Journal of Non-Newtonian Fluid Mechanics 235 (2016) 64–75

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: www.elsevier.com/locate/jnnfm

A systematic approximation of discrete relaxation time spectrum from the continuous spectrum

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ARTICLE INFO

Article history:
Received 28 March 2016
Revised 3 May 2016
Accepted 16 July 2016
Available online 19 July 2016

Keywords:
Discrete relaxation spectrum
Linear viscoelasticity
Levenberg–Marquardt method

ABSTRACT

Most of viscoelastic models contain linear and nonlinear viscoelastic parameters and the linear viscoelastic parameters correspond to relaxation time spectra of materials. The relaxation spectra can be classified into continuous and discrete ones. Discrete relaxation time spectrum is more convenient to simulate multi-mode models than continuous one because it demands shorter calculation time. It has been demonstrated that the continuous spectrum is uniquely determined in views of theoretical (Fuoss and Kirkwood, 1941; Davies and Anderssen, 1997) [1,23] as well as empirical approaches (McDougall et al., 2014) [5]. Whereas, it is reported that different algorithms for discrete spectrum infer the different results from the same data (Malkin and Masalova, 2001) [14]. This is the study on a systematic method for discrete spectrum on the basis that a discrete spectrum must be consistent with the continuous one. We suggest a simple method to extract discrete relaxation spectrum as a systematically approximated continuous spectrum by means of the Levenberg–Marquardt method. The new algorithm is tested and compared with previous algorithms using synthesized model spectra and experimental data.

1. Introduction

Fuoss and Kirkwood [1] derived an analytical equation that relates a continuous relaxation time spectrum to loss modulus. Since the derivation was made by Fourier transform, the equation of Fuoss and Kirkwood (FK equation) implies the uniqueness of continuous relaxation spectrum. Several attempts have been made to develop reliable algorithms for continuous spectrum. The representative algorithms are the nonlinear regularization method (NLRG) of Honerkamp and Weese [2], the cubic Hermite spline method (CHS) of Stadler and Bailly [3], the fixed–point iteration (FPI) of Cho and Park [4]. McDougall et al. [5] demonstrated the uniqueness of continuous relaxation spectrum by use of aforementioned algorithms.

Continuous spectra allow us to calculate various viscoelastic functions by means of integrations while discrete spectra do the same thing by summations. Hence, discrete spectra save calculation time and the calculation is simpler than that using continuous spectra. The simplicity of discrete spectra is emphasized when a nonlinear viscoelastic flow is simulated by numerical methods. Therefore, there have been a number of efforts to develop algorithms for a discrete relaxation spectrum such as Baumgärtel and Winter [6], Tschoegl and Emri [7], Fulchiron et al. [8], Simhambhatla and Leonov [9], Malkin and Kuznetsov [10], Jensen [11] and Cho [12]. The allocation method is a common method to determine discrete relaxation time spectrum. It assigns the relaxation times of spectrum with equidistance on logarithmic time axis and then determines appropriate intensities of the spectrum at each relaxation time. Although, it reduces the number of variables that should be determined, the sufficient distance between each point should be considered. If the spacing is too large or too close, it is found that this method gives negative spectrum. Baumgärtel and Winter [6] suggested a new method to determine a discrete relaxation spectrum. This method adjusts not only intensities but also positions of a relaxation spectrum. The number of modes is another important parameter and it is adjusted to provide the best fit for input experimental data. It is found that the negative intensities of the spectrum appear as the number of modes increases. In order to overcome this problem, their method merges or eliminates the unnecessary components of the spectrum.

Malkin and Kuznetsov [10] developed a method using linearization of functional of errors. It is based on the searching the proper coefficients of series which give the minimum of root mean square error. Kaschta and Schwarzl [13] have suggested a method determining retardation time spectrum from creep or recovery data with equidistance spacing of retardation strengths.

Although there have been valuable efforts to develop an efficient method for inferring discrete relaxation time spectrum, Malkin and Masalova [14] found that the different discrete spectra are obtained by different algorithms from the same data.

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http://dx.doi.org/10.1016/j.jnnfm.2016.07.004
0377-0257/

It can be said that a discrete spectrum is one of various approximations of a unique continuous spectrum. Hence, it may be natural that the discrete spectrum cannot be determined uniquely. If the uniqueness of the discrete spectrum is denied, what would be the conditions for a good algorithm of discrete spectra? Since a discrete spectrum is an approximation of the continuous one, a good algorithm must give a discrete spectrum consistent with the continuous one. The points of discrete spectrum such as (λ_i, g_i) must be aligned on the curve of continuous spectrum whenever a suitable scaling of relaxation intensity, g_i is applied. The other condition is that calculated discrete spectrum should be parsimonious. In other words, the number of relaxation time λ_i must be small as possible as the spectrum fits experimental data well. It is obvious that fewer relaxation times shorten the calculating time for multimode viscoelastic models.

Since there have been a number of reliable algorithms for continuous spectrum, we investigated a systematic approach to extract discrete spectrum from the well-determined continuous spectrum with satisfying the two conditions for a good algorithm for discrete spectra.

2. Theoretical background

Concrete foundation of linear viscoelasticity connects various linear viscoelastic functions and relaxation spectrum. These relations are able to be expressed by simple viscoelastic functions. For example, storage and loss moduli can be written as [15]

$$G'(\omega) = \int_0^\infty \frac{\lambda^2\omega^2}{1+\lambda^2\omega^2}\frac{H(\lambda)}{\lambda}\,\mathrm{d}\lambda\,;\quad G''(\omega) = \int_0^\infty \frac{\lambda\omega}{1+\lambda^2\omega^2}\frac{H(\lambda)}{\lambda}\,\mathrm{d}\lambda \tag{1}$$

where ω is angular frequency and λ is relaxation time. $H(\lambda)$ is continuous relaxation time spectrum. For convenience, substitutions of $\tau = \log\lambda$ and $\nu = \log\omega$ convert Eq. (1) into the followings.

$$G'(\nu) = \int_{-\infty}^\infty H(\tau)K_\mathrm{E}(\tau+\nu)\mathrm{d}\tau\;;\quad G''(\nu) = \int_{-\infty}^\infty H(\tau)K_\mathrm{V}(\tau+\nu)\mathrm{d}\tau \tag{2}$$

where

$$K_\mathrm{E}(x) = \frac{e^{2x}}{1+e^{2x}};\quad K_\mathrm{V}(x) = \frac{e^{x}}{1+e^{2x}}. \tag{3}$$

When the relaxation spectrum is given as discrete one, they can be calculated as below:

$$G'(\nu) = \sum_{i=1}^N g_i K_\mathrm{E}(\tau_i+\nu);\quad G''(\nu) = \sum_{i=1}^N g_i K_\mathrm{V}(\tau_i+\nu) \tag{4}$$

where g_i is intensity of discrete relaxation spectrum with the unit of moduli. The number of relaxation times is noted as N.

As mentioned before, there is no clue for a uniqueness of discrete relaxation spectra. Determination of discrete relaxation spectra deals with much higher degree of freedom compared with continuous relaxation spectra. It means that the algorithm should determine not only relaxation times and intensities at each time but also the number of modes assuring the best fit for input data concurrently. In order to reduce the degree of freedom, we assumed that intensities of a discrete relaxation spectrum are closely related to the continuous relaxation time spectrum by means of a suitable scaling relation. Assumed constraint can be written as

$$\sigma H(\tau_k) = g_k. \tag{5}$$

Scaling factor σ is derived as

$$\varphi \equiv \int_{\tau_\mathrm{min}}^{\tau_\mathrm{max}} H(\tau)\,\mathrm{d}\tau = \sum_{k=1}^N g_k;\ \sigma = \frac{\int_{\tau_\mathrm{min}}^{\tau_\mathrm{max}} H(\tau)\,\mathrm{d}\tau}{\sum_{k=1}^N H(\tau_k)} = \frac{\varphi}{\sum_{k=1}^N H(\tau_k)} \tag{6}$$

for any k. Note that φ is a constant calculated by the integration of the continuous relaxation spectrum, while σ depends on the discrete relaxation times. The scaling factor σ should be kept updating when τ_k moves. Eq. (6) indicates that the intensities of a discrete relaxation spectrum can be spontaneously inferred from the well–determined continuous relaxation spectrum. Attention should be paid to distinguish σ and the scaling factor h_o demonstrated by Baumgärtel and Winter [16]. Detailed derivation is introduced by McDougall et al. [5] for equally spaced relaxation times.

$$H(\tau = \tau_k) = g_k/h_\mathrm{o} \tag{7}$$

An algorithm of Baumgärtel and Winter [16] is based on the nonlinear regression to determine a discrete relaxation spectrum, while a newly developed algorithm is to solve the inverse problem based on a constraint of Eq. (5). The method of Baumgärtel and Winter [16] keeps the number of relaxation times small. This method is included in a widely distributed software called IRIS. It determines the optimum discrete relaxation spectrum iteratively and this algorithm is patronized by many researchers in academic and industrial fields, but details of the algorithm are not published.

In the case of newly developed algorithm, we have only to determine the positions of discrete relaxation times based on least square error, when the continuous relaxation spectrum is plausibly determined. In order to solve the inverse problem of least square error, the Levenberg–Marquardt method is adopted. The objective function can be written as

$$\psi(\{\tau_k\}) = \sum_{\alpha=1}^M \left[1 - \frac{\varphi}{G'(\omega_\alpha)}\sum_{k=1}^N K_\mathrm{E}(\tau_k+\nu_\alpha)\tilde{H}(\tau_k)\right]^2 + \sum_{\alpha=1}^M \left[1 - \frac{\varphi}{G''(\omega_\alpha)}\sum_{k=1}^N K_\mathrm{V}(\tau_k+\nu_\alpha)\tilde{H}(\tau_k)\right]^2 \tag{8}$$

where M is the number of storage or loss modulus and $\tilde{H}(\lambda_k)$ is scaled relaxation spectrum as below

$$\tilde{H}(\tau_k) = \frac{\sigma}{\varphi}H(\tau_k) = \frac{g_k}{\varphi} \tag{9}$$

Because the intensities of the discrete relaxation spectrum are decided spontaneously, the relaxation time should be imposed where the minimum value of objective function is obtained. The normal equation can be derived as

$$\tilde{H}'(\tau_k) = \frac{1}{\sum_{m=1}^N H(\tau_m)}\left.\frac{\mathrm{d}H(x)}{\mathrm{d}x}\right|_{x=\tau_k} \tag{10}$$

$$\frac{\partial}{\partial\tau_i}\sum_{k=1}^N K(\tau_k+\nu_\alpha)\tilde{H}(\tau_k) = \sum_{k=1}^N K'(\tau_k+\nu_\alpha)\tilde{H}(\tau_k)\,\delta_{ik} + \sum_{k=1}^N K(\tau_k+\nu_\alpha)\tilde{H}'(\tau_k)\delta_{ik} - \sum_{k=1}^N K(\tau_k+\nu_\alpha)\tilde{H}(\tau_k)\tilde{H}'(\tau_i) \tag{11}$$

where $K(x)$ can be K_E or K_V of Eq. (3). $K'(x)$ can be

$$K'_\mathrm{E}(x) = \frac{2e^{2x}}{\left(1+e^{2x}\right)^2}\ \text{ or }\ K'_\mathrm{V}(x) = \frac{e^{x}-e^{3x}}{\left(1+e^{2x}\right)^2} \tag{12}$$

For convenience, the objective function of Eq. (8) can be written in a matrix notation. Let us define M–dimensional column vectors