Engineering viscoelasticity in biomaterials

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***Abstract*— Biomaterials have an intrinsic viscoelastic behaviour. However, their elasticity is actually the most studied aspect of these materials and different techniques to modulate stiffening have been developed. In order to address the viscous component, agarose gels in dextran solutions with different concentrations were tested using the epsilon-dot method. Our results show that varying the viscosity of the liquid phase influences the mechanical behaviour of the gels and also enables modulation of material viscoelasticity.**

***Keywords*—viscoelasticity, biomaterials, epsilon-dot.**

I. Introduction

Tissues and biomaterials are characterised by a biphasic structure: a solid network, responsible for elastic properties, surrounded by an aqueous solution, related with the viscous properties [1].

Despite the intrinsic viscoelasticity of these materials, most of the studies in the literature are focused on the control and modulation of the elastic behaviour using different stiffening techniques (i.e. physical, chemical, enzymatic) [1]. The few works in the literature on the modulation of viscoelasticity are based on rheological measurements. The resultant dynamic modulus reflects the elastic storage modulus G') and viscous behaviour (loss modulus G'').

For example, in [2], nanoparticles (NP) containing gels resulted in an increasing yield stress with increasing NP concentration. Moreover, as described in [3] the macroscopic viscoelastic properties of a physical hydrogel are reversibly modulated by tuning the microscopic hydrogen bonding interactions with pH. As a result, the yield stress of the hydrogel is greatly enhanced reducing the pH from 7.0 to 5.0, and also G′ and G″ indicate an enhanced rigidity and stability of the gel. Mensitieri et al. show that auto-crosslinked polysaccharide (ACP) polymers can be modulated both by varying the degree of crosslinking and the weight concentration [4]. At a fixed concentration, the elasticity can be increased by increasing the level of chemical crosslinking substantially without increasing the viscous dissipation.

However, these studies are not focused on a specific modulation of the viscous component, but on the measurement of the resultant effect on both the elastic and viscous component. To address this gap, our aim was to modulate material viscoelasticity by acting on the damping component, i.e. through tuning of the liquid phase viscosity of alginate gels with different dextran concentrations.

II. Material and Methods

A. Sample Preparation

Agarose powder was dissolved in water and aqueous solutions of 2 and 5% w/v dextran respectively to give a final agarose concentration of 1% w/v. The solution was stirred under boiling and then cast into custom moulds, obtaining 13mm diameter-8mm heights cylindrical samples.

B. Viscosity Measurements

The viscosity of the different dextran solutions was measured with an AMVN Automated micro-viscosimeter.

C. Mechanical Testing

The viscoelastic properties of the gels were tested using the epsilon-dot method [5]. Unconfined bulk compression tests were performed at different strain rates (0.0005 – 0.001 – 0.005 and 0.01s^{-1}) with a ProLine Z005 Zwick/Roell.

The mean stress-time curves were globally fitted using the relative equation for the standard linear solid model (Fig. 1), obtaining E1, E2 and η and consequently the equilibrium modulus E_{eq}, the instantaneous modulud E_{ist} and the relaxation time τ as described in [5].

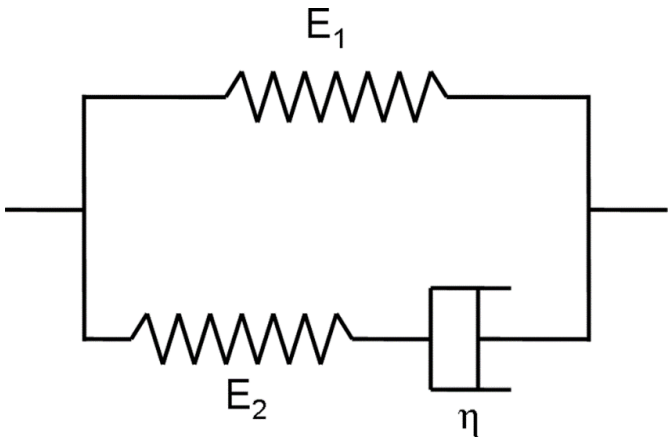


Figure 1. SLS model

III. Results And Discussions

A. Viscosity Measurements

In Table I, the measured density and viscosity for water (control) and the 2% and 5% dextran solution measured at different temperatures (16, 21 and 26°C) are shown. As

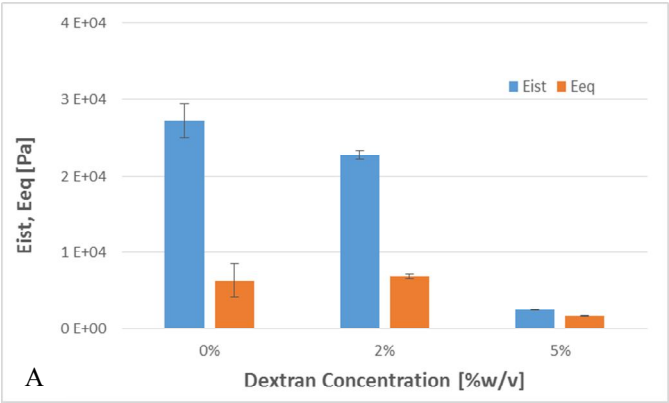
expected, viscosity increases with increasing dextran concentration.

TABLE I
VISCOSITY MEASUREMENTS

Dextran (%w/v)	*Density [g/cm^3]*	*Viscosity [mPa*s]*
0% - 16°C	0,9989	1,1039
0% - 21°C	0,9979	0,9774
0% - 26°C	0,9967	0,8729
2% - 16°C	1,0062	2,6775
2% - 21°C	1,00523	2,347
2% - 26°C	1,0039	2,0761
5% - 16°C	1,0163	6,8656
5% - 21°C	1,0152	5,9734
5% - 26°C	1,014	5,2314

B. *Viscoelastic Properties*

The results in Figure 2 show how the increased viscosity of the liquid phase is reflected in a reduction of both instantaneous and equilibrium moduli and in a reduction of the relaxation time. As expected, a variation of one component, in this case the viscous element, implies the variation of the other. In fact, in biomaterials viscoelastic and elastic properties are intrinsically related through the time constant. Thus it is possible to modulate the mechanical behaviour of the material acting only on the damping component instead of on the crosslinking of the 'elastic network'. In particular, the more significant variations of the instantaneous elastic modulus, E_{ist} and the time constant, τ with respect to the equilibrium modulus, E_{eq} reflect the action on E2 and η, which are placed in series in the SLS parallel model.



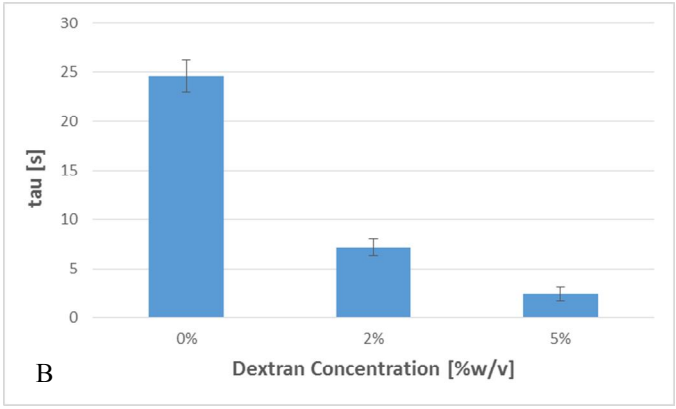


Figure 2. Viscoelastic measurements: a) Instantaneous and Equilibrium modulus; b) Relaxation time.

IV. CONCLUSION

Considering the importance of the viscous component in tissues and biomaterials, in this study the viscosity of the liquid phase of agarose samples was modulated. Specifically, the liquid phase viscosity was varied by adding increasing concentrations of dextran to the aqueous medium. Mechanical tests showed that the modulation of the liquid phase is reflected in the viscoelastic properties of the gels. Increasing the viscosity of the solution resulted in an increase of the viscous component and a reduction of elastic one. Therefore, this method is suitable to specifically modulate material viscoelasticity.

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