JOURNAL OF THE MECHANICAL BEHAVIOR OF BIOMEDICAL MATERIALS 37 (2014) 286–298

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jmbbm

Research Paper

Identification of the viscoelastic properties of soft materials at low frequency: Performance, ill-conditioning and extrapolation capabilities of fractional and exponential models

J. Ciambella[a,b,*], *A. Paolone*[b], *S. Vidoli*[b]

[a]*Advanced Composites Centre for Innovation and Science, University of Bristol, University Walk, BS8 1TR, Bristol, United Kingdom*

[b]*Dipartimento di Ingegneria Strutturale e Geotecnica, Sapienza Università di Roma, Via Eudossiana 18, 00184 Rome, Italy*

ARTICLE INFO

Article history:
Received 19 March 2014
Received in revised form
28 May 2014
Accepted 30 May 2014
Available online 10 June 2014

Keywords:
Soft-tissues
Polymers
Fractional viscoelasticity
Dynamic moduli
Biorheology

ABSTRACT

We report about the experimental identification of viscoelastic constitutive models for frequencies ranging within 0–10 Hz. Dynamic moduli data are fitted for several materials of interest to medical applications: liver tissue (Chatelin et al., 2011), bioadhesive gel (Andrews et al., 2005), spleen tissue (Nicolle et al., 2012) and synthetic elastomer (Osanaiye, 1996). These materials actually represent a rather wide class of soft viscoelastic materials which are usually subjected to low frequencies deformations.

We also provide prescriptions for the correct extrapolation of the material behavior at higher frequencies. Indeed, while experimental tests are more easily carried out at low frequency, the identified viscoelastic models are often used outside the frequency range of the actual test.

We consider two different classes of models according to their relaxation function: Debye models, whose kernel decays exponentially fast, and fractional models, including Cole–Cole, Davidson–Cole, Nutting and Havriliak–Negami, characterized by a slower decay rate of the material memory. Candidate constitutive models are hence rated according to the accurateness of the identification and to their robustness to extrapolation. It is shown that all kernels whose decay rate is too fast lead to a poor fitting and high errors when the material behavior is extrapolated to broader frequency ranges.

*Corresponding author.
E-mail addresses: jacopo.ciambella@bristol.ac.uk (J. Ciambella), achille.paolone@uniroma1.it (A. Paolone), stefano.vidoli@uniroma1.it (S. Vidoli).

http://dx.doi.org/10.1016/j.jmbbm.2014.05.031
1751-6161/

1. Introduction

Adequate modeling of the viscoelastic properties of soft materials is of paramount interest to medical applications (e.g., Kyriacou et al., 2002; Klatt et al., 2007; Rashid et al., 2012a), tissue engineering (e.g., Limbert and Middleton, 2004; Freed and Diethelm, 2006), polymers science (e.g., Tee and Dealy, 1975; Oakley et al., 1998) and food rheology (e.g., Singh et al., 2006). The evaluation of rheological properties has recently gained an increasing interest from biologists and engineers (see the comprehensive review in Humphrey, 2003).

In the laboratory, the mechanical properties are usually probed through experiments involving strain controlled elongation/compression (Ciambella et al., 2010; Rashid et al., 2012c, 2012b), torsion (Yoo et al., 2011) or shear (Nicolle et al., 2012; Chatelin et al., 2011). In all cases, sinusoidal strain histories allow a more accurate assessment of the frequency dependence of the material parameters. If the strain amplitude is small, and the material behaves linearly, the response can be characterized through the dynamic moduli: the storage modulus, G', and the loss modulus, G'', as determined from the components of the force in-phase (storage) and in-quadrature (loss) with the applied deformation.

Fig. 1 shows experimental data for porcine liver tissue (Chatelin et al., 2011), bioadhesive gel composed by cellular derivative (Andrews et al., 2005), porcine spleen tissue (Nicolle et al., 2012) and ethylene–propylene–diene termopolymer (EPDM) (Osanaiye, 1996). On such soft viscoelastic materials both storage and loss moduli display a high sensitivity with respect to frequency variations. A similar behavior is indeed observed in many other materials such as polyurethane foam (Gottenberg and Christensen, 1964), foods (Singh et al., 2006), filled rubber (Ciambella et al., 2011) and in a number of soft-tissues, both biological and synthetic (Moresi et al., 2004; Umut Ozcan et al., 2011; Yoo et al., 2011).

All materials in Fig. 1 in the frequency range considered display a power law-like dependence (f^{γ}) with $\gamma<1$. This sublinear behavior corresponds to a high sensitivity in linear-linear plots, where the same curves would stem from $\omega \simeq 0$ with very high rates.

In Ciambella et al. (2011) a thorough analysis of the relationship between the frequency behavior of the dynamic moduli and the time behavior of the viscoelastic kernel, *i.e.*, the material's memory, has been carried out for both linear and nonlinear viscoelastic models. It is shown that, if the rate of decay of the material memory is too fast, *e.g.* exponential, the resulting storage modulus has a vanishing rate for $\omega \simeq 0$ apparently in contrast to Fig. 1.

Accordingly, we analyze here how the time dependence of the material memory affects the identification of viscoelastic parameters in the frequency domain. While the exponential kernel (Debye model) is by far the most used in the literature, we show that its exponential decaying rate can cause the ill-conditioning of the identification problem. Indeed, for those materials with a high frequency sensitivity, kernels characterized by slower decay rates should be preferred; within this class, we compare the Cole–Cole, Havriliak–Negami and Nutting kernels.

The paper is organized as follows: in Section 2, the different viscoelastic constitutive models considered are analyzed and for each of them the definition of storage and loss moduli are explicitly evaluated; accordingly the complex susceptibility χ and the rate of decay of the viscoelastic kernel are computed. Section 3 outlines the identification procedure, whilst the identification results are reported, compared and discussed in Section 4. In particular, the optimal value of the objective function, the condition number near the minimum point, Akaike's information criterion and the robustness to frequency extrapolation are used to rate the performances of the considered models.

2. Constitutive modeling

We consider a linear viscoelastic constitutive equation, *i.e.*,

$$\sigma(t) = \int_{-\infty}^{t} G(t-s)\dot{\varepsilon}(s)\,ds, \tag{1}$$

where σ designates either the tensile or the shear stress. Here ε represents the strain and G the relaxation function, or *viscoelastic kernel*. It is important to note that the fading memory property guarantees that the stress always reaches a steady state with the same frequency of the input, if the corresponding strain is oscillatory, *e.g.*, sinusoidal or cosinusoidal. This guarantees that the storage and loss moduli can be formally defined.

By using the Fourier transform, from (1) one obtains the constitutive equation in the frequency domain, *i.e.*,

$$\tilde{\sigma}(\omega) = G^{*}(\omega)\tilde{\varepsilon}(\omega), \tag{2}$$

where $\tilde{\sigma}$ and $\tilde{\varepsilon}$ are the Fourier transforms of the corresponding time variables (Tschoegl, 1989). When $G^{*}(\omega) = G = \text{const.}$, the material is elastic and G represents the elastic modulus (Young's modulus or shear modulus, according to the type of test performed). In general, G^{*} is a complex function of the frequency ω and is called the complex modulus of the viscoelastic materials. G^{*} can be conveniently rewritten in terms of its real and imaginary parts:

$$G^{*}(\omega) = G'(\omega) + jG''(\omega) \tag{3}$$

i.e., the *storage*, G', and *loss*, G'', moduli. The relationships between the relaxation function and the dynamic moduli readily follow from Eq. (1) (see, e.g., Ciambella, 2012)

$$G'(\omega) = G_{\infty} + \int_{0}^{+\infty} \hat{G}(s)\omega\,\sin(\omega s)\,ds,$$
$$G''(\omega) = \int_{0}^{+\infty} \hat{G}(s)\omega\,\cos(\omega s)\,ds. \tag{4}$$

with $\hat{G}(t) = G(t) - G_{\infty}$ and $G_{\infty} = \lim_{t\to\infty} G(t)$. For solid materials the long-term relaxation modulus G_{∞}, mimicking the static elastic modulus, is strictly positive, while for liquids G_{∞} is zero. Equivalently the storage and loss moduli could be defined in terms of the instantaneous modulus G_0; however, this latter is very difficult to determine experimentally.

An equivalent quantity sometimes used is the complex susceptibility $\chi(\omega)$, that is linked to the complex modulus G^{*} by

$$\chi(\omega) = 1 - \frac{G^{*}(\omega) - G_{\infty}}{G_0 - G_{\infty}}, \quad G_0 := G(0). \tag{5}$$