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Gelatin-Based Hydrogels Containing Microcrystalline and Nanocrystalline Cellulose as Moisture Absorbers for Food Packaging Applications

Vanessa Acevedo-Puello [1], Kelly J. Figueroa-López [1,2] and Rodrigo Ortega-Toro [2,*]

[1] Food Packaging and Shelf Life Research Group (FP&SL), Food Engineering Department, Universidad de Cartagena, Cartagena de Indias 130001, Colombia; vacevedop@unicartagena.edu.co (V.A.-P.); kjfigueroal@unal.edu.co (K.J.F.-L.)
[2] Be Circular Consulting, Calle 21 16B 35, San Juan de Arama 504047, Colombia
* Correspondence: rortegap1@unicartagena.edu.co

Abstract: Sustainable hydrogels are an innovative biodegradable alternative to traditional packaging materials. They offer exceptional water absorption capacity and high biocompatibility, making them ideal food absorbents to reduce plastic waste, extend shelf life and ensure the safety and quality of packaged foods. In this study, hydrogels based on gelatin, microcrystalline cellulose (MCC), and nanocrystalline cellulose (NCC) were developed, characterized, and applied in the packaging of chicken breasts. For this, MCC was isolated from the banana pseudostem and commercial NCC was incorporated into a gelatin solution to produce the hydrogel materials by film casting. The resulting hydrogels were analyzed in terms of morphology, structural properties, water absorption capacity, mechanical strength, and color properties. The results showed that the incorporation of MCC and NCC significantly improved the mechanical integrity of the hydrogels, which prevented premature deformation of the hydrogels when they absorbed moisture. In addition, changes in the color properties of chicken breast samples in contact with the hydrogels were observed, indicating their ability to preserve food quality. Subsequently, the effectiveness of the hydrogels for chicken breast storage at 4 °C for 4 days was validated. The results demonstrated that the hydrogels developed in this study are a sustainable and environmentally friendly alternative to traditional packaging materials that can extend the shelf life of food products while maintaining their physical and microbiological integrity.

Keywords: mechanical properties; water absorption; chicken; exudates; swelling; shelf life

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1. Introduction

The need for sustainable and eco-friendly materials in food packaging design and food preservation has become increasingly critical in recent years. Packaging plays a crucial role in ensuring food security by protecting and preserving food throughout the supply chain [1]. Adequate packaging prevents spoilage, contamination, and physical damage, extending the shelf life of food products and making them more accessible and available to consumers. Traditional packaging materials, such as plastics derived from fossil fuels, have raised concerns due to their negative environmental impact and potential health risks [2].

The United Nations Environment Program (UNEP), in its resolution 5/14, recognizes plastic pollution as a major environmental problem affecting global sustainable development [3]. Recent studies have detected micro- and nano-scale plastic particles in human blood from materials commonly used in beverage bottles and food packaging [4]. Previous research has already detected microplastics in the ecosystem, food and drink, feces, and human placenta [5–9]. As a result, there is a growing interest in the development and utilization of renewable materials for food packaging applications.

In this context, hydrogels, three-dimensional networks of hydrophilic polymers, have gained significant attention in various industries, such as in biomedicine, cosmetics, pharmacology, agriculture, and food [10,11]. These materials offer unique advantages for food packaging, such as excellent mechanical strength and high water absorption capacity [12]. By acting as moisture barriers, hydrogels can help improve the shelf life of food products, whereas their ability to regulate gas permeability ensures the optimal conditions for food preservation [13]. Additionally, hydrogels have the potential to encapsulate active compounds, providing controlled release mechanisms and protecting food products from oxidation and microbial degradation [14]. The versatility of hydrogels allows for their formulation with different materials, including gelatin and cellulose, which further expands their applications in food packaging [15].

Among the most studied renewable polymers for biodegradable packaging and hydrogel design is gelatin derived from collagen through hydrolysis [16]. This natural, protein-based material offers excellent film-forming properties, making it suitable for enhancing packaging materials' mechanical strength and barrier properties [17]. Gelatin films have shown promise in applications such as coatings and edible films, contributing to improved food preservation [18].

Another bio-based material with interesting properties is cellulose derived from renewable sources such as agro-industrial by-products including banana crops, which has emerged as a sustainable alternative to conventional packaging materials [19]. It displays high mechanical strength and low gas permeability and offers significant advantages such as biodegradability and biocompatibility [20]. Microcrystalline and nanocrystalline cellulose are forms of cellulose that have gained significant attention in various industries, including food packaging [21]. Microcrystalline cellulose is produced by the controlled hydrolysis of cellulose, resulting in the formation of small, rod-shaped particles with a crystalline structure [22]. Microcrystalline cellulose offers desirable properties such as a high surface area, excellent flowability, and moisture absorption, making it a versatile ingredient in food formulations and as a bulking agent in pharmaceutical tablets [23]. On the other hand, nanocrystalline cellulose refers to cellulose particles with nanoscale dimensions. Nanocrystalline cellulose exhibits exceptional mechanical strength, optical transparency, and barrier properties, which make it suitable for enhancing the mechanical and barrier performance of food packaging materials [24]. Its potential applications range from nanocomposites and coatings to films and sensors, contributing to the development of sustainable and advanced food packaging solutions.

In general, gelatin, MCC, and NCC are derived from natural and renewable sources; their use promotes a more sustainable approach to the manufacture of various products and packaging materials that are usually made from plastic, thus reducing the amount of plastic waste in landfills and in the environment, as well as reducing the dependence on non-renewable resources and promoting the use of sustainable raw materials. The extraction of microcrystalline cellulose from the banana pseudostem (Musa paradisiaca) allows the use of agro-industrial waste, generating added value to the hydrogels manufactured from them, preserving the environment, and promoting the circular economy. Hydrogel materials based on gelatin and cellulose present numerous benefits for food preservation, particularly for chicken products [25]. Chicken meat is highly perishable and susceptible to microbial contamination, posing food safety risks [26]. Hydrogels, with their mechanical strength and ability to act as barriers against bacterial invasion, moisture loss, and oxidation, can significantly extend the shelf life of chicken products. The application of hydrogels in chicken packaging demonstrates their potential in addressing food safety concerns and reducing food waste in the poultry industry [27,28].

In this context, this research aimed at the development and characterization of hydrogels based on gelatin and microcrystalline and nanocrystalline cellulose for chicken breast preservation. Firstly, the microcrystalline cellulose was isolated from banana pseudostems. Secondly, hydrogel materials were developed by solvent casting and were assessed in terms

of their morphology, optical and thermal properties, and crystallinity. Thirdly, the hydrogel materials were applied to chicken breasts to extend their shelf life.

2. Materials and Methods

2.1. Materials

Banana pseudostem (Musa paradisiaca) (Finca El Olimpo, Antioquia, Colombia), gelatin 250 bloom (Factores & mercadeo, Bogotá, Colombia), cellulose nanocrystals (Celluforce, Montreal, QC, Canada), sodium hydroxide (NaOH), hydrogen peroxide (H_2O_2), glycerol, and glutaraldehyde were provided by Panreac (Bogotá, Colombia).

2.2. Cellulose Microcrystalline Isolation

Banana pseudostem samples were cut vertically into thin strips and were dried in an oven at 100 ± 5 °C. They were then ground and sieved to obtain microfibers (MFs) [29]. To obtain microcrystalline cellulose (MCC), an alkaline stage and a bleaching stage were carried out [30]. The alkaline treatment was carried out with NaOH for 45 min at 75 °C under stirring, the MFs were washed after each alkaline treatment with distilled water. Subsequently, the MFs were subjected to additional bleaching using NaOH (4 wt.%) and H_2O_2 (5 wt.%) solution for 60 min at 80 °C under stirring. The isolated microcrystalline cellulose (MCC) was washed several times with distilled water until a neutral pH (pH~7) (Figure 1).

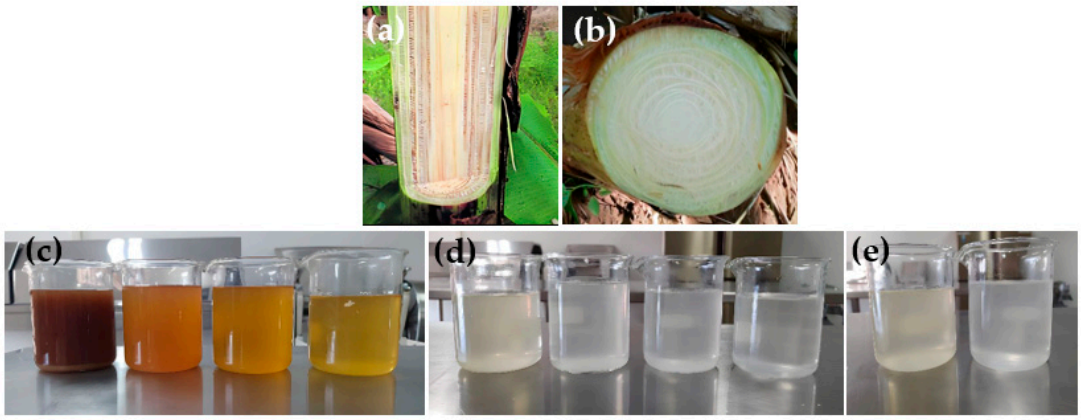

Figure 1. Banana pseudostem (Musa paradisiaca): (**a**) longitudinal section, (**b**) cross section. Microcrystalline cellulose isolation stages: (**c**) alkaline stage, (**d**) bleaching stage, and (**e**) combined treatments.

Cellulose Yield (%)

The yield of cellulose extracted from banana pseudostems was determined by the gravimetric method and calculated using Equation (1).

$$\% \text{ Cellulose yield} = \left(\frac{\text{Weight of obtained cellulose}}{\text{Weight of fiber used}} \right) \times 100 \quad (1)$$

2.3. Hydrogel Development

The hydrogels were developed following the procedure reported by [31], with some modifications. Table 1 shows the experimental design for obtaining hydrogels, expressed in mass fractions. Different gelatin solutions were prepared by dissolving the biopolymer at different concentrations in water. The microcrystalline and nanocrystalline cellulose were dissolved in water at 0.25 and 0.5 wt.% in relation to the gelatin and then added to the gelatin solution. Glycerol was also added as a plasticizer agent at 15 wt.% and glutaraldehyde as a cross-linking agent at 3 wt.% with the gelatin mixture. The hydrogels were obtained by solvent casting at 50 °C for 6 h.

Table 1. Experimental design for obtaining hydrogels expressed in mass fraction.

Sample	Gelatin	MCC	NCC	Glycerol	Glutaraldehyde
F1	0.847	0	0	0.127	0.025
F2	0.830	0.021	0	0.124	0.025
F3	0.813	0.041	0	0.122	0.024
F4	0.830	0	0.021	0.124	0.025
F5	0.813	0	0.041	0.122	0.024

2.4. Hydrogel Characterization

2.4.1. Thickness

The hydrogel thickness was measured using a digital micrometer (TL268 TOP EU) with an accuracy of 0.01 mm. Eight measurements were taken at 3 random positions and the average was calculated.

2.4.2. Morphology

The hydrogel samples were examined by SEM in a Hitachi SU8010 (Tokyo, Japan). For cross-sectional observations by SEM, the samples were previously cryo-fractured by immersion of the sample in liquid nitrogen. The SEM micrographs were taken at an accelerating voltage of 10 kV and a working distance of 8–10 mm; the samples were previously sputtered with a gold–palladium mixture for 3 min under a vacuum.

2.4.3. Fourier Transform Infrared Spectroscopy (FTIR)

Identification of the principal functional groups was conducted via spectroscopy equipment (IR Prestige 21 Shimadzu, Kyoto, Japan). Spectra were obtained within a wave number range of 4000 to 500 cm^{-1} by averaging 20 scans at a resolution of 4 cm^{-1} [32].

2.4.4. Moisture Content

Hydrogel moisture content was determined immediately after drying and after a conditioning period (58% RH during 48 h) according to the gravimetric method by the ASTM D644-99 standard test method for the moisture content of paper and paperboard by oven drying (ASTM, 2007) [33].

2.4.5. Swelling Test

Fully swollen hydrogel samples were dried with filter paper and their weight was recorded (W_1). Subsequently, the samples were dried at 100 °C under vacuum and reweighed (W_2). The swelling ratio was calculated using Equation (2) [34].

$$\% \text{ Swelling } = \frac{(W_1 - W_2)}{W_2} \times 100 \quad (2)$$

2.4.6. Color

Hydrogel color was measured using a Colorimeter CS-10 (Jiliang, China), according to the method in [35]. The CIELAB coordinates (L*, a*, b*) and tone density in terms of chroma (c*) and hue (h*) were evaluated. The color difference (ΔE*) was calculated by using Equation (3).

$$\Delta E = \left[\left(\Delta L^* \right)^2 + \left(\Delta a^* \right)^2 + \left(\Delta b^* \right)^2 \right]^{0.5} \quad (3)$$

where ΔL*, Δa*, and Δb* corresponded to the differences between the color parameters of the hydrogel samples and the values of the reference sample on day 0 for each formulation.

2.4.7. Gloss

Hydrogel gloss was measured according to the ASTM D523-89 standard test method for specular gloss (ASTM, 1999) [36]. The hydrogel samples were measured on the free

surface at an angle of incidence of 60° using a flat surface gloss meter (3NH YG268 Multi-angle, Minolta, Langenhagen, Germany). The results are expressed as gloss units (GU) with respect to the highly polished surface of a black glass standard with a value close to 100 GU.

2.4.8. Mechanical Test

The mechanical properties testing of hydrogel samples was carried out in a universal mechanical testing machine (AGS-X 500 N) from Shimadzu Corporation (Kyoto, Japan) in accordance with the ASTM D882-02 standard test method for the tensile properties of thin plastic sheeting (ASTM, 2001) [37]. Tensile strength (TS), elastic modulus (EM), and deformation capacity (E) were calculated from the stress–strain curves estimated from the force–distance data. A minimum of three specimens were measured for each sample and the average results with standard deviations are reported.

2.5. Application of Hydrogels in the Preservation of Chicken Breast

2.5.1. Chicken Breast Samples

Chicken breast cubes of approximately 50 g were covered with a top and bottom layer of the chosen hydrogel samples (F1, F3, and F5) and placed in airtight glass containers. The chicken cubes without hydrogel were used as a control sample. All samples were stored at 4 °C for 4 days [38].

2.5.2. Weight Loss (%)

The initial weight loss of the chicken samples was determined according to the official method of AOAC International (1994). The initial weight of the chicken samples (day 0) and the final evaluation weight (day 4) were recorded. The difference between the initial and final weights of the samples was considered as the total weight loss.

2.5.3. Color

The color of the chicken samples was measured according to the method in [39] using a Colorimeter CS-10 (Jiliang, China). The CIELAB coordinates (L*, a*, b*) and tone density in terms of chroma (c*) and hue (h*) were evaluated. The color difference (ΔE*) was calculated by using Equation (3).

2.5.4. pH

The pH of the chicken samples was evaluated following the methodology reported by Liu et al. [25]. The samples were crushed and mixed with distilled water, then the pH was measured using a pH meter (PHS-3C, Ciedutec, Bogotá, Colombia).

2.6. Statistical analysis

All data were evaluated through analysis of variance (ANOVA) with a 95% significance level ($p \leq 0.05$) and a multiple comparison test (Tukey) to identify significant differences among the treatments. Each treatment was performed in triplicate. For this purpose, Statgraphics Centurion XVI software (Manugistics Corp., Rockville, MD, USA) was used.

3. Results and Discussion

3.1. Morphology

The morphologies of the cross sections of the hydrogel samples containing MCC and NCC are shown in Figure 2. The F1 sample presented a thickness of ~352 µm. The F2 and F3 samples containing MCC exhibited a continuous structure with thicknesses in the 326–335 µm range; some pores and cracks were observed in their cross sections that were attributed to the particle size of MCC, which could form clumps [40,41]. On the other hand, samples F4 and F5 presented more homogeneous and smooth surfaces compared with samples F2 and F3, with thicknesses in the 363–337 µm range; this greater homogeneity is attributed to a better dispersion of the NCC due to the smaller size [42].

This increase in thickness has been reported by several authors, who have concluded that the addition of particles or fillers can also generate certain discontinuities in the polymer matrix [39,40,43,44].

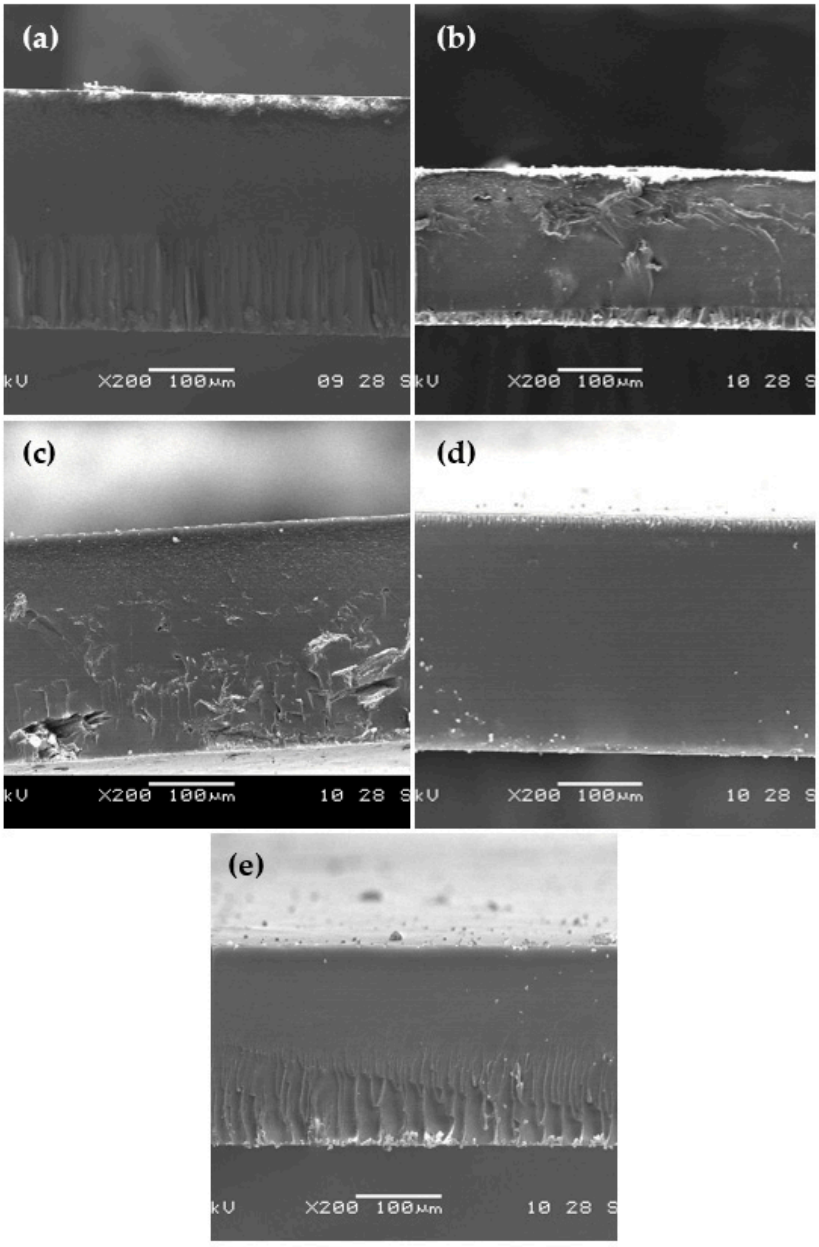

Figure 2. Scanning electron microscopy (SEM) images of hydrogel samples: (**a**) F1, (**b**) F2, (**c**) F3, (**d**) F4, and (**e**) F5. Scale bar 100 μm and magnification X200.

3.2. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR spectral data were used to confirm the cross-linking of the gelatin chains (caused by glutaraldehyde) and to identify changes in the gelatin functional groups after adding MCC and NCC to the hydrogels. Figure 3 shows the FTIR spectra recorded for the hydrogel samples. For F1, the absorption peaks at 3274 and 2938 cm^{-1} correspond to N-H stretching and aliphatic C-H stretching, respectively. The absorption peaks at 1451 and 1334 cm^{-1} correspond to C-H bending and C-N stretching, respectively. Three absorption peaks corresponding to amide groups were also observed. These three absorption peaks were at 1628 (amide I), 1539 (amide II), and 1237 cm^{-1} (amide III), coinciding with amide C=O stretching, amide N-H bending (with C-N stretching), and amide N-H bending (or CH_2

vibration), respectively. The 1628 cm^{-1} absorption peak of F1 showed values equal to those reported by Yin et al. [45] after comparing the FTIR spectra of neat gelatin and gelatin hydrogels, indicating that the absorption peak increased from 1627 to 1628 cm^{-1}, evidencing a cross-linking of the gelatin chains when using glutaraldehyde. This peak overlaps with the amide CO stretching peak of the gelatin itself. Therefore, the overall intensity of this peak increases after cross-linking [45].

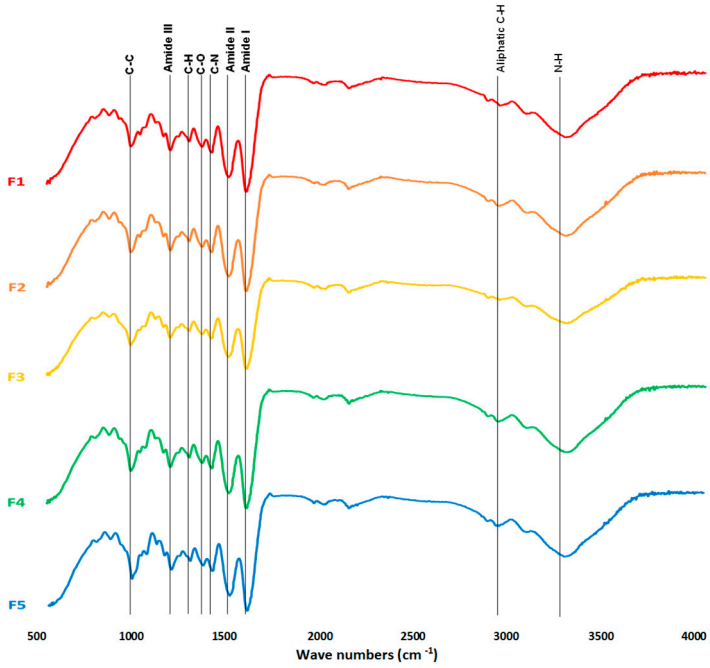

Figure 3. FTIR spectra of hydrogel samples. The spectra were recorded ranging from a 4000 to 500 cm^{-1} wavenumber.

All formulations did not show new peaks, only those corresponding to gelatin, MCC, and NCC. For the formulations with NCC and MCC, two characteristic peaks of cellulose were observed at 1402 and 1033 cm^{-1} that were caused by C-C vibrations within the cellulose structure and C-O tensile vibrations in the ester and carboxylic groups due to the existence of the same functional groups in the two compounds (MCC and NCC) [39]. The peaks of amide I, II, and III in sample F3 were slightly increased compared with sample F1. Indeed, the vibrational frequency of C=O (amide I) is affected by the hydrogen bonds between the amide units, the slight frequency shift of the amide I peak in sample F3 reflects the disruption of hydrogen bonds in the carbonyl groups of gelatin by the competitive binding of MCC. Therefore, the polarity could decrease as more hydrogen bonds form between gelatin and MCC [41].

The increased amount of hydrogen bonding interactions between the hydroxyl groups of cellulose and the carboxyl and amino groups of gelatin influenced the physical and mechanical properties of the final materials [40,41].

3.3. Moisture Content and Swelling

Table 2 shows the moisture content of the hydrogel samples and their swelling after being immersed in water for 24 h. F1 presented the highest moisture content after drying, with 12.522% moisture content and a higher swelling capacity of 375.97%, whereas F3 and F5 presented lower moisture content after drying with values of 11.61% and 11.98%, respectively, and lower swelling capacity with values of 300.95% and 291.85%, respectively. The incorporation of MCC and NCC effectively led to the creation of a rigid structure in the hydrogel [41]. In general, hydrogels containing NCC swell more than those containing MCC due to the larger contact surface and greater ability to interact with water [46,47].

Table 2. Moisture content and swelling of hydrogels.

Sample	W1 (g)	W2 (g)	W3 (g)	Absorbed Water (%)	Moisture Content (%)	Swelling (%)
F1	0.53 ± 0.07	0.47 ± 0.06	2.23 ± 0.30	1.769	12.522	375.912
F2	0.44 ± 0.03	0.39 ± 0.03	1.71 ± 0.14	1.319	12.252	334.435
F3	0.39 ± 0.02	0.35 ± 0.02	1.40 ± 0.13	1.056	11.611	300.950
F4	0.42 ± 0.06	0.37 ± 0.05	1.68 ± 0.28	1.316	12.104	354.122
F5	0.48 ± 0.09	0.42 ± 0.08	1.66 ± 0.32	1.237	11.979	291.847

W1: Weight of the conditioned sample. W2: Weight of the sample dried at 110° for 1 h. W3: Weight of the sample immersed in water for 24 h.

With respect to the amount of water absorbed by the samples and their swelling, for F1, a value of 1.769 g for water absorbed and the largest size (diameter) with respect to the other formulations were observed, corresponding to 62.23 mm, whereas for F3 and F5, values of 1.056 and 1.237 g, respectively, for water absorbed and diameters of 47.53 and 50.57 mm, respectively, are observed. These results are in agreement with those reported by Boughriba et al. [41]. It is evident that the formulations with MCC (F2 and F3) have the lowest swelling values, as they are stiffer formulations, with greater irregularities and less capacity to absorb water [39]. On the other hand, formulations with NCC (F4 and F5) have higher values than those presented by formulations with MCC; this is because the particle size of cellulose has a significant influence on the swelling of hydrogel samples. NCC particles are smaller and form a finer and more homogeneous network within the hydrogel, allowing greater water absorption and, therefore, greater swelling [46,48].

3.4. Color and Gloss

Table 3 gathers the values for the color coordinates (L*, a*, b*), the values of ΔE, and gloss of the hydrogels containing MCC and NCC. The hydrogel samples exhibit lightness values (L*) between 74.91 and 78.84; reddish (+a*) and yellowish (+b*) shades are in the ranges of 7.25–11.88 and 39.24–50.44, respectively. The chroma (c*) and hue (h*) values were in ranges of 39.90–51.82 and 76.76–79.75, respectively. The CIELAB color space was calculated to determine the color variation (ΔE) of the hydrogel samples with respect to the control sample (F1). The lowest variation was obtained for sample F5 and the highest for sample F3, with values of 3.445 and 8.140, respectively. According to Boughriba et al. [41] and Ventura-Cruz, et al. [46], the presence of NCC in the hydrogel matrix may have a greater impact on color variation than MCC due to its extremely small size and greater ability to disperse uniformly into the hydrogel matrix.

Table 3. Color parameters (L*, a*, b*, c*, h*, and ΔE*) and gloss characteristics of hydrogels.

Sample	L*	a*	b*	c*	h*	ΔE*	Gloss (GU)
F1	78.84 ± 3.38	8.67 ± 2.54	44.09 ± 7.84	44.94 ± 8.20	79.01 ± 1.16	0	94.8 ± 3.89
F2	78.88 ± 0.49	7.25 ± 0.70	39.24 ± 2.54	39.90 ± 2.63	79.54 ± 0.32	5.047	75.6 ± 4.92
F3	74.91 ± 1.43	11.88 ± 1.21	50.44 ± 2.54	51.82 ± 2.74	76.76 ± 0.65	8.140	78.6 ± 7.23
F4	76.27 ± 2.89	10.36 ± 2.46	48.31 ± 6.98	49.42 ± 7.34	77.99 ± 1.17	5.226	97.8 ± 1.78
F5	78.25 ± 2.61	7.41 ± 1.24	40.93 ± 5.91	41.6 ± 6.03	79.75 ± 0.31	3.445	71.4 ± 7.76

Sample F1 presented a gloss of 94.8 GU. Samples F2 and F3 presented gloss values of 75.6 and 78.6 GU, respectively. The highest gloss value was for sample F4 (97.8 GU), and the lowest gloss value was for sample F5 (71.4 GU). The samples containing NCC (F4 and F5) showed higher gloss due to their high crystallinity (between 60 and 90%) compared with samples containing MCC (F2 and F3), which had a crystallinity between 50 and 80%. The gloss could also be affected by factors such as the surface roughness, texture uniformity, presence of imperfections, and quality of illumination [46]. Sample F5 exhibited lower gloss values due to an aggregation of NCC particles that turned into MCC; this aggregation could have been caused by several factors, such as concentration, size, and the chemical properties of the particles [46].

3.5. Mechanical Properties of Hydrogels

The mechanical properties of the hydrogel samples are shown in Table 4. Sample F1 presented a TS value of 22.2 MPa, an EM value of 350 MPa, and an E value of 93%. The incorporation of MCC and NCC increased the mechanical values; for sample F2, the TS value was 26.2 MPa, EM was 361 MPa, and E was 90.6%. For sample F3, the TS value was 28.3 MPa, EM was 366 MPa, and E was 86%. Sample F4 presented a TS value of 42.2 MPa, an EM of 420 MPa, and an E of 83%. Sample F5 showed a TS value of 46.1 MPa, an EM of 435 MPa, and an E of 79.2%. This increase in mechanical properties can be attributed to the reinforcement effect of MCC and NCC. This effect could have been influenced by different factors such as particle dispersion, particle concentration, and variation in matrix crystallinity [41,46,48]. Samples containing NCC (F4 and F5) showed higher mechanical values due to smaller and more uniform structures compared with MCC, which improved the reinforcement effect in the gelatin matrix [39,41,46,49,50].

Table 4. Mechanical properties of hydrogels in terms of tensile strength (TS), elastic modulus (EM), and deformation capacity (E).

Sample	Thickness (μm)	TS (MPa)	EM (MPa)	E (%)
F1	352 ± 7^{b}	22.2 ± 0.8^{a}	350 ± 5^{a}	93.0 ± 2.0^{a}
F2	326 ± 5^{a}	26.2 ± 0.9^{b}	361 ± 7^{b}	90.6 ± 1.2^{a}
F3	335 ± 6^{a}	28.3 ± 1.0^{b}	366 ± 3^{b}	86.0 ± 2.0^{b}
F4	363 ± 8^{b}	42.2 ± 2.1^{c}	420 ± 8^{c}	83.0 ± 2.0^{c}
F5	337 ± 5^{a}	46.1 ± 0.9^{d}	435 ± 4^{d}	79.2 ± 1.3^{d}

[a–d] Different letters in the same column indicate a significant difference ($p < 0.05$).

3.6. Application of Hydrogels in the Preservation of Fresh Chicken Breast

Figure 4 shows the chicken breast cubes covered with the chosen hydrogel samples, F1, F3, F5, and the control sample (chicken without hydrogel). The hydrogels absorbed water from the surface of the chicken breast and adhered to it to form a protective film.

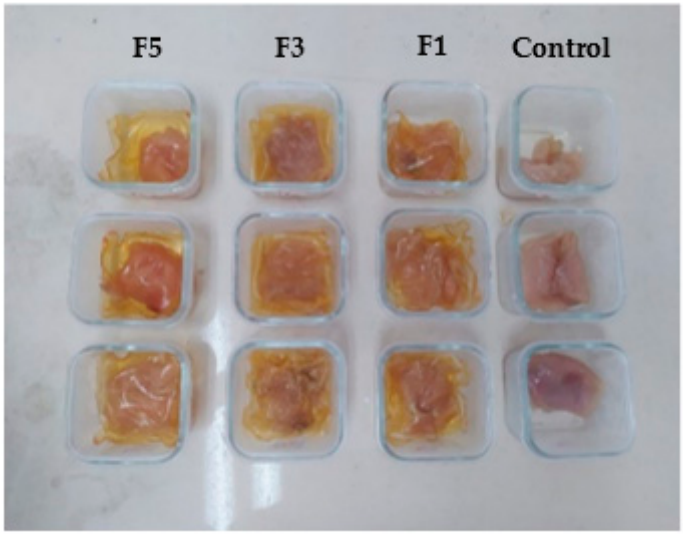

Figure 4. Chicken breast cubes cover with hydrogels.

3.6.1. Weight Loss (%)

The weight loss and swelling results of the chicken breast samples are shown in Table 5. It was observed that the chicken samples covered with the hydrogels had a lower weight compared with the control sample. Likewise, samples F1, F3, and F5 showed higher losses than the control sample, with values of 67.52, 66.77, 65.52 and 54.47%, respectively. F5 showed the highest swelling values (310.13%), which were associated with the higher

specific surface area of NCC allowing greater interaction with water and a higher water-holding capacity [46,50].

Table 5. Weight loss of chicken breast covered with hydrogels and stored at 4 °C for 4 days.

Sample	Total Weight Loss (g)	Initial Weight Loss (%)	Weight Gain of Hydrogel (g)
Control	26.996	54.47	---
F1	33.240	67.52	6.141
F3	33.489	66.77	5.913
F5	33.152	65.52	6.281

3.6.2. Color

Table 6 shows the color parameters of chicken breast samples stored at 4 °C for 4 days. Color changes in the chicken samples are influenced by various factors, including oxidation, enzymatic reactions, and microbial activity. As can be observed for all the samples, as storage progressed, the brightness (L*) and redness (+a*) values of the chicken breast decreased, whereas the chroma (c*) and hue (h*) values increased [51]. The control sample (without hydrogel) exhibited the highest color variation on day 4 (ΔE = 7.224). Sample F1 presented a ΔE of 5.059 on day 4. In contrast, the chicken samples covered with hydrogels (F3 and F5) demonstrated less color variation, which can be attributed to the varying barrier properties and interactions between the hydrogel and the chicken samples. Sample F5 showed the lowest color variation on day 4 (ΔE = 1.391); this hydrogel created a protective barrier, reducing exposure to oxygen and enzymatic reactions and resulting in milder color changes. These findings indicate that hydrogels containing microcrystalline cellulose (MCC) and nanocrystalline cellulose (NCC) could effectively reduce oxygen permeability and enzymatic activity in chicken samples, thereby preventing significant color alterations. On the other hand, the control sample and sample F1, with lower barrier properties, were more prone to triggering reactions that significantly affected the color of the chicken breast [45,52].

Table 6. Color parameters (L*, a*, b*, c*, h*, and ΔE*) of chicken breast covered with hydrogels and stored at 4 °C for 4 days.

Sample	Day	L*	a*	b*	c*	h*	ΔE*
Control	0	52.71 ± 3.11	−0.4 ± 1.30	4.12 ± 0.68	4.34 ± 0.59	95.18 ± 18.66	0
	1	52.95 ± 1.92	−1.3 ± 1.89	4.69 ± 0.42	5.15 ± 0.84	103.70 ± 19.82	1.120
	2	53.80 ± 1.61	−2.0 ± 1.62	4.18 ± 0.97	4.87 ± 1.11	114.30 ± 18.16	1.953
	3	51.35 ± 2.63	−5.2 ± 2.87	4.48 ± 0.73	7.06 ± 2.56	135.70 ± 13.10	5.050
	4	51.21 ± 1.52	−7.4 ± 1.96	5.06 ± 1.15	9.09 ± 1.72	144.90 ± 9.17	7.224
F1	0	54.21 ± 0.92	0.28 ± 1.33	5.59 ± 0.43	5.73 ± 0.50	87.32 ± 12.78	0
	1	51.16 ± 1.84	−0.40 ± 1.31	5.90 ± 0.96	6.06 ± 0.84	95.45 ± 13.72	3.163
	2	51.19 ± 2.15	−1.80 ± 1.93	5.56 ± 1.41	6.17 ± 1.31	109.7 ± 18.57	3.717
	3	50.41 ± 1.87	−3.40 ± 2.90	5.10 ± 1.17	6.72 ± 1.29	130.2 ± 7.42	5.344
	4	52.45 ± 2.11	−4.40 ± 1.15	5.05 ± 1.07	6.85 ± 0.58	131.3 ± 12.40	5.059
F3	0	52.35 ± 2.60	0.75 ± 1.10	4.51 ± 0.70	4.70 ± 0.68	80.15 ± 14.05	0
	1	50.83 ± 1.71	−1.50 ± 0.67	5.73 ± 1.26	5.98 ± 1.16	105.70 ± 8.07	2.992
	2	49.40 ± 1.77	−1.60 ± 1.70	5.46 ± 1.09	5.96 ± 0.94	107.40 ± 17.50	3.937
	3	50.69 ± 1.90	−2.70 ± 1.31	5.46 ± 1.12	6.24 ± 0.98	116.40 ± 13.20	3.950
	4	50.57 ± 1.93	−3.30 ± 1.23	4.98 ± 1.07	6.10 ± 1.00	112.50 ± 39.47	4.453
F5	0	49.93 ± 2.59	−1.50 ± 0.98	5.09 ± 0.91	5.41 ± 0.85	107.20 ± 11.32	0
	1	48.58 ± 3.57	−1.70 ± 1.62	6.09 ± 1.12	6.51 ± 1.27	105.50 ± 13.57	1.697
	2	49.04 ± 4.15	−1.80 ± 2.83	5.49 ± 1.07	6.37 ± 1.12	114.80 ± 18.03	1.021
	3	48.3 ± 2.31	−3.20 ± 0.71	6.04 ± 1.07	6.96 ± 0.85	118.60 ± 8.56	2.535
	4	49.74 ± 2.35	−2.80 ± 0.90	5.50 ± 0.53	6.29 ± 0.37	117.80 ± 8.60	1.391

3.6.3. pH

Table 7 gathers the pH values of the chicken breast samples covered with hydrogels. The pH of chicken breasts tends to be slightly acidic, usually ranging from 5.5 to 6.5. During storage at 4 °C, the pH of chicken can undergo changes due to microbiological and biochemical processes. The results indicate that the hydrogel-coated chicken breast samples experienced a slight decrease in pH values, ranging from 6.00 to 6.06 over the four-day period. In contrast, the control sample exhibited slightly higher pH values (6.44) compared with the coated samples.

Table 7. pH values of chicken breast stored at 4 °C for 4 days.

Sample	Day 0	Day 1	Day 2	Day 3	Day 4
Control	6.00	6.10	6.21	6.30	6.44
F1	5.97	6.06	6.03	5.99	6.00
F3	6.03	6.07	6.03	6.02	6.00
F5	6.20	6.05	6.10	6.12	6.06

The increased pH values in the chicken samples on day 1 are attributed to the combination of the breast with the hydrogel components. After day 2 of storage, the chicken breast may have undergone fermentation of muscle glycogen, resulting in the production of acidic substances, such as lactic acid and phosphoric acid, due to cellular respiration. This led to a subsequent decrease in the pH of the samples [51]. Likewise, the semi-permeable barrier created by the hydrogels influenced the gas exchange between the chicken samples and the surrounding environment [25]. On the other hand, the availability of oxygen can significantly impact various metabolic processes in meat, encompassing protein alterations and the generation of alkaline by-products. The decreased oxygen supply in the coated chicken samples could have influenced the microbial activity and the resulting pH changes, leading to slightly lower pH values in comparison with the control chicken samples [27].

4. Conclusions

This study examined various formulations of hydrogels based on gelatin, microcrystalline cellulose (MCC), and nanocrystalline cellulose (NCC). The NCC-based formulations exhibited denser structures and higher swelling capacities. Subtle changes were observed in the FTIR spectra, indicating interactions between the hydrogel components. Increasing the concentration of cellulose derivatives enhanced the mechanical properties such as tensile strength and Young's Modulus while maintaining deformation capacity. Regarding optical properties, color and gloss variations were primarily found in formulations with added NCC. In the assessment of moisture absorption in a food matrix, NCC-reinforced hydrogels better preserved pH and showed less color variation in chicken breasts, along with greater swelling capacity. The broad potential of applying hydrogels in packaged products presents an environmentally friendly alternative that enhances the presentation of meat products during sale by controlling the presence of exudates in the packages. The results demonstrated that NCC-reinforced gelatin-based hydrogels are a suitable option for moisture control and extending the shelf life of packaged chicken breast, offering improved presentation and eliminating exudates.

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