Journal of Food Measurement and Characterization (2026) 20:8829–8843
https://doi.org/10.1007/s11694-026-04234-9

ORIGINAL PAPER

Physically cross-linked hydrogel based on sorghum starch: development and characterization

Ana Luiza Santos Vieira[1] · Vinícius Tadeu da Veiga Correia[1] · Nayana Hayss Araújo da Silva[1] · Valéria Aparecida Vieira Queiroz[2] · André Augusto Gomes Faraco[3] · Washington Azevêdo da Silva[4] · Cleverson Fernando Garcia[5] · Maria Aparecida Vieira Teixeira Garcia[1] · Raquel Linhares Bello de Araújo[1]

Received: 24 November 2025 / Accepted: 23 February 2026 / Published online: 14 March 2026
© The Author(s) 2026

Abstract

The objectve of this work was to develop a hydrogel based on white sorghum starch and characterize its visual aspects, behavior in the presence of water, morphology, and texture profile. Six hydrogel samples were developed from sorghum starch, chitosan, and different proportions of citric acid through thermal gelatinization. The hydrogels were then physically crosslinked by freeze-thawing. All six samples showed high water absorption capacity and low syneresis rate, ranging from 0.35% (sample 5, with the highest concentration of citric acid) to 9.75% (sample 6, without the addition of citric acid). After lyophilization, all six samples exhibited stable and intact structures after immersion in water; however, those with citric acid maintained greater structural integrity after 24 h. The water activity values among the samples ranged from 0.995 ± 0.002 to 0.999 ± 0.001, characterizing a high a_w, as they were close to 1. Regarding water release from the hydrogels, the six samples presented values ranging from 0.08 ± 0.03 to 25.35 ± 1.90%. Water loss values ranged from 0.03 to 0.44 g among the six hydrogel samples. Regarding the pH of the hydrogel samples, the results ranged from 3.36 ± 0.01 to 7.66 ± 0.01. Morphological analysis of the hydrogels revealed that the crosslinking process promoted the formation of a three-dimensional polymeric matrix, resulting in highly porous hydrogels with greater water absorption capacity. The texture profile of the developed hydrogels revealed the formation of firm and cohesive gels, with good integrity and high resistance. Fourier transform infrared spectroscopy (FTIR) identified the characteristic functional groups of starch and citric acid. This study presents, for the first time, the development of hydrogels based on sorghum starch, a raw material still unexplored for this application, highlighting the potential of sorghum as a sustainable and environmentally friendly alternative. Furthermore, the developed material stands out as a promising matrix for the incorporation of bioactive compounds, expanding its possible technological applications.

Keywords Polymeric gel · biopolymer · Sorghum bicolor (L.) Moench · Citric acid · Chitosan

✉ Ana Luiza Santos Vieira
anavieiranutricionista@gmail.com

[1] Department of Food, School of Pharmacy, Universidade Federal de Minas Gerais, Belo Horizonte, MG, Brazil

[2] Brazilian Agricultural Research Corporation (EMBRAPA Maize and Sorghum), Sete Lagoas, MG, Brazil

[3] Department of Pharmaceutical Sciences, School of Pharmacy, Universidade Federal de Minas Gerais, Belo Horizonte, MG, Brazil

[4] Department of Food Engineering (DEALI), Universidade Federal de São João del-Rei, Sete Lagoas Campus, Sete Lagoas, MG, Brazil

[5] Department of Chemistry CEFET MG, Belo Horizonte, MG, Brazil

Introduction

Starch hydrogels are characterized as a polymeric matrix with a highly hydrophilic three-dimensional structure, which has the ability to absorb and retain large amounts of water without dissolving. This occurs through the formation of a cross-linked three-dimensional network, in which water is able to enter the polymer granules, forming strong and stable interconnected structures [1, 2].

The cross-linking of starch hydrogels can occur through physical or chemical processes, in which water penetrates the polymer granules, forming interconnected structures that are highly resistant [3, 4]. Physical crosslinking offers an advantage over chemical methods because it is a process

that does not involve the use of potentially toxic reagents, resulting in a more natural and ecologically sustainable product [5].

The physical cross-linking of starch gels can be achieved through freeze-thawing cycles. During freezing, when the paste or gel is frozen, phase separation occurs as ice crystals form. After thawing, the polymer paste and gel undergo syneresis, i.e., water is released. These cycles promote increased molecular associations between polymer chains, resulting in the formation of a porous system [5, 6].

In recent years, there has been a notable rise in the use of both conventional and unconventional starch-based hydrogels as matrices for releasing bioactive compounds [7]. Examples include potato starch [8], rice [9], mung beans [10], wheat [11], tapioca [12], and cassava-parsley [13], with corn starch being the most widely used and recognized as the predominant commercial starch globally [14–16].

Unconventional starch sources are pivotal in addressing the demand for biodegradable materials and packaging, serving as an alternative to promote ecologically sustainable systems [3]. Sorghum emerges as a promising alternative source of starch, which is produced worldwide, including in Brazil. It is rich in starch (70 to 80%) and is available all year round. As a cereal crop, sorghum is cultivated in very dry, hot areas, and under different environmental conditions, being considered an important contributor to the security of the food supply. Given these conditions, sorghum has an advantage over other cereal crops, which often struggle with low productivity in such climates [17–19].

To date, there are no reports in the literature on the development of hydrogels based on sorghum starch, which gives the present investigation a novel and relevant character. The exploration of this starch for such an application stands out as a promising alternative, especially given the scarcity of studies involving unconventional starches in the formulation of hydrogels, despite their high potential for sustainable and environmentally compatible systems. Therefore, the objective of this study was to develop a hydrogel based on sorghum starch, chitosan and varying concentrations of citric acid, physically cross-linked, for different applications, with a particular focus on providing a matrix for the incorporation of extracts rich in bioactive compounds.

Materials and methods

Material

Sorghum grains [Sorghum bicolor (L.) Moench] of the BR 501 genotype (white pericarp and no tannin) were donated by the National Maize and Sorghum Research Center (CNPMS) of EMBRAPA, located in Sete Lagoas, Minas Gerais, Brazil (latitude 19º28'S, longitude 44º15'W). The hydrogels were produced by extracting starch from sorghum grains, which served as the base material. Chitosan (Xingcheng Biochemical, China) and citric acid (Dinâmica Química Contemporânea LTDA, Brazil) were also used in the production process.

Methods

Sorghum starch extraction

The starch extraction followed the method described by Vieira et al. [19] with modifications, as shown in Fig. 1. The sorghum grains (500 g) were soaked in an aqueous solution containing 0.15% sodium metabisulfite and incubated in a BOD oven (BOD Incubator with Photoperiod SL – 224, SOLAB) at 40 °C for 24 h. After draining and washing extensively, the grains were ground in an industrial blender (Poli Metalúrgica Siemsen, model TR-02, frequency 60 Hz, power 245 W) and sifted through 80 mesh and 120 mesh sieves for standardization. The resulting material was then rested at 10 °C for 20 h. The decanted material was resuspended in distilled water and centrifuged (Mod Jouan BR4) at 3500 rpm for 15 min until pure starch, visually confirmed by color, was obtained. The centrifuged material was dried in a forced air circulation oven (Mod 320-SE – Mechanical Circulation, FANEM) at 40 °C for 12 h. Following drying,

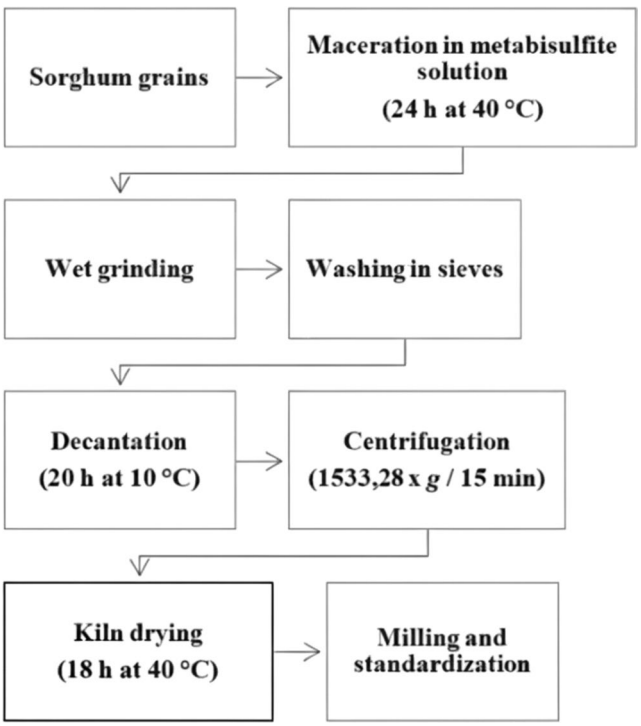

Fig. 1 Flowchart of the sorghum starch production process
Source: Author (2024)

Table 1 Experimental design for the development and characterization of hydrogels made with sorghum starch, chitosan and citric acid (CA)

Sample	Sorghum starch (g)	Chitosan (g)	Citric acid (%)
S1	5	0.5	2
S2	5	0.5	3
S3	5	0.5	4
S4	5	0.5	6
S5	5	0.5	10
S6	5	0.5	0

the starch was sieved through a mesh with an opening of 0.153 mm to standardize particle size and appearance.

Experimental design

The hydrogel samples were defined through pre-tests, based on the formulation 5 g of sorghum starch to 0.5 g of chitosan, varying the proportion of citric acid in terms of percentage of weight (% w/w of starch), according to the method described by Demitri et al. [20], with some modifications. A completely randomized design (CRD) was used, as shown in Table 1.

Elaboration of starch hydrogels

The hydrogels were prepared by the heating gelatinization method, as described by Biduski et al. [9], with some modifications. The sorghum starch suspensions (5 g of starch and 50 ml of distilled water - values established according to preliminary tests) were heated in a water bath with constant agitation until reaching 90 °C (GMA MedicalTM, Brasil). Subsequently, 0.5 g of chitosan, 50 ml of water and the different proportions of citric acid (as previously described in Table 1 to improve chitosan gelatinization) were homogenized using a magnetic stirrer and added to the starch and water suspensions. The mixture was then heated for 30 min to complete the starch gelling process. The resulting dispersions were stored in polystyrene Petri dishes measuring 15 cm in diameter and allowed to cool to room temperature (25 ± 2 °C).

Physical cross-linking of hydrogels

The hydrogels underwent physical crosslinking through five cycles of freezing and thawing, following the method described by Silva et al. [21]. The 6 hydrogel samples were frozen at -20 °C, and then completely thawed at 30 ± 2 °C.

Evaluation of visual aspects, syneresis rate and water absorption capacity of hydrogels

After preparing the six hydrogel samples and subjecting them to the physical cross-linking process through freezing and thawing, they were photographed for evaluation of their visual aspects.

Syneresis rate The syneresis rate of hydrogels following physical crosslinking (freeze/thaw treatment) was determined according to the method described by Li et al. [22] with some modifications. After undergoing physical crosslinking, the hydrogels were stored at 10 ± 2 °C for 24 h, weighed (*pi*), and then the released water was decanted and the hydrogels were weighed again (*pf*). The syneresis rate was determined according to Eq. 1.

$$SR\,(\%) = \frac{P_i - P_f}{P_f} x100 \tag{1}$$

After this step, the six samples were lyophilized for 72 h. Both lyophilized and swollen hydrogels were photographed and evaluated for structural integrity after being immersed in distilled water for 24 h.

Determination of water activity, water release and water holding capacity of hydrogels

Water Activity (a_w) The water activity (aw) was determined by directly reading the samples, using the AquaLab Lite 2T equipment, from Decagon Devices Inc, (Pullman, USA), operating at a temperature of 25 °C. Calibration was conducted using distilled water and saturated lithium chloride solution (aw 0.112 ± 0.003). The samples were placed in plastic capsules, which were inserted into the equipment to measure the aw values. The analyses were performed in triplicate.

Water release (%) The water release rate was determined according to the method described by Li et al. [22], with some modifications. Briefly, suspensions of starch, chitosan, and citric acid (Table 1) were subjected to gelatinization by heating. The hydrogels were then stored at 0 °C for 24 h and subsequently brought to room temperature for analysis. The water release rate was measured gravimetrically as the amount of water released (g of water/100 g of gel) after centrifugation at 1500 x *g* for 15 min. All analyses were conducted in triplicate.

Water Holding Capacity (WHC) The water holding capacity (WHC) was determined following the method outlined by Li et al. [22], with some modifications. Samples of hydrogels (2 g, *w1*) were dried in an oven at 40 °C for 2 h and then centrifuged at 4000 rpm for 20 min. After removing excess water with filter paper, the remaining water content

was weighed (w_2). The WHC was then calculated using Eq. 2.

$$WHC = w_1 - w_2 \quad (2)$$

Hydrogen potential (pH)

The pH readings were obtained using a benchtop pH meter (Bante, model 920). Each reading was conducted in triplicate.

Morphological evaluation of hydrogels

The morphology of the hydrogels was evaluated using a Scanning Electron Microscope (JEOL JSM −6360LV). Prior to the analysis, the samples were freeze-dried, and subsequently, a small fraction of each sample was collected, fixed on carbon tape, pulverized and coated with gold. Visualization analysis was conducted using magnifications ranging from 50 to 350 × with a voltage of 5.0 V.

Texture Profile Analysis (TPA)

The TPA of the hydrogels was performed following the method described by Biduski et al. [9] with some modifications. A TAXTPLUS (*Stable Micro Systems*) texture analyzer was used, and data were collected using the *Exponent Lite* program, version 5.1.1.0, 2010. The analysis involved two compression cycles. The samples, placed in 100 ml plastic containers (5 cm diameter x 7 cm height) at a height of 2 cm, were tested in quadruplicate. Each sample underwent compression to 50% of its initial height at a speed of 5 $mm.s^{-1}$ using a cylindrical probe of 35 mm in diameter. A defined delay period (10s) between the beginning of the second and the end of the first compression was applied. The attributes hardness, cohesiveness, gumminess, resilience and adhesiveness were analyzed.

Fourier-Transform Infrared Spectroscopy (FTIR)

The spectra of the hydrogels were acquired using a Fourier Transform Infrared Spectrophotometer (Shimadzu, model IR-Prestige 21) equipped with an Attenuated Total Reflectance (ATR) sampler. This enabled direct insertion of each sample for analysis without prior preparation. The equipment was configured to perform 20 scans, with a resolution of 1 cm^{-1}, over the range of 4000 and 400 cm^{-1}.

Statistical analysis

The results underwent analysis of variance (ANOVA) and comparison of means using Tukey's test at a significance level of 5%, after evaluation of normality and homogeneity. All analyses were performed using SPSS 15.0 (SPSS Inc., USA).

Results and Discussion

Evaluation of visual aspects, syneresis rate and water absorption capacity of hydrogels

The photographs of the hydrogels, prepared from sorghum starch and chitosan with varying proportions of citric acid, after gelatinization by heating, are presented in Fig. 2.

From the photographs taken (Fig. 2) of the 6 hydrogel samples, noticeable differences in the visual aspects can be observed, particularly in terms of color intensity and gel consistency.

After the preparation of the six hydrogel samples, they were submitted to the physical crosslinking process, and the resulting visual aspects are shown in Fig. 3.

It was observed that, after being gelatinized again in a water bath at 90 °C for 30 min, samples S1, S2 and S5 presented a firmer consistency with the presence of groats noted. Sample S6 (0% citric acid) also displayed a firm consistency and darker color compared to the others, in addition to the presence of lumps.

In contrast, samples S3 and S4 formed a fluid, homogeneous, smooth gel without the presence of lumps. Therefore, these two samples presented better visual aspects compared to the others, meaning those with lower concentrations of citric acid (S1 and S2), higher concentration of citric acid (S5) and without the addition of citric acid (S6). This observation demonstrates that intermediate levels of citric acid were sufficient to improve the gelatinization process of

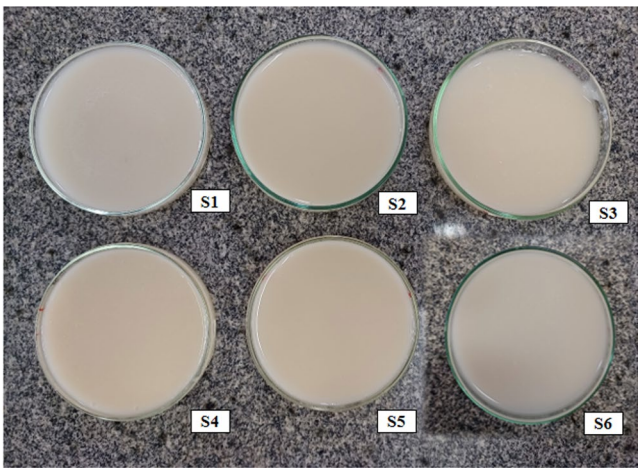

Fig. 2 Visual aspects of the six samples of hydrogels prepared with sorghum starch, chitosan and varying proportions of citric acid (CA). S1: 2% CA; S2 3% CA; S3: 4% CA; S4: 6% CA; S5: 10% CA; S6: 0% CA

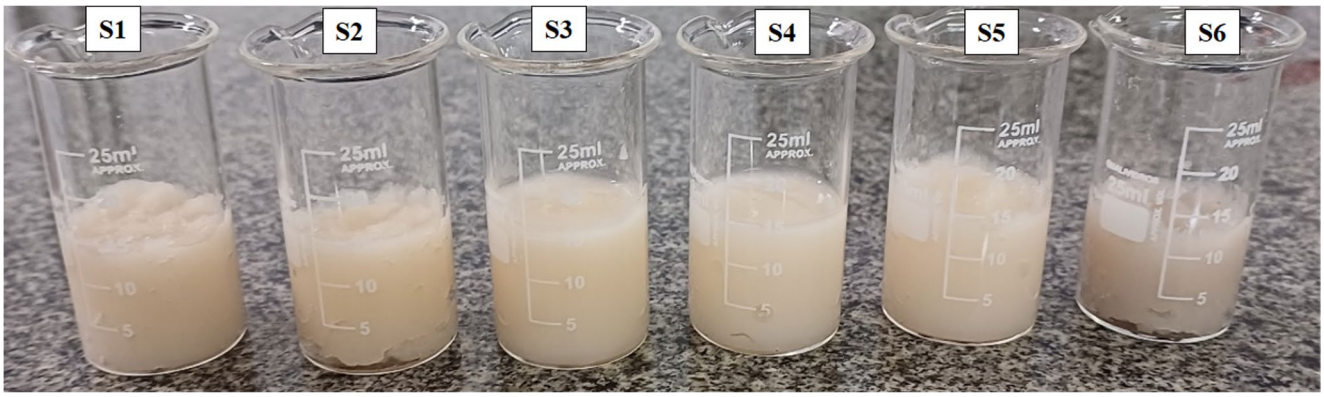

Fig. 3 Visual aspects of sorghum starch and chitosan hydrogels with different citric acid ratios after physical crosslinking. S1: 2% CA; S2 3% CA; S3: 4% CA; S4: 6% CA; S5: 10% CA; S6: 0% CA

the hydrogels composed of sorghum starch and chitosan, because it provides an acidic medium that promotes a better solubility of chitosan [23].

The visually observed differences between the samples may be attributed to the concentrations of citric acid (CA), which is an efficient crosslinker used in hydrogels, not only due to its non-toxicity, but also due to the stable cross-links it forms with chitosan and starch. In addition, CA is widely accessible and inexpensive, which makes it a very interesting reagent for industrial applications [24, 25].

A study carried out by da Costa et al. [26], who developed a hydrogel based on corn starch, demonstrated that the amylose content influences the degree of crosslinking, thermal stability and water absorption, with the hydrogel developed with starch with low amylose content (1.8%) showing a worsening of these parameters, unlike the hydrogels developed with starch with high amylose content (70%).

The white sorghum starch used in this study has 26.88% amylose [19] and this content is considered average and was sufficient to promote starch gelatinization, forming a fluid, homogeneous hydrogel with good stability, according to visual assessment.

Syneresis rate

The mean results of the syneresis rates of the six hydrogel samples are shown in Fig. 4.

As can be seen, the syneresis rates of the six samples of hydrogels, with different proportions of citric acid, varied statistically from each other, with sample 6, without the addition of citric acid, exhibiting the highest rate of syneresis, and sample 5 with 10% citric acid, displaying the lowest. Samples 1, 2, 3 and 4 exhibited syneresis rates ranging from 3.39 to 6.16%.

Li et al. [22], reported syneresis rates ranging from 1 to 9% in their study on hydrogels prepared with corn starch and tapioca, values close to those found in the present study. The authors attributed these values to the presence of starches in the system, which had a strong effect

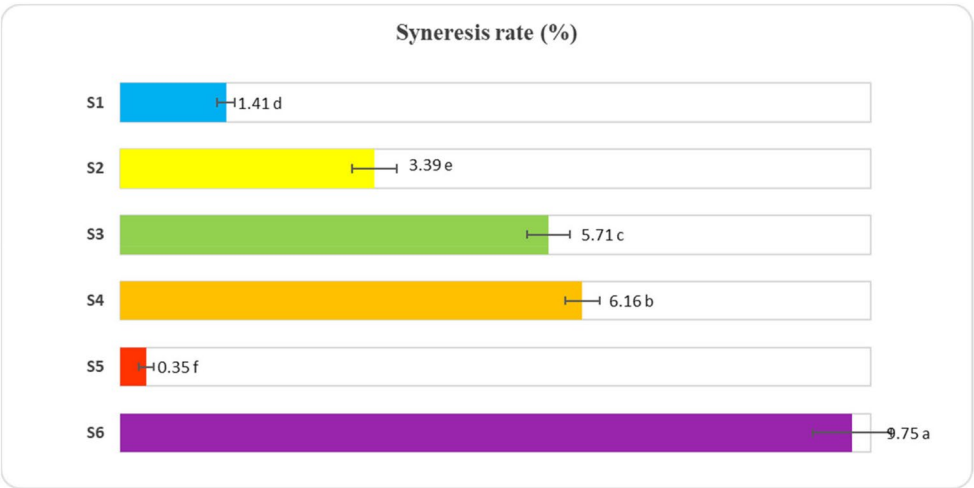

Fig. 4 Syneresis rates (%) of the six samples of hydrogels made with sorghum starch, chitosan and different proportions of citric acid (CA). Sample 1: 2% CA; Sample 2 3% CA; Sample 3: 4% CA; Sample 4: 6% CA; Sample 5: 10% CA; Sample 6: 0% CA. *Means followed by the same lowercase letter did not differ from each other in the mean-comparison test ($p \leq 0.05$)

on minimizing syneresis during the freeze-thaw process, reducing the water available to form ice crystals, leading to a significant decrease in the syneresis rates.

In this study, it was observed that sample 5, containing the highest concentration of citric acid at 10%, exhibited the lowest syneresis rate. On the other hand, sample 6, without the addition of citric acid in its composition, showed the highest syneresis rate, reaching 9.75%.

The level of citric acid exerts a direct and significant influence on syneresis rates in starch- and chitosan-based hydrogels, since it simultaneously affects the crosslinking density, the ionization state of the polymers, and the water retention capacity of the polymer network. At low to moderate concentrations, citric acid tends to reduce syneresis, while at high levels it can promote the opposite effect, depending on the system conditions. These findings demonstrate the influence of CA on the water retention of hydrogels, showing it plays a crucial role in the crosslinking process and formation of a three-dimensional network, contributing to a high water retention capacity [24, 27].

After the preparation of the 6 hydrogel samples, they were lyophilized, photographed (Fig. 5a), and then swollen in water for 24 h before being photographed again (Fig. 5b).

During the visual evaluation of the lyophilized hydrogels (Fig. 5a) and the swollen hydrogels (Fig. 5b), it was observed that the 6 samples presented a stable and intact structure after immersion in water. However, the samples with the addition of citric acid (1b, 2b, 3b, 4b and 5b) demonstrated greater resistance after swelling, highlighting the role of citric acid in enhancing gelatinization [9, 23].

Although synthetic polymer hydrogels exhibit excellent mechanical properties, they do not degrade easily, are toxic, and cause environmental pollution. Therefore, starch-based hydrogels have attracted great interest due to their biodegradability, biocompatibility, and non-toxicity. Chemical crosslinking usually requires toxic crosslinking agents and reacts at high temperatures. Therefore, the preparation of starch-based hydrogels by physical crosslinking is more attractive in the life sciences and biomedicine fields.

The preparation of starch-based hydrogels by physical methods is environmentally friendly and a healthy option for consumers.

Water Absorption Capacity of Hydrogels

The weight values in grams of the lyophilized (dry weight) and swollen hydrogels, as well as the difference between them, are shown in Fig. 6. All six formulations exhibited high water absorption, evidencing the characteristic high water absorption capacity of hydrogels.

Water absorption capacity is an important characteristic of hydrogels. However, water retention capacity, characterized by a lower syneresis rate, is a determining factor to evaluate hydrogel quality and the efficiency of the crosslinking process, which results in the formation of a three-dimensional network with high water absorption and retention capacity [22, 28].

These two samples (S3 and S4) presented superior visual characteristics compared to the others, because they formed a fluid, homogeneous, smooth gel without the presence of lumps, as shown in Fig. 3.

The physical cross-linking process and citric acid concentrations can interfere with the starch-chitosan interaction. As this interaction occurs only by hydrogen bonds, the presence of citric acid can increase the number of hydrogen bonds, thus improving the stability of the three-dimensional lattice characteristic of hydrogels [9, 22].

The solubility of starch is a result of the leaching of amylose, which dissociates and diffuses out of the starch granule during swelling, and the retrogradation process of starch, influenced by the amylose content and chain length of the molecules [29, 30].

In a study assessing the amylose content in white sorghum starch, cultivar BR 501, the same used in this study, Vieira et al. [19] found an amylose content of 26.88%, characterizing it as medium amylose content. This finding aligns with the results of water absorption capacity reported by Biduski et al. [9], who investigated water absorption capacity in low, medium and high amylose rice starch-based hydrogels.

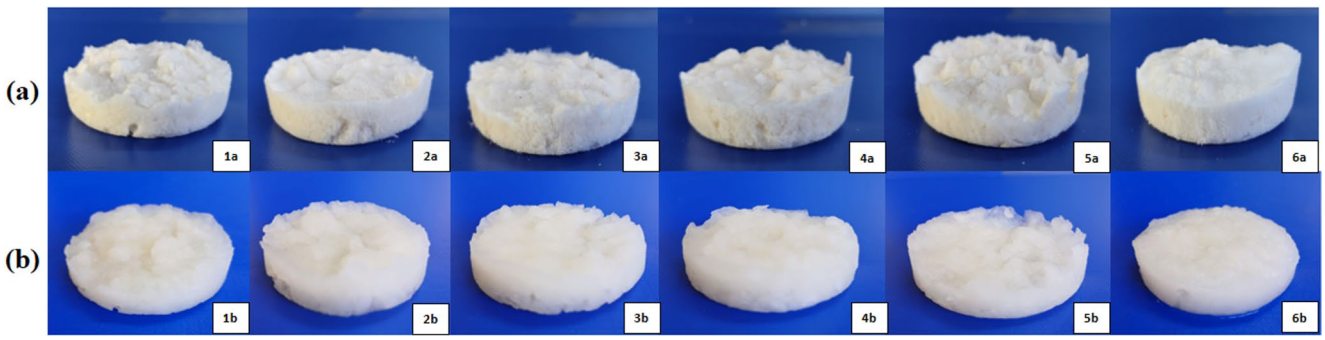

Fig. 5 Lyophilized (**a**) and swollen (**b**) hydrogels. 1a and 1b: 2% CA; 2a and 2b: 3% CA; 3a and 3b: 4% CA; 4a and 4b: 6% CA; 5a and 5b: 10% CA; 6a and 6b: 0% CA

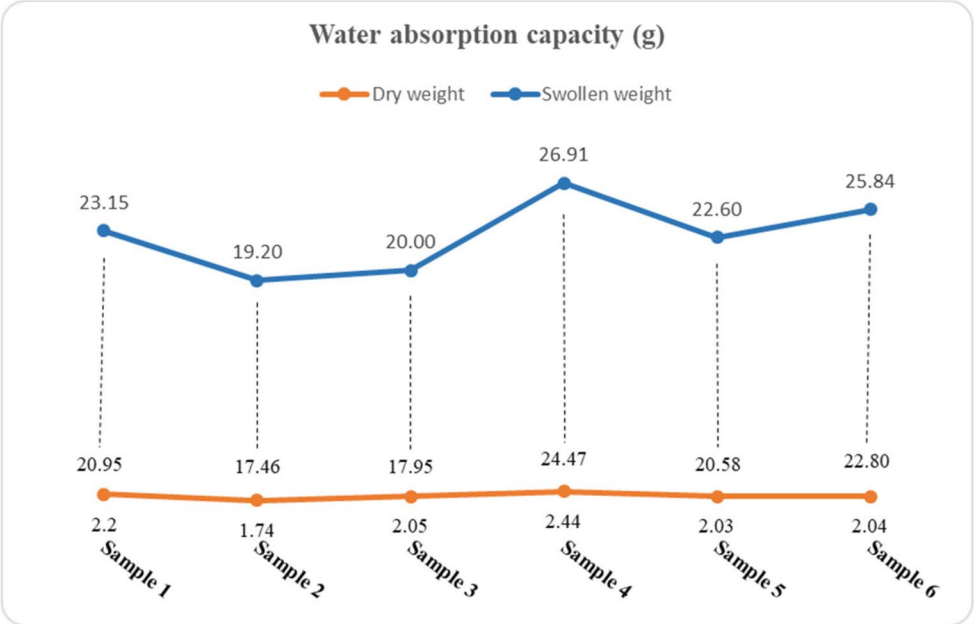

Fig. 6 Water absorption capacity of the six samples of hydrogels made with sorghum starch, chitosan and different proportions of citric acid (CA). Sample 1: 2% CA; Sample 2 3% CA; Sample 3: 4% CA; Sample 4: 6% CA; Sample 5: 10% CA; Sample 6: 0% CA

Hydrogels derived from low and medium amylose starch presented the best results, whereas the hydrogel containing high amylose content dissolved completely in water.

In a study that evaluated the degree of swelling of hydrogels based on corn starch and carboxymethylcellulose, values of 6.10 to 11.13 g/g of water absorption were reported [31], values that are lower than those found in the present study.

These factors demonstrate that sorghum starch (cultivar BR 501) is an excellent alternative for the elaboration of hydrogels with high water absorption capacity. In applications such as the controlled release of compounds, hydrogels with partial solubility and high water absorption capacity can be employed to regulate the release of bioactive agents [9]. Thus, the sample presenting the most favorable parameter concerning water absorption capacity in this study, with a low syneresis rate, was S3 with 4% citric acid.

Determination of water activity, water release and water holding capacity of hydrogels

Table 2 shows the values of water activity (aw), water release rate expressed in grams of water per 100 g of gel, and water holding capacity (WHC)

Water Activity (a_w)

Water activity (a_w) is an important measure of the free water available for microbial growth in food matrices. a_w is rated on a scale from 0 to 1, where 1 represents pure water [32, 33].

The a_w values among the samples ranged from 0.995 ± 0.002 to 0.999 ± 0.001, indicating a high a_w, as they were close to 1. Samples S1 (2% CA), S2 (3% CA), S3 (4% CA), S4 (6% CA), and S5 (10% CA) presented a_w values ranging from 0.995 ± 0.002 to 0.998 ± 0.001 and did not statistically differ from each other.

The a_w (0.995 ± 0.002) of sample S3 (4% CA) was only statistically different from the a_w (0.999 ± 0.001) of sample S6 (0% CA). High water activity values in hydrogels are expected, since they are polymer suspensions - in this study, starch - with high water retention capacity. In addition, starch hydrogels are characterized as a hydrophilic polymeric matrix with a three-dimensional structure cross-linked through physical or chemical processes. This structure allows for significant water or aqueous solution absorption while maintaining integrity without dissolving [3, 34].

Considering the high water activity values of the developed hydrogels, indicating a product highly susceptible to microbial growth, it is essential to incorporate compounds

Table 2 Water activity (Aw), water release rate, and water holding capacity (WHC) of hydrogels

Sample	Aw	Water release (%)	WHC (g)
S1 (2% AC)	0.996 ± 0.001 ab	0.08 ± 0.03 d	0.03 ± 0.01 b
S2 (3% AC)	0.996 ± 0.002 ab	0.13 ± 0.03 d	0.03 ± 0.00 b
S3 (4% AC)	0.995 ± 0.002 b	16.03 ± 0.21 c	0.16 ± 0.08 b
S4 (6% AC)	0.997 ± 0.001 ab	25.35 ± 1.90 a	0.41 ± 0.07 a
S5 (10% AC)	0.998 ± 0.001 ab	21.90 ± 1.11 b	0.44 ± 0.11 a
S6 (0% AC)	0.999 ± 0.001 a	0.14 ± 0.12 d	0.05 ± 0.02 b

* Mean ± standard deviation. Means followed by the same lowercase letter in the column did not differ from each other in the mean-comparison test ($p \leq 0.05$)

such as chitosan, which has been shown to have antifungal activity and antimicrobial properties during storage [35]. When it is solubilized in an acidic medium (e.g., citric acid), it adds another barrier to the growth of microorganisms [36], imparting an acidic characteristic to the developed hydrogels. Both chitosan and citric acid were used in the elaboration of the hydrogels in the present study.

Water release (%)

The release of water from the hydrogels varied significantly among the six samples, with values ranging from 0.08 ± 0.03 to 25.35 ± 1.90. Samples S1 (2% CA), S2 (3% CA) and S6 (0% CA) showed the lowest water release values (0.08 ± 0.03, 0.13 ± 0.03 and 0.14 ± 0.12%, respectively).

Maniglia et al. [28] developed hydrogels based on dry-heat modified wheat starch (DHT) and reported water release values ranging from 3.62 ± 0.13 to 7.38 ± 0.71 g water/100 g gel, which are similar to the findings of this study in formulations exhibiting lower syneresis.

Among all samples, S4 (6% CA) showed the highest water release value (25.35 ± 1.90%), while S3 (3% CA) and S5 (10% CA) showed 16.03 ± 0.21 and 21.90 ± 1.11% water release, respectively. All these values differed statistically from each other.

Wu wt al. [37] developed acryloyloxyethyl trimethylammonium chloride (AM/DAC) copolymer hydrogels and reported that water release values varied from 0 to 40.5% in the different formulations as a function of temperature. These findings are close to those found in this study, where the highest water release from the hydrogels reached 25.35%.

During the cooling of starch gels, molecular retrogradation and the tendency of amylose crystallization release water from the gels [28]. This parameter is crucial to evaluate the stability of the formed gel. Furthermore, starch-based hydrogels exhibit a strong effect in minimizing water release after the freeze-thaw process, reducing the availability of water for ice crystal formation and resulting in a significant decrease in the water release rate [22].

Water Holding Capacity (WHC)

The water holding capacity of the hydrogels was determined by the water loss value (WL) in grams, as shown in Table 2. WL values ranged from 0.03 to 0.44 g among the six hydrogel samples. Samples S1 (2% CA), S2 (3% CA), S3 (4% CA) and S6 (0% CA) exhibited the lowest WL (0.03 ± 0.01, 0.03 ± 0.00, 0.16 ± 0.08 and 0.05 ± 0.02 respectively), and these values are statistically equal. Such values differed from the WL of samples S4 (6% CA) and S5 (10% CA), which had the highest WL (0.41 ± 0.07 and 0.44 ± 0.11 respectively), and are statistically equal.

These results indicate that lower concentrations of citric acid are sufficient to enhance the water holding capacity of hydrogels, and may be related to the water release rate, since, as previously presented (Table 2), formulations S4 and S5 also showed a higher water release (25.35 and 21.90%, respectively).

This demonstrates that the water holding capacity is influenced by the concentration of citric acid among the different samples [9, 29]. However, higher concentrations (6 to 10%) worsen this capacity, indicating that concentrations between 2 and 4% of CA are sufficient to maintain the water holding capacity, as observed for the sample without the addition of CA, while improving other aspects of the gels (as discussed in the results presented in this study) and maintaining the water holding capacity.

Hydrogen potential (pH)

The hydrogel samples exhibited pH values ranging from 3.36 ± 0.01 to 7.66 ± 0.01, with statistical differences, as shown in Table 3. The acidic pH observed in the formulations with higher concentrations of CA (S2, S3, S4 and S5) may be attributed to the gelatinization process through heating with addition of citric acid.

The pH of the samples decreased as the concentration of citric acid increased, with S5 (10% CA) displaying the lowest pH value, indicating greater acidity (3.36 ± 0.01) and S1 (2% CA) presenting the highest pH (6.23 ± 0.01) among the samples with added citric acid. S6 (0% CA), serving as the control sample without citric acid, had a pH closer to neutrality (7.66 ± 0.01).

Lima et al. [38] developed hydrogels with cassava-parsley starch modified by ozone technology, resulting in pH ranging from 4.0 to 8.5, similar to those found in this study. Additionally [28], developed hydrogels based on wheat starch modified by dry heating treatment (DHT), and found pH values ranging from 5.90 ± 0.10 to 6.06 ± 0.15, similar

Table 3 Hydrogen Potential (pH) of hydrogels. *Mean ± standard deviation. Means followed by the same lowercase letter in the column did not differ from each other in the mean-comparison test ($p \leq 0.05$)

Sample	pH
S1 (2% AC)	6.23 ± 0.01 b
S2 (3% AC)	5.19 ± 0.00 c
S3 (4% AC)	4.30 ± 0.01 d
S4 (6% AC)	3.82 ± 0.01 e
S5 (10% AC)	3.36 ± 0.01 f
S6 (0% AC)	7.66 ± 0.01 a

*Mean ± standard deviation. Means followed by the same lowercase letter in the column did not differ from each other in the mean-comparison test ($p \leq 0.05$)

to the values from samples S1 (2% CA) and S2 (3% CA) of the present study.

In another study developed by Maniglia et al. [38] with ozonated cassava starch hydrogels, the authors reported pH values of 5.23 ± 0.11, 5.32 ± 0.17 and 5.45 ± 0.19, values close to those measured in this study for sample S2 with 3% citric acid.

Maniglia et al. [39] also developed hydrogels based on ozonated cassava starch and found pH values of 4.00 and 5.00. The authors associated the reduction in pH with a higher content of carboxylates, a functional group of acidic nature.

It is well known that the amylose and amylopectin (glucose chains) of starch are hydrolyzed by the addition of acids [40]. The addition of an acidic medium, such as citric acid dissolved in distilled water, can aid in the development of pH-sensitive hydrogels, which can be used to modulate the release of bioactive compounds, besides minimizing the growth of microorganisms [6, 36].

Therefore, the elaboration of starch hydrogels in acidic medium (with the addition of citric acid, for example), shows promise in improving the gelatinization of starch and chitosan suspensions, as demonstrated in the present study.

Morphological evaluation of hydrogels

Morphology enables the subjective evaluation of the formation of porous or non-porous structures in the three-dimensional network of hydrogels [9]. Figure 7 illustrates the internal structure of hydrogels made with physically cross-linked sorghum starch, chitosan and different concentrations of citric acid.

In general, the hydrogels presented micropores with irregular and regular shapes, as well as a spongy and rough surface. According to Lima-Tenório et al. [41], this type of morphology within hydrogels creates a pathway along the entire structure, allowing the transport of water through the hydrogel. This phenomenon possibly increases water absorption and may also result from the formation and growth of ice crystals during freezing [42, 43].

A study by Azadikhah & Karimi [44] evaluating polyvinyl alcohol (PVA), chitosan and tannic acid hydrogels, demonstrated that all hydrogels presented spongy structures, with nanometric pore walls. The authors attributed this behavior to physical crosslinking, suggesting that the freezing and thawing of PVA-based hydrogels promoted the formation of hydrogen bonds between polymer chains,

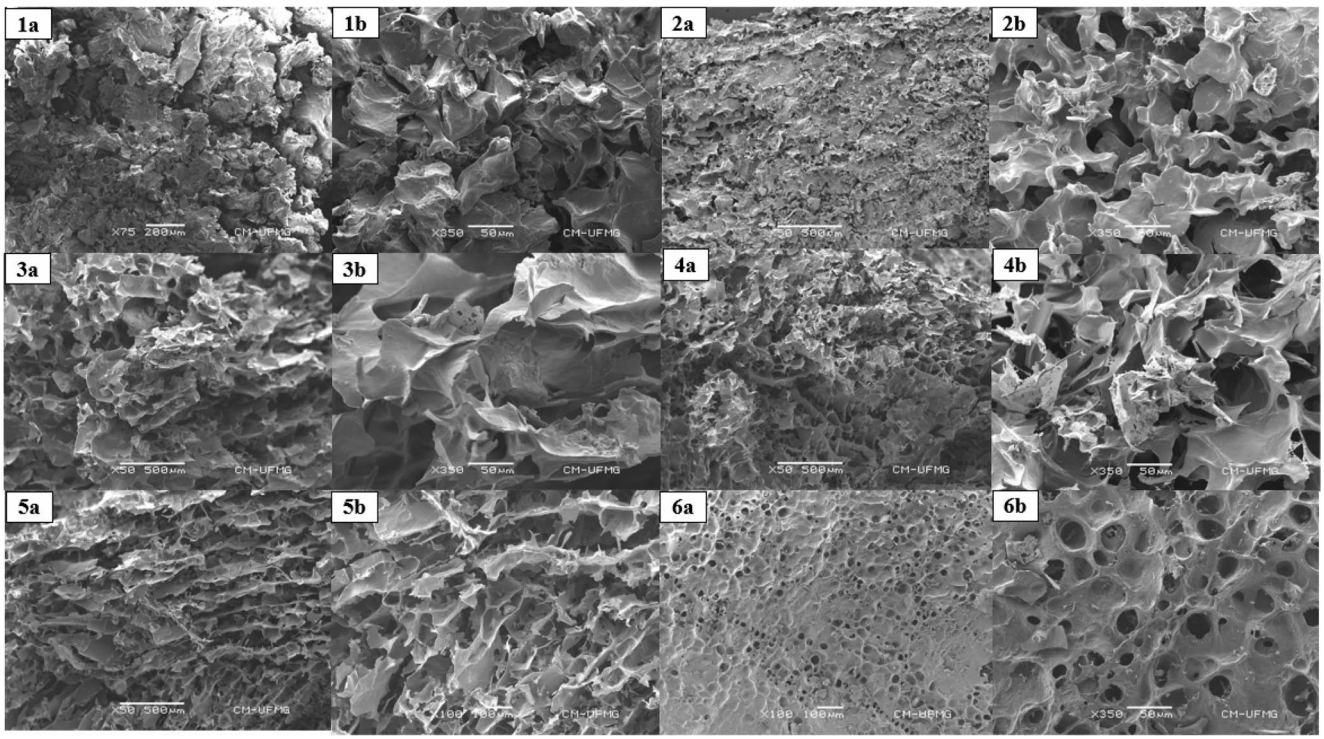

Fig. 7 Morphology of sorghum starch hydrogels with chitosan and different proportions of citric acid (CA). Images at different magnifications (1a 2% CA X75; 1b 2% CA X350; 2a 3% CA X50; 2b 3% CA X350; 3a 4% CA X50; 3b 4% CA X350; 4a 6% CA X50; 4b 6% CA X350; 5a 10% CA X50; 5b 10% CA X100; 6a 0% CA X100; 6b 0% CA X350)

Table 4 Texture analysis profile of hydrogels made with sorghum starch, chitosan and different proportions of citric acid (CA)

Sample						
Parameters	S1 (2% CA)	S2 (3% CA)	S3 (4% CA)	S4 (6% CA)	S5 (10% CA)	S6 (0% CA)
Hardness	438.16 ± 194.70 a	199.09 ± 44.22 b	335.14 ± 79.39 ab	261.72 ± 66.69 ab	136.58 ± 12.29 b	218.58 ± 46.86 b
Cohesiveness	0.34 ± 0.01 b	0.40 ± 0.03 b	0.37 ± 0.02 b	0.36 ± 0.07 b	0.56 ± 0.07 a	0.38 ± 0.01 b
Gumminess	144.81 ± 58.63 a	80.00 ± 16.78 b	122.63 ± 22.97 ab	93.00 ± 19.96 ab	75.33 ± 5.09 b	83.57 ± 15.11 ab
Resilience	0.04 ± 0.01 a	0.04 ± 0.00 a	0.05 ± 0.00 a	0.05 ± 0.01 a	0.04 ± 0.00 a	0.04 ± 0.01 a
Adhesion	124.36 ± 29.61 a	129.37 ± 49.88 a	177.57 ± 65.86 a	96.02 ± 24.51 a	138.12 ± 45.14 a	102.50 ± 44.22 a
Flexibility	0.95 ± 0.02 a	0.94 ± 0.02 ab	0.95 ± 0.03 a	0.90 ± 0.03 b	0.94 ± 0.01 ab	0.92 ± 0.01 ab

*Mean ± standard deviation. Means followed by the same lowercase letter in the column did not differ from each other in the mean-comparison test ($p \leq 0.05$)

resulting in the formation of highly porous structures. These characteristics were also observed in the present study.

In Fig. 5 (a) and (b), associated with the higher concentration of citric acid (CA), it is possible to observe overlapping micropores that form an interconnected network, which is more numerous and arranged in a more orderly manner. This morphological characteristic, as explained by Biduski et al. [9], allows the formation of a continuous path throughout the structure, facilitating water transport through the hydrogel and, consequently, contributing to the increase in its absorption capacity. Moreover, the network of interconnected pores facilitates solute diffusion and efficient mass transfer in various applications [45].

The hydrogel made with sorghum starch and chitosan, without the addition of CA, presented more uniform interconnected micropores, with regular rounded pore geometry. These pores are arranged on a denser and smoother surface, as evidenced in Fig. 5 (a) and (b). Chen et al. [46] highlighted in their research that the native corn starch hydrogel also displayed an interconnected network in its internal morphology, similar to that observed in this study.

However, the presence of larger pores makes it easier for water to penetrate the structure of the hydrogels, increasing their swelling capacity [47], This was observed in all hydrogels made with citric acid (Figs. 1A and B, 2A and B, 3A and B, 4A and B and 5A and B), highlighting the crucial role of this component in intensifying crosslinking, which promotes the formation of a three-dimensional network in the developed hydrogels with high water absorption capacity [20, 48].

Figure 5a and b show that as the concentration of citric acid increases, the formation of the network becomes more pronounced, resulting in larger and more numerous pores. Consequently, the water absorption and retention capacity of the hydrogels increase, as previously shown in Tables 2 and 3.

Currently, citric acid has been gaining a lot of attention from researchers because of its biocompatibility, versatile chemistry, availability, and low cost, and has been considered a green crosslinker. Due to its three carboxylic groups (COO-) and one hydroxyl group (-OH), citric acid can actively participate in the interactions of hydrogen bonds with other polymer networks and improve their characteristics [25, 27].

Thus, the morphological analysis of physically cross-linked hydrogels made from sorghum starch, chitosan and varying concentrations of citric acid, demonstrated the efficiency of the physical cross-linking process in promoting the development of the three-dimensional polymeric network. Furthermore, the addition of citric acid enhanced this formation, resulting in the creation of highly porous hydrogels, with a high capacity for water absorption and retention.

Texture Analysis Profile (TPA)

The texture profiles of the hydrogels based on sorghum starch, chitosan and different citric acid concentrations are presented in Table 4. The 6 hydrogel samples showed similar behavior in terms of resilience and adhesion, with no significant differences observed among them ($p > 0.05$). However, there was a significant difference between the samples in terms of hardness, cohesiveness, gumminess and flexibility.

The texture of gels is assessed based on several parameters such as hardness, cohesion, gumminess, resilience, adhesion, and flexibility. Hardness and cohesion determine the structural integrity of the gels, while the other parameters derive from them. Hardness is defined as the maximum force required to deform a specimen during initial compression [49, 50].

The hardness values of the hydrogels ranged from 136.58 ± 12.29 to 438.16 ± 194.70. Sample S1, featuring the lowest CA concentration (2%), exhibited a statistical difference from S2 (3% CA), S5 (10% CA) and S6 (0% CA), indicating higher hardness. This demonstrates that even a minor addition of CA was sufficient to produce a firmer gel, due to the crosslinking process intensified by this acid. However, this result also demonstrated that increasing the concentration of citric acid did not influence the firmness of the hydrogels, with the values remaining statistically

equivalent between the S1, S3 and S4 samples, containing 2, 4 and 6% of CA, respectively.

These values were higher and statistically equal in the samples S1 (2% CA), S3 (4% CA) and S4 (6% CA), which shows that these samples presented a firmer gel compared to the others. Zhu et al. [51], observed hardness values ranging from 20.9 to 62.7 when evaluating gels based on potato, sweet potato and quinoa starches. These values were lower than those found in the present study, which demonstrates that hydrogels made with sorghum starch are firmer.

In a study by Biduski et al. [9] that evaluated the texture profile of rice starch hydrogels, both native and intercrossed, through the heating gelatinization method, hardness values of 41.94 for the native starch and 563.24 for the intercrossed one were reported. This demonstrates that the intercrossing process enabled the formation of more rigid hydrogels. This result corroborates the findings of the present study regarding the same parameter, indicating that even with native sorghum starch, physical crosslinking via freeze-thaw cycles efficiently produced firm gels. In addition, gels with higher hardness are associated with greater structural integrity, while those with lower hardness tend to lose structure over time [50].

The textural parameter of cohesiveness, also evaluated in the TPA of hydrogels, is related to the measurement of the strength of internal bonds forming the final structure of the product. This parameter can be used as an index to assess the overall acceptability of starchy food products. A lower degree of cohesion indicates high plasticity or low elasticity of the gels [50, 52].

Cohesiveness values ranged from 0.34 ± 0.01 to 0.56 ± 0.07 among the analyzed samples. In a study that evaluated this same parameter, in rice starch hydrogels, both native and intercrossed, using the heating gelatinization method, Biduski et al. [9] found values of 0.56 for the native starch and 0.71 for the intercrossed one. They associated this result with the low cohesiveness of the native starch hydrogels and the weak intermolecular association.

This result was similar to that found in this study, where the highest cohesiveness value (0.56 ± 0.07) was observed for sample S5, which featured the highest concentration of citric acid compared to the others. This demonstrates that a higher concentration of CA was effective in intensifying the internal bonds of sorghum starch and chitosan, promoting more robust interactions between the chains. In addition, the pH value may also have influenced this result. The reduction in pH, resulting from the greater addition of citric acid in the present study, may lead to the formation of gels that are difficult to deform [40]. Zhu et al. [51] observed cohesiveness values ranging from 0.34 to 0.68 when assessing gels based on potato, sweet potato and quinoa starches, which are similar to those found in this study.

Regarding gumminess, this textural parameter is important for semi-solid food products, and it is calculated by multiplying the hardness and cohesiveness values. Higher gumminess values indicate greater hardness and less cohesion in food products [53].

Biduski et al. [9] reported a gumminess value of 19.71 when evaluating the same parameter in rice starch hydrogels, which is lower than the value reported in this study. In another study that developed hydrogels based on millet starch [50], found gummy texture parameter values ranging from 11.81 to 52.01. This demonstrates that the hydrogels based on sorghum starch, chitosan and different concentrations of citric acid, elaborated in the present research, resulted in firmer gels.

Gumminess refers to the energy required for the disintegration of food and is directly related to the amylose content of starch, that is, the higher the amylose content, the greater the force needed for the disintegration of the gel. This phenomenon occurs because linear chains of amylose tend to form stronger intermolecular bonds. However, it is worth mentioning that excessively high gumminess values can make the product unacceptable to consumers [50].

The sorghum starch used in the present study for hydrogel production contains an average of 26.88% of amylose, categorizing it as having a medium amylose content [19]. This characteristic justifies the high gumminess values found in the samples. Thus, the samples with the lowest gumminess values were S2, S3, S4, S5 and S6, which were statistically equivalent. This shows that the addition of citric acid did not affect the gumminess of the hydrogels, and these samples presented the most favorable results for the gummy texture parameter.

Regarding the textural parameters, resilience and adhesion, no statistically significant difference was observed among the six hydrogel samples. This suggests that the concentration of citric acid did not affect these parameters.

Resilience measures the ability of the sample to recover after compression, concerning both derived force and velocity, while adhesion is related to the formation of a more resistant gel structure. In other words, it is related to the force that the gel exerts on the equipment, preventing it from reverting to its original state [9].

These results highlight that hydrogels based on sorghum starch, chitosan, with varying concentrations of citric acid, were able to resist the compressive forces and recover their initial size and shape after the TPA tests. This indicates good integrity and high strength, regardless of the citric acid concentration [54].

When analyzing the textural parameter of flexibility, the observed values ranged from 0.92 ± 0.01 to 0.95 ± 0.02, with a statistically significant difference only between samples S1 (2% CA) and S4 (6% CA), the former being higher than

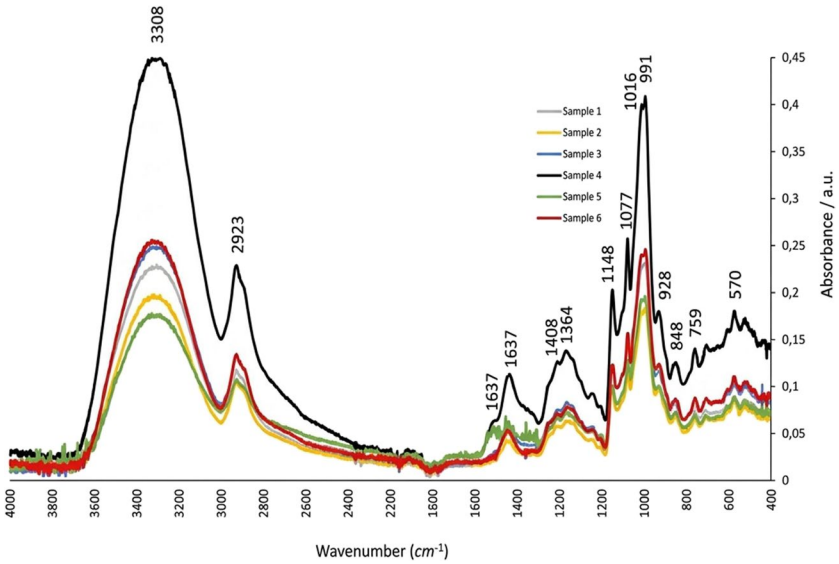

Fig. 8 FTIR spectra of the six samples of hydrogels made with sorghum starch, chitosan and different proportions of citric acid (CA). Sample 1: 2% CA; Sample 2: 3% CA; Sample 3: 4% CA; Sample 4: 6% CA; Sample 5: 10% CA; Sample 6: 0% CA

the latter. These results indicate that the concentration of citric acid did not influence the flexibility of the hydrogels, since the sample without citric acid addition (S6) presented results statistically equivalent to the sample with the highest concentration of citric acid (S5, 10% CA). Flexibility corresponds to the ability of the sample to return to its original state after compression.

Fourier-transform Infrared Spectroscopy (FTIR)

The ATR-FTIR spectra of the six formulations based on sorghum starch, chitosan and different citric acid (CA) ratios are shown in Fig. 8.

The infrared analysis of the developed hydrogels made it possible to identify specific characteristics of starch and citric acid. The identification of starch proved to be simpler due to its higher proportion.

The presence of starch was indicated by the following bands: 3308 cm^{-1}, attributed to the O-H stretch, having high amplitude due to intermolecular interactions; 3000 and 2800 cm^{-1}, assigned to the C-H stretch; 1637 cm^{-1}, assigned to a bending band of the hydroxyl group of the water, indicating the hygroscopic profile of the starch; 1459 and 1350 cm^{-1}, assigned to the angular deformation of C-H groups; 1060 and 990 cm^{-1}, being characteristic of polysaccharides and attributed to deformations of the C-O-C group and flexion of the O-H group; 1016 cm^{-1}, attributed to the C-OH stretch; 575 cm^{-1}, attributed to the vibration mode of the pyran ring of starch glucose.; 1639 cm^{-1}, attributed to the C = O stretch; and 994 cm-^{1}, assigned to the stretch of the C-O-H group [55, 56].

Citric acid was identified by the band at 1696 cm^{-1}, attributed to one of its C = O stretches. The inability to identify chitosan was due to its low proportion in the mixture and the similarity of the frequency values of some of its bands with those of starch [57].

Finally, when comparing the proportions of the three constituents of the samples and the results of the infrared spectra, it was observed that samples 4 and 5, which had a higher proportion of citric acid, exhibited the band at 1696 cm^{-1}, assigned to the stretch of one of its C = O groups. Therefore, this acid can be identified via infrared spectroscopy with ATR sampler for mixtures containing 0.3 g or more of citric acid in 5 g of starch and 0.5 g of chitosan.

The combination of starch, chitosan, citric acid, and water results in the formation of a hybrid hydrogel whose three-dimensional structure is stabilized by different types of intermolecular and intramolecular interactions. The polymeric network formed is mainly supported by hydrogen bonds between the hydroxyl groups of starch, water, and chitosan; by electrostatic interactions resulting from the protonation of the amine groups of chitosan in an acidic medium; and by ester-type covalent bonds formed between the carboxyl groups of citric acid and the hydroxyl groups of the polysaccharides. This combination of interactions gives the hydrogel greater structural, thermal, and mechanical stability, as well as allowing control of water absorption capacity and the degree of swelling [15, 44].

Conclusion

The developed hydrogels exhibited homogeneous morphology, a smooth surface, and high water absorption and retention capacity, demonstrating the efficiency of physical crosslinking through freeze-thaw cycles, reinforced by the addition of citric acid. FTIR analyses confirmed the

composition of the formulations, while the texture profile demonstrated the formation of firm, cohesive gels resistant to compression. Among the formulations evaluated, S3 and S4, containing intermediate concentrations of citric acid (4–6%), stood out for presenting better homogeneity, adequate pH, and absence of lumps, being considered the most promising for different applications.

Finally, the results obtained demonstrate that sorghum starch presents high viability as a polymeric matrix for the development of hydrogels. The combination with chitosan and physical crosslinking with citric acid conferred promising characteristics to the material, such as stability and potential for modulating the release of active ingredients. In this sense, future studies should explore the incorporation of bioactive compounds, as well as evaluate the performance of these hydrogels in food applications, including encapsulation systems and controlled release of functional ingredients, aiming to expand their use in sustainable and value-added technologies.

Limitations

Developing a sorghum starch-based hydrogel faces specific limitations that impact both the technical feasibility and the scientific contribution of the work. Firstly, the low extraction yield of sorghum starch constitutes a significant initial barrier. Compared to conventional sources such as corn or potato, sorghum often provides smaller amounts of starch per unit of biomass, due to the inherent characteristics of its grain composition and the greater presence of non-starch components. As a future perspective, the sorghum starch extraction process needs to be optimized in order to increase the extraction yield.

As this is the first study to develop a sorghum starch-based hydrogel, another relevant limitation is the scarcity of comparable studies in the scientific literature. While hydrogels based on starches from widely studied sources (such as corn, cassava, and potato) have extensive bodies of data on physicochemical properties, crosslinking mechanisms, and functional performance, sorghum starch is less represented.

These limitations, however, do not impede research, but they do require more careful approaches to experimental design, data extraction optimization, and critical interpretation, often necessitating the establishment of indirect comparison criteria or the development of internal reference models. Overcoming these challenges can, on the other hand, position studies on sorghum starch hydrogels as an original and relevant contribution to the scientific community, especially in areas focused on sustainable materials and alternative polymer sources.

Acknowledgements The authors would like to thank the Microscopy Center of the Universidade Federal de Minas Gerais, the Department of Chemistry of the Federal Center for Technological Education of Minas Gerais, and the Department of Pharmaceutical Products of the School of Pharmacy, Universidade Federal de Minas Gerais, for the analysis of microscopy, FTIR and water activity, respectively. The authors would also like to thank Embrapa Maize and Sorghum for the supply of sorghum grains and to the Coordination for the Improvement of Higher Education Personnel - CAPES (88887.503309/2020-00) for the financial support.

Funding The Article Processing Charge (APC) for the publication of this research was funded by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) (ROR identifier: 00x0ma614).

Data availability Data sharing does not apply to this article as no new data were created or analyzed in this study.

Declarations

Competing interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

References

1. P. Dahal, S. Janaswamy, Hydrocolloid-based nutraceutical delivery systems: Potential of κ-carrageenan hydrogel beads for sustained release of curcumin. Food Res. Int. **183**, 114223 (2024). https://doi.org/10.1016/J.FOODRES.2024.114223
2. K. Liu, Y.-Y. Chen, X.-Q. Zha, Q.-M. Li, L.-H. Pan, J.-P. Luo, Research progress on polysaccharide/protein hydrogels: Preparation method, functional property and application as delivery systems for bioactive ingredients. Food Res. Int. **147**, 110542 (2021). https://doi.org/10.1016/j.foodres.2021.110542
3. J.S. dos Santos, B. Biduski, R. Colussi, V.Z. Pinto, L.R. dos Santos, Hydrogel properties of non-conventional starches from guabiju, pinhão, and uvaia seeds. Food Res. Int. **173**, 113243 (2023). https://doi.org/10.1016/J.FOODRES.2023.113243
4. Y. Qin, J. Wang, C. Qiu, X. Xu, Z. Jin, Cross-Linked Strategy to Construct Moldable Hydrogels with High Stretchability, Good Self-Recovery, and Self-Healing Capability. J. Agric. Food Chem. **67**, 3966–3980 (2019). https://doi.org/10.1021/acs.jafc.8b05147
5. C. Cui, Y. Jia, Q. Sun, M. Yu, N. Ji, L. Dai, Y. Wang, Y. Qin, L. Xiong, Q. Sun, Recent advances in the preparation, characterization, and food application of starch-based hydrogels. Carbohydr. Polym. **291** (2022). https://doi.org/10.1016/j.carbpol.2022.119624
6. G.G. De Lima, R.O. De Souza, A.D. Bozzi, M.A. Poplawska, D.M. Devine, M.J.D. Nugent, Extraction Method Plays Critical

Role in Antibacterial Activity of Propolis-Loaded Hydrogels. J. Pharm. Sci. **105**, 1248–1257 (2016). https://doi.org/10.1016/J.XPHS.2015.12.027
7. A.L.S. Vieira, R.C.S. De Sousa, V.T. Correia, V.A.V. Queiroz, W.A. Da Silva, M.A.V.T. Garcia, R.L.B. De, Araújo, Starch-based hydrogels: current status and applications in food. Contribuciones Las Ciencias Sociales. **16**, 28707–28727 (2023). https://doi.org/10.55905/revconv.16n.11-237
8. K.C. Guedes Silva, G. Feltre, M.D. Hubinger, A.C. Kawazoe Sato, Protection and targeted delivery of beta-carotene by starch-alginate-gelatin emulsion-filled hydrogels. J. Food Eng. **290** (2021). https://doi.org/10.1016/j.jfoodeng.2020.110205
9. B. Biduski, W.M.F. da Silva, R. Colussi, S.L. de Halal, L.T. Lim, Á.R.G. Dias, E. Zavareze, Starch hydrogels: The influence of the amylose content and gelatinization method. Int. J. Biol. Macromol. **113**, 443–449 (2018). https://doi.org/10.1016/J.IJBIOMAC.2018.02.144
10. S. Mun, Y.-R. Kim, M. Shin, D.J. McClements, Control of lipid digestion and nutraceutical bioaccessibility using starch-based filled hydrogels: Influence of starch and surfactant type. Food Hydrocoll. **44**, 380–389 (2015). https://doi.org/10.1016/j.foodhyd.2014.10.013
11. Z. Huang, C.S. Brennan, M.S. Mohan, L. Stipkovits, H. Zheng, D. Kulasiri, W. Guan, H. Zhao, J. Liu, Milk lipidin vitrodigestibility in wheat, corn and rice starch hydrogels. Int. J. Food Sci. Technol. **55**, 3361–3371 (2020). https://doi.org/10.1111/ijfs.14607
12. R. Meng, Z. Wu, H.-Q. Xie, G.-X. Xu, J.-S. Cheng, B. Zhang, Preparation, characterization, and encapsulation capability of the hydrogel cross-linked by esterified tapioca starch. Int. J. Biol. Macromol. **155**, 1–5 (2020). https://doi.org/10.1016/j.ijbiomac.2020.03.141
13. D.C. Lima, J. Villar, N. Castanha, B.C. Maniglia, M.D. Matta Junior, P.E. Duarte, Augusto, Ozone modification of arracacha starch: Effect on structure and functional properties. Food Hydrocoll. **108**, 106066 (2020). https://doi.org/10.1016/j.foodhyd.2020.106066
14. A. López Córdoba, L. Deladino, M. Martino, Effect of starch filler on calcium-alginate hydrogels loaded with yerba mate antioxidants. Carbohydr. Polym. **95**, 315–323 (2013). https://doi.org/10.1016/j.carbpol.2013.03.019
15. L. de Soares, B.T. Gomes, G.L. Milião, R.A. da Rocha, A.V.N. de Teixeira, J.S.D.R. Coimbra, E.B. de Oliveira, Mixed starch/chitosan hydrogels: elastic properties as modelled through simulated annealing algorithm and their ability to strongly reduce yellow sunset (INS 110) release. Carbohydr. Polym. **255**, 117526 (2021). https://doi.org/10.1016/j.carbpol.2020.117526
16. B. Wu, D.J. McClements, Microgels formed by electrostatic complexation of gelatin and OSA starch: Potential fat or starch mimetics. Food Hydrocoll. **47**, 87–93 (2015). https://doi.org/10.1016/j.foodhyd.2015.01.021
17. V. Correia, D. D'Angelis, V. Queiroz, A.L. Ramos, M.C. Macedo, R. Augusti, J. Melo, A. Ferreira, I. Lacerda, C. Fante, Impact of Gamma Irradiation on Physicochemical, Technological, Antioxidant and Microbiology Properties of Whole Sorghum Flours. J. Braz Chem. Soc. (2022). https://doi.org/10.21577/0103-5053.20220097
18. B. Biduski, F.T. da Silva, W.M. da Silva, S.L. de Halal, V.Z. Pinto, A.R.G. Dias, E. Zavareze, Impact of acid and oxidative modifications, single or dual, of sorghum starch on biodegradable films, Food Chem. **214,** 53–60 (2017) https://doi.org/10.1016/J.FOODCHEM.2016.07.039
19. A.L.S. Vieira, G.B. Duarte, V.A.V. Queiroz, T.R. Correa, V.D.M. Silva, R.L.B. Araújo, M.A.V.T. Garcia, C.A. Fante, Caracterização do amido isolado de diferentes cultivares de sorgo (Sorghum bicolor L. Moench). Brazilian J. Dev. **6**, 24704–24718 (2020). https://doi.org/10.34117/bjdv6n5-067
20. C. Demitri, R. Del Sole, F. Scalera, A. Sannino, G. Vasapollo, A. Maffezzoli, L. Ambrosio, L. Nicolais, Novel superabsorbent cellulose-based hydrogels crosslinked with citric acid. J. Appl. Polym. Sci. **110**, 2453–2460 (2008). https://doi.org/10.1002/app.28660
21. F.T. da Silva, J.P. de Oliveira, L.M. Fonseca, G.P. Bruni, E.R. Zavareze, A.R.G. Dias, Physically cross-linked aerogels based on germinated and non-germinated wheat starch and PEO for application as water absorbers for food packaging. Int. J. Biol. Macromol. **155**, 6–13 (2020). https://doi.org/10.1016/J.IJBIOMAC.2020.03.123
22. J. Li, M. Zhu, L. Gu, Y. Su, Y. Yang, C. Chang, Q. Han, Freeze-thaw stability of konjac glucomannan hydrogels supplemented with natural tapioca/corn starch. LWT. **182**, 114841 (2023). https://doi.org/10.1016/J.LWT.2023.114841
23. M.N. Saqib, B.M. Khaled, F. Liu, F. Zhong, Hydrogel beads for designing future foods: Structures, mechanisms, applications, and challenges. Food Hydrocoll. Health. **2**, 100073 (2022). https://doi.org/10.1016/J.FHFH.2022.100073
24. A.P.C. Almeida, J.N. Saraiva, G. Cavaco, R.P. Portela, C.R. Leal, R.G. Sobral, P.L. Almeida, Crosslinked bacterial cellulose hydrogels for biomedical applications. Eur. Polym. J. **177**, 111438 (2022). https://doi.org/10.1016/J.EURPOLYMJ.2022.111438
25. S.H. Aswathy, U. NarendraKumar, I. Manjubala, The influence of molecular weight of cellulose on the properties of carboxylic acid crosslinked cellulose hydrogels for biomedical and environmental applications. Int. J. Biol. Macromol. **239**, 124282 (2023). https://doi.org/10.1016/J.IJBIOMAC.2023.124282
26. T.B. da Costa, P.H. Camani, R.R. Ferreira, R.F.S. Barbosa, D. dos S. Rosa, Enhancing corn starch hydrogels for effective sorption of potentially toxic metals: the role of amylose and amylopectin content, J. Polym. Environ. 33 (2025) 1615–1635. https://doi.org/10.1007/s10924-025-03495-7
27. R. Salihu, S.I. Abd Razak, N. Ahmad Zawawi, M. Rafiq Abdul Kadir, N. Izzah Ismail, N. Jusoh, M. Riduan Mohamad, Hasraf Mat Nayan, Citric acid: A green cross-linker of biomaterials for biomedical applications. Eur. Polym. J. **146**, 110271 (2021). https://doi.org/10.1016/J.EURPOLYMJ.2021.110271
28. B.C. Maniglia, D.C. Lima, M. da Matta, A. Júnior, P. Oge, P.E.D. Le-Bail, A. Augusto, Le-Bail, Dry heating treatment: A potential tool to improve the wheat starch properties for 3D food printing application. Food Res. Int. **137** (2020). https://doi.org/10.1016/j.foodres.2020.109731
29. C. Jiang, Y. Chao, W. Xie, D. Wu, Using bacterial cellulose to bridge covalent and physical crosslinks in hydrogels for fabricating multimodal sensors. Int. J. Biol. Macromol. **263**, 130178 (2024). https://doi.org/10.1016/J.IJBIOMAC.2024.130178
30. B. Klein, V.Z. Pinto, N.L. Vanier, E.D.R. Zavareze, R. Colussi, J.A. Do Evangelho, L.C. Gutkoski, A.R.G. Dias, Effect of single and dual heat–moisture treatments on properties of rice, cassava, and pinhao starches. Carbohydr. Polym. **98**, 1578–1584 (2013). https://doi.org/10.1016/J.CARBPOL.2013.07.036
31. C. Cagnin, B.M. Simões, F. Yamashita, A.C. Andrello, G.M. de Carvalho, M.V.E. Grossmann, Hydrogels of starch/carboxymethyl cellulose crosslinked with sodium trimetaphosphate via reactive extrusion. J. Appl. Polym. Sci. **138** (2021). https://doi.org/10.1002/app.50194
32. A. Zhang, H. Wang, X. Li, J. Li, Development of certified reference materials of lithium chloride solution for water activity. Food Chem. **437**, 137824 (2024). https://doi.org/10.1016/J.FOODCHEM.2023.137824
33. S. Sun, R. Yang, Y. Xie, M.-J. Zhu, S. Sablani, J. Tang, The effect of water activity on thermal resistance of Salmonella in chocolate

products with different fat contents. Food Control. **162**, 110443 (2024). https://doi.org/10.1016/J.FOODCONT.2024.110443
34. Y. Chen, Y. Hao, S. Li, Z. Luo, Q. Gao, Preparation of hydroxybutyl starch with a high degree of substitution and its application in temperature-sensitive hydrogels. Food Chem. **355**, 129472 (2021). https://doi.org/10.1016/J.FOODCHEM.2021.129472
35. L. Fernandes, E.L. Pereira, M. do Céu, A. Fidalgo, E. Gomes, Ramalhosa, Physicochemical properties and microbial control of chestnuts (Castanea sativa) coated with whey protein isolate, chitosan and alginate during storage. Sci. Hortic. **263**, 109105 (2020). https://doi.org/10.1016/J.SCIENTA.2019.109105
36. B. Wu, Q. Ren, L. Xia, Y. Liu, X. Cui, A. Dai, T. Wei, Y. Zhou, pH-dependent microbial niches succession and antibiotic resistance genes distribution in an oxygen-based membrane biofilm reactor treating greywater. Environ. Res. **216**, 114725 (2023). https://doi.org/10.1016/J.ENVRES.2022.114725
37. W. Wu, J. Hou, G. Li, L. Chen, Effect of temperature and inorganic salts concentration on syneresis rate of AM/DAC hydrogel, Colloids Surf. Physicochem Eng. Asp. **641**, 128561 (2022). https://doi.org/10.1016/j.colsurfa.2022.128561
38. B.C. Lima, D.C. Maniglia, M.D. Matta Junior, P. Le-Bail, A. Le-Bail, P.E.D. Augusto, Preparation of cassava starch hydrogels for application in 3D printing using dry heating treatment (DHT): A prospective study on the effects of DHT and gelatinization conditions. Food Res. Int. **128**, 108803 (2020). https://doi.org/10.1016/j.foodres.2019.108803
39. B.C. Maniglia, D.C. Lima, M.D. Matta Junior, P. Le-Bail, A. Le-Bail, P.E.D. Augusto, Hydrogels based on ozonated cassava starch: Effect of ozone processing and gelatinization conditions on enhancing 3D-printing applications. Int. J. Biol. Macromol. **138**, 1087–1097 (2019). https://doi.org/10.1016/j.ijbiomac.2019.07.124
40. M. Hirashima, R. Takahashi, K. Nishinari, The gelatinization and retrogradation of cornstarch gels in the presence of citric acid. Food Hydrocoll. **27**, 390–393 (2012). https://doi.org/10.1016/J.FOODHYD.2011.10.011
41. M.K. Lima-Tenório, E.T. Tenório-Neto, M.R. Guilherme, F.P. Garcia, C.V. Nakamura, E.A.G. Pineda, A.F. Rubira, Water transport properties through starch-based hydrogel nanocomposites responding to both pH and a remote magnetic field. Chem. Eng. J. **259**, 620–629 (2015). https://doi.org/10.1016/j.cej.2014.08.045
42. F. Zhu, Starch based Pickering emulsions: Fabrication, properties, and applications. Trends Food Sci. Technol. **85**, 129–137 (2019). https://doi.org/10.1016/J.TIFS.2019.01.012
43. M.D.R. Lenie, S. Ahmadzadeh, F. Van Bockstaele, A. Ubeyitogullari, Development of a pH-responsive system based on starch and alginate-pectin hydrogels using coaxial 3D food printing. Food Hydrocoll. **153**, 109989 (2024). https://doi.org/10.1016/J.FOODHYD.2024.109989
44. F. Azadikhah, A.R. Karimi, Injectable photosensitizing supramolecular hydrogels: A robust physically cross-linked system based on polyvinyl alcohol/chitosan/tannic acid with self-healing and antioxidant properties. React. Funct. Polym. **173**, 105212 (2022). https://doi.org/10.1016/J.REACTFUNCTPOLYM.2022.105212
45. S.A. Alkhursani, H.R. Barai, D.M. Alshangiti, S.A. Al-Gahtany, M.M. Ghobashy, G.A.N. Atia, M. Madani, M.K. Kundu, S.W. Joo, pH-Sensitive Hydrogel Membrane-Based Sodium Alginate/Poly(vinyl alcohol) Cross-Linked by Freeze–Thawing Cycles for Dye Water Purification. ACS ES&T Water. **4**, 509–519 (2024). https://doi.org/10.1021/acsestwater.3c00567
46. X. Chen, N. Ji, F. Li, Y. Qin, Y. Wang, L. Xiong, Q. Sun, Dual Cross-Linked Starch–Borax Double Network Hydrogels with Tough and Self-Healing Properties. Foods. **11**, 1315 (2022). https://doi.org/10.3390/foods11091315
47. C. Chang, B. Duan, J. Cai, L. Zhang, Superabsorbent hydrogels based on cellulose for smart swelling and controllable delivery. Eur. Polym. J. **46**, 92–100 (2010). https://doi.org/10.1016/J.EURPOLYMJ.2009.04.033
48. M.G. Raucci, M.A. Alvarez-Perez, C. Demitri, D. Giugliano, V. De Benedictis, A. Sannino, L. Ambrosio, Effect of citric acid crosslinking cellulose-based hydrogels on osteogenic differentiation. J. Biomed. Mater. Res. A **103**, 2045–2056 (2015). https://doi.org/10.1002/jbm.a.35343
49. L.M. Barrangou, M. Drake, C.R. Daubert, E.A. Foegeding, Textural properties of agarose gels. II. Relationships between rheological properties and sensory texture. Food Hydrocoll. **20**, 196–203 (2006). https://doi.org/10.1016/J.FOODHYD.2005.03.013
50. P. Mahajan, M.B. Bera, P.S. Panesar, Modification of Kutki millet (Panicum sumatrense) starch properties by the addition of amino acids for the preparation of hydrogels and its characterization. Int. J. Biol. Macromol. **191**, 9–18 (2021). https://doi.org/10.1016/J.IJBIOMAC.2021.09.072
51. F. Zhu, Y. Hua, G. Li, Physicochemical properties of potato, sweet potato and quinoa starch blends. Food Hydrocoll. **100**, 105278 (2020). https://doi.org/10.1016/J.FOODHYD.2019.105278
52. L.Y. Teng, N.L. Chin, Y.A. Yusof, Rheological and textural studies of fresh and freeze-thawed native sago starch–sugar gels. II. Comparisons with other starch sources and reheating effects. Food Hydrocoll. **31**, 156–165 (2013). https://doi.org/10.1016/J.FOODHYD.2012.11.002
53. M.V. Chandra, B.A. Shamasundar, Texture Profile Analysis and Functional Properties of Gelatin from the Skin of Three Species of Fresh Water Fish. Int. J. Food Prop. **18**, 572–584 (2015). https://doi.org/10.1080/10942912.2013.845787
54. D. Larrea-Wachtendorff, G. Di Nobile, G. Ferrari, Effects of processing conditions and glycerol concentration on rheological and texture properties of starch-based hydrogels produced by high pressure processing (HPP). Int. J. Biol. Macromol. **159**, 590–597 (2020). https://doi.org/10.1016/J.IJBIOMAC.2020.05.120
55. J.F.-V.J. García-Echeverr, M.V. Vidal, J. Pasqualino, P. Meza-Castellar, Lambis-Miranda, Chemical Modification and Characterization of Starch Derived from Plantain (Musa paradisiaca) Peel Waste, as a Source of Biodegradable Material. Chem. Eng. Trans. **65**, 763–768 (2018)
56. A.O. Odiongenyi, N.B. Essien, R.A. Ukpe, Corn Starch as a Substitute for Commercial Food Starch: FT-IR and Rheological Characterization. J. Sci. Eng. Res. **3**, 494–501 (2016)
57. P.S. Garcia, M.V.E. Grossmann, F. Yamashita, S. Mali, L.H. Dall'Antonia, W.J. Barreto, Citric acid as multifunctional agent in blowing films of starch/PBAT. Quim. Nova. **34**, 1507–1510 (2011). https://doi.org/10.1590/S0100-40422011000900005

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.