ON THE EXISTENCE AND UNIQUENESS OF THE RELAXATION SPECTRUM OF VISCOELASTIC MATERIALS
PART I: THE MAIN THEOREM

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Summary. Over the last 30 years many advances have been made which proved that the relaxation spectrum is a very convenient characteristic quantity describing the properties of the linear viscoelastic materials, especially of polymer solutions and polymer melts as well as soft biological materials. Given the spectrum it is very easy to convert one mechanical material function used in engineering calculations into another one, such as the relaxation modulus or the creep compliance, the constant and time-variable bulk and shear modulus or Poisson’s ratio. The relaxation spectrum can be also used to validate experiments by cross-checking results, e.g., from creep and stress relaxation test. In this paper the main necessary and sufficient condition of the existence and uniqueness of the relaxation spectrum of viscoelastic material is given using fundamental for such materials concept of fading memory and based on the notion of completely monotone functions. Other relaxation spectrum existence conditions are given in the second part of the paper.

Keywords: viscoelasticity, relaxation modulus, relaxation spectrum, completely monotone function, existence and uniqueness

INTRODUCTION

Inferring models of the materials from observations and studying their properties is really the important stage of modern engineering design and the predilection to the synthesis of the production process control systems. Rigorous predictions of the materials behaviour in different conditions, especially under different loading, is essential both for the optimal design of the machines and vehicles operation and the assurance of the structural integrity of buildings, as well as for process of system maintenance, in particular to guarantee maintenance safety. Among all the materials, for which the linear and isotropic properties hypothesis is quite enough for a lot of engineering purposes within a small deformations, significant is viscoelastic materials for which energy dissipation occurs in a result of “internal friction” between, for example, polymeric molecules or cells of plant materials. Viscoelastic models are used before all to modelling of different polymeric liquids and solids [Elder et al. 1991, Brabec et al. 1997], concrete [Desai and Zienk 1968], soils [Lemaître 2001]. Research studies conducted during the past few decades proved that those models are also an important tool for studying the behaviour of viscoelastic plant materials (wood [Lemaître 2001], fruits, vegetables
Rheological models which are good for characterizing strain-stress dependences, creep and stress relaxation within a small deformation are applied for rigorous predictions of the plant materials behavior in an accurate engineering methods used for food processing machines, harvest and storage engines as packing and granulating engine design.

The mechanical properties of linear viscoelastic materials are fully characterized by relaxation or retardation spectra [Christensen 1971, Brabec et al. 1997, Andersen and Loy 2002]. The spectra are vital not only for constitutive models but also for the insight into the properties of a viscoelastic material since from the relaxation or retardation spectra other linear material functions can be calculated without difficulty [Brabec et al. 1997].

The purpose of this paper is to give the necessary and sufficient conditions, which guarantee the existence and uniqueness of the relaxation spectrum of linear viscoelastic materials. Relaxation modulus is said to have “fading memory”, if changes in the past have less effect now than equivalent more recent changes. A brief review of this fundamental for viscoelastic materials concept is contained. Next, the Boltzmann relaxation moduli are constrained to be a completely monotone function, what gives considerable mathematical machinery. Some resulting theoretical implications are discussed. The necessary and sufficient condition of the existence and uniqueness of nonnegative integrable relaxation spectrum is given. To make the idea of relationship between the fading memory of relaxation modulus and relaxation spectrum properties a little clear, we give several examples.

LINEAR VISCOELASTIC MATERIALS

The uniaxial and isotropic stress-strain equation for a linear viscoelastic material subjected to small deformations can be represented by a constitutive integral equation [Christensen 1971]:

\[ \sigma(t) = \int_{-\infty}^{t} G(t - \lambda) \dot{\epsilon} (\lambda) d\lambda, \]  

(1)

which is based on the Boltzmann superposition principle. Here, \( \sigma(t) \) denotes the stress corresponding to given strain rate \( \dot{\epsilon}(t) \) and \( G(t) \), \( t \geq 0 \), is the linear (Boltzmann) relaxation modulus. The modulus \( G(t) \) or uniaxial relaxation function [Derek and Zembek 1963] equivalently, is the stress, which is induced in the viscoelastic material described by eq. (1) when the unit step strain \( \dot{\epsilon}(t) \) is imposed. In the result, \( G(t) \geq 0 \) for any \( t \geq 0 \). The isothermal conditions are assumed and only the states of uniaxial stress and strain are considered here. The equation (1) describes how, in each materials, the stress \( \sigma(t) \), at time \( t \), depends not only on the strain \( \dot{\epsilon}(t) \) at time \( t \), but also on its earlier history of the strain, i.e. on the strain rate \( \dot{\epsilon} \). This is the essence of the memory of viscoelastic materials.

CONCEPT OF FADING MEMORY

The properties of linear viscoelastic material, in particular the kind its memory, depends on the kernel of eq. (1), i.e. on the form and structure of the relaxation modulus \( G(t) \). Appropriate conditions must be imposed on the Boltzmann modulus in order to guarantee that the constitutive relationship (1) makes sense physically. In the first place, \( G(t) \) must be such that the stress \( \sigma \) at the time \( t \) depends on the time derivative \( \dot{\epsilon}(t) \) of the strain history for \( t \geq t_s \), but do not depend on the strain in the future. This causality requirement is already included in the convolution equation (1) in which the time \( t \) is the upper limit in the integral. Formally, the relaxation modulus is causal iff (throughout, if and only if) \( G(t) = 0 \) for any \( t < 0 \).
The next, in the modelling of linear viscous elastic materials is a fundamental concept of fading memory of the relaxation modulus \( G(t) \) that back to Boltzmann [1876]. When this regularity requirement is imposed on \( G(t) \) the changes in the strain rate in the distant past must have no effect now that the same changes in the more recent past. Thus, by virtue of the structure of the convolution equation (1) it is easy to see that the fading memory relaxation modulus \( G(t) \) is monotonically decreasing function (in the strict non-increasing), and in the result \( dG(t)/dt \leq 0 \) for any \( t > 0 \). Obviously, this is only necessary, but not sufficient conditions for the relaxation modulus \( G(t) \) to have fading memory, so much that as clear from the literature, there is no universal and unique definition of fading memory. A review of the concept of fading memory can be found in work of Andriessen and Loy [2002]. A wide variety of views are summarized as follows.

In purely mechanistic approach, it is assumed that for the fading memory material two conditions the nonnegative definiteness of the relaxation modulus \( G(t) \geq 0 \) and the weak dissipation principle are satisfied. This approach examined in details in [Havryly [2005], naturally imply the requirement that the modulus \( G(t) \) is strongly positive definite function. The above is satisfied if, for example, \( G(t) \) is nonnegative non-increasing convex function for \( t > 0 \), i.e. the following three conditions are satisfied:

\[
G'(t) \geq 0, \quad G''(t) \geq 0, \quad G'''(t) \geq 0 \quad \text{for} \quad t > 0. \tag{2}
\]

which are given for example in [Galvich et al. 2004].

The rheological approach is based on the assumption that the Boltzmann relaxation modulus \( G(t) \) and the molecular weight distribution are related by some integral rules. This approach is discussed in details in Cocchini and Nobile [2003], in this work the next references can be found.

The systems science approach is the point of view, which is supported most strongly by the mathematical advantages of the linear dynamical systems theory. It is assumed that the response of fading memory behaviour material can be modelled by a monotony decreasing and suitable smooth function \( G(t) \). The most restrictive assumption is that \( G \in L^2(0, \infty) \cap L^1(0, \infty) \) [Fabrizio and Moro 1992]. A comprehensive summary of this approach can be found in work by Fabrizio and Moro [1992].

The concept of complete monotonicity of the Boltzmann relaxation modulus which is adopted in this paper and is described in the next section.

Both the purely mechanistic approach which yields the conditions (2) as well as the systems science approach are not sufficient to draw a distinction viscous elastic materials from other materials. This, in turn, means that the definitions of fading memory of terms of physics principles or systems sciences categories are unnecessary [Andriessen and Loy 2002]. Unlike the purely mechanistic or systems science approaches to fading memory, the rheological approach allows the possibility of distinguishing between linear viscous elastic materials and, in particular, between polymers to which this approach is addressed and other materials.

**THE CONCEPT OF COMPLETELY MONOTONIC BOLTZMANN MODULUS**

The strongest assumption about fading memory appears to date back to the seventies of XX century and was introduced by Day [1972]. Day's choice, for both practical and theoretical reasons, was to define \( G(t) \) to have fading memory if it is completely monotone, i.e. if the following conditions are satisfied (see Appendix A):

\[
(-1)^n \frac{d^n G(t)}{dt^n} > 0 \quad \text{for} \quad n > 0 \quad \text{and} \quad t > 0, \tag{3}
\]
The properties of the systems and materials of completely monotone fading memory are widely discussed in work of Cobae and Mroz [1967].

Remark 1. The relaxation modulus $G(t)$ of fading memory for which conditions (3) are satisfied may be singular for $t = 0$.

Remark 2 [Hanyga 2005]. For completely monotonic relaxation modulus $G(t)$ there exists the limit:

$$\lim_{t \to \infty} G(t) - G_{\infty} = 0. \quad (4)$$

In the formula (4) $G_{\infty}$ is the long-term modulus. Modulus $G_{\infty} = 0$ for solids and $G_{\infty} = \bar{G}$ for liquid materials.

Remark 3. If completely monotonic relaxation modulus is such that $G \in L^2(0, \infty)$ or $G \in L^1(0, \infty)$ then $G(t) < \infty$ and the limit $\lim_{t \to \infty} G(t) = G_{\infty} = 0$.

To make the idea a little clear we give the following example.

Example 1. An infinite Derjaguin-Polya [Zi and Baczant 2002] series is chosen as the relaxation modulus:

$$G(t) = \sum_{i=1}^{\infty} \frac{E_i}{\nu_i} e^{-\nu_i t} + E_{\infty} \quad (5)$$

where the parameters $E_i \geq 0$, $\nu_i \geq 0$ and $E_{\infty} \geq 0$. The structure of such generalized discrete Maxwell model (5) representing well the linear viscoelastic materials in most cases is given on Figure 1. The elastic modules $E_i$ and the partial viscosity $\eta_i$ associated with the $j$-th Maxwell mode determine the relaxation frequencies $\nu_i = E_i / \eta_i$. For physically realistic materials these parameters must be positive. The parameters $E_j$ and $\nu_j$ completely represent the viscoelastic spectrum of the material (see example 3 in the second part of the paper). It is easily seen that for the relaxation modulus (5) conditions (2) are satisfied, thus $G(t)$ is strongly positive definite function. It is also easy to check the next remark.

Remark 4. If the parameters $E_i \geq 0$ and $\nu_i \geq 0$, then the relaxation modulus (5) is completely monotonic function. If additionally $E_{\infty} = 0$, then $G \in L^1(0, \infty) \cap L^2(0, \infty)$, as well as $G \in H^1(0, \infty)$ for any $0 < \nu \leq \infty$. In this case we have exponential fading memory.

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**Fig. 1.** Generalized discrete Maxwell model with additional elastic element $E_i$. 

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RELAXATION SPECTRUM

In the rheological literature it is commonly assumed that the modulus $G(t)$ has the following relaxation spectrum representation [Dresner and Ziembia 1988, Christensen 1971],

$$G(t) = \int_0^\infty H(v)\nu^\alpha d\nu,$$  \hspace{1cm} (6)

where a nonnegative relaxation spectrum $H(v)$ characterizes the distribution of relaxation frequencies $v > 0$ in the range $[0, v_{\text{max}}]$. Equation (6) yields a formal definition of a relaxation spectrum [Christensen 1971, Rao 1999, Andersen and Loy 2002].

The spectrum representation of eq. (6) guarantees, in particular, that the relaxation modulus $G(t)$ is a monotonically decreasing function on $[0, \infty)$ and infinitely differentiable function on $(0, \infty)$, i.e., function of $C^n([0, \infty)$ class. However, the obvious necessary conditions for the relaxation spectrum existence are not the sufficient one. In the next section the relaxation spectrum necessary and sufficient existence condition is given based on the notion of complete monotonic functions.

RELAXATION SPECTRUM EXISTENCE AND UNIQUENESS

It is easy to verify that, if the eq. (6) is satisfied then

$$(-1)^n \frac{d^nG(t)}{dt^n} = (-1)^n \int_0^\infty H(v)v^{n+\alpha} d\nu.$$  \hspace{1cm} (7)

Thus, the next remark is not surprising.

**Remark 5.** If nonnegative spectrum of relaxation frequencies $H(v)$ defined by the eq. (6) there exists, than the relaxation modulus $G(t)$ is a completely monotonic function.

From the above remark it follows that for the spectral representation (6) the complete monotonic fading memory of the Boltzmann modulus is established. What is more, the complete monotonicity of $G(t)$ is not only the necessary but also the sufficient conditions of the relaxation spectrum existence of linear viscoelastic modulus. Clearly, in view of the Hausdorff-Bernstein-Widder theorem of complete monotonic functions (see Appendix A), for any real completely monotonic function $G(t)$ defined on $(0, \infty)$ for which the condition $G(0^+) = \infty$ is satisfied, there exists nonnegative finite (Boyle) measure $\mu$ on $[0, \infty)$ such that

$$G(t) = \int_0^\infty \nu^\alpha d\mu(\nu).$$

Lying $d\mu(\nu) = H(v)d\nu$ on the basis of the Hausdorff-Bernstein-Widder theorem we can state the following condition of the integrable relaxation spectrum existence.

**Theorem 1.** Nonnegative integrable spectrum of relaxation frequencies $H(v)$ defined by the eq. (6) there exists iff the linear relaxation modulus $G(t)$ is completely monotonic function of fading memory and $G(0^+) = \infty$.

The condition that $G(0^+) = \infty$ is required to ensure the integrability of the relaxation spectrum. Using definition (6) it is easily seen that nonnegative spectrum $H(v)$ - if there exists - is integrable if the integral (6) is convergent for $t \to 0^+$, i.e. if and only if:

$$G(0^+) = \lim_{t \to 0^+} G(t) = \int_0^\infty H(v)\nu d\nu < \infty$$
The conditions of theorem 1 are obviously the necessary and sufficient conditions for non-negligible integrable function to be the inverse Laplace transform of non-negative real function. The uniqueness of the relaxation spectrum \( G(t) \) follows immediately from the invertibility of the Laplace transformation, therefore the following assertion can be formulated.

**Assertion 1.** If nonnegative relaxation spectrum \( H(v) \) of linear viscoelastic material there exists, then the spectrum is unique.

**Example 2.** Consider the relaxation modulus:

\[
G(t) = \frac{1}{(1 + a)^t}
\]

where the parameters \( a > 0 \) and \( p > 0 \). Since for any \( x \geq 0 \),

\[
(-1)^r \frac{d^r G(t)}{dt^r} = (-1)^r \int_0^\infty \frac{t^r}{(t + a)^{r+p}} dt > 0
\]

the relaxation modulus (7) is a completely monotonic function. On the other hand \( G(0^+) = 1/a^p \), thus on the basis of theorem 1 the integrable relaxation frequencies spectrum \( H(v) \) there exists and in view of assertion 1 is uniquely determined. It is easy to verify that spectrum corresponding to (7) has the form:

\[
H(v) = \frac{1}{\Gamma(p)} v^{p-1} e^{-av}
\]

where \( \Gamma(p) \) is classical Euler's gamma function. Relaxation spectrum \( H(v) \) (9) for two parameters \( a \) and for a few values of \( p \) is plotted on Figure 2.

![Figure 2](attachment:image.png)

**Example 3.** Consider the relaxation modulus:

\[
G(t) = t^{-1}
\]

Since in view of (3) \((-1)^r \frac{d^r G(t)}{dt^r} = (-1)^r v^r f^{r-1} > 0\), the modulus (10) is a completely monotonic function. On the other hand, the condition \( G(0^+) = \infty \) of theorem 1 is not satisfied here. On the basis of the definition formula (6) the relaxation spectrum \( H(v) = 1 \) for any relaxation frequency \( v \geq 0 \). This is an example of the relaxation modulus for which the corresponding relaxation spectrum is bounded but not integrable.
FINAL REMARKS

In the paper based on the properties of completely monotone functions the main necessary
and sufficient condition of the existence and uniqueness of nonnegative integrable relaxation spectrum
of linear viscoelastic materials is formulated. Other relaxation spectrum existence conditions
are given in the second part of the paper, where the conditions under which the square integrable
relaxation spectrum there exist are also derived. The last is important for the construction of the
scheme of the relaxation spectrum identification known in the literature, for example [Stanisliczcz
2004, 2005, 2009] and other works cited therein. The conditions of theorem 1, as well as all the
existence conditions presented in the second part of the paper refer to the Boltzmann relaxation
modulus, which is accessible in experiment.

APPENDIX A – COMPLETELY MONOTONE FUNCTIONS

Definition A. A function \( f: R \rightarrow R \), where \( R = (0, \infty) \), is said to be completely monotone
(monotone) on \( (0, \infty) \), if it belong to the class \( C^n([0, \infty)) \) and \( (-1)^n f^{(n)}(t) \geq 0 \) for any \( n \geq 0 \) and \( t > 0 \)
[Bochner, 1955, Girsberg et al. 1990].

Completely monotonic functions, also known as Bernstein functions, very important in selected
sections of functional analysis [Auja, 2000, Berg and Pedersen 2001], appear naturally in various
fields, like, for example, probability theory [Richters, 1985] and in technical sciences [Crim-
stein et al. 2000]. The main properties of these functions are given in [Widder, 1946]. We also refer
to contemporary work of Alzer and Berg [2002], where a detailed list of references on completely
monotonic functions can be found.

Hausdorff-Bernstein-Widder Theorem [Bernstein 1928, Widder 1946]. A function
\( f: R \rightarrow R \), of a class \( C^n([0, \infty)) \), is completely monotone if:

\[
f(t) = \int_0^\infty e^{-t s} d\mu(s), \quad t > 0, \tag{A.1}
\]

where \( \mu \) is nonnegative Borel measure on \([0, \infty)\) such that the integral (A.1) is convergent for
any \( t > 0 \). The measure \( \mu \) is finite on \( [0, \infty) \) if \( \int_0^\infty f(s) \, ds < \infty \).

The above theorem is known also as Bernstein theorem or Bernstein-Widder theorem.

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O ISTNIENIU I JEDNOZNAČNOSTI SPECTRUM RELAKSACJI
MATERIAŁÓW LEPKOSPRZĘŻYSTYCH

CZĘŚĆ 1: PODSTAWOWE TWIERDZENIE

Streszczenie. Badania prowadzone w ciągu ostatnich trzydziustu lat wykazały, że zgodnym narzędziem badania własności linowych materiałów lepkosprężystych jest spektrum relaksacji. Znajduje ono zastosowanie w analizie zjawisk relaksacyjnych i stażowych zachodzących w tych materiałach, w szczególności w polymerach, ale także materiałach pochodzenia biologicznego. Znając spektrum relaksacji można wyczyścić inne, powyższe stosowane w obliczeniach inżynieryjnych, charakterystyki materiałowe takie jak moduł relaksacji czy funkcja pełzenia, stałe i zmienne w czasie moduły odkształcenia podstawowego objętościowego oraz współczynnik Poissona. Jego znaczenie uwzględnia także weryfikację zgodności danych pochodzących z różnych eksperymentów na podstawie teorii szpadzenia krzywiznowego. W pracy wykorzystano fundamentalnego dla materiałów lepkosprężystych pojęcia zależności jasników i wykorzystując własności funkcji w pełni monotonicznych sformułowano podstawy w zakresie koniecznej i dodatkowo istnienia i jednoznaczności spektrum relaksacji materiałów linowych lepkosprężystych. Inne wielkości istnieją spektrum relaksacji podanych w części drugiej pracy.

Słowa kluczowe: lepkosprężistość, moduł relaksacji, spektrum relaksacji, funkcja w pełni monotoniczna, istnienie i jednoznaczność