The qualities of an ideal edible packaging material are listed as follows.

Janjarasskul & Krocha, 2010; Bourtoom, 2008, Lipids, on the other hand, are hydrophobic in nature and they exhibit good resistance to water transmission. But, lipids are not capable of forming the self-supporting film structures. Hence, lipids can be used as a edible coating or as an additive along with polysaccharides and/or proteins in order to form the composite films.

Ploypetchara & Gohtani.,2018 Sugars improved the crystallinity and homogeneity of the edible films for native corn, waxy corn, native rice and waxy rice. However, the all sugar plasticized films showed the reduced breaking stress as compared to that of the films without plasticizers.

Balqis et al.,2017 Glycerol plasticized films had higher thickness and higher moisture content compared to sorbitol plasticized films. Sorbital plasticized films showed better mechanical strength and better water barrier properties than glycerol plasticized films to kappa-carrageenan

Saberi et al.,2017 All plasticized films were smooth, transparent and homogeneous, except the one formed with polyethylene glycol 400. Glycerol plasticized films produced better flexibility among other films. However, monosaccharides were suggested to have better film properties based on the optical, mechanical and barrier characteristics to pea strach gum.

El-Miri et al.,2018 Glycerol plasticized alginate film showed lower mechanical strength, lower thermal properties and higher water sensitivity as compared to those of diethylene glycol and polyethylene glycol plasticized films for cellulose nanocrystals reinforced alginate.

Edhirej et al.,2016 Fructose plasticized films exhibited smooth surfaces with free of pores with high water resistance and high density compared to other plasticized films for cassava starch.

Mkandawire & Aryee, Zhang et al.,2017 Multifunctionality is the key requirement of the future edible films and coatings. In addition to contain the food, the edible films should also serve many functions. For example, the edible films should be heat- resistant, water-resistant, air resistant, corrosive resistant, oil-resistant, disease- resistant .

Shit & Shah, Han,2014 As the packaging can also be eaten, nothing will be left out for disposal by virtue after the food consumption . Even if the packaging is not eaten, as they are produced from easily digestible materials, they do not harm to the environment; they degrade more readily than both synthetic and 6 biodegradable packages. Edible packaging is safe for human consumption, renewable, environmental friendly and does not require large landfill sites. As a result, they have high potential to fully or partially replace both synthetic and degradable plastics used in food packaging in the near future.

Thompson et al.,2009 Though degradable polymers are beneficial to reduce the waste disposal problems, the degradable polymers are not yet good for environment and 5 health.

Angellier et al., 2006 To addition of glycerol could improve the elongation of the films, but the tensile strength of the bionanocomposite edible films would be lowered.

Kaushik & Kaur, 2016; Muller et al., 2009; Fahma, 2011; Crosby & Lee, 2007 The stress transfer between the matrix and the filler would increase, which, in turn could increase the tensile strength. Also, because the nanofillers occupy the

spaces between the starch network, the stress required to make any deformation would increase.

Zainuddin et al., 2013 The formation of aggregates from the weak bonding between the matrix-filler interactions, and, therefore, the efficacy of the stress transfer would reduce, which, in turn, could have reduced the mechanical strength of the bionanocomposite edible films with more than 10% nanofiller addition.

Kim-Kang, Li et al., 2006 most of the literature reported the permeability of water vapours and gases, but the migration of residual monomers, food additives, plasticizers, printed inks, etc. can also affect the film properties (such as, textural, nutritional and sensory properties) of the contained food. Hence, research on the permeability of oil, aroma, and other additives and food components, etc. should be evaluated for the suitability of edible films for the selected applications.

Soykeabkaew et al., 2012; Gea et al., 2010 The bionanocomposite films could experience higher transparency as compared to the control films, which could result no significant difference in the colour parameters at lower concentrations of nanofillers. Further, the addition of nanofillers would fill the voids between the starch molecules of the control films, making the surface more even, which would allow the light rays scattered making the film brighter.

Xu et al., 2017 At lower concentration of nanofiller addition, the change of colour parameters, 'L' as well as 'b', of the bionanocomposite films was very low. However, when the addition of nanofiller became significant, the transparency of the nanocomposite films was affected, and the films became darker.

CHAPTER 3

AIM AND SCOPE

3.1. AIM:

- i. To Design the film unit for effects of nanocellulose on film properties of edible film.
- ii. To fabricate the film unit for continuous edible film making machine.
- iii. To evaluate the effects of design parameters on the effects of nanocellulose on film properties of edible film.

3.2. SCOPE:

An edible films could be characterized as essential packaging produced using edible parts.

In this process thin layer of edible material can be straightforwardly covered to a food or shaped into a film and be utilized as a food wrap without changing the first original ingredients or the handling strategy.

One significant benefit of utilizing edible films and coatings is that few dynamic ingredients can be joined into the polymer framework and devoured with the food, along these lines improving wellbeing or even dietary and tactile traits.

It is exceptionally valuable for people in the future for edible film making machine.

It was clear that the addition of nanocellulose had improved the film properties, making the rich starch based edible films a promising choice for food packaging application.

CHAPTER 4

MATERIALS AND METHODS

This section examines about the materials utilized and the fabrication strategies followed to make the edible films, systems followed to remove the nanofillers and the strategies followed to quantify the film properties of the edible films.

4.1. MATERIALS

Various sources of food grade edible polymers were utilized to form the local edible films. Potato starch, custard starch, wheat starch, rice starch furthermore, corn starch with no less than 98% immaculateness were bought from Urban Platter Food Corporation, Mumbai, India. Whey protein disengage and soy protein disengage, and coconut oil with somewhere around 98 % immaculateness were bought from nearby market in and around Chennai, India. Different food grade plasticizers were utilized to further develop the film adaptability. Polyols (glycerol, maltitol, sorbitol and xylitol), glycol (polyethylene glycol) and sugars (Sucrose) were bought from Sri Amman Scientific And Surgicals, Chennai, India. Nanostarch was removed from a starch source (ricestarch, for instance) through corrosive hydrolysis response. Nanocellulose was removed from banana fiber through antacid treatment followed by corrosive hydrolysis response. Banana fiber was gotten from neighborhood ranchers close Chennai, India. Different synthetics, refined water and dish sets expected to separate the nanofillers were bought from Sri Amman Scientific And Surgicals, Chennai, India.

4.2. EXTRACTION OF NANOFILLERS

4.2.1. *Extraction of Nanocellulose*

Nanocellulose was separated from banana pseudo stems in two stages: (1) cellulose extraction by soluble treatment followed by blanching process, and (2) nanocellulose confinement from cellulose mash by corrosive hydrolysis response. The technique detailed by Mueller et al. (2014) and Elanthikkal et al. (2010) was utilized to remove the cellulose mash from banana pseudo stems. The banana stems were gathered from the neighborhood ranchers, cut into little pieces (10-15 cm), and dried. The dried pieces were then ground to make the fine powder (500 micron).

Antacid treatment was utilized to eliminate pectins and the hemicellulose, while the blanching system was utilized to eliminate lignin (the mash becomes more white). In the first place, we start with the antacid treatment. In this technique, the combination of the processed banana stem powder and the sodium hydroxide (NaOH) arrangement (3 M, for example 120 g for every 1 liter of water) were taken in a glass measuring glass. The arrangement was warmed to a temperature of around 80 °C and kept up with for around 2 h. The arrangement was persistently blended utilizing a mechanical stirrer. This interaction solubilised the hemicellulose and gelatin from the banana stem mash. After 2 h of antacid treatment, the subsequent arrangement was poured in a cotton cheddar material and washed in refined water a few times to eliminate hemicellulose and gelatin parts. Washing in refined water was rehashed until the pH esteem became 7. The basic treatment and washing cycle was rehashed threefold. The alkaline treated banana stem mash was then dried in daylight prior to fading process.

Dying cycle was done to eliminate lignin from the banana stem mash. In this interaction, the soluble treated banana stem mash was blended in sodium hypochlorite (5 wt% of NaOCl in 100 mL water) arrangement in a receptacle. The arrangement was then warmed to a temperature of about at 45 °C and kept up

with for about for 2 h. The subsequent mash was washed in refined water a few times till the pH arrived at 7. During 45 washing, the shade of the mash became more white demonstrating that the shading specialist, lignin, had been eliminated. The blanching system was rehashed threefold to eliminate the ligin parts totally. The coming about white mash was cellulose and was dried in daylight for 1 day. Subsequent to drying, the cellulose was put away in the natural chamber for the future use.

Acid hydrolysis was utilized to seclude nanocellulose from cellulose in light of the methodology announced by Beck-Candanedo et al. (2005) with slight adjustments. The banana stem cellulose was taken in Erlenmeyer jar furthermore, the sulfuric corrosive arrangement (64 wt% fixation in water) was added to itThe corrosive to-cellulose proportion was around 8.75 mL/g. The arrangement was warmed at 45 °C for 30 min to break the long cellulose chains and structure nanocellulose. After corrosive hydrolysis response, the nanocellulose suspension was shaped. The suspension was washed in ice water a few times till it arrives at the pH of 7, and along these lines centrifugated at about 2000 rpm for 30 min to make the concentrated nanocellulose suspension. The concentrated nanocellulose suspension was then ultrasonicated utilizing a ultrasonicator for around 15 min to get the colloidal nanocellulose. The collodal nanocellulose was then separated utilizing the channel papers of around 20 µm pore size. The subsequent nanocellulose was freezedried and put away before use.

4.2.2. Extraction of Nanostarch

Nanostarch was produced from a starch source through acid hydrolysis based on the procedure reported by Angellier et al. (2004), Kim et al. (2012) and Piyada et al. (2013) with slight changes. In this method, 36.5 g of starch (14.6 wt%) was taken in an Erlenmeyer flask and the sulfuric 46 acid solution (3.16 M, 178 mL of sulphuric acid and 822 mL water) was added to it. The solution was kept in a shaker and kept in an environmental chamber for 5 days.

During the period, the reaction takes place and the nanostarch would be formed. After 5 days of acid hydrolysis, the resulting nanostarch suspension was centrifuged (at 2000 rpm for 30 min) and washed several times by successive centrifugations. The centrifugation and washing in distilled water were continued till the suspension reached the pH of 7. The resulting concentrated nanostarch suspension was freeze-dried and stored before use.

4.3. FORMATION OF EDIBLE FILMS

Solution casting was the film forming method used in this research to form the edible films. This section explains the procedure to form the edible films from the starch based films. The steps followed in the making of bionanocomposite edible films. In this method, an aqueous solution was prepared initially by mixing 5 g of starch in 100 mL distilled water in a glass beaker/ vessel. A food grade plasticizer (30 wt%/100 vol% of solution) and/or different proportions of a nanofiller were also added into the aqueous solution, and mixed well with a hand stirrer. The beaker containing the aqueous solution was then heated in a water bath maintained at 80 °C while stirred continuously using a mechanical stirrer for about 20 min. After 20 min, the gelatinization reaction was taking place resulting a non-reversible gel-like film forming solution. After cooling this film forming solution to about 40 °C, it was poured onto a flat plate, spread evenly over it, and dried. After drying through evaporation mode, the edible films were formed, which were gently peeled off from the plates manually. The edible films were stored in environmental chamber at 23°C and 50% RH.

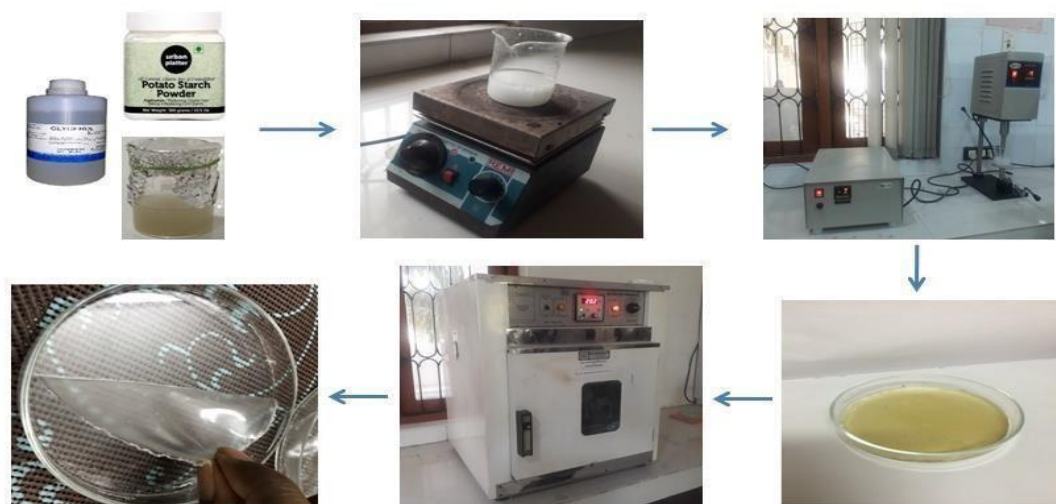


Fig:4.1: Making of edible films using solution casting method

4.4. TESTING OF FILM PROPERTIES

4.4.1. Film Thickness



Fig:4.2: Measuring the film thickness of a sample edible film

The thickness of the edible films was measured using a digital micrometer (Insize, sensitivity = 0.001 mm) as shown in Fig. 2.5. The film thickness of each film was measured at five random places, and the average thickness (in mm) was considered for the analyses.

4.4.2. Water Vapour Transmission Rate (WVTR)

The procedure as mentioned in ASTM E96 standard (Standard Test Methods for Water Vapor Transmission of Materials) was followed to determine the WVTR of the edible films. Fig. 2.6. shows the steps followed in determining the WVTR of the bionanocomposite edible films. Gravimetric method (also called upright dry cup method) was used in this research work, where each edible film was covered over a cup filled with water, and the weight loss was measured. The test films were cut into circular pieces of diameter, 7.5 cm. Each test film was then covered over the glass cup containing distilled water (100 ml) in it. The glass cups with test films were sealed with a gasket and screwed tightly using a metal lid. Before testing, the initial weight of each test cup with test film, water, gasket and lid, and the time of taking the reading were noted. A weighting balance (sensitivity = 0.001 g) was used to measure the weight of the test cups. The test cups were then kept in an environmental chamber at 50 ± 10 %RH and 23 ± 2 °C, where the water vapour inside the test cup escapes out through the edible film thereby losing the weight of the test cup. The weight of the test cups were measured every 2-3 hours along with the recording time. This weight minus the initial weight would give the weight loss.

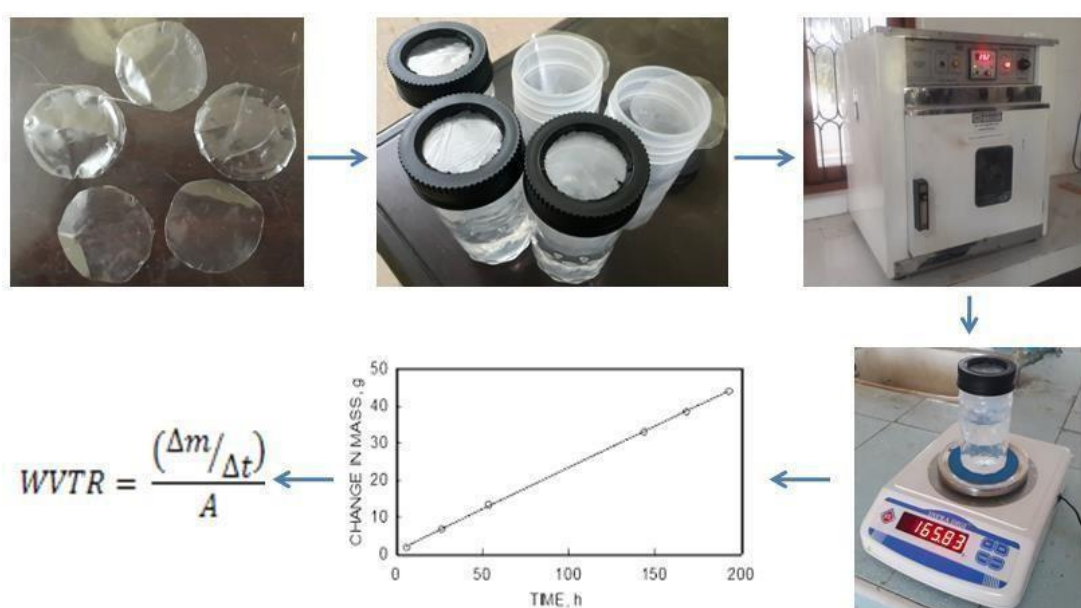


Fig:4.3 : Measuring WVTR of the sample edible films

The weight loss of the test film was marked in a graph having the weight loss in X-axis and the time interval in Y-axis. The subsequent readings at different time intervals were marked in this graph. The weight loss would be non-linear initially with the time, and then becomes linear. The number readings depends on the linearity of the graph. That means, the weight loss measurements were taken till the weight loss versus time curve became linear. From the weight loss versus time graph, the slope of the straight line was calculated, which is weight loss versus time ($\Delta m/\Delta t$). WVTR was calculated from the following formula:

$$\text{WVTR} = (\Delta m/\Delta t) A \text{ (1)}$$

where,

WVTR is the water vapour transmission rate (g/m² /day).

Δm is the weight loss (g).

Δt is the duration of weight loss, and

A is the exposed area of the test film (m²) to the environment.

A minimum of 5 replicates were tested for each test case, and the average WVTR values were used for the result analysis.

4.4.3. Tensile Properties

ASTM D882 standard (Standard Test Method for Tensile Properties of Thin Plastic Sheeting) was used to determine the tensile properties of the edible films. The films were cut into strips of width, 25.4 mm, and the length, 150 mm. The test specimens were conditioned in an environmental chamber at 50±10 %RH and 23±2 °C for at least 2 days before use.

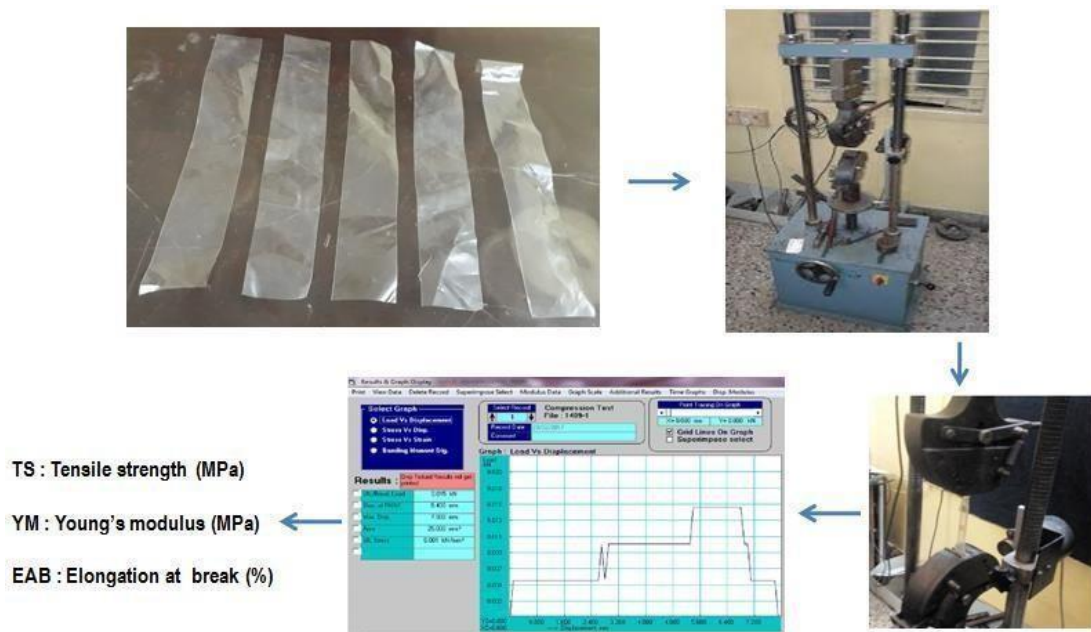


Fig:4.4 : Measuring tensile properties of a sample edible film

Tensile testing machine (Met Mech, Chennai) was used to determine the mechanical properties (TS, EAB and YM). The test films were firmly clamped between the grippers. The initial grip separation and rate of grip separation were maintained to be 100 mm and 50 mm/min, respectively. The load was then applied and the strain was measured, and the microcomputer attached to the tensile testing machine recorded the stress–strain curves. TS, YM and E were plotted by the minicomputer based on the following formulas:

$$TS = \text{Maximum load before failure} / \text{Initial area of cross section} \text{ ----- (2)}$$

$$YM = \text{Change in stress} / \text{Change in strain} \text{ ----- (3)}$$

$$E = \text{Change in length at rupture} / \text{Initial gage length} \times 100 \text{ ----- (4)}$$

A minimum of 5 replicates were tested for each test case, and the average values of TS, YM and E were considered for the result analysis.

4.4.4. Film Colour

The digital photography method reported by Afshari-Jouybari and Farahnaky et al. (2013) was used in this research work to measure the film colour. In this method, the film colour of the edible films was measured from their digital photograph taken with the help of a camera. For this purpose, a good quality digital camera was used. Before the photograph of edible films were clicked, the camera was set to 1000 dpi of resolution, 62 % of contrast and 62 % lightness, respectively. The test films were kept over a white platform, and the digital images of the test films were captured in JPEG file format.

The digital images were then opened in the Adobe Photoshop software (Version 8). and the colour parameters were observed by selecting at least 5 random locations in the image. Film colour is usually expressed in either CIE or LAB parameters. In this research work, the 'LAB' parameters were used to describe the film colour, where, three colour parameters, L, a and b, were observed from the images. In 'LAB' parameters, L represents the degree of darkness/lightness, a represents the degree of redness/greenness and b represent the degree of blueness/yellowness. The values of 'L' range from 0 (black) to 100 (white). The values of 'a' range from -80 (greenness) to 100 (redness). And, the values of 'b' range from -80 (blueness) to 70 (yellowness).

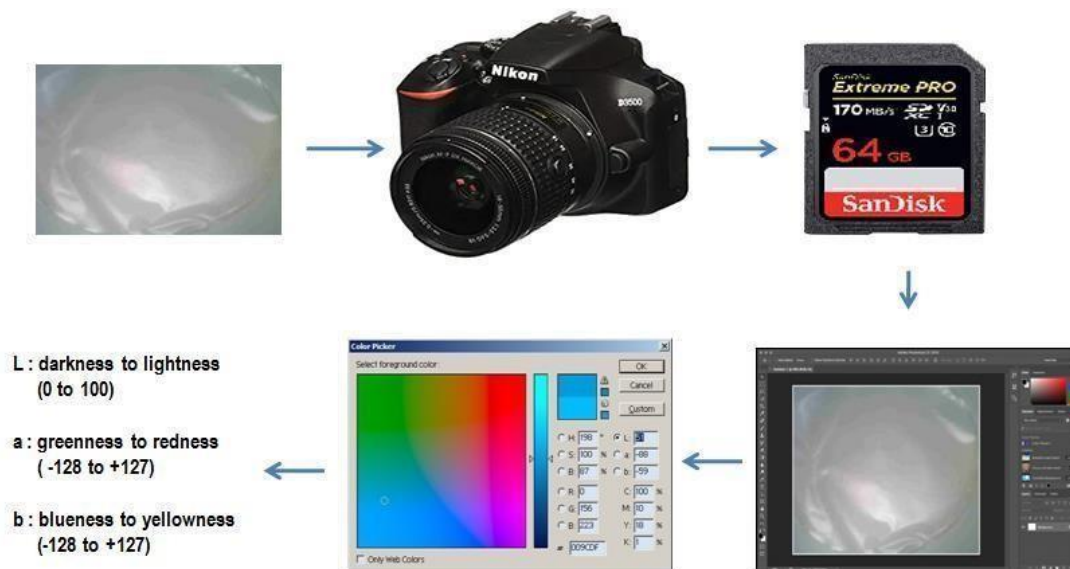


Fig:4.5 : Measuring the film colour using digital photography

The colour parameters obtained from the digital photography method give some insights on the change of film colour with respect to one other. The total colour difference (ΔE) is one of the useful parameters which tells us whether or not the film colour was changed from the reference film parameters. ΔE was determined from the following equation:

$$\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2} \text{----- (7)}$$

where,

L^* , a^* and b^* represent the colour parameters of the standard white colour background (For this research work, these values were obtained as: $L^*=96.74$, $a^*=0.09$, $b^*=2.20$). L , a and b represent the colour parameters of the test samples.

The test procedure was repeated for at least 5 replicates and the average colour parameters were considered for the comparative analysis.

CHAPTER 5

VARIOUS EFFECTS OF FILM FORMATION

5.1. EFFECTS OF STARCH SOURCES

This chapter discusses the effects of different starch sources on the film properties of bionanocomposite edible films. Five starch sources, namely, (1) rice starch, (2) potato starch, (3) corn starch, (4) wheat starch, and (5) tapioca starch, were used to prepare the bionanocomposite films in this research work. Starch, glycerol, and nanocellulose were mixed as shown in Table 5.1. The rice starch based edible films reinforced with nanocellulose were considered as the control films.

Table 5.1: Film compositions of the nanocellulose incorporated starch edible films

Composition	Starch (w/v H ₂ O)	Glycerol (w/w RS)	Nanocellulose (w/w RS)
XX*+G+NC-0%	5	30	0
XX*+G+NC-2%	5	30	2
XX*+G+NC-5%	5	30	5
XX*+G+NC-10%	5	30	10
XX*+G+NC-15%	5	30	15

*XX indicates the starch source, where RS - Rice starch, PS - Potato starch, WS - Wheat starch, CS - Corn starch, and TS - Tapioca starch.

Before understanding the influence of starch sources on the film properties of the bionanocomposite films, we should first understand the differences that make up the different starch sources. Around 98-99% of starch is made of two

Glucose polymers, namely, amylose and amylopectin. While amylose is a linear polymer with low molecular weight (in the range of 10^5 - 10^6) and low degree of polymerization (in the range of 1,000-10,000), amylopectin is a highly branched polymer with high molecular weight (in the range of 10^8) and high degree of polymerization ($>10,00,000$). From the molecular weight, it was found that the molecular weight of amylopectin is approximately 1000 times higher than that of amylose (Copeland et al., 2009). The amount of amylose and amylopectin varies with the type of starch source. For example, the amount of amylose of potato starch (21%), rice starch (17%) and tapioca starch (17%) is lower than that of corn starch (26-28%) and wheat starch (25-28%). Though the percentage variation seemed to be insignificant, it was not so. The number of amylose molecules present in one gram of corn starch or wheat starch is approximately 5 times higher than that of potato starch and tapioca starch (Thakur et al., 2019; Swinkels, 1985; Liu, 2005; Domene-Lopez et al., 2019). These differences among the starch sources might have influenced the film properties of the bionanocomposite films produced from them.

5.2. EFFECTS OF PLASTICIZERS

This chapter discusses the effects of plasticizers on the film properties of edible films. There were four plasticizers considered in this research work. They are (1) glycerol, (2) xylitol, (3) sorbitol, and (4) maltitol. The edible films prepared from native rice starch plasticized with glycerol were considered as the control films. The bionanocomposites were prepared from potato starch (5g/100ml H_2O) reinforced with 10% nanocellulose. The amount of plasticizer was varied from 0% to 60% with an increment of 15% with respect to the starch content. The film composition used in the research work is presented in Table 5.2.

Composition	Starch (w/v H ₂ O)	Glycerol (w/w PS)	Nanocellulose (w/w PS)
PS+X*0+NC	5	0	10
PS+X*15+NC	5	15	10
PS+X*30+NC	5	30	10
PS+X*45+NC	5	45	10
PS+X*60+NC	5	60	10

Table 5.2: Film compositions of the plasticized bionanocomposite films

*X indicates the plasticizer type, where G - glycerol, X - xylitol, S - Sorbitol, and M - Maltitol.

The edible films made from the native bio-based materials are generally brittle and hard, which need to be overcome in order to improve the functional properties of the films (Thakur et al., 2019; Suderman et al., 2018; Suderman et al., 2019). In order to improve the film flexibility, a plasticizer is added to the edible polymer during the preparation of film forming solution. The type of plasticizer could significantly affect the film properties of the bionanocomposite films produced. Hence, we should first compare the properties of the plasticizers used. Table 5.3. shows the properties of the plasticizers used in this research. When moving from glycerol to maltitol in the table, the molecular weight, the number of hydroxyl group and the melting point increases.

Plasticizer	Formula & Shape	Molecular Wt. (g/mol)	Specific Gravity	No. of hydroxyl group	Melting Pt. (°C)	Hygroscopy
Glycerol	C ₃ H ₈ O ₃ , Straight Chain	92.09	1.25	3	20	High
Xylitol	C ₅ H ₁₂ O ₅ , Straight Chain	152.15	1.52	5	94	High
Sorbitol	C ₆ H ₁₄ O ₆ , Straight Chain	182.17	1.28	6	100	Average
Maltitol	C ₁₂ H ₂₄ O ₁₁ , Ring Structure	344.31	1.39	9	157	Average

Table 5.3: Properties of the plasticizers

CHAPTER 6

RESULT AND DISCUSSION

6.1. EFFECTS OF NANOFILLERS

This chapter discusses the effects of nanofillers on the film properties of edible films. There were two nanofillers considered in this research work. They are (1) nanocellulose obtained from banana pseudo stems, and (2) nanostarch obtained from rice starch. The edible films, prepared from native rice starch, were considered as the control films. Glycerol was used as the plasticizer. The amount of starch and glycerol used to prepare the edible films were fixed, while the amount of nanofiller was varied from 0% to 15%. The film compositions are given Table 6.1. and Table 6.2.

Table 6.1: Film compositions of the nanocellulose incorporated rice starch edible films

Composition	Starch (w/v H ₂ O)	Glycerol (w/w RS)	Nanocellulose (w/w RS)
RS+G+NS-0%	5	30	0
RS+G+NS-2%	5	30	2
RS+G+NS-5%	5	30	5
RS+G+NS-10%	5	30	10
RS+G+NS-15%	5	30	15

Table 3.2: Film compositions of the nanostarch incorporated rice starch edible films

Composition	Starch (w/v H ₂ O)	Glycerol (w/w RS)	Nanostarch (w/w RS)
RS+G+NS-0%	5	30	0
RS+G+NS-2%	5	30	2
RS+G+NS-5%	5	30	5
RS+G+NS-10%	5	30	10
RS+G+NS-15%	5	30	15

6.2. EFFECT OF NANOFILLERS ON WVTR

Fig 6.1 shows the effects of the nanocellulose and nanostarch on WVTR of rice starch based bionanocomposite edible films. The WVTR of the control rice starch edible films was found to be 141.14 g-mm/m²-day. Literature showed that WVTR of rice starch films varied from 1.29 g.mm/m².day to 213 g.mm/m².day. Among them, produced the rice starch edible films with WVTR as 127.49 ± 0.14 g.mm/m².day and 95.1 ± 9.12 g.mm/m².day respectively, which are in close agreement with the WVTR of the control films produced in this research work. As both rice starch and glycerol are hydrophilic in nature, the edible films produced from them would allow water vapour to permeate through the film. The dimensional dissimilarities and the weak interactions between the starch molecules and the plasticizer could increase the moisture adsorption causing relatively higher WVTR for the control films.

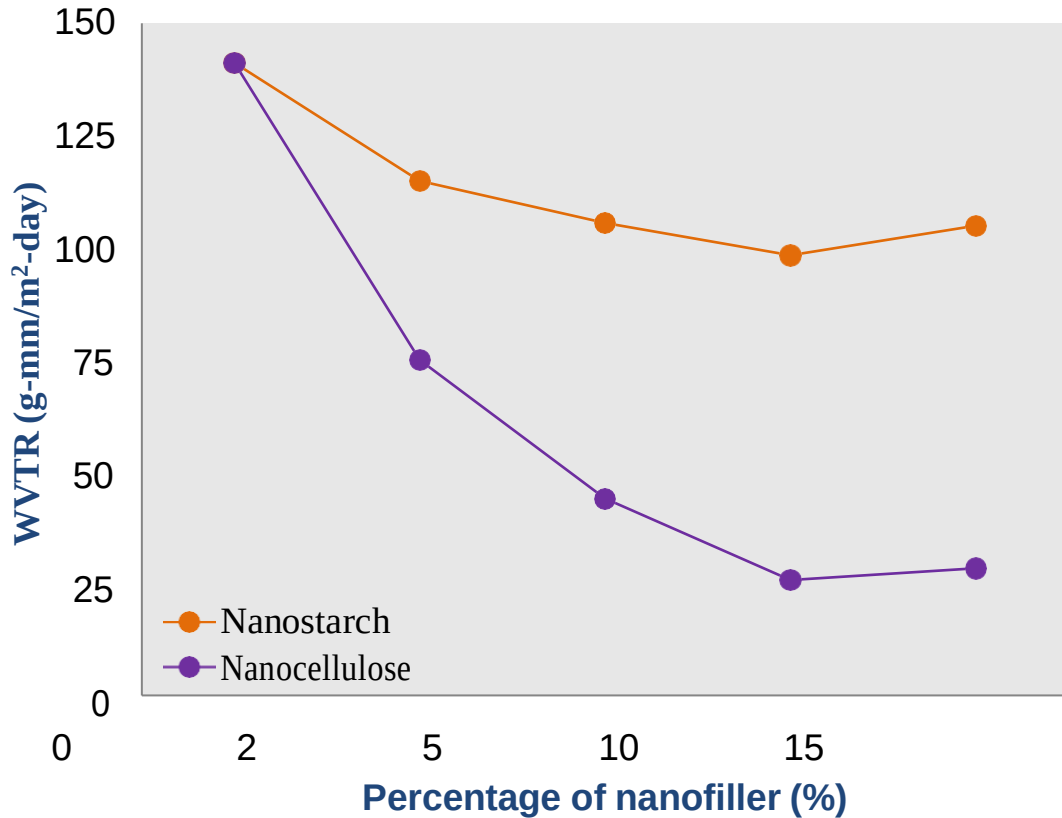


Fig. 3.1: WVTR of nanofiller incorporated rice starch films

From the figure, it could be seen that WVTR of the nanofiller incorporated bionanocomposite edible films was higher than that of the control edible films. The trend showed that the water vapour resistance of the rice starch based edible films was increased due to the addition of nanofillers irrespective of whether the nanofiller was nanocellulose or nanostarch. As the size of the nanofillers are very less, the surface area to volume ratio was very high as compared to the macro- or micro-scaled particles. Therefore, both nanocellulose or nanostarch could make strong bonding with the hydroxyl group of the rice starch, and hence the nanofiller incorporated bionanocomposite edible films could delay the water interaction with the rice starch films. With the addition of the nanoparticles within rice starch edible films, there could be an increased number of hydroxyl groups available. These hydroxyl groups could interact with the unbound water vapour molecules resulting the reduced water activity by making the hydrogen bonds. This, in turn, reduced the WVTR. Furthermore, the addition of nanofillers could also act as the obstacles to prevent the permeation of water molecules. So, the water vapour passing

through a bionanocomposite film might have to travel a long path than that of the macromolecule film. As a result, the addition of nanofiller in rice starch edible films increased the travel length or provided a tortuous path that slowed down the diffusion of water molecules through the edible films. Hence, the addition of nanofillers reduced the WVTR. As both nanostarch and nanocellulose exhibit the chemical and structural similarities with the rice starch, there would be less chance of dimensional changes, which, in turn, could lower the number of defects and create strong adhesion. Therefore, the resistance to diffusion of water vapours increased, and the nanofiller incorporated edible films showed the reduced WVTR as compared to the control films. Among nanocellulose and nanostarch, the addition of nanocellulose produced the rice starch edible films with lower WVTR than that of nanostarch based edible films. According to nanostarch was less likely to create the tortuosity to allow the water vapour due to its platelet morphology. Hence, the WVTR of nanostarch incorporated films was found to be relatively high as compared to the nanocellulose incorporated films.

The nanocellulose incorporated rice starch edible films showed higher resistance to water vapours than that of the control films. WVTR values of bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 74.82 ± 6.43 g.mm/m².day, 43.9 ± 8.07 g.mm/m².day, 25.72 ± 2.85 g.mm/m².day, and 32.7 ± 2.75 , respectively. From the results, it was observed that, as the nanocellulose content increased from 0% to 10%, the WVTR of the bionanocomposite edible films decreased, after which it started decreasing. Similar reduction of WVTR was found in bionanocomposite edible films produced from alginate, sugarcane bagasse, cassava starch, whey protein. Due to the dimensional differences of starch and glycerol, the adhesion between the starch-glycerol interfaces is poor. Hence, the edible films produced from rice starch with glycerol as plasticizer might promote adsorption of moisture and hence the increased WVTR. However, because starch and nanocellulose are chemically similar to each other, the interactions between them are more stronger. Due to the chemical similarity, the structural and interface defects of the produced edible films decreased, and the resistance to the passage of water molecules increased. Hence, the addition of nanocellulose with rice starch reduced the WVTR of

bionanocomposite edible films. The nanostarch incorporated rice starch edible films also showed higher resistance to water vapours than that of the control films, but relatively lower resistance than that of the nanocellulose based edible films. WVTR values of nanostarch incorporated bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 114.8 ± 8.96 g.mm/m².day, 105.4 ± 2.97 g.mm/m².day, 98.2 ± 4.32 g.mm/m².day, and 104.8 ± 5.45 , respectively. As the amount of the nanostarch increased, the WVTR of the bionanocomposite films was reduced up to 10%, above which WVTR was increased at 15%. The reduction in water vapour resistance could have been resulted due to the nanosize and the material compatibility. Due to the nanoscale, the nanostarch could have dispersed well as well as made the intermolecular bonding among the starch molecules. As both matrix and nanofiller were derived from the same botanical origin (rice starch, in this research work), they have similar polarity and the nanofiller-matrix interactions were stronger, which could have weakened the matrix-water interactions. As a result, a slight decrease in WVTR could be observed in the starch edible films reinforced with nanostarch. The WVTR of bionanocomposite edible films was decreasing when the nanostarch was above 10%. This could be attributed to the formation of aggregates at higher addition of nanostarch. Similar observations were found in the work of where the WVTR was increased above 20% of nanostarch. Due to high density of effective OH groups in nanostarch, they tend to form aggregates and strongly associate with each other through strong hydrogen bonding and create the preferential paths for allowing water vapours to diffuse through. Therefore, the permeability of the bionanocomposite edible films increased above 10% of nanostarch addition.

6.3. EFFECT OF NANOFILLERS ON TENSILE PROPERTIES

Fig 6.2 and Fig 6.3 show the effects of the nanocellulose and nanostarch on TS and YM of rice starch based bionanocomposite edible films. The TS and YM of the control rice starch edible films was found to be 7.92 ± 0.58 MPa and 19.04 ± 1.28 MPa, respectively. Literature showed that TS of rice starch films varied from 1.29 MPa to 8.9 MPa and YM varied from 0.19 MPa to 21.3 produced the rice starch based edible films with TS as 8.9 MPa and 7.12 MPa, respectively, which are in

close agreement with the TS of the control films produced in this research work. YM reported by Vargas et al. (2017) and Dias et al. (2010) were 19.08 ± 3.16 MPa and 21.3 ± 5.0 MPa, respectively, and were in close agreement with the YM values of the control films reported in this research work.

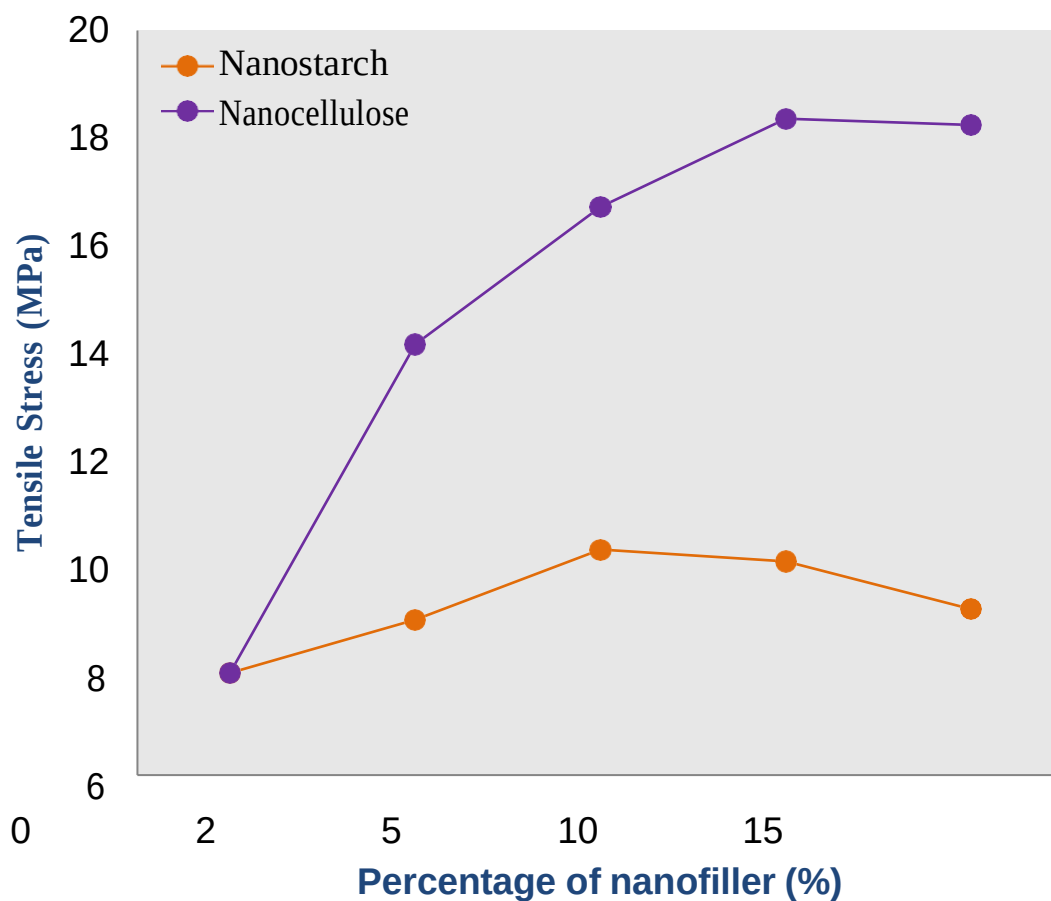


Fig: 6.2: TS of nanofiller incorporated rice starch films

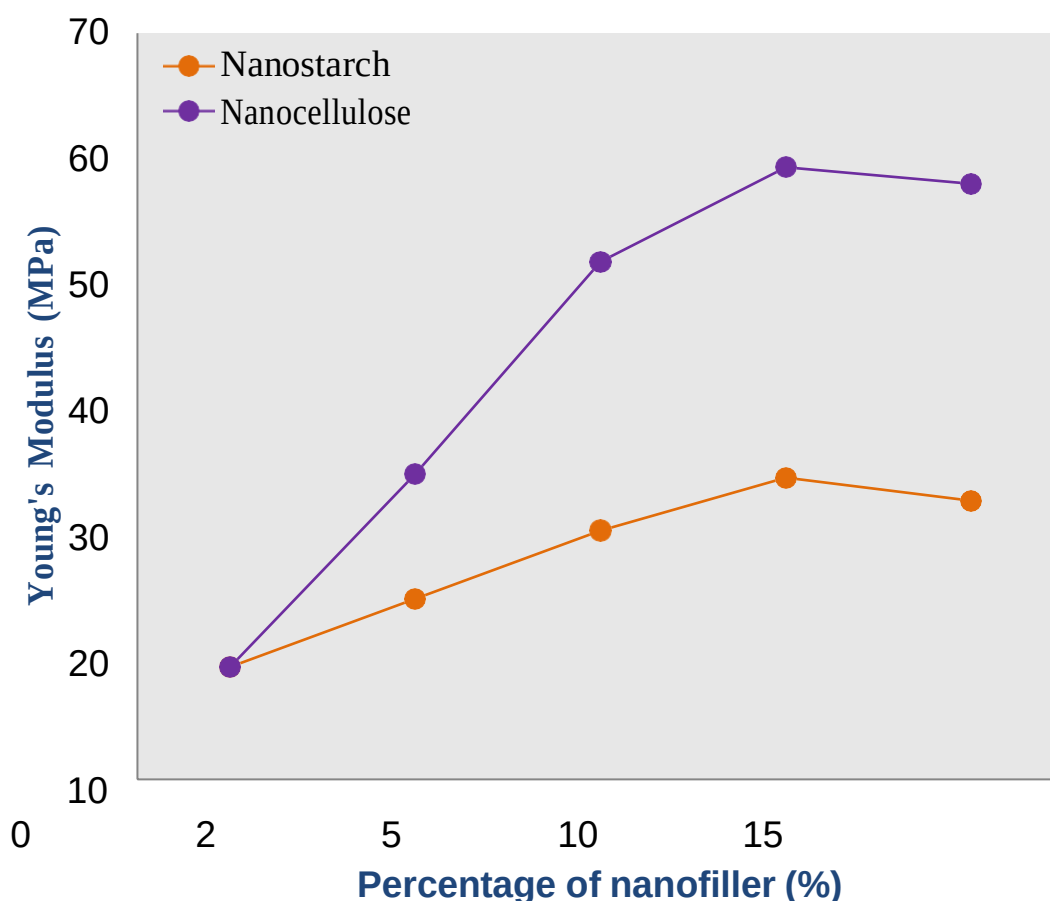


Fig: 6.3: YM of nanofiller incorporated rice starch films

It was observed that the addition of nanofiller increased the mechanical strength of the bionanocomposite edible films up to 10% of the nanofiller addition. In the control films, as glycerol has the smaller molecular structure, the glycerol molecules could enter into the starch network and breaks the matrix-matrix interactions, which promotes the plasticity of the edible films. As a result, the addition of glycerol could improve the elongation of the films, but the tensile strength of the bionanocomposite edible films would be lowered. When the nanofiller was added, the tensile strength increased. In order to achieve the higher mechanical strength from a composite, good dispersion of filler is. As both nanofillers (nanocellulose and nanostarch) are similar in chemical structure as that of the starch matrix but the filler size is extremely small, the addition of nanofiller would achieve the well dispersion into the matrix and develops relatively large quantity of filler-matrix interactions as compared to the control films. As a result, the stress transfer between the matrix and the filler would increase, which, in turn,

could increase the tensile strength. Also, because the nanofillers occupy the spaces between the starch network, the stress required to make any deformation would increase. Therefore, the Young's modulus of the nanofiller incorporated bionanocomposite edible films was found to be higher than the control films. However, when the percentage of nanofiller was above 10%, the mechanical strength started decreasing, which might be a result of the formation of aggregation. The formation of aggregates form the weak bonding between the matrix-filler interactions, and, therefore, the efficacy of the stress transfer would reduce, which, in turn, could have reduced the mechanical strength of the bionanocomposite edible films with more than 10% nanofiller addition.

The results showed that the addition of nanocellulose increased both TS and YM of the bionanocomposite edible films. TS values of nanocellulose incorporated bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 10.7 MPa, 13.48 MPa, 14.94 MPa and 13.92 MPa, respectively. YM values of nanocellulose incorporated bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 34.56 MPa, 41.22 MPa, 47.22 MPa and 46.3 MPa, respectively. As the percentage of nanocellulose increased from 0% to 10%, the TS and YM of the bionanocomposite edible films were increasing. The increase of the mechanical strength due to the addition of nanocellulose was reported in literature for the bionanocomposite edible films produced from the polymeric materials, such as cassava starch, sodium caseinate, potato starch and chitosan. The mechanical strength of the bionanocomposite is affected by morphology and dimensions of the nanofiller, the film forming method, and the interactions between the matrix and matrix/filler. From the literature, it was observed that the tensile strength and the Young's modulus of single nanocellulose fibrils are in the ranges of 2–7.7 GPa and 100-220 GPa, respectively, for which, the strength-to-weight ratio of nanocellulose is about eight times higher than that of stainless steel. Hence, the addition of nanocellulose had improved the overall mechanical strength of the bionanocomposite edible films. When the percentage of nanocellulose was above 10% threshold, both TS and YM of the nanocellulose incorporated bionanocomposite films were decreasing slightly. Above 10% of nanocellulose addition, nanocellulose started to aggregate, which might have

weakened the adhesion between the nanofiller-matrix interfaces. Therefore, for higher nanofiller addition, nanocellulose could have self-aggregated to destroy the structure orderliness leading to lower mechanical strength.

The addition of nanostarch in rice starch films had resulted in the increase of TS and YM. TS values of nanostarch incorporated bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 8.92 MPa, 10.24 MPa, 10.02 MPa and 9.12 MPa, respectively. YM values of nanostarch incorporated bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 24.52 MPa, 30 MPa, 34.26 MPa and 32.4 MPa, respectively. As the percentage of nanostarch increased from 0% to up to 10%, the TS and YM of the bionanocomposite edible films were increasing. Above this threshold value, a slight decrease in the TS and YM was observed. The increase of TS and YM were reported in literature Liu et al. (2015), Condés et al. (2015), Angellier et al. (2006), Zheng et al. (2009), Da-Silva et al. (2017) and Ren et al. (2016) by incorporating nanostarch in edible films, and the results presented in this research work were in good agreement. As the nanoparticles have high surface area to volume ratio, the mechanical properties of the bionanocomposite films are strongly influenced by the nanofiller–matrix interfacial interactions. Uniformly distributed nanoparticles with strong interfacial bonding with the matrix offer the effective stress transfer from matrix to nanoparticles, thus increasing the modulus and tensile strength of the bionanocomposites. However, when the concentration of starch nanoparticles is above a threshold value (10% in this research), due to self-aggregation of nanoparticles, there was a slight reduction in TS and YM of the bionanocomposite films.

Among the two nanofillers, the TS and YM of nanocellulose reinforcement were found to be higher than those of nanostarch reinforcement. In order to have better mechanical performance, the nanofiller should have high aspect ratio. While nanocellulose has the aspect ratio of about 11-67, nanostarch has poor aspect ratio due to its spherical or platelet morphology. Additionally, the mechanical strength of individual nanocellulose fibers was found to be very high. Another factor considered here was the degree of crystallinity that should be higher for

maximizing the mechanical performance. While the crytallinity of nanocellulose was about 70-85%, it is 45–50 % for the nanostarch. As a result, the nanocellulose incorporated bionanocomposite films showed higher TS and YM as compared to nanostarch incorporated films.

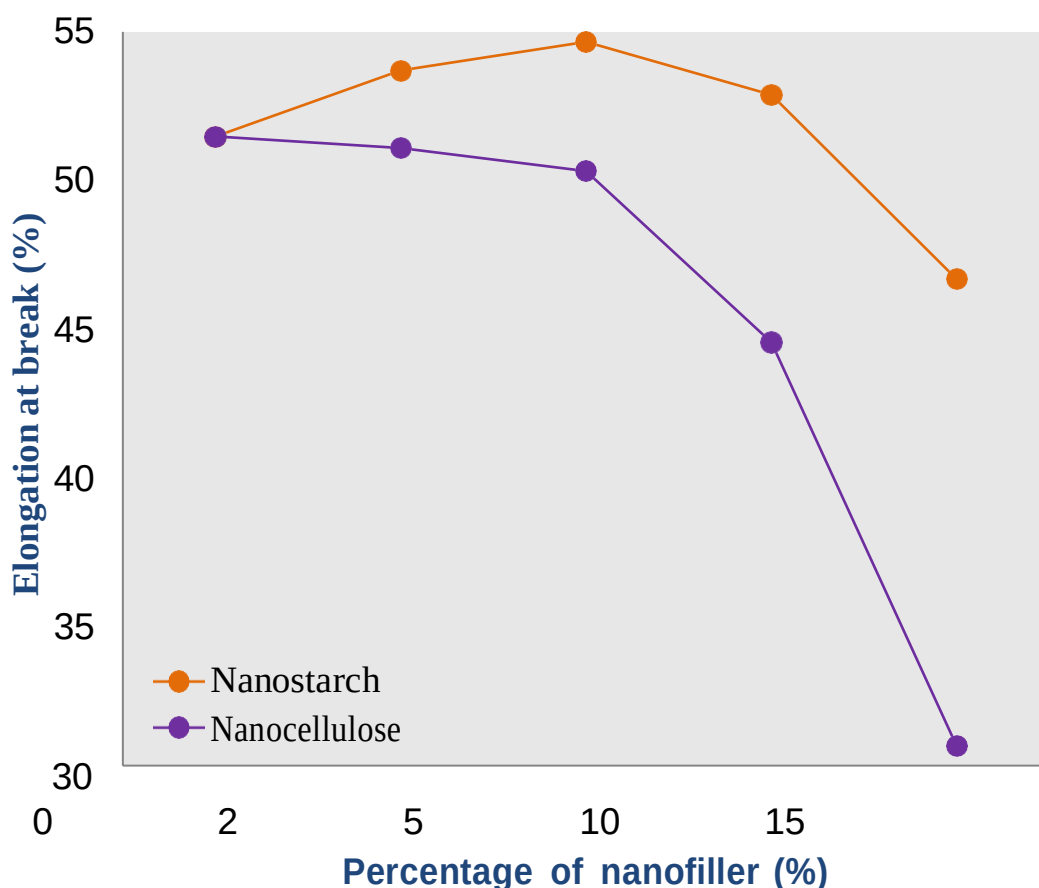


Fig: 6.4: Elongation at break of nanofiller incorporated rice starch films

Fig 3.4 shows the effects of nanofillers on elongation at break (E) of the bionanocomposite films. The control films have the elongation of 51.44 ± 1.25 %. Literature have shown that the elongation of the native rice starch films varied from 0.2% to 116%. Among these had produced the rice starch films with the elongation of 49.73 ± 11.00 %, $\sim 53.46\%$ and $59.8 \pm 9.9\%$, which were close to the elongation values presented in this research work, and hence, the results were in good agreement.

From the results, it was observed that the addition of nanostarch initially increased

the elongation at break of the bionanocomposite films up to 10% addition, above which the elongation started decreasing. Elongation at break values of nanostarch incorporated bionanocomposite edible films at 2%, 5%, 10% and 15% were found to be 53.68%, 54.66%, 52.86% and 46.58%, respectively. This showed that the film flexibility of the edible films was improved with up to 10% addition of nanostarch. Similar increase of elongation at break was observed due to the addition of nanofillers, where 1% nanostarch addition increased the elongation at break of the potato starch films. The addition of nanostarch on pea starch edible films reduced the elongation at break from 29.23% (at control films) initially to 12.58% at 7% reinforcement, but the elongation at break then increased to 26.7% at 9% reinforcement. The increase of elongation at break due to the addition of. Therefore, these results are in good agreement with the results of the bionanocomposites prepared in this research work. Due to the extremely small size of the nanofiller, the adhesion between the matrix and filler is not 100%. So, the mobility increases slightly, which can cause the bionanocomposite films to possess higher elongation at break as compared to the control films. However, when the nanostarch addition was higher than 10%, the elongation at break of the bionanocomposite edible films started reducing. At higher nanostarch addition, the free volume of the bionanocomposite films, which could have reduced the mobility of polymer chains. Due to limited mobility, the brittleness of the bionanocomposite films increased, which, in turn, had reduced the elongation at break at higher percentage of nanostarch addition.

We have seen that the addition of nanostarch initially increased the elongation at break of the bionanocomposite films up to 10% nanostarch addition, but the addition of nanocellulose reduced the elongation at break. Elongation at break values of nanocellulose incorporated bionanocomposite films at 2%, 5%, 10% and 15% were found to be 51.04%, 50.26%, 44.42% and 30.68%, respectively. The results showed that, with the increase of nanocellulose content, the elongation at break of the bionanocomposite films gradually decreased. The decreasing elongation due to the addition of nanocellulose. The addition of nanocellulose up to 5% did not have significant decrease in the elongation of bionanocomposite films. Up to 5% addition of nanocellulose, the addition of nanoparticles could have filled the voids between the starch matrix, and, therefore, the elongation at break of the bionanocomposite films was not affected at low concentration up to 5%.

When the nanocellulose concentration was higher than 5% in the bionanocomposite films, the elongation at break started decreasing significantly. With the addition of nanocellulose in the starch matrix, the polymer network becomes more rigid restricting the mobility of the starch molecular chain. Due to the restricted mobility, the elongation of the bionanocomposite films was reduced with the addition of nanocellulos.

6.4. EFFECT OF NANOFILLERS ON FILM COLOUR

Fig 6.5, Fig 6.6, Fig 6.7 and Fig 6.8 show the effects of the nanocellulose and nanostarch on colour parameters of rice starch based bionanocomposite edible films. The results showed that the film colour parameters, L, A and B, of the control films are 89.2 ± 1.3 , 2.2 ± 0.45 and 3.6 ± 1.14 , respectively. In the work of Ashwar et al. (2015), the values of L, A and B of the native rice starch films were reported as 90.68 ± 0.20 , -0.37 ± 0.05 and 4.89 ± 0.70 , respectively. In the work of Thakur et al. (2016), the L, a & b parameters were found to be 95.52, -0.31 & 5.03, respectively. To produced the rice starch films with L, a & b values are approximately 87, -1.3 and 2.5 respectively. The values of L, A and B of the native rice starch films were reported as 87, -1 and 1.2 (approximately), respectively. Hence, the results reported for the control films in this research work are in good agreement with the results reported in literature.

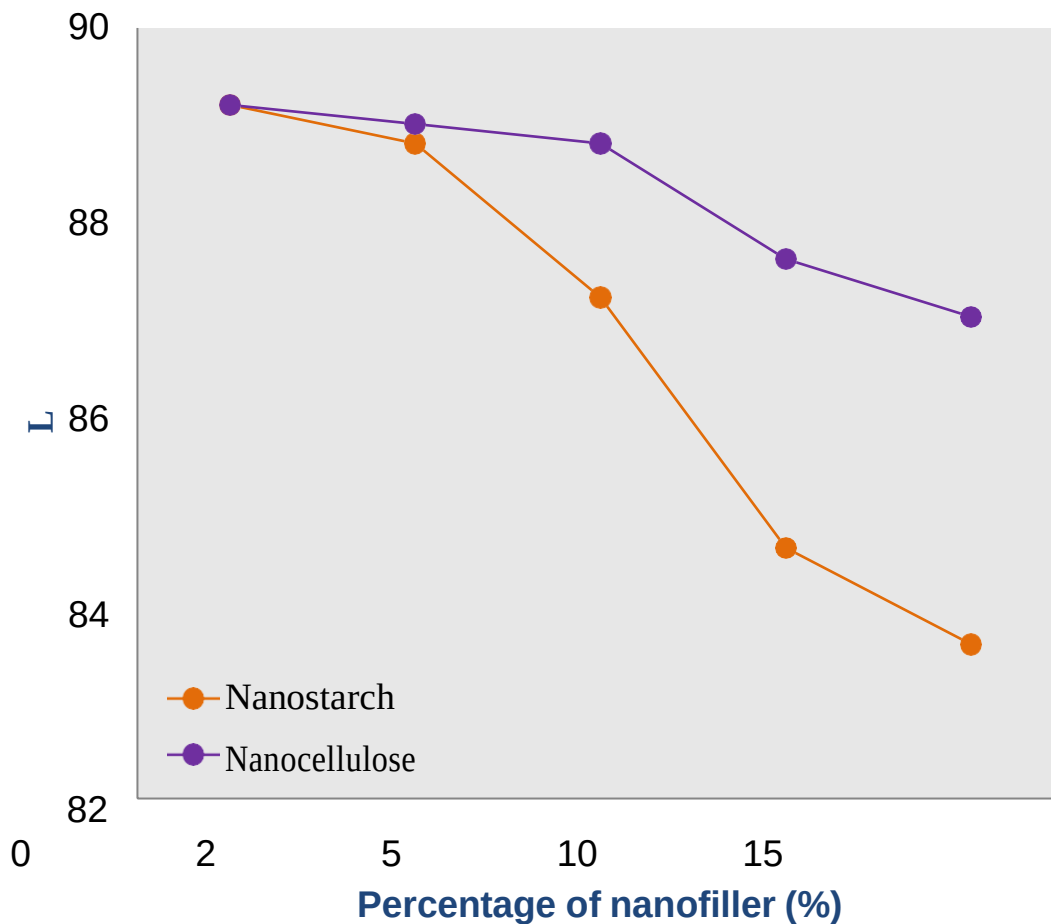


Fig: 6.5: 'L' values of nanofiller incorporated rice starch films

From the results, it was understood that the addition of nanofillers did not affect the film colour parameters greatly, but a slight variations were noticed. With the addition of nanofiller, no significant difference was found in the parameter 'a', but the colour parameters, 'L' and 'b' were slightly increasing. Due to nano-size, the diameter of the nanofiller would be lower than the wavelength of light and, the scattering of visible light would be decreased. As a result, the bionanocomposite films could experience higher transparency as compared to the control films, which could result no significant difference in the colour parameters at lower concentrations of nanofillers. Further, the addition of nanofillers would fill the voids between the starch molecules of the control films, making the surface more even, which would allow the light rays scattered making the film bright. Therefore, at lower concentration of nanofiller addition, the change of colour parameters, 'L' as well as 'b', of the bionanocomposite films was very low. However, when the

addition of nanofiller became significant, the transparency of the nanocomposite films was affected, and the films became darker.

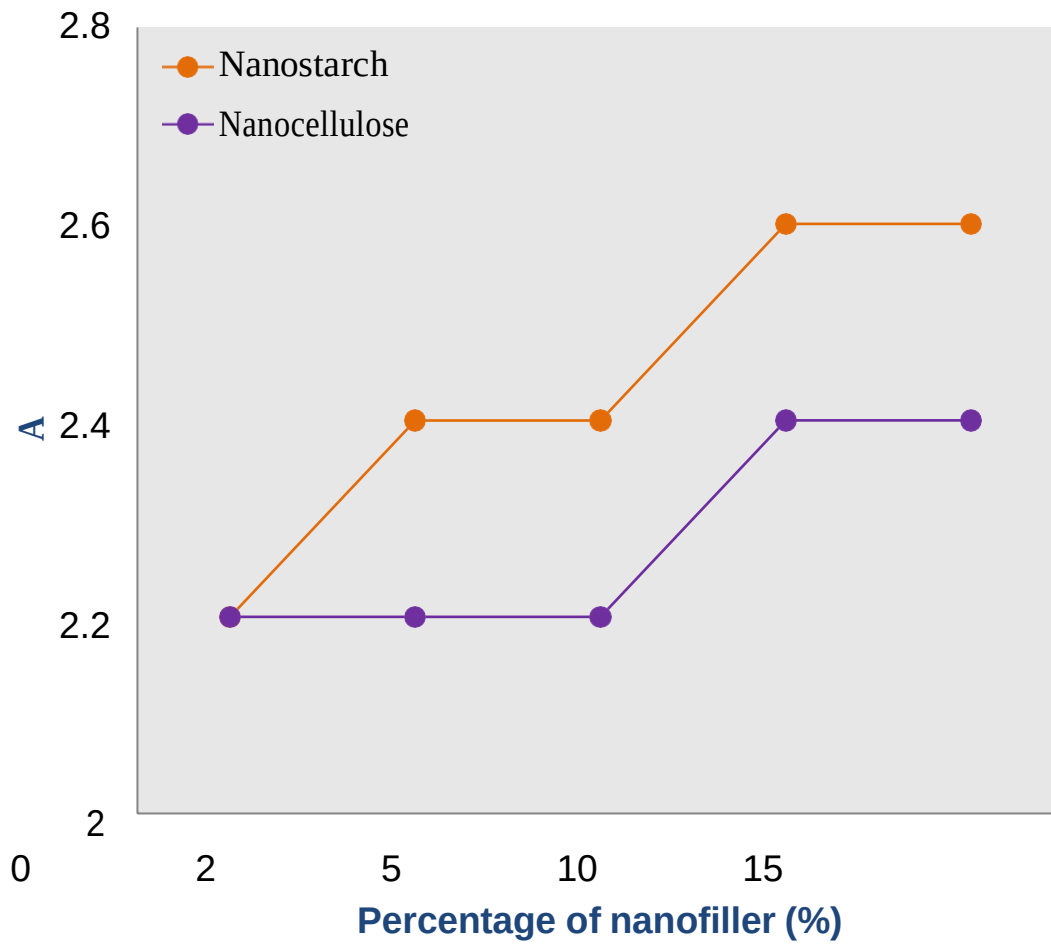


Fig: 6.6: 'A' values of nanofiller incorporated rice starch films

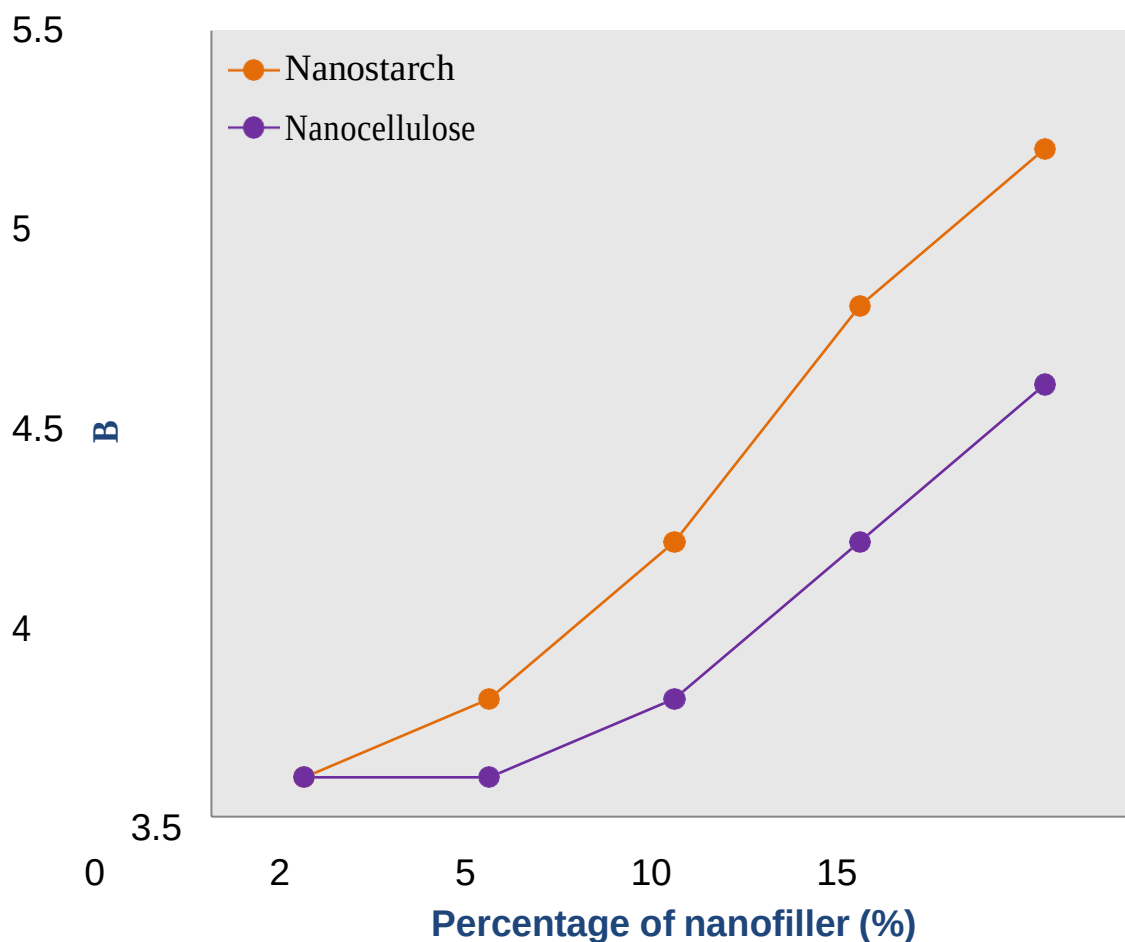


Fig: 6.7: 'B' values of nanofiller incorporated rice starch films

While the addition of nanocellulose slightly decreased the 'L' colour parameter and slightly increased the 'b' colour parameter, there was no noticeable change in 'a' values observed in the bionanocomposite films. Increasing 'L' and decreasing 'b' values due to the addition of nanocellulose. Therefore, it was understood from the results that the addition of nanocellulose had made the bionanocomposite films slightly darker and yellowish. Similar to nanocellulose, the incorporation of nanostarch on rice starch based bionanocomposite films reduced the colour parameters, 'L', and increased 'b' slightly. Hence, the bionanocomposite films reinforced with nanostarch turns the films slightly darker and yellowish. Similar observations due to the addition of nanostarch on other starch films.

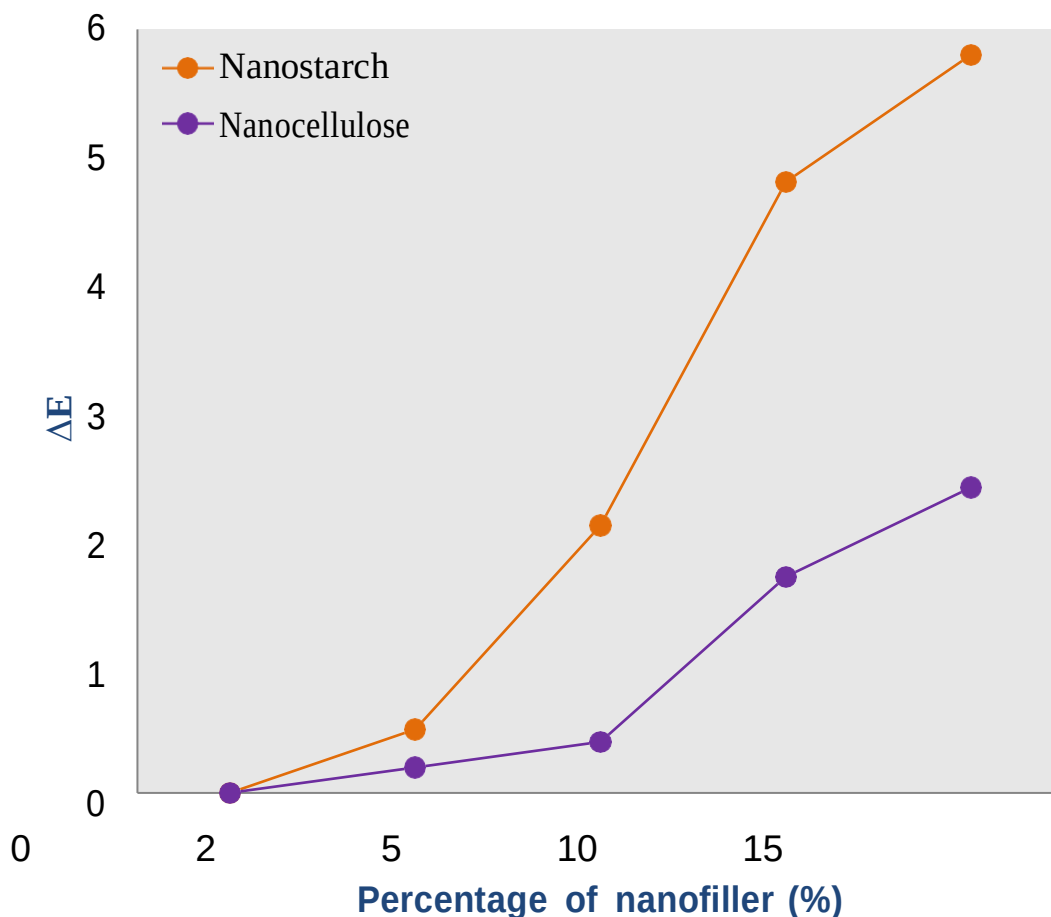


Fig: 6.8: ' ΔE ' values of nanofiller incorporated rice starch films

The overall colour change (ΔE) of the nanocellulose incorporated films was increasing from 0.5 ± 0.11 at 2% nanocellulose to 2.4 ± 0.53 at 15% nanocellulose. As the highest ΔE was lesser than 5%, the change in colour due to nanocellulose was found to be insignificant. On the other hand, ΔE of the nanostarch incorporated films was significantly changing when the nanostarch addition was 15%. which is found to be higher than that of the nanocellulose incorporated films. At 15% concentration, ΔE of the nanostarch incorporated films was found to be 5.8 ± 0.47 . Also, the ΔE values of nanostarch incorporated films were higher at all nanofiller concentrations as compared to those of nanocellulose incorporated films. Nanostarch contains both crystalline and amorphous regions, and the degree of crystallinity of nanostarch was about 40-60% . But, the crystallinity of nanocellulose was about 70-85%. Due to higher amorphous regions

of nanostarch, the light absorbed or diffracted would be greater in the nanostarch incorporated films as compared to the nanocellulose incorporated films.

CHAPTER 7

CONCLUSIONS

In this project, the effects of two nanofillers, nanocellulose obtained from the banana pseudo stems and nanostarch obtained from the native rice starch, on the film properties of the bionanocomposite edible films were investigated. Based on the results and discussion, the following findings are summarized.

The WVTR of the control rice starch edible films was found to be 141.14 g- mm/m²-day. Both nanocellulose as well as nanostarch are hydrophilic in nature. However, the WVTR was reduced with the addition of nanofillers of up to 10% concentration due to improved dispersion, increased number of interactions and the tortuosity offered by the nanofiller. However, at 10% nanofiller addition, the elongation of the nanocellulose incorporated bionanocomposite films was 50.26%, which was lower than 52.86%, the same of nanostarch incorporated films. From these findings, despite the fact that there is a slight penalty on the elongation at break in the nanocellulose films, it is found that the nanocellulose addition was more effective to produce the stronger bionanocomposite edible films as compared to nanostarch films. The highest overall colour change (ΔE) was only 2.4 ± 0.53 for the nanocellulose incorporated films, while it was 5.8 ± 0.47 for the nanostarch incorporated films. Despite the overall colour change of both bionanocomposites, due to high crystallinity and high transparency, the nanocellulose was found to be more effective in not deteriorating the film colour as compared to nanostarch.

REFERENCES

- [1] Abdollahi M, Alboofetileh M, Behrooz R, Rezaei M & Miraki R. Reducing water sensitivity of alginate bio-nanocomposite film using cellulose nanoparticles. *Int J Biological Macromolecules* 2013; 54: 166–173.
- [2] Adamu, A. D., Jikan, S. S., Talip, B. H. A., Badarulzaman, N. A., & Yahaya, S. (2017). Effect of Glycerol on the Properties of Tapioca Starch Film. *Materials Science Forum*, 888, 239–243
- [3] Afshari-Jouybaria H and Farahnaky A. Evaluation of Photoshop software potential for food colorimetry. *J Food Engineering* 2013; 106: 170-175.
- [4] Alcantara, C.R., Rumsey, T.R., & Krochta, J. M. (1998). Drying rate effect on the properties of whey protein films. *Journal of Food Process Engineering*, 21(5), 387–405.
- [5] Al-Hassan, A. A., & Norziah, M. H. (2012). Starch–gelatin edible films: Water vapor permeability and mechanical properties as affected by plasticizers. *Food Hydrocolloids*, 26(1), 108–117.
- [6] Andrady, A. L., & Neal, M. A. (2009). Applications and societal benefits of plastics. *Philosophical Transactions of the Royal Society B: Biological Sciences*, 364(1526), 1977–1984
- [7] Angellier H, Choisnard L, Molina-Boisseau S, Ozil P, Dufresne A (2004) Optimization of the Preparation of Aqueous Suspensions of Waxy Maize Starch Nanocrystals Using a Response Surface Methodology. *Biomacromolecules* 5: 1545–1551.
- [8] Arun, S., Kumar, K. A. A., & Sreekala, M. S. (2012). Fully biodegradable potato starch composites: effect of macro and nano fiber reinforcement on mechanical, thermal and water-sorption characteristics. *International Journal of Plastics Technology*, 16(1), 50–66.
- [9] Arvanitoyannis, I., Psomiadou, E., & Nakayama, A. (1996). Edible films made from sodium caseinate, starches, sugars or glycerol. Part 1. *Carbohydrate Polymers*, 31(4), 179–192.
- [10] Arvanitoyannis, I.S. and Bosnea, L. (2004). Migration of Substances from Food Packaging Materials to Foods, *Critical Reviews in Food Science and Nutrition*, 44(2), 63-76.

- [11] Ashwar, B.A., Shah, A., Gani, A., Shah, U., Gani, A., Wani, I.A., Wani, S.M., Masood, F.A. (2015). Rice starch active packaging films loaded with antioxidants—development and characterization, *Starch/Stärke*, 67: 294–302
- [12] Azeredo HMC, Mattoso LHC, Avena-Bustillos RJ, Filho GC, Munford ML, Wood D & McHugh TH. Nanocellulose Reinforced Chitosan Composite Films as Affected by Nanofiller Loading and Plasticizer Content. *J Food Science* 2010; 75: N1–N7.
- [13] Bagde, P., & Vigneshwaran, N. (2019). Mechanical, antibacterial and biodegradable properties of starch film containing bacteriocin immobilized crystalline nanocellulose. *Carbohydrate Polymers*, 115021.
- [14] Balqis, A. M.I., Nor Khaizura, M. A. R., Russly, A. R., & Nur Hanani, Z. A. (2017). Effects of plasticizers on the physicochemical properties of kappa-carrageenan films extracted from *Eucheuma cottonii*. *International Journal of Biological Macromolecules*, 103, 721–732.
- [15] Barrow, M. (2005). An Economic Analysis of the UK Landfill Permits Scheme. *Fiscal Studies*, 24(3), 361–381.
- [16] Basiak, E., Galus, S., & Lenart, A. (2014). Characterisation of composite edible films based on wheat starch and whey-protein isolate. *International Journal of Food Science & Technology*, 50(2), 372–380.
- [17] Basiak, E., Lenart, A., & Debeaufort, F. (2017). Effect of starch type on the physico-chemical properties of edible films. *International Journal of Biological Macromolecules*, 98, 348-356.
- [18] Beck-Candanedo S, Roman M, Gray DG. Effect of reaction conditions on the properties and behavior of wood cellulose nanocrystal suspensions. *Biomacromolecules*. 2005; 6: 1048–1054.
- [19] Begley, T.H. (1997). Methods and approaches used by FDA to evaluate the safety of food packaging materials, *Food Additives & Contaminants*, 14(6-7), 545-553.
- [20] Börjesson, M., & Westman, G. (2015). Crystalline nanocellulose—preparation, modification, and properties. *Cellulose-fundamental aspects and current trends*, 159-191.
- [21] Bourtoom, T. (2008). Edible films and coatings: characteristics and properties. *International Food Research Journal*, 15(3), 237-248.
- [22] Bourtoom, T. (2008). Plasticizer effect on the properties of biodegradable blend film from rice starch-chitosan. *Songklanakarin Journal of Science & Technology*, 30.

- [23] Bourtoom, T., Chinnan, M. S., Jantawat, P., & Sanguandeeikul, R. (2006). Effect of Plasticizer Type and Concentration on the Properties of Edible Film from Water-Soluble Fish Proteins in Surimi Wash-Water. *Food Science and Technology International*, 12(2), 119–126.
- [24] Callegarin, F., Gallo, J.A.Q., Debeaufort, F. and Voilley, A. (1997). Lipids and Biopackaging, *JAOCs*, 74(10), 1183-1192.
- [25] Chaichi, M., Hashemi, M., Badii, F., & Mohammadi, A. (2017). Preparation and characterization of a novel bionanocomposite edible film based on pectin and crystalline nanocellulose. *Carbohydrate Polymers*, 157, 167–175.
- [26] Charreau HL, Foresti M and Vazquez A (2013) Nanocellulose Patents Trends: A Comprehensive Review on Patents on Cellulose Nanocrystals, Microfibrillated and Bacterial Cellulose, *Recent Patents on Nanotechnology* 7: 56–80.
- [27] Chaudhary, A. L., Torley, P. J., Halley, P. J., McCaffery, N., & Chaudhary, D. S. (2009). Amylose content and chemical modification effects on thermoplastic starch from maize – Processing and characterisation using conventional polymer equipment. *Carbohydrate Polymers*, 78(4), 917–925.
- [28] Chen, Q., Liu, Y., & Chen, G. (2019). A comparative study on the starch-based biocomposite films reinforced by nanocellulose prepared from different non-wood fibers. *Cellulose*. doi:10.1007/s10570-019-02254-x
- [29] Condés, M. C., Añón, M. C., Mauri, A. N., & Dufresne, A. (2015). Amaranth protein films reinforced with maize starch nanocrystals. *Food Hydrocolloids*, 47, 146–157.
- [30] Copeland, L., Blazek, J., Salman, H., & Tang, M. C. (2009). Form and functionality of starch. *Food hydrocolloids*, 23(6), 1527-1534.
- [31] Crosby, A. J., & Lee, J. Y. (2007). Polymer nanocomposites: the “nano” effect on mechanical properties. *Polymer reviews*, 47(2), 217-229.
- [32] Cuq, B., Gontard, N. and Guilbert, S. (1998). Proteins as Agricultural Polymers for Packaging Production, *Cereal Chemistry*, 75(1), 1–9.
- [33] Cutter, C.N. (2006). Opportunities for bio-based packaging technologies to improve the quality and safety of fresh and further processed muscle foods, *Meat Science*, 74, 131-142.
- [34] Da Silva, N. M. C., de Lima, F. F., Fialho, R. L. L., Albuquerque, E. C. D. M. C., Velasco, J. I., & Fakhouri, F. M. (2018). Production and Characterization of Starch Nanoparticles. *Applications of Modified Starches*, 41.

- [35] Dai, L., Qiu, C., Xiong, L., & Sun, Q. (2015). Characterisation of corn starch-based films reinforced with taro starch nanoparticles. *Food chemistry*, 174, 82-88.
- [36] Debeaufort, F. and Voilley, A. (1994). Aroma Compound and Water Vapor Permeability of Edible Films and Polymeric Packagings, *J. Agric. Food Chem.*, 42, 2071-2075.
- [37] Debeaufort, F., & Voilley, A. (2009). Lipid-Based Edible Films and Coatings. *Edible Films and Coatings for Food Applications*, 135–168.
- [38] Debeaufort, F., Quezada-Gallo, J.A. and Voilley, A. (1998). Edible Films and Coatings: Tomorrow's Packagings: A Review, *Critical Reviews in Food Science and Nutrition*, 38(4), 299-313.
- [39] Demirbas, A. (2007). Biodegradable Plastics from Renewable Resources. *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects*, 29(5), 419–424.
- [40] Detduangchan, N., Wittaya, T. (2011). Effect of UV-Treatment on Properties of Biodegradable Film From Rice Starch, *World Academy of Science, Engineering and Technology, International Journal of Materials and Metallurgical Engineering*, 5(9): 829-834.
- [41] Domene-López, D., García-Quesada, J. C., Martín-Gullón, I., & Montalbán, M. G. (2019). Influence of Starch Composition and Molecular Weight on Physicochemical Properties of Biodegradable Films. *Polymers*, 11(7), 1084.
- [42] Elanthikkal S, Gopalakrishnanapanicker U, Varghese S & Guthrie JT. Cellulose microfibrils produced from banana plant wastes: Isolation and characterization. *Carbohydrate Polymers* 2010; 80: 852–859.
- [43] Fahma F, Imamoto S, Hori N, Iwata T, Takemura A. Effect of pre-acid-hydrolysis treatment on morphology and properties of cellulose nanowhiskers from coconut husk. *Cellulose*. 2011;18:443–450.
- [44] Farahnaky, A., Saberi, B., & Majzoobi, M. (2013). Effect of Glycerol on Physical and Mechanical Properties of Wheat Starch Edible Films. *Journal of Texture Studies*, 44(3), 176–186.
- [45] Farajpour, R., Djomeh, Z. E., Moeini, S., Tavahkolipour, H., & Safayan, S. (2020). Structural and physico-mechanical properties of potato starch-olive oil edible films reinforced with zein nanoparticles. *International Journal of Biological Macromolecules*, 149, 941-950.

- [46] Federation of Indian Chambers of Commerce and Industry (FICCI). (2016). A Report on Plastic Industry, 2nd National Conference on Plastic Packaging – The Sustainable Choice, 32 Pages.
- [47] Fernández-Pan, I., Ziani, K., Pedroza-Islas, R., & Maté, J. I. (2010). Effect of Drying Conditions on the Mechanical and Barrier Properties of Films Based on Chitosan. *Drying Technology*, 28(12), 1350–1358.
- [48] Figge, K. (1980). Migration of Components from Plastics-packaging Materials into Packed Goods - Test Methods and Diffusion Models, *Progress in Polymer Science*, 6, 187-252.
- [49] Flores, S., Famá, L., Rojas, A. M., Goyanes, S., & Gerschenson, L. (2007). Physical properties of tapioca-starch edible films: Influence of filmmaking and potassium sorbate. *Food Research International*, 40(2), 257-265.
- [50] García NL, Ribba L, Dufresne A, Aranguren M, Goyanes S (2011) Effect of glycerol on the morphology of nanocomposites made from thermoplastic starch and starch nanocrystals. *Carbohydrate Polymers* 84: 203–210.

