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Digital estimators of relaxation spectra

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Abstract

Determination of the distribution of relaxation times (DRT) from a wide variety of the time- and the frequency-domain material functions, such as polarization current and charge, real and imaginary parts of complex dielectric permittivity and complex dielectric modulus, the appropriate mechanical and magnetic counterparts is generalized as a filtering problem on a logarithmic time or frequency scale. Algorithms of the appropriate digital DRT estimators are derived. A novel regularization strategy is proposed based on choosing sampling rate for the input data, which ensures acceptably low random error of the recovered spectra. Optimum frequency ranges and sampling rates are found for determination of the relaxation spectrum from the real part of complex permittivity and complex modulus. A multi-filter DRT recovery strategy is suggested by a bank of filters with different smoothing abilities.

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1. Introduction

Determination of distribution of relaxation times (DRT) without any exaggeration may be categorized as one of all the time the most challenging and hard ill-posed inversion problems, which despite considerable effort devoted cannot be considered to be completely solved.

In the present paper, an attempt is made to consider and analyze the problem of determination of the relaxation spectrum from the perspective of up-to-date signal processing [1].

Motivation of this work is to solve DRT recovery problem based on the novel data processing technologies [1] and to derive accurate, robust and computationally efficient algorithms operating without employing numerical integration and extrapolation of the data outside the measured range.

2. Theoretical background

Mathematically, determination of the function of DRT or the relaxation spectrum $F(\tau)$ from various time- and frequency-domain material functions $x(u)$ reduces to the inversion of an integral transform [2–4]

$$x(u) = \int_0^\infty F(\tau)K(u,\tau)\mathrm{d}\tau/\tau, \quad 0 < u < \infty \tag{1}$$

with kernels $K(u,\tau)$ of the type

$$K(u,\tau) = \begin{cases} \exp(-u/\tau)/\tau & \text{polarization current} & \text{(a)} \\ \exp(-u/\tau) & \text{stress relaxation} & \text{(b)} \\ 1-\exp(-u/\tau) & \text{polarization current} & \text{(c)} \\ 1/(1+u^2\tau^2) & \text{real part of permittivity} & \text{(d)} \\ u\tau/(1+u^2\tau^2) & \text{loss factor} & \text{(e)} \\ u^2\tau^2/(1+u^2\tau^2) & \text{real part of modulus} & \text{(f)} \end{cases}, \tag{2}$$

where variable u represents time or frequency.

Since kernels $K(u,\tau)$ depend on the ratio or product of arguments u and τ, Eq. (1) may be converted in the form of the Mellin convolution type transform

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$$x(u) = F \overset{\mathrm{M}}{*} k = \int_0^\infty F(\tau)k(u/\tau)\mathrm{d}\tau/\tau, \tag{3}$$

where $\overset{\mathrm{M}}{*}$ denotes the Mellin convolution and $k(u)$ are kernels (2a)–(2f) modified in the form needed for converting Eq. (1) into Eq. (3).

Material functions $x(u)$ used for inversion (1) extend usually over many decades of time or frequency and are typically considered on a logarithmic scale [2–4]

$$u^* = \log_q u/u_0, \tag{4}$$

where u_0 is an arbitrary normalization constant often chosen equal to 1. For logarithmic arguments (4), to remember that $u = u_0 q^{u^*}$, Eq. (3) alters into the appropriate Fourier convolution type transform

$$x(q^{u^*}) = F(q^{u^*}) \overset{\mathrm{F}}{*} k(q^{u^*})$$

having kernels depending on the difference of logarithmic arguments. Consequently, DRT may be formally determined by the appropriate deconvolution

$$F(q^{u^*}) = x(q^{u^*}) \overset{\mathrm{F}}{*} k^{-1}(q^{u^*}), \tag{5}$$

where $k^{-1}(q^{u^*})$ are inverse kernels existing in the sense of generalized functions. One can derive the Mellin transforms of $k^{-1}(q^{u^*})$ as the reciprocals of the Mellin transforms of kernels $k(u)$

$$H(\mathrm{j}\mu) = 1/M[k(u); -\mathrm{j}\mu] = 1\bigg/\int_0^\infty k(u)u^{-\mathrm{j}\mu-1}\mathrm{d}u, \tag{6}$$

where $\mathrm{j} = \sqrt{-1}$ and parameter μ named the Mellin frequency [5] represents the frequency of a function, whose independent variable is logarithmically transformed.

Deconvolution (5), which can be considered as an ideal DRT estimator, represents an ideal filter [1] operating on a logarithmic time- or frequency-domain. It may be implemented by a digital filter [1]

$$F[m] = \sum_{n=-\infty}^{\infty} h[n]x(m-n) \tag{7}$$

operating with equally spaced data on a logarithmic scale, which manifest as the data spaced according to geometric progression $u_n = u_0 q^n$ on a linear scale (logarithmic sampling [5–10]). For the logarithmically sampled data, Eq. (7) modifies into the following general algorithm

$$F(u_0 q^m) = \sum_{n=-\infty}^{\infty} h[n]x(u_0 q^{m-n}), \tag{8}$$

where $h[n]$ is a set of filter coefficients, which, of course, must be limited to the finite number in practice. Depending on kernels (2a)–(2f), general algorithm (8) modifies into the following three sub-algorithms or digital DRT estimators

$$F(u_0 q^m) = \begin{cases} u_0 q^m \sum_n h[n]x(u_0 q^{m-n}) & \text{(a)} \\ \sum_n h[n]x(u_0 q^{m-n}) & \text{(b)} \\ \sum_n h[n]x(q^{-m-n}/u_0) & \text{(c)} \end{cases}, \tag{9}$$

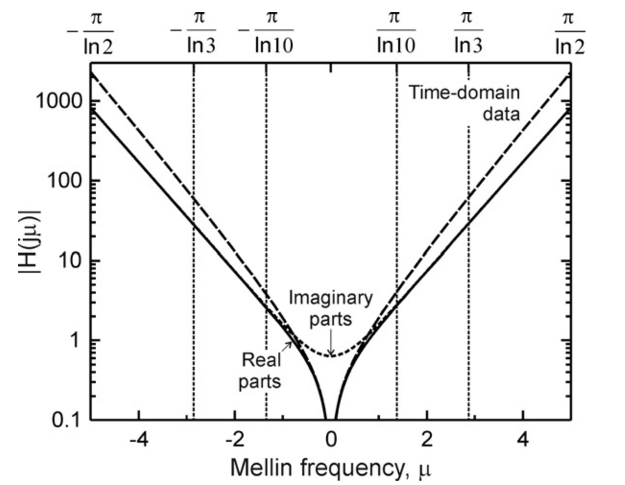

Fig. 1. Magnitude responses of the three ideal DRT estimators. Vertical lines show bandwidths $[-\pi/\ln q, \pi/\ln q]$ corresponding to different q.

where estimator (9a) relates to the polarization current (kernel (2a)), (9b) – to the polarization charge and the stress relaxation (kernels (2b) and (2c)), and (9c) – to the frequency-domain data (kernels (2d)–(2f)).

Digital estimators have periodic frequency responses

$$H(\mathrm{e}^{\mathrm{j}\mu}) = \sum_{n=-\infty}^{\infty} h[n]\exp(-\mathrm{j}\mu n \ln q) \tag{10}$$

in the Mellin transform domain, which approximate the appropriate frequency responses (6) of ideal estimators.

It has been shown [8,9] that inversion (1) for six kernels (2a)–(2f) reduces to the ideal filters with the three following frequency responses

$$H(\mathrm{j}\mu) = \begin{cases} -1/\Gamma(-\mathrm{j}\mu) & \text{for (2a)–(2c)} & \text{(a)} \\ \pm 2\sin(\mathrm{j}\pi\mu/2)/\pi & \text{for (2d) and (2f)} & \text{(b)} \\ 2\cos(\mathrm{j}\pi\mu/2)/\pi & \text{for (2e)} & \text{(c)} \end{cases}, \tag{11}$$

i.e. for the time-domain data, for the real parts, and for the imaginary parts, respectively (Fig. 1). Consequently, only three independent sets of coefficients $h[n]$ are necessary for determination of the relaxation spectrum from the material functions described by kernels (2a)–(9f).

3. Performance of digital DRT estimators

Accuracy of a digital DRT estimator is assessed by summed square error E between the samples of an exact relaxation spectrum $F_{\mathrm{exact}}(\tau_m)$ and the corresponding sequence $F(\tau_m)$ of the recovered spectrum

$$E = \sum_{m=1}^{M} [F_{\mathrm{exact}}(\tau_m) - F(\tau_m)]^2, \tag{12}$$

while noise behavior is described by noise coefficient S transforming input noise (random error) variance σ_x^2 into the output noise variance σ_y^2