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COMPREHENSIVE RESEARCH ON BIODEGRADABLE FILMS AND COMPOSITE COATING

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Abstract

Plastics are the most widely used and easily accessible packaging material, yet it also has serious environmental consequences. The increasing global demand for packaging has prompted a reevaluation of plastic's negative effects on both wildlife and humans. Since it is typically used only once before being discarded, it does not biodegrade and hence accumulates in enormous quantities over time. For this reason, research into edible biodegradable films and coatings is crucial. One of the most exciting new trends in food packaging is edible packaging, which represents a crucial stage in the food production process. Materials on the research and

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588
development of new composite materials based on sodium alginate and chitosan for the fabrication of edible coatings and films are presented in this article. Presentation of the findings on physical, chemical, and antibacterial properties of these compounds.
Keywords: Biodegradable food films and coatings, edible films and coatings, sodium alginate, physicochemical qualities, antibacterial properties, and protection of the environment.

Introduction

Synthetic polymer packaging recycling is a pressing issue. The annual discharge of non-biodegradable garbage into the ocean is estimated at 8 million tons. Synthetic polymers can take decades, if not centuries, to degrade in soil. According to European Parliament Directive 94/62/EC on the decrease in the use of light (thickness 50 m) and ultralight (15 m) plastic bags, the average European should use no more than 40 light bags in a year by the end of 2025. The Directive further stresses that oxo-biodegradable and oxo-degradable plastic bags are not truly biodegradable since hydroxy compounds merely aid in breaking down plastic into tiny particles that are not biodegradable and persist in the environment for a very long period.

Research on environmentally friendly biodegradable edible films and coatings is therefore one of the most pressing areas of study. One of the exciting new trends in the evolution of food packaging is the use of edible materials. Edible films and coatings are the only biodegradable polymer packaging that may be thrown away with regular trash without first having to be separated from other recyclables. Protecting the product against mechanical damage, physical and chemical causes, and microbiological damage is the job of both edible and synthetic films and coatings. Several signs must be present before they can be used in the food industry: organoleptic indicators; structural resistance to exposure to microorganisms; light, heat, and frost resistance; vapour, gas, and scent permeability; optical qualities (transparency, tarnish); and mechanical properties (flexibility, stretching, tensile strength) that meet expectations.

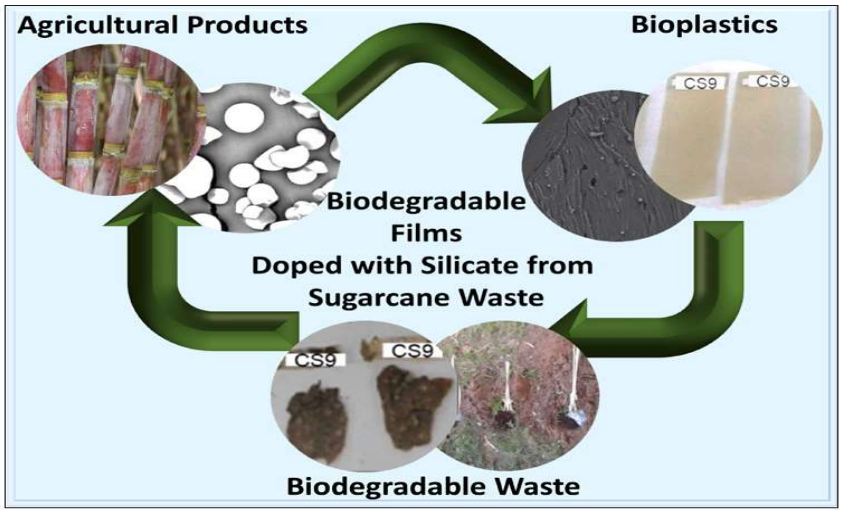

Figure 1: Biodegradable Films Derived from Corn and Potato Starch

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

Assimilable edible films and coatings are those made from digestible macronutrients like proteins and fats, while non-assimilable ones are made from substances like natural waxes, water-soluble natural and synthetic gums, water-soluble derivatives of cellulose, polyvinyl alcohol, polyvinyl pyrolidone, etc. There is a lifetime safe daily intake for non-assimilable coatings in the human body [1-5].

Research into edible films and their applications is still in its infancy. The primary reason for this is that their physicochemical and/or sensory qualities fall short of expectations. Edible packaging must meet stringent standards for non-toxicity, environmental safety, and sanitation. Non-preservative packaging with antibacterial characteristics is of particular importance.

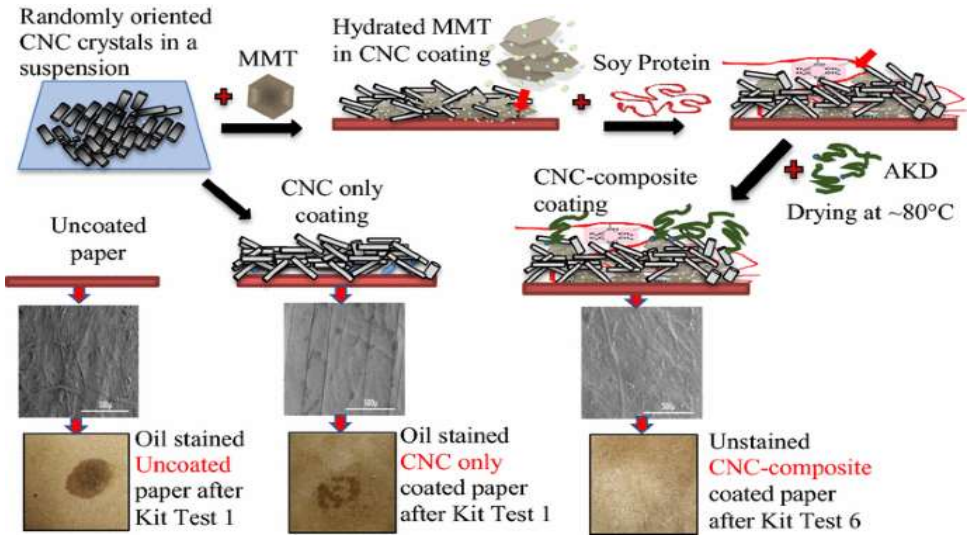

Figure 2: High performance nanocellulose-based composite coatings

The possibility of producing films and coatings out of a chitosan-biodegradable polymer with improved antibacterial and sorption properties and the required rheological features has been examined. Cleansing the body of accumulated toxins and wastes, its sorption properties allow for the elimination of heavy metals and radionuclides that have built up over time. Weight loss can be aided by the use of chitosan since it is low in calories and can be used to make stable gels that can selectively bind molecules of 'harmful fats' like triglycerides and cholesterol without absorbing unsaturated fatty acids. The films formed from chitosan are sufficiently robust, albeit inelastic.

Because of their cellular structure, both sodium alginate and chitosan polysaccharide polymer are capable of forming stable thermostable gels. Alginate is safe for human consumption, has antacid qualities, reduces blood cholesterol, is used in many weight reduction products and dietary supplements, and is an effective radionuclide and heavy metal detoxifier. Like chitosan, alginate has no calories. Curing and forming form-stable elastic hydrogels under the action of divalent metal ions is an undeniable benefit of alginates.

Because of how long it takes for the gel to dry at high temperatures, gel polymers are not suitable for usage as coatings on perishable foods like meat, fish, and other proteins. Therefore, it is prudent to create an edible coating based on sodium alginate and chitosan, which would integrate the benefits of both polymers: rapid alginate film formation (curing), biocidal properties, chitosan's

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

selective absorption of harmful fats, eco-rehabilitation of the body from toxins, low calorie content, and biodegradability. Because chitosan films are strong and alginate gels are elastic, the new coating is likely to have improved physico-mechanical properties.

Because of their thermodynamic incompatibility, combining polymers of sodium alginate and chitosan in a single coating had previously proven unfeasible. It is well known that coatings lose physical and mechanical properties and stratify into two phases during the drying process. Surfactants and plasticizers can be used to modify thermodynamically incompatible polymers, allowing for their possible joint use in composite materials. New composite materials based on sodium alginate and chitosan were formulated and manufactured with the goal of obtaining edible coatings and films, with the eventual goal of studying the physicochemical features of these materials through their application to products [6-9].

Materials and Methods

Materials

- The following sources were used in this investigation:
- Crustacean shell chitosan, which is of high molecular weight (Table 1 lists its physicochemical qualities). Bio progress, LLC, the supplier, is a limited liability company. Sodium alginate, extracted from Japanese kelp algae (the powder's physicochemical parameters are listed in Table 1);
- E472 is an ester of sorbitol hexahydrate and stearic acid, and it is used as a surfactant.
- Able to be used in the food production sector;
- White crystalline powdered citric acid (p.a.);
- Purified water
- Calcium chloride, a white granular substance.

Table 1. Physicochemical properties of sodium alginate and chitosan.

Polymer	Sodium alginate	Chitosan
Solubility in 2% acetic acid,%	100.0	99.7
Solubility in water,%	100.0	insoluble
Humidity,%	13.5	10.0
Ash content,%	17.38	0.46
Amine nitrogen,%	–	8.48
-COOH, %	17.60	–
Degree of deacetylation,%	–	98.0
Kinematic viscosity of a 2% solution, cPs	13.98	3173.6
Molecular mass, kDa	63.0	224.5

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

Analytical Methods

The formol titration method was used to quantify amine nitrogen. The approach involves the creation of methylene compounds, acids, through the reaction of amino acids with formalin, and their subsequent titration with alkali. Titrating with alkali allows one to determine an amine's concentration. We used an Ecotest 2000 ion meter to perform potentiometric titration on chitosan solutions in 0.1 N hydrochloric acid and 0.1 N potassium hydroxide to ascertain the extent to which the chitosan had been deacetylated. Chitosan's molecular weight was measured in a solution containing 1L of the polymer. 0.33 M acetic acid and 0.33 M sodium chloride. Sodium alginate's MW is as follows was measured in a 0.1 M sodium chloride solution [10].

Previous methods [10] using a potentiometric approach on an ANION 4100 ionometer with a glass combination electrode (pH = 0.05, T = 23 C) were followed to ascertain the functional group content in polymers. Using Eq.1, we were able to determine the percentage of free carboxyl groups (Kc) in sodium alginate.

$$Kc = \frac{C_{NaOH} \cdot V_{NaOH} \cdot 45}{Valg \cdot 1000} \cdot \frac{V_{bottle}}{V_{alg}} \cdot 100\%, \quad (1)$$

Where m_{alg} is the weighed amount of polysaccharide [g], 45 is the alkali equivalence coefficient for carboxyl groups and V_{NaOH} is the volume of alkali used for titration (in mL).

Film Preparation

The film-forming procedure was enabled by applying the prepared solution to the surface and mixing it with a calcium chloride solution. The amount of the combined ingredients dictated the final film thickness. The film-forming composition was sprayed onto a flat, clean surface and allowed to cure for 24 hours. A film-forming solution was placed into 89-mm-diameter Petri dishes. For 24 hours, we dried at 50 degrees Celsius inside a BinderED-115 thermostat.

Film Thickness Measurement

The film thickness was measured directly using an MK 50-1 micrometer. After taking ten measurements in triplicate over three distinct regions of the video, an average was calculated.

Determining Bactericidal Properties

Staphylococcus aureus ATCC 25923 and *Escherichia coli* ATCC 25922 were used to test the bactericidal effects. The 'grass' was created by streaking microorganisms on Givental-Vedmina nutrient agar medium, a finely divided, hygroscopic yellow powder that contains a variety of dry components. The feeding medium contained 21.0 g/L of pancreatic hydrolyzate of fish meal, 10.0 g/L of enzymatic peptone, 1.5 g/L of starch, 3.0 g/L of sodium chloride, and 12.0 3.0 g/L of microbiological agar.

A bacterial lawn culture was placed under a film with a diameter of 4 mm and incubated for 24 hours. The bacterial activity was measured by the size of the inhibitory zone surrounding the test film sample. Five independent samples were averaged to get this conclusion.

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

Determining the Degree of Water Absorption

Three by two centimeter film samples were air dried at 50 degrees Celsius during the day, then chilled in a desiccator to 23° Celsius before being weighed (W1). After that, 10 mL of distilled water was added to each sample in a 50 mL test tube. The samples were kept for a full day.

Samples were kept at room temperature and slowly mixed on a regular basis before being weighed and the amount of dry matter (W2) measured. The solution was then filtered, and the precipitate on filter paper was dried in the thermostat at 105°C for 24 hours.

Solubility was calculated by Eq. 2:

$$P = \frac{W_1 - W_2}{W_1} \cdot 100\%, \quad (2)$$

where P is the solubility (%), $\mathbf{W_1}$ is the mass of the sample dried in a thermostat before filtering and $\mathbf{W_2}$ is the mass of the sample dried in a thermostat after filtration.

Determining Physical and Mechanical Properties

On the Instron 3343 electromechanical testing machine, 35 mm 50 mm samples were subjected to tests of strength (St) and elongation in tension (L). The device computerized display showed the maximum force needed to tear each film. Film sample strength (S) was determined by multiplying the width (W) by the average thickness (T) of the film strips, and dividing the result by the maximum tensile strength (F). Eq. 3 was used to determine the strength:

$$St = \frac{F}{S}, \quad (3)$$

where **F** is the maximum strength of the break and **S** is the cross-sectional area of the film sample.

The elongation in tension (L) was calculated by dividing the increase in the length of the film strips at break (b) by the initial length of the film strips before loading (a), according to Eq. 4:

$$L = \frac{b}{a} \cdot 100\%. \quad (4)$$

Results and Discussion

Biodegradable film samples were made using solutions of chitosan, surfactant E472, sodium alginate, and liquid vegetable oil. The chitosan was dissolved in a citric acid solution between 3% and 5%. There was no more than 1.0% insoluble precipitate. Chitosan citrate was blended for 2 minutes with an aqueous combination containing 3-7% surfactant and vegetable oil. Then, an aqueous sodium alginate solution (between 1% and 3% by weight) was added and blended thoroughly. This resulted in a film-forming slurry, which was subsequently applied to the primed substrate and cured with a 2%-5% calcium chloride solution for 60 minutes at 20°C (Fig. 3).

The ideal proportions of the composite constituents were investigated (Table 2). Because of its widespread application in the food business, citric acid was selected as the solvent for chitosan.

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

The framework of the polymer (film) being formed is made up of layers (fibers) of high-molecular-weight chains, and the vegetable oil in the mixture is employed as the filler (plasticizer). Vegetable oil, which is safe for human consumption, is added to films to enhance their physical and mechanical qualities. This includes making the polymer more elastic and resistant to fracture when bent repeatedly.

Because the quantity of components may vary depending on the film's desired qualities, for example, greater elasticity or greater strength, the composition of the film-forming mixture is shown as intervals (Table 2). But they would not go beyond the ranges shown. The film's organoleptic qualities matched those of the containers they came in. Matte white in color, the film had a smooth, crack-free surface, a dense texture free of undissolved and foreign particles, and a low sheen.

Figure 3. Chitosan-alginate film-forming mixture

Table 2. The composition of the film-forming mixture.

Material	**Quantity,%**
Chitosan solution in citric acid	6–10
Vegetable oil	18–23
Surfactant solution E472	11.2–15.6
Sodium alginate solution	53–76
Calcium chloride solution	Unlimited

Table 3. Bactericidal properties of four-component chitosan-alginate films.

Chitosan concentration, %	**Growth inhibition zone diameter, mm**	
	Escherichia coli	Staphyloccous aureus

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

0.090	8.1±0.4	7.2±0.4
0.135	8.6±0.3	7.9±0.7
0.160	9.2±0.5	11.3±0.5
0.200	11.0±0.3	12.6±1.0
0.240	11.0±0.4	12.4±2.0

Tasteless, odourless and presented good chewiness. As a result of mixing, air bubbles were formed, which were removed by vacuuming the film-forming mixture before solidification. The biodegradable film made by this method with the introduction of chitosan into the mixture was0.19–1.20 mm thick.

The bactericidal and sensory properties of the four-component chitosan-alginate films, depending on the concentration of chitosan, are presented in Table 3. The bactericidal properties were better in films where the chitosan content is at least 0.2%.

To study the influence of the thickness of the developed film materials on their consumer properties, their strength and elastic characteristics were determined (Table 4). The obtained biodegradable packaging material had satisfactory quality characteristics and thus could be used as a coating applied directly to the product. As a test experiment, the developed chitosan-alginate composition was applied to chilled 6 cm^3 pieces of cod and catfish. Fig. 4 shows product samples after immersion in a calcium chloride solution to cure the coating. The chitosan-alginate coating was not destroyed during heat treatment (cooking, frying) and prevented the loss of moisture in fish samples.

Table 4. Physico-mechanical characteristics of four-component chitosan-alginate films of different thicknesses.

Chitosan con, %	**Alginate concentration, %**	**Film dimensions, mm**	**Average thickness, mm**	**Tensile strength, MPa**	**Relative extension, %**
0.2	1.5	35 × 50	1.18	32	155
		35 × 50	1.16	55	135
		35 × 50	1.14	41	139

Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

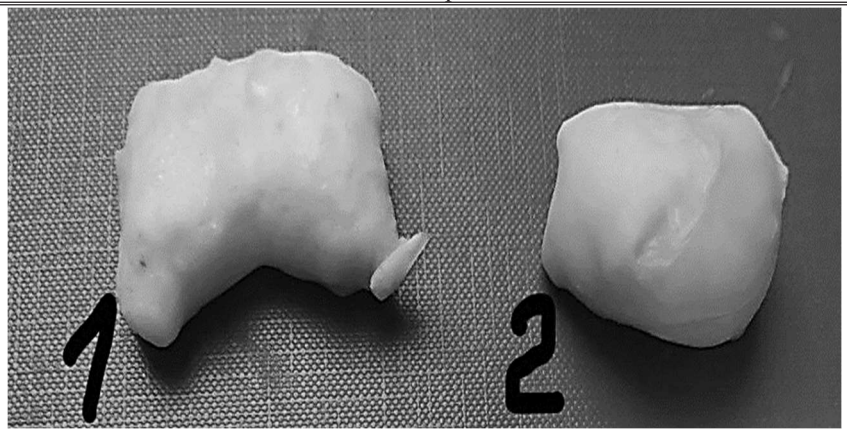

Figure 4. Samples of cod (1) and catfish (2) pieces with chitosan-alginate film applied to them.

Conclusion

Sodium alginate and chitosan were combined to create a composite material with desirable physicochemical and antibacterial properties for use in the manufacture of biodegradable food packaging films. Moussed items (sausages, meatballs) and fillets of meat and fish can both benefit from having the biodegradable film applied to their exteriors. When a film-forming component is wetted onto molded items or fillets and then placed into a hardener (calcium chloride solution) at room temperature, the film hardens instantly. The biodegradable film has the properties of biologically active additives; therefore, it can be used in food products to protect their surfaces from hazardous elements, increase their shelf life, and reduce their environmental impact. It can withstand high temperatures without melting. The film incorporates the benefits of both chitosan and sodium alginate, including but not limited to antibacterial activity, low caloric content, the ability to eco-rehabilitate the human body through sorption and excretion of hazardous pollutants, and the improvement of intestinal function.

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Catalyst Research Volume 23, Issue 2, September 2023 Pp. 1579-1588

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