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점탄성 유체에 관한 조성방정식 이론 연구

권 영 돈

성균관대학교 섬유공학과
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Recent Development in the Constitutive Theory of Viscoelastic Fluid

Youngdon Kwon

Department of Textile Engineering, Sung Kyun Kwan University,
300 Chunchun-dong, Jangan-gu, Suwon, Kyunggi-do 440-746, Korea
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1. Introduction

The history of theoretical rheology is not a long one, but in our opinion, it is blotted with a number of controversial propositions. Many scientists in this field of study have hoped to derive or find a reliable and decisive constitutive equation (CE) for non-Newtonian fluid. Since no fundamental relation is believed to have yet been discovered, at least more than ten popular CEs are in competition at present without any clue to a preferable type. In independent works performed by many researchers over recent decades, various types of pathological behavior of CEs have been observed. This situation raises some serious doubts about ongoing theoretical study of highly nonlinear rheological phenomena which usually exist in the modern processing of polymeric liquids. The application of bad formulation of CEs to real flow simulation makes all the efforts in vain, and further, the unphysical results have already incurred a great deal of speculation which in fact have nothing at all to do with the true flow phenomenon.

For several decades, there have been many attempts to derive CEs of viscoelastic materials from the viewpoints of mechanics, mathematics and physics. However, a deep understanding on the nature of viscoelasticity has not yet been reached.

In chronological order, the approaches used by rheologists to obtain a fundamental CE or a class of CEs for viscoelastic liquids can be roughly described in several stages. The first attempt was initiated from the viewpoint of continuum mechanics. Pioneering work in this direction was carried out by Oldroyd [1,2] who postulated quasilinear and nonlinear CEs of differential and integral types to relate external observable variables such as stress tensor $\underline{\underline{\sigma}}$ and strain rate tensor $\underline{\underline{e}}$, and also elucidated important principles of invariance. Later, it was recognized that many rheological equations derived from different approaches by different scientists, are associated with the equations proposed by Oldroyd. Until now, a great many rheological equations, both of differential and integral types have been suggested, and they are able to describe some properties of viscoelastic liquids. However, the disadvantage of this work is that there is lack of thermodynamic analysis, and hence important phenomena such as dynamic birefringence, non-isothermal flow, and diffusion cannot be considered, and the resultant equations are often non-evolutionary, having solutions that grow exponentially to infinity with time, which is not physically plausible.

This approach was then, followed by rational continuum mechanics approach which was developed mainly by mathematicians [3-6]. They treated the rheology simply as a branch of mathematics and found the most general form, which relates kinematic variables to dynamic ones. In essence, the basic system involving the constitutive and thermodynamic equations is constructed using strict mathematics, with the CEs under the restriction of causality (or determinism), material objectivity and local action principles. In this way, the properties of all viscoelastic liquids can be described by a set of hereditary functionals with "fading memory", whose invariance and thermodynamic consistency are explicit. Unfortunately, there is no unique way to specify a deterministic form for the memory functionals and hence predictions are not possible. In other words, the restrictions imposed on the constitutive functionals by thermodynamics and basic principles are still loose enough to allow enormous choice of the memory functionals. Thus, even though the class of perceivable CEs has been narrowed and some rigorous understanding has been achieved, the results are still too general to produce any specific model for the comparison with experimental data.

The third approch is purely physical and explains the behavior of polymeric liquid in terms of the intra- and inter-molecular dynamics. In the beginning, this approach was used to study the behavior of dilute polymer solutions by Kargin, Slonimsky, Kirkwood, Riseman, Rouse, Zimm and so on (see for a review [7]). For a long time, concentrated polymer solutions and melts have also been considered as "temporary networks" of entangled chains that can move over each other [8]. Green and Tobolsky [9], Lodge [10], and Yamamoto [11,12,13] developed semi-phenomenological theories that extend the theory for rubber elasticity. This idea was enhanced by the rise of the "reptation" theories, which are due to the work of de Gennes [7], Edwards [14], and Doi and Edwards [15-17]. The creation and decay of the molecular entanglements are studied by the statistical description of a polymer molecule moving along its own axis within a "tube" created by surrounding molecules, and then the motion of the molecule is averaged over high frequency transverse Brownian motion. Another reptational or statistical approach can be found in the works by Giesekus [18], Curtiss *et al.* [19-22], and Volkov and Pokrovsky [23,24].

In contrast to the mathematical approach, this statistical method has generated a number of specific outcomes, i.e. CEs, and has been generally successful for explaining viscoelastic behavior of polymer liquids in linear or weakly nonlinear deformations. Even though this approach was assumed to be based on the fundamental principles of molecular physics, apart from their poor description of the experimental data in highly nonlinear region it also suffers from high empiricism involved in formulation and some arbitrary attempts to overcome mathematical difficulties. Additionally, recent mathematical analyses and numerical simulations revealed numerous examples of unphysical and unstable behavior of those CEs. Hence, it is questionable whether the CEs formulated by these theories are consistent with the Second Law of thermodynamics.

The fourth method of deriving CEs has been devised by Leonov [25]. It considers all nonlinear viscoelastic phenomena using quasi-linear irreversible thermodynamics and introducing a "recoverable strain tensor" as an internal parameter. By this approach, Leonov proposed a class of Maxwell-type differential CEs under strict stability constraints which are based mainly on thermodynamics. Later, similar approach was adopted by Dashner and Vanarsdale [26,27] to formulate the class of CEs almost equivalent to Leonov's in their most general forms.

Almost all CEs proposed in the literature have a limited ability to describe start-up, steady state and relaxation phenomena of polymer fluids in standard simple shear and extensional flows, within a relatively narrow region of strain rates usually employed in viscometric tests [28]. There are, however, two frustrating problems in this field of study: (i) no specific CE proposed could describe the whole set of available experimental data with one set of parameters specified, (ii) in real modern processing the values of Deborah number may be at least two orders of magnitude higher than those in usual rheological tests and in that flow regime almost all CEs exhibit various numerical instabilities, the reason for which still remains unclear.

There are a lot of speculations in the literature about relations between the instabilities in CEs and those observed in the polymer fluid flow (see, e.g. Ref. [29] and for the counter example see Ref. [30]). There also exist contrary opinions about the physical meaning of non-evolutionary (Hadamard unstable) behavior of rheological equations. A major motivation to relate the unstable CE to real flow instability may consist in the perception which has spread among the scientists that no rheological constitutive model is globally stable.

A few books [22,28,31-33] and a lot of papers devoted to the rheology of polymer fluids do not answer the question: "Which CE should be chosen to solve fluid mechanics problems of polymer processing in the usual case of large recoverable deformations where nonlinear effects of elasticity are important?" (One can find some discussion on this topic in the recent book [34].) Therefore, general investigation to seek not only descriptive but also reliable CEs seems more necessary. The principle of choosing CEs should be based on such fundamental properties of the equations as formulations of dissipation and free energy, stability conditions, the boundedness of variables and the relation of these to thermodynamics with subordination to the Second Law.

To study these fundamental properties of CEs, even in the relatively simple case of general Maxwell-like or time-strain separable single integral models, one has to employ some framework of a general formalism within which it is feasible to establish some general constraints imposed by the fundamental macroscopic laws of thermodynamics. Relatively recently, there have been suggested in the literature several formal approaches. One is the local approach of non-equilibrium thermodynamics by Leonov [25, 35]. As mentioned before, from this idea, he could obtain a class of Maxwell-like CEs with some constraints. Almost 10 years later, the Poisson-bracket formalism has been established, which was first introduced to viscoelasticity by Grmela [36,37] and then extended by Beris and Edwards [38,39]. This approach extends the Hamiltonian formalism in classical mechanics to the case of continuum mechanics employing functional approach and variational derivatives and also including the dissipative functional into consideration. In a later paper by Leonov [40], it was proved that both approaches result in the same formulation, thereby giving the general Maxwell-like model, and then its canonical form was established. One can find another two general formalisms developed by Kwon and Shen [41,42], and by Jongschaap *et al.* [43,44]. Both are based on the irreversible thermodynamics, but the first one uses the notion of evolution of the temporary network structure and the second employs a matrix representation with intensive use of the time reversal principle. Regardless of all differences in their detailed scheme of derivation, their equivalence to Leonov's in the most general form is evident.

Another important objective that the rheologists should keep in mind, is in solving flow problems that contain complicated geometry under high deformation rate, especially the ones in industrial processing. Complexity of flow problems is enhanced by the fading memory of polymeric liquids that does not exist in viscous or elastic material. Hence, the viscoelastic polymer fluids show many unique features such as kinetics or evolution of stress variables under steady deformation, which cannot be examined in the other fluid systems. Owing to this, even problems with simple geometry which may be solved analytically for viscous fluids, have often to be treated numerically in the viscoelastic case.

As mentioned above, in order to solve flow problems, the first crucial decision to be made beforehand must be the proper choice of a CE for viscoelastic liquids. The following principles of choice can be suggested for the selection of CEs for practical use:

(i) Stability. However well an unstable CE can describe rheometric tests, it is impossible to use it in modeling of polymer processing, since the Deborah numbers there may be at least two order of magnitude higher and flow in a much more complicated manner. Extrapolation of most CEs to the region of high Deborah numbers and 3D flows can result in several types of instabilities during numerical simulation. These instabilities reflect the mathematical structure of the CEs proposed. In most cases, they are not related to physical instabilities observed in the flows of polymeric fluids, or poor numerical algorithms, but rather, to violations of some fundamental principles.

(ii) Descriptive ability and flexibility. It is now well recognized that polymer melts with similar linear viscoelastic spectra can show qualitatively different nonlinear behavior. For the proper description of various flows, this requires some functions of the kinematic variables, and the associated nonlinear parameters in the CE, to be specified within the stability constraints. Once these functional forms and parameters are specified for a particular polymer, the CE must simultaneously describe the entire set of available experimental data fairly accurately.

(iii) Computational economy. The proposed CE should allow for numerical calculations in complex flows as little computational effort as possible. For example, despite the good descriptive ability, it is rather cumbersome to work with models in which the elastic potential is specified in terms of the principal values of a strain measure, and it is usually conceived that working with CEs of differential type is preferable for numerical calculation than with integral ones.

(iv) Extensibility. Real polymer processing is confronted with a variety of complications such as compressibility, non-isothermality, wall slip, phase transitions and separations, chemical effects (degradation, curing), etc. In principle, the CE of choice should be amenable to extension in order to accommodate these phenomena.

Among the four principles listed above, the first two can be regarded as the most fundamental properties which the CEs should possess. This review article is chiefly focused on the first principle, i.e. the stability of CEs, and it is organized as follows. First, in section 2 we review almost all popular viscoelastic CEs of both differential and integral types, and then represent them in some unified form. In section 3, all the stability results are collected, and the most general form is presented for compressible as well as incompressible flow system. In addition, all available results of stability analysis are summarized in a table, where one can easily observe in view of stability the behavior of most of renowned CEs. Next, description of experimental data by the CE which satisfies all stability criteria, is demonstrated for the most thoroughly characterized poymer liquid. Finally, conclusions follow with some comments on recommended future study of theoretical rheology.

2. Formulation of Viscoelastic Constitutive Equations

In this section, many of popular viscoelastic CEs are reviewed. Even though there are numerous ways to classsify viscoelastic CEs, here we separate them with respect to their mathematical structure for the specific purpose of stability analysis. They are of either differential or integral type and are suggested mainly in the incompressible case. At the end, the unified form including rheological models applicable to compressible flow is presented for the general stability analysis valid for most CEs.

2.1. Incompressible Differential Constitutive Equations

The simplest class of differential nonlinear CEs for incompressible viscoelastic liquids is the Maxwell-type. It can be written in the "canonical" form [40]:

$$\theta \overset{\square}{\underline{\underline{c}}}_{\zeta} + \underline{\underline{\psi}}_{\zeta}(T, \underline{\underline{c}}) = \underline{\underline{0}}, \quad \underline{\underline{\sigma}} = -p\underline{\underline{\delta}} + \frac{1}{\zeta}\underline{\underline{\tau}}(T, \underline{\underline{c}}) \qquad (-1 \le \zeta \le 1), \tag{1}$$

$$\text{where } \overset{\square}{\underline{\underline{c}}}_{\zeta} \equiv \frac{d\underline{\underline{c}}}{dt} - \underline{\underline{c}}\cdot\underline{\underline{\omega}} + \underline{\underline{\omega}}\cdot\underline{\underline{c}} - \zeta(\underline{\underline{c}}\cdot\underline{\underline{e}} + \underline{\underline{e}}\cdot\underline{\underline{c}}). \tag{2}$$

Here $\underline{\underline{\sigma}}$ is a total stress tensor; p is isotropic pressure; $\underline{\underline{\delta}}$ is a unit tensor; $\underline{\underline{c}}$ which is supposedly positive definite, is the symmetric second rank "configuration" or "elastic Finger strain" tensor; $\underline{\underline{e}} = (\nabla\underline{v} + \nabla\underline{v}^T)/2$ and $\underline{\underline{\omega}} = (\nabla\underline{v} - \nabla\underline{v}^T)/2$ are strain rate and vorticity tensors of the velocity field $\underline{v}$ with the notation $(\nabla\underline{v})_{ij} = \partial v_j/\partial x_i$ in the Cartesian coordinate x_i, and the strain rate tensor is subject to the incompressibility condition, $tr\underline{\underline{e}} = 0$; $\underline{\underline{\psi}}_{\zeta}$ and $\underline{\underline{\tau}}$, the dimensionless dissipative term and "extra stress" tensor, are isotropic tensor functions of the tensor $\underline{\underline{c}}$ that provide the CEs with a regular limit to a linear viscoelastic case; θ is relaxation time which sharply depends on temperature T, and can also depend on the basic scalar invariants I_j of the tensor $\underline{\underline{c}}$; ζ is a numerical parameter. The operation $\overset{\square}{\underline{\underline{c}}}_{\zeta}$ defined in eq. (2), which determines the evolution equation in (1), is called the "mixed" convected time derivative of $\underline{\underline{c}}$. Depending on the value of ζ, it takes the forms of the upper convected, $\overset{\nabla}{\underline{\underline{c}}}(\zeta = 1)$, corotational, $\overset{\circ}{\underline{\underline{c}}}(\zeta = 0)$, and lower convected, $\overset{\triangle}{\underline{\underline{c}}}(\zeta = -1)$ time derivatives. The evolution equation in (1) is subject to the following initial condition:

$$\underline{\underline{c}} = \underline{\underline{\delta}} \quad at \quad t = 0. \tag{3}$$

Also, the relation between the tensors $\underline{\underline{\tau}}$ and $\underline{\underline{c}}$ is usually assumed to be potential: $\underline{\underline{\tau}} = 2\rho\underline{\underline{c}}\cdot\partial\underline{\underline{c}}$ (that is, the hyper-viscoelastic case where there exists a thermodynamic potential relation), where F is the Helmholtz free energy, ρ is the constant density, and it has been first obtained by Murnaghan [45] for elastic solids. In the case of isotropic materials,

$$\underline{\underline{\tau}}=2\rho[\varphi_1\underline{\underline{c}}+\varphi_2(I_1\underline{\underline{c}}-\underline{\underline{c}}^2)+\varphi_3 I_3\underline{\underline{\delta}}]\quad\left(\varphi=\frac{\partial F}{\partial I_j}\right), \quad (4)$$

where

$$I_1=\mathrm{tr}\underline{\underline{c}},\ I_2=\frac{1}{2}(I_1^2-\mathrm{tr}\underline{\underline{c}}^2),\ I_3=\det\underline{\underline{c}} \quad (5)$$

are basic invariants of the tensor $\underline{\underline{c}}$. Non-potential formulations are also formally possible, if φ_j are appropriately specified for individual models. A special class of CEs proposed in papers [25,35] corresponds to a particular case of potential CEs (1) with conditions

$$\det\underline{\underline{c}}=1,\quad \zeta=1, \quad (6)$$

where the tensor $\underline{\underline{c}}$ is treated as a recovery strain tensor (or elastic Finger strain tensor) which in principle can be measured in experiments. In this case, the extra stress tensor is expressed by the Finger formula:

$$\underline{\underline{\tau}}=2\rho(\varphi_1\underline{\underline{c}}-\varphi_2\underline{\underline{c}}^{-1}). \quad (7)$$

When $\zeta=1$ and the potential case is under consideration, eqs.(1) and (2) give rise to the energetic relation:

$$\rho\frac{dF}{dt}+D=\mathrm{tr}(\underline{\underline{\tau}}\cdot\underline{\underline{e}}),\ D=\mathrm{tr}(\underline{\underline{c}}^{-1}\cdot\underline{\underline{\tau}}\cdot\underline{\underline{\psi}})/2,\ \underline{\underline{\psi}}=\underline{\underline{\psi}}_\zeta\Big|_{\zeta=1}. \quad (8)$$

Here D is the mechanical dissipation which according to the Second Law must be positive definite in any flow situation and vanishes in the rest state.

In order to better describe the data, multimodal approaches are usually employed with a finite discrete spectrum of independent nonlinear relaxation modes:

$$F(T,\underline{\underline{c}}_1,\cdots,\underline{\underline{c}}_N)=\sum_{k=1}^{N}F_{(k)}(T,\underline{\underline{c}}_k),\ \underline{\underline{\tau}}=\sum_{k=1}^{N}\underline{\underline{\tau}}_k(T,\underline{\underline{c}}_k), \quad (9)$$

and the evolution equation in (1) holds for every mode. The above nonlinear multimodal approach can be justified only when the discrete relaxation times in the limit of linear viscoelasticity are well (say, in order) separated.

In the particular case of $\zeta=1$, the derivation of eq. (1) was given by Beris and Edwards [38] who employed the Poisson-bracket formalism developed by Grmela [36,37]. A brief derivation and extensive discussion of the above equations from the viewpoint of thermodynamics, along with many examples, were given in Ref. [40]. Some general derivations of viscoelastic CEs were also suggested recently by Jongschaap *et al.* [44].

We now illustrate how particular viscoelastic CEs can be obtained from the above general equations by specifying the terms $\underline{\underline{\psi}}_\zeta$ and $\underline{\underline{\tau}}$ in eqs. (1), or F in eqs. (4).

i) the interpolated Maxwell model

$$\theta=\mathrm{const.},\ \underline{\underline{\psi}}_\zeta=\underline{\underline{c}}-\underline{\underline{\delta}},\ (2\rho/G)F=I_1-3,\ \underline{\underline{\tau}}=G\underline{\underline{c}}, \quad (10)$$

where G is the shear modulus. The set of eqs. (1), (4) and (10) are also called the Gordon-Schowalter [46] or the Johnson-Segalman [47] which includes upper convected ($\zeta=1$), lower convected ($\zeta=-1$) and corotational ($\zeta=0$) Maxwell models. These models were derived from some microscopic arguments introducing nonaffine motion based on the Ericksen's theory for anisotropic fluids or the slippage of strand in continuum. If we add the Newtonian viscosity term to the stress tensor, then eqs.(4) constitute a form of the Oldroyd A ($\zeta=-1$) or the Oldroyd B ($\zeta=1$) model.

ii) the Phan Thien-Tanner model [48,49]

$$\theta=f(I_1),\ \underline{\underline{\psi}}_\zeta=\underline{\underline{c}}-\underline{\underline{\delta}},\ (2\rho/G)F=I_1-3,\ \underline{\underline{\tau}}=G\underline{\underline{c}}. \quad (11)$$

It is derived from the Yamamoto's theory [50]. Here, the function f is assumed as a linear or exponential function of I_1. It is evident that eq. (10) is the particular case of eq. (11) when $f\equiv 1$. When $\zeta=1$, it is particularly called as an upper convected Phan Thien-Tanner model.

iii) the White-Metzner model [51]

$$\theta = f(II_e),\ II_e = 2\mathrm{tr}(\underline{\underline{e}}^2),\ \zeta = 1, \underline{\underline{\psi}}_{\zeta} = \underline{\underline{\psi}} = \underline{\underline{c}} - \underline{\underline{\delta}},$$

$$(2\rho/G)F = I_1 - 3,\ \underline{\underline{\tau}} = G\underline{\underline{c}}. \tag{12}$$

It is introduced by modification of an upper convected Maxwell model, and does not belong to the class of quasilinear (linear in derivatives) CEs.

iv) the FENE model (section 8.5.3 of Ref. [28])

$$\theta = \mathrm{const.},\ \zeta = 1,\ \underline{\underline{\psi}} = (K\underline{\underline{c}} - \underline{\underline{\delta}}),$$

$$(2\rho/G)F = (I_c - 3)\ln K,\ \underline{\underline{\tau}} = GK\underline{\underline{c}},\ K = \frac{I_c - 3}{I_c - I_1}. \tag{13}$$

where $G = nkT$ is the elastic modulus, n is the number of polymer chains per unit volume, k is the Boltzmann constant, T is temperature, $\underline{\underline{c}} = \langle \underline{RR} \rangle / \langle R_o^2 \rangle$, $\langle \cdot\cdot \rangle$ denotes the average over configuration space, $I_c = R_c^2 = \mathrm{const.}$, $I_1 = \mathrm{tr}\underline{\underline{c}}$, and $\underline{R}$ is the end-to-end vector with mean-square equilibrium length R_o and finite length R_c. Above canonical form is obtained in Ref. [40].

v) the simplest Giesekus model [18,52]

$$\theta = \mathrm{const.},\ \zeta = 1,\ \underline{\underline{\psi}} = \alpha\underline{\underline{c}}^2 + (1-2\alpha)\underline{\underline{c}} - (1-\alpha)\underline{\underline{\delta}},$$

$$0 \le \alpha \le 1,\ (2\rho/G)F = I_1 - 3,\ \underline{\underline{\tau}} = G\underline{\underline{c}}. \tag{14}$$

where α is a positive numerical constant. This CE phenomenologically describes orientation phenomena in elastic liquids.

vi) the Marrucci [53] and the Larson [54] models

$$\theta = \mathrm{const.},\ \zeta = 1,\ \underline{\underline{\psi}} = B(I_1)(\underline{\underline{c}} - \underline{\underline{\delta}}),$$

$$(2\rho/G)F = (3/\xi)\ln B(I_1)\ \ 0 \le \xi \le 1,\ \underline{\underline{\tau}} = G\underline{\underline{c}}/B(I_1),$$

$$B(I_1) = 1 + \frac{\xi}{3}(I_1 - 3), \tag{15}$$

where they are presented in a canonical form [40]. The Larson model is derived from modification of the Doi-Edwards reptation model using the concept of so called "partially extending strand convection".

vii) the general class of the Leonov CEs [25,35]

$$\underline{\underline{\psi}} = \frac{1}{2}\underline{\underline{c}} \cdot \left\{ b_1(I_1, I_2)\left[\underline{\underline{c}} - \frac{I_1}{3}\underline{\underline{\delta}}\right] - b_2(I_1, I_2) \left[\underline{\underline{c}}^{-1} - \frac{I_2}{3}\underline{\underline{\delta}}\right]\right\},\ \theta = \mathrm{const.},\ \zeta = 1,$$

$$\underline{\underline{\tau}} = 2\rho(\ \varphi_1\underline{\underline{c}} - \ \varphi_2\underline{\underline{c}}^{-1}),\ b_1 > 0,\ b_2 > 0,\ \det\underline{\underline{c}} = 1. \tag{16}$$

Here the functions $b_i(I_1, I_2)$ should have a proper linear viscoelastic limit and their positiveness suffices the positive definiteness of the dissipation. This model is introduced by using a local equilibrium approach of irreversible thermodynamics. The convexity constraints

$$\varphi_1 > 0,\ \varphi_2 > 0,\ \varphi_{11}\varphi_{22} > \varphi_{12}^2\ (\varphi_{ij} = \partial\varphi_i/\partial I_j) \tag{17}$$

imposed on the general form of potential F, were also suggested [25,40]. The important implication of inequalities (17) and the proper use of this class of CEs are discussed in detail in Refs. [34,55]. In the simple case of $b_1 = b_2 = 1$, with the neo-Hookean potential for F, it reduces to the simplest Leonov model which does not include any nonlinear parameter.

2.2. Incompressible Integral Constitutive Equations

From a wide class of viscoelastic CEs of the integral type, only the single integral ones have been experimentally tested. In the common incompressible case, its general form is represented as [56]:

$$\underline{\underline{\sigma}} = -p\underline{\underline{\delta}} + \underline{\underline{\tau}} = -p\underline{\underline{\delta}} + 2\rho \int_{-\infty}^{t} [\tilde{\varphi}_1(t-t_1, I_1, I_2)\underline{\underline{C}} - \tilde{\varphi}_2(t-t_1, I_1, I_2)\underline{\underline{C}}^{-1}]dt_1. \quad (18)$$

Here $\underline{\underline{C}}$ is the Finger total deformation tensor, which has the following evolution and invariant relations for incompressible media:

$$\frac{d\underline{\underline{C}}}{dt} = \underline{\underline{C}} \cdot \underline{\nabla v} + \underline{\nabla v}^T \cdot \underline{\underline{C}}, \quad \underline{\underline{C}}\Big|_{t=t_1} = \underline{\underline{\delta}}, \quad (19)$$

$$I_1 = \mathrm{tr}\underline{\underline{C}}, \; I_2 = \frac{1}{2}(I_1^2 - \mathrm{tr}\underline{\underline{C}}^2), \; I_3 = \det\underline{\underline{C}} = 1, \quad (20)$$

$\tilde{\varphi}_1$ and $\tilde{\varphi}_2$ are generally independent functions specified by the constitutive model and sometimes they are correlated by an elastic potential as in the case of the K-BKZ model [57,58]. Many specific equations of this class are quite popular and are frequently used in the rheology of viscoelastic liquids. Experiments suggested that one simplification of $\tilde{\varphi}_1$ and $\tilde{\varphi}_2$, so called "time-strain separability" (see Ref. [28] p.81) is assumed to hold (its invalidity is discussed in the later section). It factors out one common functional term dependent purely on time from $\tilde{\varphi}_1$ and $\tilde{\varphi}_2$, and it can be shown as

$$\varphi_k(t-t_1, I_1 I_2) = m(t-t_1)\; \varphi_k(I_1, I_2),$$
$$(k=1,2), \; m(t-t_1) = \frac{dG(t-t_1)}{dt_1}. \quad (21)$$

Here $G(t)$ is the dimensionless relaxation modulus defined in the linear viscoelasticity theory, which is a smooth function of time with common properties of

$$G(t) > 0, \; G'(t) < 0, \; G''(t) > 0, G(0) < \infty$$
$$\text{and } G(\infty) = 0. \quad (22)$$

Including the condition of time-strain separability, we can rewrite eq.(18) as

$$\underline{\underline{\sigma}} = p\delta + 2\rho \int_{-\infty}^{t} m(t-t_1)[\;\varphi_1(I_1, I_2)\underline{\underline{C}} - \varphi_2(I_1, I_2)\underline{\underline{C}}^{-1}]\,dt_1. \quad (23)$$

Among many specifications in the type of eq.(18) or (23), some popular forms are enumerated here.

i) the CE by Wagner and coworkers (the Wagner model I) [59]

$$(2\rho/G)\;\varphi_1 = f \cdot \exp(-n_1\sqrt{I-3}) + (1-f) \cdot \exp(-n_2\sqrt{I-3}),$$
$$\varphi_2 = 0, \; I = \beta \cdot I_1 + (1-\beta)I_2 \quad (24)$$

where G is the modulus, and f, n_1, n_2 and β are positive fitting parameters. It is proposed based on some empirical reasons.

ii) the CE by Wagner and Demarmels (the Wagner model II) [60]

$$(2\rho/G)\;\varphi_1 = (1-\beta)\cdot h(I_1, I_2),$$
$$(2\rho/G)\;\varphi_2 = \beta \cdot h(I_1, I_2),$$
$$h(I_1, I_2) = [1 + a\sqrt{(I_1-3)(I_2-3)}]^{-1}, \quad (25)$$

where a and b are positive fitting parameters.

iii) the model by Papanastasiou et al. [61]

$$(2\rho/G)\;\varphi_1 = \alpha \cdot [\alpha - 3 + \beta I_1 + (1-\beta)I_2]^{-1}, \; \varphi_2 = 0, \quad (26)$$

where α and β are numerical parameters.

iv) the model by Luo and Tanner [62]

$$(2\rho/G)\;\varphi_1 = \alpha \cdot [\alpha - 3 + \beta I_1 + (1-\beta)I_2]^{-1},$$
$$(2\rho/G)\;\varphi_2 = \kappa \cdot \varphi_1. \quad (27)$$

It is proposed as a modification of eq.(26) in order to include a nonzero value of the second normal stress difference in simple shear flow. Here the additional parameter κ is positive and it relates the first and second normal stresses as $N_2/N_1 = -\kappa/(1-\kappa)$ in simple shear flow.

v) the class of the K-BKZ CEs [57,58]

$$\tilde{\varphi}_1 = \frac{\partial \tilde{F}}{\partial I_1}, \quad \tilde{\varphi}_2 = \frac{\partial \tilde{F}}{\partial I_2}, \quad (28)$$

or in the case of the time-strain separability, i.e.

eq.(23):

$$\varphi_1 = \frac{\partial F}{\partial I_1}, \quad \varphi_2 = \frac{\partial F}{\partial I_2}. \tag{29}$$

This is derived on the basis of thermodynamics. In eqs.(28), the potential $\tilde{F}$ denotes the thermodynamic free energy F with relaxation effects taken into account. Note that the Wagner models I (24) and II (25), the model by Papanastasiou *et al.* (26) and its modification (27) do not satisfy this potential relation (29) except for some trivial cases.

Several specifications of the elastic potential F in eq.(29) are proposed in the literature.

vi) the Oldroyd-Lodge model [63-65]

$$(2\rho/G)\ \varphi_1 = 1, \quad \varphi_2 = 0, \tag{30}$$

where it results from the neo-Hookean potential $(2\rho/G)F = (I_1 - 3)/2$.

vii) the Larson and Monroe potential [66]

$$(2\rho/G)F = \frac{3}{2\alpha}\ln\left[1 + \frac{\alpha}{3}(I-3)\right]$$
$$I = (1-\beta)I_1 + \sqrt{1+2\beta \cdot I_2} - 1,$$
$$\alpha = k_0 + k_2 \cdot \tan^{-1}\left[\frac{k_1(I_2 - I_1)^3}{1+(I_2-I_1)^2}\right]. \tag{31}$$

Here k_0, k_1, k_2 and β are numerical fitting constants. It is proposed in order to describe not only simple shear and simple elongational but also biaxial elongational data.

viii) the Doi-Edwards model [15-17]

$$\underline{\underline{\sigma}} = -p\underline{\underline{\delta}} + 2\rho \int_{-\infty}^{t} m(t-t_1) \cdot \underline{\underline{Q}}(t_1, t)dt_1 \text{ and}$$
$$(2\rho/G)\underline{\underline{Q}} = 5\overline{<uu>}. \tag{32}$$

Here $\underline{u}$ is the unit end-to-end vector of a strand during deformation, and ⟨ · ⟩ denotes average over configuration space. It assumes reptating motion of polymer chains inside a tube created by surrounding polymer chains. Even though it has the form of single integral type, it contains a functional of statistical averaging inside this integral.

ix) the Currie potential [67]

$$(2\rho/G)F = \frac{5}{2} \cdot \ln[(J-1)/7],$$
$$J = I_1 + 2(I_2 + 13/4)^{1/2}. \tag{33}$$

It is derived as a close approximation to the reptation model shown in eqs. (32).

x) a potential by Yen and McIntire [68]

$$2\rho\tilde{F} = \frac{G_1}{2\alpha\theta_1} \cdot \exp\left(-\frac{t}{\theta_1}\right) \cdot \ln[1+\alpha(I_1-3)] + \frac{G_2}{2\beta\theta_2} \cdot \exp\left(-\frac{t}{\theta_2}\right) \cdot \ln[1+\beta(I_2-3)] + \frac{G_3}{2\theta_3} \cdot \exp\left(-\frac{t}{\theta_3}\right) \cdot (I_1-3). \tag{34}$$

It is a linear combination of simple potential forms, which introduces a partially time-strain separable version of the general potential presented by Zapas [69]. Here G_i's are moduli, θ_i's are relaxation times, α and β are nonlinear parameters and the relationship between the potential $\tilde{F}$ and CE is shown in eqs. (28).

Before concluding this section, several potential forms suggested in the rubber elasticity theory are mentioned below, since they can be applied in principle for the K-BKZ class of integral CEs as well as for the hyper-viscoelastic CEs of differential type. It seems conventional that they are in many cases presented as a function of principal values rather than invariants of the total Finger deformation tensor.

xi) a potential by Ogden [70]

$$2\rho F = \sum_n \frac{G_n}{2\alpha_n}(C_1^{\alpha_n/2} + C_2^{\alpha_n/2} + C_3^{\alpha_n/2} - 3), \tag{35}$$

where α_n is a numerical parameter which can be negative or positive, G_n is a constant with a dimension of modulus and C_i is a principal value of the Finger tensor. It is noticeable that it becomes identical to the Mooney potential [71] if it contains only two specific terms corresponding to $\alpha_1 = 2$ and $\alpha_2 = -2$.

xii) a potential by Valanis and Landel [72]

$$2\rho F = \sum_{i=1}^{3}\left[A \cdot \sqrt{C_i}(\ln\sqrt{C_i} - 1) + \alpha \cdot \ln\sqrt{C_i}\right], \qquad (36)$$

where A and α are parameters. It is based on the concept that the elastic potential for isotropic hyperelastic materials can be represented as a sum of three separate functions of each principal values of the Finger tensor.

xiii) a BST potential [73]

$$2\rho F = \frac{A}{n}I_E + B \cdot I_E^m, I_E = \frac{1}{n}(C_1^{n/2} + C_2^{n/2} + C_3^{n/2}), \qquad (37)$$

where A, n, B and m are all parameters.

For the time-strain separable viscoelastic CEs with potential F (the K-BKZ class), the basic functionals such as the stored free energy W, the extra stress tensor $\underline{\underline{\tau}}$ and the dissipation D are of the form [74]:

$$W = \rho\int_{-\infty}^{t} m(t-t_1)F(I_1, I_2; t, t_1)\,dt_1,$$
$$\underline{\underline{\tau}} = 2\rho\int_{-\infty}^{t} m(t-t_1)\underline{\underline{C}} \cdot \frac{\partial F}{\partial \underline{\underline{C}}}(I_1, I_2; t, t_1)\,dt_1,$$
$$D \equiv \mathrm{tr}(\underline{\underline{\tau}} \cdot \underline{\underline{e}}) - \frac{dW}{dt} = \rho\int_{-\infty}^{t} \frac{dm}{dt_1}(t-t_1) F(I_1, I_2; t, t_1)\,dt_1. \qquad (38)$$

As to the non-potential viscoelastic (or nonhyper-viscoealstic) CEs such as the Wagner equations (24,25), and the models by Papanastasiou *et. al* (26) and by Luo and Tanner (27), their formulation is completely unphysical. When working on very rapid deformations, it is possible to create a perpetual motion machine from a hypothetical material subordinate to this type of rheology [75]. Nevertheless, we summarize the results on this type of CEs too, considering it as mathematical abstraction for the superficial curve fitting of data

2.3. Unified form of Compressible and Incompressible Constitutive Equations

We now introduce a unified set of notations for both differential and single integral types of viscoelastic CEs, which is in principle applicable to the compressible case. For the differential models, we employ only upper convected time derivatives in the evolution equations due to the following reason. It has been proven several times [40,76-78] that the evolution equations with mixed time derivatives are Hadamard unstable (the definition is given in the next section) except for the cases of upper and lower convected derivatives, and it is also well-known that the lower convected time derivative can be equivalently rewritten in the form of upper convected one (e.g. see Ref. [40]).

Introducing a modified pressure term defined as

$$p' = \begin{cases} p & \text{for differential CEs} \\ p + \int_{-\infty}^{t} \bar{\varphi}_2 I_2\,dt_1 & \text{for integral CEs} \end{cases} \qquad (39)$$

and using the Cayley-Hamilton identity and the invariance of rheological variables (say, the extra stress $\underline{\underline{\tau}}$) under arbitrary addition of isotropic terms in the case of incompressibility, we represent both classes of CEs in eqs. (1), (4) and (18), (19) as follows:

$$\underline{\underline{\sigma}} = -p'\underline{\underline{\delta}} + \underline{\underline{\tau}}, \quad \underline{\underline{\tau}} = 2\rho\underline{\underline{c}} \cdot \frac{\delta F}{\delta \underline{\underline{c}}},$$

$$\underline{\underline{\tau}} = \begin{cases} \underline{\underline{E}} & \text{for differential CEs} \\ \int_{-\infty}^{t} \bar{\underline{\underline{E}}}\,dt_1 & \text{for non-separable integral CEs} \\ \int_{-\infty}^{t} m(t-t_1)\underline{\underline{E}}\,dt_1 & \text{for separable integral CEs} \end{cases} \qquad (40)$$

where $\delta/\delta\underline{\underline{c}}$ is in general the partial Fréchet derivative with respect to $\underline{\underline{c}}$, and $\underline{\underline{E}} = 2\rho[\varphi_1\underline{\underline{c}}+\varphi_2(I_1\underline{\underline{c}}-\underline{\underline{c}}^2)+\varphi_3 I_3\underline{\underline{\delta}}]$ with the definition for φ_i in eqs. (4) and (29) while for $\tilde{\underline{\underline{E}}}$ φ_i should be replaced by $\tilde{\varphi}_i$ in eq.(28). In this notation, $\underline{\underline{c}}$ becomes the total Finger strain tensor $\underline{\underline{C}}$ in the case of integral CEs, and thus for the integral and the Leonov CEs it is directly related to volume change, i.e. $I_3 = \det\underline{\underline{c}} = (\rho_0/\rho)^2$ with $\rho_0 = \rho(t=0$ or $t=t_1)$. Here we can formally include the CEs for compressible materials, if we assign $p'=0$ in eqs. (40) since the isotropic pressure is no more indeterminate in the compressible system. Even though the set (40) is written for hyper-viscoelastic equations, the nonhyper-viscoelatic formulation ($\varphi_{ij} \neq \varphi_{ji}$, with a definition for φ_{ij} in eqs. (17)) can also be included by properly specifying φ_i or $\tilde{\varphi}_i$.

At this point, we can find some logical imperfection for differential CEs. In the case of general differential hyper-viscoelastic CEs other than the Leonov class, the formulation of compressibility (otherwise incompressibility) in the constitutive model is not known because of the lack of the relation between the density and the tensor $\underline{\underline{c}}$. In other words, if we choose the potential $F = F(\rho, I_1, I_2, I_3)$ without the relation $I_3 = (\rho_0/\rho)^2$, Murnaghan's relation in eqs.(40) is no longer valid. Hence, strictly speaking, hyper-viscoelastic differential CEs except for the Leonov class lose their physical meaning, and some other substitute for Murnaghan's relation should be sought, which has not yet been discovered.

The (elastic or total) strain tensor $\underline{\underline{c}}$ is the solution of the following evolution problem:

$$\theta\overset{\nabla}{\underline{\underline{c}}}+\underline{\underline{\psi}}(\underline{\underline{c}})=\underline{\underline{0}},\quad \begin{cases} \underline{\underline{c}}\,\Big|_{t=0}=\delta & \text{for differential models} \\ \underline{\underline{c}}\,\Big|_{t=t_1}=\underline{\underline{C}}\,\Big|_{t=t_1}=\underline{\underline{\delta}},\ \underline{\underline{\psi}}(\underline{\underline{c}})=\underline{\underline{0}} & \end{cases}$$

for integral models,

$$\overset{\nabla}{\underline{\underline{c}}}=\frac{\partial\underline{\underline{c}}}{\partial t}+\underline{v}\cdot\underline{\nabla}\underline{\underline{c}}-\underline{\nabla}\underline{v}^T\cdot\underline{\underline{c}}-\underline{\underline{c}}\cdot\underline{\nabla}\underline{v}. \tag{41}$$

Here $\underline{\underline{\psi}}$ is the dissipative term which vanishes for integral CEs, and from now on we consider only upper convected time derivatives even for differential models.

Before concluding this section, we want to modify above unified form especially for the compressible formulation. In mechanics of compressible materials, one usually employs the following transformations to separate volumetric and shear components of deformations:

$$\underline{\underline{c}}=\left(\frac{\rho_0}{\rho}\right)^{2/3}\hat{\underline{\underline{c}}},\ I_1=\left(\frac{\rho_0}{\rho}\right)^{2/3}\hat{I}_1,\ \hat{I}_2=\left(\frac{\rho_0}{\rho}\right)^{4/3}\hat{I}_2,$$

$$I_3=\left(\frac{\rho_0}{\rho}\right)^2,\ \hat{I}_1=\mathrm{tr}\hat{\underline{\underline{c}}},\ \hat{I}_2=,\ \hat{I}_2=(\hat{I}_1^2-\mathrm{tr}\hat{\underline{\underline{c}}}^2)/2=\mathrm{tr}\hat{\underline{\underline{c}}}^{-1},$$

$$\hat{I}_3=\det\hat{\underline{\underline{c}}}=1 \tag{42}$$

In the hyper-viscoelastic case represented in eqs. (40), the potential F and hence the stress tensor $\underline{\underline{E}}$ are now dependent upon a different set of variables such that

$$F(T,I_1,I_2,I_3)=\hat{F}(T,\hat{I}_1,\hat{I}_2,\rho),$$

$$\underline{\underline{E}}=2\rho[-(\rho/2)\ \hat{\varphi}_\rho\underline{\underline{\delta}}+\ \hat{\varphi}_1(\hat{\underline{\underline{c}}}-\hat{I}_1\underline{\underline{\delta}}/3)-\ \hat{\varphi}_2(\hat{\underline{\underline{c}}}^{-1}-\hat{I}_2\underline{\underline{\delta}}/3)] \tag{43}$$

where $\hat{\varphi}_1=\partial\tilde{F}/\partial\tilde{I}_1$, $\hat{\varphi}=\partial\tilde{F}/\partial\tilde{I}_2$ and $\hat{\varphi}_\rho=\partial\tilde{F}/\partial\rho$ of which the relationships with φ_i's are derived in Ref. [79]. Here the nonhyper-viscoelastic case can again be included.

In this type of formulation, the evolution relation (41) is rewritten as

$$\theta\left\{\overset{\nabla}{\hat{\underline{\underline{c}}}}+(2/3)(\underline{\nabla}\cdot\underline{v})\hat{\underline{\underline{c}}}\right\}+\hat{\underline{\underline{\psi}}}(\hat{\underline{\underline{c}}},\rho)=\underline{\underline{0}}, \tag{44}$$

where $\overset{\nabla}{\hat{\underline{\underline{c}}}}$ is the upper convected time derivative of $\hat{\underline{\underline{c}}}$ and $\hat{\underline{\underline{\psi}}}(\hat{\underline{\underline{c}}}, \rho) = (\rho_0/\rho)^{2/3}\underline{\underline{\psi}}(\underline{\underline{c}})$ introduces dissipation to differential CEs.

3. Mathematical Stability of Viscoelastic Constitutive Equations

3.1. Background

In numerical simulation of non-Newtonian flows, degradation of the numerical solution or lack of convergence of computational schemes has been frequently observed for large or even modest values of Deborah numbers. It is thought that the main cause of this instability is the bad choice of a CE for numerical applications (see, e.g. p.314 of Ref. [80]).

The mathematical instability of rheological CEs can be distinguished into two types: (i) Hadamard and (ii) dissipative. Hadamard instability, which shows the unboundedly increasing amplitude of short waves as the wavelength tends to zero, is associated with the nonlinear rapid response of CEs, hence it depends on the type of differential operator in the evolution equation for differential models and the configuration tensor-stress relation, i.e. the elastic potential in the hyper-viscoelastic case. However, the dissipative instability which is inherent only in viscoelastic equations, by definition, results from the dissipative terms of CEs in the case of differential type. For integral equations, the dissipative terms are hidden inside the hereditary integral, and thus the type of instability for integral CEs originated from the characteristics same with those for dissipative unstable differential CEs, will also be called dissipative.

The most distinct is Hadamard instability, which is mathematically understood as the ill-posedness of the Cauchy problem. The Hadamard instability results in catastrophic disaster in numerical calculations: the finer the mesh is, the worse the results degrade [81]. The first example of this type is illustrated by Hadamard for the initial value problem of the elliptic (Laplace) partial differential equation. In the case of more complex equations, this ill-posedness occurs when the time-dependent partial differential equation changes its type from hyperbolicity to ellipticity. Similar instability may be also observed in a steady problem, if the differential equation manifests the change of type from ellipticity to hyperbolicity. We can examine some phenomena in nature associated with this change of type such as transonic flows of compressible media and phase transitions like melting or phase separation. However, the change of type in transonic flows should be distinguished from that in Hadamard unstable CEs. The transonic flow occurs when the velocity exceeds the characteristic wave velocity, but the change of type by unstable CEs is due to the stress level. Many scientists in the field of viscoelastic rheology have also made attempts to associate this type of unstable behavior in CEs to certain physical instabilities of the polymer fluid flow like melt fracture [82,83]. On the other hand, Dupret and Marchal [76] and Leonov [40] pointed out that non-evolutionary models for viscoelastic fluids are no good and should be discarded from any further consideration as in other fields of physics and mechanics. Illustrating one example of flow with singular geometries, involved in most practical applications [76], where a solution is hardly expected to exist for unstable equations, they conclude that only models which never lose evolution should be used.

In viscoelastic fluid mechanics, the Hadamard stability analysis has been first carried out by Rutkevich [84,85] in the case of the Maxwell model, and to some extent by Godunov [86]. Some significant results have been obtained relatively recently by Dupret and Marchal [76] and Joseph and coworkers (see the monograph [81] by Joseph).

Rutkevich considered the Maxwell model [84, 87], and proved that the upper convected, the lower convected and the corotational Maxwell models with a Newtonian viscosity term are always evolutionary. He also obtained the stability conditions for all three equations. Later, it is shown that those stability criteria for the upper and the lower convected Maxwell models are always satisfied [77].

In addition to the results by Rutkevich, for the interpolated Maxwell model which involves the mixed time derivative in the evolution equation, Joseph and coworkers [77,78], Dupret and Marchal [76] and Leonov [40] independently proved that the Hadamard instability always occurs except for the marginal cases of upper and lower convected time derivatives. The ill-posedness of the Johnson-Segalman or Gordon-Schowalter [76] and the original Phan Thien-Tanner [78] CEs is subject to this cause of instability.

Using the general method of characteristic cone orientation, Dupret and Marchal [76] showed that the White-Metzner model exhibits loss of evolution, which has been again justified by Verdier and Joseph [88] who employed a perturbation method in their analysis. It is found that the dependence of the relaxation time on the second invariant of the strain rate tensor causes nonevolutionary behavior. For this model, Verdier and Joseph [88] also noticed one type of dissipative instability similar to the one of an upper convected Maxwell model in elongational flow: the solution goes to infinity whenever the extensional strain rate exceeds half of the reciprocal relaxation time.

As to the mathematical procedure of stability analysis, a general method of characteristics [76] is sometimes complicated and cumbersome. However, in almost every interesting cases, it can be simplified by the introduction of the method called "frozen coefficient" (see, e.g. [81]) when we consider only extremely short and high frequency wave disturbances propagating with a finite speed. Those cases are related to all the CEs of the quasilinear differential type as well as the time-strain separable single integral CEs. Then, the linear stability analysis of the problem is studied locally without considering boundary conditions. Although by following Kreiss' examples [89] this simplified local stability condition is neither necessary nor sufficient for the overall stability in the general nonlinear differential case, in the case of quasilinear differential and time-strain separable single integral equations the local stability analysis with the method of frozen coefficients can be employed without loss of generality [90].

We can see that a number of papers on the evolution of viscoelastic CEs have been published, but the stability analysis has been confined to only simple flow situations for specific differential models. Besides the above-mentioned results, one can see various limited results on the Hadamard stability and on the change of type [91~97].

The stability analysis of Hadamard type has not been developed to produce any concrete result on the evolution character for single integral CEs of both potential and non-potential types until recently. The most general attempt in this direction has been made by Joseph and coworkers [77,81] by formulating rate equations from single integral models. This complete generality is, however, difficult to utilize because the general conditions for stability can be presented only in the form of a hereditary functional, which needs to be analyzed for any particular, seemingly simple flow.

There have been some contradictory arguments about the meaning of nonevolutionary behavior related to thermodynamics. On the basis of local equilibrium postulate, Rutkevich [98] proposed that evolution criteria for the Maxwell fluid are identical to the Second Law of thermodynamics, i.e. the Clausius-Duhem inequality. From this, he concluded the loss of evolution is a consequence of incorrect assignment of the internal energy or the free energy. On the contrary, Dupret and Marchal [76] showed the possibility that the Second Law may be preserved even in the nonevolutionary case. This illustrates the difference between the Second Law and the evolution requirements. For single integral CEs, Renardy suggested and proved a sufficient condition for the Hadamard stability imposed on the potential in the K-BKZ model [99]. This condition may suggest a clue to the implication of evolution criteria associated with thermodynamics.

In the theory of nonlinear elasticity, the method of stability analysis has been well established and the implications of stability are understood in great detail. Among many conditions suggested, the simplest stability constraint known as the Baker-Ericksen inequality [100] means that the greater principal stress occurs always in the direction of the greater principal stretch. The thermodynamic stability condition for hyperelastic solids called the GCN^+ condition (see the section 52 in [3]) is identical to the convexity of the thermodynamic potential with respect to the Hencky strain measure, which has also been known as a condition for the strong monotonicity of stress with respect to strain. The Hadamard stability of field equations in theoretical elasticity, corresponds to the strong ellipticity, the condition of which implies the stability requirement in dynamic situation, whereas the ordinary ellipticity produces the constraint in static relaxed state [101].

The GCN^+ condition has a close relationship with the strong ellipticity condition. The lack of symmetry of a second rank tensor in the representation of stability conditions causes more restrictive conditions for the strong ellipticity than for the GCN^+ condition [3]. Therefore, such inequalities as the Baker-Ericksen and the GCN^+ may be regarded as necessary conditions for the strong ellipticity or for the Hadamard stability.

For isotropic compressible hyperelastic solids, the necessary and sufficient conditons of ordinary and strong ellipticity have been obtained in the case of finite plane equilibrium deformations by Knowles and Sternberg [102]. In the general 3-D case, the necessary and sufficient condition of strong ellipticity for equations governing an isotropic compressible material has also been established in the paper [103]. Both ordinary and strong ellipticity conditions are formulated in a form of algebraic inequalities by Zee and Sternberg [101] in the equilibrium theory of isotropic incompressible hyperelastic solids. Additionally, they demonstrated a close relationship between the ordinary and strong ellipticity, such that the Baker-Ericksen inequality combined with the ordinary ellipticity constraint is equivalent to the strong ellipticity condition.

In the viscoelastic case, general results on global Hadamard stability, i.e. stability for any type of flow and for any Deborah number, has been recently proposed for both general classes of quasilinear Maxwell-like CEs [40] and time-strain separable single integral CEs [104]. Later, it has been proved that the algebraic criteria for Hadamard stability are in reality the necessary and sufficient conditions for thermodynamic stability, that is, the GCN^+ or the convexity conditions for thermodynamic potential in the hyperelastic case, which impose weaker constraints on CEs than the criteria for Hadamard stability [105]. The global Hadamard stability condition has been first obtained for 2-dimensional flow system in Ref. [74] for the separable single integral CEs. Then the complete 3-dimensional conditions for the global Hadamard stability in the incompressible case are derived in the paper [105] by introducing the constitutive formalism in Refs. [40,104] and following the algebraic procedure in the paper [101], and many of popular CEs are analyzed by the application of obtained criteria. In the compressible flow of viscoelastic fluid, the global Hadamard stability is also considered for the formalism including both differential and separable single integral CEs [79].

Results regarding the dissipative instability of CEs have been very rare. 1-D instability of the Giesekus model within a certain range of a parameter is reported for the plane Couette [106] and the Poiseuille flows [107]. Those papers show that the instability occurs when the numerical parameter is greater than 1/2 and shear rate exceeds a certain critical value. This type of dissipative instability is related to the decreasing branch of the steady shear flow curve. Similar results can be seen for the Larson as well as the Giesekus CE in Refs. [108,109], where another type of "blow-up" instability of the Larson, the simplest Leonov and the Giesekus models under step shear stress exceeding some bounded stress maximum (or supremum) is demonstrated. As for the Larson model which is later proved Hadamard unstable [105], the breakage of the sample specimen is also calculated in simple elongational flow under long wave disturbance [108].

In Ref. [40], one can also find two theorems proven for the Maxwell-type CEs, one of which illustrates positive definiteness of the configuration tensor in some restricted situation, which was first proven by Hulsen [110] in a different way. The positive definiteness of the configuration tensor is required for stability, since its violation immediately yields the Hadamard instability. Another theorem gives a useful sufficient condition for the boundedness of variables in the Maxwell-type CEs, the satisfaction of which guarantees the dissipative stability also for a limited flow history. On the other hand, the necessary and sufficient condition for the boundedness of the solution for the integral CEs is established in the paper [74]. However, both the theorems assume a predefined strain history, hence whenever mixed stress-strain history is given, they cannot be applied to dissipative stability analysis. It should be also mentioned that this type of instability can occur even if the dissipation is positive definite.

3.2. Hadamard Stability Criteria for Viscoelastic Constitutive Equations

In this section, following the procedure of papers [79,105], we outline the mathematical scheme of obtaining the global Hadamard stability conditions for quasilinear differential and time-strain separable single integral CEs in incompressible or compressible isothermal flow of viscoelastic liquid.

The total set of equations in this stability problem consists of viscoelastic CEs (40) with (39) and (41) or with (39), (43) and (44), and the following momentum and mass balance equations:

$$\rho\frac{d\underline{v}}{dt}=\underline{\nabla}\cdot\underline{\underline{\sigma}},\quad \frac{d\rho}{dt}+\rho\underline{\nabla}\cdot\underline{v}=0. \qquad (45)$$

Here for simplicity, we neglect the force exerted on the mass of the body. Then assuming that the set has a solution $\{\underline{\underline{c}},\ \underline{v},\ p',\ \rho\}$ which satisfies some proper initial and boundary conditions, we impose on the solution extremely short and high frequency, infinitesimal waves of disturbances

$$\{\delta\underline{\underline{c}}, \delta\underline{v}, \delta p', \delta\rho\} = \varepsilon\{\bar{\underline{\underline{c}}}, \bar{\underline{v}}, \bar{p}, \bar{\rho}\} \cdot \exp[i(\underline{k} \cdot \underline{x} - \omega t)/\varepsilon^2], \tag{46}$$

where ε is a small amplitude parameter ($|\varepsilon| \ll 1$) and also implies the short wavelength and high frequency of the waves. $\bar{\underline{\underline{c}}}$, $\bar{\underline{v}}$, $\bar{p}$ and $\bar{\rho}$ are (generally complex) amplitudes of the corresponding disturbances, $\underline{k}$ is a wave vector, and ω is the frequency. For incompressible system, the density is constant, and thus its fluctuation vanishes, i.e. $\delta\rho = 0$.

Considering the local linear stability analysis, we can easily find the following "dispersion relation", that is, the dependence of the frequency ω on the wave vector and the parameters of the basic flow:

$$\frac{1}{2}\Omega^2\bar{v}^{-2} = \begin{cases} B_{ijmn}\bar{v}_i k_j \bar{v}_m k_n & \text{for differential models} \\ \int_{-\infty}^{t} m(t-t_1) B_{ijmn} dt_1 \cdot \bar{v}_i k_j \bar{v}_m k_n & \text{for integral models.} \end{cases} \tag{47}$$

Here $\Omega = \omega - \underline{k} \cdot \underline{v}$ is the frequency of oscillations with Doppler's shift on the basic velocity field $\underline{v}$ taken into account, and the fourth rank tensor B_{ijmn} is defined in the principal axes of $\underline{\underline{c}}$ with principal values c_i as

$$\begin{aligned} B_{ijmn} &= \delta_{im}\delta_{jn}G_{ij} + \delta_{ij}\delta_{mn}L_{im} && \text{(no sum)}, \\ G_{ij} &= [\varphi_1 + \varphi_2(I_1 - c_i - c_j)] \cdot c_j && \text{(no sum)}, \\ L_{ij} &= [\varphi_2 + 2\varphi_{11} + 2\varphi_{22}(I_1 - c_i)(I_1 - c_j) \\ &\quad + 2\varphi_{12}(I_1 - c_j) + 2\varphi_{21}(I_1 - c_i)]c_i c_j \\ &\quad + \chi[\varphi_3 + 2\varphi_{33}I_3 + 2\varphi_{23}(I_1 - c_i)c_i \\ &\quad + 2\varphi_{32}(I_1 - c_j)c_j + 2\varphi_{13}c_i + 2\varphi_{31}c_j]I_3 && \text{(no sum)}, \end{aligned}$$

$$\begin{cases} \chi = 0, \ \bar{v}_i k_i = 0 & \text{in the case of incompressibility} \\ \chi = 1, \ \bar{v}_i k_i \neq 0 & \text{in the case of compressibility.} \end{cases} \tag{48}$$

In this analysis, the difference between stability problems of incompressible and compressible systems consists in the possibility of longitudinal wave propagation. For the incompressible system, the speed of the longitudinal wave approaches infinity, whereas the speed of the transverse wave is finite. Hence, perturbation of basic solutions by the longitudinal wave is not considered in this stability problem, and the wave vector is always orthogonal to the vector disturbing the velocity field ($\bar{v}_i k_i = 0$). However, for the compressible material, the speeds of both waves have finite values. Thus for stability, the initially infinitesimal amplitude of distrubing waves of either type (or a mixed type) should remain small at subsequent moments.

Now from eq. (46) with the definition of Ω, one can see that $\Omega^2\bar{v}^2/2 > 0$ is required for stability. Therefore, the necessary and sufficient condition for global Hadamard stability reduces to

$$\frac{1}{2}\Omega^2\bar{v}^{-2} = B_{ijmn}\bar{v}_i k_j \bar{v}_m k_n > 0. \tag{49}$$

On the other hand, the convexity of potential or the GCN$^+$ condition can be represented as

$$\bar{B}_{ijmn}\beta_{ij}\beta_{mn} > 0, \tag{50}$$

where

$$\bar{B}_{ijmn} = \frac{\partial^2 F}{\partial h_{mn}\partial h_{ij}} = 4c_{mq} \cdot \frac{\partial}{\partial c_{qn}}\left(c_{ip} \cdot \frac{\partial F}{\partial c_{pj}}\right). \tag{51}$$

h_{ij} is the Hencky strain measure with $\underline{\underline{h}} = (1/2)x \ln\underline{\underline{c}}$, and the inequality (50) should be satisfied for thermodynamic stability with respect to an arbitrary symmetric tensor β_{ij} (for incompressible system $\text{tr}\underline{\underline{\beta}} = 0$). In the potential case (hyper-viscoelastic case), the identity $B_{ijmn} = \bar{B}_{ijmn}$ holds. When comparing inequalities (49) and (50), one can notice that due to the symmetry of the tensor β_{ij}, the condition (50) imposes weaker stability constraints than the inequality (49), that is, the Hadamard stability conditions are stronger than those of GCN$^+$.

Employing the algebraic procedure applied in hyperelasticity [101,103], we can finally obtain the necessary and sufficient condition for the global Hadamard stability as the following:

Incompressible System

(i) $\mu_i > 0$ or $\varphi_1 + \varphi_2 c_i > 0$,

(ii) $\zeta_i + 2\mu_i > 0$,

(iii) $\left[\sqrt{\zeta_i + 2\mu_i} + \sqrt{\zeta_j + 2\mu_j}\right]^2 > \zeta_k - 2\mu_k$,

$(i \neq j \neq k)$, (52)

where

$$\mu_i = (\varphi_1 + \varphi_2 c_i)\sqrt{c_j c_k} \quad (i \neq j \neq k),$$

$$\zeta_i = (I_1 - c_i)(\varphi_1 + \varphi_2 c_i) + 2\left(I_1^2 - 2I_2 - c_i^2 - \frac{2I_3}{c_i}\right)$$

$$[\varphi_{11} + (\varphi_{12} + \varphi_{21})c_i + \varphi_{22}c_i^2]. \quad (53)$$

Compressible System

In the compressible case, we present the conditions written only in separated variables $\hat{\underline{\underline{c}}}$ and ρ defined in eqs.(42). Representation in terms of the original configuration tensor $\underline{\underline{c}}$ can be found in Ref. [79].

(i) $\alpha_i > 0$ and $\gamma_i > 0 \quad (i = 1, 2, 3)$,

(ii) $w_i \equiv \alpha_i\sqrt{\hat{c}_j \hat{c}_k} + \sqrt{\gamma_j \gamma_k} > |\beta_i|, \quad (i \neq j \neq k)$,

(iii) $\left(\alpha_1\sqrt{\hat{c}_2\hat{c}_3} + \beta_1\theta_1\right)\sqrt{\gamma_1} + \left(\alpha_2\sqrt{\hat{c}_1\hat{c}_3} + \beta_2\theta_2\right)\sqrt{\gamma_2}$

$+ \left(\alpha_3\sqrt{\hat{c}_1\hat{c}_2} + \beta_3\theta_1\theta_2\right)\sqrt{\gamma_3} + [2(w_1 + \beta_1\theta_1)$

$(w_2 + \beta_2\theta_2)(w_3 + \beta_3\theta_1\theta_3)]^{1/2} + \sqrt{\gamma_1\gamma_2\gamma_3} > 0$

for all four choices of $\theta_1, \theta_2 = \pm 1$. (54)

Here

$$\alpha_i = \hat{\varphi}_1 + \hat{c}_i\hat{\varphi}_2,$$

$$\beta_i = \left[\frac{5}{9}\hat{I}_1 - \frac{2}{3}(\hat{c}_j + \hat{c}_k)\right]\hat{\varphi}_1 + \left(\frac{2}{9}\hat{I}_2 - \frac{1}{3}\hat{c}_j\hat{c}_k\right)\hat{\varphi}_2$$

$$+ \rho\hat{\varphi}_\rho + 2\left[\alpha_m^{(j)}\right]\left[\hat{\Phi}_{mn}\right]\left[\alpha_n^{(k)}\right]^T, \quad (i \neq j \neq k),$$

$$\gamma_i = \left(\frac{5}{9}\hat{I}_1 - \frac{1}{3}\hat{c}_1\right)\hat{\varphi}_1 + \left[\frac{14}{9}\hat{I}_2 - \frac{5}{3}(\hat{I}_1 - \hat{c}_i)\hat{c}_i\right]\hat{\varphi}_2$$

$$+ \rho\hat{\varphi}_\rho + 2\left[\alpha_m^{(i)}\right]\left[\hat{\Phi}_{mn}\right]\left[\alpha_n^{(i)}\right]^T,$$

$$\left[\hat{\Phi}_{ij}\right] = \begin{bmatrix} \hat{\varphi}_{11} & (\hat{\varphi}_{12} + \hat{\varphi}_{21})/2 & (\hat{\varphi}_{1\rho} + \hat{\varphi}_{\rho 1})/2 \\ (\hat{\varphi}_{12} + \hat{\varphi}_{21})/2 & \hat{\varphi}_{22} & (\hat{\varphi}_{2\rho} + \hat{\varphi}_{\rho 2})/2 \\ (\hat{\varphi}_{1\rho} + \hat{\varphi}_{\rho 1})/2 & (\hat{\varphi}_{2\rho} + \hat{\varphi}_{\rho 2})/2 & \hat{\varphi}_{\rho\rho} \end{bmatrix}$$

$$\left[\alpha_m^{(i)}\right] = \left[\alpha_1^{(i)}\alpha_2^{(i)}\alpha_3^{(i)}\right] = \left[\hat{I}_1/3 - \hat{c}_i, -(\hat{I}_2/3 - \hat{c}_i^{-1}), \rho/2\right],$$

$$\hat{\varphi}_{ij} = \partial\hat{\varphi}_i/\partial\hat{I}_j, \quad \hat{\varphi}_{i\rho} = \partial\hat{\varphi}_i/\partial\rho, \quad \hat{\varphi}_{\rho j} = \partial\hat{\varphi}_\rho/\partial\hat{I}_j,$$

$$\hat{\varphi}_{\rho\rho} = \partial\hat{\varphi}_\rho/\partial\rho \quad (i, j = 1, 2). \quad (55)$$

Hence, the criteria (52) and (54) are obtained in the form of algebraic inequalities, which should be imposed on CEs to prevent unphysical instabilities in the whole range of Deborah numbers. Regarding the biquadratic form (49) for the compressible system, a little algebraic manipulation yields

$$\frac{1}{2}\Omega_v^{2-2} = \hat{B}_{ijmn}\bar{v}_i k_j \bar{v}_m k_n + [y_i][m_{ij}][y_j]^T > 0, \quad (56)$$

where

$$2m_{ij} = m_i + m_j, \quad [y_i] = [\bar{v}_1 k_1 \ \bar{v}_2 k_2 \ \bar{v}_3 k_3],$$

$$m_i = \left(\frac{5}{9}\hat{I}_1 - \frac{4}{3}\hat{c}_i\right)\hat{\varphi}_1 + \left[\frac{14}{9}\hat{I}_2 - \frac{8}{3}(\hat{I}_1 - \hat{c}_i)\hat{c}_i\right]\hat{\varphi}_2$$

$$+ \rho\hat{\varphi}_\rho + \hat{I}_1\left(\frac{2}{9}\hat{I}_1 - \frac{4}{3}\hat{c}_i\right)\hat{\varphi}_{11} + \hat{I}_2\left[\frac{8}{9}\hat{I}_2 - \frac{8}{3}(\hat{I}_1\right.$$

$$\left. - \hat{c}_i)\hat{c}_i\right]\hat{\varphi}_{22} + \frac{1}{2}\rho^2\hat{\varphi}_{\rho\rho} + \left[\frac{4}{9}\hat{I}_1\hat{I}_2 - \frac{2}{3}\hat{I}_1(\hat{I}_1 - \hat{c}_i)\right.$$

$$\left.\hat{c}_i - \frac{4}{3}\hat{I}_2\hat{c}_i\right](\hat{\varphi}_{12} + \hat{\varphi}_{21}) + \rho\left(\frac{1}{3}\hat{I}_1 - \hat{c}_i\right)(\hat{\varphi}_{1\rho} + \hat{\varphi}_{\rho 1})$$

$$+ \rho\left[\frac{2}{3}\hat{I}_2 - (\hat{I}_1 - \hat{c}_i)\hat{c}_i\right](\hat{\varphi}_{2\rho} + \hat{\varphi}_{\rho 2}) \text{ (no sum)} \quad (57)$$

The fourth rank tensor $\hat{B}_{ijmn}$ is the same as B_{ijmn} for the incompressible system in eqs. (48) but represented by $\hat{c}_j$, $\hat{I}_j$, $\hat{\varphi}_j$ and $\hat{\varphi}_{ij}$ instead of c_j, I_j, φ_j and φ_{ij}, respectively. If we consider at this point one particular system perturbed only by shear waves ($\bar{v}_i k_i = 0$), then it can readily be seen that positiveness of the biquadratic form $\hat{B}_{ijmn}\bar{v}_i k_j \bar{v}_m k_n$ in eq.(28) is the condition for the global Hadamard stability, since $m_{ij}y_i y_j = 0$ under the disturbance of these equivolumetric waves. Hence, the above consideration simply indicates that the necessary and sufficient condition (52) with eqs.(53) becomes the necessary one for the Hadamard stability of compressible systems when it is rewritten in terms of $\hat{c}_j$, $\hat{I}_j$, $\hat{\varphi}_j$ and $\hat{\varphi}_{ij}$ instead of c_j, I_j, φ_j and φ_{ij}.

Before the necessary and sufficient condition of global Hadamard stability being established for viscoelastic fluid, there have been suggested sufficient conditions for the incompressible system as the following, which are of importance especially in formulating new CEs:

① Leonov's condition (17): the thermodynamic potential F for the Leonov class of viscoelastic models is a monotonically increasing convex function of invariants I_1 and I_2 [25,40].

② Renardy's condition: the thermodynamic potential F for the K-BKZ class of CEs is a monotonically increasing convex function of invariants $\sqrt{I_1}$ and $\sqrt{I_2}$ [99].

Even though Renardy's condition is proved only for the K-BKZ class, it is also sufficient for Hadamard stability when it is applied to differential Maxwell-like CEs with upper convected time derivatives [105]. Since the condition by Leonov is stronger than Renardy's, it also guarantees the global Hadamard stability for the K-BKZ class. As to the compressible CEs, one sufficient condition for the global stability is suggested [79], but its complexity still prevents it from being easily utilized as a criterion for formulating new stable CEs.

3.3. Dissipative Stability Criteria for Viscoelastic Constitutive Equations

As mentioned before, there exists another type of instability related to the specifications of dissipative terms in viscoelastic CEs. This instability may happen due to a poor formulation of the dissipative term $\underline{\underline{\psi}}_\zeta$ (or $\underline{\underline{\psi}}$ when $\zeta = 1$) in eqs. (1) even for the Hadamard stable CEs where the dissipation is also positive definite. For single integral CEs the instability results from fading memory effects in eq. (18). Although the global criteria for dissipative stability of viscoelastic CEs are far from complete (if it is in general possible), we discuss in this section two specific criteria that have already been proven. In the case of compressible flow, no theorem on disspative stability is known yet, but the following theorems are presumably valid also for the compressible CEs, when they are represented and applied to CEs in separated variables such as $\hat{\underline{\underline{c}}}$.

<u>Criterion I of Dissipative Stability</u>

Theorem 1.1: The case of differential CEs [40]
Consider the set of upper convected Maxwell-like CEs (40) and (41) with the positive dissipation $D = D(T, I_1, I_2, I_3)$ defined in eqs. (8). Let the free energy F be a non-decreasing smooth function of three invariants I_k. If for any positive number E, the asymptotic inequality

$$D > E \cdot \|\underline{\underline{\tau}}\|, \quad \|\underline{\underline{c}}\| \to \infty \quad \left(\|\underline{\underline{b}}\| \equiv (\mathrm{tr}\,\underline{\underline{b}}^2)^{1/2} \right) \tag{58}$$

holds, then in any regular flow, the configuration tensor $\underline{\underline{c}}$ and the stress tensor $\underline{\underline{\tau}}$ are limited.

Theorem 1.2: The case of single integral CEs [74]
In any regualr flow, the functionals of free energy and dissipation in eqs. (38) are bounded if

and only if the thermodynamically or Hadamard stable potential function $\bar{F}(H_1, H_2, H_3)$ expressed in terms of principal Hencky strains H_k increases more slowly than exponentially.

In theorem 1.2, principal values of Hencky strain tensor and Finger tensor for the total deformation are related as

$$H_i = (1/2)\ln C_1 \ \ or \ \ \underline{\underline{H}} = (1/2)\ln \underline{\underline{C}}, \ \ \mathrm{tr}\underline{\underline{H}} = 0. \qquad (59)$$

Detailed proofs and definitions of terminology are given in the cited papers or in Ref.[105]. While Theorem 1.1 has been proved for differential CEs as a sufficient condition close to the necessary one, Theorem 1.2 is a necessary and sufficient condition for integral CEs.

Establishing above theorems was initially motivated by the fact that the upper convected Maxwell model, globally Hadamard stable, displays the unbounded growth of stress in simple extension when the elongation rate exceeds the half of the reciprocal relaxation time. In consequence, the theorems result in the following: (i) the upper convected Maxwell model which violates Criterion I [40], and (ii) the K-BKZ class with a potential F represented as an increasing rational polynomial function of basic invariants [74], are dissipative unstable. Therefore, the Mooney and the neo-Hookean potentials as well as the potentials for the K-BKZ class of CEs which are subordinate to Renardy's sufficient evolution criterion also violate Criterion I of dissipative stability [74].

Regarding differential CEs, one cannot guarantee even the positive definiteness of the tensor $\underline{\underline{c}}$, the violation of which immediately causes Hadamard instability. In some limited situation, one theorem on the positive definiteness has been proved by Hulsen [110] and in Ref. [40]. It states that for any given piecewise smooth strain history with the initial condition $\underline{\underline{c}} = \underline{\underline{\delta}}$, the principal values of tensor $\underline{\underline{c}}$ are positive. Hence, the Criterion I of dissipative stability as well as the theorem on positive definiteness of $\underline{\underline{c}}$ in differential CEs assumes the pre-defined strain history (or the regular flow). In the usual case of complex flow where not a strain history but a mixture of strain and stress histories are given, Criterion I cannot be applied.

Since the satisfaction of Criterion I alone cannot prevent the severe dissipative instability, an additional criterion for dissipative stability is introduced.

Criterion II of Dissipative Stability [105]

For the stability of Maxwell-like and time-strain separable single integral CEs, it is necessary that both the steady flow curves in simple shear and in simple elongation have to be monotonically and unboundedly increasing with respect to the strain rate.

In Refs. [108,109], it is shown that the violation of Criterion II incurs "blow-up" instability or even negative values of diagonal components of $\underline{\underline{c}}$ in simple shear. Here we show some examples of this type of instability for the simpest Leonov, the simplest Giesekus and the Larson models.

For those three CEs, the steady flow curves in simple shear (dimensionless shear stress $\hat{\sigma}$ vs. dimensionless shear rate Γ) are shown in Figs. 1 and 2. Evidently, they violate Criterion II of dissipative stability. When we apply step stress (therefore the stress history is prescribed) greater than the maximum (or the supremum) in the flow curves, we can observe severe blow-up instability illustrated in Figs. 3-5, where rheological variables go to infinity in a finite time [109].

These examples clearly demonstrate the validity of Criteria I and II. Even though all three models satisfy Criterion I [40], under specified stress history they show dissipative instability

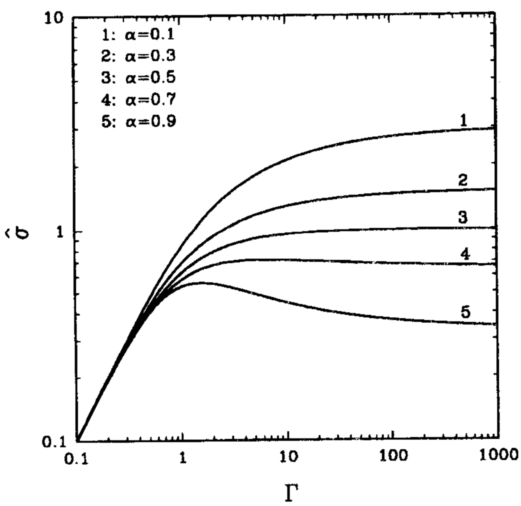

Fig. 1. Dimensionless shear stress of the Giesekus model or the Leonov model ($\alpha = 1/2$) plotted versus dimensionless shear rate in steady simple shear flow.

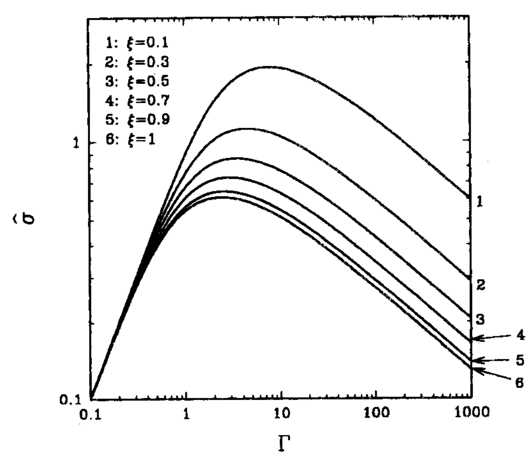

Fig. 2. Dimensionless shear stress of the Larson model plotted versus dimensionless shear rate in steady simple shear flow.

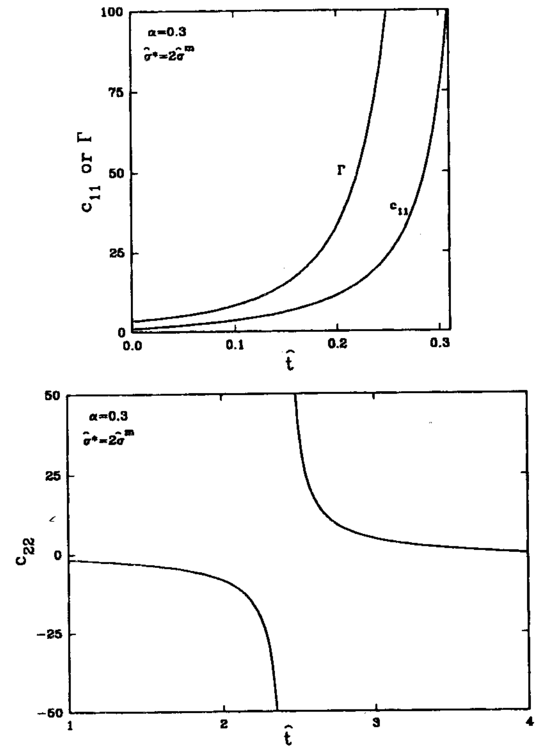

Fig. 3. Blow-up instability of various rheological variables exhibited by the Giesekus model in creep shear flow ($\alpha = 0.3$, $\hat{t}$: dimensionless time).

since they violate Criterion II of dissipative stability. In the paper [105], it is assumed that the subordination to the combined criterion "I+II" is presumably sufficient for the dissipative stability of both differential Maxwell-like and time-strain separable single integral CEs, at least in simple flows.

3.4. Application Results and Discussion

The problem of global Hadamard stability for quasilinear differential and time-strain separable single integral models is completely resolved recently in both cases of incompressible and compressible isothermal flows of viscoelastic fluid. Concerning dissipative stability, even after the study performed in recent years, the global analysis is far from being completed. However, two distinct patterns of dissipative instability have been revealed.

In fact, it has been very controversial on how to distinguish the unstable behaviors caused by poor modeling of CEs and the observed physical instabilities which CEs should also describe. So far there have been a lot of attempts in the literature to apply unstable CEs to real flow instabilities like melt fracture. For example, the

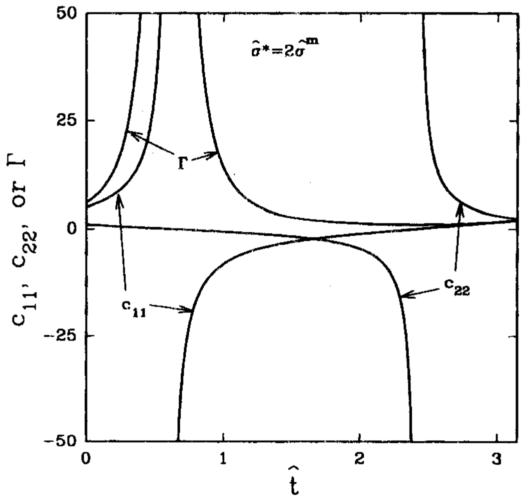

Fig. 4. Blow-up instability of various rheological variables exhibited by the Leonov model in creep shear flow.

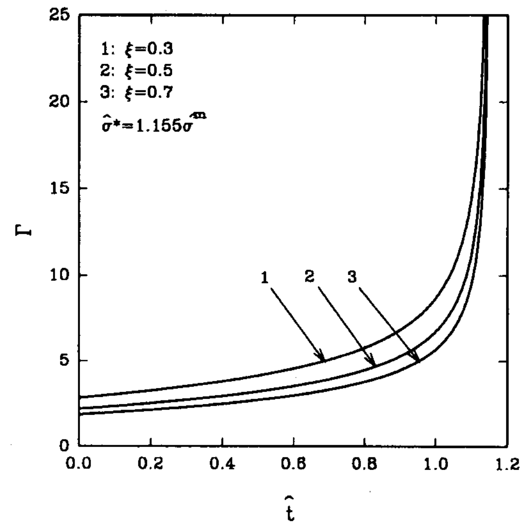

Fig. 5. Blow-up instability of dimensionless shear rate exhibited by the Larson model in creep shear flow.

hypothesis of short memory is employed for the explanation of these physical phenomena [111], but it turned out that this instability is related to the change of type and furthermore some inconsistency appeared due to the use of different equations for the basic flow [82]. Another approach can be found in the work by Dunwoody and Joseph [112], where they obtained stability criteria of shear flows by applying long wave perturbations to CEs.

It is generally agreed that the melt fracture is a phenomenon related to stick-slip process of polymeric fluids along the wall. Therefore, this problem has nothing to do with the unstable behavior of CEs, and it should be treated as an adhesion problem of liquid on the wall under intensive flow. In author's opinion [105], Hadamard and dissipative instabilities of viscoelastic CEs are the genetic flaws incurred by a bad formulation of various terms in CEs, and the occurrence of either Hadamard instability or/and ill-posedness in 1-D situations without such physical reasons as phase transition, etc., is a distinct sign of inappropriateness in the CEs. Hence we can treat the instabilities demonstrated in these works as being associated not with the real instabilities observed in flows of polymer melts, but rather with the improper modeling of terms in CEs. Any CE with any type of instability described herein, should be discarded from any further application, however well it describes viscometric data and however deep physical meaning it may contain. An attempt to apply the unstable behavior of equations to real flow instability would cause inconsistency with other sets of experimental data, and this kind of contradiction has been already demonstrated recently in the paper [113].

In numerical simulation of complex flows with unstable CEs, when the flow rate becomes high enough, occurrence of various types of unphysical instabilities is inevitable. Even in the range of the moderate Deborah number, the existence of singular points in flow geometry such as the corner singularity in die entrance region, is sufficient to spoil the whole numerical procedure.

All the results of the stability analyses found in various studies for popular viscoelastic CEs, are summarized in Table 1. Details of applications of stability criteria can be seen in the cited papers. Table 1 also shows the Hadamard instability of the Doi-Edwards model which is not proved anywhere. However, if we are aware of the behavior of the Doi-Edwards model that in the elastic limit the shear stress exhibits decreasing branch with respect to step shear strain, the proof of instability becomes trivial (the instability of the decreasing branch in the case of time strain separability is explained in Ref. [114]). It is noteworthy that such CEs derived from molecular approaches as the Larson and the Currie models including the Doi-Edwards CE, exhibits the most unstable behavior. Surprisingly enough, none of the time-strain separable single integral models are evolutionary (it is also found that simultaneous satisfaction of the Hadamard stability criterion and Criterion I of dissipative stability is almost impossible for time-strain separable single integral CEs). Recently, Simhambhatla [114] analyzed extensively the time-strain separ-

Table 1. Stability of viscoelastic constitutive equations

Model (Eq. #)	Type of CE	Type of Instability	Reference
Upper convected Maxwell (10) ($\zeta=1$)	Quasilinear differential	Dissipative unstable (Criterion I)	e.g. 40
Interpolated Maxwell (Johnson-Segalman, Gordon-Schowalter) (10)	Quasilinear differential	Hadamard unstable	40, 76, 77, 78
General Phan Thien-Tanner (11)	Quasilinear differential	Hadamard unstable	40, 76, 77, 78
Upper convected Phan Thien-Tanner (11) ($\zeta=1$)	Quasilinear differential	Hadamard stable; dissipative stability depends on dissipative terms	105
White-Metzner (12)	Nonlinear differential	Hadamard and dissipative unstable (Criterion I)	76, 88
FENE (13)	Quasilinear differential	Globally Hadamard and dissipative stable	105
Giesekus (14)	Quasilinear differential	Dissipative unstable (Criterion II)	108
Simplest Leonov (16) ($b_1=b_2=1$)	Quasilinear differential	Dissipative unstable (Criterion II)	108
Leonov CE (16) under specified stability constraints (17)	Quasilinear differential	Globally Hadamard and dissipative stable	105
Larson differential (15)	Quasilinear differential	Hadamard and dissipative unstable (Criterion II)	105, 108
Wagner I (24)	Separable single integral	Hadamard unstable	105
Wagner II (25)	Separable single integral	Hadamard unstable	105
Papanastasiou (26)	Separable single integral	Hadamard unstable	74
Luo-Tanner (27)	Separable single integral	Hadamard unstable	105
Lodge (30)	Separable K-BKZ	Dissipative unstable (Criterion I)	74
K-BKZ class under Renardy's condition	Separable K-BKZ	Dissipative unstable (Criterion I)	74
Larson-Monroe potential (31)	Separable K-BKZ	Hadamard and dissipative unstable (Criterion II)	74, 105
Currie potential (33)	Separable K-BKZ	Hadamard and dissipative unstable (Criterion II)	105
Yen-McIntire (34)	Quasi-separable K-BKZ	Dissipative unstable (Criterion I)	74
Doi-Edwards (32)	separable integral	Hadamard unstable	128

ability concept for viscoelastic CEs. He verified the invalidity of time-strain separability, in more detail, the Hadamard stable CEs of the separable type cannot properly describe the experimental data of stress relaxation after stepwise loading. Interestingly, such violation of separability in short time scale has already been observed experimentally by Einaga *et al.* [115]. Astonishingly, many CEs become Hadamard unstable even in viscometric flow region.

For CEs of the differential type, only 3 stable specifications exist. These are the FENE, the upper convected Phan Thien-Tanner models, and the Leonov class of CEs under convexity constraint (17) for elastic potential. However, besides thermodynamic inconsistency (invalidity of Murnaghan's relation explained in the previous section), both the FENE and the upper convected Phan Thien-Tanner models predict zero value for the second normal stress difference in simple shear flow, which contradicts the experimental evidence for polymer melts and concentrated polymer solutions. Hence, it may be expecting too much for the FENE and the upper convected Phan Thien-Tanner models to reasonably describe whole set of experimental data. The difficulty of describing the experimental data has been reported in quite a few publications and it is briefly discussed in the following section.

In addition, for any successful numerical modeling of high Deborah numer flows of polymer melts, compliance with the stability criteria must be taken very seriously. The importance of stability may be implied by the fact that (within author's knowledge) the high Deborah number flow of abrupt contraction in the die entrance region has been satisfactorily simulated only with the Leonov model until now [116].

It should be noted that all the necessary and sufficient conditions described for single mode CEs become only sufficient for the multi-modal approach if they are applied to each Maxwellian mode. Even though the necessity is not proven, it is thought that the threshold of instability would be only delayed to some higher Deborah number region in multi-modal approach if the single mode case is unstable.

Table 1 shows that the combined stability criteria impose very tough constraints on viscoelastic CEs (the Hadamard stability constraints for the compressible CEs are even stronger than for the incompressible CEs). Therefore, the serious question arises whether there exists a CE or a class of CEs which can properly describe all the available rheometric data for concentrated polymer solutions and melts, when satisfying all the stability constraints. Recently, the paper [55] has demonstrated that such a class does exist, and it is discussed in the following.

4. Rheological Modeling of Viscoelastic Polymer Liquids

Although the descriptive ability is another crucial feature of viscoelastic CEs, its review will not be presented in detail. The flow effects predicted by constitutive models can be found, e.g. in the monographs [22,28,31-34]. A good review on this field of study may be in the monograph by Larson [28], which also contains brief derivations of CEs and some comparisons of the descriptions of various models with experimental data.

In the field of rheology of viscoelastic liquids, there has been a prevailing perception that none of CEs may properly describe the whole set of experimental data obtained for polymer melts or solutions. It was right in some sense until recent years. Indeed, almost none of rheological models could describe consistently the whole set of data with one set of parameters (see chapter 7 in Ref. [28]). As to the single integral CEs, the same difficulty was again recently reported [117,118]. However, all pessimistic precautions on rheological modeling notwithstanding, Leonov and Simhambhatla [55] recently verified the existence of desirable CEs. With the Leonov class of CEs proposed in 1976 [25] which satisfies all stability criteria, they demonstrated excellent correspondence of model descriptions with all experimental data available for such extensively characterized polymer melts as Melt I (LDPE), HDPE, polystyrene and polyisobutylene. In this review, we examine those results.

In their study of model description [55], instead of the simplest Leonov model which uses only the parameters of the discretized linear viscoelastic spectrum, they employed a highly nonlinear specification of the general Leonov class of CEs. This choice eliminates some of the often discussed deficiencies of the simplest model. Of all polymer liquids, LDPE Melt I is the most extensively characterized in rheological testing. Differnt batches of the same resin referred to as IUPAC A and IUPAC X have virtually indistinguishable rheological properties. This polymer with its long side branches, has also proven to be one of the most difficult for viscoelastic modeling. Hence, here we present only the comparison between the description of the Leonov CE and the experimental data on Melt I.

For LDPE Melt I of which material specification is listed in Table 2, they specified the following functional form for the dissipative

Table 2. Molecular Weight and molecular weight distribution of LDPE Melt I

	M_w	M_w/M_n	Reference
LDPE Melt I	460000	22	129

term in eqs. (16) and the neo-Hookean potential:

$$b_1(I_1, I_2) = b_2(I_1, I_2) = (I_2/I_1)^m,\ (2\rho/G)F = I_1 - 3,\quad m > 0. \tag{60}$$

The parameter m was chosen to be 1.4 for properly describing the extensional stress growth data. The parameters of the linear viscoelastic spectra are shown in Table 3, which have been independently obtained by Laun [119].

All model descriptions are reproduced in Figs. 6-16, where experimental data have been obtained independently by Laun [119,120], Wagner and Laun [121], Giacomin *et al.* [122], Meissner [123], Laun and Munstedt [124], and Khan *et al.* [125]. Even though the short explanation for the experiments is given in figure

Table 3. Linear viscoelastic spectra of LDPE Melt I [119]

θ_i (sec)	G_i (Pa)
10^3	1.00×10^0
10^2	1.80×10^2
10^1	1.89×10^3
10^0	9.80×10^3
10^{-1}	2.67×10^4
10^{-2}	5.86×10^4
10^{-3}	9.48×10^4
10^{-4}	1.29×10^5

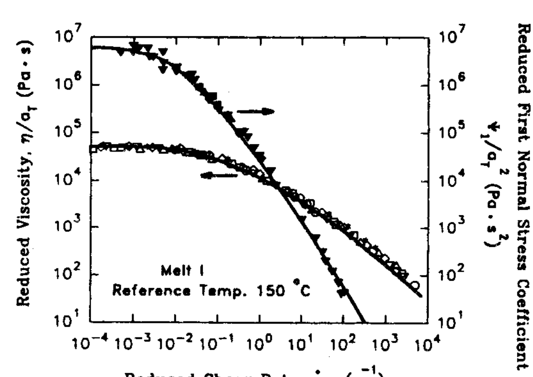

Fig. 6. Steady state shear viscosity and first normal stress coefficient. Various symbols correspond to experiments performed at different temperatures.

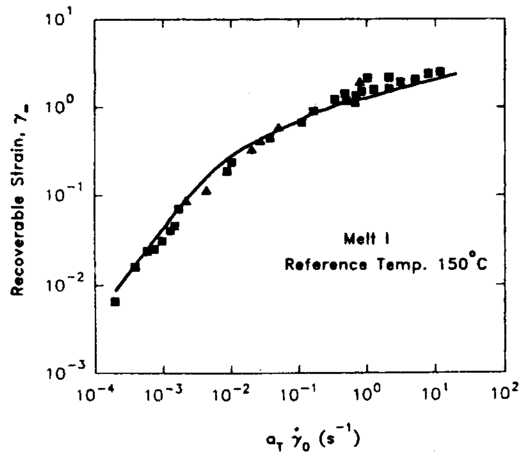

Fig. 7. Equilibrium recoverable shear strain. Various symbols correspond to experiments performed at different temperatures.

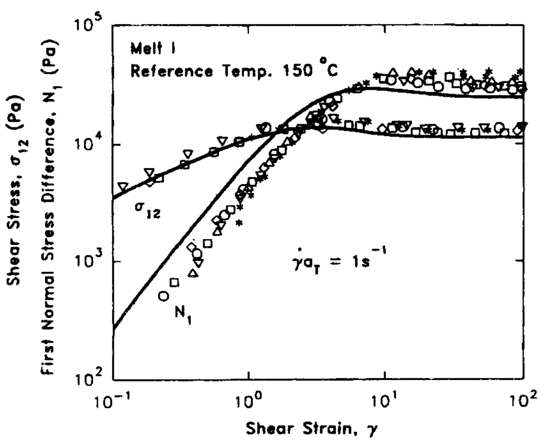

Fig. 8. Transient stress growth at a shear rate of 1. Various symbols correspond to experiments performed at different temperatures.

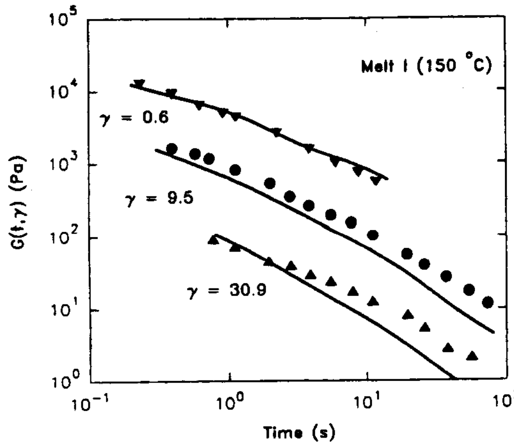

Fig. 9. Shear relaxation moduli.

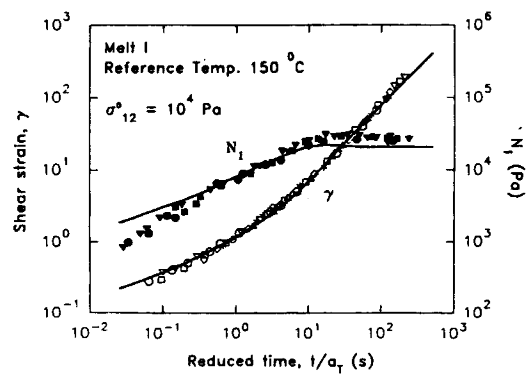

Fig. 10. Shear strain and primary normal stress difference in creep tests at a constant shear stress. Various symbols correspond to experiments performed at different temperatures.

captions, for detailed testing scheme and model calculations one would rather refer to the original paper [55]. If we keep in mind possible experimental errors, it can be asserted that the model descriptions of the whole set of experimental data are excellent. If we compare this result with the work by Larson and Monroe [66], the good desciptive ability of the Leonov CEs becomes evident. While in their study Larson and Monroe described only part of the experimental data for Melt I with 4 nonlinear parameters shown in eqs. (31) (even without considering the heavy instability inherent in their potential form), the specification (60) of the Leonov class could successfully reproduce the whole set of available data with only one nonlinear parameter m. The work by Simhambhatla and Leonov [55] is the only publication known to the current author, which contains good experimental comparison of CEs inside the global stability range.

Furthermore, one can observe another good description of the Leonov CEs in the paper [126],

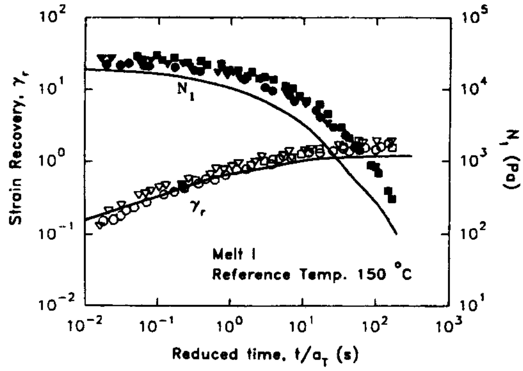

Fig. 11. Shear strain recovery and primary normal stress difference after unloading following the experiment in Fig. 10. Various symbols correspond to experiments performed at different temperatures.

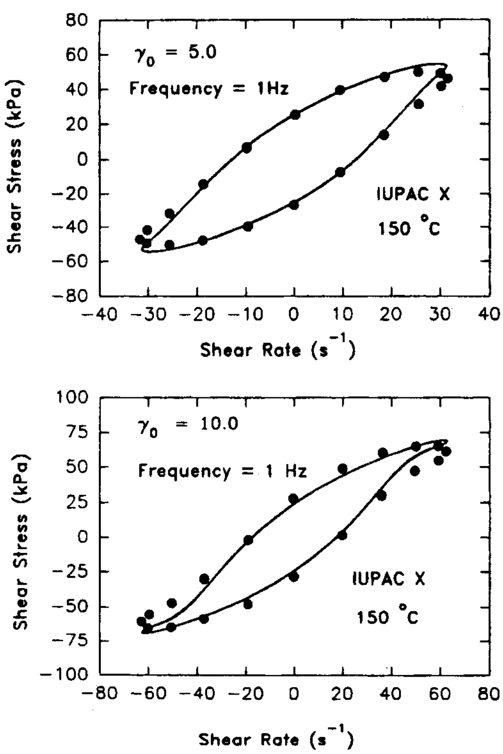

Fig. 12. Response of IUPAC X to large amplitude oscillatory shearing.

where highly nonlinear data for orthogonal superposition of two shear flows (obtained by Simmons [127]) are satisfactorily matched by the

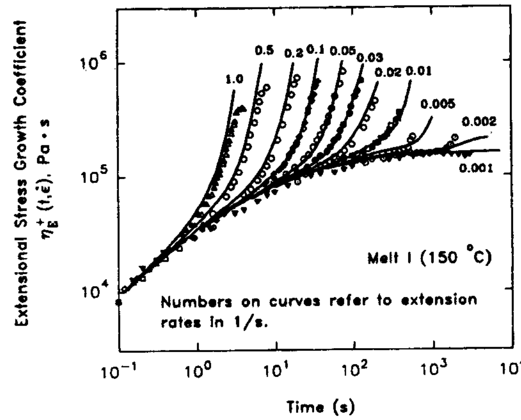

Fig. 13. Extensional stress growth coefficient as a function of time.

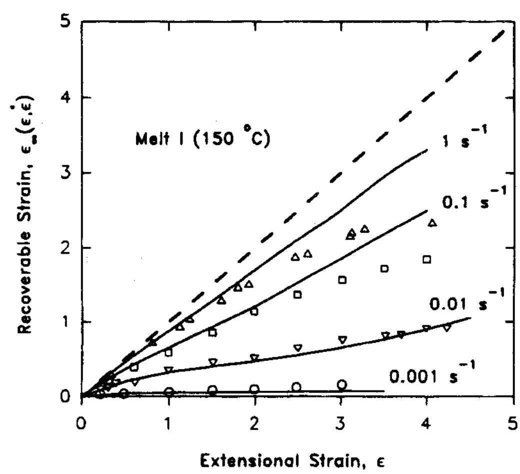

Fig. 14. Equilibrium recoverable strain as a function of extensional strains applied at constant strain rates.

simplest Leonov model involving a retardation term.

5. Conclusions

In this paper, recent studies on the mathematical stability of viscoelastic CEs are reviewd. By definition, there are two types of instabilities such as the Hadamard and the dissipative instabilities. The instability considered here is not at all related to the real physical unstable

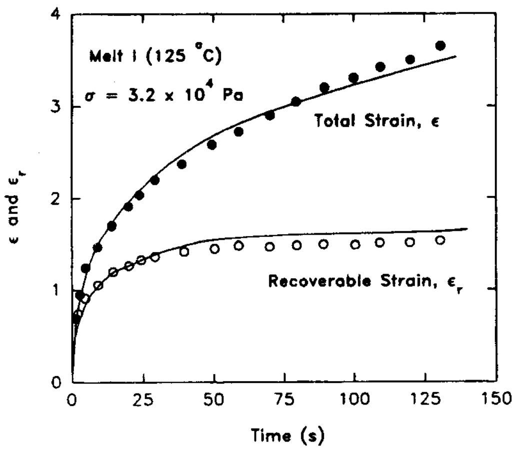

Fig. 15. Total and recoverable strains during constant stress tensile creep tests.

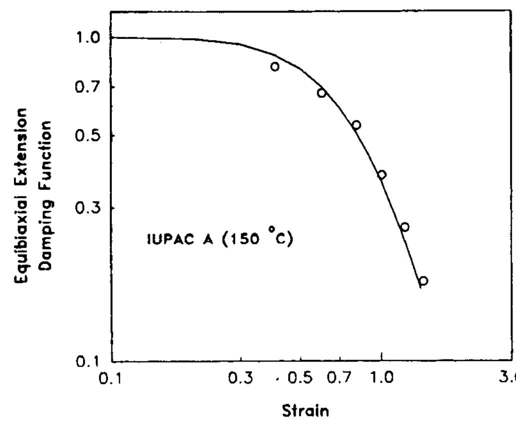

Fig. 16. Equibiaxial extension damping function for IUPAC A.

phenomena exhibited by viscoelastic fluid especially in high Deborah number flows, and should be avoided in formulating new CEs and in modeling complex flow effects.

The problem of the global Hadamard stability has been completely resolved for two such broad classes of viscoelastic CEs as quasilinear differential and factorable single integral models which are the only ones in practical use at the present time. The distinction between thermodynamic and Hadamard stabilities is also clarified. The necessary and sufficient condition obtained in an algebraic form, imposes constraints on parameters or functional terms in CEs for the Hadamard stability in any type of flow and in any Deborah number. Regarding the dissipative stability, two necessary conditions have been found. Even though the problem of the global dissipative stability still remains unsolved, it seems that the satisfaction of those two conditions is sufficient for the dissipative stability of the viscoelastic CEs.

It is shown that the combined stability criteria imposes very tough constraints on viscoelastic CEs. As a result, no separable single integral CE is found to be stable, and only three specifications of quasilinear differential CEs are proven to be globally evolutionary. Recently, only the Leonov class of CEs is shown to be able to consistently describe all the experimental data when simultaneously satisfying every stability condition.

Based on the fundamental study reviewed in this article, it is discovered that the general Leonov class of CEs is the only stable one with the required predictive ability. In author's opinion, it is not at all accidental, since the Leonov class has been derived from the theory with a firm basis on thermodynamics. In addition, it may be the only type of CEs which possess the strict stability constraints imposed from the formulation. However, there still remains one defect in constitutive modeling derived on the basis of continuum mechanics: the arbitrariness in the specification of parameters and functional terms in CEs. For its remedy, many scientists have been trying to formulate viscoelastic CEs from the concept of molecualar (or statistical) physics. However, in spite of their limited success in the linear viscoelastic limit, CEs derived from the molecular physics exhibit the most unstable behavior. Hence, we may conclude that our present understanding of rheology in view of molecular physics is in the primitive stage.

Apart from very few exceptions, solutions for the problems of viscoelastic fluid mechanics are not available when the Deborah number exceeds a certain value, usually equal to the order of one. However, the Leonov class of CEs seems promising for numerical simulation of complex flows at higher Deborah numbers. With such stable CEs, the study of highly nonlinear phenomena exhibited under strong flow regimes and in a complex flow geometry is desirable as a next step of development in this field. Furthermore, such complicated flow phenomena as phase separation, polymerization, curing and degradation during isothermal or non-isothermal flows are the challenging problems to move towards, which may be readily solved if we utilize good thermodynamic consisitency present in the Leonov class.

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