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Production of Biocomposite Films using Gum Arabic and Chitosan from Luffa Gourd for Food Packaging Applications

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Abstract

Most common food packaging are from nonbiodegradable and nonrenewable sources making them to be toxic to human, animals and their environment. It becomes paramount to search for alternate sources of food packaging materials. This study attended to reveal chitosan extracted from renewable agricultural waste (luffa sponge) chitosan was produced via three stages; proteinization, demineralization and deacetylation. The Produced chitosan was characterized for yield of 41.07%, which was a very high value from plant. The pH of 6.7 and degree of deacetylation of 33.2%. The FTIR result of the produced chitosan indicated the presence of –OH and $–NH_2$ groups which are the main groups that constitute the active sites of the biopolymer. The TGA indicated that the chitosan is thermally stable while the morphological study of the chitosan derivative has shown it has a non-porous structure. The effect of chitosan reinforcement on the Gum Arabic (GA) matrix on mechanical and barrier properties has been studied for food packaging applications. Four different mixtures at chitosan (C) % were used to prepare the

composite film with Gum Arabic: C-GA (5-95%), C-GA (10-90%), C-GA (15-85%) and C-GA (25-75%), respectively. The prepared composite films were subjected to morphological, mechanical strength, film thickness, water vapour permeability (WVP), and water uptake analysis. It has been observed that C-GA (15-85%) is well dispersed in the matrix (Image analysis via. SEM). Mechanical properties of the C-GA (15-85%) composite film evaluated indicated an increase in the strength. A film with more matrix showed less absorption of water as well as less water vapour permeability. The WVP test analysis revealed that film C-GA (15-85%) possesses the best barrier properties.

Introduction

Packaging is an important factor in the food industry and is dominated by petroleum derived polymers. However, an abundance of research is now being conducted on the development and evaluation of biodegradable materials, mainly due to concerns over potential damage posed by synthetic packaging materials. Several biopolymers have been exploited to develop eco-friendly food packaging materials. As known, chitosan as a natural polymer is like starch and cellulose, consisting of deacetylated d-glucosamine and *N*-acetyl-d-glucosamine in an irregularly dispersed manner with β-(1–4)-linear polysaccharide linkages. Moreover, chitin/chitosan could be also acquired from fungi or from plants.

Additionally, the most important chitin/chitosan from fungi and plants is a more attractive alternative for consumers of veganism and allergic to seafood (Chiu et al., 2017; Sebastian et al., 2020). Joseph et al. (2021) mentioned that “The chitosan market is escalating exponentially, while crustaceans are majorly exploited to meet the growing demand, and fungal chitosan exhibit more functional properties than marine and terrestrial sources.” Crognale et al. (2022) also noted that

Chitosan could be obtained from chitins via deacetylation. As known, chitin is typically derived from the exoskeletons crustaceans and fungal cell wall, but this biopolymer can also be found in other sources, such as plants. In fact, *Luffa* spp. is popularly utilized as Asian edible plants abundant in chitin/chitosan. This

first-attempt study tended to reveal chitosan extracted from renewable agricultural waste (*i.e.*, *Luffa* leaf waste), through comparative analysis upon the contents of the different portions of *Luffa*. *Luffa* is a genus of tropical and subtropical vines classified in the cucumber (Cucurbitaceae) family. The fruits of these species are cultivated and eaten as a vegetable. When the fruit is fully ripened, it is very fibrous. The fully developed fruit is the source of the loofah scrubbing sponge which is used in bathrooms and kitchens as a sponge tool. *Luffa* has fruits possessing a net-like fibrous vascular system (*Luffa* sponges) consisting of cellulose and lignin (1.4% and 2.9%, respectively, of the sponge dry weight) (Chang s.c. et al., 2019). To the present, *Luffa* sponges have been applied to immobilize biocatalysts such as enzymes, microorganisms, organelles, and plant and animal cells in bioreactors (Pazzetto et al., 2022), scaffolds for tissue engineering (Chen J.-P et al., 2023), and dye absorbents from aqueous solutions (Altinis et al., 2020) and for developing biofiber-reinforced bio-nano-composites (Siqueira et al., 2023).

Smart packaging serves additional crucial purposes, such as maintaining food quality, in addition to safeguarding food from the outside environment by giving packed food superior barrier properties. Additionally, smart packaging can lower the danger of pathogen development and offer effective surface contamination control, which is beneficial for foods with high water activity, like fish, to prevent microbial growth on their surfaces (Turgis et al., 2008).

Gum Arabic is a water-soluble polysaccharide and its biocompatible characteristics are a result of its favorable hydrophilic and hydrogen bond characteristics. It has a number of advantageous applications, including food reinforcing, preservative, emulsifier, stabilizer, and is a source of dietary fiber. Moreover, it could be used as a component to increase a material's physical and structural characteristics, such as hydration, oil-holding capacity, viscosity, texture, sensory properties, and shelf life (Rezaei, et-al., 2016). Several types of edible material produced from Gum Arabic, or mixtures of these bioplastics have recently been produced as a result of earlier studies. The lack of toxicity and excellent stiffness characteristics make it ideal for application in the industries of medicine, cosmetics, and food packaging. In addition, almond gum is found to exhibit surface, interfacial and emulsification activities. Although chitosan and other polymers have been utilized in multiple studies for food packaging applications, these two combinations are not described in any of them (Farooq et al., 2022).

The aim is to investigate how loading Gum Arabic affects the dissimilar physiochemical characterization of chitosan from Luffa Gourd. The biocomposites characterized for the thermal, mechanical, and antimicrobial activities. Thus, it represents an ideal material for active food packaging applications.

MATERIALS AND METHODS

Materials

Fresh Luffa gourd was harvested around fences of building at Kurmi Mashi, Kaduna south. It was the sun dried for 2 weeks after which it was then dried in the oven at 103 ^{0}C for 8 hours. Sodium hydroxide (NaOH), Hypochlorite, Glucose and galactose were obtained from Cybex laboratory store in Kaduna town

Deproteinization and Deacetylation of Luffa Membranes

The dried Luffa fruit fibers were pulverized to 100 μm and autoclaved at 90^{0}C for 20 min to soften the fibrous structure and then were blended to make a paste. The paste was then deproteinized by digestion with 1 M NaOH at 85◦C for 4h, to give the solids, alkali-insoluble materials (AIMs) containing chitin. The AIMs residue was collected and washed with deionized water to remove any residual NaOH. The fibers of lengths ranging between 10 to 50 μm were collected and dispersed in deionized water to form a suspension. The suspension was filtered using filter paper under aseptic conditions. The membrane formed on the filter paper was then dried in laboratory Oven (BDH 1200, China) to obtain the final product for further Synthesis.

Demineralization Decolorization of Chitin to Chitosan

Furthermore, due to plant chitin containing low minerals (e.g., $CaCO_3$), AIMs would be demineralized usually with dilute hydrochloric acid (0.25 M) to dissolve calcium carbonate, and followed by decolorization to remove pigments (*e.g.*, astacene, astaxanthin, canthaxanthin, lutein, and β-carotene) with KMnO (Elsoud & Kady, 2019). Finally, the pHs of the filtrates were adjusted to alkaline pHs >9 to form the chitosan precipitates.

Preparation of CHITOSAN-Gum Arabic Film

The chitosan film-forming solutions (C) were prepared according to a method described by Xu et al. (2019) with some modifications. Certain amounts of

chitosan powder as shown in Table 1 were dispersed in the aqueous solution of acetic acid (1%, v/v), and glycerol as a plasticizer, at a concentration of 0.75 g glycerol/g chitosan, was added to the solution. The resultant mixture was then stirred at 60 °C by a heater–stirrer for 3 h. In parallel, to prepare the Gum Arabic (GA) solution, the method of Choi et al. (2019) was used. In brief, the gum was powdered in a high-speed mechanical blender (Model 1000, Asan Toos Shargh, Tehran, Iran) to obtain a smooth powder. Then, OG solutions were prepared in distilled water by adding certain amounts of OG and stirring overnight at ambient temperature. The resultant mixture was centrifuged at 1500 g for 10 minutes. Subsequently, the supernatant was again centrifuged at 2500 g for 10 min and successively at 10,000 g for 20 minutes to remove insoluble materials.

Characterization of the Films

Water Solubility

Films solubility was measured in accordance with the methodology described by Gontard et al. (1994). Approximately 2 cm in diameter of the films were cut and weighed before and after the oven drying process at 105 °C for 24 h. Then, the samples were immersed in 50 mL of distilled water at 25 °C and subjected to constant stirring at 125 rpm for 24 h. Afterward, in order to determine the dry matter content of the solubilized films, the CH films were dried and weighed as previously explained. The water solubility of the films was expressed as percentage of the dry matter of films solubilized in distilled water.

Mechanical Analysis

The mechanical properties of the films were measured using a SANTAM machine (STM-5, SANTAM, Karaj, Iran), according to the ASTM method D882. Before testing, the prepared films were equilibrated at 50% RH in a desiccator containing saturated magnesium nitrate solution at 25 °C for 24 h. The initial grip separation and crosshead speed was adjusted at 50 mm and 5 mm/min, respectively. Two mechanical parameters, including tensile strength (TS) and elongation at break (EB), were calculated from force extension curves. TS (MPa) was determined by dividing the maximum load on the film before rupture by the cross-sectional area of the initial sample, and EB (%) was defined as the ratio of the final length at the point of specimen failure to the initial length of a sample and expressed as a percentage.

Antibacterial Properties

The antibacterial impacts of the fabricated films were tested against two important foodborne pathogens, Staphylococcus aureus (ATCC 25923) and Salmonella Typhimurium (ATCC 14028) by the disc diffusion method [26]. In brief, the stock cultures were prepared overnight in brain heart infusion (BHI) agar (Merck, Darmstadt, Germany) at 37 °C. For the preparation of inoculums, the tested bacteria were transferred into BHI broth tubes and, after 18 h incubation at 35 °C, the bacterial density was adjusted to approximately 108 CFU/mL using a spectrophotometer (Optizen 2120UV plus, Mecasys Co. Ltd., Daejeon, Korea) at 600 nm. Then, the CH films were cut into a 5 mm diameter circular disc and placed on the surface of pre-inoculated (with 100 mL of each bacterial suspension) Muller Hinton agar (MHA) Petri dishes and incubated at 37 °C for 24 h. Inhibition zones were then measured using a caliper and recorded.

Fourier-Transform Infrared (FTIR) Spectroscopy

The interactions of CH and OG molecules in the structure of the developed composites were determined using a Bruker Equinox spectrometer (Bruker Banner Lane, Coventry, Bremen, Germany). In this regard, two samples, including pure CH and CH1/OG0.625 films, were selected and analyzed. Analyses were carried out in the range of 4000 to 400 cm^{-1} at a resolution of 4 cm^{-1}. Sixteen scans were co-added for each spectrum.

Film Morphology

The morphology of the films was studied by a scanning electron microscope (LEO-1450 VP, Zeiss, Oberkochen, Germany). Film's samples were mounted on the specimen holder and sputter-coated with gold (10 mm) under vacuum, and SEM photographs were obtained

RESULTS AND DISCUSSION

Chitosan Results

Table 1: Physicochemical Parameters of Chitin and Chitosan

S/N	Parameters	Value (g)	Value (% Raw Luffa)
1	Chitin yield	53.22	53.22
2	Chitosan yield	41.07	41.07
3	Chitosan pH	6.7	6.7
4	Chitosan degree of deacetylation	73.12	73.12

Table 1 shows the results of physicochemical analysis of chitin and chitosan, and it was observed that the amount of the produced chitinwas categorized as the yield of chitin from the raw Luffa gourd before extraction process quantity of chitin from 100% of processed luffa gourd was 53.22%. This showed a high amount of chitin and give sign of a positive begin for chitin from plant sources. The yields also indicates that the product can be highly commercialized since the abundant of Luffa gourd is very high in Nigeria, making a room for local production. From the chitin produced the process went further by deacetylation for the production of chitosan. From the yield calculated from 100 g basis of raw luffa gourd, 41.07% chitosan was gotten. This is a big breakthrough from Luffa having such a high amount of chitosan. This will lead to having good catalyst, adsorbent and drug carriers from chitosan from plant sources, making the vegetarians more comfortable and having a cleaner source of chitosan since Plant sources seems cleaner.

Degree of Deacetylation of Chitosan

The degree of deacetylation (DD) of chitosan determines the amount of acetyl groups that have been removed from the chitosan structure, leaving behind free amino groups on the polysaccharide. The greater the DD, the more available amine groups are exposed as potential binding sites for contaminants. Studies have concluded that temperature, reaction time and NaOH concentration have a significant effect on the DD of the synthesized chitosan. Deacetylation process is achieved by treating chitin with concentrated NaOH or KOH (40 to 50%) usually at around 100 ^{o}C for several hours. A similar trend was observed in the study by Palpandi et al. (2019), they find out that synthesized chitosan by treating chitin with 40% NaOH solution at 110 ^{o}C for 6 h, Kumari et al. (2017) synthesized chitosan from fish scales, shrimp and crab shells using 40% KOH at various temperatures for 6 h, and Hussain et al. (2019) synthesized chitosan by treating chitin with 40% NaOH at 80 ^{o}C for 4 and 8 h. In the present case, the reaction time was kept constant at 6 h and temperature (80 ^{o}C, 100 ^{o}C and 120 ^{o}C) and NaOH (20%, 40% and 60%) concentration was varied to the synthesis chitosan samples having different DD. Table 1 shows the determined DD of chitosan samples synthesized in this work. The general trend observed was that DD was 73.12% with NaOH concentration of 40% when the synthesis temperature was kept constant at 80 ^{o}C, which promotes the degradation of the acetyl group and formation or exposure of more amine groups, thus increasing

the DD of the sample. Increasing NaOH concentration to more than 40% will induced reduction in DD of the chitosan. DD was reported as 73.12% when the NaOH was 40% at 80 °C.

FT-IR Analysis

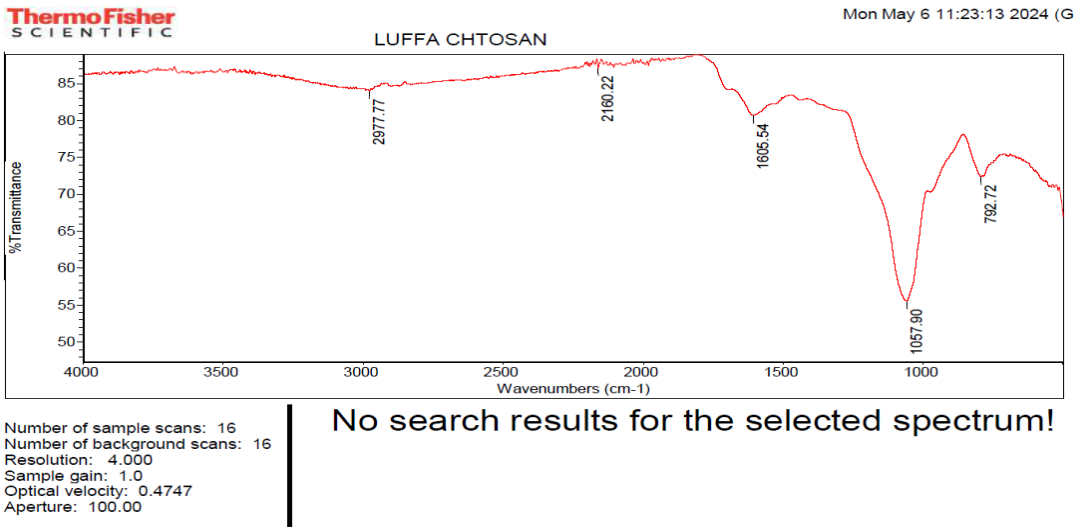

Fig. 1: FT-IR Analysis of Chitosan from Luffa Gourd

Figure 1 present the FTIR analysid of the chitosan from luffa gourd, and it was seen that the FTIR spectra, characteristic absorption bands from functional groups of chitosan can be observed, at 2870 cm^{-1} derived from the stretching vibrations of the C–H bond occurring in this polysaccharide. Next, stretching vibrations of C=O group at wavelength number 1724 cm^{-1} may have originated from Luffa gourd. The low intensity absorption band at 1447 cm^{-1} came from the hydroxyl group of chitosan. The band at 1350 cm^{-1} can be attributed to the stretching vibrations of C–O group. Also, the band at 1095 cm^{-1} with relatively high intensity compared to the other ones can be attributed to the stretching vibrations of –C–O–C– groups' characteristic for polysaccharides. Bands at 953 cm^{-1} and 840 cm^{-1} may have come from the amine group of chitosan.

TGA of Chitosan

From Figure 2, the first weight loss step was seen in the temperature range of 22-100 °C corresponding to the loss of moisture (around 6.7%). Under the flow of Nitrogen, a Non-oxidative thermal degradation occurs in chitosan which was observed in the temperature range of 248-600 °C, indicating the deacetylation

of chitosan, vapourization and elimination of volatile products. The degradation of chitosan starts with amino groups forming unsaturated structures as a result of the pyrolysis of the polysaccharides leading to a random split of the glycosidic bonds occurred at 850 °C, which are further followed by decomposition forming acetic and butyric acids and a series of lower fatty acids.

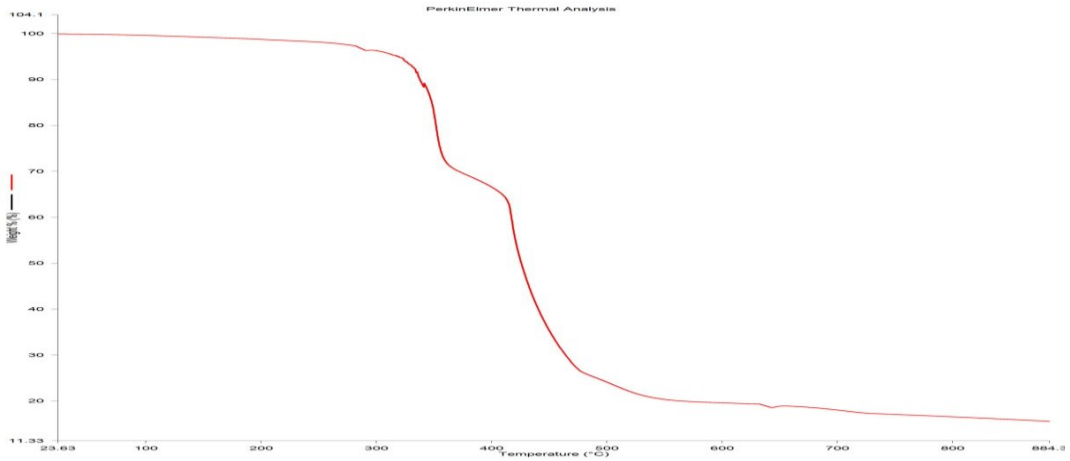

Fig. 2: Thermogravic Analysis of Chitosan

Scanning Electron Microscopy (SEM)

The SEM images of the pure chitosan (Figure 3) gives the morphology of the Luffa gourd chitosan which indicates a nonporous, smooth membranous phase which consists of cross-section of randomly oriented grains, micro fibrils and crystallite.

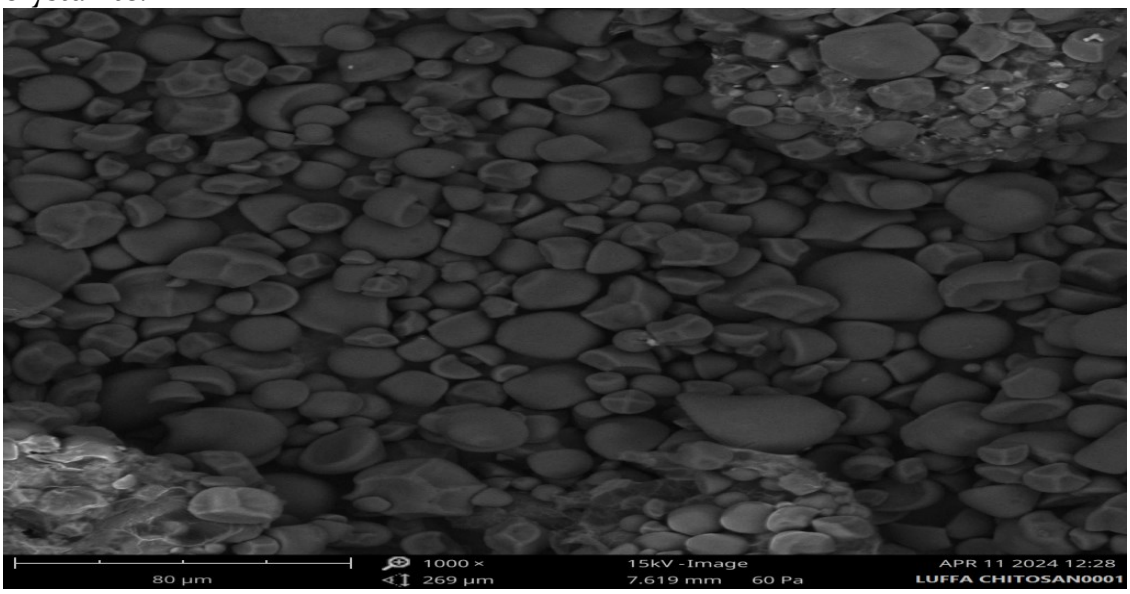

Fig. 3: Scanning Electron Microscopy (SEM) of Luffa Gourd

Composite Results

Water absorption.

Figure 4 shows effect of CH loading on water sorption of C-GA composites. It was clearly detected that water sorption increases with decreasing chitosan loading due to increasing hydrophilicity of the composite as expected. The increase is almost in a linear fashion. Linearity in the graph proves that the composites did not exhibit agglomeration which would complicate water sorption tendency of composites with increasing fiber loading. It was observed that increasing the amount of chitosan reduced the hydrophilic nature of the sample. C-GA 25% better composite, making it the least water absorbed sample. This implies that the Chitosan from Luffa has a good treatment by macerations, making it a good fiber that resist water absorption and will also be able to resist bioactivities.

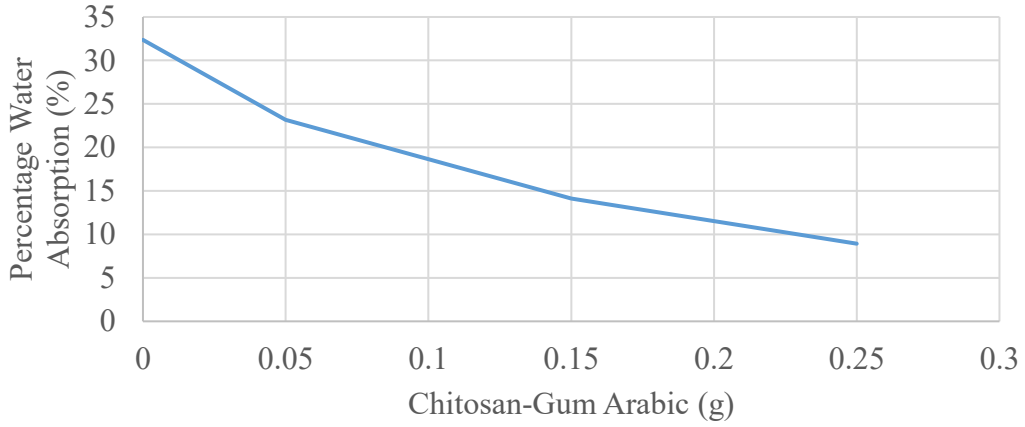

Fig. 4: Graphical Representation of Water Absorption

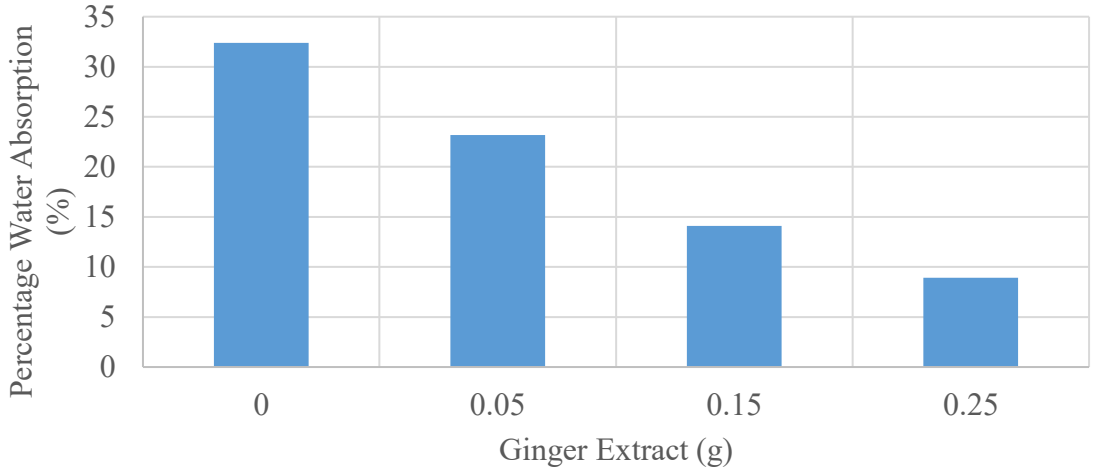

Fig. 4.: Histogram Representation of Water Absorption

FTIR analysis of fibers and composites

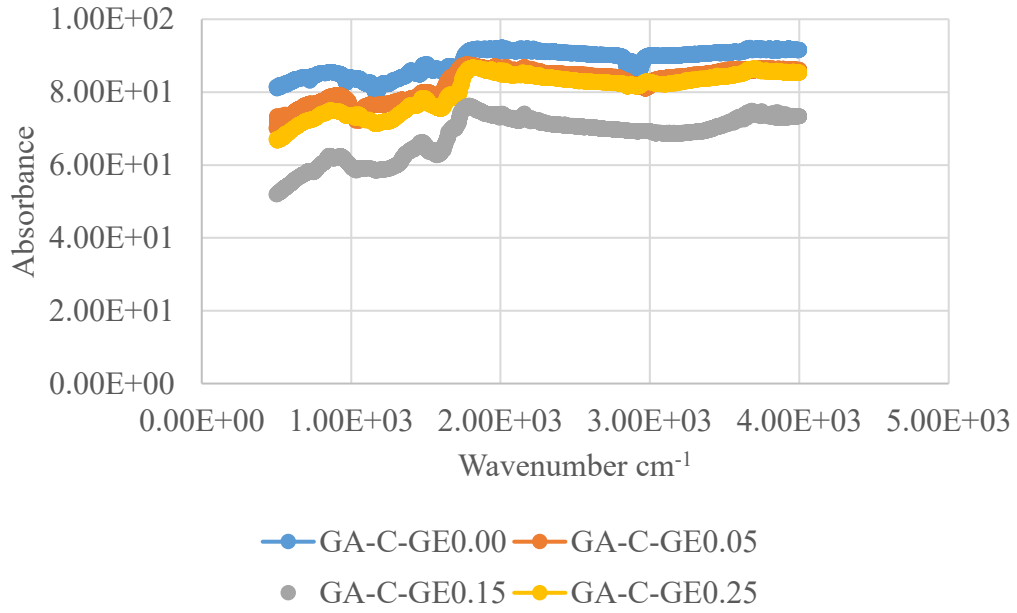

Figure 5: FTIR Analysis of *Fibers and Composites*

As indicated in Figure 5, the functional groups and the nature of molecular interactions between the C-GA were studied by using Fourier Transform Infrared (FT-IR) spectroscopy. The FT-IR spectra of C-GA displayed principal points in wavenumber length around 1413.31 cm^{-1}, 1603.21 cm^{-1} (symmetrical, asymmetric stretching vibration of –COO), 2953.42cm^{-1} (C–H stretching) and 3287.12 cm^{-1} (O–H stretching, characteristic of a glucosidic ring). Concerning the addition of different concentrations from Chitosan to Gum Arabic (GA) composite films, the resulted spectral analysis for C-GA films showed the same typically wavenumber length compared to GA films evidence for only physically interaction occurred between GA and C-GA polymer composite. The physically interaction is favourable to maintain the bioactivity of incorporated extracts by impeding the disappearance of their active groups by chemical bonding. The incorporation of varying Ginger extracts dosage inside polymer composite proved the physically interaction with small changes in wavenumber length. The peak around 1734 cm^{-1} is specific to GA-C assigned to C=O stretching in little hemicellulose remaining. The peaks observed GA-C at around 1460 cm^{-1} and 1420 cm^{-1} are assigned to CH deformation of lignin. These results prove that only constituent of C is cellulose but GA and C are polymeric in nature.

Antioxidant Effect of Produced composite Films

The antioxidant capacity of produced composite films was determined based on DPPH method. From the results obtained, this method indicated the suitability of the composite films to scavenge free radicals. The results showed that DPPH radical scavenging values increased with the increase of Ginger extract concentrations. Comparatively, the C-GA displayed the maximum antioxidant activity by reduction up

to 92.48% of stable DPPH radicals. C-GA25% and C-GA10% reduced the radical of DPPH by 68.32 and 41.19% respectively. While the C-GA which was the control was able to reduce the radical by 15.3%. this indicates that the Luffa is also playing a large role in radical reduction. From the analysis done, reasonable effect of films as antioxidants is due to the presence of Chitosan (G) within the folds of Gum Arabic composite films.

CONCLUSIONS

Chitosan and Gum Arabic can be used to produce a composite film with desirable packaging characteristics. The C-GA composite materials were successfully prepared by solution cast method. In the Biocomposite, the concentration of Chitosan varied from 5.0 to 25.0 wt. %. FTIR spectra demonstrated good interaction among the materials. The film solubility values showed that it has good solubility and low porosity. Gum Arabic was discovered to improve tensile strength and film thickness when combined with Chitosan, but had a reversed result on the film's water solubility. This green composite film could find potential applications in transparent biodegradable food packaging.

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