Preparation and Characterization of Starch Aerogels Loaded with Senna alata (L.) Roxb Leaves Extract

NADIA FADHILAH BINTI AZIZ
J20A0531

A Thesis submitted in fulfilment of the requirements for the Degree of Bachelor of Applied Science (Materials Technology) With Honours

FACULTY OF BIOENGINEERING AND TECHNOLOGY
UNIVERSITY MALAYSIA KELANTAN

2024

DECLARATION

I hereby declare that this thesis entitled "Preparation and Characterization of Pectin Aerogels Loaded with *Senna Alata (L.) Roxb*. Leaves Extract" is the result of my research except as cited in references.

Signature :

Student's Name : NADIA FADHILAH BINTI AZIZ

Date : 11 JANUARY 2024

Verified by:

Signature :

Supervisor's Name : DR. ASANAH BINTI RADHI

Stamp :

Date :

ACKNOWLEDGEMENT

I want to thank everyone who has helped me finish this study project. Thank you so much to my supervisor, Dr. Asanah binti Radhi, for all the help, comments, and support that she has given to me along the way. Her huge experience and knowledge have motivated me and helped me grow as a researcher. Also, I want to thank all the lab workers and support staff who helped me with my work in the lab. Their desire to give their time and knowledge has been very important. I would also like to acknowledge my friends and coursemates for making this experience more enjoyable. Our discussions and exchanges of ideas have motivated me along the way. Finally, my deepest thanks go to my family - my parents, Aziz bin Abd Rahman and Ramlah binti Ibrahim, and my siblings, for their endless love, prayers, and support that have sustained me thus far. This achievement would not have been possible without them.

PENYEDIAAN DAN PENCIRIAN AEROGELS KANJI YANG DIISI DENGAN EKSTRAK DAUN *SENNA ALATA (L.)* *Roxb*.

ABSTRAK

Daripada kajian ini, pendekatan yang lebih baik untuk pengeluaran dan pencirian aerogel kanji yang diselitkan dengan ekstrak daripada daun Senna alata (L.) Roxb. Aerogel kanji ubi kayu telah disintesis pada kepekatan 10 wt% dan 15 wt%. Daun Senna alata tertakluk kepada pengekstrakan Soxhlet menggunakan etanol sebagai pelarut. Ekstrak tumbuhan kemudiannya disepadukan ke dalam aerogel kanji dengan kandungan kanji 15% berat pada kepekatan 1% dan 3%. Sifat fizikal, kimia, biologi dan terma aerogel kanji yang dimuatkan telah dinilai menggunakan mikroskop elektron pengimbasan (SEM), spektroskopi inframerah transformasi Fourier (FTIR), ujian resapan cakera antimikrob dan analisis termogravimetrik (TGA). Penyiasatan SEM menunjukkan kehadiran struktur berliang di dalam matriks aerogel kanji. Analisis FTIR mengesahkan kewujudan kanji dan ekstrak dalam komposit aerogel. Aerogel, yang dipenuhi dengan bahan, mempamerkan sifat antibakteria terhadap bakteria E. coli. Data analisis termogravimetrik (TGA) menunjukkan bahawa aerogel kekal stabil dari segi haba sehingga suhu 250°C. Hasilnya menunjukkan kejayaan sintesis aerogel kanji dengan menggunakan ekstrak Senna alata. Aerogel ini berpotensi untuk digunakan sebagai pad penyerap dengan sifat antimikrob dalam pembungkusan makanan aktif. Ujian tambahan dinasihatkan untuk memeriksa kadar di mana ekstrak dikeluarkan, untuk mencapai penghantaran terkawal dalam sistem makanan.

Kata Kunci: Kanji aerogel, *Senna Alata (L.) roxb*, antimikrobial, TGA, FTIR

PREPARATION AND CHARACTERIZATION OF STARCH AEROGELS LOADED WITH *SENNA ALATA (L.) Roxb.* LEAVES EXTRACT

ABSTRACT

From this study, an improved approach for production and characterization of starch aerogels infused with extracts from Senna alata (L.) Roxb leaves. Tapioca starch aerogels were synthesised at concentrations of 10 wt% and 15 wt%. The leaves of Senna alata were subjected to Soxhlet extraction using ethanol as the solvent. The plant extracts were subsequently integrated into starch aerogels with a starch content of 15 wt% at concentrations of 1% and 3%. The physical, chemical, biological, and thermal properties of the loaded starch aerogels were assessed using scanning electron microscopy (SEM), Fourier transform infrared (FTIR) spectroscopy, antimicrobial disc diffusion tests, and thermogravimetric analysis (TGA). The SEM investigation indicated the presence of porous structures inside the starch aerogel matrices. The FTIR analysis verified the existence of starch and extracts in the aerogel composites. The aerogels, which were filled with substances, exhibited antibacterial properties against E. coli bacteria. The thermogravimetric analysis (TGA) data demonstrated that the aerogels remained thermally stable up to a temperature of 250°C. The results demonstrate the successful synthesis of starch aerogels by using Senna alata extracts. These aerogels have the potential to be used as absorbent pads with antimicrobial properties in active food packaging. Additional testing is advised to examine the rate at which the extracts are released, in order to achieve controlled delivery in food systems.

Keywords: Starch aerogel, *Senna Alata (L.) roxb*, antimicrobial, TGA, FTIR

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

WT%	**percentage by weight**
°C	**degree Celsius**
ML	**millimetre**
RPM	**Revolutions per minute**
FTIR	**Fourier transform infrared spectroscopy.**
TGA	**thermogravimetric**

LIST OF SYMBOLS

%	**PERCENTAGE**
°	**DEGREE**
CM	**CENTIMETER**
M	**METER**
ML	**MILLIMETER**

CHAPTER 1

INTRODUCTION

1.1 BACKGROUND OF STUDY

Biodegradable products are now being used in many businesses around the world, including the food industry. The food business tries to cut down on waste and use old materials to make new things, like food packaging. aerogels made from polysaccharides that can be used to absorb a lot of water in food packing. It's hard to understand carbohydrates because they are made up of many long strings of sugar molecules. There are many kinds of plants that have them, and they do many things, like hold up the structure. Aerogels are made of materials that are very light, have a lot of surface area, and are porous (Zheng et al., 2020). This part is called a "aerogel" because it is made up of a gel structure and a filling medium (air). Kistler said in 1931 that solid materials with a lot of holes and a lot of surface area can be made by putting air into wet gel instead of liquid and drying them in a more advanced way. It's hard to describe aerogels because they have a low density, a large surface area, and a structure full of holes. They are often used in a lot of different scenarios because of these traits. Since aerogels have these qualities, they can be used to absorb wetness and add bioactive compounds.

Aerogels can be added or directly or indirectly to food packaging which can interact with food and can extend the shelf life of food packaging. The study shows that starch aerogels can be used as absorbent food packaging (Lebedev et al., 2021). Aerogels is a very useful material in the food industry because of their ability that can absorb moisture. This property can help to prevent the spoilage and extend shelf life of food products. This can provide health benefits to the consumers. The primary of absorbent pack of food packaging is to contain the products and protect them from external factors such as contamination, moisture and air. Nowadays, efforts are being made to find alternatives and ecological substitutes for currently known polymers and food packaging, and as a solution starch may be used. Starch is common used in bioaerogel that has been used in many applications (Rangaraj et al., 2021).

Starch, a food-grade biodegradable gelling agent of low cost, could form an integrated gel network structure in the absence of cross-linkers (Mikkonen et al., 2013; Ubeyitogullari & Ciftci, 2017). Chemically, starch is a homopolysaccharide made up of glucose. Its divided into two types which is amylose and amylopectin. There are differences in the structures and also properties which may have an effect on how starch-based aerogels made from them behave. Starch-based aerogels are made from natural starch sources and can have low density and low thermal conductivity. The variety of starch raw materials used to make these aerogels has expanded to include corn, potato, tapioca, pea, and wheat, and different drying methods have been adopted to obtain high-performance starch-based aerogels with low density, high specific surface area, and high porosity. To produce food packaging containing antioxidant, absorbent pads do not need to have a crosslink with extraction, but it needs to be incorporated with active compound that have antioxidant and antimicrobial properties (Manzocco et al., 2021). Alternatively, plant extract that rich with antioxidant can incorporated with starch aerogel which means absorbent pad contain of bioactive. To be absorbent pad in starch aerogel of food packaging, starch aerogel should consist active compound that can be protect the food from damage which its have antioxidant. This antimicrobial agent is available consisting of synthetic polymer but also available from natural resources such as from plant extract. Study say that among the plant extracts that have a beneficial antioxidant to be used in preparing absorbent pads are among them are the Senna alata (L.) Roxb tree (Laura Martins Fonseca et al., 2021).

The Senna Alata plant Roxb, which is also called "cassia alata" or "ringworm bush," is a tropical plant that has been used for medicine for a long time. There are antioxidants in *Senna alata (L.) Roxb* products, which may be one of their health benefits. Some secondary chemicals in the plant have been shown to have antifungal, antitumor, antioxidant, antibacterial, anti-inflammatory, analgesic, and immune-stimulating effects. These effects have been seen in extracts from the plant's leaves, stems, flowers, root bark, and seeds. Based on this study, the antioxidant-rich *Senna alata (L.) Roxb* extract could be a good choice for use as an active ingredient in food packaging absorbent pads.

In this study, starch aerogel can be loaded with *Senna alata (L.) Roxb* leaves extract that rich with antioxidant and antimicrobial. So the properties will be prepare to see the effect of *Senna alata (L.) Roxb* incorporation into the starch aerogel will be evaluate based on their physical, chemical, thermal and biological properties.

1.2 PROBLEM STATEMENT

Due to the widespread use of non-biodegradable polymers which have been around for hundreds of years in the production of food packaging, this practise has the potential to pollute the environment. The incorporation of *Senna alata (L.) Roxb* extract into starch aerogels for food packaging enhances their antioxidant and antibacterial characteristics, hence promoting bioactive effects.

1.3 OBJECTIVES

In this research, there two objectives which need to be achieved. The objectives are:

1. To prepare starch aerogel loaded with *Senna Alata (L.) Roxb* extraction.
2. To study the effect of *Senna Alata (L.) Roxb* extraction on the physical, chemical, biological, and thermal properties of aerogels.

1.4 SCOPE OF STUDY

The scope of the study involves the preparation of starch aerogel using tapioca starch and extraction of plant materials from *Senna Alata (L.) Roxb* leaves using Soxhlet extraction. The starch aerogel was combined with varying concentrations (1% and 3%) of the Senna alata plant extract to produce an environmentally sustainable product with antimicrobial advantages. The physical, chemical, thermal, and biological properties of the starch aerogels were studied after different amounts of Senna alata extracts were added. The aim was to assess the the effect of *Senna Alata (L.) Roxb* leaf extracts and findings would determine the feasibility of using such aerogel composites as absorbent pads with active properties for food packaging applications.

1.5 SIGNIFICANCE OF STUDY

The significance of study was to see whether starch can be good as aerogels since it is a natural polymer. Starch is a well-known as polysaccharide that has been utilised in aerogels for many years. Studies have demonstrated that aerogels made from starch have high biocompatibility, biodegradability and also non-toxic. The mechanical characteristics and biocompability of this aerogels were also discovered to be quite good for self-healing, viscocity and toughness. It has potential to be food packaging to be much safer and good for environment since its natural and also affordable. Therefore, starch and *Senna Alata (L.) Roxb* extraction is the most suitable alternatives to be more efficient antioxidant in food packaging.

CHAPTER 2

LITERATURE REVIEW

2.1 STARCH

A lot of things, like bread, pasta, potatoes, and rice, contain starch. Starch is safe to eat because it is usually recognised as safe (GRAS), doesn't cause allergies, and isn't toxic. Starch is also easy to find and doesn't cost much, which makes it a good material for many uses. It is the main thing that plants store in polysaccharides (Hoseney, 1994). Starch comes from plants and can be made again and again. It is soluble and made up of long chains of glucose molecules that are joined together chemically. Polysaccharides are long chains of carbohydrates made up of many simple sugars connected by glycosidic bonds. Natural, renewable sources of starch are separated out and mostly used as building blocks for active food packaging. Most polysaccharide products are hydrophilic, which means they don't keep water vapour out (Maria José Fabra, Talens, & Chiralt, 2008).

From chemically sides, starch consists of semi-crystalline granules composed of amylose and amylopectin components. The shape, size, structure and chemical composition of the starch mainly depends on its source from which is it derived (Cazón et al., 2017). For the normal starches, amylose makes up around 20-30% of the starch while amylopectin makes up remaining 70-80% (Svihus et al. 2005). Amylose is primaly a linear molecular that made up of glucose units that linked together by alpha- 1,4 glycosidic bonds. It has a helical structure and is responsible for the formation of a semi-crystalline region in the starch granule while amylopectinis a highly branched molecule structure and is responsible for the amorphous region in the starch granule and its made up of glucose units linked together by alpha-1,4 glycosidic bonds and alpha-1,6 glycosidic bonds.. The variation of amylose content in starch source influencers the granule size distribution and the molecular characteristics of amylose and amylopection and the functional properties.

Amylose is linear starch molecule that consists of 1-4 linked a α D glucopyranosyl units and had a low molecular weight (1.03-4.89 x 105) and it behaves as a non-branched molecule while amylopectin is highly branched starch molecule of α D glucopyranosyl units primarily linked by 1-4 bonds with branches resulting from 1-6 linkages. Its has a higher molecular weight (7.08-9.88 x 107) and is heavily branched. As a result, the structures and characteristics of amylose and amylopectin differ which may have an impact on the characteristics of starch-based aerogels that made from them (García-Guzmán et al., 2022).

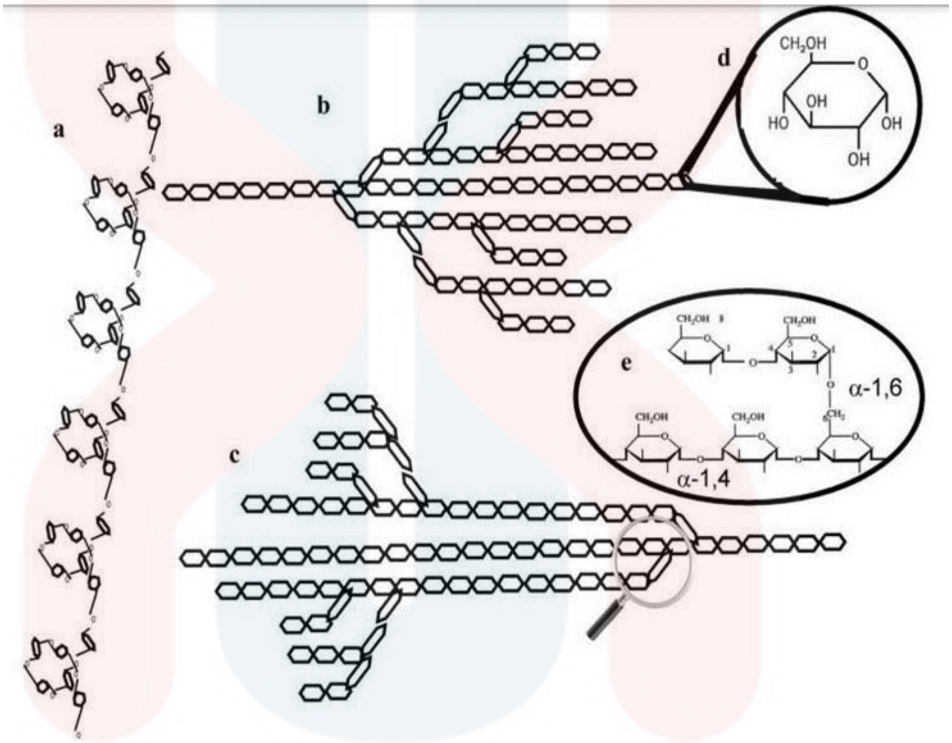


Figure 2.1: *Schematic representation of a the structure of amylose and amylopectin* (The Structural Characteristics of Starches and Their Functional Properties, 2018)

Figure 2.1 shows a type of glucan that can be found in starch. Large (A), long (B1), mid-length (B2), short (B3), and B4 chains are the different categories for these chains. Depending on where the starch came from, these chains' lengths can change. The length or degree of polymerization of the glucan chains can differ due to genetic variations, environmental and nutritional conditions during plant growth and seed formation, and the activity of enzymes involved in the synthesis of starch granules.

The branches of amylopectin link every 20–25 glucose linear units. Also discussed is the anomeric carbon of glucose molecules, which can function in metabolic processes as either a proton receptor or an electron donor. Through o-glycoside links, which are alpha-1,4 and alpha-1,6 links, the reducing end of glucose is useful for the biosynthesis of amylose and amylopectin. They both have a reducing end. Figure 2, also mentioned in the abstract, may provide a visual representation of the structure of amylose and amylopectin.

For starch granules, amylose typically provides amorphous lamealle, whereas amylopectin develops ordered crystalline lamealle. As a result, the structures and characteristics of amylose and amylopectin differ, which may have an impact on the characteristics of starch-based aerogels made from them.

2.2 STARCH AEROGEL

Among natural polysaccharides, starch has become popular choice for making aerogels. Aerogels such as advanced biomaterials that have low density and also high specific surface area. Starch-based aerogels are nanoporous, meaning they have tiny pores at the nanoscale level, and have many potential applications in different areas. Starch-based aerogels are typically made using a combination of processes such as starch gelatinization, retrogradation, organic solvent exchange, and supercritical CO_2 drying. These processes help to create the nanoporous structure of the aerogel. Its have been used for various applications such as encapsulation and controlled release of bioactive compounds, biomedical uses and tissue engineering, as packaging and thermal materials, and as templates for the synthesis of novel functional materials.

Because of their special characteristics, starch-based aerogels have a variety of potentialuses. They can be used for a variety of purposes, including food packaging, drug delivery, oil spill cleanup, and thermal insulation in buildings. They are also environmentally friendly and biodegradable. They also have potential in fields like aerospace, electronics, and automotive, which are looking for lightweight materials with superior insulation qualities. Starch aerogels are versatile and a promising material for many applications because they can be further altered by adding other components or by applying chemicalprocesses to improve their properties.

2.3 PROPERTIES OF STARCH AEROGELS

2.3.1 PHYSICAL PROPERTIES

Aerogels are typically characterized by low density solid, low optical index of refraction, low thermal conductivity, low speed of sound through materials, high surface area, and low dielectric constant. For physical properties of starch-based aerogels, there are a porous structure that highly porous materials. It's with three-dimensional network interconnected pores. The porosity of aerogels is typically in the range of 90-99% that gives extremely large surface area per unit volume.

2.3.2 CHEMICAL PROPERTIES

Aerogels are 99.8% porous, have low thermal conductivity, density, dielectric constant, and refractive index. Aerogels' startling properties enable many technological uses. Pectin was selected for the synthesis of pectin aerogels via dissolution-solvent-exchange-drying with supercritical carbon dioxide due to its tolerance to a broad spectrum of conditions that affect solution viscosity and gelation. Thus, because of the chemical properties of the pectin aerogels changes in external parameters (pH, polymer content, non-solvent type, and concentration of monovalent and polyvalent metal ion compounds) influenced the aerogel's shape, porosity, pore size, surface area, and mechanical properties (Groult & Budtova, 2018).

2.3.3 BIOLOGICAL PROPERTIES

As well as being biocompatible, porous, water-absorbent, and biodegradable, starch aerogels that incorporated with *Senna Alata (L.) Roxb* also have properties that help them gel. These qualities make them desirable materials for use in agricultural, environmental, and industrial fields as absorbent pads for food packaging. A promising new material with many potential uses in food packaging is starch aerogels. Because they are biocompatible, biodegradable, and non-toxic, starch aerogels are a good option for absorbent pads in food packaging due to their biological characteristics. Starch aerogels also have a number of other qualities, such as antioxidant and antimicrobial that make them perfect for absorbent pads in food packaging.

2.3.4 THERMAL PROPERTIES

Such as major application of starch-based aerogels was a thermal property. The study found that the thermal conductivity of starch aerogels and foams was similar to that of commercial insulation samples. This means that starch aerogels and foams can beused as insulation materials to prevent heat transfer. The thermal conductivity of starch aerogels was affected by the starch type and processing conditions (Druel et al., 2017). For example, among the aerogels made from waxy and regular potato, high amylose maize, and pea starches, the pea starch aerogels had the lowest thermal conductivity, which was even lower than that of air. The low thermal conductivity of starch aerogels is attributed to two factors. Firstly, the pore size of starch aerogels is very small, less thanthe mean free path of air molecules. This means that air molecules have a difficult time moving through the small pores, which reduces heat transfer. Secondly, the density of starch aerogels is low, which also reduces heat transfer. The smaller the pore size, the more difficult it is for heat to pass through the material. Therefore, optimizing the type of starch and processing methods used can help to further improve the thermal conductivity of starch-based aerogels.

In another aspect of thermal properties of starch aerogels is thermal stability. The studied from Ubeyitogullari and Ciftci (2016a) shows thermal stability refers to the ability of a material to resist changes in its physical and chemical properties when exposed to high temperatures. However, a rapid thermal degradation occurred at some point during the analysis, which means that the material started to break down and lose weight quickly. A rapid thermal degradation occurred at 280-330°C. However, the study also found that starch aerogels had a faster formation of ash and char compared to starch alone. This could be due to the more open structure of the aerogels, which allows for more efficient heat penetration. The open structure of the aerogels refers to their nanoporous nature, which allows for a high specific surface area.

This means that there is a large surface area available for reactions to occur, which can lead to faster formation of ash and char when exposed to high temperatures. This study prove that starch-based aerogels have potentials as a thermal property due to their thermal stability and efficient heat penetration

2.4 SENNA ALATA (l.) ROXB

Figure 2.2: Senna Alata (L.) Roxb leaves

Senna Alata (L.) Roxb able to grow up to 3 - 4 m tall. Pinnately compound leaves are composed of 6 - 12 pairs of leaflets (30 - 60 cm long). Oblong leaf lets are smooth and thinly leathery (6 - 15 cm long, 3.5 - 7.5 cm wide). Mostly it has been traditionally used in medicine for various purposes, including its antioxidant properties. But its also can be use as absorbent pad in food packaging as long as *Senna Alata (L.) Roxb* have extraction of antioxidant. Theres some about *Senna Alata (L.) Roxb* which is it can have antioxidant activity. The leaf *Senna Alata (L.) Roxb* contains of active chemical compound called anthraquinones, which exist in two forms: aglycone and glycoside. Anthraquinones are known for their antioxidant and antimicrobial properties and are the active ingredients in *Senna Alata (L.) Roxb* leaves that can make them useful for absorbent pad (Rahmawati et al., 2022). Absorbent pad of food packaging is typically designed to absorb excess moisture or gases released by food products to maintain freshness and prevent spoilage. They may contain materials such as activated carbon or other absorbent materials that derived from *Senna Alata (L.) Roxb* extraction.

2.5 BIOLOGICAL PROPERTIES OF SENNA ALATA

2.5.1 ANTIOXIDANTS ACTIVITY

The antioxidant are commonly used in absorbent food packaging because it can prevent or delay the oxidation of food products and also can extending the shelf life their quality of food. These antioxidants can be obtained from a variety of sources, including plant extracts, essential oils extracted from herbs and spices, and polyphenolic concentrates made from degraded biosources for example this antioxidant can be found in the leaves of the *Senna Alata (L.) Roxb* leaves. These antioxidants from *Senna Alata (L.) Roxb* are complex molecules that can release active compounds from the packaging to protect food from harmful substances such as free radicals, oxidative intermediates, and secondary breakdown products. Several studies had reported the biological activity of *Senna Alata (L.) Roxb* leaves extract has a robust antioxidant activity. It is because *Senna Alata (L.) Roxb* contains flavonoid compounds especially in the leaves. These compounds also possess antimicrobial and antifungal properties that can help preserve the quality of food (Brewer, 2011). Recent research has demonstrated that these antioxidants from natural sources such as *Senna Alata (L.) Roxb* can be incorporated into the absorbent pad of food packaging and released to the food at specific times. This aids in lowering lipid oxidation and preserving the food's nutritional value and quality. Antioxidants can prevent or slow down this process by neutralizing the free radicals that cause oxidation. Overall, with the presence of antioxidant in *Senna Alata (L.) Roxb* will be an alternative for absorbent pad of food packaging.

2.5.2 ANTIMICROBIAL ACTIVITY

Antimicrobial can be used as absorbent packs for food packaging to help inhibit the growth of microorganisms, such as bacteria, fungi, and mold, which can cause spoilage or foodborne illness. *Senna Alata (L.) Roxb* is believed to possess a variety of chemicals, including anthraquinones, flavonoids, and tannins, which all have antimicrobial properties. Anthraquinones are a class of substances with laxative properties as well as antimicrobial properties. A class of substances known as flavonoids also has antimicrobial activity in addition to its anti-inflammatory and antioxidant properties. The group of substances known as tannins also has antimicrobial activity in addition to their astringent and anti-inflammatory properties. Both gram-positive and gram-negative bacteria as well as fungi have been shown to be resistant to *Senna Alata (L.) Roxb* antimicrobial activity. *Senna Alata (L.) Roxb* has a promising property that could be used to create new antimicrobial agents: antimicrobial activity. *Senna Alata (L.) Roxb* is a plant that has been used for centuries in traditional medicine, and also can be used as absorbent pad in food packaging because it is safe and effective. It is a possible source of fresh antimicrobial substances that might be applied to the treatment of various infections. These antimicrobial agents can be incorporated into the absorbent materials used in the packaging to provide additional protection for the food products.

2.6 APPLICATION OF STARCH AEROGELS IN FOOD PACKAGING

Applications for starch-based aerogels in food packaging have been thoroughly investigated. The starch aerogels could be used as food packaging absorbers. During in vitro tests, quercetin-impregnated starch-based aerogels were used. A substance known as quercetin has antimicrobial and antioxidant properties. According to the tests, quercetin released from aerogels at a much slower rate than it did from pure quercetin. This implies that the quercetin- containing aerogels might be used as functional packaging (Fang, 2019). The aerogels' nanoporous structure is probably to blame for the slower rate of quercetin release. It's possible that the tiny pores in the aerogels will act as a barrier, slowing the diffusion of quercetin from the substance. Due to this characteristic, quercetin-containing aerogels can be used as active packaging materials. Active packaging components are made to communicate with the package's contents.

CHAPTER 3

MATERIALS AND METHOD

3.1 MATERIALS

The materials was used needed to produce starch aerogel is starch powder, distilled water, *Senna alata (L.) Roxb*. leaves extraction, Since this research is based by using starch powder with the extraction of *Senna alata (L.) Roxb* leaves, there will be 4 different amounts of concentration of extraction.

3.2 METHODS

3.2.1 PREPARATION OF SENNA ALATA EXTRACTION

Senna Alata (L.) Roxb leaves was collected Lata Keding, Jeli, Kelantan, Malaysia. *Senna Alata (L.) Roxb* leaves were then rinse with distilled water to remove any dirt or debris. The leaves was spread on a clean drying tray and dried in oven for 24 hours at temperature of 40-45°C for 24 hours as shown in Figure 3.1.

Figure 3.1 : *Senna Alata (L.) Roxb* Leaves were dried in oven for 24 hours

After the leaves already dry, the leaves were grinded into the powder completely and put the powder into the airtight containers. By using soxhlet extraction, the apparatus was set up and the sample material containing the fine powder of *Senna Alata (L.) Roxb* (30.0g) was loaded into the thimble. Then, the timble was placed into the main chamber and the ethanol was added (300 ml) of 80% in Soxhlet apparatus a round of bottom flask and place onto a heating mantle. Then, the soxhlet extractor was put on top of the round-bottom flask and the reflux condenser on top of the extractor. The cold water enters from the bottom and stays above for about 6 hours.

Figure 3.2 : Soxhlet Extraction of Senna Alata (L.) Roxb

After that, put the leaves extraction into the 250ml of reagent bottle. The extraction solvent, distilled water and ethanol at the concentration of 95% been used to remove the ethanol by using the rotary evaporator. 20g of *Senna Alata (L.) Roxb* powder been weigh in conical flask and mix with heat of room temperature which is 30°C. The mixture needed an hour to settle in a water reservoir that was heated to 40, 60, and 80°C. After that, Whatmann no. 1 filter paper been used to filter the extract. To obtain the crude extracts, residual solvent has evaporate at 45°C and 65 rpm in rotary evaporator. The extracts been stored in the freezer at 4°C for further use. The leftover of extraction been put in borosilicate glass with cap (Gritsanapan & Mangmeesri, 2009).

3.2.2 PREPARATION OF STARCH AEROGEL

Firstly, tapioca starch was weighed which 30g by using a scale and been put in a clean beaker. 170ml of distilled water was prepared and been mixed with the starch that has been weighed to produce 10 and 15 wt% concentration of starch solution. After that, the solution being stir until its become homogeneous (about 20 minutes) by using magnetic stirrer in the hot stir plate. Then, the solutions being heat up to 90°C for 45 minutes under stirring in thermostatic bath to gelatinize the starch. Solution of 1% and 3% of Senna Alata (L.) Roxb bring added into the homogenous starch then mix well with the glass rod. After that, the solutions was added into the silica moulding to being prefreeze and let it be cool down for 10 to 15 minutes. The sample being put in -80°C freezer and left for 24 hours for retrogradation process which is to reduce the possibility of moisture absorption (Hu et al., 2019). After the retrogradation is done, the drying step being carried out for 3 days. When the drying process was completed, starch aerogel was formed.

Sample	Starch Powder (wt %)	Concentration of Senna Alata (g)
S1	10 wt %	0 %
S2	15 wt %	0 %
S3	15 wt %	1 %
S4	15 wt %	3 %

Table 3.1 : Concentration of Senna Alata (L.) Roxb into the starch aerogels

3.2.3 CHARACTERIZATION OF AEROGELS

3.2.3.1 Scanning electron microscopy (SEM)

To examine the physical properties of starch aerogels, an analysis was conducted on their surface shape and composition attributes. Observable or quantifiable characteristics of a substance that do not alter its fundamental nature. The characterization can be performed using a Scanning Electron Microscope equipped with a JSM-IT200 InTouchScope SEM model. The SEM should be operated at an accelerating voltage of 3 kV, and the diaphragm sizes used should be 50, 100, and 250 µm. Starch aerogels can be used to see the surface of objects at high magnification. *Senna Alata (L.) Roxb* will envelop the starch core. This research reveals the porosity and shape of starch aerogels. The morphology of the aerogel, encompassing its dimensions, configuration, and surface texture, may be discerned from the SEM image.

3.2.3.2 Fourier transform infrared analysis (FTIR)

FTIR was used to study the different functional groups of extracted *Senna Alata (L.) Roxb* obtained the FTIR spectra. The FTIR spectra were acquired using a Nicolet iS50 model FTIR located at University Malaysia Kelantan (UMK) at the temperature of the surrounding air, with a resolution of 4 cm^{-1} and a wavenumber range of 4000 to 400 cm^{-1}. Each spectrum was obtained by averaging the results of four separate scans, and the speed of each scan was 0.2 cm^{-1} s^{-1} . It was determined by interpreting the spectrum to which functional group the extractives belonged.

3.2.3.3 TGA ANALYSIS

TGA determines the thermal stability of compounds at high temperatures. Thermogravimetric Analysis (TGA) requires controlled heating of a sample and observation of weight loss over time or temperature. The weight reduction after furnace heating and crucible insertion is measured with a balance. The material under study affects the temperature range of 25°C to 600°C and heating rates of 20 °C/min and gas flow of nitrogen atmosphere with a rate of 50 ml min^{-1}. Materials science use TGA to determine material composition, purity, and heat stability. Universiti Malaysia Kelantan used EW-25753-44 Mettler Toledo TGA2 Thermogravimetric Analyzer with Small Furnace, XP1 Balance, and TGA Sensor.

3.2.3.4 ANTIMICROBIAL TESTING

Antimicrobial activities of preparation being used by the disc diffusion method. To perform the antimicrobial process, the disk diffusion method has been used. The diameter of clear zone of inhibition around the samples after 24 hours of incubation has been measured. For this method, firstly cut the samples into circular shapes with a diameter of 10mm. Then, the samples being placed to the sterilized with ultraviolet light for 60 minutes. After E. coli grow on agar plates with lots of nutrients, *Senna Alata (L.) Roxb* will be added to starch aerogels that are clean. The dishes being put incubate for 37°C. After incubation process is done, the diameter of inhibition zone has been measured where the bacteria did not grow. They were able to determine through this measurement how the aerogels were preventing the bacterial growth (Ye et al., 2018).

RESEARCH FLOWCHART

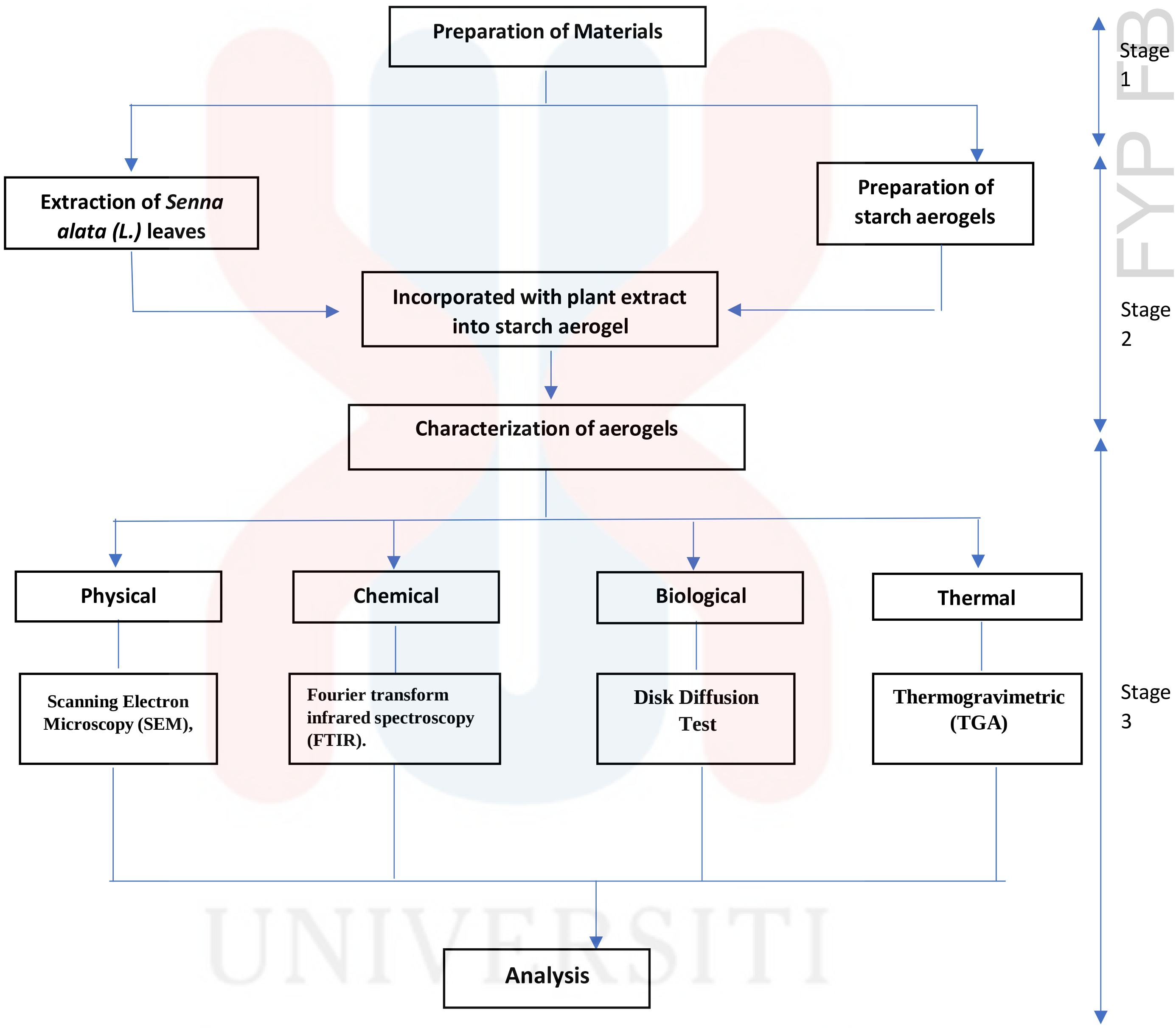


Figure 4: Flowchart of Starch Aerogels.

CHAPTER 4

RESULTS & DISCUSSIONS

4.1 MORPHOLOGY

The examination of aerogels microstructure unveiled a uniform three-dimensional porous arrangement distinguished by overlapping layers that are associated by *Senna alata (L.) Roxb*. This entails analyzing the external appearance and arrangement of the material at a microscopic or nanoscopic level. This technique yields the overall microstructure ofthe composite material.

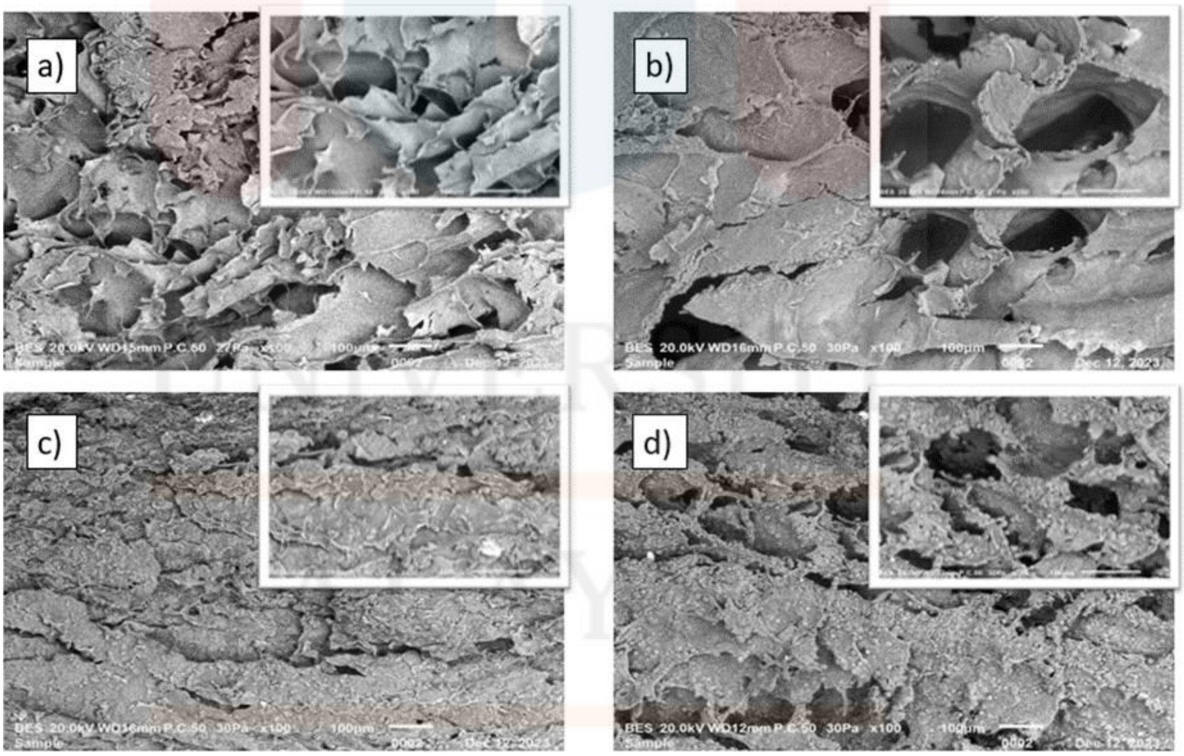


Figure 4.1 : SEM images of Starch Arogels 10 wt% (a), SA 15 wt% (b), SA 15 wt% 1%, (c) and SA 15 wt% 3% (d) at 100x and 250x magnificent (100x and 250x).

An analysis was conducted on the structural morphology of a material containing 10 and 15 wt % of starch. This analysis involved the use of both high and low magnification. Upon close examination at a low level of magnification which is 100x (Figure 5 (a) and (b)), it is evident that starch aerogels with a concentration of 10 wt % have a rough structure. The close-up view of starch aerogels clearly reveals a rough surface structure with irregular shape of porous that appeared on the sample of starch aerogels.

Both types of aerogel samples in (Figures (c) and (d)) exhibit a resemblance, namely the presence of many spots on their surfaces. This could be attributed to the regenerative properties of *Senna alata (L.) Roxb*. The surface of aerogels also appears rougher for both sample. For (Figure (c)), porous less visible while (Figure (d) shows pores that look bigger and clearer.

4.2 FOURIER TRANSFORM INFRARED SPECTROSCOPY (FTIR)

Chemical characterization and characteristics of all samples of starch aerogels involving pure and extraction of *Senna alata (L.) Roxb* 10 wt % and 15 wt %. Hence, FTIR showed in Figure illustrated that all samples mostly samples are not significantly different from each other in functional group. The absorption bond at ~1005, ~2929 and ~3313 cm^{-1} in spectrum of aerogels incorporated with *Senna alata (L.) Roxb* with different wt%.

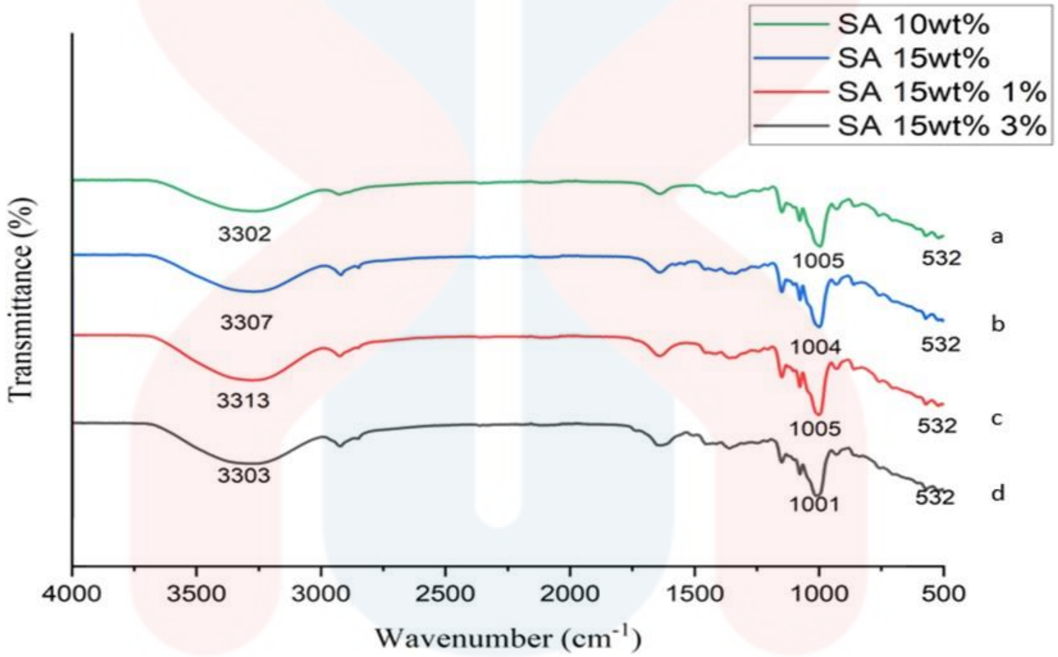


Figure 4.2. FTIR Spectra of Starch Aerogels 10 wt% (a), SA 15 wt% (b), SA 15 wt% 1% (c) and SA 15 wt% 3% (d).

For the curve (a) which is at the peak 1005 cm^{-1}, linked to the C-O stretching vibration in glycosidic bonds between glucose units, which shows that starch is present. Aliphatic phosphates, on the other hand, have unique peaks in a different part of the spectrum. These peaks are caused by phosphorus-oxygen (P-O) links. Frequencies stretching is wavenumbers between 1200 cm^{-1} to 1000 cm^{-1}. Meanwhile for the peak 3302 cm^{-1}, Its involves the stretching vibration of the hydroxyl group (O-H). The peak which are found in many substances, like alcohols. This is a broad and strong band that shows the O-H stretching vibration for primary aliphatic alcohols. It happens between 3650 cm^{-1} and 3200 cm^{-1}. The O-H stretching peak can be in different places and have different shapes based on the alcohol and its surroundings.

Another curve which (b) for the peak 1004 cm^{-1}, its showed corresponding to the lengthening vibration of C-O bonds in glycosidic chains. Glycosydic linkages are formed when the hydroxyl group (-OH) of one glucose molecule forms a C-O-C bond with the anomeric carbon of another glucose molecule. This peak also showed the existence of overall structure of the starch polymer. Meanwhile for the peak at 3307 cm^{-1} represents O-H (hydroxyl) stretching vibration. This is because the carbon atoms of a polysaccharide composed of glucose units are attached to hydroxyl groups (-OH). The vibration peak associated with O-H stretching is highly responsive to hydrogen bonding interactions. There might be intermolecular hydrogen bonds between hydroxyl groups in the aerogel structure.

For the third curve which is (c), the peak at 1005 cm^{-1}, usually linked to the vibrational stretching of the carbon-oxygen bond, more especially the stretching of the C-O bond in the glycosidic bonds between glucose units. The glycosidic linkages that connect these units are vital. This peak is precipitated by the C-O stretching vibrations present in the glycosidic linkages. While for the peak 3313 cm^{-1} showed that equivalent to the stretching vibration of hydroxyl groups O-H bonds. Starch, being a polysaccharide consisting of glucose units, is endowed with carbon atoms that are affixed to hydroxyl groups (-OH).

Last curve in the figure (d) for the peak observed at 1001 cm^{-1} is commonly ascribed to the stretching vibration of the C-O-C bonds in the ether moieties that are present in starch., the lengthening vibration revealed the C-O-C bond. Additionally, the existence of intermolecular hydrogen bonds can be deduced from the interpretation of the C-O-C stretching peaks are usually intense and between wavenumbers 1300 and 1000 cm^{-1}. Hydrogen bonding can establish connections between distinct segments of the polymer chain in starch, thereby making a significant contribution to the molecule's overall stability and structure. Meanwhile, the peak at their 3303 cm^{-1} connected to the stretching sound of O-H. This peak shows that there are hydroxyl (OH) groups in the starch molecule. The stretching waves of O-H bonds can be seen between 3600 cm^{-1} and 3200 cm^{-1}. There are hydroxyl groups in the starch molecule, as shown by its specific frequency at 3303 cm^{-1}

4.3 ANALYSIS (TGA)

Thermogravimetric (TGA) analysis of starch aerogels incorporated with Senna alata (L.) Roxb provide the information of weight loss of a sample as its heated in an inert atmosphere. From this study, TGA was used to analyzed the weight loss of starch aerogels 10 wt % and 15 wt % of pure starch and 15 wt % with concentration of 1% and 3% of Senna alata (L.) Roxb as the temperature increased. The weight loss curves of all samples were plotted in figure 4.4, showing the change in weight as function of temperature.

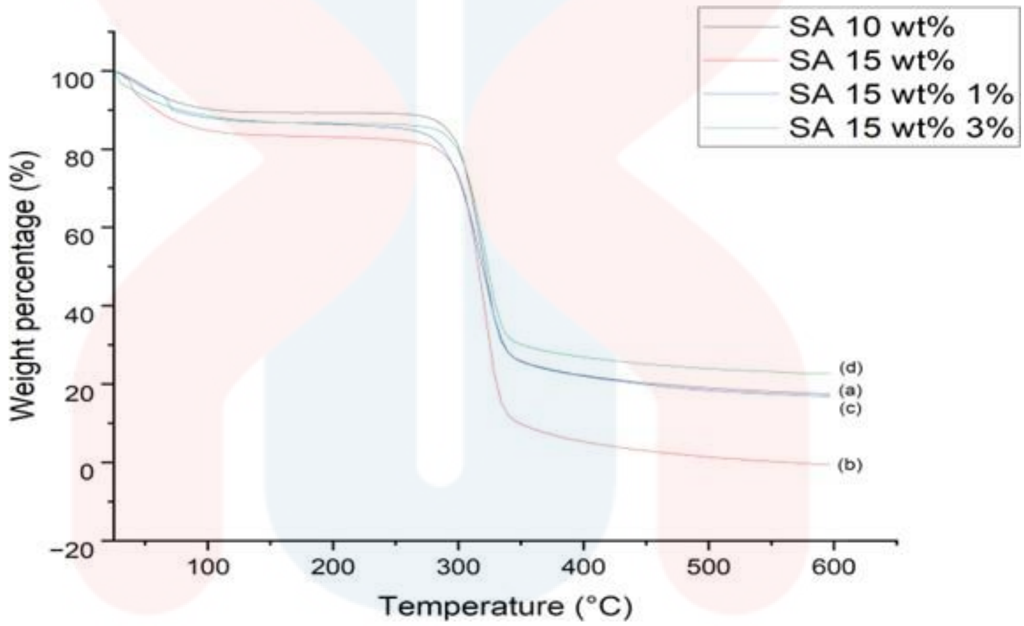


Figure 4.3. : TGA Analysis for Starch Aerogels 10 wt% (a), SA 15 wt% (b), SA 15 wt% 1% (c) and SA 15 wt% 3% (d).

At the temperature below of 100°C, all of the samples had an initial small amount of weight loss that indicates to the evaporation of water loosely bound to the surfaces of starch aerogels. At all the curves of the samples, a single remarkable decomposition associated with progressive weight loss is observed.

The decomposition of pure starch 10 wt% inside a bigger range of weight percentages occurred at from the temperature of 280°C and 310°C. While for pure starch of 15 wt% were found to degrade at a temperature lower than others. This results shows that the pure starch that contain 15 wt% is more susceptible to the degradation when it exposed to the heat. This degradation occurred in the temperature range of 250°C to 300°C. The heat breakdown of pure starch slowed down after it was subjected to acid hydrolysis, which breaks down the starch molecules into their individual parts.

While for starch 15 wt% 1% start to decompose after 250°C. This occurred when pyrolysis process happened in which big molecules break down into smaller pieces when oxygen is not present. This process may be used to break down starch. Because of this, char and volatile organic substances may form. For the decomposition of starch 15 wt% 3% took place at the temperature 250°C to 370°C and also been illustrated one step pyrolysis process. This various complex process including thermal degradation, pyrolysis and the release of volatile products occurred. It may only take a small amount of energy to break these glycosidic bonds, which breaks down the starch molecule into smaller pieces. Thermal decomposition and pyrolysis are both parts of this process. Large starch molecules break down into smaller substances, releasing volatile products.

4.4 ANTIMICROBIAL ANALYSIS

Disc diffusion was utilised to apply starch aerogels with antibacterial properties. The efficacy of the antibacterial method can be assessed by quantifying the diameter of the conspicuous zone of inhibition encircling the sample following a 24-hour exposure to ambient temperature.

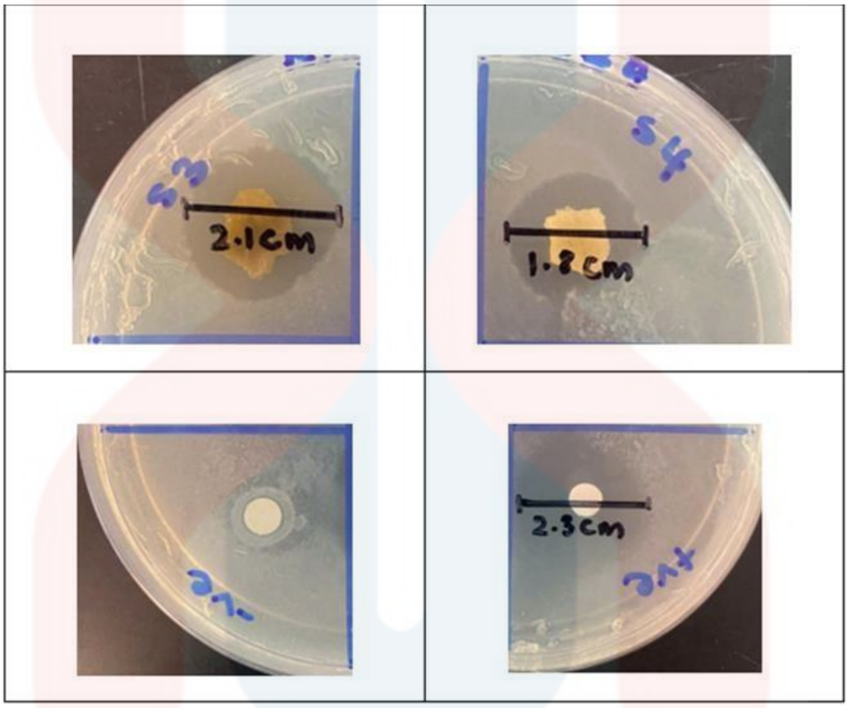


Figure 4.4. : Optical Images of Clear Zones of Starch Aerogelswith E.coli (S3) 15 wt% 1%, (S4) SA 15 wt% 3%

For the image of sample (S3) and (S4) showed that the clear zone of each sample from SA 15 wt%. The diameter of clear zone for (S3) is 2.1 cm while for (S4) is 1.8cm. For the negative (-) zones that contained distilled water while for positive (+) zones showed there are clear zone around 2.3 cm that contained the antibacterial solution which is *Chlorophenicol*. From this antimicrobial analysis, we can see the starch aerogels that contained with extraction of *Senna alata (L.) Roxb* going well worked against *E. coli*. From this study showed that aerogels contained with concentrations of the extracts created areas of inhibition, which means they had stronger antimicrobial qualities

CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

In Summary, during this research, starch aerogels loaded with extracts from *Senna alata (L.) Roxb* leaves were successfully synthesised and characterised. Both 10 and 15 wt% concentrations of tapioca starch were used in the production of aerogels. In order to obtain antioxidants from the leaves of the Senna alata plant, the Soxhlet extraction method was utilised. After that, the extracts were mixed into the aerogels that contained 15 wt% starch at loadings of 1% and 3%. The investigation using scanning electron microscopy revealed the presence of porosity networks inside the starch aerogel matrices. The FTIR analysis confirmed the existence of starch and plant extracts in the aerogel composites, as shown by the distinctive peaks. The aerogels that were loaded in antimicrobial testing exhibited inhibitory efficacy against E. coli. The TGA measurements demonstrated that the aerogels remained thermally stable up to a temperature of around 250°C.

The results suggest that starch aerogels infused with *Senna alata (L.) Roxb* extracts have been successfully prepared. These aerogels show promise for use as absorbent and antimicrobial pads in the field of active food packaging. The aerogels exhibited appropriate morphological, thermal, and antibacterial characteristics. It is advisable to conduct additional studies on the release kinetics of these antimicrobial starch aerogels in order to evaluate their potential for controlled delivery in food systems.

For recommendations, to further this study work, Examine the rate at which antioxidant and antibacterial components are released from the *Senna alata (L.) Roxb* extracts when they are incorporated into the starch aerogel matrices. This can aid in determining the release profiles and appropriate applications for controlled delivery.

Also, its can evaluate the efficacy of the starch aerogel absorbent pads in actual food systems by observing alterations in quality and the prolongation of shelf life over a period of time. This can verify the efficacy of the antibacterial and antioxidant properties when used in food packaging. For the biodegradability, its can created aerogels to verify their appropriateness for environmental applications and sustainability objectives.

REFERENCES

Chien, S. (2015). Synthesis and Characterization of Starch Aerogel - UNIMAS Institutional Repository. *Unimas.my*. http://ir.unimas.my/id/eprint/11051/1/Siaw%20Chien%2824pgs%29.pdf

De Marco, I., Baldino, L., Cardea, S., & Reverchon, E. (2015). Supercritical Gel Drying for the Production of Starch Aerogels for Delivery Systems. *CHEMICAL ENGINEERING TRANSACTIONS*, *43*. https://doi.org/10.3303/CET1543052

Ebert, H.-P. (2011). *Thermal Properties of Aerogels*. 537–564. https://doi.org/10.1007/978- 1-4419-7589-8_23

El-Naggar, M. E., Othman, S. I., Allam, A. A., & Morsy, O. M. (2020). *Synthesis, drying process and medical application of polysaccharide-based aerogels*. *145*, 1115–1128. https://doi.org/10.1016/j.ijbiomac.2019.10.037

Fang, Y. (2019). *Starch based aerogels: Production, properties and applications*. *89*, 1–10. https://doi.org/10.1016/j.tifs.2019.05.001

García-González, C. A., Uy, J. J., Alnaief, M., & Smirnova, I. (2012). *Preparation of tailor- made starch-based aerogel microspheres by the emulsion-gelation method*. *88*(4), 1378–1386. https://doi.org/10.1016/j.carbpol.2012.02.023

Hu, L., He, R., Lu, Z., Zhang, K., & Bai, X. (2019). *Step-freeze-drying method for carbon aerogels: a study of the effects on microstructure and mechanical property*. *9*(18), 9931–9936. https://doi.org/10.1039/c9ra01328h

Jane, J.-L., Maningat, C. C., & Rungtiwa Wongsagonsup. (2010, September). *Starch characterization, variety and application*. ResearchGate; unknown. https://www.researchgate.net/publication/286135326_Starch_characterization_variety_ and_application

Joaquín Gómez-Estaca, López-de-Dicastillo, C., Hernández-Muñoz, P., Ramón Catalá, & Gavara, R. (2014). *Advances in antioxidant active food packaging*. *35*(1), 42–51. https://doi.org/10.1016/j.tifs.2013.10.008

L. García-Guzmán, Cabrera-Barjas, G., Soria-Hernández, C. G., Castaño, J., A.Y. Guadarrama-Lezama, & Saddys Rodríguez-Llamazares. (2022). *Progress in Starch- Based Materials for Food Packaging Applications*. *3*(1), 136–177. https://doi.org/10.3390/polysaccharides3010007

Lebedev, A. Y., Ekaterina Suslova, Troyankin, A., & Lovskaya, D. (2021). *Investigation of Aerogel Production Processes: Solvent Exchange under High Pressure Combined with Supercritical Drying in One Apparatus*. *7*(1), 4–4. https://doi.org/10.3390/gels7010004

Luo, Q., Huang, X., Gao, F., Li, D., & Wu, M. (2019). *Preparation and Characterization of High Amylose Corn Starch–Microcrystalline Cellulose Aerogel with High*

Absorption. *12*(9), 1420–1420. https://doi.org/10.3390/ma12091420

Manzocco, L., Mikkonen, K. S., & García-González, C. A. (2021). *Aerogels as porous structures for food applications: Smart ingredients and novel packaging materials*. *28*, 100188–100188. https://doi.org/10.1016/j.foostr.2021.100188

Mirelle Dogenski, Navarro-Díaz, H. J., J. de Oliveira, & Roberta, S. (2020). *Properties of starch-based aerogels incorporated with agar or microcrystalline cellulose*. *108*, 106033–106033. https://doi.org/10.1016/j.foodhyd.2020.106033

Prabha Lahare, Rajeshwari , et al. "TLC BASED PHYTOCHEMICAL ANALYSIS and ANTIOXIDANT ACTIVITY of SENNA ALATA." *INTERNATIONAL JOURNAL of ADVANCED RESEARCH (IJAR)*, vol. 8(11),1099-1107, no. 2320-5407, Nov. 2020, https://doi.org/10.21474/IJAR01/12106.

Rahmawati, F., Prihantini, Nur Nunu, & Hady, B. C. (2022). In Vitro Bioactivity Test of Senna Alata (L.) Roxb Leaves Extract - Repositori Universitas Kristen Indonesia. *Uki.ac.id*. http://repository.uki.ac.id/6732/1/InVitroBioactivityTest.pdf Tavares, F., Jean, Laura Martins Fonseca, Graziella Pinheiro Bruni, Martins, F. T., & Renato, A. (2020). *Physically cross-linked aerogels based on germinated and non- germinated wheat starch and PEO for application as water absorbers for food packaging*. *155*, 6–13. https://doi.org/10.1016/j.ijbiomac.2020.03.123

The structural characteristics of starches and their functional properties. (2018). CyTA - Journal of Food. https://www.tandfonline.com/doi/full/10.1080/19476337.2018.1518343

Víctor Santos-Rosales, Inés Ardao, Alvarez-Lorenzo, C., Ribeiro, N., Carolina, A., & García-González, C. A. (2019). *Sterile and Dual-*

Porous Aerogels Scaffolds Obtained through a Multistep Supercritical CO2-Based Approach. *24*(5), 871–871. https://doi.org/10.3390/molecules24050871

Zou, F., & Budtova, T. (2021). *Tailoring the morphology and properties of starch aerogels and cryogels via starch source and process parameter*. *255*, 117344–117344. https://doi.org/10.1016/j.carbpol.2020.117344

Zuzanna Żołek-Tryznowska, & Kałuża, A. (2021). *The Influence of Starch Origin on the Properties of Starch Films: Packaging Performance*. *14*(5), 1146–1146. https://doi.org/10.3390/ma14051146

Carbon Aerogels and Monoliths: Control of Porosity and Nanoarchitecture via Sol–Gel routes. (2014). ACS Publications. https://pubs.acs.org/doi/abs/10.1021/cm402239e

G. Ehiowemwenguan, Inetianbor, J., & Yakubu, J. (2014). *Antimicrobial Qualities of Senna Alata*. ResearchGate; IOSR Journals. https://www.researchgate.net/publication/272986022_Antimicrobial_Qualities_of_Sen

na_Alata

Gritsanapan, W., & Mangmeesri, P. (2009). STANDARDIZED SENNA ALATA LEAF EXTRACT. *J Health Res*, *23*(2), 59–64.

https://thaiscience.info/Journals/Article/JHRE/10893386.pdf

Laura Martins Fonseca, Tavares, F., Graziella Pinheiro Bruni, Caroline Dellinghausen Borges, Martins, F. T., & Renato, A. (2021). *Aerogels based on corn starch as carriers for pinhão coat extract (Araucaria angustifolia) rich in phenolic compounds for active packaging*. *169*, 362–370. https://doi.org/10.1016/j.ijbiomac.2020.12.110

Oluwole Solomon Oladeji, Funmilayo Enitan Adelowo, Abimbola Peter Oluyori, & Deborah Temitope Bankole. (2020). *Ethnobotanical Description and Biological Activities of* Senna alata. *2020*, 1–12. https://doi.org/10.1155/2020/2580259

Rangaraj, V. M., Krishnamoorthy Rambabu, Banat, F., & Mittal, V. (2021). *Natural antioxidants-based edible active food packaging: An overview of current advancements. 43*, 101251–101251. https://doi.org/10.1016/j.fbio.2021.101251

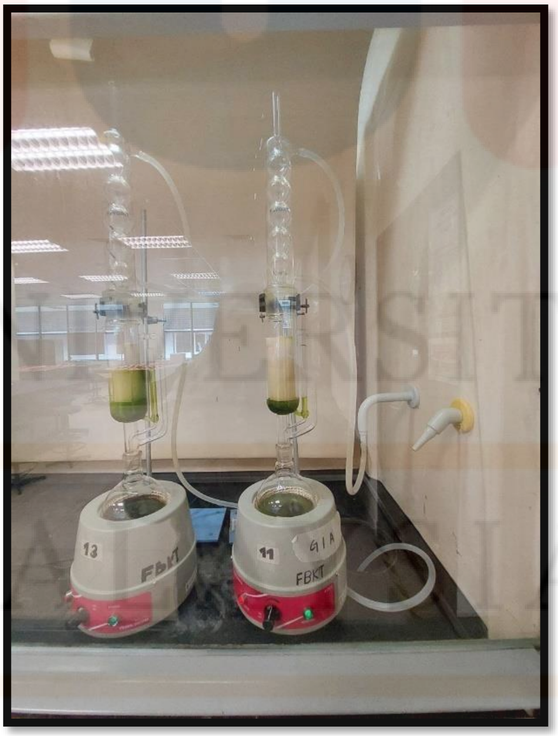
Wang, Y., Wu, K., Xiao, M., Saffa Riffat, Su, Y., & Jiang, F. (2018). *Thermal conductivity, structure and mechanical properties of konjac glucomannan/starch based aerogel strengthened by wheat straw. 197*, 284–291. https://doi.org/10.1016/j.carbpol.2018.06.009

Ye, S., He, S., Su, C., Jiang, L., Wen, Y., Zhu, Z., & Shao, W. (2018). Morphological, Release and Antibacterial Performances of Amoxicillin-Loaded Cellulose Aerogels. *Molecules*, *23*(8), 2082. https://doi.org/10.3390/molecules23082082

Zheng, Q.-Y., Tian, Y., Ye, F., Zhou, Y., & Zhao, G. (2020). *Fabrication and application of starch-based aerogel: Technical strategies*. *99*, 608–620. https://doi.org/10.1016/j.tifs.2020.03.038

APPENDICES A

A1: Senna Alata (L.) *roxb* powder.



A2: Soxhlet Extraction of Senna Alata (L.) *roxb*.

A3: Extraction of Senna Alata (L.) *roxb*