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CHITOSAN AS BIOMATERIAL - AN OVERVIEW OF FUNCTIONALISATION WITH PLANTS EXTRACT

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Abstract

Chitosan as natural biomaterial is used in tissue engineering and regenerative medicine as a biomaterial alone, as well as in combination with other polymers. The recent research to obtain functionalized chitosan has also focused on the use of environmentally friendly natural resources, introducing different plants, for which new properties and applications in various modern fields have been highlighted. The use of hydro-alcoholic extracts and essential oils from plants to the production of functionalized chitosan-based materials (membranes, films, nanoparticles) shown improved antimicrobial properties and the use of these materials in various fields (medicine, food, industry, cosmetics and environment). The most valuable sources of natural compounds come from plants, being represented by a wide class of phenolic substances that can appear in all parts of plants in fresh or in dried form, extracts or essential oils from seeds, nuts, fruits, vegetables, leaves, roots or even from the stem and bark. The characterisation of membranes and films incorporating chitosan and plants extracts are referring of physical characterisation, structural, morphological structure, mechanical and biological properties based on their antimicrobial potential.

Keywords: *chitosan; biomaterial; plants extract; membranes; films; applications.*

Introduction

The intense development of modern technologies brings special interest for the research of biomaterials, in obtaining of new materials through various manufacturing processes. Biomaterials are synthetic or natural compounds able to interact with biological systems being used in the medical field especially for various and complex diseases [1,2]. The basic characteristic for biomaterials consists in the increased ability to integrate and be tolerated by the organism where they are implanted, coming into contact with natural biological fluids and human tissues, with minimization of adverse and unwanted reactions [3,4,5]. Biocompatible materials, for medical applications, must be primarily non-toxic, biodegradable, bioresorbable, not to produce essential changes to the biological pH, not to produce essential blood transformations, without allergic effects, etc. Biomaterials are diverse, classified for its chemical structure (metals, ceramic, polymeric, composite) or for their properties in several categories according to different criteria [6,7,8,9].

Chitosan is biopolymer with huge structural possibilities of chemical and mechanical modifications to generate new properties, functions and biomedical applications. Chitosan as natural biomaterial is used in tissue engineering and regenerative medicine as alone, as well as in combination with other polymers. Chitosan could introduce valuable properties such as

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antimicrobial activity, mucoadhesiveness, and biocompatibility, which are in demand for biomedical use [10]

Chitosan and its derivatives are preferred components as the biomaterial for applied medicine [11,12,13,14]. Studies, based on chitosan and chitosan-derivative activities against microorganism, clearly indicate the diversity and relevance of the research and use of chitosan and its derivatives as antimicrobial agents [15,16,17].

In many studies the bioactivity of compounds has not provided detailed molecular mechanisms, because is difficult to explain exactly how these molecules exert their activities. The chitosan and its derivates are versatile new materials such composites, membranes, films, nanoparticle and in the last decades a major interest in research and applications of them using extracts or oils from plants was reported [18].

Materials and Methods

Antioxidant potential of extracts and plant's oils

Plants are inexhaustible sources of biologically active substances with various properties in various fields used in food, as traditional remedies for treating various diseases, but also for obtaining various materials and others [19].

Antioxidants as the majority of compounds in plants are intensively studied and used for the specificity of their characteristics and are currently being introduced for the functionalization of chitosan [20]. Numerous studies on the antioxidant activity of chitosan films are difficult to compare due to the differences between the type of material used, the analytical methods and the results. Various studies confirm that chitosan-based films have low antioxidant activity and that their enrichment with various plant extracts significantly increases their antioxidant properties [21]. The study of antioxidants as well as their importance in various fields (medicine, pharmacy, food industry, but also materials engineering) is of major interest to the scientific community. In determination of the antioxidant activity, a lot of relevant tests have been described and introduced, these tests are based on chemical reactions and the evaluation of kinetics or equilibrium based on spectrophotometry, assuming the appearance of characteristic colors or discoloration of the analyte solutions that are processes monitored by specific wavelength adsorption [22].

The best known and most commonly used tests are mixed tests that include the transfer of both a hydrogen atom and an electron, such as the 2,2 ”-Azinobis- (3-ethylbenzothiazolin-6-sulfonic acid (ABTS) test) and [2,2-di (4-tert-octylphenyl) -1-picrylhydrazyl] (DPPH) test [22]. Specific tests based on the transfer of a hydrogen atom are the Oxygen Radical Absorption Capacity (ORAC) test, Hydroxyl Capacity Tests Radical Antioxidant Test (HORAC) and Electron Transfer-Based Tests include Copper Reducing Antioxidant Power (CUPRAC) performed in the basic environment, Happiness Reduction Antioxidant Power Test (FRAP), and others [23]. Tests have been successfully applied in the antioxidant analysis or determination of the antioxidant capacity of complex samples, such as extracts from various plants of therapeutic or food interest, but also in the analysis of various materials [24,25].

The most valuable sources of natural compounds come from plants, being represented by a wide class of phenolic substances that can appear in all parts of plants in fresh, dried form, extracts or essential oils from fruits, vegetables, seeds, nuts, leaves, roots or even from the stem and bark. Plants produce a wide range of secondary metabolites, such as flavonoids, alkaloids, lignin, terpenes, terpenoids, tocopherols, phenolics, phenolics, peptides, polyfunctional organic acids. Phenolic acids are present in almost all plants and herbal foods and supplements, accounting for a significant share of human diets [26].

The most important characteristic antioxidant compounds from extracts and plant’s oil are summarised in the Table 1.

Table 1. Antioxidant compounds from extracts and plant's oil and methods of detection

Plants	Chemical composition	Methods of detection	References
Boswellia carteri, Citrus paradise, Lavendular angustifolia, Lemon Citrus limon, Mentha piperita, Rosmarius officinalis	monoterpenes: linalyl acetate menthol ,1.8-cineole, limonene, p-menth-2-en-ol	DPPH, ABTS, FTC	[27]
Melissa officinalis L., Mentha peritae L., Ocimum basilicum L.	bicyclic sesquiterpenoids: α-trans-bergamotene; monoterpenes: geranial, neral, piperitenone oxide, cineole, linalool, 1.8-cineole	DPPH, ABTS	[28,29,30]
Argylia radiate (roots)	benzoic acid, gallic acid, vanillic acid, syringic acid, cinammic acids: caffeic acid, cumaric acid, cinnamic acid, ferulic acid, flavones: apigenin, quercetin, myricetin, luteolin, chrysin, flavon-3-ols: catechin, epicatechin; polyphenols: resveratrol, rutin, chlorogenic acid	ORAC, CAA, ESR, HORAC	[31]
Ganoderma lucidum , Ganoderma applanatum (fung)	flavon-3-ols: catechin, epicatechin; catechin, protocatechuic acid; polyphenols: 3-hydroxybenzoic acid, rutin terpenoids: 20-OH lucidenic acid A, 20-OH lucidenic acid N, ganoderic acid A, gaoderiol A , ganoderiol D	ABTS, RT, LPO, Nitric oxide scavenging activity	[32]
Eugenia patrisii, E. punicifolia, and Myrcia tomentosa Essential oils (EOs)	terpenoids: (E)-caryophyllene, bicyclogermacrene, germacrene D, γ-elemene oxygenated sesquiterpenes spathulenol, selin-11- 4α-ol	DPPH , TAC	[33]
Green tea *(Camellia sinensis)*	flavon-3-ols: epicatechin, epigallocatechin, epicatechin gallate, catechin, gallocatechin gallat	DPPH	[34]
	benzoic acid, gallic acid	DPPH, FRAP	[35]
Aristotelia chilensis	anthocyanins, flavonoids, phenolic compounds	DPPH, FRAP	[21]
Viola odorata Linn.	flavonoids	DPPH	[36]
Aronia melanocarpa, family Rosaceae (Rosaseae), sub-family pome (Pomoideae)	benzoic acid: gallic acid; anthocyanins	ABTS FRAP	[40]

DPPH = (2,2-diphenyl-1-picrylhydrazyl) assay; TAC= total antioxidant capacity; FRAP = Ferric Reducing Antioxidant Power; HORAC= Hydroxyl Radical Antioxidant Capacity; ABTS-radical scavenging; ORAC = Oxygen Radical Absorption Capacity; ORAC-FL - oxygen radical absorbance capacity-fluorescein; CAA -cellular antioxidant activity; ESR -spin trapping technique evaluated hydroxyl elimination capacity.

DPPH, ABTS and FRAP tests were used to investigate the in vitro antioxidant activity of hot water extracts from lotus flowers. The resulting antioxidant activities were similar to the synthetic antioxidant BHT (butylated hydroxytoluene) [37]. Extraction of antioxidant compounds from the various anatomical parts of N. alba (fruit, flower, leaf, stem and root) from the Danube, a method of obtaining extracts exhaustively by ultrasound was used, due to the chemical composition rich in polyphenols or obtained remarkable antioxidant results [38].

Lamiaceae species *Melissa officinalis, Origanum vulgare* used as spices both fresh and dried due to the varied composition of polyphenols and volatile compounds show promising values in DPPH, ABTS, ORAC tests [39]. The antioxidant activity of *E. purpurea, T. vulgare, Achillea millefolium, Hypericum performatum* and *O. vulgare* is the result of the content of phenolic acid, especially caffeic and p-coumaric acid [40].

Basil species, lemon balm and lemon mint, the species have a great antioxidant potential [28,30]. Essential oils from various plant species also have antioxidant properties, such as basil species (Ocimum spp.) Used as a traditional medicine analyzed by DPPH and cyclic voltammetry [29,33]. Nutmeg (*Myristica fragrans*) and cloves (*Syzygium aromaticum*) through their components are used to inhibit the oxidative whitening of b-carotene, as measured by the b-carotene agar diffusion test [34]. The highest DPPH radical scavenging activity was obtained from lavender and limonene essential oil, with RC50 values of 2.1 0.23% and 2.1 0.04%, respectively. The radical scavenging activity against the ABTS radical was the highest in peppermint essential oil (1.6 = 0.09). Lavender oil was most effective in inhibiting linoleic acid peroxidation after 10 days [27].

Tests such as CUPRAC, FRAP and DPPH, which are the most sensitive methods to phenolic compounds, have shown the antioxidant potential of different cucumber species *Cucumis pepo, Cucumis moschata, Cucumis ficifolia* [41].

Studies show that fresh fruits and juices have antioxidant properties on the antioxidant activity of fresh apple peels and show promising potential, with small differences between species plant debris [42]. Fresh juices of the Cucurbitaceae family have a strong antioxidant activity, especially the species of *Momordica charantia, Cucumis metuliferus* and *Trichosantes cucumerina* [43].

One of the known and widely used plants is *Aloe vera* extracts which have shown antioxidant capacity by permanganate and CV technique [44].

Specimens of *Eugenia punicifolia* and *Eugenia patrisii* showed the highest and, respectively, the lowest antioxidant capacities, using the DPPH method. In the TAC method, *Morchella tomentosa* specimens had the highest antioxidant potential due to the compounds extracted with ethanol [33].

The absence of differences in the antioxidant activity of the extracts in the species studied by *Ganoderma lucidum, Ganoderma applanatum* (fungus) the presence of reishi-like terpenes makes *Ganoderma applanatum* a promising raw material for the production of food supplements.

For the species *Argylia radiate* (roots), the results suggest the potential application of in vitro plant extracts as antioxidant compounds for various applications, ie food packaging, cosmetics, nutraceuticals, biopharmaceuticals and other biotechnological uses [33]. Finally, this paper will open an opportunity for the use of extremophilic plants in the driest desert in the world as a source of valuable compounds.

The study indicated that an active film from chitosan-based film could be achieved by incorporation with aqueous green tea extract, as a natural antioxidant. Addition of aqueous green tea extract improved mechanical, water vapor barrier and antioxidant properties of the resulting films [34].

The incorporation of maqui berry extracts to chitosan edible films may have supplementary applications in food packaging to delay microbial growth and to improve the oxidative stability of foodstuffs [21].

The new materials obtained from ground peanuts packed in chitosan and grafted bag of gallic acid-chitosan have antioxidant properties due to the incorporated compounds and have various useful properties [35]. The chitosan-alginate matrix retained the antioxidant compounds in the *Viola* extract that can be used as a natural antioxidant in food [36]. In the evaluation of antioxidant potential of extracts are important and methods to prepare them. In study of was determined the effect of different drying methods (by freeze-drying (FD), vacuum (VD), convective drying (CD), microwave (VMD) and combined method (CVM)) on the quality factors of chokeberry fruit, including phenolic compounds and antioxidant activity [45].

Chitosan functionalized with extracts and plant's oils

Chitosan remain most attractive material for research and applications and several reviews concluded the results from last decades [1,5]. Recent research to obtain functionalized chitosan has also focused on the use of natural resources that are as environmentally friendly as possible, prepared from different plants, for which new properties and applications in modern fields have been highlighted. In both situation oil or aqueous plants extracts were used in obtaining of new materials for functionalized of chitosan. Membranes and films of chitosan with plant extracts having antimicrobial activity are most common materials.

Table 2 summarizes information in the last 10 years, on the use of new materials with the main source of chitosan with different parts of plants.

Table 2. New materials obtained through functionalized chitosan with plants extracts and applications

Plants/ parts of plants used in extracts	Materials obtained	Filed of aplications	References
turmeric *(Curcuma longa)*	membranes	medicine (wound dressings)	[60]
neem seed *(Azadirachta indica)*	composite	medical textile	[51]
henna leaves	nanofibres	skin tissue engineering	[61]
aloe-vera	membrans	biomedical applications	[52]
Euphorbia umbellata	membranes	medical textile	[48]
Hibiscus Sabdariffa L.	chitosan matrix	antimicrobial	[64]
Salvia officinalis and *Satureja montana*	nanoparticles	nanomedicine	[58]
Senna auriculata and *Achyranthes aspera*	nanoparticles	medical textile	[59]
esential oils (ginger, rosemary, sage, tea tree, thyme) and hydroethanolic diferent extracts (ginger, rosemary, sage, black tea, green tea, kenaf leaves)	films	food packaging industry	[62,63]
esential oils rosemary (*Rosmarinus officinalis L.*), thyme (*Thymus vulgaris*), licorice (*Glycyrrhiza glabra L.*) centaury (*Centaurium erythraea*) and mugwort (*Artemisia vulgaris*)	coating	food packaging industry	[65]
pomegranate peel	coating	food packaging industry	[66]
purple corn extract	films	food coating and packaging industry	[67]
black rice	films	food packaging industry	[68]
purple-fleshed sweet potato	pH-sensing films	food packaging materials industry	[55]
red grape seed extract and *Ziziphora clinopodioides* essential oil	films	biodegradable active food packaging	[70]

pomegranate (*Punica granatum L.*) peel extract	coating	food coating	[56]
hops *(Humulus lupulus)*	films	food packaging materials, medicine (wound dressings)	[74]
Quercus	films	food packaging industry, environment	[76]
nettle *(Urtica dioica L.)* and sage *(Salvia officinalis L.)* leaves	films	food packaging industry, environment	[77]
Artemisia campestris	films	food packaging industry	[78]
pine needles (*Cedrus deodara*) extract	films	food packaging industry	[79]
Pistacia terebinthus	films	food coating and packaging industry	[49]
carvacrol and grape seed	films	food packaging materials	[50]
pine nut shell, peanut shell and jujube leaf	films	food packaging industry	[80]
spice (thyme, cloves, prickly ash, fennel, geranium and cinnamon)	films	food preservation, environment	[83]
Viola-odorata Linn.	microcapsules	food products and environment	[36]
chokeberry extract	films	food packaging industry, environment	[73]

The plants extract incorporated in chitosan is intensively studied, the most used being the methanolic extracts from fresh or dried plants. Methods of obtaining for chitosan-based membranes and films have been reported with procedures varied, most of them reporting plants extracts from different fraction of plants.

Methods of production for membranes and films include:

i. direct synthesis using chitosan with alcoholic extracts (methanolic, ethanolic, hydroethanolic);
ii. direct synthesis using chitosan with aqueous plants extracts.

The most characterisation of membranes and films incorporating chitosan and plants extract are referring of physic-chemical properties, structural, morphological structure, mechanical and biological using different methods (Table 3).

Table 3. Methods used in the characterisation of functionalised chitosan

Biomaterials / functionalized chitosan	**Methods**				**References**
	spectroscopy	**structural/ morphological**	**mechanical**	**biological activity**	
membranes	FTIR, AFM, UV-VIS, colorimetry	SEM	DMA, contact angle	bacterial cell suspension, agar diffusion method	[48]
films	FTIR	SEM, XRD, TGA	thickness, density, degree of swelling, humidity, color-light transmission	antioxidant, antibacterial	[83]

Biomaterials / functionalized chitosan	Methods				References
	spectroscopy	structural/ morphological	mechanical	biological activity	
nanofibres	FTIR	SEM	porosity, swelling degree, weight loss	antibacterial, cell biocompatibility, in vivo wound healing activity	[62]
nanoparticles	FTIR, photonic spectroscopy, doppler laser anemometry	SEM, DSC, HPLC		antioxidant activity (ABTS, ORAC)	[58]

Following the study conducted by Lemes et al. chitosan-based membranes and methanolic fractions other than *Euphorbia umbellata* (10%, 50% and 100%) are obtained in order to use the material as medicine application. Chitosan is dissolved in acetic acid and sodium benzoate, emulsifying agents (Tween – 80 and PEG 400) and plant methanolic fractions are added, and the control membrane is prepared in parallel. The resulting membranes are stored for 72 hours in a controlled environment before performing physic-chemical analyses.

Physic-chemical characterization was performed by SEM, density, DSC, TGA, XRD, FTIR and colorimetry techniques. The phenolic compounds released from the membranes were evaluated using the Folin-Ciocalteu method, and the antioxidant and antimicrobial activity was also studied. The increase in the amount of methanolic fraction added to the polymeric matrix led to the appearance of a different number of pores on the membrane surface, changes in calorimetric, spectroscopic and crystalline properties, as well as changes in color compared to the control membrane. These changes can be attributed to the chemical interactions that took place between the structure of chitosan and the phenolic compounds present in the methanolic fraction of *Euphorbia umbellata* extract. The samples with 50 and 100% of the methanolic fraction showed different profiles of release of phenolic compounds from membranes (controlled mode), promising antioxidant and antimicrobial activity for medical applications [48].

An experiment conducted by Kaya et al. demonstrated that chitosan films with improved antioxidant and antimicrobial properties can be obtained using methanolic extracts from *Pistacia terebinthus* (stem, leaves and seeds) which have a very high content of polyphenols. The films were obtained by dissolving chitosan in acetic acid and water over which methanolic extract and glycerol are added, the components being very well homogenized. Each mixture is dried in Petri dishes at 30 ℃ for 48 h. The films were analyzed by FT-IR, SEM, TGA, DSC, contact angle measurement and UV-Vis spectroscopy and also for mechanical properties [49].

Studies have been reported by Rubilar et al. on the properties of films based on chitosan**,** carvacrol and grape seed extract which may vary according to the combination ratio of the components. The chitosan film-forming solution was prepared by dissolving chitosan in glacial acetic acid and homogenizing at 9500 rpm for 20 min. The solution was kept at room temperature for 12 hours. After filtration, the solution was heated to 40°C and stirred for 30 min by adding plasticizer, glycerol and Tween-80 emulsifying agent homogenizing for 1 hour. Combine different amounts of the film components homogenizing for 5 min at 9500 rpm, after cooling to room temperature disintegrate under vacuum for 5 min. The mixtures thus obtained are passed on the plates and dried for 48 hours at room temperature (25 ℃). The properties of the films were compared with the control, and the mechanical, structural, barrier and colour properties were evaluated. The results suggest that grape seed extract and carvacrol influence the structure of the film and its mechanical properties due to the hydrophilic compounds (grape seed extract) and hydrophobic (carvacrol) respectively [50].

Composite materials synthesized from chitosan and aqueous extract from neem seeds and cotton fabrics treated therewith were obtained and characterized by FTIR for confirmation of functional groups, XRD for crystalline behavior determination, UV-VIS spectroscopy analysis for optical properties and SEM for surface morphological properties [51].

Membranes obtained by Silva et al. incorporating *Aloe Vera* in chitosan can be used successfully in obtaining dressings. The structural differences between chitosan and aloe vera, as well as the changes that occur in the developed membranes were analyzed by FTIR. Changes in the surface topography of membranes at the nanometer level were assessed by AFM. The contact angle and the degree of enzymatic degradation using lysozyme were determined. The mechanical properties of the resulting membranes were investigated by DMA [52]. Characterization of the mechanical properties of dressing materials is essential, as dressings obtained from films must be durable, resistant to stress and flexible to facilitate handling [53].

The potential for infection with various bacteria is always present, which has led to the development of improved dressings by incorporating various antibacterial agents. Evaluation of the antibacterial activity of the developed membranes was investigated against *S. aureus*, a Gram-negative pathogen present in wounds, using two methods: bacterial cell suspension and agar diffusion method. *Staphylococcus aureus* was chosen for this study due to its frequent presence in skin wounds [54].

Yong et al. synthesized antioxidant and intelligent pH detection films by incorporating sweet potato pulp with violet pulp (PSPE) rich in chitosan matrix anthocyanins. The effects of PSPE incorporation on the physical, structural, antioxidant and pH-detecting properties of films have been established. PSPE can significantly increase the thickness, water solubility; UV-vis light barrier property and thermal stability of chitosan film but at the same time can decrease the humidity, elongation at break and the crystalline character of chitosan film. The water vapor barrier properties and tensile strength of the film were almost unchanged by the incorporation of PSPE. Microstructure analysis confirmed the homogeneity of the films. The existence of intermolecular interactions between PSPE and the chitosan matrix was confirmed by infrared spectroscopy. Antioxidant tests have shown that chitosan-PSPE films can eliminate free radicals depending on the dose and time of action. Moreover, chitosan-PSPE films showed visible color changes with pH change due to the presence of anthocyanins identified based on mass spectroscopic analysis. Due to their properties relevant to the food industry, the films can be used as antioxidant and intelligent films [55].

Chitosan and gelatin-based materials incorporating pomegranate peel hydroethanol extract were synthesized. Both the viscosity and the activation energy of materials containing natural compounds are highly dependent on temperature, a statement demonstrated by the analysis of rheological properties. SEM analysis revealed the influence of pomegranate peel hydroethanol extract concentration on scaffold porosity. The results of the study showed that pomegranate peel extract is able to improve the functional characteristics of chitosan / gelatin-based materials for use in the food industry [56].

Chitosan films with different spice extracts were obtained in order to prolong the shelf life of the food. With the addition of spice extracts, significant changes were observed on the physical properties, such as thickness, density, degree of swelling, humidity, color parameters and light transmission; verified by SEM analysis of the film. FTIR, XRD and TGA spectra indicated that there were intermolecular interactions between chitosan and the various spice extracts. The antioxidant and antibacterial properties of composite films were also analyzed, which indicated the ability to remove radicals, while showing a prominent inhibitory effect against *Staphylococcus aureus* (S. aureus) and *Escherichia coli* (E. coli) [57].

Results - Applications

Chitosan and their derivates compounds obtained using plant extracts or oils as major natural biopolymer, nontoxic and biodegradable have many applications in such diverse fields: nanotechnology, medical, food, environment etc**.** There is difficult to made a rigorous classification, that materials have applications in one or more fields.

Medical materials

Abbas et al. reported obtaining of membranes composed of a mixture of chitosan, polyvinyl alcohol and curcumin extracted from turmeric (*Curcuma longa*) with antimicrobial properties. The potential for wound healing was analysed in the laboratory animals, with membranes showing significant properties against pathogens such as *Escherichia coli, Pasturella multocida, Bacillus subtilis, Staphylococcus aureu*s and repairing properties on damaged fibrous connective tissue following grade 2 burns [60].

A new material is the textile fabric made of cross-linked cotton with two different agents (glutaraldehyde and citric acid) and composed of chitosan and aqueous neem seed extract (CS-NS) [51]. The antibacterial activity of the CS-NS-coated cotton fabric and the CS-NS composite-coated cotton fabric with crosslinking agents was tested against gram-positive and gram-negative bacteria. CS-NS composite on cross-linked cotton has a higher antibacterial activity than the fabric with CS-NS composite on non-crosslinked cotton. Thus, the chitosan composite and neem seed extract can be applied on medical textiles.

Yousefi et al. conducted a study on the structure, mechanical properties, morphology, and swelling or weight loss of chitosan-based nanofibers and henna leaf extract that have been shown to have potential for use as biodegradable and antibacterial dressings for wound healing. Nanofibers have been studied for antibacterial activity, cell biocompatibility and in vivo wound healing activity as well as FT-IR [61].

Natural compounds extracted from plants with therapeutic properties such as those of aloe vera have contributed to the production of chitosan-based membranes for which determinations have been made on degradation, roughness, wettability and mechanical properties from which satisfactory results have been obtained for their use in wound dressing materials [52].

It was reported by Abdelghany et al. that African mallow has antimicrobial properties due to the chemical compounds identified in the prepared extracts. Films based on chitosan (CS), polyvinyl alcohol (PVA) and mallow extract were obtained which were characterized optically, structurally and morphologically by analyses such as: UV-Vis, FT-IR, XRD, FESEM and antimicrobial activity [64].

Nanotechnology/ nanomaterials

Biodegradable chitosan nanoparticles reported by Baptista da Silva et al. can be used in innovative medical therapies as carriers and protectors of compounds with strong antioxidant activity such as rosmarinic acid present in sage (*Salvia officinalis*) and winter thyme (*Satureja montana*) extracts. The chitosan nanoparticles were prepared by ionic gelation with sodium tripolyphosphate at pH 5.8 with a mass ratio of 7:1, with a theoretical antioxidant charge of 40–50%, having small dimensions, around 300 nm. Chitosan nanoparticles were then characterized by various methods, such as photon correlation spectroscopy, Doppler laser anemometry, scanning electron microscopy (SEM), differential scanning calorimetry (DSC), Fourier transform infrared spectroscopy (FTIR), high performance liquid chromatography performance (HPLC), combination efficiency and antioxidant activity. FTIR analysis found interactions between antioxidant and chitosan after encapsulation without the addition of other binding chemicals [58].

To increase the effectiveness of biological, chemical and physical properties, such as antibacterial activity and biocompatibility, cotton fabrics were finished with chitosan and plant nanocomposites (*Senna auriculata* and *Achyranthes aspera*). The study by Chandrasekar et al.

demonstrates that nanocomposites can provide better functional properties than bulk finished fabrics. Nanometer-sized particles in composites have been considered optimal for its applications in hospital tissues to prevent the transmission of nosocomial infections [59].

Food packaging materials

In studies by Souza et al. essential oils (ginger, rosemary, sage, tea tree and thyme) and hydro-alcoholic extracts (ginger, rosemary, sage, black tea, green tea and kenaf leaves) were used to obtain new materials based on functionalized chitosan with improved antimicrobial properties and their use in the food industry [62,63]. However, in order to identify whether these new materials can be used in the food packaging industry, further research is needed to evaluate the behaviour of these biopolymers when in contact with food.

Obtaining edible films to cover food with preservative properties depends on the method of production and the natural chemical compounds present in the plant extracts used. There is information in the literature that edible coatings based on chitosan have been obtained using essential oils and plant extracts to evaluate their effect on fruit preservation [65].

Films obtained from chitosan, alginate and pomegranate peel extract have been shown to be effective in maintaining the quality of guava fruit and the concentration of natural antioxidant compounds present in them during several days of storage at low temperatures [66].

Research by Qin et al. led to a smart foil, useful as a packaging in the food industry, consisting of chitosan / silver nanoparticles / purple corn extract that can change color depending on pH due to the high concentration of anthocyanins in purple corn [67].

Chitosan films were synthesized with sweet potato extracts, black rice and purple rice that can be used as antioxidant and intelligent materials to detect pH because they show visible color changes in order to extend shelf life and monitor food quality [55,68].

Moradi et al. used essential oils from the plant specific to Southwest Asia called *Zataria multiflora Boiss* and grape seed extracts in various proportions to obtain antioxidant films based on chitosan. Films incorporating both *Zataria multiflora Boiss* essential oil and grape seed extract showed increased antioxidant activity compared to the control sample containing only chitosan [69].

Studies to improve the antibacterial, antioxidant, physical and mechanical properties of chitosan films by incorporating ethanolic grape seed extract and *Ziziphora clinopodioides* essential oil confirm its use as an active biodegradable packaging [70].

For the recovery of industrial residues, pomegranate peel extract (*Punica granatum L.*) was incorporated in different concentrations in chitosan / gelatin gels to contribute to the significant increase of the structural and functional characteristics of the materials obtained from these mixtures [56]. The incorporation of the extract in the polymeric system has led to the improvement of the antioxidant activity due to the high content of phenolic compounds in the plant and to the increase of the viscosity which offers it the potential to be used as a material for food packaging.

Coatings based on chitosan and cranberry leaf extract can be used to extend shelf life and maintain a high nutritional value of fresh blueberries during storage after harvest [71]. The treated samples contained polyphenols and higher radical scavenging activity than the control. The phenolic compounds in the extracts were determined based on HPLC analysis. Syringic acid, well known for its antimicrobial activity, was the most present phenolic compound in leaf extracts. Leaf extracts showed antimicrobial activity against Gram-positive, Gram-negative bacteria and fungi.

The combination of mint extract with good antioxidant properties with chitosan which has excellent antimicrobial activity has led to a new preservative that extends the shelf life of meat and meat products [72].

Chitosan can be used as a carrier of active substances such as polyphenols extracted from chokeberry aronia to develop films with high antioxidant properties for active food packaging [73].

The study by Xu et al. aimed at obtaining films with various bioactive agents (hop extract, hop α-acids, or hop β-acids) incorporated in chitosan and gelatin. The physical, mechanical, optical, structural and barrier properties as well as the antioxidant and antibacterial activity were investigated. Bioactive agents improve the properties of films, which show better ultraviolet barrier properties than films without bioactive agents. Interactions between bioactive agents and polymers are supported by FTIR analysis. The films have antioxidant properties and antimicrobial action against Gram-positive bacteria. The films obtained can be used as organic packaging, to store fruit after harvest or as wound dressings due to the high degree of oxygen permeability [74].

Soares et al. investigated the effect of different concentrations of *Cenostigma nordestinum* extracts (leaves, bark and exudate bark) on the physicochemical, antioxidant and antimicrobial properties of chitosan films. The results show that bioactive chitosan films incorporated with *C. Nordestinum* extracts are quite promising for obtaining environmentally sustainable packaging for the food industry [75].

One of the recent trends is the development of materials obtained from food processing waste and their use as an alternative to plastic packaging. There are studies investigating the effect of various plant extracts on the properties of chitosan films to obtain biodegradable packaging.
In the study developed by Oberlintner et al. the biodegradability of the films obtained from chitosan with *Quercus* polyphenol extract in different soil types was monitored; films that have the potential for use as food packaging due to rapid degradation [76].

Another study on obtaining biodegradable films was conducted by Bigi et al. which introduced extracts of sage leaves and nettle into the matrix of chitosan/hydroxypropyl methylcellulose. SEM and FTIR analyzes confirmed the microstructure and compatibility between the polymer and the incorporated leaf extracts. With the addition of extracts, the films became opaquer, with increased water solubility and water vapor permeability compared to the control film. The films thus obtained can be used as an environmentally friendly alternative to the partial replacement of synthetic plastics and to extend the shelf life of food products [77].

Moalla et al. obtained active film packaging films based on chitosan and a mixture of hydroalcoholic extract, aqueous extract and *Artemisia campestris* essential oil. The structural properties of the films were evaluated using FTIR, XRD and SEM analyzes. The addition of A. campestris extracts to chitosan films significantly increased the antioxidant and UV-Vis barrier properties [78].

Kadam et al. studied the effect of extracts from pine needles containing curcuminoid pigments on the properties of chitosan films. Films with improved antioxidant properties by incorporating pine needle extracts with applicability in food packaging [79].

Agriculture

The effect of post-harvest treatment with chitosan and thyme essential oil on table grapes of the “Shahroudi” variety over a period of 90 days of storage was investigated [81]. The mixture of chitosan with essential oil has better effects in reducing fungal rot after harvest and maintaining the quality of table grapes compared to applying only the essential oil.

The study, published by Silva et al. shows the influence of vine treatment by spraying with chitosan solution and chitosan nanoparticles on the polyphenol content, antioxidant and antimicrobial activity. After the application of the treatment, ethanolic extracts were prepared from parts of the vine (stem, seeds and husks) which were analysed, registering a slight increase in the values of the vine treated with chitosan due to the antioxidant intake [82].

Environment

In recent decades, biodegradable polymers such as chitosan have been an alternative to petroleum-based products, and many extensive investigations have been conducted to minimize waste disposal. However, biopolymers face some constraints on their mechanical and structural barrier properties. Numerous studies have been performed to test the incorporation of several natural antioxidant compounds, both oil-based and aqueous, into the chitosan matrix to evaluate the effect on the physical properties of the resulting biopolymers [36,83].

Chitosan-based films are used as carriers of active compounds extracted from chokeberry to obtain biodegradable materials with increased antioxidant activity. The physical and chemical properties of chitosan films were investigated. Chitosan extract films showed better antioxidant properties than the control sample. Chokeberry extracts have improved the properties of UV-vis barrier and water vapor barrier films and reduced their oxygen permeability [73].

Conclusions

Chitosan remain a unite biomaterial by its properties and processed in promising of future materials. The main purpose of our review is to present the recent progress in the functionalised chitosan using different plants extract which show a remarkable antioxidant potential. Several methods were reported to prepare such biomaterials of functionalised chitosan with diversity in structure, analysed for physical characterisation, structural, morphological structure, mechanical and biological properties. This review is an attempt to show that functionalized chitosan with plants extract or oils are value-added for use in different modern technological fields. New studies are necessary to enlarge the number of functionalized chitosan-materials, to be characterized as potential biomaterials in many applications.

References

[1] C. Zhang, D. Hui, C. Du, H. Sun, W. Peng, X. Pu, Z. Li, J. Sun, C. Zhou, *Preparation and application of chitosan biomaterials in dentistry – Review*, **International Journal of Biological Macromolecules, 167,** 2021, pp.1198-1210.

[2] F. Furlani, A. Rossi, M.A. Grimaudo, G. Bassi, E. Giusto, F. Molinari, F. Lista, M. Montesi, S. Panseri, *Controlled liposome delivery from chitosan-based thermosensitive hydrogel for regenerative medicine*, **Int. J. Mol. Sci., 23(2)**, 2022, pp. 894, https://doi.org/10.3390/ijms23020894.

[3] S. Husain, K.H. Al-Samadani, S. Najeeb, M.S. Zafar, Z. Khurshid, S. Zohaib, S.B. Qasim, *Chitosan biomaterials for current and potential dental applications*, **Materials, 10,** 2017, pp. 602, https://doi.org/10.3390/ma10060602.

[4] L. Guo, Y. Guan, P. Liu, L. Gao, Z. Wang, S. Huang, L. Peng, Z. Zhao, *Chitosan hydrogel, as a biological macromolecule-based drug delivery system for exosomes and microvesicles in regenerative medicine: a mini review*, **Cellulose, 29**, 2022, pp. 1315–1330, https://doi.org/10.1007/s10570-021-04330-7.

[5] S. Cao, Y. Deng, L. Zhang, M. Aleahmad, *Chitosan nanoparticles, as biological macromolecule-based drug delivery systems to improve the healing potential of artificial neural guidance channels: A review,* **International Journal of Biological Macromolecules**, **201**, 2022, pp. 569-579.

[6] M. S. Ahsan, M. Thomas, K. K. Reddy, S. G. Sooraparaju, A. Asthana, I. Bhatnagar, *Chitosan as biomaterial in drug delivery and tissue engineering,* **International Journal of Biological Macromolecules, 110**, 2018, pp. 97-109.

[7] S. P. Bakshi, D. Selvakumar, K. Kadirvelua, N. S. Kumar, *Chitosan as an environment friendly biomaterial – a review on recent modifications and applications*, **International Journal of Biological Macromolecules**, **150**, 2020, pp. 1072-1083.

[8] G. D. Ivanova, Z. L. Yaneva, *Antioxidant properties and redox-modulating activity of chitosan and its derivatives: biomaterials with application in cancer therapy*, **BioResearch Open Access, 9(1),** 2020 pp. 64-72, http://doi.org/10.1089/biores.2019.0028.

[9] B. Tian, Y. Liu, J. Liu, *Chitosan-based nanoscale and non-nanoscale delivery systems for anticancer drugs: A review*, **European Polymer Journal, 154**, 2021, 110533.

[10] B. Sultankulov, D. Berillo, K. Sultankulova, T. Tokay, A. Saparov, *Progress in the development of chitosan-based biomaterials for tissue engineering and regenerative medicine*, **Biomolecules, 9(9),** 2019, pp. 470.

[11] M. M. Islam, M. Shahruzzaman, S. Biswas, M. N. Sakib, T. U. Rashid, *Chitosan based bioactive materials in tissue engineering applications-A review*, **Bioactive Materials 5(1)**, 2020, pp. 164–183.

[12] W. Wang, Q. Meng, Q. Li, J. Liu, M. Zhou, Z. Jin, K. Zhao, *Chitosan derivatives and their application in biomedicine*, **Int. J. Mol. Sci., 21(2),** 2020, pp. 487.

[13] F. S. Rezaei, F. Sharifianjazi, A. Esmaeilkhanian, E. Salehi, *Chitosan films and scaffolds for regenerative medicine applications: A review*, **Carbohydrate Polymers, 273**, 2021, 118631.

[14] R. A. Pandey, U. S. Singh, M. Momin, C. Bhavsar, *Chitosan: Application in tissue engineering and skin grafting*, **J Polym Res, 24(8)**, 2017, pp 1-22.

[15] H. Tan, R. Ma, C. Lin, Z. Liu, T. Tang, *Quaternized chitosan as an antimicrobial agent: antimicrobial activity, mechanism of action and biomedical applications in orthopedics*, **Int. J. Mol. Sci., 14(1),** 2013, 1854-1869, https://doi.org/10.3390/ijms14011854.

[16] E. Oyervides-Muñoz, E. Pollet, G. Ulrich, G. J. Sosa-Santillán, L. Avérous, *Original method for synthesis of chitosan-based antimicrobial agent by quaternary ammonium grafting*, **Carbohydrate Polymers, 157**, 2017, pp. 1922-1932.

[17] C. L. Ke, F. S. Deng, C. Y. Chuang, C. H. Lin, *Antimicrobial actions and applications of chitosan*, **Polymers, 13(6)**, 2021, pp. 904. https://doi.org/10.3390/polym13060904.

[18] R. E. Villegas-Rascón, M. Plascencia-Jatomea, E. C. Rosas-Burgos, Y. L. López-Franco, J. C. Tánori-Córdova, A. K. López-Meneses, M. O. Cortez-Rocha, *Chitosan / essential oils biocomposites for suppressing the growth of Aspergillus parasiticus*, **International Food Research Journal, 27(2),** 2020, pp. 316 – 326.

[19] A. M. L. Dans, M. V. C. Villarruz, C. A. Jimeno, M. A. U. Javelosa, J. Chua, R. Bautista, G. G. B. Velez, *The effect of momordica charantia capsule preparation on glycemic control in type 2 diabetes mellitus needs further studies*, **Journal of Clinical Epidemiology, 60(6),** 2007, pp. 554–559. https://doi.org/10.1016/j.jclinepi.2006.07.009.

[20] M. E. Abd El-Hack, M. T. El-Saadony, M. E. Shafi, N. M. Zabermawi, M. Arif, G. E. Batiha, A. F. Khafaga, Y. M. Abd El-Hakim, A. A. Al-Sagheer, *Antimicrobial and antioxidant properties of chitosan and its derivatives and their applications: A review*, **International Journal of Biological Macromolecules, 164**, 2020, pp. 2726–2744, https://doi.org/10.1016/j.ijbiomac.2020.08.153.

[21] E. Genskowsky, L. A. Puente, J. A. Pérez-Álvarez, J. Fernandez-Lopez, L. A. Muñoz, M. Viuda-Martos, *Assessment of antibacterial and antioxidant properties of chitosan edible films incorporated with maqui berry (Aristotelia Chilensis)*, **LWT - Food Science and Technology, 64(2)**, 2015, pp. 1057–1062, https://doi.org/10.1016/j.lwt.2015.07.026.

[22] I. Gulcin, *Antioxidants and antioxidant methods: An updated overview*, **Archives of Toxicology, 94(3),** 2020, pp. 651-715. https://doi.org/10.1007/s00204-020-02689-3.

[23] M. N. Alam, N. J. Bristi, M. Rafiquzzaman, *Review on in vivo and in vitro methods evaluation of antioxidant activity*, **Saudi Pharmaceutical Journal, 21(2),** 2013, pp. 143–152. https://doi.org/10.1016/j.jsps.2012.05.002.

[24] I. G. Munteanu, C. Apetrei, *Analytical methods used in determining antioxidant activity: A review*, **International Journal of Molecular Sciences, 22(7),** 2021, 3380. https://doi.org/10.3390/ijms22073380.

[25] L. Diniz Do Nascimento, A. A. Barbosa de Moraes, K. Santana da Costa, J. M. P. Galúcio, P. S. Taube, C. M. L. Costa, J. N. Cruz, E. H. de Aguiar Andrade, L. J. Guerreiro de Faria, *Bioactive natural compounds and antioxidant activity of essential oils from spice plants: new findings and potential applications*, **Biomolecules, 10(7),** 2020, pp. 1–37, https://doi.org/10.3390/biom10070988.

[26] F. Shahidi, P. Ambigaipalan, *Phenolics and polyphenolics in foods, beverages and spices: antioxidant activity and health effects - A Review*, **Journal of Functional Foods, 18**, 2015, pp. 820–897, https://doi.org/10.1016/j.jff.2015.06.018.

[27] S. A. Yang, S. K. Jeon, E. J. Lee, C. H. Shim, I. S. Lee, *Comparative study of the chemical composition and antioxidant activity of six essential oils and their components*, **Natural Product Research, 24(2),** 2010, 140–151, https://doi.org/10.1080/14786410802496598.

[28] Z. S. Ilić, L. Milenković, N. Tmušić, L. Stanojević, J. Stanojević, D. Cvetković, *Essential oils content, composition and antioxidant activity of lemon balm, mint and sweet basil from Serbia*, **Lwt, 153**, 2022, pp. 112210, https://doi.org/10.1016/j.lwt.2021.112210.

[29] F. Balanescu, M. D. Ionica Mihaila, G. Cârâc, B. Furdui, C. Vînătoru, S. M. Avramescu, E. L. Lisa, M. Cudalbeanu, R. M. Dinica, *Flavonoid profiles of two new approved romanian ocimum hybrids*, **Molecules, 25(19)**, 2020, pp. 4573, https://doi.org/10.3390/molecules25194573.

[30] A. Ghasemi Pirbalouti, F. Malekpoor, A. Salimi, A. Golparvar, *Exogenous application of chitosan on biochemical and physiological characteristics, phenolic content and antioxidant activity of two species of basil (Ocimum Ciliatum and Ocimum Basilicum) under reduced irrigation*, **Scientia Horticulturae, 217**, 2017, pp. 114–122, https://doi.org/10.1016/j.scienta.2017.01.031.

[31] A. Giordano, P. Morales-Tapia, M. Moncada-Basualto, J. Pozo-Martínez, C. Olea-Azar, A. Nesic, G. Cabrera-Barjas, *Polyphenolic composition and antioxidant activity (orac, epr and cellular) of different extracts of argylia radiata vitroplants and natural roots*, **Molecules, 27(3),** 2022, pp. 610, https://doi.org/10.3390/molecules27030610.

[32] A. Ermoshin, I. Kiseleva, I. Nikkonen, S. Duan, *Chemical composition and antioxidant activity of two fungi species from the genus Ganoderma*, **AIP Conference Proceedings 2390(1),** 2022, pp. 030017.

[33] C. D. J. P. Franco, O. O. Ferreira, Â. Antônio Barbosa de Moraes, E. L. P. Varela, L. D. D. Nascimento, S. Percário, M. Santana de Oliveira, E. H. D. A. Andrade, *Chemical Composition and Antioxidant Activity of Essential Oils from Eugenia Patrisii Vahl, E. Punicifolia (Kunth) Dc., and Myrcia Tomentosa (Aubl.) Dc., Leaf of Family Myrtaceae*, **Molecules, 26(11),** 2021, https://doi.org/10.3390/molecules26113292.

[34] U. Siripatrawan and B. R. Harte, *Physical properties and antioxidant activity of an active film from chitosan incorporated with green tea extract*, **Food Hydrocolloids, 24(8),** 2010, pp. 770–775. https://doi.org/10.1016/j.foodhyd.2010.04.003.

[35] S. B. Schreiber, J. J. Bozell, D. G. Hayes, S. Zivanovic, *Introduction of primary antioxidant activity to chitosan for application as a multifunctional food packaging material*, **Food Hydrocolloids, 33(2),** 2013, pp. 207–214. https://doi.org/10.1016/j.foodhyd.2013.03.006.

[36] M. Yousefi, E. Khanniri, M. Shadnoush, N. Khorshidian, A. M. Mortazavian, *Development, characterization and in vitro antioxidant activity of chitosan-coated alginate microcapsules entrapping Viola odorata Linn. extract*, **International Journal of Biological Macromolecules, 163,** 2020, pp. 44–54. https://doi.org/10.1016/j.ijbiomac.2020.06.250.

[37] D. Tungmunnithum, P. Kongsawadworakul, C. Hano, *A cosmetic perspective on the antioxidant flavonoids from Nymphaea lotus L.*, **Cosmetics, 8(1),** 2021, pp. 1–9, https://doi.org/10.3390/cosmetics8010012.

[38] M. Cudalbeanu, I. O. Ghinea, B. Furdui, D. Dah-nouvlessounon, R. Raclea, T. Costache, I. E. Cucolea, F. Urlan, R. M. Dinica, *Exploring new antioxidant and mineral compounds from Nymphaea alba wild-grown in Danube Delta biosphere*, **Molecules, 23(6),** 2018, pp. 1–16. https://doi.org/10.3390/molecules23061247.

[39] E. Capecka, A. Mareczek, M. Leja, *Antioxidant activity of fresh and dry herbs of some lamiaceae species*, **Food Chemistry, 93(2),** 2005, pp. 223–226, https://doi.org/10.1016/j.foodchem.2004.09.020.

[40] A. Wojdyło, J. Oszmiański, R. Czemerys, *Antioxidant activity and phenolic compounds in 32 selected herbs*, **Food Chemistry, 105(3),** 2007 pp. 940–949, https://doi.org/10.1016/j.foodchem.2007.04.038.

[41] A. Kostecka-Gugała, M. Kruczek, I. Ledwożyw-Smoleń, P. Kaszycki, *Antioxidants and health-beneficial nutrients in fruits of eighteen Cucurbita cultivars: analysis of diversity and dietary implications*, **Molecules, 25(8),** 2020, pp. 1792.

[42] K. Wolfe, X. Wu, R. H. Liu, *Antioxidant activity of apple peels*, **Journal of Agricultural and Food Chemistry, 51 (3),** 2003, pp. 609–614, https://doi.org/10.1021/jf020782a.

[43] A. Cazanevscaia Busuioc, A. V. Dediu Botezatu, B. Furdui, C. Vinatoru, F. Maggi, G. Caprioli, R. M. Dinica, *Comparative study of the chemical compositions and antioxidant activities of fresh juices from romanian Cucurbitaceae varieties*, **Molecules, 25(22),** 2020, https://doi.org/10.3390/molecules25225468.

[44] A. Carac, R. Boscencu, S. Patriche, R. M. Dinica, G. Carac, C. E. Gird, *Antioxidant and antimicrobial potential of extracts from Aloe Vera leaves*, **Revista de Chimie, 67(4),** 2016, pp. 654–658.

[45] J. Samoticha, A. Wojdyło, K. Lech, *The influence of different the drying methods on chemical composition and antioxidant activity in Chokeberries*, **Lwt, 66,** 2016, pp. 484–489, https://doi.org/10.1016/j.lwt.2015.10.073.

[46] N. V. Majeti, R. Kumar, *A review of chitin and chitosan applications*, **Reactive and Functional Polymers, 46(1),** 2000, pp. 1-27.

[47] B. Evros, D. Aycan, N. Alemdar, Production of ciprofloxacin loaded chitosan/gelatin/bone ash wound dressing with improved mechanical properties, **Carbohyd. polym., 222,** 2019, pp. 115007.

[48] M. B. Lemes, A. Novatski, P. C. Ferrari, B. R. Minozzo, A. D. S. Justo, V. E. K. Petry, J. C. R. Vellosa, S. D. R. F. Sabino, J. V. Gunha, L. A. Esmerino, F. L. Beltrame, *Physicochemical, biological and release studies of chitosan membranes incorporated with Euphorbia umbellata fraction*, **Revista Brasileira de Farmacognosia, 28(4),** 2018, pp 433-443.

[49] M. Kaya, S. Khadem, Y. S. Cakmak, M. Mujtaba, S. Ilk, L. Akyuz, A. M. Salaberria, J. Labidi, A. H. Abdulqadir, E. Deligöz, *Antioxidative and antimicrobial edible chitosan films blended with stem, leaf and seed extracts of Pistacia terebinthus for active food packaging*, **RSC Advances, 8**, 2018, pp. 3941-3950.

[50] F. J. Rubilar, R. M. S. Cruz, H. D. Silva, A. A. Vicente, I. Khmelinskii, M. C. Vieira, *Physico-mechanical properties of chitosan films with carvacrol and grape seed extract*, **Journal of Food Engineering, 115(4),** 2013, pp. 466-474.

[51] T. Revathi, S. Thambidurai, *Synthesis of chitosan incorporated neem seed extract (Azadirachta indica) for medical textiles,* **International Journal of Biological Macromolecules, 104**, 2017, pp. 1890–1896.

[52] S. S. Silva, E. G. Popa, M. E. Gomes, M. Cerqueira, A. P. Marques, S. G. Caridade, P. Teixeira, C. Sousa, J. F. Mano, R. L. Reis, *An investigation of the potential application of chitosan/aloe-based membranes for regenerative medicine*, **Acta Biomaterialia, 9(6)**, 2013, pp. 6790-6797.

[53] J.S. Boateng, K.H. Matthews, H.N.E. Stevens, G.M. Eccleston, *Wound healing dressings and drug delivery systems: A review,* **J. Pharm. Sci., 97,** 2008, pp. 2892-2923.

[54] P. Bowler, B. Duerden, D. Armstron, *Wound microbiology and associated approaches to wound management*, **Clin. Microbiol. Rev., 14**, 2001, pp. 244-269.

[55] H. Yong, X. Wang, R. Bai, Z. Miao, X. Zhang, J. Liu, *Development of antioxidant and intelligent pH-sensing packaging films by incorporating purple-fleshed sweet potato extract into chitosan matrix,* **Food Hydrocolloids, 90**, 2019, pp. 216-224.

[56] M. R.V. Bertolo, V. C. A. Martins, M. M. Horn, L. B. Brenelli, A. M. G. Plepis, *Rheological and antioxidant properties of chitosan/gelatin-based materials functionalized by pomegranate peel extract*, **Carbohydrate Polymers, 228**, 2020, 115386.

[57] T. Liu, L. Liu, X. Gong, F. Chi, Z. Ma, *Fabrication and comparison of active films from chitosan incorporating different spice extracts for shelf life extension of refrigerated pork*, **LWT, 135**, 2021, 110181.

[58] S. B. D. Silva, M. Amorim, P. Fonte, R. Madureira, D. Ferreira, M. Pintado, B. Sarmento, *Natural extracts into chitosan nanocarriers for rosmarinic acid drug delivery*, **Pharm. Biol., 53(5)**, 2015, pp. 642–652.

[59] S. Chandrasekar, S. Vijayakumar, R. Rajendran, *Application of chitosan and herbal nanocomposites to develop antibacterial medical textile,* **Biomedicine & Aging Pathology 4(1),** 2014, pp. 59-64.

[60] M. Abbas, T. Hussain, M. Arshad, A. R. Ansari, A. Irshad, J. Nisar, F. Hussain, N. Masood, A. Nazir, M. Iqbal, *Wound healing potential of curcumin cross-linked chitosan/polyvinyl alcohol,* **International Journal of Biological Macromolecules, 140**, 2019, pp. 871–876.

[61] I. Yousefi, M. Pakravan, H. Rahimi, A. Bahador, Z. Farshadzadeh, I. Haririan, *An investigation of electrospun Henna leaves extract-loaded chitosan based nanofibrous mats for skin tissue engineering,* **Materials Science and Engineering: C, 75**, 2017, pp. 433-444

[62] V. G. L. Souza, A. L. Fernando, J. R. A. Pires, P. F. Rodrigues, A.A.S. Lopes, F. M. B. Fernandes, *Physical properties of chitosan films incorporated with natural antioxidants*, **Industrial Crops&Products, 107**, 2017, pp. 565-572.

[63] V. G. L. Souza, P. F. Rodrigues, M. P. Duarte, A. L. Fernando, *Antioxidant migration studies in chitosan films incorporated with plant extracts*, **J. Renew. Mater., 6(5),** 2018, 548-58.

[64] A. M. Abdelghany, A. A. Menazea, A. M. Ismail, *Synthesis, characterization and antimicrobial activity of Chitosan/Polyvinyl alcohol blend doped with Hibiscus Sabdariffa L. extract,* **Journal of Molecular Structure, 1197**, 2019, pp. 603-609.

[65] S. E. Quintana, O. Llalla, M. R.García-Risco, T. Fornari, *Comparison between essential oils and supercritical extracts into chitosan-based edible coatings on strawberry quality during cold storage,* **The Journal of Supercritical Fluids, 171**, 2021, pp. 105198.

[66] M. S. Nair, A. Saxena, C. Kaur, *Effect of chitosan and alginate based coatings enriched with pomegranate peel extract to extend the postharvest quality of guava (Psidium guajava L.)*, **Food Chemistry, 240**, 2018, pp. 245-252.
[67] Y. Qin, Y. Liu, L. Yuan, H. Yong, J. Liu, *Preparation and characterization of antioxidant, antimicrobial and pH-sensitive films based on chitosan, silver nanoparticles and purple corn extract*, **Food Hydrocolloids, 96**, 2019, pp. 102-111.
[68] H. Yong, J. Liu, Y. Qin, R. Bai, X. Zhang, J. Liu, *Antioxidant and pH-sensitive films developed by incorporating purple and black rice extracts into chitosan matrix*, **International Journal of Biological Macromolecules, 137**, 2019, pp. 307-316.
[69] M. H. T. Moradi, S. M. R. Rohani, A. R. Oromiehie, H. Malekinejad, J. Aliakbarlu, M. Hadian, *Characterization of antioxidant chitosan film incorporated with Zataria multiflora Boiss essential oil and grape seed extract*, **LWT - Food Science and Technology, 46(2),** 2012, pp. 477-484.
[70] Y. Shahbazi, *The properties of chitosan and gelatin films incorporated with ethanolic red grape seed extract and Ziziphora clinopodioides essential oil as biodegradable materials for active food packaging*, **International Journal of Biological Macromolecules, 99**, 2017, pp. 746–753.
[71] G. Yang, J. Yue, X. Gong, B. Qian, H. Wang, Y. Deng, Y. Zhao, *Blueberry leaf extracts incorporated chitosan coatings for preserving postharvest quality of fresh blueberries*, **Postharvest Biology and Technology, 92,** 2014, pp. 46–53.
[72] S. R. Kanatt, R. Chander, A. Sharma, *Chitosan and mint mixture: A new preservative for meat and meat products*, **Food Chemistry, 107**, 2008, pp. 845–852.
[73] S. Sady, A. Błaszczyk, W. Kozak, P. Boryło, M. Szindler, *Quality assessment of innovative chitosan-based biopolymers for edible food packaging applications*, **Food Packaging and Shelf Life, 30,** 2021, pp. 100756.
[74] D. Xu, T. Chen, Y. Liu, *The physical properties, antioxidant and antimicrobial activity of chitosan–gelatin edible films incorporated with the extract from hop plant*, **Polymer Bulletin, 78**, 2021, pp. 3607–3624, https://doi.org/10.1007/s00289-020-03294-1.
[75] J. M. A. Soares, E. D. da Silva Júnior, B. Oliveira de Veras, R. Yara, P. B. Sales de Albuquerque, M. Pessoa de Souza, *Active biodegradable film based on chitosan and Cenostigma Nordestinum' extracts for use in the food industry*, **Journal of Polymers and the Environment, 30**, 2022, pp. 217-231, https://doi.org/10.1007/s10924-021-02192-5.
[76] A. Oberlintner, M. Bajić, G. Kalčíková, B. Likozar, U. Novak, *Biodegradability study of active chitosan biopolymer films enriched with Quercus polyphenol extract in different soil types*, **Environmental Technology & Innovation, 21,** 2021, pp. 101318.
[77] F. Bigi, H. Haghighi, H. W. Siesler, F. Licciardello, A. Pulvirenti, *Characterization of chitosan hydroxypropyl methylcellulose blend films enriched with nettle or sage leaf extract for active food packaging applications*, **Food Hydrocolloids, 120**, 2021, pp. 106979.
[78] S. Moalla, I. Ammar, M. L. Fauconnier, S. Danthine, C. Blecker, S. Besbes, H. Attia, *Development and characterization of chitosan films carrying Artemisia campestris antioxidants for potential use as active food*, **International Journal of Biological Macromolecules, 183,** 2021, pp. 254-266.
[79] A. A. Kadam, S. Singh, K. K. Gaikwad, *Chitosan based antioxidant films incorporated with pine needles (Cedrus deodara) extract for active food packaging applications*, **Food Control, 124**, 2021, pp. 107877.

[80] X. Zhang, H. Lian, J. Shi, W. Meng, Y. Peng, *Plant extracts such as pine nut shell, peanut shell and jujube leaf improved the antioxidant ability and gas permeability of chitosan films*, **International Journal of Biological Macromolecules, 148**, 2020, pp. 1242-1250.

[81] M. Dehestani Ardakani, Y. Mostofi, *Postharvest application of chitosan and Thymus essential oil increase quality of the table grape cv. 'Shahroudi'*, **Journal of horticulture and postharvest research, 2(1),** 2019, 31-42.

[82] V. Silva, R. K. Singh, N. Gomes, B. G. Soares, A. Silva, V. Falco, R. Capita, C. Alonso-Calleja, J. E. Pereira, J. S. Amaral, G. Igrejas, P. Poeta, *Comparative insight upon chitosan solution and chitosan nanoparticles application on the phenolic content, antioxidant and antimicrobial activities of individual grape components of sousão variety,* **Antioxidants, 9(2),** 2020, pp. 178, https://doi.org/10.3390/antiox9020178.

[83] T. Liu, L. Liu, X. Gong, F. Chi, Z. Ma, *Fabrication and comparison of active films from chitosan incorporating different spice extracts for shelf life extension of refrigerated pork*, **LWT, 135,** 2021, pp. 110181.

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