International Journal of Advances in Engineering and Management (IJAEM)
Volume 6, Issue 02 Feb 2024, pp: 516-521 www.ijaem.net ISSN: 2395-5252

Analysis of the solubility of edible films based on nopal mucilage (Opuntia ficusindica): the modulating effect of pH.

Montalvo-Paquini C[1]., Lozada-Lechuga J[1]. Alvarado-Castillo M. G[1]., Borráz-Argüello M. T[1].

[1,]*Universidad Politécnica de Puebla, tercer carril del ejido serrano s/n, 72640 Juan C. Bonilla, Puebla, México.*
Corresponding Author: claudia.montalvo@uppuebla.edu.mx

Date of Submission: 20-02-2024 Date of Acceptance: 04-03-2024

ABSTRACT: The mucilage was extracted from fresh cladodes of Opuntia ficusindica and utilized for the preparation of edible films, additionally formulated with sodium alginate and glycerol. The effect of pH-adjusted nopal mucilage edible film formulations at different pH values (6.0, 9.0, and 11.0) on the solubility percentage of the films was evaluated. A second-order nonlinear response effect with positive concavity was observed, where the formulation at pH 9 exhibited lower water solubility (96.5%), while at more alkaline pH values (11.0), solubility values significantly increased ($p< 0.05$). It is relevant to assess the pH effect of mucilage in films, optimizing the physical properties of films formulated based on nopal mucilage.

KEYWORDS: Solubility, pH, Opuntiaficusindica, edible films.

I. INTRODUCTION

[1].Edible films are thin coatings, approximately 3mm in thickness, made from biopolymers that are ingestible and have versatile applications in the food, pharmaceutical, and packaging industries.
[2].These materials offer significant advantages over traditional packaging: they act as effective barriers against moisture loss, oxygen absorption, and the release of food volatile components, thereby enhancing shelf life.
[3].Moreover, being derived from renewable natural sources and completely biodegradable, they represent eco-friendly packaging solutions that promote the circular economy and mitigate the generation of contaminating plastic waste.
[4].Edible films are crafted from a variety of natural biopolymers, including proteins (collagen, casein, gluten), polysaccharides (starch, cellulose and derivatives, chitosan, alginate), and lipids (waxes, fatty acids). These biologically sourced compounds form structured matrices capable of creating thin edible coatings with desirable barrier, mechanical, and optical properties for food and pharmaceutical applications.
[5].Additionally, the functionality of these biodegradable films can be enhanced through the controlled incorporation of nanoparticles, plasticizers, and other additives.
[6].The surge in research on innovative materials based on biopolymers is focused on developing eco-friendly films to replace contaminating synthetic plastics, thus reducing the environmental impact of food and product packaging.
[7].Water solubility is considered one of the fundamental parameters for characterizing the quality and effectiveness of edible films intended for food and pharmaceutical uses. Controlled solubility provides them with mechanical strength as a protective barrier in aqueous environments, while also allowing for their disintegration to release nutrients or drugs when needed.
[8].Highly soluble films would have limited functionality as protective coatings, whereas completely insoluble formulations would not release encapsulated active components. Solubility is also closely related to other physicochemical properties such as water vapor permeability, transparency, and texture, which determine the specific application of these films in food.
[9].The pH is a parameter that indicates the acidity or alkalinity of an aqueous solution. This value regulates the ionization and thus the electric charge of ionizable functional groups of macromolecules such as the biopolymers used in edible films.
[10].The protonation or deprotonation of groups such as amines, amides, and carboxyl in proteins, as well as the hydroxylation of polysaccharides, is strongly influenced by changes in pH. The resulting charge modifies the electrostatic interactions

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between biopolymer chains and their miscibility, affecting the three-dimensional structure of film matrices and their consequent final properties, such as their solubilization kinetics in aqueous media.

[11].Determining how the pH value of the mucilage used in formulations affects the solubility properties of the resulting films would allow for a better understanding and prediction of their functionality for practical applications. Establishing this relationship would aid in the development of edible films with controlled barrier properties and nutrient release, according to specific product requirements. Understanding these effects would guide the formulation and processing of films with tailor-made solubilities, enhancing their final performance in food systems.

[12].The pH of the nopal mucilage in the formulation would modulate the interactions between polymeric chains during gelation, drying, and structural matrix formation, determining critical solubility parameters such as kinetics, degree of swelling, dispersibility, or film integrity.

[13].The nopal mucilage (Opuntia ficus-indica) has proven to be an attractive biopolymeric source for the development of innovative edible films, owing to its film-forming properties.

[14, 15].The heteropolymeric composition of its galacturonans, arabinogalactans, and xyloglucans endows it with excellent functional properties, as well as modifiable solubility. To enhance this property, other biopolymers such as gelatin and alginate are used in the formulation of nopal films. These agents are employed as thickeners and texturizers in the food industry. Due to their good gelling properties, they have a high capacity to form edible films that generally exhibit acceptable solubility properties.

[16].The nopal mucilage is obtained from an abundant and renewable source found in many arid regions of Mexico, where 90% of this resource is produced, requiring minimal nutrients for its growth. It is non-toxic, biodegradable, and biocompatible, characteristics that confer advantages as an alternative for sustainable food preservation and packaging. In the case of nopal, mucilage is an undesirable byproduct which, combined with the high demand from consumers for biodegradable products, results in a potential source for the generation of films and coatings.

The variations in pH during the formulation of edible films based on nopal (Opuntia ficus-indica) would modify the electrostatic interactions between the biopolymers comprising its matrix, primarily mucilage such as acidic galacturonans and proteinaceous arabinogalactans, thereby altering the structural properties and consequent solubility of the resulting films.

The importance of the solubility properties of films made with this polymer lies in understanding how variations in the pH value of the mucilage can affect the solubility of the formed polymer, thereby inferring its potential use as packaging in the food industry.

II. METHODS

Raw materials.

The Nopal (Opuntia ficus-indica) plant material used in this study was acquired from the municipal market of San Pedro Cholula, Puebla.

Mucilage extraction.

To extract the nopal mucilage, fresh spineless cladodes were used. These were washed, brushed, cut into 1x1 cm pieces, and mixed with distilled water in a ratio of 1:1 (w/v). The mixture was ground for 2 minutes and filtered. The filtrate was then subjected to a water bath at 80°C for 2 hours, followed by centrifugation at 3500 rpm for 20 minutes to separate the mucilage.

Film production

To prepare the edible films, the nopal mucilage were adjusted to pH values of 6.0, 9.0, and 11.0 using 3N KOH. Film-forming solutions were prepared with 1.5% (w/v) sodium alginate, 5.5% (w/v) glycerol, and a proportion of raw nopal mucilage at 15% (w/v) in distilled water. The solutions were homogenized for 5 minutes at 10,000 rpm and subjected to a temperature of 121°C for 15 minutes. The Casting method was used to prepare the films, with solvent removal by drying at 45°C for 24 hours. The generated films were stored in a system with water activity of 0.46 for a minimum of 24 hours before evaluating their properties.

Watersolubility determination.

[17]Samples of 2x2 cm film were used, and their initial dry weight (W1) was recorded. The samples were placed in vessels with 40 ml of distilled water with 0.01% (w/v) potassium sorbate and subjected to agitation at 100 rpm for 24 hours at 25°C. Subsequently, the residue was filtered through filter paper and dried at 90°C for 24 hours to record its final dry weight (W2). Total solubility was calculated using the equation:

Total Solubility (%) = ((W1 – W2) / W1) x 100.

Statistical analysis

An ANOVA was conducted considering the pH of the formulation as a factor, with the percentage of total solubility as the response

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variable. Significant differences between means (α=0.05) were analyzed, and the variables were correlated by fitting a second-order nonlinear model.

III. RESULTS

For the present study, edible films were prepared using a formulation based on nopal mucilage, where the dependent variable corresponded to the adjusted pH value at three treatment levels, ranging from slightly acidic (pH = 6.0) to moderately alkaline values (pH = 11.0), with an intermediate point (pH = 9.0). The measured dependent variable corresponded to the percentage of film solubility in water. Each experiment was conducted using four replicates.

The results of the study (Figure1) demonstrate a non-linear dependency effect, fitting a second-order curve with positive concavity ($p<0.05$). Based on the results, a second-order polynomial fitting model was constructed, where the equation is as follows: $y = 0.251x^2 - 4.385x + 115.57$; where y corresponds to the percentage of solubility; x corresponds to the pH value.

The films formulated with nopal mucilage adjusted to an acidic pH of 6.0 and 11.0 achieved the highest solubility values (98.3 ± 0.26% and 97.7 ± 0.22%, respectively), while lower values were observed in films with mucilage at pH 9 (96.4 ± 0.20%). The results indicate that at extreme pH values, the ionization of carboxyl groups in the mucilage would be favored, increasing intermolecular electrostatic repulsions. This would prevent associations between polymer chains and the formation of a cohesive network, resulting in higher solubility of the films in aqueous media. In contrast to what was observed at pH 9, where we can infer that there is less repulsion between the polymer chains, generating a more stable three-dimensional network.

[18].Medina-Torres et al. reported that nopal mucilage at a pH higher than 9.0 decreased its gelation properties, attributing it to strong repulsion caused by the electrostatic charges of sugars when they become ionizable, resulting in an expanded molecule that forms loose gels retaining large amounts of water.

[19].This phenomenon is also noted by Nieto, who indicates that in anionic polymers, the presence of ionized groups increases their polarity and weakens the intermolecular associations between polymer chains due to repulsion. However, when molded and dried, they retain their structure, forming good films.

In addition to the above, the high solubility values obtained in the films may also be related to the hydrophilic nature of nopal mucilage.

[20].The nopal mucilage is an anionic polymer with a high content of neutral sugars such as arabinose (35-44%), galactose (20-29%), glucose (2.5%), xylose (22.8%), and fructose, and acidic sugars such as galacturonic acid (6.4%). The neutral sugars form highly branched chains.

[19].The presence of multiple side chains along the central chain generates a steric effect that negatively affects the intermolecular association of the polymer. Additionally, the negative charge of the molecules prevents the formation of a tight or compact structure in the dry state, allowing the polymers to easily absorb water during the hydration process. Depending on the extent and distribution of the charges of the molecules in the polymer, the film can be completely soluble.

[21].In addition to the above, it has been reported that nopal mucilage can contain slightly over 50% of low molecular weight sugars (2.4 x 102 Da). This can significantly contribute to the formation of a weak three-dimensional structure that easily dissolves in aqueous solutions.

The reported solubility values for nopal mucilage films can vary widely, depending on the components of the formulation and the film-making conditions.

[22].González-Sandoval et al., who conducted a mucilage extraction process like that described in the methodology, found values ranging from 81.26% to 91.03% for different Opuntia cultivars in nopal mucilage films with pectin.

[23].On the other hand, in nopal mucilage and potato starch films prepared at a pH of 8.7, solubility values ranging from 24.03% to 51.65% were obtained, thanks to the retrogradation properties exhibited by the starch.

[24,25].To improve the physical and chemical properties of nopal mucilage films, various authors have added various polymers to the formulations such as gelatin, starch, pectin, and alginate.

In this work, sodium alginate was added to the formulation. This polymer favored the texture of the films because, being a linear unbranched anionic polymer, it forms strong but flexible films, unlike if only nopal mucilage were used. Since nopal mucilage is a highly branched polymer, the films were very brittle.

[19].Alginate is primarily composed of mannuronic acid and guluronic acid. Its carboxyl groups make alginate highly soluble in water, likely contributing to the high solubility values obtained.

Furthermore, glycerol used as a plasticizer has hydroxyl groups in its structure that could also favor water retention in the polymeric matrix.

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[26].It has been observed that the type of plasticizer used in the preparation of nopal mucilage films influences solubility properties. When glycerol is used, solubility values are higher than those obtained with sorbitol.

This is because glycerol, due to its low molecular weight, can easily insert itself into the polymer chains, leading to a decrease in interactions between polymer chains in favor of plasticizer-polymer interactions. Additionally, due to its hydrophilic nature, glycerol readily retains water.

As a complement to the current analysis, a one-way analysis of variance (ANOVA) was conducted, demonstrating the presence of significant differences ($p<0.05$) in the solubility of edible films formulated with nopal mucilage at different pH values. These results suggest that pH is a factor that significantly affects the quality of edible films, ruling out the presence of random effects. Table 1 summarizes the findings from the ANOVA test for the different treatments. Consequently, it is verified that film formulation by controlling pH values allows for the regulation of solubility properties in these edible matrices, presenting a strategy for the development of plant-based bio-packaging with specific properties.

In this case, by obtaining films with high solubility, they could be used to create coatings that can be ingested along with the food, or they can be used to package ingredients where the film needs to completely dissolve when subjected to aqueous preparations.

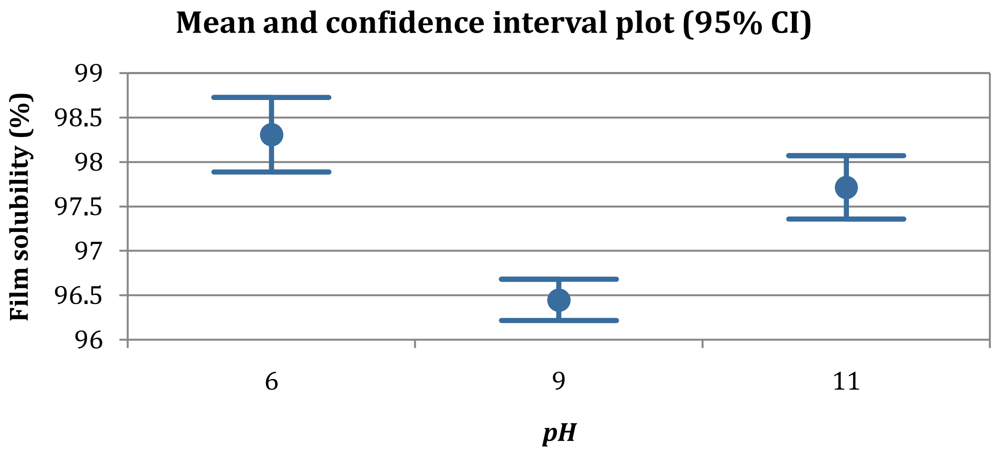

FIGURE 1. GRAPHICAL REPRESENTATION DEPICTING THE MEANS AND CONFIDENCE INTERVALS OF FILM SOLUBILITY IN CONTRAST OF OPUNTIA'S MUCILAGE pHVALUE.

ANOVA

Sourceof Variación	d.f.	SS	MS	F	p-value
Between Groups	2	7.2317	3.6159	76.86	2.20E-06
WithinGroups	9	0.4234	0.0470		
Total	11	7.65			

TABLE 1. ONE-WAY ANOVA TEST AMONGFILM SOLUBILITYUNDER THE EFFECT OFpH.

IV. CONCLUSION

The heightened global concern over the increase in non-biodegradable plastic waste has prompted the development of new packaging materials from sustainable raw materials to emerge as an interesting alternative. Nopal mucilage represents a promising option to produce edible films, particularly in regions with high production rates at low cost, such as Mexico.

In this study, highly soluble edible films were obtained from nopal mucilage with alginate. The pH adjustment in the nopal mucilage, used in film-forming solutions, has a significant effect on the solubility of the resulting films. Edible films with solubility values exceeding 97% were achieved when the mucilage pH was adjusted to an alkaline pH (11.0). It is important to evaluate the conditions affecting the film properties as they will define their final application. Exploring ways in which mucilage can be incorporated into everyday foods aims to add value to nopal by-products that would otherwise be considered waste.

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