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Concentration as a Factor of the Gelatin-Based Hydrogel Mechanical Properties from Gel-Like to Solid-Like Behavior

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Abstract

Hydrogels are soft materials that exhibit mechanical properties between liquids and solids, making them attractive for applications in biomedical engineering, pharmaceutics, and food science. In this work, we studied how the mechanical properties of gelatin-based hydrogels depend on concentration, focusing on the transition from a soft solid with a gel-like behavior to an elastic solid. We prepared hydrated gelatin samples by dissolving determined mass quantities of dry bovine gelatin powder in hot deionized water and allowing them to solidify under reproducible thermal conditions. Compression tests were performed using a custom mechanical press to determine the Young's modulus and Poisson's ratio, while torsion pendulum experiments were used to obtain the shear modulus. The results show that concentration plays a critical role in the rigidity and deformation response of the material. For samples with concentrations below approximately 0.075 g/ml, the Young's modulus and shear modulus remain very low, and the Poisson's ratio exceeds 0.5, indicating a soft solid behavior dominated by gel-like behavior. In contrast, for concentrations above this threshold, both elastic moduli increase significantly, while Poisson's ratio gets below 0.5, showing the emergence of an elastic solid network capable of storing mechanical energy. This identifies a clear mechanical transition governed by gelatin concentration, even under constant temperature conditions (24.1˚C). The three mechanical properties exhibit consistent trends, confirming that the microstructure of the gelatin network evolves with concentration: weak and sparsely connected chains at low concentrations, and a strongly cross-linked structure at higher concentrations. The identification of critical concentration provides insight into how gelatin hydrogels can be adjusted to achieve desired stiffness, deformability, and stability. These findings contribute to the understanding of soft organic matter and offer a quantitative basis for producing gelatin systems in practical appli-

cations. Furthermore, the results are relevant for future research on phononic crystals made from hydrogels, since their elastic properties directly influence mechanical wave propagation and the resulting band structures.

Keywords

Soft Matter, Hydrogel, Gelatin, Young's Modulus, Poisson's Ratio, Shear Modulus

1. Introduction

Mechanical characterization of soft matter has an intrinsic relevance, due to its multiple applications in biomedical [1], pharmaceutical and food engineering [2] [3]. Additionally, soft materials have been considered for applications in phononic crystals [4]. Understanding its behavior under mechanical and thermal external agents is essential to set up limits of these applications [5]. Soft matter exhibits properties between liquids and solids; this characteristic allows us to study how the mechanical properties of a hydrogel such as the gelatin changes as a function of the gelatin concentration, from a quasi-liquid at low concentrations to a quasi-solid at high concentrations. The mechanical properties change from soft solid with significant viscous behavior to elastic solid with low energy dissipation.

The dry basis of gelatin is the result of a controlled hydrolysis process of animal tissues rich in collagen such as skin, tendon and bones. This dry basis contains up to 99% of protein, at chemical level, the gelatin is a natural polymer formed by single, double and triple covalently crosslinked chains [6].

The aqueous solution is prepared by dissolving a known mass m of dry gelatin in a determined volume of hot water ($V = 120$ ml). For concentration larger than ($c \approx 2\% m/V$) the gelatin has a first order thermo-reversible transition at the gelation temperature ($T_g \approx 30^{\circ}\text{C}$) [7]. This gelation temperature T_g and the viscoelastic properties of the hydrated gelatin also depend on concentration.

In this paper, we study the mechanical behavior of hydrated gelatin as function of concentration, that is the ratio of dry gelatin mass to a certain volume of hot water. The dry basis of gelatin selected was bovine origin and the gelatin samples were in solid phase at the laboratory temperature (24.1˚C). We present the experimental procedure about hydrogel preparation and the methodology to determine the Young's and shear modulus, and the Poisson's ratio at different concentrations of gelatin. In discussion and analysis of results section, we show and analyze our results and compare them with other ones reported in the literature.

2. Experimental Method and Results

2.1. Gelatins

We focus on a particular type of hydrogel obtained from a reversible physical process of dissolution of dry gelatin of bovine origin in hot deionized water. The wa-

ter was obtained from a pressure container, and the quantity of air bubbles was reduced at atmospheric pressure and at room temperature. All mixtures were prepared with different masses ($m \pm 0.001$ mg) of dry gelatin and a constant mass of water ($M = 120 \pm 0.001$ mg) at 92˚C of temperature (The boiling point in Mexico City). The water evaporation was avoided by covering the container. All homogeneous mixtures were refrigerated at 2.0˚C.

To determine the concentration $c[\mathrm{g/ml}]$, we considered that the density of the water is $\rho \approx 1.0\,\mathrm{g/ml}$, in the temperature range in which the dissolutions were prepared. Figure 1 and Figure 2 show the containers of the hydrogels at different concentrations in the liquid and the solid phase, respectively, from the lower concentration, at the left, to the higher concentration, at the right. The data reported in this work were obtained from multiple trials that we carried out under same conditions, which increases reproducibility of our experiments.

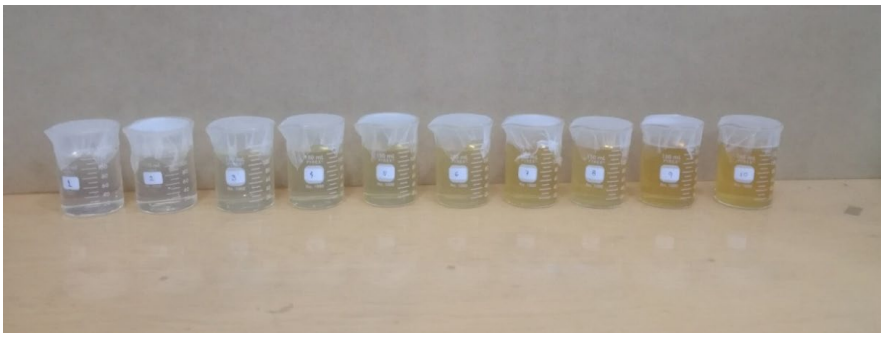

Figure 1. Liquid water-gelatin mixtures at different concentrations of dry gelatin, from lower to higher from left to right.

Figure 2. Solid water-gelatin mixtures at different concentrations of dry gelatin, from lower to higher from left to right.

2.2. Young's Modulus and Poisson's Ratio Measurement

Hydrogel samples were removed from the mold at 2.0˚C. The compression measurements were made at laboratory temperature (24.1˚C), using a press designed in our laboratory. Figure 3 shows a gelatin solid sample under compression by means of the press. Figure 4 shows the parts of the press that consisted of a steel rod, a spring, and a micrometer screw. The compression of the spring was engaged

to the advance of the screw in the press. To quantify the force, the parameters of the press components were calibrated through compression trials in the laboratory. The advance of the screw indicated the compression of the spring, and therefore, the force F that was applied.

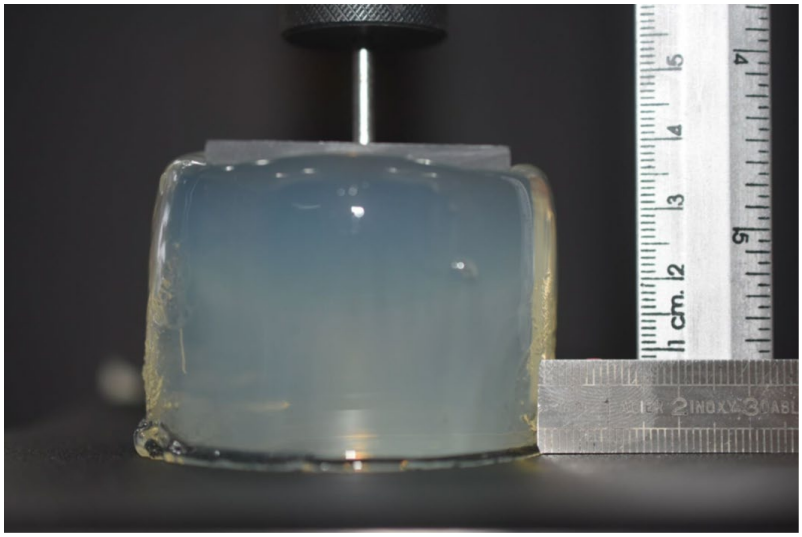

Figure 3. Gelatin solid sample under compression by means of a mechanical press.

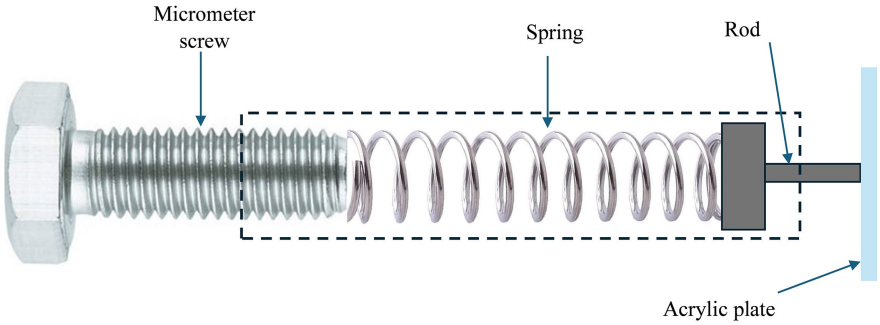

Figure 4. Diagram of the essential components of the press: a steel rod, a spring, and a micrometer screw.

As the elastic behavior of the samples is undetermined, we must be careful with the Young's modulus definition, since the load may not be evenly distributed in the sample, therefore we use the real values of the axial stress and strain instead of being nominal values.

To determine the Young's modulus $Y = \sigma/\varepsilon$, we use the definition of ratio between the real axial stress $\sigma = F/A$, and the real axial strain $\varepsilon = \Delta L/L_0$; where A is the area of the transversal middle section of the sample during compression and L_0 is the axial length of the sample, measured in the middle axis of the sample. Within the range of stress exerted on each of the samples with different concentration, we find a linear relationship between stress and strain. The graph of real axial stress against real axial strain for a concentration $c = 0.091\,\mathrm{g/ml}$ is shown in **Figure 5**. The correlation coefficient of the lineal fit is $R = 0.9976$.

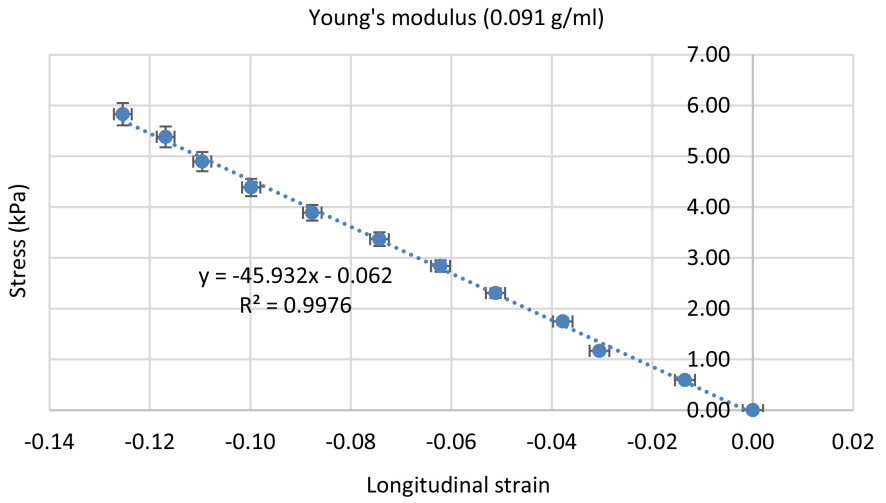

Figure 5. Real axial stress against real axial strain for a sample with $c = 0.091$ g/ml. The Young's modulus is the absolute value of the slope of the line.

The stress trails were made with gelatin samples with different concentrations under the same experimental conditions. The dependence of the Young's modulus with the concentration is shown in **Figure 6**.

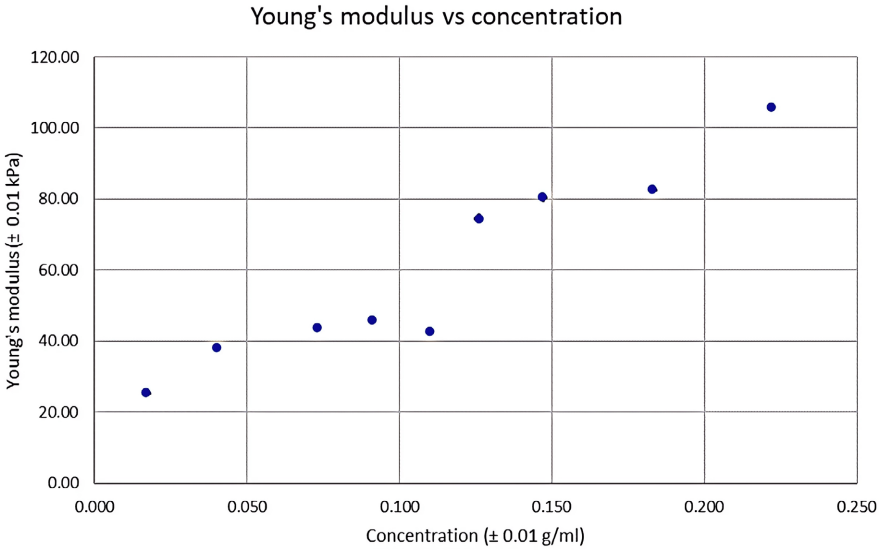

Figure 6. Graph of the Young's modulus determined for each gelatin sample against the respective concentration. The experiments were performed at 24.1˚C.

The Poisson's ratio ν was determined in the same compression experiments, defined as the negative of the ratio of transversal strain ε_x to axial strain ε_y. The length transversal was measured as the diameter of the cross section at mid height of the sample. **Figure 7** shows the transversal strain against the axial strain obtained for the sample with concentration $c = 0.091\,\text{g/ml}$. The interrupted line corresponds to the linear fit with a correlation coefficient $R^2 = 0.9949$, the negative value of the slope is the Poisson's ratio of this gelatin.

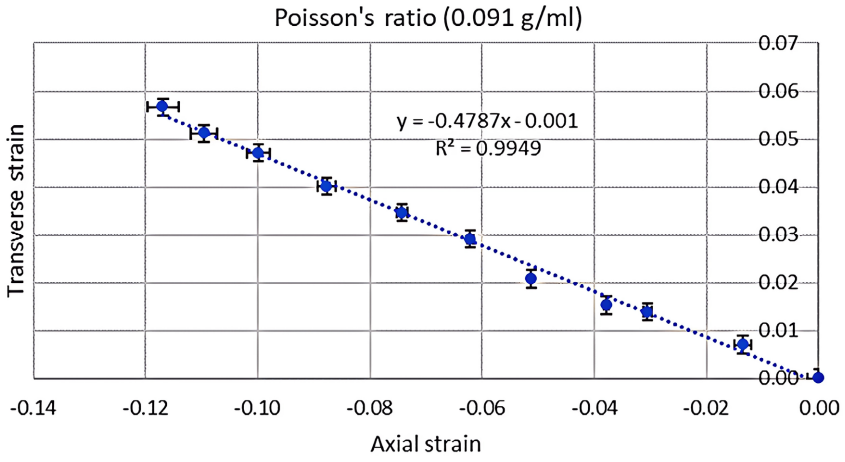

Figure 7. Transversal strain against axial strain for a sample with $c = 0.091$ g/ml. The points correspond to experimental data and the interrupted line to the linear fit. The experiment was performed at 24.1˚C. The negative of the slope is the Poisson's ratio.

The Poisson's ratio determined for samples with different concentrations against the corresponding concentration is shown in **Figure 8**. All the experiments were carried out under the same conditions.

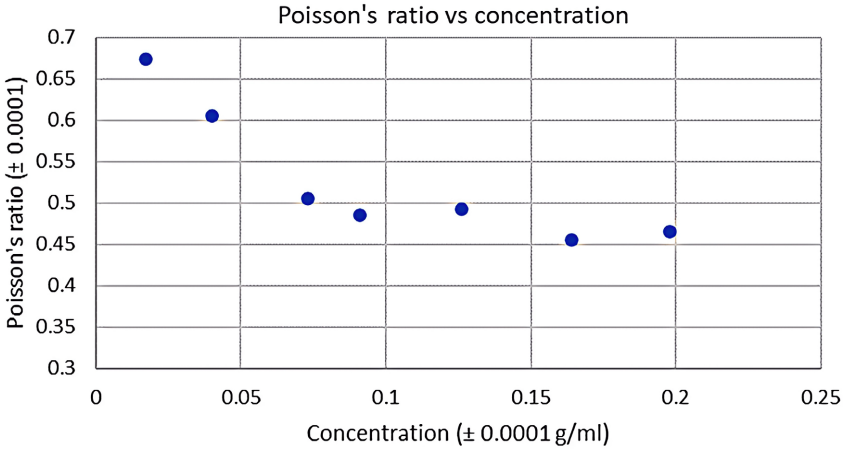

Figure 8. Poisson's ratio for gelatin samples with different concentrations against the respective concentration. The experiments were performed at 24.1˚C.

2.3. Shear Modulus

To determine the shear modulus of a particular sample of gelatin, we use a torsional oscillation bar, where the oscillation system consists of a gelatin bar and an acrylic disk with a mass $m_d = 212.26$ g and a diameter $D_d = 19.5$ cm. This lightweight material with a diameter larger than that of the gelatin bar was selected with the purpose of not deforming the gelatin with its weight, in special samples with low concentrations. **Figure 9** shows a diagram of the torsional oscillation bar at the left, and a top view of the device at the right, with a gelatin sample with $c = 4.7$ g/ml. In this image the acrylic disk is rotated with respect to its equilibrium position indicated by the red interrupted lines.

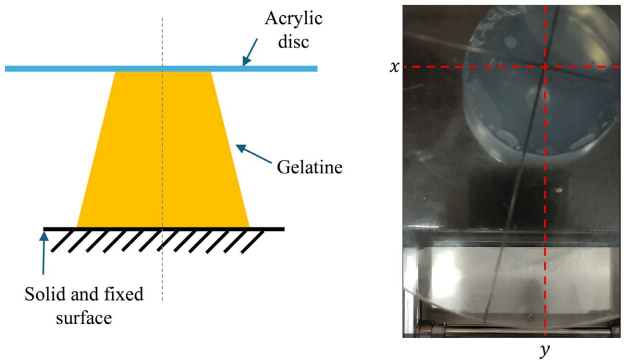

Figure 9. Diagram of the torsional oscillation bar at the left, and a top view of the device at the right, with a gelatin sample with $c = 4.7$ g/ml. On the left, the lower surface of the sample is adhered to a fixed solid surface, and the top surface is adhered to the oscillating acrylic disc. In this figure the shape of the sample has been exaggerated. On the right, the top image of the acrylic disk is rotated with respect to its equilibrium position indicated by the red interrupted lines.

In this experiment we use a truncated cone-shaped sample as a torsion bar. The oscillation of the acrylic disk produced by the torque T generates torsion in the bar. The volume of the bar can be divided into layers of differential volume elements with thickness $\mathrm{d}r$ and height $H/\cos\alpha$, as shown in Figure 10. A differential force $\mathrm{d}F$ uniformly distributed acts over the area differential $\mathrm{d}A$ of the concentric rings with radius r and thickness $\mathrm{d}r$ on the upper surface of each differential volume element, see Figure 10. Therefore, the shear stress τ over the differential volume element is given by

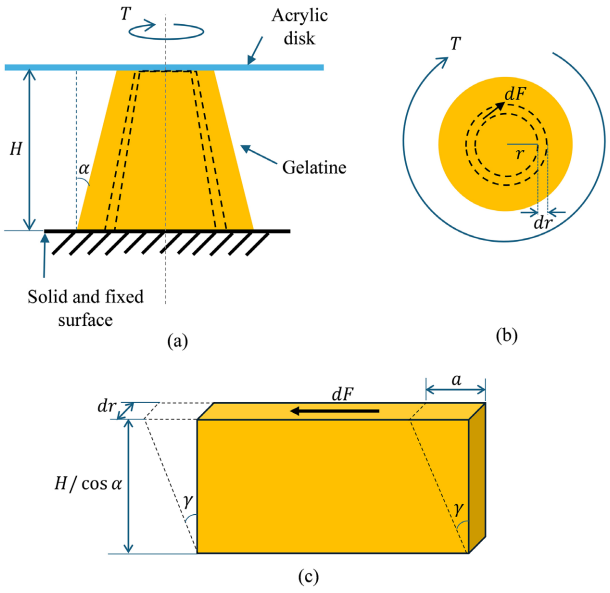

Figure 10. Diagram of the torsion bar. (a) Side view, the dashed line represents a differential volume element of the truncated cone. (b) The top view of the sample, the differential area element $\mathrm{d}A$ is represented with the dashed ring with radius r and thickness $\mathrm{d}r$. (c) The force $\mathrm{d}F$ tangent to the surface $\mathrm{d}A$ generates a shear strain γ on the differential volume element shown in (a), this layer has a thickness $\mathrm{d}r$ and a height $H/\cos\alpha$.

$$\tau = \frac{\mathrm{d}F}{\mathrm{d}A} = \mu\gamma \tag{1}$$

where μ is the shear modulus of the torsion bar (gelatin) and γ is the shear strain of differential volume element.

The displacement a between the upper and lower surfaces of the differential volume element for small shear strain γ is given by

$$a \cong \frac{\gamma H}{\cos\alpha} \tag{2}$$

While the displacement a along the ring with radius r can be written as

$$a = r\varphi \tag{3}$$

where φ is the disk rotation angle, it is the same torsion angle of the upper extreme with respect to the lower extreme of the torsion bar (gelatin).

For small oscillation angles, from Equations (1), (2) and (3) we obtain

$$\mathrm{d}F \cong \mu\frac{r\varphi\cos\alpha}{H}\mathrm{d}A = 2\pi\mu\frac{\varphi\cos\alpha}{H}r^2\mathrm{d}r \tag{4}$$

Therefore, the torque on the differential volume element is

$$\mathrm{d}T = r\mathrm{d}F = 2\pi\mu\frac{\varphi\cos\alpha}{H}r^3\mathrm{d}r \tag{5}$$

The total torque T on torsion bar is obtained by integration Equation (5) from $r=0$ to R_s, with R_s the radius of the upper surface of the truncated cone. So, the total torque is given by

$$T = \frac{\mu\pi\varphi\cos\alpha R_s^4}{2H} \tag{6}$$

For small oscillations, $T = \kappa\varphi$, where κ is the torsion constant of the torsion bar, and the period of oscillation P is given by:

$$P = 2\pi\sqrt{\frac{I}{\kappa}} \tag{7}$$

with $I = I_D + I_G$, where I_D and I_G are the inertia moments of the acrylic disk and the bar torsion (gelatin) respect to the rotation axis, respectively. Therefore, the gelatin shear modulus μ is calculated from the oscillation period P as

$$\mu = \frac{8\pi H\left(I_D + I_G\right)}{\cos\alpha R_s^4 P^2} \tag{8}$$

In our experiments with torsion pendulums, initial torsion angles were less than 15˚ to ensure small oscillations. The oscillation movement of each torsion pendulum was recorded with a fixed camera at 60 frames per second. The video was analyzed via Tracker Video Analysis and Modelling Tool software [8], and we obtained the relative amplitude of oscillation in function of the time. **Figure 11** shows the plot obtained from the sample with a concentration $c = 4.7\ \mathrm{g/ml}$, which had an oscillation period $P = 0.43\ \mathrm{s}$.

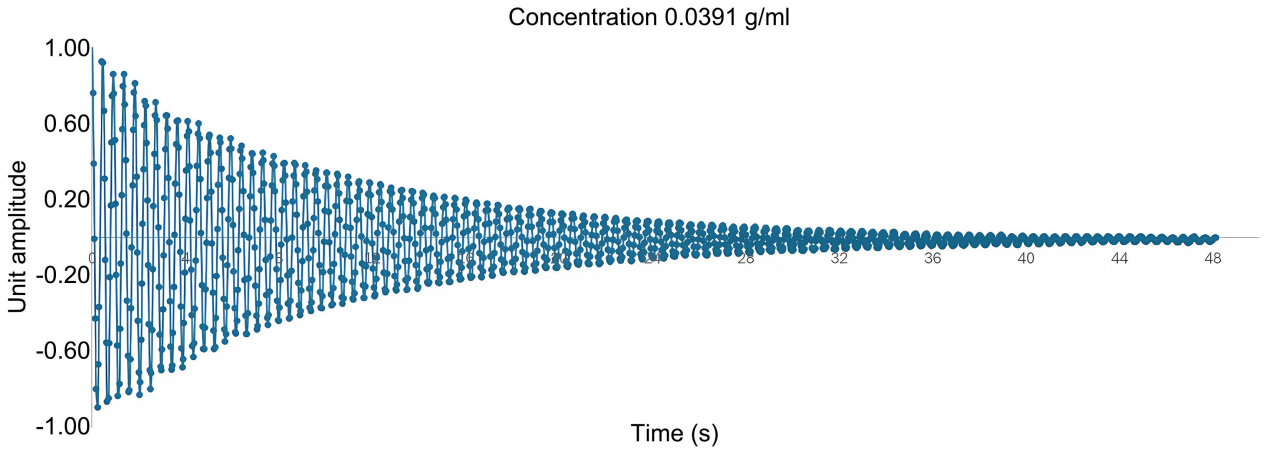

Figure 11. Plot of the amplitude oscillation (normalized to 1) versus the time. Obtained for a torsion pendulum with a torsion bar of gelatin with a concentration 4.7 g/ml at 24.1˚C.

From the data obtained with the Tracker software we obtained the oscillation period for each sample with different concentation and from Equation (8) the respective shear modulus μ. **Figure 12** shows the shear modulus versus the concentration.

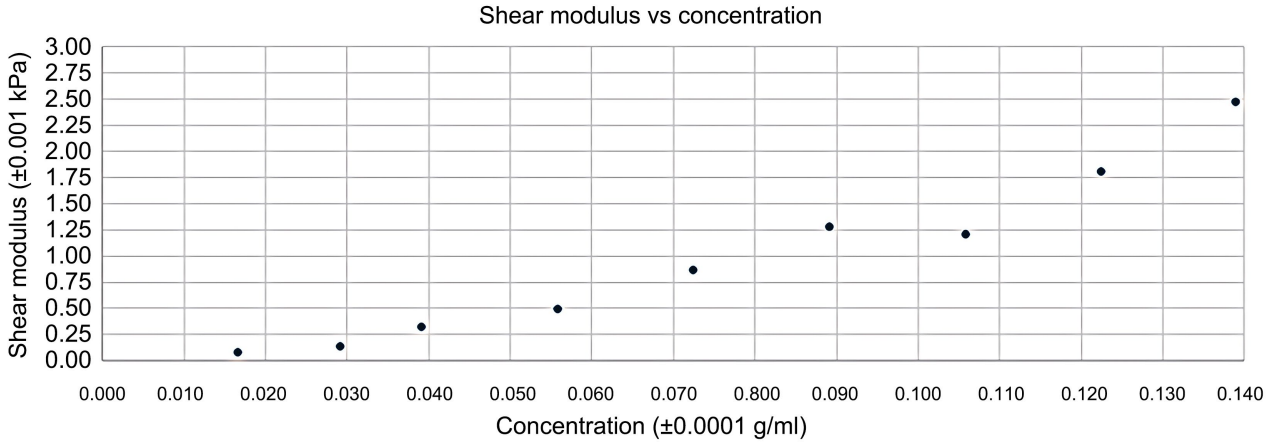

Figure 12. Shear modulus for the gelatin samples with different concentrations against the respective concentration. The experiments were performed at 24.1˚C.

3. Discussion and Analysis of Results

Our experimental results for Young's modulus and shear modulus for bovine gelatin in deionized water as a function of concentration are comparable in magnitude to those reported by Sand [9] for porcine gelatin in water, as well as by Chen *et al.* [10] for GelMA30 hydrogel. Our results for Poisson's ratio against concentration are comparable to those obtained by Cappello *et al.* [11] for polyethylene glycol (PEG) hydrogels showing that Poisson's ratio of their samples varies over a wide range of values (0.165 to 0.5).

It is well known that Poisson's ratio of a homogeneous and isotropic solid has an upper limit of 0.5, which is obtained by an analysis of the compressive stresses

and the isothermal compressibility coefficient, which corresponds to this value in its liquid phase. Materials formed by mixtures present phase transitions that depend on pressure and temperature, as well as on the composition [12], making the phase transition process more complex. Furthermore, soft matter has the characteristic of presenting an intermediate phase between the solid and liquid phases, in which the properties of both phases combine to give behaviors between solid and liquid. On the other hand, rheological and microstructural studies of hydrogel fluids, using measurements of physical properties such as viscosity, demonstrate that the viscoelastic properties of the microgel particles are affected by the microstructure of the gel particles because of varying concentrations in hydrogels. It is shown that the storage and loss modulus increases with increasing concentration [13].

Poisson's ratio greater than 0.5 in soft solid is a result of its viscoelastic behavior is the dynamic response of non-equilibrium thermodynamic states, where energy dissipation and relaxation times allow values of Poisson's ratio forbidden by theory of elasticity. Recently, a unifying van der Waals-like model has been proposed to describe the dynamic solid-liquid transition of these materials [14].

Analyzing our results, we found that Poisson's ratio of the gelatin samples, which are a mixture of deionized water and dry mass of gelatin, presents Poisson's ratios greater than 0.5, and the samples can be separated into two regions of mechanical behavior determined by the dry mass of gelatin concentration values, as shown in **Figure 13**. The first region corresponds to Poisson's ratio values greater than 0.5 and concentration values less than 0.075 g/ml. The dashed line fits this data in the figure. Meanwhile, the second region corresponds to Poisson's ratio values less than 0.5 and concentration values greater than 0.075 g/ml, and these data are fit by dotted line in the same figure.

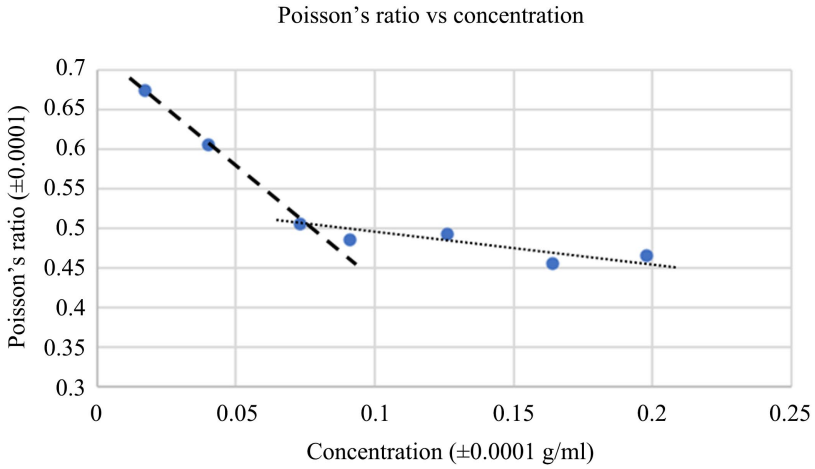

Figure 13. Regions of mechanical behavior. Points on the dashed line exhibit soft solid behavior, and points on the dotted line exhibit elastic solid behavior.

Furthermore, our results indicate that for samples with concentrations lower than 0.075 g/ml the Young's moduli are less than 45 kPa, while samples with con-

centrations between 0.075 and 0.125 g/ml have a Young's modulus has small changes, and for samples with higher concentrations, the Young's modulus increased significantly, see **Figure 6**. Similarly, our results indicate that the shear moduli values of the samples have a similar behavior, that is, for concentrations lower than 0.075 g/ml, the shear moduli are less than 1.0 kPa, while for concentrations between 0.075 g/ml and 0.125 g/ml, the shear modulus has small changes, and for higher concentrations, the shear modulus increased significantly.

Therefore, our results indicate that the concentration of 0.075 g/ml is a value that marks a change in the mechanical behavior of the gelatin samples at a temperature of 24.1˚C, from an elastic solid behavior for concentrations greater than 0.075 g/ml to a soft solid behavior for concentrations between 0.016 g/ml and 0.075 g/ml at a temperature of 24.1˚C. This behavior is consistent with our experimental procedure the gelatin samples at concentrations of 0.016 g/ml solidify, but at a temperature of 24.1˚C they turned unstable mechanically and became a gel.

The three mechanical properties measured exhibit consistent trends, confirming that the microstructure of the gelatin network evolves with concentration: weak and sparsely connected chains at low concentrations, and a strongly cross-linked structure at higher concentrations. The identification of critical concentration provides insight into how gelatin hydrogels can be adjusted to achieve desired stiffness, deformability, and stability.

Our results are relevant to understanding the soft organic materials and their applications in areas such as biomedical, pharmaceutics and food engineering. In particular, the results presented in this research will be very important for continuing our research on phononic crystals formed with gelatin samples of different concentrations, because the allowed bands of mechanical waves depend on the propagation velocity, which depends on the mechanical moduli of the material.

4. Conclusions

In this work, we have experimentally found that gelatin hydrogels undergo a clear transition in their mechanical response as a function of concentration, evolving from gel-like behavior at low concentrations to an elastic solid-like behavior at higher concentrations. By independently determining the Young's modulus, the Poisson's ratio, and the shear modulus for each sample, we identified a critical concentration of 0.075 g/ml at which the mechanical properties change significantly at temperature of 24.1˚C. Below this threshold, the hydrogels exhibit low rigidity and Poisson's ratios greater than 0.5, consistent with a soft solid dominated by gel-like characteristics. Above this concentration, both elastic moduli increase significantly, and Poisson's ratio drops below 0.5, indicating a mechanically stable elastic network.

Our findings confirm that the mechanical behavior of physical gelatin gels is highly sensitive to concentration, and that the transition from soft to elastic solid behavior is continuous but well defined. These results provide a quantitative basis for adjusting the mechanical response of gelatin-based hydrogels for practical ap-

plications. Because gelatin is widely used in biomedical engineering, pharmaceuticals, and food science, and the present characterization contributes to improving the design of hydrogel-based systems, especially in applications requiring precise control of stiffness and deformability. Moreover, the mechanical trends reported here will be particularly useful for future studies on phononic crystals and wave propagation in structured soft matter, where elastic moduli directly determine the acoustic band structure.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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