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Time-dependent rheological behaviour of bacterial cellulose hydrogel

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ABSTRACT

This work focuses on time-dependent rheological behaviour of bacterial cellulose (BC) hydrogel. Due to its ideal biocompatibility, BC hydrogel could be employed in biomedical applications. Considering the complexity of loading conditions in human body environment, time-dependent behaviour under relevant conditions should be understood. BC specimens are produced by *Gluconacetobacter xylinus* ATCC 53582 at static-culture conditions. Time-dependent behaviour of specimens at several stress levels is experimentally determined by uniaxial tensile creep tests. We use fraction-exponential operators to model the rheological behaviour. Such a representation allows combination of good accuracy in analytical description of viscoelastic behaviour of real materials and simplicity in solving boundary value problems. The obtained material parameters allow us to identify time-dependent behaviour of BC hydrogel at high stress level with sufficient accuracy.

1. Introduction

Bacterial cellulose (BC) hydrogel (Fig. 1a) is known for its excellent biocompatibility [26]. It makes BC hydrogels a potential material for various bioengineering applications, such as implant replacement of human tissues [16,27,28], encasement for tissue regeneration [2,3,9,13], etc. From a structural point of view, BC hydrogel is a typical nonwoven bio-material. It consists of high-crystalline (70–80% [12]) long nanofibres that are randomly distributed in its radial-transverse plane (Fig. 1b). Fibrous layers with some cross-links form a multi-layer structure in the out-of-plane direction (Fig. 1c). Such a fibrous scaffold with high porosity (~99vol.%) is capable to hold a large amount of interstitial water, forming the hydrogel. The growing interest to BC is not only thanking to its excellent biological properties but also because of attractive mechanical properties of BC nanofibres [10]. BC fibre-reinforced bio-composites with optimized mechanical properties have found various applications for artificial tissues, such as a BC/fibrin composite for artificial blood vessel [4].

A matrix of the BC composite usually acts as a damper in loading-bearing processes; therefore, the BC hydrogel or the composite based on it demonstrates viscoelastic behaviour. Since BC is exposed to complex loading conditions of human-body environment, it is extremely important to characterize its viscoelastic properties in an explicit analytical form. Brown et al. [4,5] quantified elastic and viscoelastic properties of a BC fibre-reinforced bio-composite with tensile and cyclic creep tests, demonstrating its suitability as a potential implant for artificial blood vessels. Nimeskern et al. [18] evaluated viscoelastic properties of BCs with various cellulose contents by means of stress–relaxation indentation; then, comparing the obtained data with those for ear cartilage. It was demonstrated that BC could be used as ear-cartilage replacement. Accurate prediction of viscoelastic behaviour using an appropriate theoretical approach is necessary for optimization of mechanical performance of BC under application-relevant conditions. A spring/dashpot combination model is often used to depict creep and stress–relaxation behaviour. Kim et al. [11] used a model formed by parallel spring-dashpot combinations and a dashpot in series to describe cyclic creep behaviour of cellulose electro-active paper at various temperatures. Frensemeier et al. [8] used the Burgers viscoelastic model that accounts for water release to describe a viscoelastic response of a BC hydrogel. Lopez-Sanchez et al. [15] studied viscoelastic behaviour of a BC hydrogel with a linear transversely isotropic poroelastic model based on its compression response to loading with various strain rates.

The use of oversimplified spring/dashpot combinations is caused by the fact that a standard approach to solve problems of viscoelasticity is based on elasticity–viscoelasticity correspondence principle (see, for example [6]). The problem is formulated in a Fourier or Laplace domain, treated as the elastic one, and then, an inverse transform gives the desired viscoelastic solution. The main challenge in this approach is obtaining analytical formulas for the inverse transform. It can be achieved only for some particular cases corresponding to various combinations of springs and dashpots. Note, however, that the governing relations of

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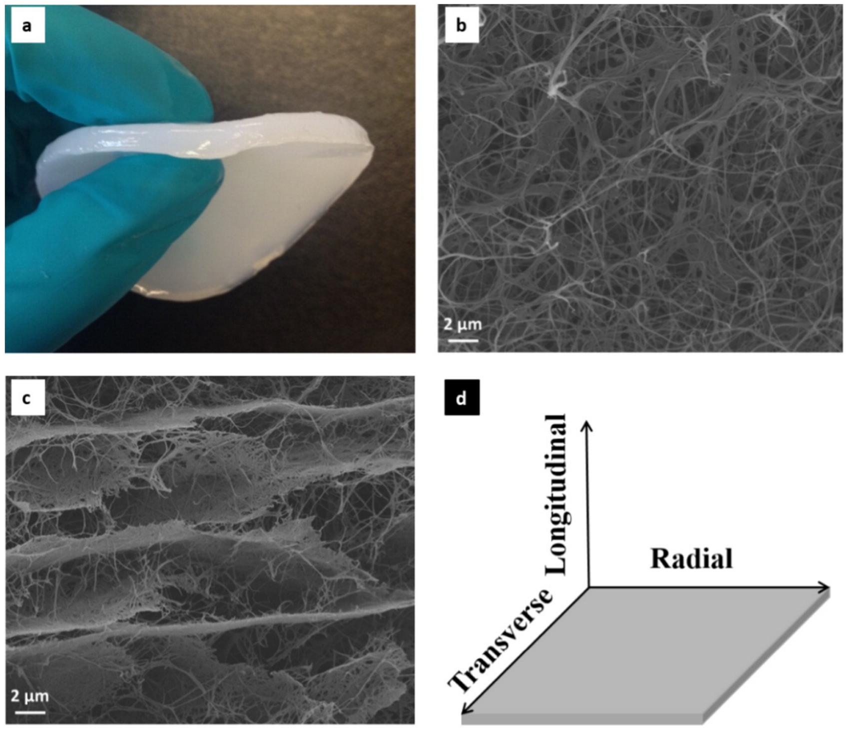

Fig. 1. (a) Disc sample of wet BC hydrogel's. SEM images of BC hydrogel show a random distribution of cellulose fibres in the radial-transverse plane (b) and multiple-layer structure with some links between layers along out-of-plane direction (c). (d) Schematic diagram of radial, transverse and out-of-plane directions.

viscoelasticity (as all other relations of this kind) are of a phenomenological nature — they are chosen to match experimental data obtained in standard tests on creep and relaxation. Unfortunately, simplest models describing combinations of springs and dashpots are not sufficiently flexible to match experimental data for real materials while more complex ones do not allow analytical expressions for inverse Fourier or Laplace transforms appearing in the solution.

An alternative description of viscoelastic behaviour was proposed by Scott Blair and Coppen [22,23] and Rabotnov [20]. They suggested using fraction-exponential operators that, on the one hand, can describe experimental data of real materials with sufficient accuracy and, on the other hand, allow analytical Laplace transformations. Recently, the use of these fraction-exponential operators attracted attention of researchers in the area of solid mechanics (especially mechanics of heterogeneous materials) again; the respective detailed discussion can be found, for instance, in the book of Podlubny [19]. A connection between fraction-exponential operators and various simplistic models involving combinations of dashpots and springs was analysed in the work of Di Paola and Zingales [7]. Applications of fraction-exponential operators to various heterogeneous viscoelastic materials are discussed in Levin and Sevostianov [14], Sevostianov and Levin [24] and Sevostianov et al. [25]. In this paper, a fraction-exponential model is employed to describe a time-dependent behaviour of the BC hydrogel. Tensile creep tests at various stress levels were performed to quantify its mechanical behaviour. A numerical procedure was developed in MATLAB to recover the model parameters from the experimental data.

2. Fraction exponential operators

To describe viscoelastic properties of the material, a most general form of the respective governing equation is used in the form of Stieltjes convolution:

$$\varepsilon_{ij}(x,t) = S_{ijkl}\sigma_{kl}(x,t) + \int_0^t K_{ijkl}(t-\tau)\sigma_{kl}(x,\tau)d\tau \quad (1)$$

where ε_{ij} and σ_{kl} are the strain and stress tensors, respectively, S_{ijkl} is a fourth rank tensor of instantaneous elastic compliance and $K_{ijkl}(t)$ is time-dependent forth-rank tensor (creep kernel) satisfying the fading-memory principle: $K_{ijkl}(t)\underset{t\to\infty}{\to}0$. It is assumed that volume changes during deformation are purely elastic. Then, for an isotropic material, expression (1) can be written as

$$\varepsilon_{ij}(x,t) = \frac{1}{3K_0}\left(\frac{1}{3}\delta_{ij}\right)\sigma_{kk}(x,t) + \frac{1}{2}(\mu^*)^{-1}\left(\sigma_{ij}(x,t) - \frac{1}{3}\delta_{ij}\sigma_{kk}(x,t)\right) \quad (2)$$

where

$$(\mu^*)^{-1}[f(x,t)] = \frac{1}{\mu_0}f(x,t) + \int_0^t J(t-\tau)f(x,\tau)d\tau, \quad (3)$$

K_0 is the bulk elastic modulus of the material, μ_0 is instantaneous shear modulus, and creep kernel $J_{(t-\tau)}$ satisfies the fading memory principle. To model this kernel, we follow Scott Blair and Coppen [22,23] and Rabotnov [20] who independently proposed to use fraction-exponential functions for this goal in the form:

$$J(t-\tau) \equiv \mathfrak{I}_\alpha(\beta, t-\tau) = (t-\tau)^\alpha \sum_{n=0}^{\infty} \frac{\beta^n (t-\tau)^{n(1+\alpha)}}{\Gamma[(n+1)(1+\alpha)]}, \quad (4)$$

As mentioned in the introduction, this representation allows analytical expression for an inverse Laplace transform in solving boundary-value problems, and, at the same time it is sufficiently general to match experimental data with good accuracy. To satisfy the fading memory principle, the following restrictions on the parameters entering Eq. (4) have to be satisfied:

$$\beta < 0; \quad -1 < \alpha \leq 0 \quad (5)$$