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Mechanical Characterization of Biopolymer-Based Hydrogels Enriched with Paulownia Extracts Recovered Using a Green Technique

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Abstract: This study deals with the development and mechanical characterization of innovative starch- and gelatin-based hydrogels enriched with hydrothermal aqueous extracts from Paulownia (i.e., bark, leaves, petioles). The color, rheology, and texture properties of formulated biopolymer-based hydrogels depending on the processing conditions of the extracts was evaluated for the different Paulownia fractions. Results indicated that a clear impact on color features of the tested hydrogels was observed with the incorporation of Paulownia extracts. Rheological testing showed that weak and intermediate strength hydrogels were identified for starch- and gelatin-based hydrogels independently of used extract. A relevant softening of all gelled matrices was observed in the presence of recovered liquor extracts involving the following trend (leaves > barks > petioles). For all fractions, the highest viscoelastic features were achieved for hydrogels made with extracts recovered at 140 °C. Texture outcomes confirmed the rheological achievements. No syneresis was observed in developed hydrogels after two weeks of cold storage.

Keywords: paulownia; autohydrolysis; hydrogels; starch; gelatin; rheology

1. Introduction

Currently, there is a growing social demand in obtaining bioactive compounds because they have a positive effect on health, aiding in the prevention and/or mitigation of some chronic diseases, due to their antioxidant related activities [1]. Bioactives are derived from the secondary metabolism of plant sources [2]. The search for natural sources that allow obtaining bioactives has gained attention of the scientific community. In new investigations, authors report the recovery of the bioactive compounds from a variety of sources such as cornelian cherry [3], coffee cherry pulp [4], apple pomace [5], rose hip [6], or Pavlova pinguis [7].

A source of potential interest in obtaining bioactive compounds could be Paulownia. This genus is native to China and Southeast of Asia and includes tree varieties that are characterized by a rapid growth. For this reason, its main use is focused on biomass exploitation for timber use [8,9]. However, there are studies that report that fractions such as leaves, bark, flowers, and fruits are rich in antioxidant compounds among which phenols, flavonoids, terpenoids, and lignans stand out [10,11]. Paulownia plant was traditionally used because of its medicinal properties [12]. Therefore, an interesting strategy could be to consider exploitation of this natural source from two points of view, such as the use of the wood; on the other hand, as valorization of residual fractions to obtain bioactive compounds with environmentally friendly technologies.

There are numerous extraction technologies for obtaining compounds of interest but the development of greener efficient processes is required and the use of water as solvent is especially promising [13]. The autohydrolysis is one of them, and consists of the treatment of the raw material with water as solvent at high temperature and pressure in order to solubilize the interest compounds. Autohydrolysis or subcritical water is widely used in the fractionation of lignocellulosic materials. In this catalytic process the acid groups from the raw material cause the solubilization of carbohydrates [14].

Bioactive compounds have a notable role in different areas such as food and pharmaceutical industries [15]. A novel application of the bioactive fraction from the extracts would be its use as a solvent for the production of hydrogels. The formulation of hydrogels from natural matrices such as gelatin and starch stand out. Gelatin is one of the most commonly used bases and starch has boomed in recent years. Gelation is a common phenomenon in foods, and the molecules responsible for the gelation are typically proteins or polysaccharides [16]. In this context, the main objective of this study is the development of biopolymer-based hydrogels enriched with extracts from natural underused sources and the corresponding thermomechanical characterization. In a parallel work, the bioactive features of used extracts here were comprehensively detailed [17].

2. Materials and Methods

Paulownia elongata x fortunei is a sterile hybrid clone created by Cotevisa (COTEVISA 2®) placed in Valencia, Spain and harvested by Maderas Álvarez Oroza S.L. in Nois, Lugo, Galicia, Spain. Leaves, petioles, and bark were collected in June 2017. Petioles were manually separated from the leaves. All raw materials were dried at room temperature. Then, they were milled (Retsch, Schneidemühle, SM1; Haan, Germany) and stored in a cold and dry place. Note here that samples were periodically characterized before starting any experimental treatment in order to ensure nondegradation of the raw materials.

Agria variety potatoes were provided by the Instituto Ourensán de Desenvolvemento Económico (INORDE). Further information about the composition of this raw material was previously reported in a parallel work [17–20].

2.1. Autohydrolysis and Severity Factor

All Paulownia materials proposed in this work were extracted by autohydrolysis. For it, ground samples were mixed with distilled water at different liquid-to-solid mass ratio of 7:1 (*w/w*) for bark and 15:1 (*w/w*) for leaves and petioles. A Parr Instrument Company reactor (Series 4560, Moline, IL, USA) with 600 mL of capacity was used. The experiments were subjected to different temperatures in the range 140–240 °C with a constant agitation (150 rpm). The extractions were performed under nonisothermal conditions so that, once the selected temperature was reached, the system was immediately cooled. Finally, the solid and liquid phases were separated by filtration.

Severity factor (log R_0) was calculated using Equation (1). This parameter was introduced by Overend and Chornet (1987). R_0 includes the effect of time and temperature parameters and provides an easy way to compare results among experiments carried out under different operational conditions. Usually, the severity, S, is expressed as the decimal logarithm of R_0.

$$R_0 = \int_0^t \exp\left(\frac{T - T_r}{\omega}\right) dt \quad (1)$$

where T: temperature (°C); Tr: reference temperature (°C); t: reaction time; ω: activation energy of reaction (constant = 14.75).

2.2. Protein Determination

The quantification in protein content of bark liquors was determined following the method proposed by Bradford (1976). Note here that protein content for leaves and petioles liquors were previously determined and included here for comparison [17]. Commercial Bovine Serum Albumin (BSA, Sigma, Aldrich, St Louis, MO, USA) was used as standard. Briefly, Bradford reagent

(1 mL) was added to the sample (100 µL) and incubated at room temperature for 5 min. Subsequently, absorbance was measured at 595 nm. Results were expressed in µg BSA equivalents/L. Analyses were undertaken at least in triplicate.

2.3. Potato Starch Extraction

Potato starch from an underused source was selected for the hydrogel preparation. Particularly, Agria potato variety was chosen as raw material for the starch extraction based on the results previously found from different underused potato varieties [20]. The recovery of potato starch included the following steps. Firstly, a manual peeling of potatoes was undertaken in order to obtain the flesh fraction. This portion was cut and milled, and contacted with distilled water in a solid:liquid mass ratio of 1:2 for 1 h with constant agitation (200 rpm). After this, the flesh fraction was separated from the liquid filtrate with a sieve (71 µm) and water-washed several times to enhance the starch fraction. The liquid filtrate containing the starch was put in the fridge to promote the starch decantation. After 24 h, starch fraction was recovered and placed in an oven at 40 °C for 48 h to dry it. Finally, to obtain a homogeneous sample in terms of moisture content, the starch in petri dishes was stored in a desiccator with $MgCl_2$ until constant weight.

2.4. Hydrogel Preparation

Two biopolymers were proposed as gelling agents in the preparation of hydrogels. Namely, commercial gelatin (Scharlau, CAS number 9000-70-8) which is a commonly used biopolymer and extracted starch from underused potato sources. Hydrogels were prepared with autohydrolysis liquors (5 mL) of leaves, petioles, and bark as solvents. A fixed content of biopolymer was set, in the case of the gelatin a concentration of 2% was used and the starch was used at 15%. The autohydrolysis extracts were mixed with the gelling agents in the appropriated amounts at 90 °C until homogeneity was noticed. Formulated systems were stored at 4 °C to ensure gel maturation for 24 h. Note here that the biopolymers content and the processing conditions were selected based on the results reported in a previous work [20].

2.5. Color Measurements

Color properties of the liquid extracts determined using a Minolta CR-400 colorimeter (Japan) based in the CIELab system (L*, a*, and b*). The chromaticity responsible parameters were given by L* (degree of whiteness, $0 < L^* < 100$, brightness), a* (degree of redness ($a^* > 0$) or greenness ($a^* < 0$), and b* (degree of yellowness ($b^* > 0$) or blueness ($b^* < 0$). Note here that for ΔE* determination, 140 °C was selected as the reference temperature.

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (2)$$

All samples were measured at room temperature under the same light conditions at least four times.

2.6. Mechanical Characterization of the Formulated Matrices

The viscoelastic features of developed hydrogels, in terms of elastic (G′) and viscous (G′′) moduli, were determined using a controlled-stress rheometer (MCR302, Anton Paar Physica, Austria). For this purpose, small-amplitude oscillatory shear (SAOS) testing was performed using a plate-plate geometry (25 mm of diameter and 1 mm of gap). Hydrogels were placed on the measuring system and the edges sealed with paraffin. Samples were rested for 5 min before rheological measurements in order to favor the thermal and structural equilibration. Afterwards, stress sweeps were run (1 Hz, from 0.1 to 100 Pa, 25 °C) to define the linear viscoelastic region < 20 Pa for gelatin hydrogels and < 45 for starch hydrogels. Subsequently, the mechanical spectra were monitored from 0.1 to 10 Hz at 25 °C within the linear regime (15 Pa). Rheological measurements were made at least in triplicate.

Macro structural features of above hydrogels (25 mm diameter) were determined using a texture profile analysis (TPA) on a TA-XT2 texture analyzer (Stable Microsystems, Godalming, UK). A stainless steel probe (P50) was used for the textural testing in compression mode (5 kg load cell). The testing conditions were 1.0 mm/s crosshead speed and 2 mm compression, compacting the hydrogels twice in a reciprocating motion. Firmness (kgf), adhesiveness (kgf s), cohesiveness (-), and springiness (-) of the formulated hydrogels were determined from the texture plots. Note here that the springiness was measured as the distance of height during second compression by the first compression distance Textural trials were undertaken at least five times.

2.7. Statistical Analysis

Data sets were statistically assessed using one-factor analysis of variance (ANOVA) employing the PASW Statistics v.22 software (IBM SPSS Statistics, New York, NY, USA). A post-hoc Scheffé test was performed to differentiate average values with a 95% confidence ($p < 0.05$).

3. Results and Discussion

3.1. Severity in Autohydrolysis Treatment

Figure 1 presents the heating and cooling temperature profiles for the nonisothermal autohydrolysis treatment of Paulownia leaves, petioles, and barks. In all cases, the temperature curves exhibited similar thermal behaviors between tested Paulownia fractions. Namely, the time and temperature influence on the autohydrolysis processing was analyzed based on the severity factors (Table 1). It should be indicated that severity values ($S = \log R_0$) were determined using 80 °C as initial and final temperature. As expected, severity values increased with increasing autohydrolysis temperature. At fixed temperature, no notable differences were identified between petioles, leaves, and barks treatments. This is consistent with the results previously reported for *Lentinus edodes* [14] and wheat straw [21] treated using autohydrolysis at similar experimental conditions.

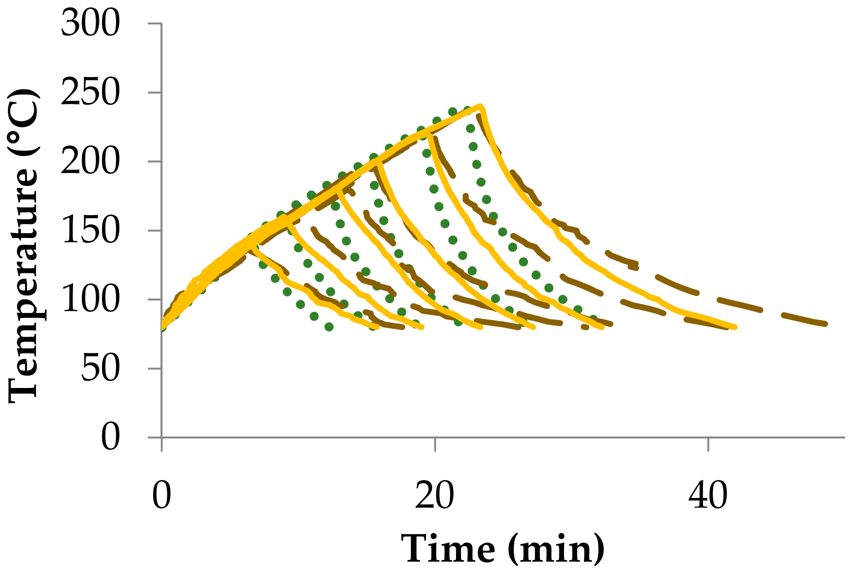


Figure 1. Temperature profile of different experiment of Paulownia bark (dashed line), leaves (dotted line), and petioles (continuous line).

Table 1. Severity factor of autohydrolysis extraction of different parts of Paulownia.

		Bark	*Leaves*	*Petioles*
140 °C	R_0	23.13	24.64	26.75
	Severity	1.36	1.39	1.43
160 °C	R_0	151.1	122.61	139.14
	Severity	2.18	2.09	2.14
180 °C	R_0	560.47	569.57	537.4
	Severity	2.75	2.76	2.73
200 °C	R_0	2188.13	2356.25	2013.74
	Severity	3.34	3.37	3.3
220 °C	R_0	9545.34	9696.99	10,073.78
	Severity	3.98	3.99	4
240 °C	R_0	35,207.12	36,810.67	42,454.67
	Severity	4.55	4.57	4.63

Note here that for all temperatures the standard deviations were <2.5%.

3.2. Color Measurements

Table 2 summarizes the color parameters for tested starch-based and gelatin-based hydrogels. In general, lightness values were higher for starch-based hydrogels. In both matrices, no clear tendencies were identified for the individual color coordinates; whereas the color differences parameter (ΔE*) exhibited a marked behavior. Note here that in all cases the color differences were calculated regarding the lowest autohydrolysis processing temperature (140 °C). In general, it was observed that ΔE* showed a random trend with increasing temperature for both starch-based and gelatin-based hydrogels. For bark and petiole hydrogels, higher color differences were observed for those made with a starch matrix. A reverse tendency was identified for gelatin-based hydrogels. Following the Adekunte et al. [22] classification, very different color features (ΔE*) were identified for the majority of the samples especially for starch-based hydrogels enriched with petioles and those prepared with gelatin incorporated with leaves. The latter authors established no significant color differences for ΔE* < 1.5; distinct color values for ΔE* between 1.5 and 3.0 and very different for ΔE* > 3.0. According above classification, only starch-based hydrogels incorporated with bark (at 160 and 200 °C) or leaves (at 200 °C) as well as those gelatin-based hydrogels formulated with bark (at 180 and 200 °C) or petioles (at 240 °C) exhibited no significant color differences. Even though, the magnitudes of color parameters were consistent with those previously found for other hydrogels formulated with similar biopolymer matrices enriched with extracts from different food leaves [23] or peels [20].

Table 2. Color coordinates by CIEL*a*b* system for hydrogels formulated with starch (**a**) and gelatin (**b**).

(a) Starch Hydrogels		**Coordinates**			
	T ^{a} (°C)	**L***	**a***	**b***	**ΔE**
Bark	140	30.82 ± 1.16 ^{d}	0.37 ± 0.05 ^{g}	7.42 ± 0.14 ^{d}	-
	160	31.29 ± 0.77 ^{d}	0.71 ± 0.03 ^{e}	8.20 ± 0.18 ^{c}	0.97 ± 0.39
	180	27.33 ± 0.36 ^{f}	0.92 ± 0.05 ^{d}	6.40 ± 0.34 ^{e}	3.68 ± 0.83
	200	30.57 ± 0.74 ^{d}	0.31 ± 0.05 ^{g}	6.34 ± 0.42 ^{e}	1.11 ± 0.51
	220	34.75 ± 0.79 ^{c}	5.33 ± 0.13 ^{a}	17.70 ± 0.54 ^{a}	12.07 ± 0.55
	240	36.11 ± 0.21 ^{b}	0.58 ± 0.07 ^{f}	14.42 ± 0.37 ^{b}	8.77 ± 0.98
Leaves	140	24.96 ± 0.16 ^{g}	2.94 ± 0.06 ^{b}	2.29 ± 0.034 ^{g}	-
	160	23.96 ± 0.80 ^{g}	1.81 ± 0.11 ^{c}	1.27 ± 0.28 ^{h}	1.82 ± 0.66
	180	25.11 ± 0.90 ^{g}	−0.45 ± 0.09 ^{i}	0.20 ± 0.03 ^{j}	3.98 ± 0.74
	200	23.98 ± 0.80 ^{g}	−0.54 ± 0.07 ^{i}	−0.02 ± 0.06 ^{k}	4.29 ± 0.64
	220	25.03 ± 0.43 ^{g}	−0.62 ± 0.03 ^{i,j}	0.41 ± 0.04 ^{i}	4.02 ± 0.27
	240	25.11 ± 0.33 ^{g}	2.85 ± 0.05 ^{b}	1.52 ± 0.14 ^{h}	0.79 ± 0.21
Petioles	140	34.38 ± 0.45 ^{c}	−0.61 ± 0.04 ^{i,j}	8.18 ± 0.18 ^{c}	-
	160	31.06 ± 0.30 ^{d}	−0.75 ± 0.08 ^{j}	6.65 ± 0.18 ^{e}	3.66 ± 0.15
	180	30.93 ± 0.34 ^{d}	−0.64 ± 0.08 ^{j}	6.94 ± 0.24 ^{e}	3.67 ± 0.13
	200	30.31 ± 0.60 ^{d}	0.79 ± 0.10 ^{e}	8.17 ± 0.24 ^{c}	4.31 ± 0.18
	220	29.97 ± 0.16 ^{d}	0.41 ± 0.06 ^{g}	5.47 ± 0.03 ^{f}	7.96 ± 0.32
	240	37.22 ± 0.45 ^{a}	0.14 ± 0.03 ^{h}	14.28 ± 0.44 ^{b}	6.77 ± 0.27

Table 2. *Cont.*

(b) Gelatin hydrogels			**Coordinates**		
	T ^{a} (°C)	**L***	**a***	**b***	**ΔE**
Bark	140	25.14 ± 0.14 ^{a}	0.53 ± 0.06 ^{f}	1.30 ± 0.07 ^{d}	-
	160	24.24 ± 1.00 ^{a}	−0.46 ± 0.13 ^{h}	2.43 ± 0.23 ^{c}	1.74 ± 0.88
	180	25.64 ± 0.84 ^{a}	0.002 ± 0.09 ^{g}	0.35 ± 0.12 ^{e}	1.20 ± 0.70
	200	25.20 ± 0.69 ^{a}	0.47 ± 0.07 ^{f}	0.45 ± 0.07 ^{e}	0.86 ± 0.55
	220	25.09 ± 0.21 ^{a}	3.86 ± 0.13 ^{c}	1.45 ± 0.13 ^{d}	3.33 ± 0.12
	240	24.81 ± 0.58 ^{a}	6.93 ± 0.45 ^{a}	3.74 ± 0.05 ^{b}	6.85 ± 0.59
Leaves	140	20.02 ± 0.86 ^{c}	−0.42 ± 0.24 ^{h}	2.64 ± 0.13 ^{c}	-
	160	24.65 ± 0.66 ^{a}	−0.34 ± 0.04 ^{h}	0.29 ± 0.08 ^{f}	5.20 ± 0.28
	180	24.49 ± 0.43 a	−0.52 ± 0.07 ^{h}	−0.13 ± 0.06 ^{g}	5.26 ± 0.46
	200	23.52 ± 0.80 ^{b}	−0.38 ± 0.09 ^{h}	3.78 ± 0.16 ^{b}	3.68 ± 0.48
	220	22.85 ± 0.39 ^{b}	−0.05 ± 0.18 ^{g}	4.37 ± 0.07 ^{a}	3.34 ± 0.48
	240	23.28 ± 1.65 ^{b}	5.37 ± 0.75 ^{b}	4.43 ± 0.56 ^{a}	6.88 ± 1.04
Petioles	140	24.96 ± 0.16 ^{a}	2.94 ± 0.06 ^{d}	2.29 ± 0.30 ^{c}	-
	160	23.96 ± 0.80 ^{a,b}	1.81 ± 0.11 ^{e}	1.27 ± 0.28 ^{d}	1.82 ± 0.69
	180	25.11 ± 0.90 ^{a}	−0.45 ± 0.09 ^{h}	0.20 ± 0.03 ^{f}	3.98 ± 0.74
	200	23.98 ± 0.80 ^{a,b}	−0.54 ± 0.07 ^{h}	−0.02 ± 0.06 ^{g}	4.29 ± 0.64
	220	25.03 ± 0.43 ^{a}	−0.62 ± 0.03 ^{h}	0.41 ± 0.04 ^{e}	4.02 ± 0.27
	240	25.11 ± 0.33 ^{a}	2.85 ± 0.05 ^{d}	1.52 ± 0.14 ^{d}	0.79 ± 0.21

Data values are given as average values ± standard deviations. Data in a column with different superscript letters are significantly different at the $p \leq 0.05$ level. Statistical analysis was separately made for starch- and gelatin-based hydrogels. Note here that for ΔE determination, 140 °C was selected as the reference temperature.

3.3. Rheological Testing

Figure 2 presents the viscoelastic behavior of starch-based systems prepared with Paulownia extracts (i.e., bark, leaves, petioles) recovered using the lowest autohydrolysis temperatures, as representative of the tested systems developed with this biopolymer. In all cases, the elastic modulus (G′) and viscous modulus (G″) remained practically constant with rising frequency. Moreover, elastic modulus was higher than the viscous one over the tested frequencies range. These tendencies indicated a typical gel behavior commonly found in similar biopolymer-based matrices [20,24]. At fixed frequency, the highest magnitudes in viscoelastic moduli were identified for hydrogels enriched with extracts from petioles followed by those incorporated with bark and leaves. This is suggested to be related with the total solid content (g/100 g) present in the different tested Paulownia fractions, which varied from 20 to 33 for petioles, from 25 to 40 for barks, and from 36 to 43 for leaves [17]. Overall, the presence of *Paulownia* extracts allowed the softening of starch-based matrices (around three times) in the absence of hydrothermal aqueous extracts. Even though, hydrogels with intermediate strength were identified in all cases. Above results were consistent with those previously reported for other similar biopolymer-based matrices [20,25,26] and those enriched with natural sources [24]. The latter authors stated that the softening of the hydrogels enriched with bioactive extracts could be explained by the competition by free water between the biopolymer-based matrix and the antioxidant extracts.

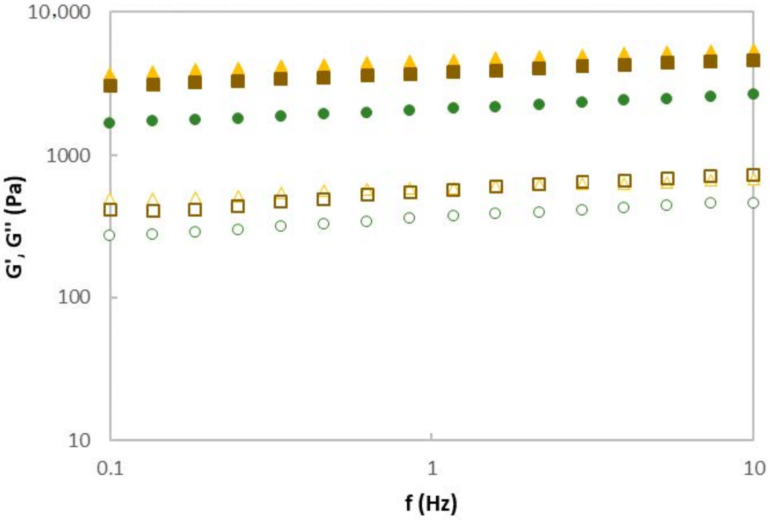


Figure 2. Mechanical spectra of starch-based hydrogels enriched with representative Paulownia extracts treated at the lowest autohydrolysis temperature (140 °C). Symbols: G′ (closed), G″ (open), bark (squares), leaves (circles), petioles (triangles).

Figure 3 shows the influence of representative Paulownia extracts on the mechanical spectra of gelatin-based hydrogels. Again, the monitoring of both elastic and viscous moduli with frequency allowed identifying characteristic gel profiles (G′ > G′′, and almost frequency independent). Similar viscoelastic behavior was observed when compared with their counterparts made with starch, although lower values (around 10 times) of both moduli were identified. Within the gelatin-based hydrogels, the strongest gelling features were found for those formulated with petioles followed by hydrogels in the presence of barks and leaves, as above for starch-based hydrogels. In this case, the presence of Paulownia extracts on the gelatin-based hydrogels involved a softening (around five times) regarding the pure matrix, which suggest to be linked with the water holding capacity. The elastic and viscous values were in good harmony with those previously reported for other gelatin-based gels incorporated with antioxidant extracts from guava leaves [24]. It should be highlighted that in both cases (i.e., starch-based and gelatin-based systems), a consistent trend between softening of the gelling features and phenolic behavior was exhibited, as analyzed in a parallel work [17]. The latter authors found increasing total phenol content with temperature for above aqueous extracts in the range of 1–4.8% for petioles, 2.2–4.2% for bark, and 5.2–7.1% for leaves. Another factor closely associated with the gel strength is the protein content [27], although in this particular case a random tendency between both properties was found except for bark hydrogels. Namely, bark protein content remained practically constant until 180 °C with average values around 195.6 ± 11.2 µg ABS eq/L, decreasing significantly ($p < 0.05$) above this temperature following the trend: 131.2 ± 16.0 µg ABS eq/L at 200 °C, 67.2 ± 3.4 µg ABS eq/L at 220 °C, and 34.6 µg ABS eq/L ± 3.1 at 240 °C. In contrast, an increase in this parameter with rising temperature (up to 200 °C) was previously reported for the corresponding leaves and petioles [17]. In this parallel work, the authors found that the leaves exhibited protein values varying between 65 and 637 µg ABS eq/L, whereas petioles ranged from 45 to 367 µg ABS eq/L. The differences observed could be related to other factors such as the different protein types or sizes, due to a denaturalization of these biopolymers during high autohydrolysis temperatures [28–30].

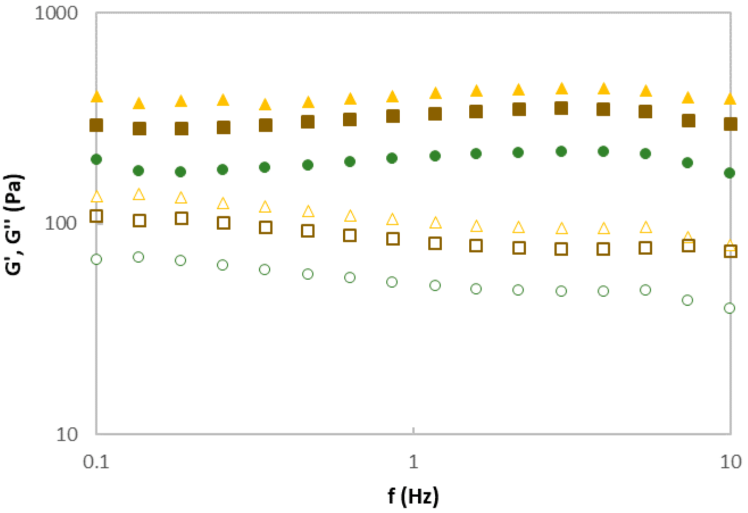
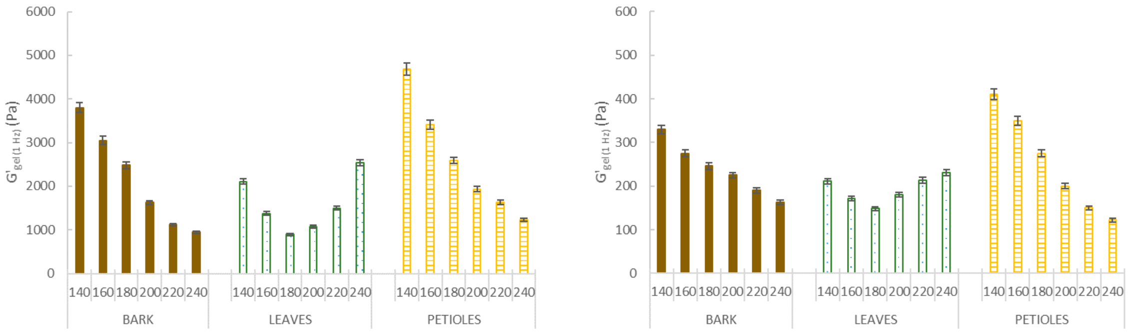


Figure 3. Mechanical spectra of gelatin-based hydrogels supplemented with selected Paulownia extracts processed at the lowest autohydrolysis conditions (140 °C). Symbols: G′ (closed), G′′ (open), bark (squares), leaves (circles), petioles (triangles).

Figure 4 provides an overview of the impact of autohydrolysis temperature during the Paulownia extracts recovery on the mechanical features of the final hydrogels developed. A relevant influence of extracts processing temperature on the viscoelastic features of the developed hydrogels was clearly observed. For bark and petiole extracts, the strength of starch-based and gelatin-based hydrogels dropped with increasing autohydrolysis temperature. For leaves extracts, the softening of the gels was noticed below 180 °C, whereas the reverse trend was identified at the highest temperatures. Although similar trends were detected for both tested matrices (i.e., starch and gelatin), the decay in the gel strength was promoted in starch hydrogels, which is suggested to be related to the highest water holding capacity of the potato starch [20]. This delivers an extensive variety of viscoelastic characteristics for the recovered extracts which enlarge the possible uses in food and nonfood sectors.



(**a**) (**b**)

Figure 4. Impact of the autohydrolysis processing temperature (°C) on the viscoelastic features, G′gel (1 Hz), of all tested hydrogels formulated with (**a**) starch and (**b**) gelatin. Symbols: bark (filled bars), leaves (dotted bars), petioles (dashed bars).

3.4. Textural Features

The main textural properties of all formulated starch-based and gelatin-based hydrogels enriched with Paulownia extracts are collected in Table 3. As expected, starch-based hydrogels incorporated with Paulownia extracts showed statistically ($p < 0.05$) higher firmness (close to four times) and springiness (about three times) values than those prepared with gelatin. The magnitude of these parameters is in the range of those previously reported for other starch-based [20] and gelatin-based [2,15] hydrogels. In both cases, similar parameters tendencies were identified with increasing autohydrolysis temperature. Namely, firmness, springiness, and cohesiveness decreased with increasing temperature for hydrogels enriched with bark and petioles extracts; whereas the above parameters decreased until 180 °C, increasing above 200 °C for leaf extracts. No significant differences ($p < 0.05$) were observed in adhesiveness values independently of extract type or temperature within each tested gelling matrix. Note here that gelatin-based hydrogels seem to tend to higher values of this parameter. In general, texture results of formulated hydrogels related to the macroscopic structure agreed with those aforementioned for the mechanical spectra linked to the microscopic structure. Again, the presence of hydrothermal aqueous extracts from Paulownia in the hydrogels led to a weakening of the gel's texture features. This behavior nicely matches with the outcomes found for other hydrogels formulated in the presence of natural antioxidant compounds [2,30]. It should be remarked that no syneresis was observed for any of the developed hydrogels after two weeks cold storage (4 °C).

Table 3. Texture parameters for hydrogels prepared with potato starch (**a**) and gelatin (**b**) enriched with Paulownia extracts.

(**a**) **Starch Hydrogels**

	T ^{a} (°C)	Firmness (kgf)	Adhesiveness (kgf s)	Cohesiveness (-)	Springiness (-)
Bark	140	6.2 ± 0.4 ^{b}	0.11 ± 0.03 ^{a}	0.18 ± 0.01 ^{b}	0.59 ± 0.02 ^{b}
	160	5.3 ± 0.3 ^{c}	0.10 ± 0.04 ^{a}	0.14 ± 0.01 ^{c}	0.55 ± 0.01 ^{c}
	180	4.3 ± 0.4 ^{e}	0.12 ± 0.02 ^{a}	0.11 ± 0.01 ^{e}	0.50 ± 0.02 ^{e}
	200	3.6 ± 0.1 ^{f}	0.13 ± 0.03 ^{a}	0.09 ± 0.00 ^{f}	0.43 ± 0.01 ^{f}
	220	2.8 ± 0.2 ^{g}	0.12 ± 0.05 ^{a}	0.07 ± 0.00 ^{g}	0.39 ± 0.01 ^{g}
	240	2.4 ± 0.4 ^{g}	0.13 ± 0.03 ^{a}	0.06 ± 0.01 ^{g}	0.38 ± 0.02 ^{g}
Leaves	140	4.1 ± 0.2 ^{e}	0.10 ± 0.02 ^{a}	0.11 ± 0.01 ^{e}	0.51 ± 0.02 ^{e}
	160	3.5 ± 0.2 ^{f}	0.11 ± 0.04 ^{a}	0.08 ± 0.00 ^{f}	0.44 ± 0.01 ^{f}
	180	2.9 ± 0.3 ^{g}	0.13 ± 0.04 ^{a}	0.06 ± 0.01 ^{g}	0.38 ± 0.01 ^{g}
	200	3.2 ± 0.2 ^{g}	0.10 ± 0.03 ^{a}	0.06 ± 0.01 ^{g}	0.37 ± 0.01 ^{g}
	220	3.8 ± 0.1 ^{f}	0.11 ± 0.02 ^{a}	0.09 ± 0.00 ^{f}	0.44 ± 0.01 ^{f}
	240	4.3 ± 0.3 ^{e}	0.10 ± 0.05 ^{a}	0.11 ± 0.01 ^{e}	0.49 ± 0.02 ^{e}
Petioles	140	8.3 ± 0.3 ^{a}	0.10 ± 0.03 ^{a}	0.22 ± 0.01 ^{a}	0.62 ± 0.01 ^{a}
	160	6.2 ± 0.2 ^{b}	0.10 ± 0.02 ^{a}	0.19 ± 0.01 ^{b}	0.59 ± 0.01 ^{b}
	180	5.5 ± 0.3 ^{c}	0.11 ± 0.04 ^{a}	0.15 ± 0.02 ^{c}	0.54 ± 0.01 ^{c}
	200	4.9 ± 0.2 ^{d}	0.12 ± 0.06 ^{a}	0.12 ± 0.00 ^{d}	0.50 ± 0.01 ^{d}
	220	4.0 ± 0.3 ^{e}	0.12 ± 0.03 ^{a}	0.10 ± 0.00 ^{e}	0.46 ± 0.02 ^{e}
	240	3.5 ± 0.1 ^{f}	0.13 ± 0.04 ^{a}	0.09 ± 0.00 ^{f}	0.41 ± 0.02 ^{f}

(**b**) **Gelatin hydrogels**

	T ^{a} (°C)	Firmness (kgf)	Adhesiveness (kgf s)	Cohesiveness (-)	Springiness (-)
Bark	140	1.7 ± 0.2 ^{b}	0.13 ± 0.04 ^{a}	0.28 ± 0.01 ^{b}	0.23 ± 0.01 ^{b}
	160	1.3 ± 0.1 ^{c}	0.12 ± 0.04 ^{a}	0.24 ± 0.00 ^{c}	0.19 ± 0.01 ^{c}
	180	1.0 ± 0.1 ^{e}	0.14 ± 0.05 ^{a}	0.18 ± 0.01 ^{e}	0.14 ± 0.01 ^{e}
	200	0.7 ± 0.1 ^{f}	0.14 ± 0.04 ^{a}	0.15 ± 0.01 ^{f}	0.10 ± 0.01 ^{f}
	220	0.4 ± 0.1 ^{g}	0.15 ± 0.03 ^{a}	0.12 ± 0.01 ^{g}	0.07 ± 0.01 ^{g}
	240	0.4 ± 0.1 ^{g}	0.15 ± 0.02 ^{a}	0.11 ± 0.02 ^{g}	0.07 ± 0.01 ^{g}
Leaves	140	1.1 ± 0.1 ^{e}	0.12 ± 0.03 ^{a}	0.19 ± 0.01 ^{e}	0.15 ± 0.01 ^{e}
	160	0.8 ± 0.1 ^{f}	0.13 ± 0.05 ^{a}	0.16 ± 0.01 ^{f}	0.10 ± 0.01 ^{f}
	180	0.4 ± 0.0 ^{g}	0.14 ± 0.04 ^{a}	0.12 ± 0.01 ^{g}	0.06 ± 0.01 ^{g}
	200	0.4 ± 0.0 ^{g}	0.11 ± 0.02 ^{a}	0.11 ± 0.01 ^{g}	0.06 ± 0.01 ^{g}
	220	0.7 ± 0.1 ^{f}	0.12 ± 0.04 ^{a}	0.16 ± 0.01 ^{f}	0.11 ± 0.01 ^{f}
	240	1.1 ± 0.1 ^{e}	0.12 ± 0.04 ^{a}	0.19 ± 0.01 ^{e}	0.15 ± 0.01 ^{e}
Petioles	140	2.1 ± 0.2 ^{a}	0.13 ± 0.04 ^{a}	0.33 ± 0.02 ^{a}	0.25 ± 0.01 ^{a}
	160	1.7 ± 0.1 ^{b}	0.12 ± 0.05 ^{a}	0.29 ± 0.02 ^{b}	0.22 ± 0.01 ^{b}
	180	1.4 ± 0.1 ^{c}	0.13 ± 0.06 ^{a}	0.25 ± 0.01 ^{c}	0.19 ± 0.01 ^{c}
	200	1.1 ± 0.1 ^{d}	0.15 ± 0.02 ^{a}	0.22 ± 0.01 ^{d}	0.16 ± 0.01 ^{d}
	220	0.8 ± 0.1 ^{e}	0.16 ± 0.02 ^{a}	0.19 ± 0.01 ^{e}	0.13 ± 0.01 ^{e}
	240	0.5 ± 0.0 ^{f}	0.16 ± 0.03 ^{a}	0.15 ± 0.01 ^{f}	0.10 ± 0.01 ^{f}

Data values are given as average values ± standard deviations. Data in a column with different superscript letters are significantly different at the $p \leq 0.05$ level. Statistical analysis was separately made for starch and gelatin hydrogels.

4. Conclusions

To conclude, it can be indicated that studied starch-based and gelatin-based hydrogels with a wide range of mechanical features can be successfully developed using different hydrothermal aqueous extracts from Paulownia. The autohydrolysis temperature during the Paulownia extracts recovery had a critically relevant impact on the color, viscoelastic and textural properties of the corresponding biopolymer-based hydrogels. The formulated hydrogels exhibit a number of advantages from the industrial point of view, not only for their weak or intermediate strength adequate for target food and nonfood applications, but also for the absence of syneresis. Finally, the assessment of above hydrogels allows the revalorization of underused natural sources in a green and sustainable way that could be expanded to other bioactive raw materials.

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