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ORIGINAL PAPER

Development and characterization of edible films from onion and garlic purees via continuous casting for scalable active food packaging

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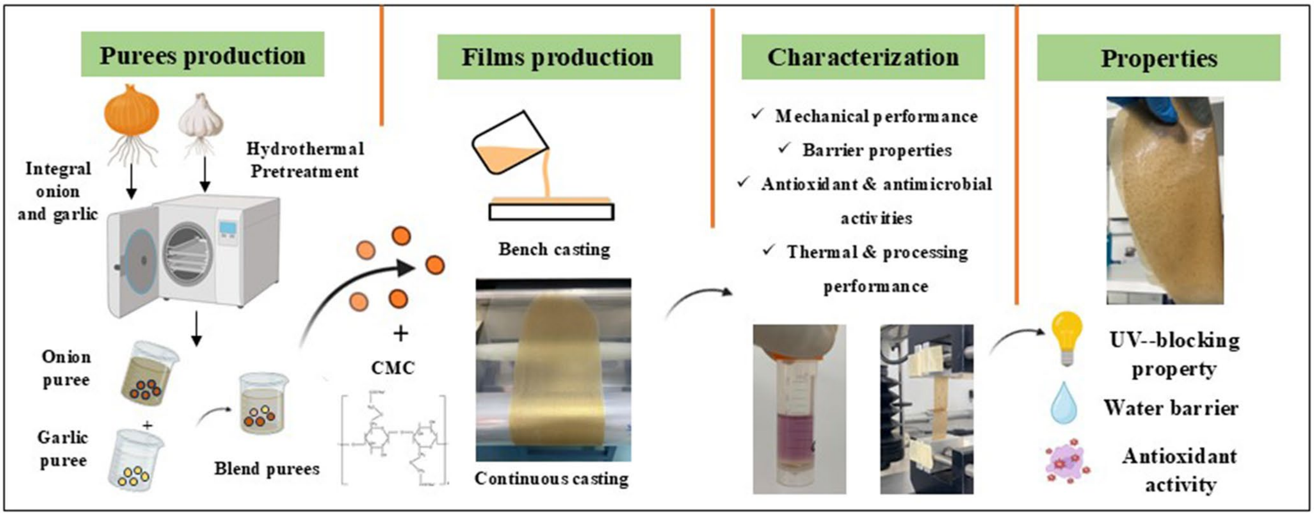
Abstract

Edible films based on *Allium* purees, such as onion (*Allium cepa* L.) and garlic (*Allium sativum* L.), are a promising alternative for food packaging due to their high levels of polysaccharides, bioactive compounds, and intrinsic flavor, which enhance the sensory and functional properties of packaged foods. However, large-scale production is essential for these packaging materials to reach the final consumer. In this study, film-based purees were developed from hydrothermally pretreated onion and garlic, with or without carboxymethylcellulose (CMC), using both continuous casting and laboratory-scale bench casting methods. The properties of films produced by continuous casting were compared to those obtained by bench casting. Films containing CMC exhibited a more cohesive structure and improved mechanical performance (Tensile Strength (TS) increased by approximately 79%), higher contact angles (increased by approximately 112%), while water vapor permeability (WVP) and water solubility decreased by approximately 27% and 8.5%, respectively ($p<0.05$ for all parameters), indicating better barrier properties. CMC also increased thermal stability, while the heat sealability of the films was unaffected by the production method. All films demonstrated strong antioxidant activity, regardless of their production method. These findings highlight the potential of CMC-enriched Allium-based edible films for packaging fatty foods prone to lipid oxidation. Hydrothermal pretreatment of whole Allium purees enhanced antioxidant properties, resulting in films with high antioxidant activity. Furthermore, the successful application of continuous casting demonstrates the feasibility of scaling up production, supporting the development of commercially viable bio-based packaging solutions.

Graphical Abstract



Keywords Hydrothermal pretreatment · Allium vegetables · Antioxidant activity · Active food packaging

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Introduction

The development of edible films from vegetable-based purees has emerged as a promising strategy for sustainable food packaging. These materials are typically composed of macromolecules such as polysaccharides and proteins, which enable the formation of dense matrices with excellent oxygen barrier properties. However, their hydrophilic nature generally results in poor water resistance, limiting broader applications [1, 2]. In addition to structural components such as cellulose, hemicellulose, and pectic substances, as well as lignin, vegetables also contain a wide range of bioactive compounds, including phenolic compounds and natural antimicrobial agents. These substances, usually classified as secondary metabolites, are part of the plants' intrinsic defense mechanisms and can function as active compounds in food packaging [2–4]. Furthermore, films produced from vegetable purees can enhance the sensory and nutritional qualities of packaged foods [1, 5].

Among plant-based matrices, Allium species, such as onions (*Allium cepa* L.) and garlic (*Allium sativum* L.), are particularly attractive because of their high levels of flavonoids (e.g., quercetin) and sulfur-containing compounds, including thiosulfinates. These compounds are responsible for their characteristic pungent flavor and aroma and contribute significantly to antimicrobial and antioxidant activities, thereby providing dual functionality when incorporated into edible films, namely preservation and potential flavor enhancement [6].

From a circular economy perspective, the valorization of Allium-based residues is particularly relevant. Onion (ON) and garlic (GA) processing generates large amounts of solid waste, which, due to their high sulfur content, are unsuitable for soil application and pose environmental concerns [7]. On the other hand, these residues are rich sources of polysaccharides and bioactive compounds, making them excellent candidates for conversion into active food packages [2, 4, 7].

Despite the clear potential, producing cohesive films directly from vegetable matrices poses significant challenges due to the heterogeneity and complex structure of plant cell walls, particularly the poor film-forming capacity of hemicellulose. Thermal hydrolysis has been demonstrated to be an effective pretreatment that partially depolymerizes hemicellulose, improves biomass dispersion, and enhances film cohesion and mechanical integrity [8]. This approach has proven successful for producing cohesive and functional films from onion [9], orange by-products [10], and banana peels [11]. Moreover, hydrothermal processing facilitates the release of phenolic compounds and other cell wall-bound bioactives, further enhancing the antioxidant properties of the films [12–14].

Recent studies have demonstrated that onion-based films produced at the laboratory scale exhibit promising active properties, including antimicrobial activity and the ability to extend shelf life in food systems such as hamburgers [15]. Additionally, the safety of these materials has been confirmed for cytotoxicity and mutagenicity [16]. However, these developments remain confined to small-scale bench casting, severely limiting their industrial viability.

The transition from laboratory to industrial-scale production is crucial for the commercialization of vegetable puree-based films. Continuous casting has emerged as an effective strategy for scaling up biopolymer films, offering significant advantages such as reduced drying time, increased production rates, and improved film uniformity [17, 18]. Previous studies have successfully applied this method to films based on tomato by-products [19], carrot residues [18], and curaua fibers [20, 21], achieving production rates suitable for industrial application. Despite these advances, no studies to date have reported the production of Allium-based edible films using continuous casting, nor has the impact of hydrothermal pretreatment on their processability and functional properties been fully elucidated. In contrast, continuous casting has emerged as an effective strategy for scaling up vegetable-based films, enabling continuous film formation with substantially reduced drying times (minutes to a few hours), higher production rates, and improved thickness uniformity. For example, it has been reported that films based on tomato puree and watermelon, guava, and passion fruit have been previously produced by continuous casting with lamination speeds of 0.11 m/min and 0.12 m/min for the latter three, forming a dry film in a few minutes [5, 19]. This is due to the infrared radiation drying steps, higher temperatures, and air circulation involved in the process [5].

Furthermore, this is the first study to develop edible films based on a synergistic combination of onion (*A. cepa* L.) and garlic (*A. sativum* L.) purees. The combination leverages the complementary functional attributes of both species, onion contributing high levels of flavonoids and film-forming polysaccharides, and garlic providing additional antimicrobial activity due to its rich sulfur-containing compounds. This unique formulation not only enhances the bioactive properties of the films but also introduces new challenges and opportunities related to sensory impact, processability, and functional performance. This study combined onion and garlic purees into a single film-forming system and incorporated carboxymethyl cellulose (CMC) as a structuring agent to enable continuous casting, while improving mechanical performance and water vapor barrier properties. This approach advances the development of Allium-based puree films produced from whole vegetables, including peels and trimmings, thereby eliminating the need for peel separation and drying step, adding value to these by-products, and aligning the process with conditions more compatible with large-scale production.

Therefore, this study aims to (i) investigate the effects of hydrothermal pretreatment on the chemical composition and structural properties of yellow onion and garlic purees, (ii) develop composite edible films based on their combination using the continuous casting technique, and (iii) compare the functional, barrier, and active properties of these films with those produced by conventional bench casting. This is the first report to demonstrate both the feasibility of combining onion and garlic in edible film production and the scalability of these films through continuous casting, offering new insights into their industrial application and contribution to sustainable food packaging solutions.

Materials and methods

Materials

Fresh onions and garlic were obtained from a local market from a single supplier of Araraquara, SP, Brazil. Folin-Ciocalteu phenol reagent (aqueous solution, 2 M) was acquired from Exodo Científica. The following analytical-grade compounds were also utilized: (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonate)) (ABTS) (≥98%), 2,2-diphenyl-1-picrylhydrazyl; di(phenyl)-(2,4,6-trinitrophenyl) iminoazanium) (DPPH), ferric chloride ($FeCl_3$, reagent grade, 97%), 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox) (97%), quercetin (≥95%, HPLC), potassium peroxydisulfate ($K_2S_2O_8$) (ACS grade, ≥99%) and sodium carboxymethylcellulose (CMC, molecular weight approximately 250 kDa, substitution degree: 0.8–0.9) were obtained from Sigma-Aldrich. Gallic acid (GA, 98%) was obtained from Perfyl Tech, glycerol (99.5%, LS Chemicals), and methanol (99.8%, Synth) were of analytical grade purity. Distilled water was employed in the preparation of all aqueous solutions.

Production of *Allium* purees using hydrothermal pretreatment

Two different purees were prepared from yellow onion (*Allium cepa* L.) and garlic (*Allium sativum* L.), according to the methodology described by Dias et al. [9] with some modifications. The experimental design, including the formulation ratios and processing routes used for film production, is summarized in Fig. 1. Onion and garlic peels were included in the purees and in the liquid fraction of the purees

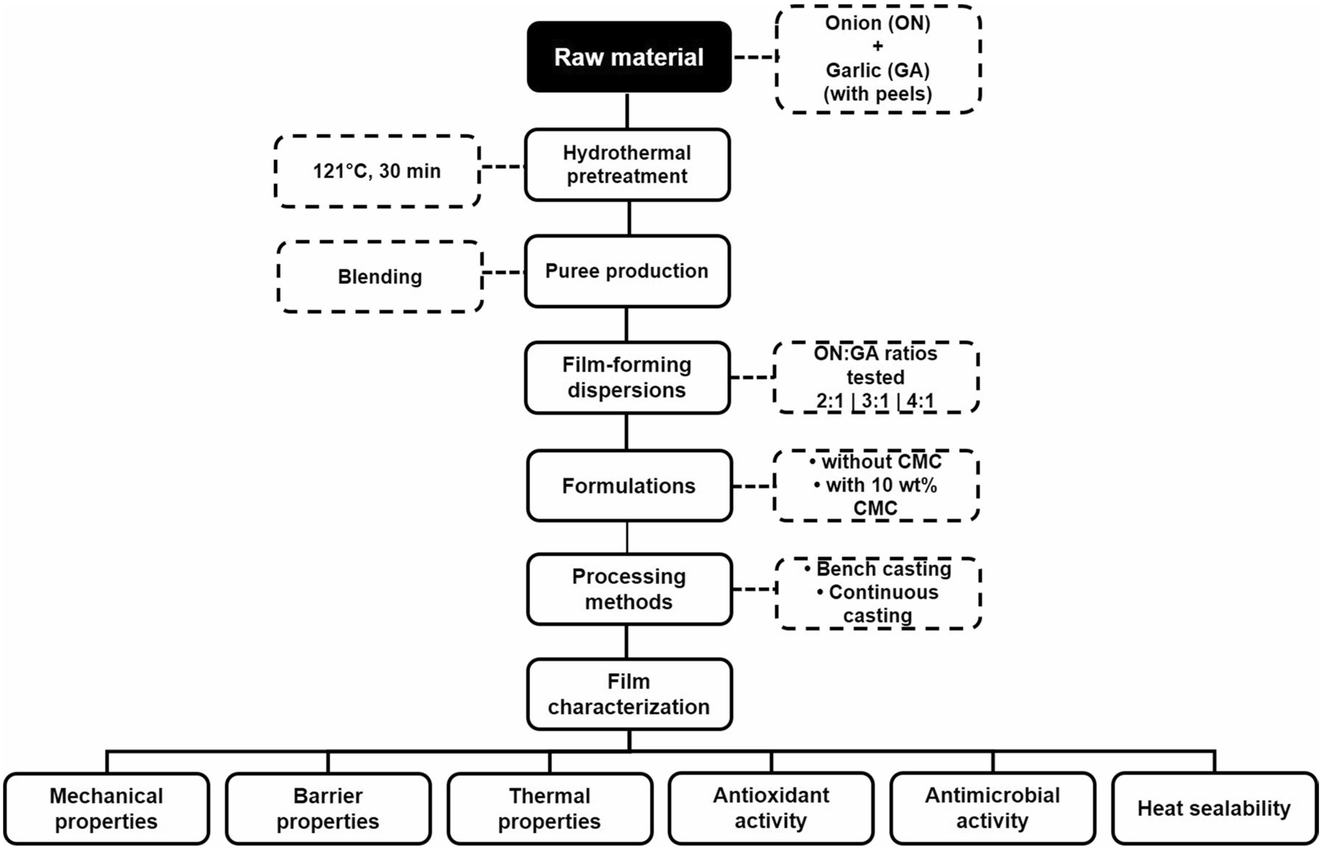

Fig. 1 Experimental design and processing routes for the production of Allium puree-based films

after pretreatment. Initially, the bulbs, with their skins, were washed, then cut in half longitudinally and transversely, resulting in four pieces per bulb. These pieces were washed again. The bulbs were mixed with distilled water at a 1:1 mass ratio and subjected to hydrothermal pretreatment. The inclusion of onion and garlic peels was rather on a dual rationale: (i) their high concentration of bioactive compounds, particularly flavonoids such as quercetin in onion outer layers [7], and (ii) their relevance as significant agro-industrial by-products generated during processing [7]. Thus, their use aimed to integrate residue valorization with the development of antioxidant-active edible films. Hydrothermal pretreatment was conducted in an AV 50 autoclave (Phoenix Luferco, Brazil) at 121 °C under 1.2 kgf/cm$^{2\ \text{pressure}}$ for 30 min, as previously described by Dias et al. [9]. After thermal pretreatment, each puree was processed in an industrial blender (model SPL-062, Spol, Brazil) with a capacity of 1.5 L, operating at 18,000 rpm for 3 min.

Characterization of *Allium* purees

The phenolic compound content (TPC) of the purees was determined before and after hydrothermal pretreatment, using the Folin-Ciocalteu method [22]. The results are presented in supplementary Table S1 The pretreated purees were analyzed for total flavonoid content (TFC) by the spectrophotometric method [23], and antioxidant activity (AA) was assessed through scavenging of the DPPH (2,2-diphenyl-1-picrylhydrazyl) radical [24], the ABTS·+ (2,2'-azino-bis-(3-ethylbenzothiazoline-6-sulfonate) radical [25], and by the ferric reducing power (FRAP) assay. The results are presented in supplementary Table S2. Furthermore, lignin and structural carbohydrates were determined according to the NREL/TP-510–42618 method (Sluiter et al., 2008). For this analysis, after hydrothermal pretreatment, the liquid fraction was filtered, and the retained solid fraction part was freeze-dried and sieved using a 20-mesh sieve.

Preparation of films: formulation, processing, and experimental design

Films were produced by bench casting using purees obtained from previously hydrothermally treated bulbs. Forming dispersions (formulations are provided in Table S3, Supplementary Information) from the purees in a 4:1 ratio (ON: GA) were homogenized using an Ultra Turrax (model T-18, Ika, Germany) at 10,000 rpm for 3 min, followed by vacuum desgassing for 20 min at – 800 mmHg. This proportion was based on preliminary tests (data not shown). Preliminary mechanical tests were performed using onion: garlic ratios of ON2:GA1, ON3:GA1, and ON4:GA1. No significant differences were observed between the formulations. Therefore, the 4:1 ratio was selected, as it provided a higher proportion of onion puree, which exhibited greater antioxidant content. It should be noted that other ratios were not further explored, representing a limitation of the present study. When included, a 4.0% CMC stock solution was incorporated into the film-forming solution to achieve a final CMC concentration of 10 wt% (based on total solids), thereby improving the films' physical properties due to this biopolymer's binding capabilities. These proportions were based on preliminary tests. No CMC was added in the first treatment, named *Allium* puree bench (AP-B). In the other two treatments, CMC was added, and they were named *Allium* puree with CMC by bench scale (AP/CMC-B) and *Allium* puree with CMC by continuous casting (AP/CMC-CC). For each film, two separate batches were produced independently. A summary of the formulations and processing routes used for each sample, including the presence or absence of CMC and the casting method employed, is presented in Table 1.

Table 1 Summary of the processing steps to which CMC was added and sample were subjected to bench casting and continuous casting processing

Sample	CMC	Bench casting (B)	Continuous casting (CC)
AP-B	No	Yes	No
AP/CMC-B	Yes	Yes	No
AP/CMC-CC	Yes	No	Yes

CMC carboxymethylcellulose

Bench casting

The dispersions were poured onto glass plates (35 × 30 cm^2) coated with polyester film for bench casting, using a casting leveling bar to an initial thickness of 2 mm. The films were dried at 40 °C in an air-circulated oven (model SL-102, SOLAB, Brazil) with air circulation for 20 h.

Continuous casting

The CMC film was manufactured by continuous casting using a KTF-S coating machine (Werner Mathis AG, Switzerland). The dispersion was deposited onto a moving Mylar® substrate (DuPont, Brazil) and passed through a precision Teflon-coated knife to a thickness of 1.80 mm. The film was moved at 0.08 m min^{-1}, through an initial infrared drying (30 cm at ca. 40 °C) and two more drying ovens set at 110 °C and 120 °C, respectively. Three film-forming dispersions were initially prepared: the *Allium* mix puree without CMC, and with 10 and 20 wt% CMC (based on total solids). The solution without CMC did not form a cohesive film. Therefore, the film-forming components in the purees were insufficient to produce self-supporting films in continuous casting. The films with CMC in both concentrations were cohesive and detachable. However, the 10 wt% formulation was selected for further characterization, as it represented the lowest effective concentration capable of forming cohesive films. Therefore, films containing 20 wt% CMC were not further characterized.

Film characterization

Scanning electron microscopy (SEM)

The morphology of the films, including flat surfaces and the cross section obtained by freeze-fracture, was analyzed using scanning electron microscopy (SEM). The samples were coated with gold in a Sputter Coater SCD 050 (LEICA) at a current of 40 mA for 60 s and mounted on metal stubs with the air-exposed during drying surface facing upwards. Fractures were created by breaking a strip of film in liquid nitrogen.

Moisture content

Moisture content was determined using the physical method of infrared radiation on a scale, with 1 g of sample heated at 105 °C in a balance (model ID-50, Marte Científica, Brazil). The water content was measured by assessing radiation absorption at infrared wavelengths of 3.0 µm and 6.1 µm (Cecchi, 2003). The analyses were performed in triplicate.

Water vapor permeability (WVP)

Water vapor permeability was performed according to the ASTM E96/E96M-22 (2022) by gravimetric method. Each film was placed in acrylic permeation cells with a diameter of 24 mm, containing 1.5 mL of distilled water. The analysis was conducted in quadruplicate. The cells remained in a climatic chamber at 50% RH and 25 °C, with eight measurements throughout 24 h.

Water solubility

Water solubility was assessed by measuring the amount of dry material dissolved after 24 h of immersion in water. The dry mass of each 2 cm² film sample, in quadruplicate, was determined by weighing them prior to and following a 24 h drying at 105 ± 1 °C (initial weight, *wi*). Subsequently, the samples were then immersed in 50 mL of distilled water and kept under agitation at 76 rpm for 24 h at 25 ± 1 °C. After that, they were subjected to drying at 105 ± 1 °C for 24 h to obtain the final dry weight (final weight, *wf*). The water solubility percentage was calculated using Eq. 1 [26].

Water contact angle (WCA)

Following the sessile drop method, the static contact angle with water measurements were performed using an Attention Theta Lite optical tensiometer from Biolin Scientific (Gothenburg, Sweden). Approximately 5 µL of ultrapure water was deposited on the film surface (the one dried in contact with air), and the contact angle values were recorded 60 s after deposition.

Mechanical tests

The thickness of the films was determined at ten different points using a KR1250 thickness gauge (AKROM), with a margin of error of ± 1 µm. All film samples were conditioned for at least 40 h in a controlled environmental cham 50% relative humidity (RH) and 25 °C before mechanical testing. The mechanical properties of the films, including tensile strength (TS), elongation at break (EB), and elastic modulus (EM), were measured using a texturometer TA. XT Plus Texture Analyzer (Stable Micro Systems, Godalming, UK). The equipment was equipped with a 100 N load cell, an initial jaw separation of 125 mm, and a deformation speed of 12.5 mm/min. For the analysis, 12 film samples (125 mm x 17.5 mm) were tested according to ASTM D882-18 (ASTM, 2018).

Thermal properties: differential scanning calorimetry (DSC) and thermogravimetry (TG)

DSC analysis was conducted using a Differential Scanning Calorimeter (model Q100, TA Instruments, EUA) under a nitrogen atmosphere with a gas flow rate of 50 $\text{mL}\cdot\text{min}^{-1}$. An aluminum crucible (40 µL) was employed to contain the sample and as a reference. Approximately 7 mg of each film was analyzed. DSC curves were recorded at a heating rate of 10 $°\text{C}\cdot\text{min}^{-1}$. The initial heating cycle was carried out up to 140 °C, where the temperature was held constant for 3 min to remove water from the film. Subsequently, the material was cooled to −50 °C and then reheated to 130 °C. The analysis focused on the second heating cycle, covering the range from −40 °C to 130 °C. Thermogravimetric analysis was performed using a TA Instruments Q500 equipment for puree flour (before and after pretreatment) and films. Approximately 7 mg of each sample was analyzed under the following conditions: heating ramp from 30 to 500 °C, nitrogen gas flow rate of 50 $\text{mL}\cdot\text{min}^{-1}$ and heating rate of 10 $°\text{C}\cdot\text{min}^{-1}$.

Heat sealing and seal strength determination

Heat sealing was accomplished using a heat-sealing machine (Barbi M300-T) equipped with a seal bar of 4,5 mm width for 4 s at power 10. Ten rectangular samples from both films with CMC were cut into 65 mm x 15 mm strips using a TA. XT Plus Texture Analyzer (Stable Micro Systems, Godalming, UK). The analysis was carried out using the same method as tensile strength, but with an initial jaw separation of 50 mm. Briefly, each end of the sample was fixed to the equipment and held perpendicular to the tensile direction, without any support (Technique A). Seal strength was assessed following the ASTM standard method F88-21, using Technique A (ASTM, 2021). The samples were conditioned at 25 ± 2 °C and 50% RH for 40 h both before and after sealing.

Fourier-transform infrared (FTIR) spectroscopy

The film spectra recorded using a Vertex 70 spectrometer (BRUKER) with attenuated total reflectance (ATR) in the range of 4000 to 500 cm^{-1} and a spectral resolution of 4 cm^{-1} wavenumber cm^{-1}. A quantitative FTIR analysis was performed by calculating the ratio between the intensity of the OH band and the reference C-H band.

Color measurement

The color parameters (L*, a*, b*) were measured with a Miniscan XE colorimeter (HunterLab, USA). The analyses were performed under the white standard plate, using color parameters CIELab. Samples with 4 × 4 cm were done in triplicate.

UV–Visible (UV–Vis) spectroscopy

In transmission mode, UV absorption capacity was measured in triplicate on rectangular film samples (2.5 × 4.5 cm) using a UV-2600 spectrophotometer (Shimadzu, Kyoto, Japan). The films were placed perpendicular to the ultraviolet light inside the spectrophotometer cell. Measurements were conducted over a scanning range of 200 to 800 nm, covering the UVA, UVB, and UVC regions, and UV shielding was calculated using Eqs. 2–4 [3], where T_λ represents the spectral transmittance at wavelength λ and Δλ corresponds to the wavelength interval. The calculated values represent the average transmittance within each UV region, and S_{UV} expresses the UV-blocking ability of the films.

Total phenolic content (TPC), total flavonoid content (TFC), and antioxidant activity (AA)

TPC was determined by the Folin-Ciocalteu method (Swain & Hills, 1959). The results were reported as mg of gallic acid equivalents per gram of sample (GAE mg g^{-1}). TFC was performed as described by Funari e Ferro [23], and results were determined for total flavonoids equivalent in quercetin. AA was analyzed by the spectrophotometric method of DPPH (2,2-diphenyl-1-picrylhydrazyl) radical scavenging, as described by [24], the 2,2'-azinobis-(3-ethylbenzothiazoline-6-sulfonate) radical (ABTS·+) method according to [25], and by the ferric reducing power (FRAP) assay. All the analyses were conducted in triplicate.

Antioxidant activity over time

The AA of the films was analyzed by the spectrophotometric discoloration method of DPPH (2,2-diphenyl-1-picrylhydrazyl) radical scavenging, adapted from Luís et al. [27]. Film discs (6 mm) were immersed in test tubes containing 3.9 mL of DPPH solution. The tubes were shaken and left to rest in the dark until reading. Only the DPPH solution was used as a control, without the film. The antioxidant activity of the films was evaluated based on the percentage of inhibition of the DPPH radical every 30 min, for 2 h.

Identification and quantification of phenolic compounds

For the analysis, methanolic extracts (80:20 v/v) were prepared from the purees after hydrothermal treatment (ON e GA) and from the film obtained by continuous casting (AP/CMC-CC). The solvent from the extract was removed using a vacuum sample concentrator (model RVC 2–33, Chist, Germany), and the dried extract was used for analysis.

First, the extract was solubilized in methanol/water (80:20, v/v) to obtain a solution with a suitable concentration for analysis. The solution was then filtered using an RC 13 mm/0.22 µm sample filter. High-performance liquid chromatography (HPLC) analysis was performed on a Shimadzu SCL-10Avp chromatograph, with LC-6AD pumps equipped with an Agilent Hypersil ODS 250 × 4.6 mm x 5 µm column and coupling to an SPD-M10Avp detector. The chromatographic conditions were as follows: 20µL of sample was injected, and the mobile phase consisted of water (Phase A) and acetonitrile (Phase B), each containing 0.1% formic acid. The separation was carried out at a 1 mL/min flow rate, employing a concentration gradient. The gradient program was as follows: start B at 5%, 5% (5 min); 20% B (23 min); 65% B (30 min); 90% B (40 min); 5% B (45 min); Stop at 55 min. Quantification was based on a quercetin standard curve (10–160 µg/mL), with a retention time of 32.2 min and detection at 370 nm.

Antimicrobial activity

The disk diffusion method (halo test) was employed. Standard strains of *Staphylococcus aureus* (ATCC 6538) and *Escherichia coli* (ATCC 11229) were used, previously reactivated in sterile liquid culture medium and grown for 16 h, as described by CLSI [28]. Petri dishes with culture medium were prepared, and 100 µL of the previously diluted inoculum (1 to 5×10^6 CFU/mL) were evenly spread across the medium surface. Samples of the films previously cut into 18 mm discs were placed on the medium surface. Filter discs soaked with a 10 mg/mL streptomycin solution were used as positive control. The plates were incubated at 35–37 °C for 24 h. If halo formation occurred, the diameters were determined using a caliper on the underside of the inverted Petri dish.

Statistical analysis

The characterization data of the developed films were analyzed by analysis of variance with post-hoc Tukey's test ($p < 0.05$) in case of significant differences. The data of the purees, before and after hydrothermal pretreatment, were analyzed using a t-test. The data were processed using Minitab® statistical software version 17 (Minitab Inc., State College, PA, USA).

Results and discussion

Characterization of *Allium* purees

Table 2 presents the biomass composition before and after (solid fraction) hydrothermal pretreatment. Before pretreatment, yellow onion puree exhibited higher cellulose and extractives than garlic, whereas garlic had a higher lignin percentage than onion. Hydrothermal pretreatment resulted in a significant decrease in cellulose, hemicellulose, acid-soluble lignin, and ash percentage in yellow onion puree, while increasing extractives, indicating its effectiveness in modifying the biomass composition. After hydrothermal pretreatment, no significant difference in cellulose levels was observed between ON and GA. In contrast, garlic's hemicellulose content was significantly higher than that of onion puree. Hemicellulose is the first component to be hydrolyzed in biomass during hydrothermal pretreatment due to its shorter glycosidic chain links. The sugars present in hemicellulose are solubilized, and their composition may vary depending on the biomass. As mentioned earlier, hydrothermal pretreatment was used to separate the structural carbohydrates of the plant cell wall (namely cellulose, hemicellulose, and gel-forming polysaccharides). During this process, hemicellulosic sugars, including pentoses (xylose, arabinose) and hexoses, are solubilized. These low-molecular-weight sugars remain in the film-forming matrix and act as internal plasticizers by increasing Polymer chain Mobility through hydrogen bonding interactions, thereby contributing to the flexibility observed in the resulting films [9]. It is essential to analyze biomass because the properties of edible films depend mainly on the composition of the raw materials used and the processing methods applied [8]. In the case of edible films, the composition of the puree plays a crucial role, contributing to the sensory characteristics of the packaged product [1, 5].

Film characterization

As previously mentioned, the *Allium* puree without CMC did not form a cohesive film by continuous casting, indicating that the film-forming components present in the onion and garlic purees were insufficient to produce independent films via this method. Therefore, CMC was incorporated as a binding agent to enable film formation. This strategy allows in films with improved mechanical resistance, extensibility, and reduced water permeability, making them more suitable for food packaging applications. Similar findings were reported by Otoni et al. [18], who obtained composite films

Table 2 Chemical composition of *Allium* purees before and after hydrothermal treatment

	Raw Puree		Pre hydrothermal Puree	
Composition (g/100 g)	ON	GA	ON	GA
Cellulose/Glucan	26.23 ± 0.74^{Aa}	20.18 ± 0.85^{Ba}	14.32 ± 0.60^{b}	15.31 ± 1.27^{b}
Hemicellulose	14.10 ± 1.87^{a}	13.40 ± 0.21^{a}	3.54 ± 0.32^{Bb}	5.89 ± 0.48^{Ab}
Acid-soluble lignin	1.49 ± 0.03^{Ba}	3.20 ± 0.18^{Aa}	0.61 ± 0.01^{Bb}	1.88 ± 0.05^{Ab}
Acid-insoluble lignin	10.50 ± 1.13^{B}	25.64 ± 8.93^{A}	8.71 ± 1.05^{B}	21.40 ± 2.95^{A}
Extractives	22.97 ± 1.39^{Ab}	13.50 ± 0.44^{Bb}	69.13 ± 2.57^{Aa}	41.83 ± 0.78^{Ba}
Ashes	5.67 ± 0.32^{a}	5.80 ± 1.0	3.97 ± 0.38^{b}	4.50 ± 0.43

[*] Values within the same treatment (raw or treated) followed by different uppercase letters (A, B) indicate significant differences between ON and GA samples (t-test, $p < 0.05$)

[**] Values in the same row with different letters (a, b) indicate significant differences between raw and treated puree (t-test, $p < 0.05$)

from carrot processing waste only continuous casting after the addition of a binding agent. Accordingly, only CMC-containing films were produced using both methods (bench and continuous casting) for process comparison. Additionally, a film obtained by bench casting without CMC (AP-B) was included to evaluate the effect of the polymer addition on film properties.

Morphological characteristics

The SEM microstructure of the films is shown in Fig. 2. The structural characterization of the film showed intact and continuous films, without visible bubbles or pores. The film surfaces were rough, which is expected due to the heterogeneity of the starting material. Similar findings were reported by Riaz et al., [31] who observed that the incorporation of Chinese chive (*Allium tuberosum*) root extract into CMC films led to a more heterogeneous surface, characterized by increased roughness and more intense points in the structure. This effect was more pronounced at the highest concentration tested (5%). The micrographs also show that the film without adding CMC presented greater roughness on its surface compared to the other two films. This result suggests that the presence of CMC in the matrix probably

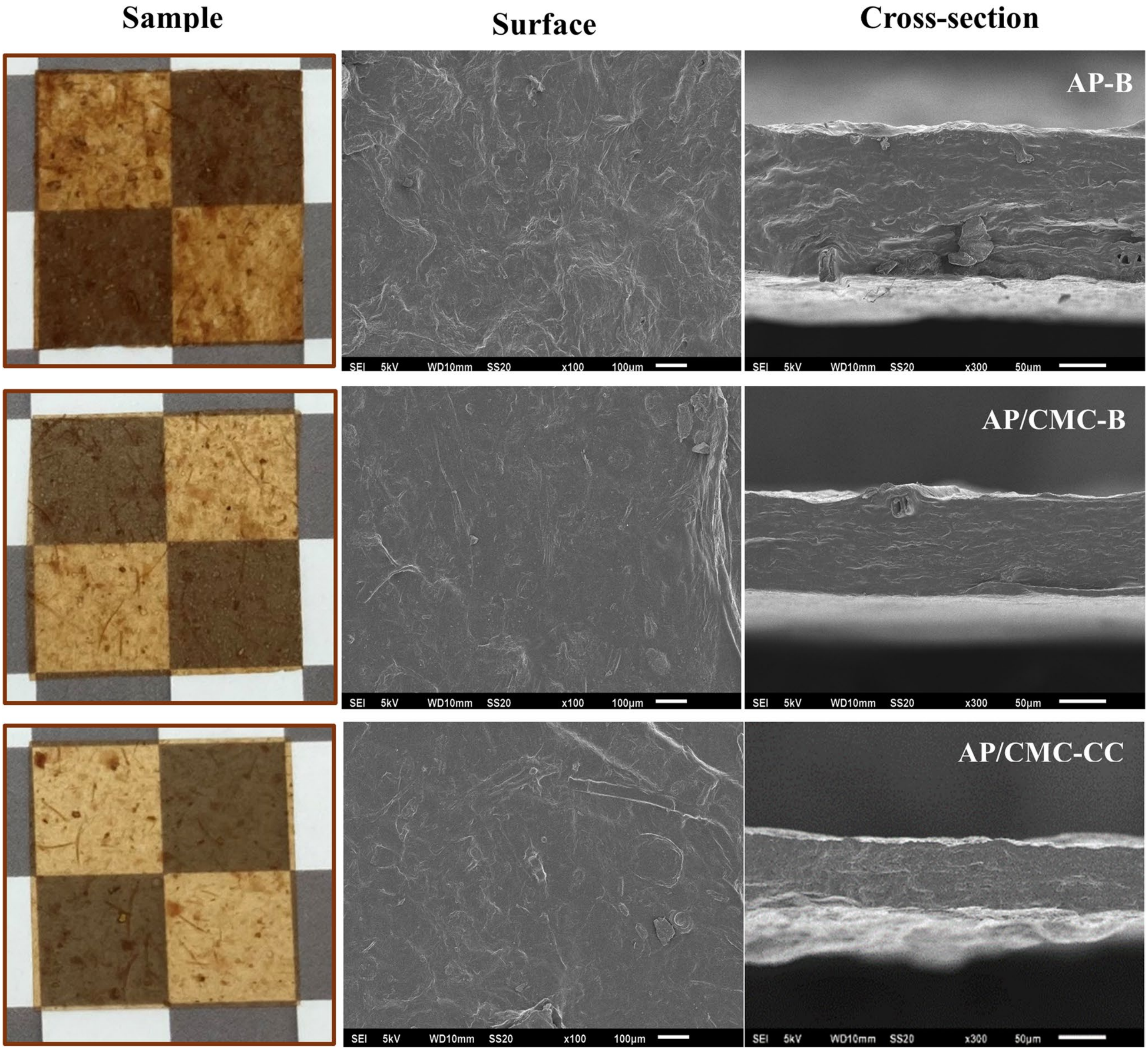

Fig. 2 Photographs and scanning electron microscopy (SEM) micrographs (surface and cross-section) of Allium films obtained by bench casting without CMC (AP-B) and with CMC produced by bench (AP/CMC-B) or continuous casting (AP/CMC-CC). Surface micrographs were obtained at x100 magnification (scale bar = 100 µm), while cross-section micrographs were obtained at x300 magnification (scale bar = 50 µm)

resulted in the development of a more cohesive structure. The same effect of CMC was observed in films produced from banana peel [11].

Fourier-transform infrared (FTIR) spectroscopy

The FTIR spectra of the films are presented in Fig. 3 (A). All spectra showed identical absorption bands, evidencing the occurrence of the same components in the three films. The spectra reveal a broadening of the band at 3290 cm$^{-1,}$ which indicates the presence of single hydrogen bonds in the OH groups of cellulose, xylans, pectic substances, and other polysaccharides and simple sugars. This broadening reflects the intra and intermolecular interactions among the various components (dos Santos Dias et al., 2020). The films also showed characteristic carboxyl single bonds with OH at 2924 and 2850 cm^{-1}, originating from the galacturonic acid of pectin present in the plant cell wall, as well as the presence of C=O double bonds at 1747 and 1690 cm^{-1}, which correspond to the carboxylic acids of the ester groups of the galacturonic acid of pectin and/or due to the ketone group in hemicellulose that may be overlapped [29]. The band at 1618 cm^{-1} is attributed to the C-C stretching in the phenolic ring of the aromatic structure, in the polyphenols in *Allium* species. The band at 1408 cm^{-1} corresponds to the asymmetric stretching of the methyl group (CH3) at 1436 cm^{-1} [32], while the band in the 1016 cm^{-1} region is typically characteristic of cellulose [29]. The addition of CMC is evident by the decrease in the intensity of the band in the regions 3290 cm^{-1} and 1408 cm^{-1}, which may suggest a lower proportion of these components due to the addition of another polymer, suggesting intermolecular interaction between the biopolymers, which would decrease the availability of hydroxyl groups through the formation of hydrogen bonds. A quantitative FTIR analysis was performed by calculating the ratio of the OH band intensity to the reference C-H band intensity. The obtained values were 1.09, 1.02, and 1.03 for films AP-B, AP/CMC-B, and AP/CMC-CC films, respectively. The decrease in the ratio (from 1.09 to 1.02 and 1.03) indicates a relative reduction in the availability of hydroxyl groups after the addition of CMC.

Tensile properties

Adding CMC (10 wt% %) increased the material's mechanical strength, regardless of the processing type (Table 3). Films produced with CMC showed higher tensile strength (TS) (8.49–10.78 MPa) than films without CMC (4.75 MPa). This result suggests that CMC acts as a binding agent, bringing polymer chains closer together and leading to a reinforcement effect confirmed by SEM (Fig. 2).

The addition of one or more polymers has been used as a strategy to improve the properties of polymeric materials [33–35]. CMC in sodium alginate films containing onion shallot waste extracts exhibited a mechanical reinforcing effect [36]. Films with banana peels produced with CMC showed higher TS and EB than films without CMC [11]. Thus, adding this component to the film-forming solution and allowing film formation through continuous casting is in line with the need to produce both stronger and more flexible films simultaneously.

The TS in *Allium* films reinforced with CMC (8.49–10.78 MPa) here is higher than edible films based in carrot puree [37] and in banana puree films [38], which exhibited

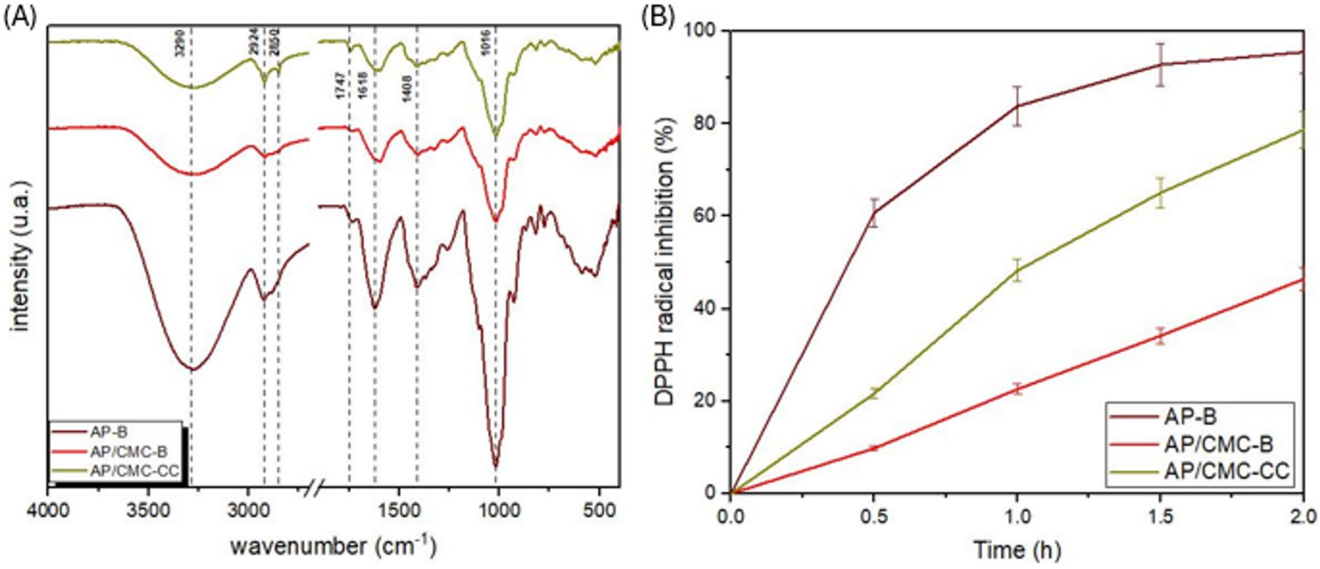

Fig. 3 (**A**) ATR-FTIR spectra and (**B**) DPPH radical inhibition (%) over time of *Allium* films obtained from bench casting without CMC (AP-B), or with the addition of CMC produced by bench (AP/CMC-B) or continuous casting (AP/CMC-CC)

Table 3 Properties of *Allium* films (AP-B), or with the addition of CMC produced by bench (AP/CMC-B) or continuous casting (AP/CMC-CC)

Property	Film				
	AP-B	AP/CMC-B	Δ (%) vs. AP-B	AP/CMC-CC	Δ (%) vs. AP-B
Thickness (µm)	125.07±26.97^{a}	106.99±7.75^{*a}	−14	71.46±3.92^{*b}	−43
TS (MPa)	4.75±1.67^{b}	8.49±1,14^{a}	+79	10.78±3.98^{a}	+127
EM (MPa)	234.1±42.8^{c}	469.1±35.2^{*a}	+100	379.51±20.73^{b}	+62
EB (%)	3.59±1.37^{a}	4.24±0,61^{a}	+18	4.34±1.99^{a}	+21
WVP (g-mm-m ^{2}-h-kPa)	2.832±0.564^{a}	2.073±0.316^{b}	−27	1.797±0.190^{b}	−37
Water solubility (%)	78.33±0.84^{a}	71.64±0.98^{b}	−9	71.46±1.41^{b}	−9
WCA [°]	19.53±3.09^{c}	46.27±0.65^{a}	+137	41.36±1.59^{b}	+112
Moisture (%)	6.68±0.44^{a}	6.62±0.53^{a}	−1	7.33±0.38^{a}	+10
L*	56.63±3.91^{c}	65.53±2.46^{b}	+16	72.29±1.57^{a}	+28
a*	12.17±1.76^{a}	7.57±0.95^{b}	−38	5.57±1.50^{b}	−54
b	36.96±1.18^{a}	38.49±1.80^{a}	+4	36.89±1.52^{a}	0
Croma	38.96±1.15^{a}	39.24±1.86^{a}	+1	37.63±1.91^{a}	−3
Hue (*)	71.68±2.54^{c}	78.86±1.22^{b}	+10	81.69±1.44^{a}	+14
SUVA (%)	99.92±0.05^{a}	99.92±0.02^{a}	0	99.92±0.01^{a}	0
SUVB (%)	99.94±0.03^{a}	99.93±0.01^{a}	0	99.94±0.00^{a}	0
SUVC (%)	99.84±0.06^{a}	99.81±0.02^{a}	0	99.83±0.01^{a}	0
TPC mg GAE/100 g dw	8590±180^{a}	7367±128.0^{b}	−14	8643±647^{a}	+1
TFC mg Q/100 g dw	110.00±10.44^{a}	72.7±17.4^{b}	−34	95.67±2.08^{ab}	−13
DPPH (%)	94.91±0.19^{a}	95.19±0.29^{a}	0	95.3±0.1^{a}	0
ABTS (%)	94.03±1.47^{b}	97.683±0.86^{a}	+4	95.588±1.27^{ab}	+2
FRAP µM FeSO4/g dw	18,798±1183^{a}	16,306±1756^{a}	−13	11,360±644^{b}	−40
Total sugar g Gly/100 g dw	9.658±1.723^{a}	8.4299±0.1652^{a}	−13	10.613±1.042^{a}	+10
mg Quercetin/100 g dw	-	-	-	6.03±1.68	-

Values followed by equal letters within the same row are not significantly different (teste de Tukey, $p<0.05$). TS: tensile strength; EM: elastic modulus; EB: elongation at break; WVP: water vapor permeability; WCA: water contact angle; SUVA: UV-shielding (400–320 nm); SUVB: UV-shielding 320–280 nm; SUVC: UV-shielding (280–200); Tvis: visible region (400–780 nm). TPC: Total Phenolic Content; TFC: Total Flavonoid Content; DPPH: 2,2-diphenyl-1-picrylhydrazyl; di(phenyl)-(2,4,6-trinitrophenyl); ABTS: (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonate)); FRAP: Ferric Reducing Antioxidant Power

*Δ (%) was calculated relative to the control film (AP-B), (% = ((Tratated − Control)/Control) × 100)

5.06 MPa and 6.9 MPa respectively, and are in line with tomato binding HM pectin film which exhibited 8.9 and 14.8 MPa for bench and continuous casting respectively [19]. Furthermore, they are like TS of conventional plastics widely used in food, such as LDPE, which has a TS of 8 to 10 MPa [5, 39].

The improvement in mechanical properties can also be verified by the increase in elastic modulus (EM) in CMC films. The EM of the films produced with CMC, on the bench and by continuous casting, was higher than that observed in puree-based edible sheet with 0.5 g de CMC [40] and significantly different from each other (Table 3), with the one produced by continuous casting presenting a lower EM. However, adding CMC to corn and cassava starch did not improve EM [41]. This result suggests that the addition of CMC makes the material more resistant to elastic deformation under tension, which can be attributed to the interaction of the hydroxyl groups of *Allium* purees with the carboxyl groups of CMC (as discussed in the FTIR analysis, Fig. 3 (A)), resulting in a resistant polymer network as reported previously [37, 40].

The elongation at break (EB) of *Allium* films ranged from 3.59% for the film without CMC to 4.24% and 4.34% for AP/CMC-B and AP/CMC-CC, respectively, with no statistically significant differences observed among the samples. The films produced by continuous casting exhibited an average thickness of 71 µm. In contrast, those obtained via bench casting showed greater thickness, ranging from 107 to 125 µm (Table 3). No significant difference in thickness was observed between the bench-cast films with (107 µm) or without (125 µm) CMC, indicating that the addition of CMC did not affect this parameter. The observed differences in mechanical properties between the films produced by different methods, but with the same composition, can be primarily attributed to variations in thickness. As expected, film thickness is directly influenced by factors such as the total solids content in the film-forming solution, the volume of solution applied to the substrate, and the efficiency of leveling during the drying process [34].

Furthermore, continuous casting enables the production of films with more uniform thickness. This difference between films produced with the same matrix can be explained by the drying methods used in the two processes.

While the bench film remains in the oven for 20 h, the continuous casting film exits dry at the end of a process that typically lasts around 30 min, depending on the flow speed. Thickness directly affects the uniformity, mechanical strength, and barrier properties of edible films, and variations in these properties can be linked to differences in drying dynamics and the shrinkage of the polymeric network during drying [2]. These results suggest that it is possible to produce thinner films faster and more efficiently without affecting the material's mechanical strength or extensibility, as indicated by the elongation at break.

It is important to highlight that the films were developed without the addition of external plasticizers. Therefore, any plasticizing effect observed in the materials can be attributed to soluble sugars naturally present in the Allium matrix, as discussed later. These compounds may act as internal plasticizers, increasing polymer chains and reduce interactions [30]. In the present study, the incorporation of CMC resulted in films with higher tensile strength (Table 3), suggesting a reduction in flexibility compared with formulations relying solely on the plasticizing effect of soluble sugars. Unlike conventional plasticizers, which may migrate from the film matrix over time and lead to embrittlement, internal plasticizers naturally present in plant matrices may contribute to improved structural stability [30].

Biopolymeric films derived from agrowastes, agro-food byproducts, and fruit or vegetable purees plasticized with conventional agents have been widely reported in the literature. For example, potato peel films produced with polyglycerol-3 showed a TS of 12.7 ± 1.5 MPa, EB of 18.4 ± 4.3%, and an EM of 432.1 ± 8 MPa [4]. Similar elastic modulus values (469.1 ± 35.2 and 379.51 ± 20.73 MPa) were obtained in the present study for films containing CMC produced by bench and continuous casting, respectively, suggesting that sugars released during hydrothermal pretreatment contributed to a plasticizing effect. This phenomenon is associated with increased polymer chain mobility and alignment, which may reduce the force required for rupture [4]. A similar behavior was observed in onion-based films reported by dos Santos Dias et al. [9], in which films produced from unwashed purees containing soluble sugars showed the lowest TS and EM values (4.75 ± 1.67 and 47 ± 9 MPa, respectively) compared with films prepared from washed purees without soluble sugars. These comparisons indicate that Allium-based films can achieve satisfactory mechanical performance without the addition of external plasticizers, depending on the processing conditions employed.

Water-related properties

The barrier properties of a material are essential to protect food from environmental factors and are therefore closely related to the stability of the packaged food. For example, the lowest possible WVP value helps keep food fresh for a longer period [42]. The WVP data are presented in Table 3. The addition of CMC to the films significantly decreased the WVP, which can be ascribed to the hydrogen bonding interactions between the hydroxyl groups of CMC and the vegetable puree, allowing greater intermolecular interactions, making the chain more compact, which reduces the molecular spaces and results in a more cohesive film, as observed in the SEM image (Fig. 2). Hydroxyls are characteristic of polysaccharides and are present in vegetable films, as discussed in FTIR (Fig. 3 (A)). These results are similar to those of tomato puree films containing higher-methylation pectin [19].

Although the WVP values are higher than those of conventional synthetic packaging materials (LDPE: 6,673–8,704; HDPE: 1,741–3,482 WVP x 10^{14} (g m m^{-2} s −1 Pa^{-1}) [5, 39], these films are not intended to replace high-barrier packaging. Their potential application lies primarily in edible packaging for ready-to-cook products, such as hamburger patties, where the film could be incorporated during thermal processing, acting as both a protective layer and a flavoring agent [1].

The water solubility of the films is characterized by the amount of dry matter solubilized after 24 h of immersion and reflects the material's resistance to dissolution [26]. This property is particularly important in edible films, which remain in contact with food until it is consumed. Therefore, low water solubility indicates greater material resistance, which is desirable [43]. The water solubility of the films is presented in Table 3. The water solubility of the CMC films was significantly lower than that of AP-B. The decrease in solubility with the addition of CMC can be explained by intermolecular interactions between the biopolymers, which, through hydrogen bonds, reduce the availability of hydroxyl groups, thereby decreasing the breaking of the hydrogen chain by water molecules and, consequently, making the film less soluble [36, 43]. Similar behavior has been reported in CMC-based films containing plant extracts, where solubility decreased due to hydrogen bonding between the polymer matrix and the phenolic compounds [31].

Water Contact angle (WCA) measures the wettability of a material based on the behavior of a liquid droplet on its surface. Surfaces with a contact angle < 90º are hydrophilic [17]. The WCA of the films ranges from 19.5 (AP-B) to 46.27 (AP/CMC-B) (Table 3). Contact angles below 90 °C can be attributed to the hydrophilic compounds in the vegetable purees. The AP-B film exhibited greater hydrophilicity than the CMC-containing films. This behavior may be attributed to the formation of a more cohesive polymer network resulting from interactions between the CMC and the polyphenols present in the *Allium* purees, as evidenced by the SEM micrographs (Fig. 2) and PVA values (Table 3). The addition of CMC likely promoted a more compact film structure by reducing the availability of hydroxyl groups, thereby decreasing the material's surface wettability [36]. Similar effects have been previously reported following CMC incorporation into a banana peel-based film [11] and with the addition of shallot onion waste extracts in sodium alginate films formulated with carboxymethyl cellulose [36]. Moreover, no significant differences were observed between films produced via bench casting and continuous casting, reinforcing the beneficial impact of CMC on film microstructure and surface properties.

The addition of CMC increased the contact angle, indicating reduced surface hydrophilicity due to intermolecular interactions between CMC and the Allium matrix, likely involving hydrogen bonding that limits the availability of hydroxyl groups on the film surface [44]. This interpretation is corroborated by the FTIR-ATR spectra, which exhibit an absorption band around 3500 cm^{-1}, associated to O–H stretching (Fig. 3 (A)). A similar increase in WCA was previously reported by Thyvia et al. [36] for films incorporating shallot onion waste extract, attributed to a reduction in surface hydrophilic functional groups. Comparable trends were also observed by Silva et al. [11]. Furthermore, the contact angle measured on the top face of the bench-cast film was higher than that of the continuous-cast film, while no difference was detected on the bottom face in contact with the substrate. Despite these differences, all films remained hydrophilic, indicating comparable overall wettability. This observation aligns with previous findings where the processing method had no significant effect on the wettability of biocomposite films [18].

Thermal properties

Initial temperature (Tonset), final degradation temperatures (Toffset), and residual mass at 500 °C (R500%) of Allium films are shown in Table 4. TG and DTG curves were shown in Fig. 4. From the TG and DTG curves, it is possible to observe that the mass variation for the bench and continuous casting treatments is very similar (Fig. 4). The first mass loss, below 100 °C, refers to the loss of residual water from the film after drying. Around 130 °C, the second stage involves the initial degradation of short-chain sugars, such as glucose and sucrose. The third stage occurred at approximately 170 °C and is attributed to the degradation of phenolic compounds and other organosulfur compounds, and finally, the fourth stage, the loss, between 270 and 500 °C, is related to the degradation of lignin, hemicelluloses, and cellulose. Considering that the control treatment loses more mass at the end of the analysis, it can be inferred that CMC provides greater thermal stability to the films. The mass loss of the sample is associated with water evaporation and the volatile products from reaction or decomposition.

DSC measures the energy variation between the sample and a reference material as a function of temperature and time. Firstly, the films were heated to 140 °C to remove residual moisture and then heated again. DSC curves were obtained from the second heating and are presented in Fig. 4. The glass transition temperatures (Tg), observed from the DSC curves, are shown in Table 4. The initial glass transition temperatures varied from 4.4 °C for the film without CMC and 3.3 °C and 3.2 °C for AP/CMC-B and AP/CMC-CC, respectively. Monosaccharides, such as glucose, sucrose, and fructose, reduce the material's Tg [5, 45, 46]. This behavior can be attributed to the interaction of low-molecular-weight sugars with the polymer matrix, which enhances polymer chain mobility and consequently increases film flexibility. In a related study, Dias et al. [9] reported higher Tg values for onion-based bioplastics than those reported in the present work. Films produced from raw onion exhibited a Tg of 46.7 °C, while those produced without the liquid fraction after hydrothermal pretreatment showed a significantly lower Tg of 9.6 °C. Conversely, films subjected to both hydrothermal pretreatment and a subsequent washing step displayed Tg values ranging from 42.6 to 45.0 °C. The authors attributed the lower Tg to the retention of decomposition by-products, particularly solubilized sugars generated during pretreatment, which acted as

Table 4 Initial (Tonset) and final degradation temperatures (Toffset) and residual mass at 500 °C (R500 (%)) of *Allium* films

	TG/DTG curves								
Film	ΔT_1 (°C)	Δm_1 (%)	ΔT_2 (°C)	Δm_2 (%)	ΔT_3 (°C)	Δm_3 (%)	ΔT_4 (°C)	Δm_4 (%)	Residual masses (%)
AP-B	30.0–126.53.0.53	6.9	126.5–172.4.5.4	15.6	172.4–284.5.4.5	44.2	284.5–493.8.5.8	70.6	29.4
AP/CMC-B	30.0–132.78.0.78	7.2	132.8–174.6.8.6	14.0	174.6–274.8.6.8	38.5	274.8–493.6.8.6	66.3	33.7
AP/CMC-CC	30.0–131.59.0.59	7.9	131.6–175.0	14.4	175.0–279.5.0.5	39.6	279.5–493.9.5.9	65.5	34.5

ΔT: temperature variation; Δm: mass loss variation; R500: residual mass at 500 °C

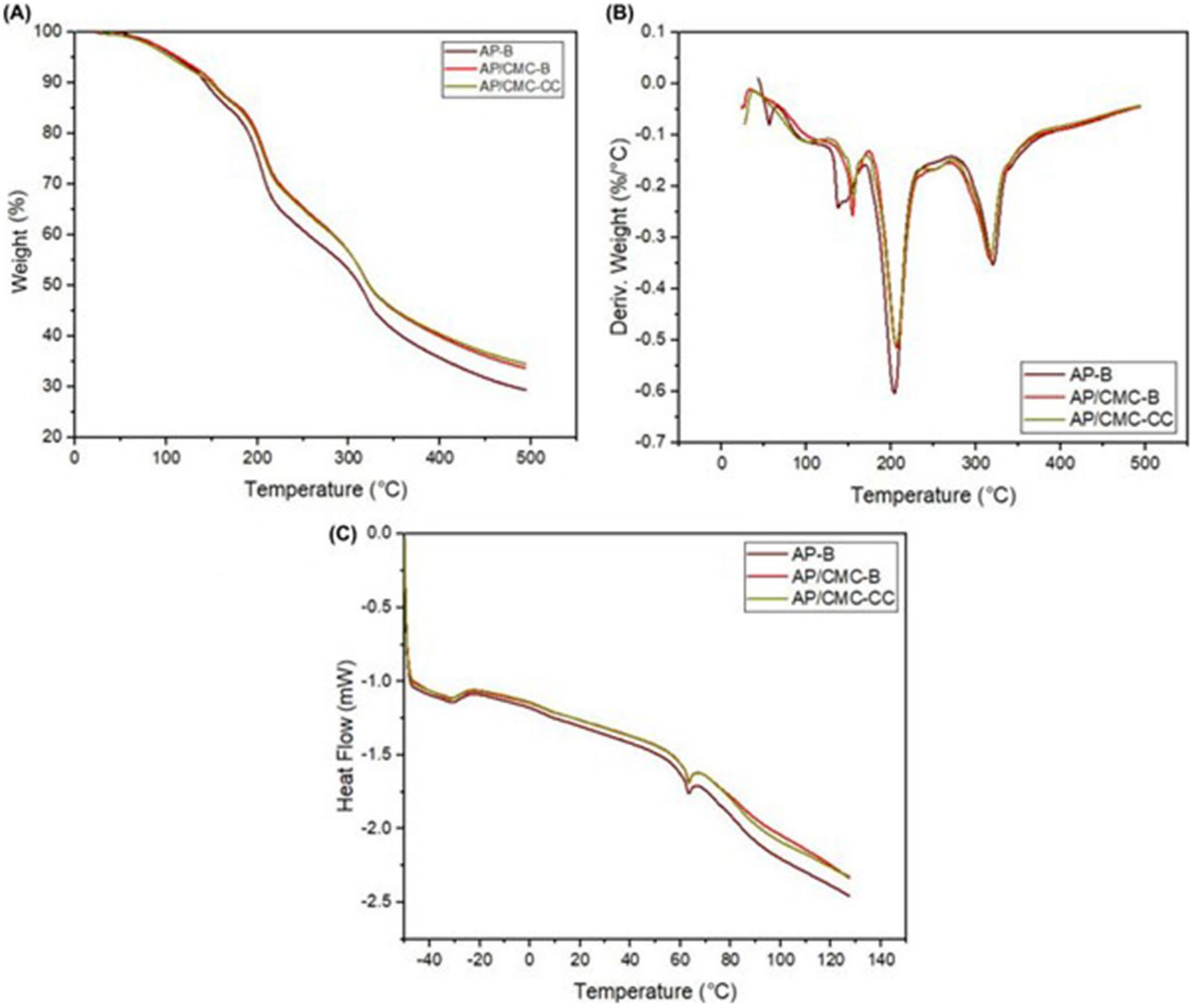

Fig. 4 (**A**) Thermogravimetric (TG), (**B**) differential TG (DTG) and (**C**) Differential Scanning Calorimetry (DSC) curves of Allium films obtained from bench casting without CMC (AP-B), or with the addition of CMC produced by bench (AP/CMC-B) or continuous casting (AP/CMC-CC)

plasticizers. Similarly, the lower Tg observed in our films is likely due to the presence of retained soluble sugars, which reduce intermolecular forces and enhance molecular mobility. This characteristic suggests that the material remains malleable over a wider temperature range before becoming brittle, providing advantages in processing and handling. The Tg of the films developed in this study (3.2–4.4 °C) was lower than values reported for onion-based films (9.6–46.7 °C), indicating of decomposition by-products solubilized, as well as soluble sugar during pretreatment [9].

Furthermore, CMC increases the material's heat capacity but does not alter the glass transition range. Despite the film production conditions, the films produced by bench and continuous methods present the same chemical structure. Thus, by modifying the production method, it is suggested that the chemical alterations are not significant, demonstrating that the formulation is robust to changes, making the film production process easily scalable. The melting event, an endothermic event, is altered by the presence of CMC. The films produced with CMC show the same melting area with the same amount of energy released to degrade the same amount of mass from the films.

Heat sealing and seal strength determination

The sealing average and maximum strength values are shown in Table 5. Considering the higher thermal resistance of *Allium* films with CMC (Table 5), the seal strength for both films was investigated. The sealing failure was due to the adhesive type and delamination. The average seal

Table 5 Glass transition temperature (Tg) obtained by differential scanning calorimetry (DSC) and seal strength of *Allium* films produced on a bench without CMC (AP-B), or with the addition of CMC produced by bench (AP/CMC-B) or continuous casting (AP/CMC-CC)

Film	Tg (°C) (onset)	Tg (°C) (mid)	T_1 (°C)	T_2 (°C)	Initial mass (mg)	Mass loss (J/g)	Seal strength (avg) (*N*/15 mm)	Seal strength (max) (*N*/15 mm)
AP-B	4.4	8.3	61.2	63.1	6.5	0.354	-	-
AP/CMC-B	3.3	8.3	61.0	63.3	6.1	0.414	0.309 ± 0.153	2.750 ± 0.762
AP/CMC-CC	3.2	8.6	61.4	63.1	6.2	0.410	0.338 ± 0.157	2.070 ± 0.775

[a]Tg (°C) (onset) = temperature to the beginning of the glass transition; Tg (°C) (mid) = midpoint glass transition temperature; T_1 (°C) = first degradation temperature; T_2 (°C) midpoint degradation; Strength (avg) = Sealing strength average; Strength (avg) = Sealing strength maximum

strength was 0.309 and 0.338 N/15 mm, and the maximum seal strength was 2.750 and 2.070 N/15 mm for AP/CMC-B and AP/CMC-CC, respectively, showing no significant difference between them (t-test, $p < 0.05$), which indicates that the sealing behavior is independent of the film production method. This result was expected, since the films have the same matrix and similar thermal stability, as demonstrated by the TG/DTG curves (Fig. 4). Similar results were found by Lu et al. [47] in the maximum sealing force in edible soy protein films (2.38 N/15 mm). The authors explain that the sealing strength is due to the intermolecular interaction in the heat-sealing area [47]. The maximum sealing strength is found when the sample is stressed to rupture and indicates the maximum resistance required to separate one flexible material from another. In contrast, the average strength indicates the average resistance for the same to occur. In edible films, opening the pouches, sachets, or 'ingredient delivery systems' sachet, is not always desirable, as the film will be prepared and consumed together with the food to which they were applied. Therefore, the sealing capacity of the films is essential because it allows the prediction of the performance of the packaging during storage and handling. The seal must be sufficient to keep the product inside the packaging and prevent food contamination [48].

Optical properties

Color is a fundamental property in food, as appearance is the first characteristic the consumer evaluates and indicates the product's freshness and quality. Therefore, packaging applications must consider this factor. The coloration results of the films are shown in Table 3. The parameter L* corresponds to the brightness and ranges from 0 - black to 100 - white, while a* and b* indicate the color hue from green (-) to red (+) and blue (-) and yellow (+), respectively. The addition of CMC significantly increased the L* values of the films, indicating that they became lighter. This effect is attributed to the higher proportion of colorless components introduced by CMC, which is intrinsically colorless. Additionally, incorporating CMC reduced the red intensity of the films (Table 3). In contrast, chroma values were not influenced by either CMC addition or processing method. However, the hue angle increased in films produced by continuous casting, indicating a shift toward a more yellowish tone (Table 3). These results suggest that while CMC incorporation primarily affects film luminosity, the processing method influences color tone. The higher hue angle observed in continuous-cast films may be associated with their lower thickness, which can alter light scattering and color perception.

Light barrier properties are fundamental in food packaging to help prevent photo-oxidation, reduce food deterioration, and improve shelf life [49]. The interaction of the *Allium* films with UV light is shown in Table 3. All the films locked over 99% of radiation in the UVA, UVB, and UVC ranges (Table 3). There were no differences in the UV values of the films, suggesting that the incorporation of CMC and the film-forming method did not alter the UV barrier properties of the *Allium* films. The high UV light protection observed in the films can be attributed to their high phenolic content (Table 3). Similar behavior was reported by Thivya et al. [36] in alginate films incorporating shallot onion waste extract, where reduced light transmittance was associated with interactions between the film matrix and polyphenols. Likewise, Barbosa et al. [49] reported comparable UV barrier performance in onion-based biocomposite films. Notably, our results demonstrate that neither the addition of CMC as a reinforcing agent nor the processing method (bench or continuous casting) significantly affected the UV barrier properties. These findings indicate that the UV protective performance of the films is maintained even under scalable production conditions.

Antioxidant properties

Total phenolic compounds (TPC) and antioxidant activities of the films are shown in Table 3. The total phenolic content of the *Allium* film without CMC was significantly higher than that of the film with CMC, also produced on the bench. The same result was observed for the Total flavonoid content (TFC) expressed as quercetin (Table 3). The TPC and TFC values of the films can be attributed to phenolic compounds present in the *Allium* species, which either resisted the heat pretreatment or were released during it (results of

TPC and TFC analyses of purees before and after pretreatment are provided in Table S1, Supplementary Information). The decrease in the TPC and TFC values in the films produced on the bench casting with the added CMC can be attributed to a possible dilution of phenolic compounds, as CMC increases the total solids without contributing bioactive components. This hypothesis is supported by the FTIR spectra (Fig. 3 (A)) in which in the 1618 cm^{-1} region, characteristic of such compounds, is visibly more intense in the film without CMC, suggesting that the phenolic compounds were more freely available in the AP-B film. Phenolic compounds present in fruits and vegetables contain oligomers or dimers linked by glycosidic bonds that undergo hydrolysis during heat pretreatment, forming Maillard reaction products, which have been reported to positively influence phenolic content and antioxidant capacity in *Allium* species [12, 13].

Multiple methods should be used to determine antioxidant activity, as the oxidation process is complex and involves multiple mechanisms [7]. In the present study, DPPH and $ABTS^+$ radical scavenging analysis and iron ion reduction analysis were performed to determine the antioxidant capacity of the films. The antioxidant activity of the *Allium* films was ~95% inhibition for all films by the DPPH method and ranged from 94 to 97.6% by the ABTS method, and for the film prepared on the bench with CMC (97.7%), it was significantly higher than for the film prepared by the same process without the addition of CMC. The FRAP method, which measures the ability of antioxidants to reduce the ferric ion complex to ferrous ions, ranged from 3157 to 18,798 µM FeSO4/g of film and was significantly higher for the films produced using bench casting. In general, the films showed excellent antioxidant activity across the three methods, regardless of the presence of CMC or the method used to form the films. Thus, the antioxidant potential of the films appears to be related to the bioactive compounds present in the *Allium* purees and their ability to act as free radical scavengers [50].

The antioxidant activity of the films was monitored over time, with a view to their potential application in fatty foods. The inhibition of the DPPH radical by the films over time is shown in Fig. 3 (B). All films increased inhibition of the DPPH radical over time, with the AP-B film showing the highest inhibition percentage at all time points evaluated. After 30 min, the AP/CMC-B and AP/CMC-CC films showed similar inhibition percentages ($p < 0.05$). However, in the following times, the inhibition percentages of all films were significantly different. Interestingly, the results indicate that the presence of CMC in the *Allium* films reduces the inhibition of the DPPH radical. This may be due to intermolecular interactions between the polymer and the *Allium* puree, as evidenced by FTIR (Fig. 3(A)), which may hinder the release of phenolic compounds from the films, or because the addition of CMC may have diluted these compounds. Similar results were obtained in a study with banana peel films by Silva et al. [11] in which the addition of CMC negatively affected the TPC and AA content determined by DPPH and FRAP. Furthermore, this result suggests that the continuous-casting drying method has a lesser effect on the antioxidant power of *Allium* films produced with CMC. HPLC analysis was performed on the two hydrothermally treated purees and on the film produced in continuous casting. Quercetin was detected only in the onion puree and the film AP/CMC-CC. This result was expected because quercetin accounts for 80% of the flavonoids in onion and can be found free or bound to other sugars in vegetables (Nile et al., 2017), whereas in GA the major bioactive compounds are allicin, alliin, and ajoene [50]. The quercetin in onion is 1497 mg/kg, while in garlic it is only 47 mg/kg, according to Lanzotti [6]. Therefore, onions are expected to exhibit greater radical-scavenging activity than garlic, especially since the films and extracts were prepared from onion peels [6]. This result also indicates the flavonoid's ability to withstand the high temperatures used during pretreatment. However, its effectiveness decreases after the production of edible film on an industrial scale, which may be due to the formation of the polymer network, favoring retention of active compounds, as verified in the DPPH test over time (Fig. 3 (B)).

In addition, these results suggest that pretreatment can increase antioxidant activity (Table S1). This is because organosulfur compounds can also contribute to the high antioxidant activity of *Allium* films [6, 50]. During hydrothermal pretreatment, autohydrolysis of these compounds also occurs, and the amino groups of these flavor precursors react with reducing sugars through the Maillard reaction. The final and intermediate products of the reaction, such as furfural, for example, are considered to have high antioxidant power [12]. Thus, the bioactive compounds in *Allium* species are increased after heating (80 to 120 °C) [12, 13, 51]. Nile et al. [7] demonstrated that the antioxidant activity in solid onion residues was lower than in this study using the DPPH (74.3%), ABTS (68.3%), and FRAP (70.3%) methods [7]. Similar results were obtained by Nile et al. [52], who reported 65.5% and 72.3% inhibition of DPPH and FRAP, respectively. Thus, the high antioxidant activity of the film is mainly associated with the presence of quercetin and bioactive organosulfur compounds found in *Allium* species, which can be either resistant to or even enhanced by hydrothermal pretreatment. The antioxidant properties of the pretreated onion (ONT) and garlic (GAT) purees are provided in Table S2 (Supplementary Information), with the onion puree showing superior results in all methods analyzed. These results also reveal that the integral use of

Allium bulbs appears to be a suitable approach for producing edible and active Allium *film*s. The sugar content was determined among the films (Table 3) and the purees (Table S2). In the films, the total sugar content ranged from 8.43 to 10.61 g of glucose/100 g of film, with no significant difference among them. In contrast, among the purees, the yellow onion puree showed the highest sugar content. This analysis aimed to assess a possible plasticizing effect of sugars on the Allium films, as mentioned in the mechanical properties section, suggesting that the plasticizing effect was mainly due to the sugars present in the onion pure.

Antimicrobial properties

The antimicrobial activity of the films against Gram-negative (*E. coli*) and Gram-positive (*S. aureus*) bacteria was evaluated by the agar diffusion method. The films did not show a zone of inhibition against any of the bacteria studied. Some flavonoids, such as quercetin, have been reported to have antimicrobial effects against *E. coli* [3]. This may have occurred because the active compounds did not migrate from the film into the medium, likely due to interactions. with the matrix. However, the active film is intended for use under refrigeration conditions, which help reduce bacterial proliferation.

Conclusions

The present study demonstrated that hydrothermal pretreatment of whole Allium purees enhances antioxidant properties, as reflected in the excellent antioxidant activity of the resulting films. In addition, hydrothermal pretreatment favored the formation of cohesive, continuous onion-based films, as previously reported, eliminating the need for conventional plasticizers, such as glycerol, and advancing toward formulations with reduced additives. The incorporation of CMC into the Allium puree improved the mechanical, water-barrier, and thermal stability of the films and enabled the production of edible films at a larger scale, with performance comparable to, and in some cases superior to, films obtained by bench casting. The films also exhibited excellent UV-light barrier properties and strong antioxidant activity, regardless of the production method. Overall, the results indicate that Allium-based edible films have potential for application in perishable, lipid-rich foods prone to lipid oxidation and photooxidation, while providing characteristic sensory attributes. Nevertheless, further studies are required to evaluate the practical application of these films and their effects on the final quality of foods.

Authors' contributions Conceptualization: Henriette Monteiro Cordeiro de Azeredo; Methodology: Raquel Alves Mauricio, Severino Matias de Alencar, Michel Brienzo, Jovan Duran Alonso, Henriette Monteiro Cordeiro de Azeredo; Formal analysis and investigation: Raquel Alves Mauricio, Josemar Gonçalves de Oliveira Filho, Leticia Bueno dos Santos, Severino M. de Alencar; Rosangela Almeida Maia; Michel Brienzo; Writing – original draft preparation: Raquel Alves Mauricio; Writing – review and editing: Henriette Monteiro Cordeiro de Azeredo, Josemar Gonçalves de Oliveira Filho; Funding acquisition: Renata Tieko Nassu; Resources: Renata Tieko Nassu, Henriette Monteiro Cordeiro de Azeredo; Supervision: Henriette Monteiro Cordeiro de Azeredo.

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Declarations

Competing interests The authors have no competing interests to declare that are relevant to the content of this article.

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