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Deconvolution Filters for Determination of the Distribution of Relaxation and Retardation Times

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***Abstract* – The paper is devoted to determination of the distribution of relaxation and retardation times (DRRT) from various time- and frequency-domain response functions. It is demonstrated that the problem in general reduces to the three deconvolution tasks for the data on a logarithmic time or frequency scale. FIR deconvolution (inverse) filters operating with geometrically sampled data are proposed to use as DRRT estimators. The frequency responses are found and the algorithms of the estimators are derived for estimating DRRT from different response functions. It is disclosed that non-linear phase filters must be used for DRRT recovery from time-domain (impulse and step) response functions, whereas linear phase filters are required for DRRT recovery from the frequency-domain responses. Simulation results are presented obtained by two estimators from the noiseless and noisy input data.**

***Keywords*: Distribution of relaxation and retardation times (DRRT), functional filters, geometric (logarithmic) sampling, integral transforms, inversion**

I. Introduction

To describe objects exhibiting aperiodic behaviour, elementary relaxation and retardation systems [1] are used having the exponential impulse responses

$$g(t)=\begin{cases}\exp(-t/\tau)/\tau & \text{(a)}\\ \delta(t)-\exp(-t/\tau)/\tau & \text{(b)}\end{cases}, \tag{1}$$

where $\delta(t)$ is delta function, τ is relaxation/retardation time, and (1a) and (1b) relate to an elementary relaxation system and an elementary retardation system, respectively. Since the responses of real objects, e.g. materials do not comply often with the simple exponential law, (1) is generalized in the form:

$$g(t)=\begin{cases}\int_0^\infty F(\tau)\exp(-t/\tau)d\tau/\tau & \text{(a)}\\ \delta(t)-\int_0^\infty F(\tau)\exp(-t/\tau)d\tau/\tau & \text{(b)}\end{cases} \tag{2}$$

by introducing nonnegative function of **d**istribution of **r**elaxation/**r**etardation **t**imes (DRRT) or relaxation/retardation spectrum $F(\tau)$

DRRT contains valuable information about structure of aperiodic objects and is one of the most important quantities in various relaxation theories, including dielectric [2], [3], viscoelastic [3], [4], paramagnetic [5] ones. DRRT is not measurable directly, however, can be calculated from various experimental response functions to solve the appropriate inverse problems. The difficulty is that the inversions belong to ill-posed inverse problems where small perturbations in input data can yield unrealistic high perturbations in the results. Due to discretization, distortion by noise and incompleteness of experimental data, exact DRRT recovery is impossible and only physically feasible estimates can be obtained.

Despite of huge effort devoted, determination of DRRT poses still theoretical as well as experimental challenges with a number of unsolved questions. This, particularly, concerns so-called non-parametric methods [6], which contrary the parametric techniques based on curve fitting techniques do not make any assumption made about the parametric form of DRRT. At present, there is the lack of computationally efficient non-parametric methods for recovery of continuous DRRT with a strong theoretical basis in the signal processing context.

Motivation of this work is to gain an understanding of the overall problem framework of DRRT recovery in light of the up-to-date signal processing [7] and to propose computationally efficient algorithms based on the *functional filtering approach* [8] – [11].

II. Background of Inverse Functional Filtering

Functional filtering approach [8] – [11] has been developed for data interconversion of the monotonic and locally monotonic signals in the relaxation experiments via direct and inverse transformations, which can be reduced to the Mellin convolution transforms. For inverse transformations, the Mellin convolution transform may be represented in the form:

$$x(u)=y(u)\overset{M}{*}k(u)=\int_0^\infty y(r)k(u/r)dr/r = \int_0^\infty y(u/r)k(r)dr/r, \tag{3}$$

where $\overset{M}{*}$ denotes the Mellin convolution, variable u represents time or frequency, $x(u)$ is some recorded relaxation signal, $y(u)$ is some unknown signal that we wish to recover, and $k(u)$ is a kernel depending on the ratio of arguments u/r. Inversion of (3) formally can be expressed through the Mellin convolution transform with inverse kernel $k_{\mathrm{inv}}(u)$

$$y(u)=x(u)\overset{M}{*}k_{inv}(u)=\int_0^\infty x(r)k_{inv}(u/r)dr/r = \int_0^\infty x(u/r)k_{inv}(r)dr/r. \tag{4}$$

The functional filtering approach is based on the fact that data in the relaxation experiments [2] – [5] are monotonic or locally monotonic functions, which are usually recorded over many decades of time or frequency and, for this reason, are typically represented on a logarithmic scale

$$u^* = \log_q (u / u_0)\,, \tag{5}$$

where u_0 is an arbitrary normalization constant (often chosen to be equal to 1). For the logarithmic variables (5), the Mellin convolution type transforms (3) and (4) alter into the appropriate Fourier convolution type transforms, which may be interpreted as linear shift-invariant systems or ideal filters operating on a logarithmic scale. This constitutes a theoretical basis for executing transforms (3) and (4) by means *discrete convolution algorithms* or *discrete-time filters* processing uniformly sampled data on the logarithmic scale. Therefore, transform (4) can be executed by the following algorithm:

$$y\left(u_0 q^{u_m^*}\right) = \sum_{n=-\infty}^{\infty} h[n]\, x\left(u_0 q^{u_{n-m}^*}\right), \tag{6}$$

where $h[n]$ is the impulse response, which for computationally realizable algorithms must contain finite number of coefficients.

However, a more elegant solution is to implement algorithm (6) on the linear scale, where uniformly sampled data on the logarithmic scale manifest as the data sampled according to geometrical progression

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

Then, algorithm (6), depending on evenness or oddness of number of coefficients of impulse response $h[n]$, takes the final form of a functional filter [8]:

$$y(u_0 q^m) = \begin{cases} \sum\limits_{n=-(N-1)/2}^{(N-1)/2} h[n] x\left(u_0 q^{m-n}\right) & \text{for odd } N \quad \text{(a)} \\ \sum\limits_{n=-(N-2)/2-1}^{(N-2)/2} h[n] x\left(u_0 q^{m-0.5-n}\right) & \text{for even } N \quad \text{(b)} \end{cases}. \tag{7}$$

Since the functional filters execute the Mellin convolution type transforms, they have frequency-domain descriptions in the Mellin transform domain. Thus, transform (3) has the following frequency-domain representation:

$$X(j\mu) = Y(j\mu) K(j\mu)\,,$$

while its inverse (4) –

$$Y(j\mu) = X(j\mu) / K(j\mu)\,, \tag{8}$$

where functions with capital letters are the Mellin transforms of the appropriate functions with small letters. For example, function $K(j\mu)$, representing the frequency response of ideal direct functional filter, is described as:

$$K(j\mu) = \mathfrak{M}\left[k(u); -j\mu\right] = \int_0^\infty k(u) u^{-j\mu-1} du,$$

where $\mathfrak{M}$ denotes the Mellin transform, $j = \sqrt{-1}$ and parameter μ, further named *Mellin frequency*, represents the angular frequency of a signal (function) on the logarithmic scale [9]. According to (8), deconvolution (inverse) functional filter (4) has frequency response, which is equal the reciprocal of the Mellin transform of kernel $k(u)$

$$H(j\mu) = 1 / K(j\mu)\,, \tag{9}$$

and so it has an increasing magnitude response

$$\lim_{|\mu| \to \infty} |H(j\mu)| = \infty$$

coming from the fact that the magnitude responses of the direct filters $|K(j\mu)|$ usually decrease with growing frequency.

The necessary condition for implementation of inversion of (3) by a computationally realizable functional filter is the bounded magnitude response of the ideal filter at zero frequency [8]

$$\left|H(j\mu)\right|\Big|_{\mu=0} = \left|H(j0)\right| \neq \infty\,. \tag{10}$$

Following the suggestion in [12], the degree of ill-posedness of deconvolution filters will be characterized here quantitatively by noise amplification coefficient

$$S = \sum_n h^2[n]$$

multiplying input noise variance σ_x^2 to give output noise variance σ_y^2

$$\sigma_y^2 = S \sigma_x^2\,.$$

III. DRRT ESTIMATORS

A. *Ideal estimators*

In practice, DRRT is determined from impulse responses (2), as well as other response functions, such as the step responses given by the integrals of (2) or the real and imaginary parts of the frequency responses expressed via the Fourier transforms of (2). To take into consideration that zero and infinitive times are located at $\pm\infty$ on a logarithmic scale, i.e. are inaccessible in the relaxation experiments, mathematically, determination of DRRT from various time- and frequency-domain response functions is considered often without the delta function in (2b) and is generalized as an inverse problem [8] – [11] in the form:

$$x(u) = \int_0^\infty F(\tau) K(u, \tau) d\tau / \tau, \quad 0 < u < \infty \tag{11}$$

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

$$K(u,\tau)=\begin{cases}\exp(-u/\tau)/\tau & \text{(a)}\\ \exp(-u/\tau) & \text{(b)}\\ 1-\exp(-u/\tau) & \text{(c)}\\ 1/(1+u^2\tau^2) & \text{(d)}\\ u\tau/(1+u^2\tau^2) & \text{(e)}\\ u^2\tau^2/(1+u^2\tau^2) & \text{(f)}\end{cases} \qquad (12)$$

where $K(u,\tau)$ describes the response functions of the elementary relaxation and retardation systems with single relaxation/retardation time (DRRT in the form of the delta function). Kernels (12a) and (12b) represent the impulse and step response of the elementary relaxation system having the frequency response with real part (12d) and imaginary part (12e). In its turn, (12a) and (12c) express the impulse and step response of the elementary retardation system having the frequency response with real part (12f) and imaginary part (12e).

Equations (11) and (12) may be rewritten as the following Mellin convolution type transforms

$$x(u)=\begin{cases}\int_0^\infty F(\tau)/\tau\{\exp(-u/\tau)\}d\tau/\tau & \text{(a)}\\ \int_0^\infty F(\tau)\begin{Bmatrix}\exp(-u/\tau)\\ 1-\exp(-u/\tau)\end{Bmatrix}d\tau/\tau & \begin{matrix}\text{(b)}\\ \text{(c)}\end{matrix}\\ \int_0^\infty F(1/\tau)\begin{Bmatrix}1/(1+u^2/\tau^2)\\ u/\tau/(1+u^2/\tau^2)\\ u^2/\tau^2/(1+u^2/\tau^2)\end{Bmatrix}d\tau/\tau & \begin{matrix}\text{(d)}\\ \text{(e)}\\ \text{(f)}\end{matrix}\end{cases} \qquad (13)$$

where the curly braces {} contain Mellin convolution kernels $k(u)$, i.e. kernels $K(u,\tau)$ modified so to give the Mellin convolution (3). In the light of the functional filtering, (13) represent ideal deconvolution filters or ideal DRRT estimators having, according to (9), the following frequency responses [13] – [16]:

$$H(j\mu)=\begin{cases}-1/\Gamma(-j\mu) & \text{for (12a)} - \text{(12c)} & \text{(a)}\\ \pm 2\sin(j\pi\mu/2)/\pi & \text{for (12d) and (12f)} & \text{(b)}\\ 2\cos(j\pi\mu/2)/\pi & \text{for (12e)} & \text{(c)}\end{cases} \qquad (14)$$

The fact that determination of DRRT from the functions described by six kernels (12) leads to three ideal filters with frequency responses (14) has the important practical consequences, such as:

(i) only three independent impulse responses or sets of filter coefficients corresponding to (14) are necessary for DRRT recovery from the functions described by six kernels (12),

(ii) the same coefficients may be used for all the time-domain functions described by kernels (12a) – (12c), and

(iii) the coefficients for the real parts of frequency-domain functions with kernels (12d) and (12f) differ only by signs.

B. Discrete-time estimators

Since condition (10) is satisfied for all three frequency responses (14)

$$|H(j0)|=\begin{cases}0 & \text{for (14a) and (14b)} & \text{(a)}\\ 2/\pi & \text{for (14c)} & \text{(b)}\end{cases}$$

computationally realisable functional filters can be constructed for DRRT recovery from all the response functions described by kernels (12). However, only convolution transforms (13b) and (13c) relates directly to $F(\tau)$ as an output function allowing to use general algorithms (7). For other transforms, algorithms (7) shall be modified. Thus, transform (13a) relates to output function $F(\tau)/\tau$ requiring that the general algorithms modified into the form $F(\tau_m)=\tau_m y(\tau_m)$ are used. Likewise, transforms (13d) – (13f) relates to output function $F(1/\tau)$, for which the general algorithms must be modified into the form $F(\tau_m)=y(1/\tau_m)$. Therefore, DRRT recovery from the functions described by kernels (12) can be implemented by the following three algorithms:

$$F(u_0q^m)=\begin{cases}u_0q^m\sum_n h[n]x(u_0q^{m+k}) & \text{for (12a)} & \text{(a)}\\ \sum_n h[n]x(u_0q^{m+k}) & \text{for (12b) and (12c)} & \text{(b)}\\ \sum_n h[n]x(q^{k-m}/u_0) & \text{for (12d)} - \text{(12f)} & \text{(c)}\end{cases} \qquad (15)$$

where $k=-n$ for odd N and $k=-0.5-n$ for even N, and summation index n, depending on even or odd filter length, runs in accordance with (7).

C. DRRT recovering from the time-domain data

Frequency response (14a) of ideal filter for producing $F(\tau)$ from the time-domain data is a complex function of μ. From the symmetry property of the Fourier transform, it follows that the appropriate impulse response has no symmetry or the estimators recovering DRRT from the time-domain data belong *non-linear phase filters* [7].

D. DRRT recovering from the real parts

Frequency response (14b) of the ideal filter producing $F(\tau)$ from the real parts is a pure imaginary function

$$H(j\mu)=\pm 2\sin(j\pi\mu/2)/\pi=\pm j2sh(\pi\mu/2)/\pi,$$

with odd symmetry enforcing anti-symmetry properties also on the appropriate impulse responses [7]. Therefore, the estimators recovering DRRT from the real parts must be *type III linear phase filter* in the case of odd N and *type IV linear phase filter* in the case of even N.

E. DRRT recovering from the from the imaginary parts

Frequency response (14c) of the ideal filter producing $F(\tau)$ from the imaginary parts is a real function with even symmetry

$$H(j\mu) = 2\cos(j\pi\mu/2)/\pi = 2ch(\pi\mu/2)/\pi\,.$$

In this case, the estimators have the symmetric impulse responses and represent *type I linear phase filters* for odd *N* and *type II linear phase filters* for even *N* [7].

F. Design of DRRT estimators

The basic design problem for DRRT estimators is the necessity of limiting the sensitivity to noise to the levels acceptable for practice. Because all noise reduction schemes make worse the accuracy, a trade-off between the amount of noise suppression and the amount of signal distortion (accuracy) must be searched. In this study, the filters have been designed according to approach [12], [13], [16], which allows obtaining the estimators with the desired values of the noise amplification coefficients.

IV. Simulation Results

Simulations have been performed by two six-point FIR estimators of type IV [13], [14] operating at $q = 3.3$ and recovering the retardation spectrum from the real part of the frequency-domain compliance (kernel (12d)). The estimators carry out algorithm (15c), which, for the selected values of *N* and *q*, takes the form

$$F(u_0 q^m) = \sum_{n=-3}^{2} h[n]x(3.3^{-0.5-n-m}/u_0).$$

Coefficients $h[n]$ are given in Table 1, they ensure the noise amplification coefficients $S = 10.62$ (estimator 1) and $S = 2.28$ (estimator 2), respectively.

A. Simulations with noiseless data

In Fig. 1, the retardation spectra are compared recovered from the noiseless input data corresponding to the Cole-Cole (CC) model with different values of spectrum width parameter α. Estimator 1 recovers almost perfectly the retardation spectrum for parameters $\alpha = 0.5$ and $\alpha = 0.7$ (see Fig. 1a)). For $\alpha = 0.5$, coincidence with the exact spectrum is so good, that it is hard to distinguish them. However, estimator 1 generates the oscillating spectrum with non-physical negative values for the delta function retardation spectrum at α=1. Contrary, estimator 2 gives the non-oscillating spectrum at α=1 (see Fig. 1(b)), but at the expense of the worse recovery quality for the broader spectra.

TABLE I

Coefficients of the Estimators

n	*h*[*n*]	
	Estimator 1	Estimator 2
-3	-0.062 133	-0.033 296
-2	0.577 504	0.129 207
-1	-2.253 640	-1.058 800
0	2.253 640	1.058 800
1	-0.577 504	-0.129 207
2	0.062 133	0.033 296

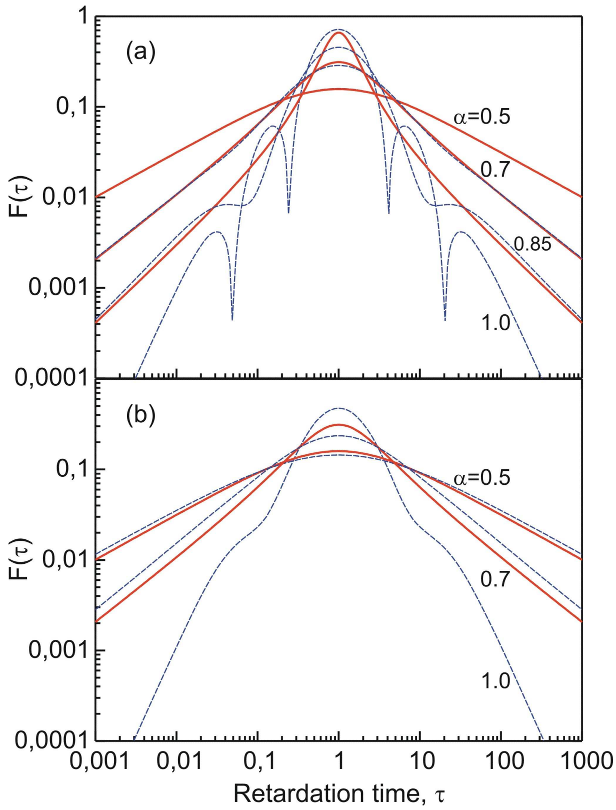

Fig. 1. The retardation spectra corresponding to CC model with different values of parameter α recovered by estimator 1 (a) and estimator 2 (b) from noiseless input data. Solid lines: exact spectra, dashed lines: recovered spectra. The exact spectrum for α=1 is the delta function (not shown).

B. Simulations with noisy data.

The effect of the noise and its potential reduction by smoothing have been investigated for noisy input data distorted by additive noise

$$x_{noisy}(\omega_m) = x_{exact}(\omega_m) + e\cdot n(m) \tag{16}$$

and multiplicative noise

$$x_{noisy}(\omega_m) = x_{exact}(\omega_m)[1 + e\cdot n(m)]\,, \tag{17}$$

where $n(m)$ is the normally distributed pseudorandom sequence within interval [-1,1] with zero mean, and *e* denotes the noise amplitude.

The noise curtails the intervals of the usable spectrum. It has been empirically estimated that, in the case of additive noise, DRRT can be obtained within the intervals of τ where

$$F(\tau) \ge F_{\min} = e\sqrt{S}/3\,. \tag{18}$$

Within these intervals, the noise can be effectively suppressed by smoothing the input data, while outside the intervals the noise effect is dominant and the spectrum is lost.

The effect of additive random noise (16) and smoothing is demonstrated in Fig. 2, where the retardation spectrum is shown recovered by the both estimators from the noisy the real

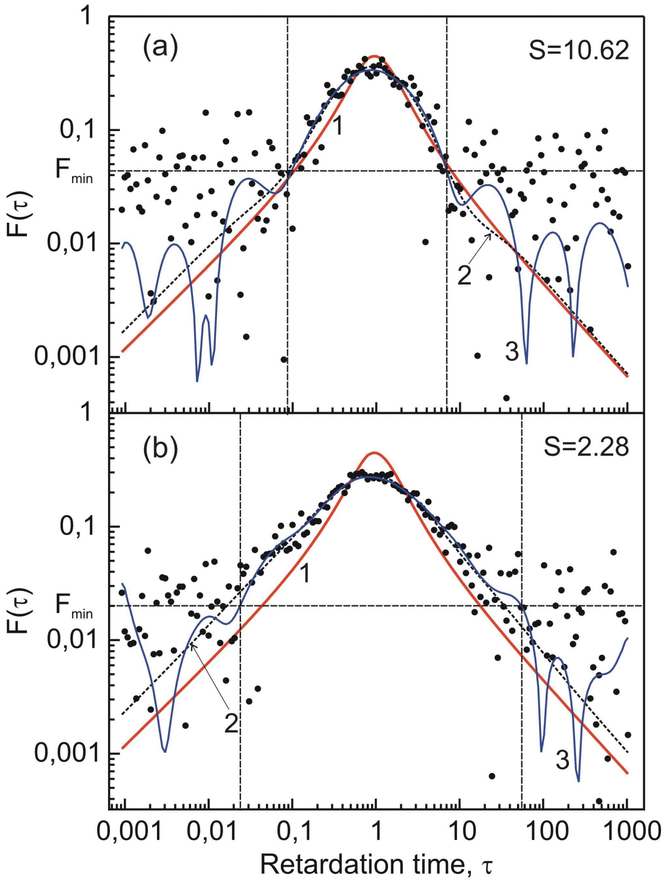

Fig. 2. Effect of additive random noise and smoothing on the retardation spectrum recovered by estimator 1 (a) and estimator 2 (b). Curves 1 – the exact spectrum; curves 2 – the spectra recovered from the noiseless input data; points – the spectra recovered from the noisy input data; curves 3 – the noisy spectra smoothed 10 times by (19). The vertical lines show intervals of the usable spectra according to criterion (18).

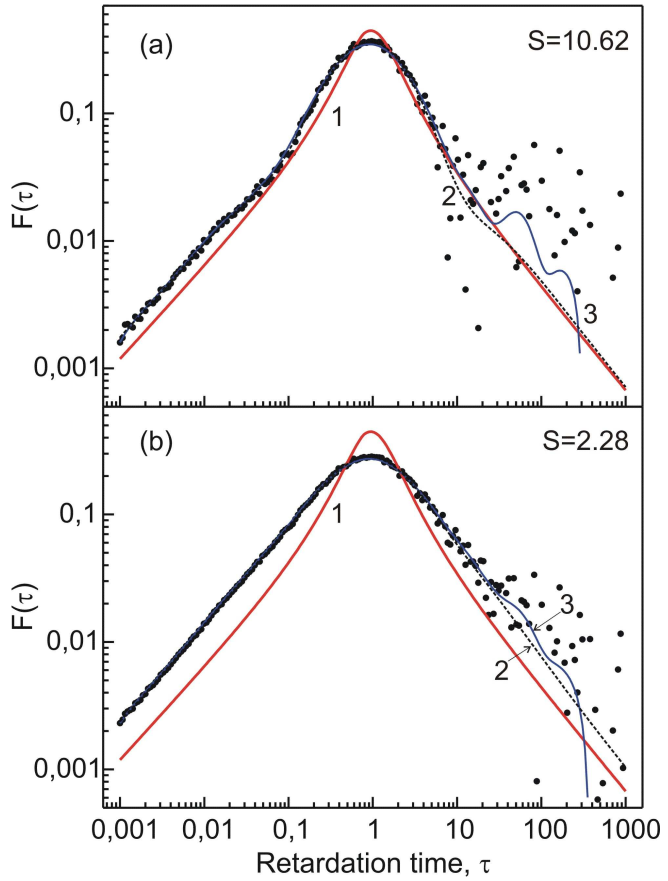

Fig. 3. Effect of multiplicative random noise and smoothing on the retardation spectrum recovered by estimator 1 (a) and estimator 2 (b). Numbering of curves – as in Fig. 2.

part corresponding to CC model with parameter $\alpha = 0.8$. Here, amplitude $e = 0.05$ has been used and the recovered noisy spectra have been smoothed by simple 5-point averaging

$$\overline{F}(\tau_m) = \frac{1}{5}\sum_{n=-2}^{2} F(\tau_{m+n}) . \quad (19)$$

The vertical lines show the usable intervals of the recovered DRRT estimated according to criterion (18). As seen, within these intervals, there is the good agreement between the recovered spectra from the noiseless data (curves 2) and the smoothed spectra (curves 3).

In Fig. 3, the same recovery situation is shown from noisy input data distorted by multiplicative noise (17) also with $e = 0.05$. In this case, the usable spectra are curtailed only at the large relaxation times. Again, positive effect of smoothing is demonstrated.

V. Conclusions

It is demonstrated that the problem of determination of the distribution of relaxation/retardation times (DRRT) from various response functions leads to three deconvolution tasks on the logarithmic time or frequency scale related to recovery of DRRT from: (i) the time-domain (impulse and step) responses, (ii) the real parts and (iii) the imaginary parts of the frequency responses. These deconvolution tasks are interpreted as ideal deconvolution (inverse) filters or ideal DRRT estimators and finite impulse response (FIR) deconvolution filters operating with geometrically sampled data are proposed for their implementation. It is demonstrated that the estimators recovering DRRT from the time-domain responses belong to non-linear phase filters, while linear-phase filters of type I or II must be used for DRRT recovery from the imaginary parts, and the filters of type III or IV – for DRRT recovery from the real parts. Three algorithms (with modifications for even or odd number of coefficients) are derived, which must be used for recovering DRRT from: (i) the impulse responses, (ii) the step responses, and (iii) the real and imaginary parts of the frequency responses. Simulation results are represented obtained by two estimators of type IV for DRRT recovery from the noiseless and noisy real parts distorted by additive and multiplicative noise.

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He has a wife Inta and a daughter Sanita. The hobby of Uldis Lomanovskis is football and motor race.

V. Štrauss, A. Kalpiņš, U. Lomanovskis. Dekonvolūcijas filtri relaksācijas un retardācijas laika sadalījuma noteikšanai
Raksts veltīts relaksācijas un retardācijas laika sadalījuma (RRLS) noteikšanai no dažādām reakcijas funkcijām laika un frekvences apgabalā. Parādīts, ka RRLS noteikšana reducējās uz trīs dekonvolūcijas uzdevumiem logaritmiskā laika vai frekvences mērogā, kuros RRLS tiek noteikts no: 1) laika apgabala reakcijas funkcijām (IPMulsa un pārejas raksturlīknēm), 2) frekvences raksturlīkņu reālām daļām un 3) frekvences raksturlīkņu imaginārām daļām. Šie dekonvolūcijas uzdevumi tiek interpretēti kā ideāli dekonvolūcijas filtri, kurus piedāvāts realizēt finitas IPMulsu raksturlīknes (FIR) dekonvolūcijas (inversu) filtru veidā, apstrādājot ģeometriski diskretizētus ieejas datus. Parādīts, ka dekonvolūcijas filtri RRLS noteikšanai no laika apgabala rekcijas funkcijām pieder nelineāras fāzes filtru klasei, bet lineāras fāzes I vai II tipa filtri jāizmanto RRLS noteikšanai no frekvences raksturlīkņu imaginārajām daļām, un 3) lineāras fāzes III vai IV tipa filtri – RRLS noteikšanai no frekvences raksturlīkņu reālām daļām. Atrasti trīs algoritmi (ar modifikācijām pāra un nepāra skaita koeficientiem) RRLS noteikšanai no: 1) IPMulsa raksturlīknēm, 2) pārejas raksturlīknēm, un 3) frekvences raksturlīkņu reālajām un imaginārajām daļām. Sniegti ar diviem IV tipa filtriem iegūti RRLS noteikšanas modelēšanas rezultāti no precīzām frekvences raksturlīkņu reālām daļām un reālām daļām, kas izkropļotas ar aditīvu un multiplikatīvu troksni.

В. Штраус, А. Калпиньш, У. Ломановскис. Фильтры обратной сверки для определения распределения времен релаксации и ретардации
Статья посвящена определению распределения времен релаксации и ретардации (РВРР) по различным функциям откликов во временной и частотной областях. Показано, что определение РВРР сводится к трем задачам обратной сверки в логарифмическом масштабе времени или частот, связанных с восстановлением РВРР по: 1) функциям откликов во временной области (импульсными и переходными характеристиками); 2) действительными частями частотных характеристик, и 3) мнимыми частями частотных характеристик. Эти задачи обратной сверки интерпретируются как идеальные фильтры обратной сверки. Для их осуществления предлагается использовать фильтры обратной сверки с конечной импульсной характеристикой (КИХ) работающие с геометрически дискретизированными данными. Показано, что фильтры, восстановляющие РВРР с функций откликов во временной области принадлежат фильтрам с нелинейной фазой, в то время как фильтры с линейной фазой типа I или II должны быть использованы для восстановления РВРР с мнимых частей частотных характеристик, и фильтры с линейной фазой типа III или IV – для восстановления РВРР с вещественных частей частотных характеристик. Получены три алгоритма (с версиями для четного и нечетного числа коэффициентов) применяемые для восстановления РВРР по: 1) импульсными характеристиками, 2) переходными характеристиками, и 3) действительными и мнимыми частями частотных характеристик. Представлены результаты моделирования восстановления РВРР полученные двумя фильтрами типа IV с бесшумных вещественных частей и шумных вещественных частей, искаженных аддитивным и мультипликативным шумом.