Measurement Systems for Distribution of Relaxation and Retardation Times

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Abstract: - There is a widespread belief in scientific literature on material research that the distribution of relaxation and retardation times (DRRT) is a non-measurable quantity. In the presented contribution, we try to revise this belief. Based on the recently developed functional filtering approach for DRRT recovery, measurement systems are proposed to develop executing an active measurement experiment by exciting material with the specific stimulus, measuring material's responses sampled geometrically in time- or frequency-domain and processing them by the appropriate DRRT recovery filters. Design and algorithms of DRRT recovery filters are considered and practical implementations of DRRT measurement systems are proposed executing measurement experiments by exploiting the standard excitations, such as the Heaviside step function, the Dirac delta function and the steady-state multi-harmonic one.

Key-Words: - Distribution of Relaxation and Retardation Times (DRRT), Measurement Systems, Excitations, DRRT Recovery Filters

1 Introduction

Distribution of relaxation and retardation times (DRRT) or relaxation/retardation spectrum is one of the most fundamental quantities in various relaxation theories, such as linear theory of viscoelasticity [1-3], dielectric relaxation theory [3-5], magnetic resonance theory [6], etc. DRRT relates to molecular structure of materials [7-9]. Knowledge of DRRT is necessary for a wide range studies and applications, such as examination of the relationship between the molecular weight distribution and properties of a material, prediction of the behaviour of materials after an arbitrary excitation, interconversion of material functions, etc.

Historically, a concept of DRRT has been introduced to allow interpreting the non-exponential behaviour of the stress decay and strain retardation of viscoelastic materials, as well as the similar processes in dielectrics after abrupt turning on or off the electrical field. To ensure agreement with experimental data, it has been accepted that the non-exponential response functions result from a superposition of exponential (Debye) processes with different relaxation/retardation times. Mathematically, this idea has been expressed by replacing terms with a single relaxation/retardation time by their distributions.

DRRT is determined from various experimental data either in time- or frequency-domain to solve appropriate inverse problems being, as a rule, ill-posed. It is no exaggeration to say that DRRT recovery is one of all the time the most hard ill-posed inversion problems. Despite that intense studies of the determination of DRRT date back more than a century and that there is extensive literature on determination of DRRT [10-16], the problem still presents a lot of theoretical and practical challenges. Most of algorithms, particularly elder ones are based on curve fitting techniques and therefore, are applicable for limited class of materials to be in compliance with the mathematical model involved.

In scientific literature on material research, it is a widespread belief that DRRT is a non-measurable quantity. The two main reasons behind this belief are as follows: (i) the lack of effective computational resources for DRRT recovery, (ii) necessity to determine DRRT from experimental data, being, as a rule, incomplete (truncated), discrete and erroneous. However, recently [17-20], the problem of determination of DRRT has been analysed from the up-to-date signal processing perspective and computationally efficient DRRT recovery filters have been developed allowing to revise the belief of non-measurability of DRRT. In the presented contribution, a concept and practical implementations are considered for developing systems for measuring DRRT by exciting material with the standard stimulus, such as the Heaviside step function, the Dirac delta function and the steady-state multi-harmonic one, and processing of material's responses by the appropriate DRRT recovery filters.

2 Determination of DRRT

2.1 Input Data

Theoretically, DRRT can be calculated from various monotonic or locally monotonic material functions either in the time- of frequency- domain. These functions have various names in the specific experiments and in special fields [1-6, 10-16], however, in most cases, the functions represent the characteristic responses of a material to the three standard excitations (loadings), such as step (the Heaviside step function), impulse (the Dirac delta function) and harmonic (the steady-state sinusoidal) ones. Due to this, the material functions will be conditionally generalized here in two categories as *compliance* and *modulus* functions depending on the *force* (stress, voltage, etc.) or *displacement* (strain, charge, etc.) excitations in the experiments. Fig. 1 shows eight material functions traditionally used for calculation of DRRT and the excitations exploited to generate them.

2.2 DRRT Recovery – a Deconvolution Problem on the Logarithmic Scale

Recently [17-19], it has been demonstrated that determination of DRRT from the eight material functions shown in Fig. 1 represents a *deconvolution* or *inverse filtering problem on a logarithmic time or frequency scale*, which can be solved by the deconvolution filters with the following three frequency responses defined in the Mellin transform domain:

(i) $H(j\mu) = -1/\Gamma(j\mu)$ (1)

to be used for DRRT recovery from the time-domain material functions ($J(t)$, $J'(t)$, $G(t)$, $G'(t)$),

(ii) $H(j\mu) = \pm 2\sin(j\pi\mu/2)/\pi$ (2)

– for DRRT recovery from the real parts of frequency-domain material functions ($J'(\omega)$ and $G'(\omega)$), and

(iii) $H(j\mu) = 2\cos(j\pi\mu/2)/\pi$ (3)

– for DRRT recovery from the imaginary parts of frequency-domain material functions ($J''(\omega)$ and $G''(\omega)$). In Eqs. (1) – (3), $j=\sqrt{-1}$, Γ represents the Gamma function, and parameter μ, named '*Mellin frequency*', may be interpreted [17-20] as the angular frequency for a function (signal) on the logarithmic scale.

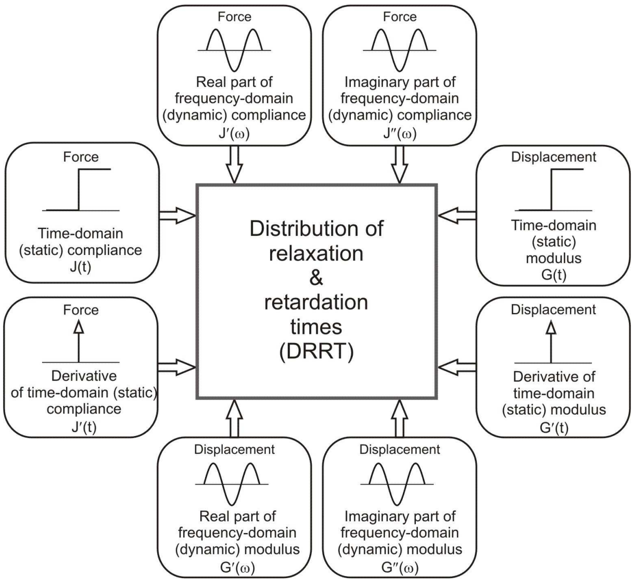

Fig. 1. Standard excitations and material functions used for calculation of DRRT.

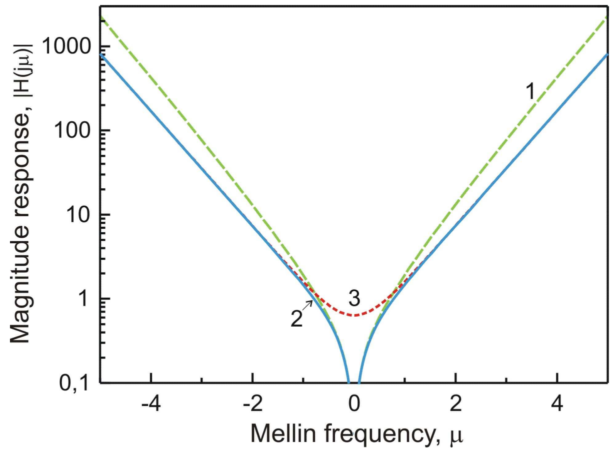

Fig. 2. Magnitude responses of ideal deconvolution filters recovering DRRT from the time-domain material functions (curve 1), real (curve 2) and imaginary (curve 3) parts of the frequency-domain functions.

In Fig. 2, the plots of magnitude responses are shown for (1) – (3). As it is seen, the magnitude responses are similar – extremely rapidly increasing functions.

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2.3 Discrete-time DRRT Recovery Filters

Formulation of determination of DRRT as a deconvolution or inverse filtering problem on the logarithmic scale gives a theoretical basis for implementation of DRRT recovery by the appropriate discrete-time deconvolution filters, which, for ensuring operation on the logarithmic scale, must process the input data sampled according to a geometric progression on the linear scale

$$u_n = u_0 q^n, \quad n = 0, \pm 1, \pm 2, ..., \quad q > 1 \tag{4}$$

where q is progression ratio specifying the sampling rate in the sense that $\ln q$ defines the distance between samples on the logarithmic scale, i.e. plays formally a role of sampling period, whereas its reciprocal describes the appropriate sampling frequency, and u_0 is an arbitrary normalization constant.

It has been found [17-19] that, for DRRT recovery from the eight material functions shown in Fig. 1, the following algorithms must be used:

$$F(u_0 q^m) = \sum_{n=-(N-1)/2}^{(N-1)/2} h[n] x\left(u_0 q^{m-n}\right) \text{ for odd } N, \tag{5}$$

$$F(u_0 q^m) = \sum_{n=-(N-2)/2-1}^{(N-2)/2} h[n] x\left(u_0 q^{m-0.5-n}\right) \text{ for even } N \tag{6}$$

for the time-domain material functions ($J(t)$ and $G(t)$) representing step responses,

$$F(u_0 q^m) = \sum_{n=-(N-1)/2}^{(N-1)/2} h[n] u_0 q^m x\left(u_0 q^{m-n}\right) \text{ for odd } N, \tag{7}$$

$$F(u_0 q^m) = \sum_{n=-(N-2)/2-1}^{(N-2)/2} h[n] u_0 q^m x\left(u_0 q^{m-0.5-n}\right) \text{ for even } N \tag{8}.$$

for the derivatives of time-domain material functions ($J'(t)$ and $G'(t)$) representing impulse responses, and

$$F(u_0 q^m) = \sum_{n=-(N-1)/2}^{(N-1)/2} h[n] x\left(u_0 q^{n-m}\right) \text{ for odd } N, \tag{9}$$

$$F(u_0 q^m) = \sum_{n=-(N-2)/2-1}^{(N-2)/2} h[n] x\left(u_0 q^{0.5+n-m}\right) \text{ for even } N, \tag{10}.$$

for the real and imaginary parts of frequency-domain material functions ($J'(\omega)$, $J''(\omega)$, $G'(\omega)$ and $G''(\omega)$). In Eqs. (5)-(10), $F(.)$ is a function of DRRT, N is filter length, $x(.)$ is a response function.

Fig. 3 summarizes DRRT recovery by the discrete-time deconvolution filters. Since a unique relation exists between both the time- and frequency-domain representations, the same ideal frequency response (1) for all the time-domain functions defines that the same filter coefficients (impulse response) $h[n]$ may be used for filters (5) and (7), and (6) and (8). In its turn, different coefficients $h[n]$ corresponding to frequency responses (2) and (3) shall be used in algorithms (9) and (10) for recovering DRRT from the real and imaginary parts.

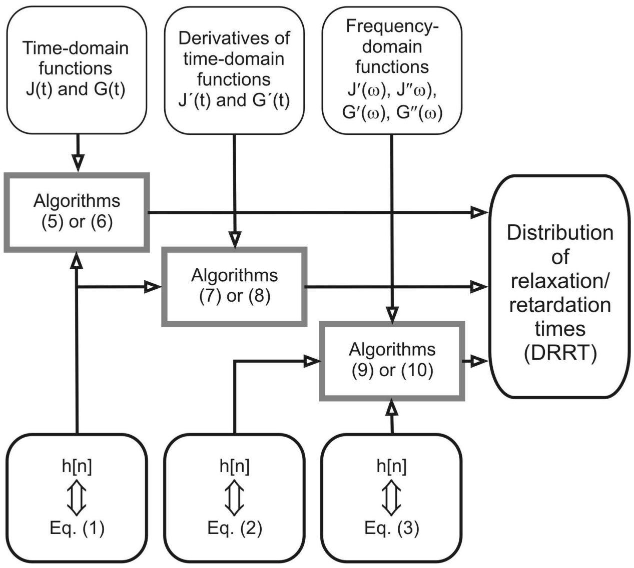

Fig. 3. Summary of DRRT recovery filters.

2.4 Design of DRRT Recovery Filters

It is well known that the determination of DRRT is a severely ill-posed problem [10-19], where small perturbations of the input signal may lead to large perturbations of the output spectra or, in other words, the solution is extremely sensitive to noise of measurement. To overcome this problem, various stabilization or regularization methods are employed to minimize the sensitivity to noise to acceptable for practice levels. Despite that a substantial body of regularization methods have been proposed, there is the lack of the techniques, which can work without human (operator) involvement necessary for implementing measurement systems.

Recently, this problem has been solved by developing a methodology for designing deconvolution filters [21-24], which integrates signal acquisition, regularization and discrete-time algorithm implementation. According to the methodology, a deconvolution filter is regularized by searching a combination of progression ratio q (sampling rate) and filter length N, which ensures the desired noise amplification and, for the found combination of q and N, it is designed by the identification method [25] performing a learning algorithm. In such way, the DRRT recovery filters are obtained with the guaranteed desired noise amplification producing maximum accurate waveforms of DRRT for available (limited) time or frequency ranges of input data.

3 DRRT Measurement Systems

We propose to develop DRRT measurement systems, which execute active measurement experiments by exciting material under test (MUT) with the specific excitations, measure MUT responses and process them by the appropriate DRRT recovery filters. A general block diagram for a DRRT measurement system is shown in Fig. 4.

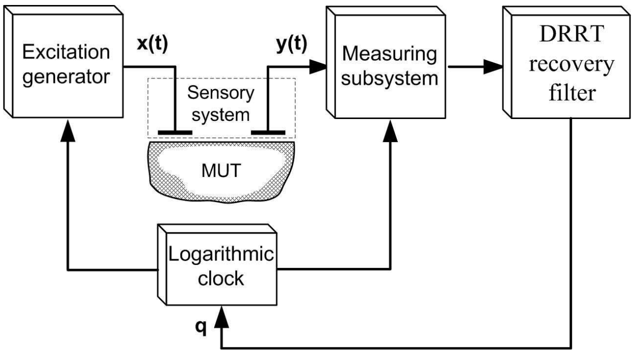

Fig. 4. General block diagram a DRRT measurement system.

An electrical excitation signal $x(t)$ from an *excitation generator* is transmitted to a *sensory system*, which produces an appropriate physical (electrical, mechanical, magnetic, thermal, etc.) excitation to *MUT* and detects and converts MUT response back into electrical response signal $y(t)$. This electrical response signal is measured and converted into discrete-time signal by a *measuring subsystem*. To calculate DRRT, discrete samples of the response signal are processed by *DRRT recovery filter*. Due to monotonicity of material responses [1-6], signal $y(t)$ is measured at the uniformly distributed instants on a logarithmic time or frequency scale manifesting as the instants distributed according to geometric progression (4) on the linear time or frequency scale. Such geometric (logarithmic) sampling is provided by a *logarithmic clock*. To ensure regularized solutions, i.e. the distributions with minimized sensitivity to noise, noise amplification of DRRT recovery filter is minimized to the desired value by finding the appropriate sampling rate (progression ratio q) [21-24] (see Subsection 2.4). The found value of q from DRRT recovery filter is transmitted to the logarithmic clock to ensure sampling of a response function with the needed sampling rate.

Implementation of a specific DRRT measurement system depends on a material response function (see Fig. 1) exploited for recovering the distributions, and so on the excitation used in the measurement experiment. DRRT measurement systems may be classified into two basic classes as time-domain and frequency-domain systems (Fig. 5). Further, formally, the time-domain systems may be divided into systems with the step or impulse excitations, while the frequency-domain systems employing harmonic excitations – as ones where DRRT is recovered through the real or imaginary part of a complex frequency-domain material function.

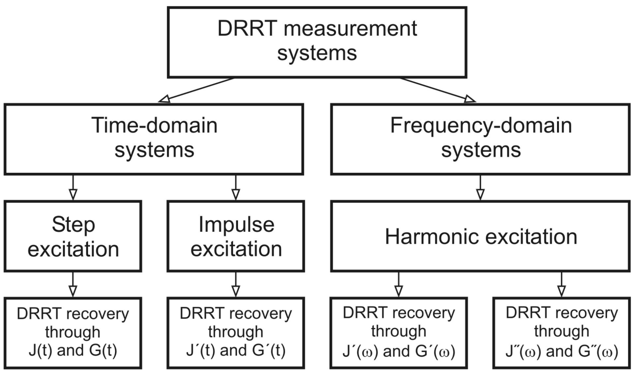

Fig. 5. Classification of DRRT measurement systems.

3.1 DRRT Measurement Systems in Time Domain

For time-domain DRRT measurement systems, the general block diagram (see Fig. 4) modifies into one shown in Fig. 6. In this case, MUT is excited by step

$$x(t) = X_0 1(t) \tag{11}$$

or impulse stimulus

$$x(t) = X_0 \delta(t) \tag{12}$$

with amplitude X_0, where $1(t)$ is unit step or Heaviside step function, and $\delta(t)$ is Dirac delta function.

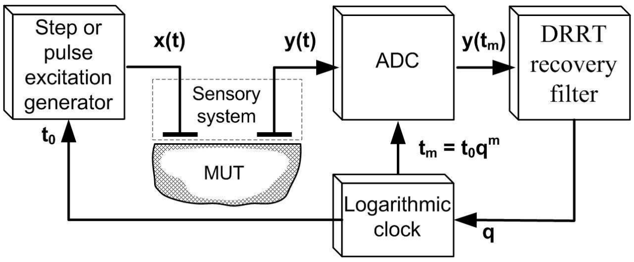

Fig. 6. Implementation of a DRRT measurement system in the time domain.

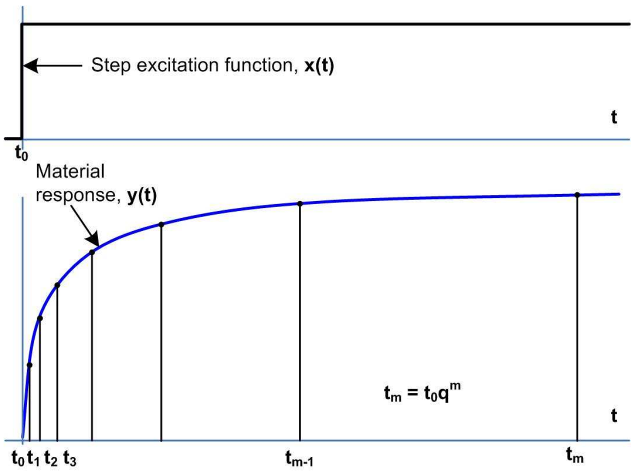

Fig. 7. Illustration of geometric sampling for a step-response function.

For excitations (11) and (12), MUT responds respectively as

$$y(t) = \begin{cases} X_0 J(t) \\ X_0 G(t) \end{cases} \tag{13}$$

or

$$y(t) = \begin{cases} X_0 J'(t) \\ X_0 G'(t) \end{cases}. \tag{14}$$

By an analog-to-digital converter (ADC), response signal (13) or (14) is measured and converted into geometrically sampled discrete-time samples at instants (Fig. 7)

$$t_m = t_0 q^m .$$

To obtain DRRT, the recorded samples of the response function are processed by DRRT recovery filter, which performs algorithms (5) or (6) for the step responses (13) and algorithms (7) or (8) for the impulse responses (14).

3.2 DRRT Measurement System in Frequency Domain

For frequency-domain DRRT measurement systems, the general block diagram (see Fig. 4) modifies into the scheme shown in Fig. 8. In this case, MUT is excited by harmonic excitations

$$x_m(t) = X_m \sin \omega_m t \tag{15}$$

at discrete frequencies ω_m altering according to geometric progression

$$\omega_m = \omega_1 q^{m-1} .$$

For excitations (15), MUT responds by harmonic responses of the same frequencies ω_m but with a different amplitudes Y_m and phases φ_m

$$y_m(t) = Y_m \sin(\omega_m t - \varphi_m) .$$

The system measures the amplitudes and phase differences between the excitations and responses by an *amplitude meter* and *phase difference meter*, from which the real or imaginary part of a complex frequency-domain material function is calculated. Further, similarly as for time-domain DRRT recovery systems (see Fig. 6), DRRT is calculated from the discrete – geometrically sampled in frequency domain samples of the real or imaginary part by DRRT recovery filters performing algorithms (9) or (10).

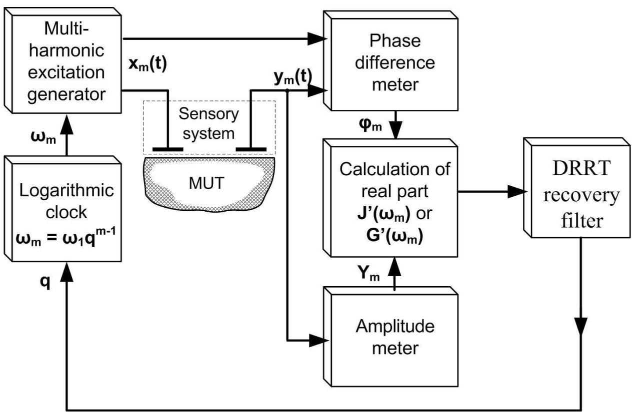

Fig. 8. Implementation of a measurement system in the frequency domain recovering DRRT from the real parts.

4 Conclusions

A widespread in scientific literature belief of non-measurability of distribution of relaxation and retardation times (DRRT) is revised. Practical

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implementation is considered for systems measuring DRRT by exciting a material with standard stimulus, such as the Heaviside step function, the Dirac delta function and the steady-state sinusoidal ones, measuring material's responses sampled geometrically in time- or frequency-domain and processing them by DRRT recovery filters.

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