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Exact asymptotic relations for the effective response of linear viscoelastic heterogeneous media

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Abstract

This article addresses the asymptotic response of viscoelastic heterogeneous media in the frequency domain, at high and low frequencies, for different types of elementary linear viscoelastic constituents. By resorting to stationary principles for complex viscoelasticity and adopting a classification of the viscoelastic behaviours based on the nature of their asymptotic regimes, either elastic or viscous, four exact relations are obtained on the overall viscoelastic complex moduli in each case. Two relations are related to the asymptotic uncoupled heterogeneous problems while the two remaining ones result from the viscoelastic coupling which manifests itself in the transient regime. These results also provide exact conditions on certain integrals in time of the effective relaxation spectrum. This general setting encompasses the results obtained in preceding studies on mixtures of Maxwell constituents [1, 2].

Keywords: viscoelasticity, composite, homogenization

1. Introduction

Homogeneous viscoelastic materials are characterized by a time-dependent response which can be expressed as a hereditary integral involving their relaxation or creep functions and a mechanical loading history. These constitutive functions describe asymptotically, at short and long times, elastic or viscous behaviours while the intermediate time response (i.e. transient regime) exhibits a viscoelastic character. The latter is contained in the relaxation and retardation spectra of the material. The effective response of a viscoelastic heterogeneous medium also presents these different regimes. For Kelvin-Voigt or Maxwell constituents, it has been demonstrated that the asymptotic effective properties are the homogenized tensors of the corresponding uncoupled elastic or viscous heterogeneous problem [3, 4]. Besides, it has been established that the overall constitutive response is solution of an integro-differential equation whereas the local response is described by a differential equation. This feature has been termed a “long memory” effect [5, 6, 3, 4]. In the case of a mixture of Maxwell materials, this implies a non vanishing retardation spectrum at the overall scale. Therefore, the effective creep response exhibits a transient regime, with a continuous decrease of the creep strain rate, whereas the local constitutive law

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does not exhibit a transient creep response. Similar features hold for the relaxation response of a mixture of Kelvin-Voigt materials.

In this context, exact relations have been more recently obtained on the overall relaxation and retardation spectra of heterogeneous materials made of Maxwell constituents [1, 2]. They supplement the classical ones previously derived on the asymptotic uncoupled regimes. These exact relations on the overall transient response involve a coupling between the local viscoelastic properties and the local fields which are solutions of the asymptotic heterogeneous problems. The present study aims at deriving similar results for any type of elementary viscoelastic constituents, assuming that the heterogeneous medium is made of elementary constituents of the same type. As with Maxwell constituents, these results can be further used to derive approximate homogenization models.

2. The different types of linear viscoelastic behaviours

According to the classical linear theory of viscoelasticity [7, 8, 9], the stress response $\boldsymbol{\sigma}(t)$ to a given derivable strain loading path $\boldsymbol{\varepsilon}(u)$, $u \in [0; t]$, with additional discontinuities (*i.e.* strain jumps) $[\boldsymbol{\varepsilon}]_i$ at times t_i and initial conditions $\boldsymbol{\sigma}(t=0) = \mathbf{0}$, reads

$$\boldsymbol{\sigma}(t) = \int_0^t \mathbf{L}(t-u) : \dot{\boldsymbol{\varepsilon}}(u) du + \sum_i \mathbf{L}(t-t_i) : [\boldsymbol{\varepsilon}]_i. \quad (1)$$

$\mathbf{L}(t)$ is the viscoelastic stiffness tensor (*i.e.* relaxation function) whose general form is

$$\mathbf{L}(t) = \mathbf{L}_{e_r} + \mathbf{L}_{v_g} \delta(t) + \int_0^{+\infty} \mathbf{G}(\tau) e^{-t/\tau} d\tau \quad (2)$$

with \mathbf{G} the relaxation spectrum. Besides, by reference to polymer materials, the viscous and elastic properties at short and long times (high and low frequencies) are respectively termed “glassy” (subindex g) and “relaxed” (subindex r). So, \mathbf{L}_{e_r} is the relaxed elastic stiffness while \mathbf{L}_{v_g} is the glassy viscous stiffness. Note also that $\delta(t)$ is the Dirac delta function. The stress response is therefore given by the time derivative of the convolution product of the functions \mathbf{L} and $\boldsymbol{\varepsilon}$. It is usually termed Stieltjes convolution product, by reference to the Stieltjes integral which generalizes the classical Riemann integral [10], and is noted \otimes in the sequel. The constitutive relation (1) can thus be written in a concise manner as

$$\boldsymbol{\sigma}(t) = \frac{d}{dt} (\mathbf{L} * \boldsymbol{\varepsilon})(t) = (\mathbf{L} \otimes \boldsymbol{\varepsilon})(t). \quad (3)$$

Similarly, the strain response $\boldsymbol{\varepsilon}(t)$ to a given derivable stress loading path $\boldsymbol{\sigma}(u)$, $u \in [0; t]$, with additional discontinuities (*i.e.* stress jumps) $[\boldsymbol{\sigma}]_i$ at times t_i and initial conditions $\boldsymbol{\varepsilon}(t=0) = \mathbf{0}$, reads

$$\boldsymbol{\varepsilon}(t) = (\mathbf{M} \otimes \boldsymbol{\sigma})(t) = \int_0^t \mathbf{M}(t-u) : \dot{\boldsymbol{\sigma}}(u) du + \sum_i \mathbf{M}(t-t_i) : [\boldsymbol{\sigma}]_i. \quad (4)$$

$\mathbf{M}(t)$ is the viscoelastic compliance tensor (*i.e.* creep function) whose general form is

$$\mathbf{M}(t) = \mathbf{M}_{e_g} + t \mathbf{M}_{v_r} + \int_0^{+\infty} \mathbf{J}(\tau) (1 - e^{-t/\tau}) d\tau \quad (5)$$

with \mathbf{M}_{e_g} the glassy elastic compliance, \mathbf{M}_{v_r} the relaxed viscous compliance and \mathbf{J} the retardation spectrum¹. The relaxation and retardation spectra characterize the viscoelastic transient response.

By considering the possible combinations of elastic or viscous asymptotic regimes, the linear viscoelastic behaviours can be classified into four categories [12] which are summarized in Table 1. The subsequent analysis is closely related to this classification. For later use, it can be noted that the Maxwell and Kelvin-Voigt models are described by two constitutive tensors, respectively elastic and viscous, whereas the Zener and anti-Zener models are described by three constitutive tensors (i.e. two elastic (resp. viscous) and one viscous (resp. elastic) tensors).

Type	\mathbf{M}_{e_g}	\mathbf{M}_{v_r}	\mathbf{L}_{e_r}	\mathbf{L}_{v_g}	Elementary constituent	Short time response	Long time response
I	> 0	0	> 0	0	Zener	Elastic	Elastic
II	> 0	> 0	0	0	Maxwell	Elastic	Viscous
III	0	0	> 0	> 0	Kelvin-Voigt	Viscous	Elastic
IV	0	> 0	0	> 0	anti-Zener	Viscous	Viscous

Table 1: The four types of linear viscoelastic response [12, 13]

3. Overall viscoelastic functions of heterogeneous media

Exact relations on the asymptotic responses of viscoelastic heterogeneous media with elementary constituents of the same type (Table 1) are derived in the following. These results encompass those obtained in [1, 2] for Maxwell constituents. By making use of stationary principles for complex constitutive behaviours [14], these relations are derived for the real and imaginary parts of the effective complex modulus, which are the quantities most commonly measured by dynamic mechanical analysis. Implications on the relaxation spectrum are also given. For conciseness, the corresponding relations for the overall complex compliance and the retardation spectrum are not reported here. They can be obtained in a similar manner by a dual analysis.

3.1. Description of a heterogeneous viscoelastic medium

The heterogeneous medium occupies a volume element Ω and comprises N different homogeneous phases of volume $\Omega^{(s)}$ with viscoelastic relaxation $\mathbf{L}^{(s)}(t)$ and creep $\mathbf{M}^{(s)}(t)$ functions, $s \in [1; N]$. Besides, it is assumed that $\Omega^{(s)} \ll \Omega$ and that the phases are perfectly bonded. The pointwise viscoelastic stiffness and compliance tensors thus read

$$\mathbf{L}(\mathbf{x}, t) = \sum_{s=1}^N \mathbf{L}^{(s)}(t) \chi^{(s)}(\mathbf{x}) \quad \text{and} \quad \mathbf{M}(\mathbf{x}, t) = \sum_{s=1}^N \mathbf{M}^{(s)}(t) \chi^{(s)}(\mathbf{x}) \quad (6)$$

with $\chi^{(s)}$ the characteristic function of phase (s) , that is

$$\chi^{(s)}(\mathbf{x}) = \begin{cases} 1 & \text{if } \mathbf{x} \in \Omega^{(s)}, \\ 0 & \text{otherwise.} \end{cases} \quad (7)$$

¹It is noted that the spectra \mathbf{G} and \mathbf{J} present several ranges of relaxation (resp. retardation) times which depend on their symmetry class [11, Appendix A]

$\mathbf{L}^{(s)}(t)$ and $\mathbf{M}^{(s)}(t)$ are inverse functions for the Stieltjes convolution product, that is $\mathbf{L}^{(s)} \circledast \mathbf{M}^{(s)} = \mathbf{I}$. The volume averages over Ω and $\Omega^{(s)}$ are respectively denoted $\bar{\bullet} = \langle \bullet \rangle$ and $\bar{\bullet}^{(s)} = \langle \bullet \rangle^{(s)}$. By definition of the characteristic function, the volume fraction of phase (s) is $c_s = \langle \chi^{(s)} \rangle$.

3.2. Local problems for general loading paths

The local problem to be solved in the volume element Ω subjected to a strain loading history $\bar{\boldsymbol{\varepsilon}}(t)$ from $t = 0$ to $t = T$ and classical boundary conditions (i.e. uniform or periodic) reads

$$\begin{cases} \boldsymbol{\sigma}(\mathbf{x}, t) = (\mathbf{L} \circledast \boldsymbol{\varepsilon})(\mathbf{x}, t), & \forall (\mathbf{x}, t) \in \Omega \times [0; T], \\ \operatorname{div} \boldsymbol{\sigma} = \mathbf{0}, \quad \operatorname{curl}({}^t \operatorname{curl} \boldsymbol{\varepsilon}) = \mathbf{0}, & \forall (\mathbf{x}, t) \in \Omega \times [0; T], \\ \langle \boldsymbol{\varepsilon}(t) \rangle = \bar{\boldsymbol{\varepsilon}}(t), & \forall t \in [0; T]. \end{cases} \quad (8)$$

Similarly, the local problem corresponding to a stress loading history $\bar{\boldsymbol{\sigma}}(t)$ reads

$$\begin{cases} \boldsymbol{\varepsilon}(\mathbf{x}, t) = (\mathbf{M} \circledast \boldsymbol{\sigma})(\mathbf{x}, t), & \forall (\mathbf{x}, t) \in \Omega \times [0; T], \\ \operatorname{div} \boldsymbol{\sigma} = \mathbf{0}, \quad \operatorname{curl}({}^t \operatorname{curl} \boldsymbol{\varepsilon}) = \mathbf{0}, & \forall (\mathbf{x}, t) \in \Omega \times [0; T], \\ \langle \boldsymbol{\sigma}(t) \rangle = \bar{\boldsymbol{\sigma}}(t), & \forall t \in [0; T]. \end{cases} \quad (9)$$

Depending on the applied loading history, the overall constitutive law is defined by

$$\bar{\boldsymbol{\sigma}}(t) = (\tilde{\mathbf{L}} \circledast \bar{\boldsymbol{\varepsilon}})(t) \quad \text{or} \quad \bar{\boldsymbol{\varepsilon}}(t) = (\tilde{\mathbf{M}} \circledast \bar{\boldsymbol{\sigma}})(t), \quad \forall t \in [0; T], \quad \text{with} \quad \tilde{\mathbf{L}} \circledast \tilde{\mathbf{M}} = \mathbf{I}. \quad (10)$$

3.3. Local problem for harmonic strain loadings

The response of a heterogeneous media to a sinusoidal loading is classically studied by making use of the Laplace-Carson (\mathcal{LC}) transform of the constitutive equations for a purely imaginary transform variable p [15]. For the particular case of an overall harmonic strain loading, that is $\bar{\boldsymbol{\varepsilon}}(t) = \bar{\boldsymbol{\varepsilon}}^* e^{i\omega t}$ (where $i = \sqrt{-1}$), the local problem corresponding to the steady-state regime at angular frequency ω reads

$$\begin{cases} \boldsymbol{\sigma}^*(\mathbf{x}, \omega) = \mathbf{L}^*(\mathbf{x}, \omega) : \boldsymbol{\varepsilon}^*(\mathbf{x}, \omega), & \forall \mathbf{x} \in \Omega, \\ \mathbf{L}^*(\mathbf{x}, \omega) = \mathcal{LC}(\mathbf{L}(\mathbf{x}, t)), & \forall \mathbf{x} \in \Omega, \\ \operatorname{div} \boldsymbol{\sigma}^* = \mathbf{0}, \quad \operatorname{curl}({}^t \operatorname{curl} \boldsymbol{\varepsilon}^*) = \mathbf{0}, & \forall \mathbf{x} \in \Omega, \\ \langle \boldsymbol{\varepsilon}^* \rangle = \bar{\boldsymbol{\varepsilon}}^*. \end{cases} \quad (11)$$

$\mathbf{L}^*(\mathbf{x}, \omega)$ is the \mathcal{LC} transform of the local relaxation function $\mathbf{L}(\mathbf{x}, t)$ with transform variable $p = i\omega$. It can be decomposed into

$$\mathbf{L}^*(\mathbf{x}, \omega) = \mathbf{L}'(\mathbf{x}, \omega) + i\mathbf{L}''(\mathbf{x}, \omega) \quad (12)$$

where \mathbf{L}' and \mathbf{L}'' are the storage and loss moduli which are respectively proportional to the stored and dissipated energies. The problem to be solved, for a given angular frequency ω , therefore corresponds to a symbolically heterogeneous elastic problem with pointwise complex fields $\boldsymbol{\varepsilon}^*$, $\boldsymbol{\sigma}^*$ and \mathbf{L}^* . The overall complex constitutive law can thus be expressed in the form

$$\bar{\boldsymbol{\sigma}}^* = \tilde{\mathbf{L}}^*(i\omega) : \bar{\boldsymbol{\varepsilon}}^* \quad \text{with} \quad \tilde{\mathbf{L}}^*(i\omega) = \tilde{\mathbf{L}}'(\omega) + i\tilde{\mathbf{L}}''(\omega). \quad (13)$$

Besides, it is worth noting that the asymptotic local fields (i.e. as $\omega \rightarrow +\infty$ and $\omega \rightarrow 0$) are solutions of the purely elastic or viscous heterogeneous problems corresponding to the glassy and relaxed regimes. It is recalled that the nature of the asymptotic states depends on the type of viscoelastic behaviour considered (Table 1). The local (complex) stress field satisfies

$$\lim_{\omega \rightarrow +\infty} \boldsymbol{\sigma}^*(\mathbf{x}, \omega) = \boldsymbol{\sigma}_g(\mathbf{x}) \quad \text{and} \quad \lim_{\omega \rightarrow 0} \boldsymbol{\sigma}^*(\mathbf{x}, \omega) = \boldsymbol{\sigma}_r(\mathbf{x}). \quad (14)$$

with $\boldsymbol{\sigma}_g$ and $\boldsymbol{\sigma}_r$ the (real) stress fields solution of the heterogeneous glassy and relaxed problems. The same asymptotic properties hold for the strain (rate) field $\boldsymbol{\varepsilon}^*(\mathbf{x}, \omega)$ with asymptotic fields $\boldsymbol{\varepsilon}_g(\mathbf{x})$ and $\boldsymbol{\varepsilon}_r(\mathbf{x})$ respectively.

3.4. Saddle-point variational principles for complex viscoelasticity

The complex constitutive relation (11)₁ can be rewritten as a system of real equations

$$\begin{pmatrix} \boldsymbol{\sigma}' \\ \boldsymbol{\sigma}'' \end{pmatrix} = \mathbb{L}_I : \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\varepsilon}' \end{pmatrix} \quad \text{with} \quad \mathbb{L}_I = \begin{pmatrix} -\mathbf{L}'' & \mathbf{L}' \\ \mathbf{L}' & \mathbf{L}'' \end{pmatrix} \quad (15)$$

where $(\boldsymbol{\sigma}', \boldsymbol{\varepsilon}')$ and $(\boldsymbol{\sigma}'', \boldsymbol{\varepsilon}'')$ are real fields. Observing that the quadratic form associated with \mathbb{L}_I is a saddle-shaped function, Cherkaev and Gibiansky [14] derived the following stationary principle for the real fields $\boldsymbol{\varepsilon}'(\mathbf{x}, \omega)$ and $\boldsymbol{\varepsilon}''(\mathbf{x}, \omega)$

$$\begin{pmatrix} \bar{\boldsymbol{\varepsilon}}'' \\ \bar{\boldsymbol{\varepsilon}}' \end{pmatrix} : \begin{pmatrix} -\tilde{\mathbf{L}}'' & \tilde{\mathbf{L}}' \\ \tilde{\mathbf{L}}' & \tilde{\mathbf{L}}'' \end{pmatrix} : \begin{pmatrix} \bar{\boldsymbol{\varepsilon}}'' \\ \bar{\boldsymbol{\varepsilon}}' \end{pmatrix} = \min_{\boldsymbol{\varepsilon}', \langle \boldsymbol{\varepsilon}' \rangle = \bar{\boldsymbol{\varepsilon}}'} \max_{\boldsymbol{\varepsilon}'', \langle \boldsymbol{\varepsilon}'' \rangle = \bar{\boldsymbol{\varepsilon}}''} \left\langle \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\varepsilon}' \end{pmatrix} : \begin{pmatrix} -\mathbf{L}'' & \mathbf{L}' \\ \mathbf{L}' & \mathbf{L}'' \end{pmatrix} : \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\varepsilon}' \end{pmatrix} \right\rangle. \quad (16)$$

The left-hand side of (16) is the imaginary part of the overall “complex energy” $\phi^* = \bar{\boldsymbol{\sigma}}^* : \bar{\boldsymbol{\varepsilon}}^*$, that is

$$\text{Im}(\phi^*) = \text{Im}(\bar{\boldsymbol{\sigma}}^* : \bar{\boldsymbol{\varepsilon}}^*) = \langle \boldsymbol{\sigma}'' : \boldsymbol{\varepsilon}' + \boldsymbol{\sigma}' : \boldsymbol{\varepsilon}'' \rangle. \quad (17)$$

By considering an alternative rewriting of the complex constitutive law (11)₁

$$\begin{pmatrix} \boldsymbol{\sigma}' \\ -\boldsymbol{\sigma}'' \end{pmatrix} = \mathbb{L}_R : \begin{pmatrix} \boldsymbol{\varepsilon}' \\ \boldsymbol{\varepsilon}'' \end{pmatrix} \quad \text{with} \quad \mathbb{L}_R = \begin{pmatrix} \mathbf{L}' & -\mathbf{L}'' \\ -\mathbf{L}'' & -\mathbf{L}' \end{pmatrix} \quad (18)$$

another saddle-point variational principle can be established

$$\begin{pmatrix} \bar{\boldsymbol{\varepsilon}}' \\ \bar{\boldsymbol{\varepsilon}}'' \end{pmatrix} : \begin{pmatrix} \tilde{\mathbf{L}}' & -\tilde{\mathbf{L}}'' \\ -\tilde{\mathbf{L}}'' & -\tilde{\mathbf{L}}' \end{pmatrix} : \begin{pmatrix} \bar{\boldsymbol{\varepsilon}}' \\ \bar{\boldsymbol{\varepsilon}}'' \end{pmatrix} = \min_{\boldsymbol{\varepsilon}', \langle \boldsymbol{\varepsilon}' \rangle = \bar{\boldsymbol{\varepsilon}}'} \max_{\boldsymbol{\varepsilon}'', \langle \boldsymbol{\varepsilon}'' \rangle = \bar{\boldsymbol{\varepsilon}}''} \left\langle \begin{pmatrix} \boldsymbol{\varepsilon}' \\ \boldsymbol{\varepsilon}'' \end{pmatrix} : \begin{pmatrix} \mathbf{L}' & -\mathbf{L}'' \\ -\mathbf{L}'' & -\mathbf{L}' \end{pmatrix} : \begin{pmatrix} \boldsymbol{\varepsilon}' \\ \boldsymbol{\varepsilon}'' \end{pmatrix} \right\rangle \quad (19)$$

which is the real part of the overall “complex energy” ϕ^* , that is

$$\text{Re}(\phi^*) = \text{Re}(\bar{\boldsymbol{\sigma}}^* : \bar{\boldsymbol{\varepsilon}}^*) = \langle \boldsymbol{\sigma}' : \boldsymbol{\varepsilon}' - \boldsymbol{\sigma}'' : \boldsymbol{\varepsilon}'' \rangle. \quad (20)$$

From the definition of ϕ^* and a lemma on the derivative of the stationary value of an energy [16], it follows that

$$\frac{\partial}{\partial \omega} (\text{Re}(\phi^*)) = \left\langle \begin{pmatrix} \boldsymbol{\varepsilon}' \\ \boldsymbol{\varepsilon}'' \end{pmatrix} : \frac{\partial \mathbb{L}_R}{\partial \omega} : \begin{pmatrix} \boldsymbol{\varepsilon}' \\ \boldsymbol{\varepsilon}'' \end{pmatrix} \right\rangle \quad \text{and} \quad \frac{\partial}{\partial \omega} (\text{Im}(\phi^*)) = \left\langle \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\varepsilon}' \end{pmatrix} : \frac{\partial \mathbb{L}_I}{\partial \omega} : \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\varepsilon}' \end{pmatrix} \right\rangle. \quad (21)$$

We can build on the two stationary principles (16) and (19) and their derivatives with respect to the angular frequency ω (21) to obtain exact asymptotic relations on the effective complex modulus of heterogeneous viscoelastic media at low and high frequencies. These relations are investigated in the sequel for the different cases of elementary viscoelastic constituents (Table 1).

3.5. Mixture of Zener constituents (Type I viscoelasticity)

3.5.1. Local and effective viscoelastic properties

The behaviour of a Zener constituent (a.k.a. standard linear solid model) is characterized by elastic responses in the asymptotic regimes (\mathbf{L}_{e_r} , \mathbf{M}_{e_g}) and presents a unique ‘‘transient’’ viscous stiffness tensor (\mathbf{L}_v). Its constitutive relation is solution of a homogeneous differential equation which reads

$$\boldsymbol{\sigma}(t) + \mathbf{L}_v : (\mathbf{L}_{e_g} - \mathbf{L}_{e_r})^{-1} : \dot{\boldsymbol{\sigma}}(t) = \mathbf{L}_{e_r} : \boldsymbol{\varepsilon}(t) + \mathbf{L}_{e_g} : \mathbf{L}_v : (\mathbf{L}_{e_g} - \mathbf{L}_{e_r})^{-1} : \dot{\boldsymbol{\varepsilon}}(t). \quad (22)$$

The viscoelastic stiffness and compliance tensors of the Zener phase (s) are

$$\mathbf{L}^{(s)}(t) = \mathbf{L}_{e_r}^{(s)} + \mathbf{G}^{(s)} e^{-t/\tau^{(s)}} \quad \text{and} \quad \mathbf{M}^{(s)}(t) = \mathbf{M}_{e_g}^{(s)} + \mathbf{J}^{(s)} \left(1 - e^{-t/\tau^{(s)}}\right) \quad (23)$$

with $\tau^{(s)}$ and $\tau'^{(s)}$ the relaxation and retardation times. It can be noted that the eigenvalues of $\mathbf{L}_v^{(s)} : (\mathbf{L}_{e_g}^{(s)} - \mathbf{L}_{e_r}^{(s)})^{-1}$ corresponds to the relaxation times of the Zener constituent (s). Also, the asymptotic elastic properties tensors obey the following relations

$$\mathbf{L}_{e_r}^{(s)} = (\mathbf{M}_{e_g}^{(s)} + \mathbf{J}^{(s)})^{-1} \quad \text{and} \quad \mathbf{M}_{e_g}^{(s)} = (\mathbf{L}_{e_r}^{(s)} + \mathbf{G}^{(s)})^{-1}. \quad (24)$$

The general expressions of the effective relaxation and creep functions, consistent with the elastic asymptotic regimes, are

$$\tilde{\mathbf{L}}(t) = \tilde{\mathbf{L}}_{e_r} + \int_0^{+\infty} \tilde{\mathbf{G}}(\tau) e^{-t/\tau} d\tau \quad \text{and} \quad \tilde{\mathbf{M}}(t) = \tilde{\mathbf{M}}_{e_g} + \int_0^{+\infty} \tilde{\mathbf{J}}(\tau) \left(1 - e^{-t/\tau}\right) d\tau \quad (25)$$

and the effective response to strain harmonic loadings with angular frequency ω is described by the complex viscoelastic stiffness tensor $\tilde{\mathbf{L}}^*(i\omega)$ given by

$$\tilde{\mathbf{L}}^*(i\omega) = \mathcal{L}\mathcal{C}(\tilde{\mathbf{L}}(t)) = \tilde{\mathbf{L}}_{e_r} + \int_0^{+\infty} \frac{i\omega\tau}{1 + i\omega\tau} \tilde{\mathbf{G}}(\tau) d\tau. \quad (26)$$

3.5.2. Exact relations on the overall complex modulus and relaxation spectrum

From the stationary principles on the complex energy ϕ^* and the asymptotic properties of the strain field $\boldsymbol{\varepsilon}^*$, it follows that

$$\left\{ \begin{array}{l} \lim_{\omega \rightarrow +\infty} \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}'(\omega) : \bar{\boldsymbol{\varepsilon}} = \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}_{e_g} : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_{e_g}^{(s)} :: \langle \boldsymbol{\varepsilon}_g \otimes \boldsymbol{\varepsilon}_g \rangle^{(s)}, \\ \lim_{\omega \rightarrow 0} \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}'(\omega) : \bar{\boldsymbol{\varepsilon}} = \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}_{e_r} : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_{e_r}^{(s)} :: \langle \boldsymbol{\varepsilon}_r \otimes \boldsymbol{\varepsilon}_r \rangle^{(s)}, \\ \lim_{\omega \rightarrow 0} \bar{\boldsymbol{\varepsilon}} : \frac{\partial \tilde{\mathbf{L}}''}{\partial \omega}(\omega) : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_v^{(s)} :: \langle \boldsymbol{\varepsilon}_r \otimes \boldsymbol{\varepsilon}_r \rangle^{(s)}, \\ \lim_{\omega \rightarrow +\infty} -\omega^2 \bar{\boldsymbol{\varepsilon}} : \frac{\partial \tilde{\mathbf{L}}''}{\partial \omega}(\omega) : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \left(\mathbf{G}^{(s)} : \mathbf{L}_v^{(s)-1} : \mathbf{G}^{(s)} \right) :: \langle \boldsymbol{\varepsilon}_g \otimes \boldsymbol{\varepsilon}_g \rangle^{(s)}. \end{array} \right. \quad (27)$$

The first two relations (27)_{1,2} classically express the effective elastic properties in the glassy and relaxed asymptotic states at high and low frequencies respectively. The last two relations

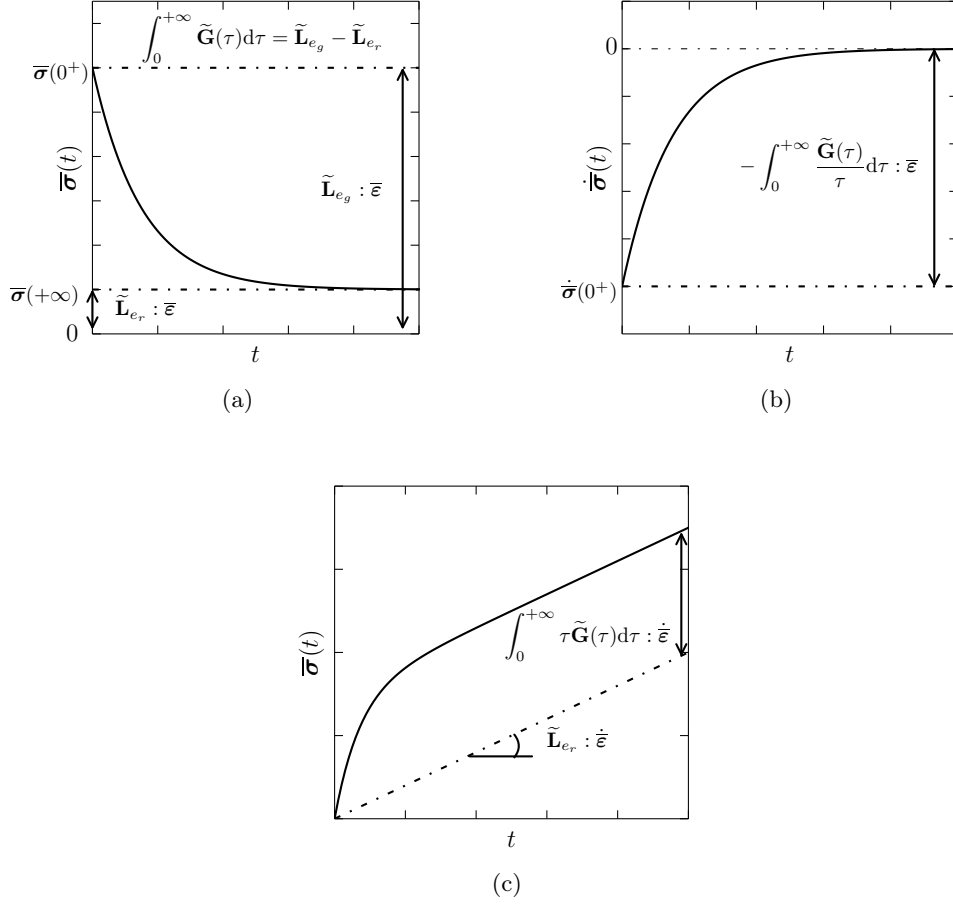


Figure 1: Physical interpretation on the overall relaxation spectrum $\tilde{\mathbf{G}}$ for Type I viscoelastic behaviour. (a) and (b): Relaxation stress and stress rate for a constant macroscopic strain $\bar{\epsilon}$. (c): Stress response for a constant macroscopic strain rate $\dot{\bar{\epsilon}}$.

(27)_{3,4} result from the viscoelastic nature of the transient response. They combine the local viscoelastic properties and the intraphase second moments of the strain fields which are solutions of the asymptotic heterogeneous problems.

From the form of the complex moduli tensor (26), it is also noted that

$$\lim_{\omega \rightarrow 0} \tilde{\mathbf{L}}''(\omega) = \lim_{\omega \rightarrow +\infty} \tilde{\mathbf{L}}''(\omega) = \lim_{\omega \rightarrow +\infty} \omega^2 \frac{\partial \tilde{\mathbf{L}}'}{\partial \omega}(\omega) = \lim_{\omega \rightarrow 0} \frac{\partial \tilde{\mathbf{L}}'}{\partial \omega}(\omega) = \mathbf{0}. \quad (28)$$

By using the expression (26), exact conditions are obtained as well from (27)_{1,3,4} on integrals in time of the effective relaxation spectrum $\tilde{\mathbf{G}}$, namely

$$\int_0^{+\infty} \tilde{\mathbf{G}}(\tau) d\tau, \quad \int_0^{+\infty} \tau \tilde{\mathbf{G}}(\tau) d\tau \quad \text{and} \quad \int_0^{+\infty} \frac{\tilde{\mathbf{G}}(\tau)}{\tau} d\tau. \quad (29)$$

Their respective physical interpretations, in the time domain, are given in Figure 1.

3.6. Mixture of Maxwell constituents (Type II viscoelasticity)

This case has been studied in details in [1, 2]. The main results are recalled here to make the present article self-contained.

3.6.1. Local and effective viscoelastic properties

The response of a Maxwell constituent is characterized by an elastic regime (\mathbf{M}_{e_g}) at short times ($t \rightarrow 0^+$) and a viscous regime (\mathbf{M}_{v_r}) at long times ($t \rightarrow +\infty$). Its constitutive relation is solution of the differential equation

$$\mathbf{M}_{v_r} : \boldsymbol{\sigma}(t) + \mathbf{M}_{e_g} : \dot{\boldsymbol{\sigma}}(t) = \dot{\boldsymbol{\varepsilon}}(t). \quad (30)$$

Hence, the viscoelastic stiffness and compliance tensors of the Maxwell phase (s) are

$$\mathbf{L}^{(s)}(t) = \mathbf{L}_{e_g}^{(s)} e^{-t/\tau^{(s)}} \quad \text{and} \quad \mathbf{M}^{(s)}(t) = \mathbf{M}_{e_g}^{(s)} + \mathbf{M}_{v_r}^{(s)} t. \quad (31)$$

The inverse of the relaxation times $\tau^{(s)}$ are the eigenvalues of $\mathbf{L}_{e_g}^{(s)} : \mathbf{M}_{v_r}^{(s)}$ while the retardation times are null.

The corresponding overall relaxation and creep functions have the following general expressions

$$\tilde{\mathbf{L}}(t) = \int_0^{+\infty} \tilde{\mathbf{G}}(\tau) e^{-t/\tau} d\tau \quad \text{and} \quad \tilde{\mathbf{M}}(t) = \tilde{\mathbf{M}}_{e_g} + \tilde{\mathbf{M}}_{v_r} t + \int_0^{+\infty} \tilde{\mathbf{J}}(\tau) (1 - e^{-t/\tau}) d\tau. \quad (32)$$

The complex viscoelastic stiffness tensor $\tilde{\mathbf{L}}^*(i\omega)$ reads

$$\tilde{\mathbf{L}}^*(i\omega) = \mathcal{LC}(\tilde{\mathbf{L}}(t)) = \int_0^{+\infty} \frac{i\omega\tau}{1 + i\omega\tau} \tilde{\mathbf{G}}(\tau) d\tau. \quad (33)$$

3.6.2. Exact relations on the overall complex modulus and relaxation spectrum

From the stationary principles on ϕ^* and the asymptotic properties of the strain field $\boldsymbol{\varepsilon}^*$, we obtain

$$\left\{ \begin{array}{l} \lim_{\omega \rightarrow +\infty} \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}'(\omega) : \bar{\boldsymbol{\varepsilon}} = \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}_{e_g} : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_{e_g}^{(s)} :: \langle \boldsymbol{\varepsilon}_g \otimes \boldsymbol{\varepsilon}_g \rangle^{(s)} \\ \lim_{\omega \rightarrow 0} \dot{\boldsymbol{\varepsilon}} : \frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) : \dot{\boldsymbol{\varepsilon}} = \dot{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}_{v_r} : \dot{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_{v_r}^{(s)} :: \langle \dot{\boldsymbol{\varepsilon}}_r \otimes \dot{\boldsymbol{\varepsilon}}_r \rangle^{(s)}, \\ \lim_{\omega \rightarrow +\infty} -\omega^2 \bar{\boldsymbol{\varepsilon}} : \frac{\partial \tilde{\mathbf{L}}''}{\partial \omega}(\omega) : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \left(\mathbf{L}_{e_g}^{(s)} : \mathbf{M}_{v_r}^{(s)} : \mathbf{L}_{e_g}^{(s)} \right) :: \langle \boldsymbol{\varepsilon}_g \otimes \boldsymbol{\varepsilon}_g \rangle^{(s)}, \\ \lim_{\omega \rightarrow 0} \dot{\boldsymbol{\varepsilon}} : \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) \right) : \dot{\boldsymbol{\varepsilon}} = \sum_s c_s \left(\mathbf{L}_{v_r}^{(s)} : \mathbf{M}_{e_g}^{(s)} : \mathbf{L}_{v_r}^{(s)} \right) :: \langle \dot{\boldsymbol{\varepsilon}}_r \otimes \dot{\boldsymbol{\varepsilon}}_r \rangle^{(s)}. \end{array} \right. \quad (34)$$

Besides, the general expression of the complex moduli tensor (33) implies that

$$\lim_{\omega \rightarrow +\infty} \tilde{\mathbf{L}}''(\omega) = \lim_{\omega \rightarrow 0} \frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) = \lim_{\omega \rightarrow +\infty} \omega^2 \frac{\partial \tilde{\mathbf{L}}'}{\partial \omega}(\omega) = \lim_{\omega \rightarrow 0} \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) \right) = \mathbf{0}. \quad (35)$$

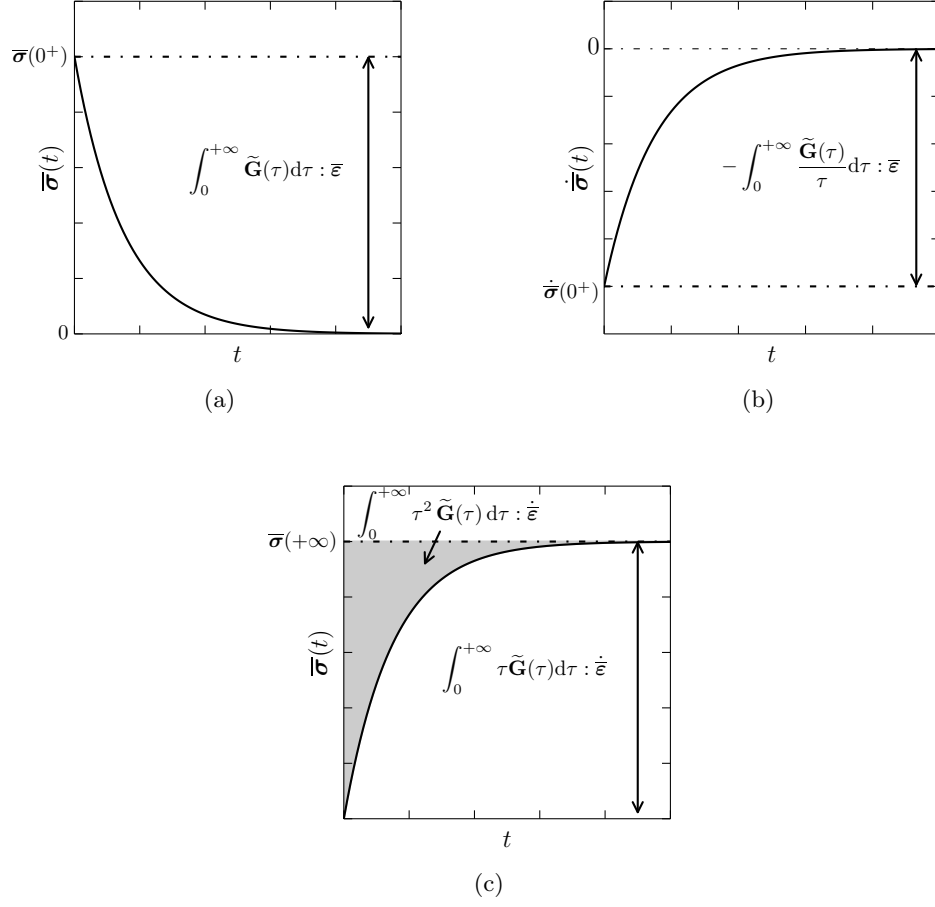


Figure 2: Physical interpretation on the overall relaxation spectrum $\tilde{\mathbf{G}}$ for Type II viscoelastic behaviour. (a), (b) Relaxation stress and stress-rate for a prescribed macroscopic strain $\bar{\boldsymbol{\varepsilon}}$ and (c) stress response for a constant applied macroscopic strain rate $\dot{\bar{\boldsymbol{\varepsilon}}}$.

By introducing the expression (33), the relations (34) give as well conditions on integrals of the overall relaxation spectrum $\tilde{\mathbf{G}}$

$$\int_0^{+\infty} \tilde{\mathbf{G}}(\tau) d\tau, \quad \int_0^{+\infty} \frac{\tilde{\mathbf{G}}(\tau)}{\tau} d\tau, \quad \int_0^{+\infty} \tau \tilde{\mathbf{G}}(\tau) d\tau \quad \text{and} \quad \int_0^{+\infty} \tau^2 \tilde{\mathbf{G}}(\tau) d\tau \quad (36)$$

whose physical meaning is shown in Figure 2 for constant macroscopic strain (rate) loadings.

3.7. Mixture of Kelvin-Voigt constituents (Type III viscoelasticity)

3.7.1. Local and effective viscoelastic properties

The response of a Kelvin-Voigt material is characterized by a viscous regime (\mathbf{L}_{v_g}) at short times ($t \rightarrow 0^+$) and an elastic regime (\mathbf{L}_{e_r}) at long times ($t \rightarrow +\infty$). Its constitutive relation is solution of

$$\boldsymbol{\sigma}(t) = \mathbf{L}_{e_r} : \boldsymbol{\varepsilon}(t) + \mathbf{L}_{v_g} : \dot{\boldsymbol{\varepsilon}}(t). \quad (37)$$

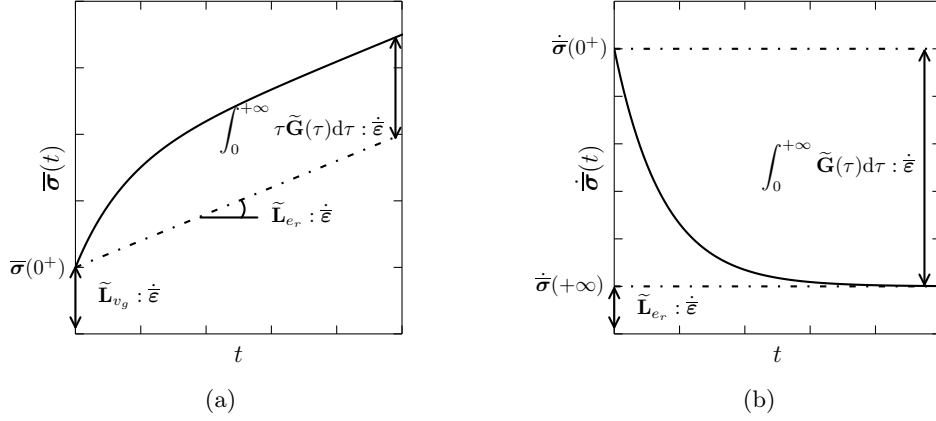


Figure 3: Physical interpretation on the overall relaxation spectrum $\tilde{\mathbf{G}}$ for Type III viscoelastic behaviour. Stress and stress rate for a constant macroscopic strain rate $\dot{\bar{\boldsymbol{\varepsilon}}}$.

Hence, the viscoelastic stiffness and compliance tensors of the Kelvin-Voigt phase (s) are

$$\mathbf{L}^{(s)}(t) = \mathbf{L}_{e_r}^{(s)} + \mathbf{L}_{v_g}^{(s)} \delta(t) \quad \text{and} \quad \mathbf{M}^{(s)}(t) = \mathbf{M}_{e_r}^{(s)} \left(1 - e^{-t/\tau^{(s)}}\right). \quad (38)$$

The retardation times $\tau^{(s)}$ are the eigenvalues of $\mathbf{L}_{v_g}^{(s)} : \mathbf{M}_{e_r}^{(s)}$ while the relaxation times are null.

In agreement with the asymptotic regimes, the general expressions of the relaxation and creep functions are

$$\tilde{\mathbf{L}}(t) = \tilde{\mathbf{L}}_{e_r} + \tilde{\mathbf{L}}_{v_g} \delta(t) + \int_0^{+\infty} \tilde{\mathbf{G}}(\tau) e^{-t/\tau} d\tau \quad \text{and} \quad \tilde{\mathbf{M}}(t) = \int_0^{+\infty} \tilde{\mathbf{J}}(\tau) \left(1 - e^{-t/\tau}\right) d\tau \quad (39)$$

and the complex viscoelastic stiffness tensor $\tilde{\mathbf{L}}^*(i\omega)$ reads

$$\tilde{\mathbf{L}}^*(i\omega) = \mathcal{LC}(\tilde{\mathbf{L}}(t)) = \tilde{\mathbf{L}}_{e_r} + i\omega \tilde{\mathbf{L}}_{v_g} + \int_0^{+\infty} \frac{i\omega\tau}{1 + i\omega\tau} \tilde{\mathbf{G}}(\tau) d\tau. \quad (40)$$

3.7.2. Exact relations on the overall complex modulus and relaxation spectrum

From the stationary principles on ϕ^* and the asymptotic properties of the strain field $\boldsymbol{\varepsilon}^*$, we get

$$\left\{ \begin{array}{l} \lim_{\omega \rightarrow 0} \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}'(\omega) : \bar{\boldsymbol{\varepsilon}} = \bar{\boldsymbol{\varepsilon}} : \tilde{\mathbf{L}}_{e_r} : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_{e_r}^{(s)} :: \langle \boldsymbol{\varepsilon}_r \otimes \boldsymbol{\varepsilon}_r \rangle^{(s)}, \\ \lim_{\omega \rightarrow +\infty} \dot{\bar{\boldsymbol{\varepsilon}}} : \frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) : \dot{\bar{\boldsymbol{\varepsilon}}} = \dot{\bar{\boldsymbol{\varepsilon}}} : \tilde{\mathbf{L}}_{v_g} : \dot{\bar{\boldsymbol{\varepsilon}}} = \sum_s c_s \mathbf{L}_{v_g}^{(s)} :: \langle \dot{\boldsymbol{\varepsilon}}_g \otimes \dot{\boldsymbol{\varepsilon}}_g \rangle^{(s)}, \\ \lim_{\omega \rightarrow 0} \bar{\boldsymbol{\varepsilon}} : \frac{\partial \tilde{\mathbf{L}}''}{\partial \omega}(\omega) : \bar{\boldsymbol{\varepsilon}} = \sum_s c_s \mathbf{L}_{v_g}^{(s)} :: \langle \boldsymbol{\varepsilon}_r \otimes \boldsymbol{\varepsilon}_r \rangle^{(s)}, \\ \lim_{\omega \rightarrow +\infty} -\omega^2 \dot{\bar{\boldsymbol{\varepsilon}}} : \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) \right) : \dot{\bar{\boldsymbol{\varepsilon}}} = \sum_s c_s \mathbf{L}_{e_r}^{(s)} :: \langle \dot{\boldsymbol{\varepsilon}}_g \otimes \dot{\boldsymbol{\varepsilon}}_g \rangle^{(s)}. \end{array} \right. \quad (41)$$

Besides, the general expression of the complex moduli tensor (40) implies that

$$\lim_{\omega \rightarrow 0} \tilde{\mathbf{L}}''(\omega) = \lim_{\omega \rightarrow +\infty} \frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) = \lim_{\omega \rightarrow 0} \frac{\partial \tilde{\mathbf{L}}'}{\partial \omega}(\omega) = \lim_{\omega \rightarrow +\infty} \omega^2 \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) \right) = \mathbf{0}. \quad (42)$$

By incorporating the definition (40) into the relations (41)_{3,4}, conditions on the following integrals of the relaxation spectrum $\tilde{\mathbf{G}}$ are obtained

$$\int_0^{+\infty} \tau \tilde{\mathbf{G}}(\tau) d\tau \quad \text{and} \quad \int_0^{+\infty} \tilde{\mathbf{G}}(\tau) d\tau. \quad (43)$$

Their physical interpretations, in the time domain, are given in Figure 3.

3.8. Mixture of anti-Zener constituents (Type IV viscoelasticity)

3.8.1. Local and effective viscoelastic properties

The behaviour of anti-Zener constituents is characterized by viscous responses in the asymptotic regimes $(\mathbf{L}_{v_g}, \mathbf{M}_{v_r})$ and shows a unique “transient” elastic compliance tensor (\mathbf{M}_e) . Its constitutive response is described by a homogeneous differential equation which may be written as

$$\boldsymbol{\sigma}(t) + (\mathbf{L}_{v_r} - \mathbf{L}_{v_g}) : \mathbf{M}_e : \dot{\boldsymbol{\sigma}}(t) = \mathbf{L}_{v_r} : \dot{\boldsymbol{\epsilon}}(t) + (\mathbf{L}_{v_r} - \mathbf{L}_{v_g}) : \mathbf{M}_e : \mathbf{L}_{v_g} : \ddot{\boldsymbol{\epsilon}}(t). \quad (44)$$

The viscoelastic stiffness and compliance tensors of the anti-Zener phase (s) are

$$\mathbf{L}^{(s)}(t) = \mathbf{L}_{v_g}^{(s)} \delta(t) + \mathbf{G}^{(s)} e^{-t/\tau^{(s)}} \quad \text{and} \quad \mathbf{M}^{(s)}(t) = \mathbf{M}_{v_r}^{(s)} t + \mathbf{J}^{(s)} \left(1 - e^{-t/\tau^{(s)}} \right) \quad (45)$$

with $\tau^{(s)}$ and $\tau'^{(s)}$ the relaxation and retardation times. The relaxation times $\tau^{(s)}$ are the eigenvalues of $(\mathbf{L}_{v_r}^{(s)} - \mathbf{L}_{v_g}^{(s)}) : \mathbf{M}_e^{(s)}$ and the asymptotic viscous properties tensors satisfy

$$\mathbf{L}_{v_r}^{(s)} = \mathbf{L}_{v_g}^{(s)} + \tau^{(s)} \mathbf{G}^{(s)} \quad \text{and} \quad \mathbf{M}_{v_g}^{(s)} = \mathbf{M}_{v_r}^{(s)} + \frac{1}{\tau'^{(s)}} \mathbf{J}^{(s)}. \quad (46)$$

It is noted that $\mathbf{G}^{(s)} = \mathbf{M}_e^{(s)-1} = \mathbf{L}_e^{(s)}$. In agreement with the viscous asymptotic regimes, the overall relaxation and creep functions can be expressed as

$$\tilde{\mathbf{L}}(t) = \tilde{\mathbf{L}}_{v_g} \delta(t) + \int_0^{+\infty} \tilde{\mathbf{G}}(\tau) e^{-t/\tau} d\tau \quad \text{and} \quad \tilde{\mathbf{M}}(t) = \tilde{\mathbf{M}}_{v_r} t + \int_0^{+\infty} \tilde{\mathbf{J}}(\tau) \left(1 - e^{-t/\tau} \right) d\tau. \quad (47)$$

The corresponding overall complex relaxation tensor $\tilde{\mathbf{L}}^*(i\omega)$ reads

$$\tilde{\mathbf{L}}^*(i\omega) = \mathcal{L}\mathcal{C}(\tilde{\mathbf{L}}(t)) = i\omega \tilde{\mathbf{L}}_{v_g} + \int_0^{+\infty} \frac{i\omega\tau}{1 + i\omega\tau} \tilde{\mathbf{G}}(\tau) d\tau. \quad (48)$$

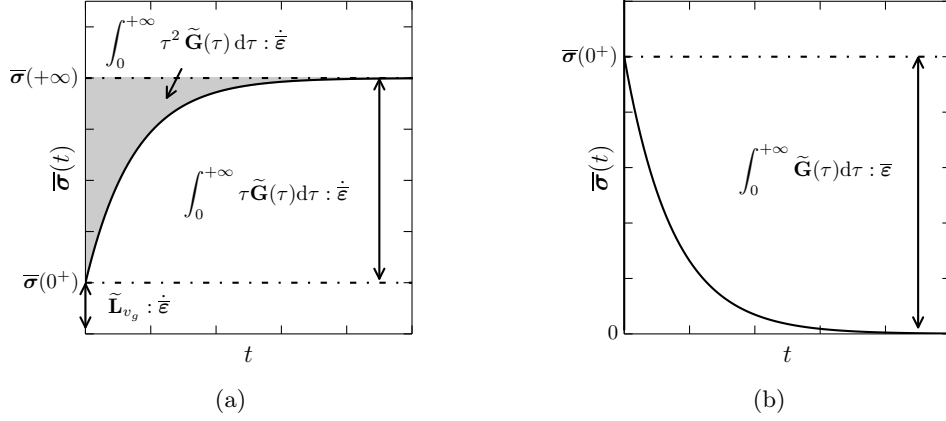


Figure 4: Physical interpretation on the overall relaxation spectrum $\tilde{\mathbf{G}}$ for Type IV viscoelastic behaviour. (a): Stress response for a constant macroscopic strain rate $\dot{\bar{\boldsymbol{\varepsilon}}}$. (b): Relaxation stress for a constant macroscopic strain $\bar{\boldsymbol{\varepsilon}}$.

3.8.2. Exact relations on the overall complex modulus and relaxation spectrum

From the stationary principles on ϕ^* and the asymptotic properties of the strain field $\boldsymbol{\varepsilon}^*$, it follows that

$$\left\{ \begin{array}{l} \lim_{\omega \rightarrow 0} \dot{\bar{\boldsymbol{\varepsilon}}} : \frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) : \dot{\bar{\boldsymbol{\varepsilon}}} = \dot{\bar{\boldsymbol{\varepsilon}}} : \tilde{\mathbf{L}}_{v_r} : \dot{\bar{\boldsymbol{\varepsilon}}} = \sum_s c_s \mathbf{L}_{v_r}^{(s)} :: \langle \dot{\boldsymbol{\varepsilon}}_r \otimes \dot{\boldsymbol{\varepsilon}}_r \rangle^{(s)}, \\ \lim_{\omega \rightarrow +\infty} \dot{\bar{\boldsymbol{\varepsilon}}} : \frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) : \dot{\bar{\boldsymbol{\varepsilon}}} = \dot{\bar{\boldsymbol{\varepsilon}}} : \tilde{\mathbf{L}}_{v_g} : \dot{\bar{\boldsymbol{\varepsilon}}} = \sum_s c_s \mathbf{L}_{v_g}^{(s)} :: \langle \dot{\boldsymbol{\varepsilon}}_g \otimes \dot{\boldsymbol{\varepsilon}}_g \rangle^{(s)}, \\ \lim_{\omega \rightarrow +\infty} -\omega^2 \dot{\bar{\boldsymbol{\varepsilon}}} : \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) \right) : \dot{\bar{\boldsymbol{\varepsilon}}} = \sum_s c_s \mathbf{L}_e^{(s)} :: \langle \dot{\boldsymbol{\varepsilon}}_g \otimes \dot{\boldsymbol{\varepsilon}}_g \rangle^{(s)}, \\ \lim_{\omega \rightarrow 0} \dot{\bar{\boldsymbol{\varepsilon}}} : \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) \right) : \dot{\bar{\boldsymbol{\varepsilon}}} = \sum_s c_s \left(\Delta \mathbf{L}_v^{(s)} : \mathbf{M}_e^{(s)} : \Delta \mathbf{L}_v^{(s)} \right) :: \langle \dot{\boldsymbol{\varepsilon}}_r \otimes \dot{\boldsymbol{\varepsilon}}_r \rangle^{(s)}. \end{array} \right. \quad (49)$$

with $\Delta \mathbf{L}_v^{(s)} = \mathbf{L}_{v_r}^{(s)} - \mathbf{L}_{v_g}^{(s)}$.

From the form of the complex moduli tensor (48), it is also noted that

$$\lim_{\omega \rightarrow 0} \frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) = \lim_{\omega \rightarrow +\infty} \frac{1}{\omega} \tilde{\mathbf{L}}'(\omega) = \lim_{\omega \rightarrow +\infty} \omega^2 \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) \right) = \lim_{\omega \rightarrow 0} \frac{\partial}{\partial \omega} \left(\frac{1}{\omega} \tilde{\mathbf{L}}''(\omega) \right) = \mathbf{0}. \quad (50)$$

By introducing the expression (48), relations (49)_{1,3,4} deliver conditions on integrals of the effective relaxation spectrum $\tilde{\mathbf{G}}$

$$\int_0^{+\infty} \tau \tilde{\mathbf{G}}(\tau) d\tau, \quad \int_0^{+\infty} \tau^2 \tilde{\mathbf{G}}(\tau) d\tau \quad \text{and} \quad \int_0^{+\infty} \tilde{\mathbf{G}}(\tau) d\tau \quad (51)$$

whose physical meaning is shown in Figure 4.

4. Concluding remarks

By making use of stationary principles for complex viscoelasticity and asymptotic properties of the solution fields of viscoelastic heterogeneous problems, exact asymptotic relations in the frequency domain have been derived on the effective storage and loss moduli tensors as well as the overall relaxation spectrum which characterizes the transient viscoelastic response. These results extend those previously obtained for Maxwell constituents [1, 2] to any kind of elementary viscoelastic constituents (see Table 1). In particular, four independent exact relations have been obtained whatever the kind of viscoelastic behaviour. This implies that the form of the overall constitutive law does not follow, in general, the one of the elementary viscoelastic constituents since the latter are described by two or three independent parameters, depending on the type of viscoelastic behaviour. This remark is consistent with the previous studies on Kelvin-Voigt [5, 6, 3] and Maxwell materials [4]. In the case of mixtures of Zener or anti-Zener constituents, this simply means that the overall relaxation (retardation) spectrum does not reduce in general to a single Dirac mass.

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