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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 tests. 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 prelude 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 are 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 [Elster at al. 1991, Brabec at al. 1997], concrete [Derski and Ziemba 1968], soils [Lemaitre 2001]. Research studies conducted during the past few decades proved that these models are also an important tool for studying the behaviour of viscoelastic plant materials (wood [Lemaitre 2001], fruits, vegetables

[Rao 1999] and other papers cited therein). Rheological models which are good for characterizing strain-stress dependence, creep and stress relaxation within a small deformation are applied for rigorous predictions of the plant materials behaviour in an accurate engineering methods used for food processing machines, harvest and storage engines as packing and granulating engines design.

The mechanical properties of linear viscoelastic materials are fully characterized by relaxation or retardation spectra [Christensen 1971, Brabec at al. 1997, Anderssen 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 spectrum other linear material functions can be calculated without difficulty [Brabec at 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 rheological 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 izotropic stress-strain equation for a linear viscoelastic material subjected to small deformations can be represented by a constitutive integral equation [Christensen 1971]:

$$\sigma(t)=\int_{\infty}^{t} G(t-\lambda)\dot{\varepsilon}(\lambda)d\lambda, \tag{1}$$

which is based on the Boltzmann superposition principle. Here, $\sigma(t)$ denotes the stress corresponding to given strain rate $\dot{\varepsilon}(t)$ and $G(t)$, $t \geq 0$, is the linear (Boltzmann) relaxation modulus. The modulus $G(t)$ or uniaxial relaxation function [Derski and Ziemba 1968] equivalently, is the stress, which is induced in the viscoelastic material described by eq. (1) when the unit step strain $\varepsilon(t)$ 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 such materials, the stress $\sigma(t)$, at time t, depends not only on the strain $\varepsilon(t)$ at time t, but also on their earlier history of the strain, i.e. on the strain rate $\dot{\varepsilon}$. 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 σ at the time t depend on the time derivative $\dot{\varepsilon}(\tau)$ of the strain history for $\tau \leq t$, 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, iff = if and only if) $G(t)=0$ for any $t<0$.

The next, in the modelling of linear viscoelastic 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 less effect now than 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 Anderssen 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 [Hanyga 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:

$$G(t) \geq 0, -\frac{dG(t)}{dt} \geq 0, \frac{d^2G(t)}{dt^2} \geq 0 \;\; for \;\; t > 0, \tag{2}$$

which are given for example in [Galucio at 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 Morro 1992]. A comprehensive summery of this approach can be found in work by Fabrizio and Morro [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 viscoelastic materials from other materials. This, in turn, means that the definitions of fading memory in terms of physics principles or systems sciences categories are unnecessary [Anderssen and Loy 2002]. Unlike the purely mechanistic or systems sciences approaches to fading memory, the rheological approach allows the possibility of distinguishing between linear viscoelastic 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 data 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):

$$(-1)^n \frac{d^nG(t)}{dt^n} > 0 \;\; for \; n > 0 \; and \; t > 0, \tag{3}$$

The properties of the systems and materials of completely monotone fading memory are widely discussed in work of Colemanand and Mizel [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:*

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

In the formula (4) G_∞ is the *long-term modulus*. Modulus $G_\infty>0$ for solid and $G_\infty=0$ 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 $G(0)<\infty$ 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 Dirichlet-Prony [Zi and Bažant 2002] series is chosen as the relaxation modulus:

$$G(t) = \sum_{j=1}^{\infty} E_j e^{-t\nu_j} + E_\infty, \tag{5}$$

where: the parameters $E_j \geq 0$, $\nu_j \geq 0$ 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 modulus E_j and the partial viscosity η_j associated with the $j-th$ Maxwell mode determine the relaxation frequencies $\nu_j = E_j/\eta_j$. For physically realistic materials these parameters must be positive. The parameters E_j and ν_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 $E_j \geq 0$ and $\nu_j \geq 0$, then the relaxation modulus (5) is completely monotonic function. If additionally $E_\infty=0$, then $G \in L^2(0,\infty) \cap L^1(0,\infty)$, as well as $G \in L^p(0,\infty)$ for any $0<p\leq\infty$. In this case we have exponential fading memory.*

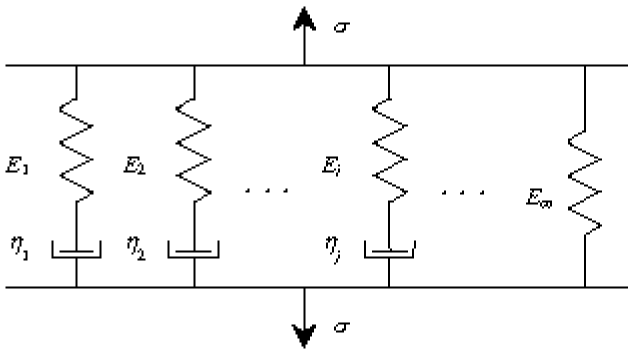

Fig. 1. Generalized discrete Maxwell model with additional elastic element E_∞

RELAXATION SPECTRUM

In the rheological literature it is commonly assumed that the modulus $G(t)$ has the following relaxation spectrum representation [Derski and Ziemba 1968, Christensen 1971]:

$$G(t) = \int_0^\infty H(\nu) e^{-t\nu} d\nu, \tag{6}$$

where a nonnegative relaxation spectrum $H(\nu)$ characterizes the distribution of relaxation frequencies $\nu \geq 0$ in the range $[\nu, \nu+d\nu]$. Equation (6) yields a formal definition of a relaxation spectrum [Christensen 1971, Rao 1999, Anderssen and Loy 2002].

The spectrum representation of eq. (6) guarantee, 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 $C^\infty(0,\infty)$ 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

$$(-1)^n \frac{d^n G(t)}{dt^n} = (-1)^{2n} \int_0^\infty H(\nu) \nu^n e^{-t\nu} d\nu.$$

Thus, the next remark is not surprising.

Remark 5. *If nonnegative spectrum of relaxation frequencies $H(\nu)$ defined by the eq. (6) there exists, then 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 (Borel) measure μ on $[0,\infty)$ such that:

$$G(t) = \int_0^\infty e^{-t\nu} d\mu(\nu).$$

Lying $d\mu(\nu) = H(\nu)d\nu$ 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 $H(\nu)$ 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 $H(\nu)$ - if there exists – is integrable if the integral (6) is convergent for $t \to 0^+$, i.e. if and only if:

$$G(0+) = \lim_{t\to 0^+} G(t) = \int_0^\infty H(\nu) d\nu < \infty.$$

The conditions of theorem 1 are obviously the necessary and sufficient conditions for nonnegative integrable function to be the inverse Laplace transform of nonnegative real function. The uniqueness of the relaxation spectrum $H(v)$ follows immediately from the invertiability 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:

$$G(t) = \frac{1}{(t+a)^p}, \tag{7}$$

where the parameters $a>0$ and $p>0$. Since for any $n\geq 0$,

$$(-1)^n \frac{d^n G(t)}{dt^n} = (-1)^{2n} \prod_{r=1}^{n} (r+p-1) \frac{1}{(t+a)^{p+n}} > 0, \tag{8}$$

the relaxation modulus (7) jest completely monotonic function. On the other hand $G(0+)=1/a^p<\infty$, 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:

$$H(v) = \frac{1}{\Gamma(p)} v^{p-1} e^{-av}, \tag{9}$$

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.

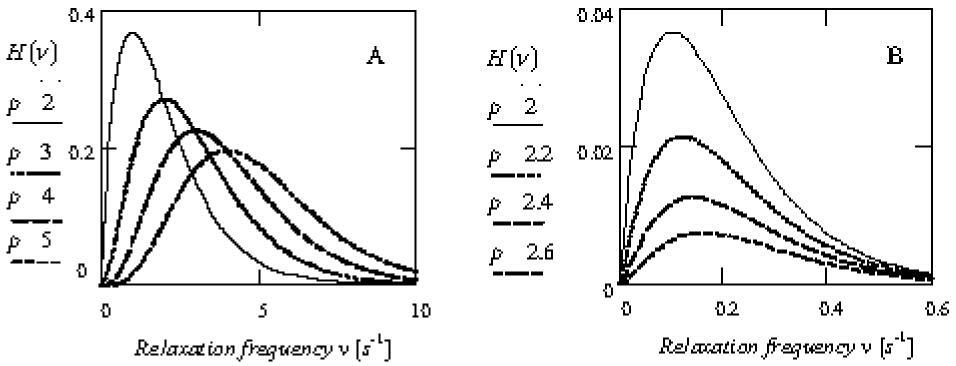

Fig. 2. Relaxation spectrum $H(v)$ (9) for parameters (*A*) $a=1$ and (*B*) $a=10$

Example 3. Consider the relaxation modulus:

$$G(t) = t^{-1}. \tag{10}$$

Since in view of (8) $(-1)^n d^n G(t)/dt^n = (-1)^{2n} n!/t^{n+1} > 0$, the modulus (10) is 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 monotonic 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 exists are also derived. The last is important for the construction of the schemes of the relaxation spectrum identification known in the literature, for example [Stankiewicz 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* $f: R_+ \to R_+$ *where* $R_+=(0,\infty)$*, is said to be completely monotonic (monotone) on* $(0,\infty)$*, if it belong to the class* $C^\infty(0,\infty)$ *and* $(-1)^n f^{(n)}(t) \geq 0$ *for any* $n \geq 0$ *and* $t > 0$ [Bochner 1955, Gripenberg at al. 1990].

Completely monotonic functions, also known as Bernstein functions, very important in selected sections of functional analysis [Aujla 2000, Berg and Pedersen 2001], appear naturally in various fields, like, for example, probability theory [Richards 1985] and in technical sciences [Grinshpan at 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 $f: R_+ \to R_+$ *of a class* $C^\infty(0,\infty)$ *is completely monotone iff*:

$$f(t) = \int_0^{+\infty} e^{-tx} d\mu(x),\ t > 0, \tag{A.1}$$

where μ *is nonnegative Borel measure on* $[0,\infty)$ *such that the integral* (A.1) *is convergent for any* $t>0$. *The measure* μ *is finite on* $[0,\infty)$ *iff* $f(0+)<+\infty$.

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

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O ISTNIENIU I JEDNOZNACZNOŚCI SPEKTRUM RELAKSACJI MATERIAŁÓW LEPKOSPRĘŻYSTYCH CZĘŚĆ I: PODSTAWOWE TWIERDZENIE

Streszczenie. Badania prowadzone w ciągu ostatnich trzydziestu lat wykazały, iż dogodnym narzędziem badania własności liniowych materiałów lepkosprężystych jest spektrum relaksacji. Znajduje ono zastosowanie w analizie zjawisk relaksacyjnych i retardacyjnych zachodzących w tych materiałach, w szczególności w polimerach, ale także materiałach pochodzenia biologicznego. Znając spektrum relaksacji można wyznaczyć inne, powszechnie stosowane w obliczeniach inżynierskich, charakterystyki materiałowe takie jak moduł relaksacji czy funkcja pełzania, stałe i zmienne w czasie moduły odkształcenia postaciowego i objętościowego oraz współczynnik Poissona. Jego znajomość umożliwia także weryfikację zgodności danych pochodzących z różnych eksperymentów na podstawie tzw. sprawdzania krzyżowego. W pracy wychodząc z fundamentalnego dla materiałów lepkosprężystych pojęcia zanikającej pamięci i wykorzystując własności funkcji w pełni monotonicznych sformułowano podstawowy warunek konieczny i dostateczny istnienia i jednoznaczności spektrum relaksacji materiału liniowo lepkosprężystego. Inne warunki istnienia spektrum relaksacji podano w części drugiej pracy.

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