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Mechanical characterization of gelatinbased hydrogels upon finite strains in tension, compression and shear

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Introduction

Hydrogels are attractive biomaterials used in a wide range of biomedical applications, such as scaffolds for cell culture in tissue engineering, biomimetic models for human tissues, and carriers in drug delivery. Hydrogels consist of hydrophilic polymeric 3D networks, able to retain high tissue-like water content without dissolution. Among the biopolymers used to process hydrogels, gelatin is very common due to its cost-effectiveness, ease of manufacturing and ability to form thermo-reversible gels [1,2]. The poor mechanical performances of standard gelatin-based hydrogels (e.g. low elastic modulus, low ultimate elongation, brittle failure) are known to be tunable and enhanced with additional physical and/or chemical crosslinking in the gels [2,3].

A proper and complete characterization of the mechanical behavior of these hydrogels is critical to evaluate the relevance of their utility for biomedical applications. However, corresponding experimental studies are often limited either to one specific loading mode (mainly compression or shear) with a single quasi-static monotonic path up to failure, or to standard infinitesimal strain DMA analyses [2-4]. These configurations are usually far from those endured *in vivo* by native tissues which are subjected to numerous complex and coupled mechanical loading upon finite strains and various strain rates. In this work, we propose a procedure to characterize and to highlight the role of the finite strain mechanical loading (tensions, compressions and shear) on the mechanical behavior of gelatin-based hydrogels.

Experimental Methods

Dry porcine gelatin (gel strength ≈ 300 g Bloom, Type A, Sigma-Aldrich®) was used to produce a series of hydrogels, mixing the powder in ultrapure water (18.2 M Ω) for 30 min at 45°C. The prepared aqueous solutions were casted into a Teflon® mold at room temperature for 1 h, and kept at 3 °C for 24 h to form a rectangular gel plate $(100 \times 100 \times 5 \text{ mm}^3)$. Two parameters were varied during the processing route: (i) the gelatin concentration in the aqueous solutions, i.e., 5, 10 and 15 %w/v [1,3]; (ii) the use of a crosslinking reagent (0.5 wt% of glutaraldehyde (GA), Sigma-Aldrich®). Samples were cut from the plates at a length-to-width ratio ranging from 5:1 to 1:5, and coated with a random pattern made of small speckles. Mechanical tests were carried out using a standard uniaxial machine (Instron® 5944) equipped with a ±10 N load cell. Five loading modes were studied: simple tension, plain strain tension (pure shear), simple and plain strain compressions and simple shear [5]. Whatever the loading mode, samples were subjected to cyclic loadings with increasing strain amplitude up to the ultimate elongation. Pictures of the deformed sample in frontal and lateral views were recorded using two high-resolution CCD cameras (JAI® BM-500GE, 5 MPixels), to quantify its dimensional changes and to allow digital image correlation for the measurement of local strain field (7D software [6]). For each loading case, two parameters were varied during the protocol: (i) the strain rates, ranging from 10^{-3} s⁻¹ to 10^{1} s⁻¹; (ii) the surrounding hygrometric conditions, regulated at ambient or saturated (RH 100%) atmosphere.

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Results and Discussion

To illustrate some of our results, we have plotted in Figure 1 stress-strain curves showing the typical cyclic mechanical behavior at a cross head speed of 0.05 mm.s⁻¹ of the processed hydrogels in simple tension and compression (here with gelatin 10% w/v, no chemical crosslinking). The figure clearly emphasizes the impact of the mechanical loading on the behavior of the hydrogels: weak hysteresis and weak residual strain in tension, these two features being pronounced in compression where, in addition, stress levels being much higher than those recorded in tension. In addition, the figure highlights the strong impact of the sample air-drying on stress-strain curves: the higher the resting time, the higher the stress levels.

Conclusion

The results of this work will help to clarify the mechanical protocols and specially the loading modes to be considered for the biomimetic design of new homogeneous materials. This knowledge is expected prior to develop any suitable fibre-reinforced biocomposites, able to mimic the histological and biomechanical features of native tissues.

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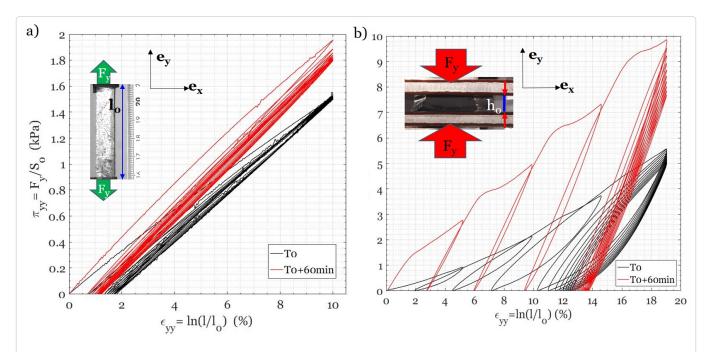


Fig1. Cyclic mechanical behavior of a physically crosslinked gelatin-based hydrogel

Typical evolutions of nominal stresses as function of Hencky strains in tension (left), in compression (right)

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