

Dynamic behaviour of rubber and rubberlike materials

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Literature survey

Dynamic behaviour of rubber and rubberlike
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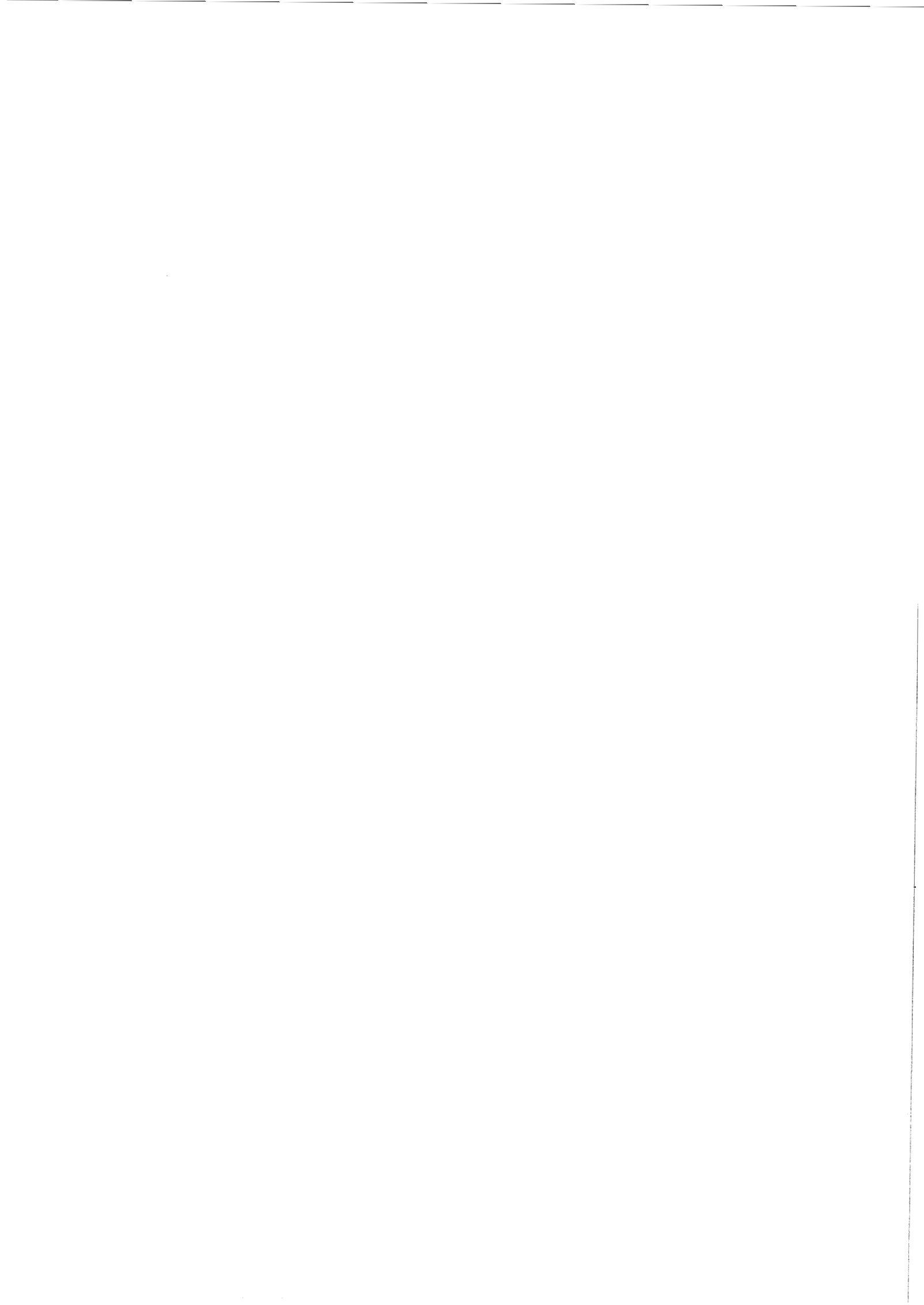
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Anniek de Bever

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Abstract

This work gives an overview of the available knowledge of the dynamic behaviour of rubber materials. It is mainly focussed on constitutive equations for rubber and rubberlike materials. Furthermore a short review is given of measurement techniques for material property evaluation, and the basic principles of vibration isolation are outlined.



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Chapter 1

Introduction

1.1 Overview

Nowadays rubber has become increasingly useful in engineering applications. One of these applications is the use of passive rubber dampers for the suspension of the laser module inside an outdoor CD player. So far the design of these dampers is largely hampered by a lack of knowledge of the mechanical behaviour of rubbers. As a consequence there are no design rules for suspensions with optimal damping properties.

Up till now the design process has been empirical. In order to change that research is conducted at the PMF and the Research Laboratories concerning the dynamic behaviour of rubber dampers. The objectives are [1]:

1. Definition of the knowledge required to predict the dynamic behaviour of rubber dampers.
2. Development of the design rules for the conception of suspension systems.

The attention is focussed on 'new' types of Silicon rubber. These types show stable damping properties over a wide temperature range. The polymers will be compounded at the PMF.

As part of that research project, this literature survey is carried out. The goal of this survey is to give an overview of the available knowledge of the dynamic behaviour of rubber and rubberlike materials. Parallel with this survey, a similar survey is conducted by Geelen [2] at the PMF.

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- [1] V.M Geelen. *Project Outline Passive Rubber Dampers for outdoor CD players*. Competence Centre Plastics, 1991. PHR-08k-813-003.

- [2] V.M Geelen. *Literature study passive rubber dampers for outdoor CD players* (in Dutch). Competence Centre Plastics, 1991. PHB-08K-013-335.

Chapter 2

General physical properties of rubbers

2.1 Definition of rubber

The original material known as rubber (or more precisely 'India-rubber') is made from the milky juice of the *Hevea Brasiliens* tree. The word rubber is derived from the ability of this material to remove pencil marks from paper. The French use the term 'caoutchouc' derived from the Maya Indian words meaning 'weeping woods'. This term is transliterated into several other European languages [1].

In current usage the term rubber is not restricted to the original natural rubber, but embraces a unique group of materials which may be identified by their ability, under certain conditions, to undergo large deformations and recover almost completely and instantaneously on release of the deforming forces [2]

Rubbers belong to the group of polymers, having long chain molecules made up of repeated units. From this an alternative name for rubber is derived, namely elastomer. The requirements for a material to be an elastomer are as follows [2]:

1. The molecules must be very long (chain-molecules) and be able to rotate freely about the bonds joining neighbouring molecular units.
2. The molecules must be joined at a number of sites to form a three-dimensional network, either by chemical bonds or by mechanical entanglements. Either of which is called crosslinks.
3. Apart from these crosslinks the molecules must be able to move readily past another; in other words the intermolecular attractions must be small.

Both natural and synthetic rubber have in their raw or unvulcanised state few or no permanent crosslinks, although there may be sufficient links formed by entanglements to give the material a certain degree of elastic behaviour. The object of the

process of vulcanisation is to introduce permanent chemical links so as to improve the elastic properties [3].

2.2 Rubber elasticity

The unique elastic properties of rubber all arise from the characteristics of elastomers, mentioned in the previous section. These properties can be explained quantitatively with the use of the first law of thermodynamics [2].

A piece of unstretched rubber is a tangled mass of long irregular molecules. When the rubber is stretched these molecules become aligned more or less in the direction of the stretching. The laws of thermodynamics state that this more orderly arrangement is less probable than the original random configuration. In other words the rubber has maximum entropy in the undeformed state and therefore will always return to that state in absence of external constraints. This may be expressed by:

$$dE = dQ + dW \quad (2.1)$$

where: dE is the change in internal energy.

dQ is change in heat input.

dW is change in work input.

With the use of:

$$dQ = TdS \quad (2.2)$$

and

$$dW = Fdl \quad (2.3)$$

it follows that:

$$F = \frac{dE}{dl} - T \frac{dS}{dl} \quad (2.4)$$

where: F is force.

T is temperature.

dS is change in entropy.

dl is change in length.

$\frac{dE}{dl}$ represents the rate of change of internal energy during deformation.

$T \frac{dS}{dl}$ represents the rate of change of entropy.

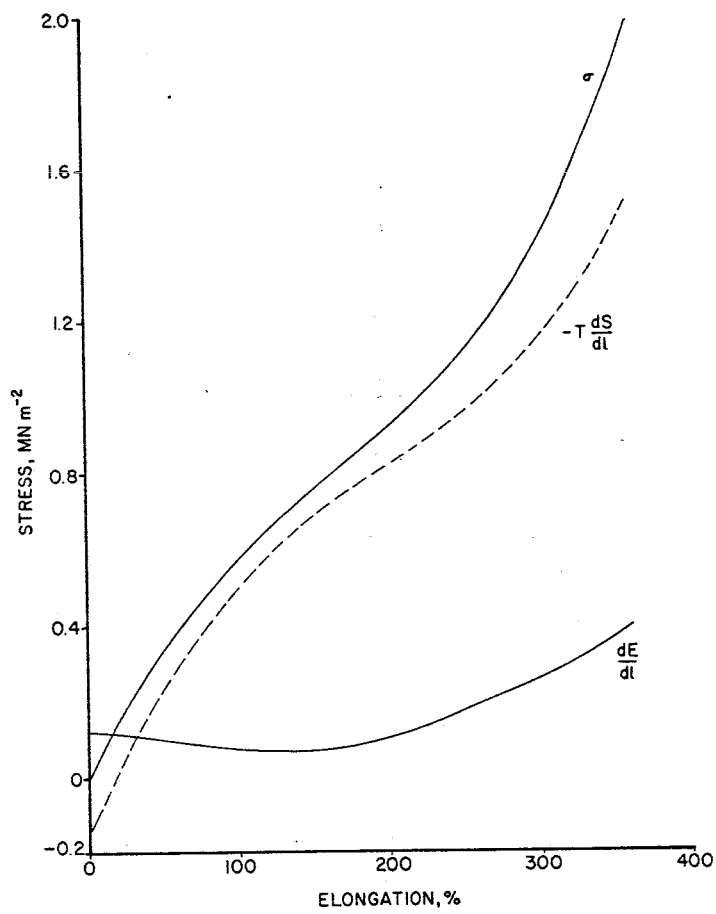


Figure 2.1: Internal energy and entropy components of tensile stress, as functions of extension (Freakley and Payne, 1978)

The force F of equation (2.4) becomes the stress σ when a unit cross-sectional area is assumed. The stress-strain curve of figure 2.1 is typical of a strain crystallising rubber. Note that the curve is markedly non-linear (i.e. Hooke's law does not apply). The major component is the entropy change due to the introduction of order to the system.

On the other hand, for metals the $\frac{dE}{dl}$ term predominates. To deform a metal, work is done in changing the interatomic distances of the molecules and not in changing the configuration of the molecules (as for rubber). Consequently the region of elastic behaviour for metal is very small in comparison with rubber.

This fundamental difference between the elastic behaviour of rubbers and metals has the following consequence: with metals, rise in temperature decreases the retractive force, while with rubbers, it increases the retractive force. From the latter it follows that when rubber is stretched heat is evolved, and vice versa, although these temperature changes are too small to be significant technically. This effect is called the Gough-Joule effect, and applies to the stress-strain relation at equilibrium. [3]

2.3 Energy storing ability

The unique stress-strain properties of rubber result from its ability to store enormous amounts of energy and to release most of this energy on retraction. The energy storing ability of rubber is e.g. 150 times as much as that for hardened steel.

Material	Energy [j/kg]
Grey cast iron	1.11
Extra soft steel	9.18
Rolled aluminium	22.6
Hardened steel	284
Hickory wood	365
Vulcanised rubber	44800

Table 2.1: Energy storing ability of materials (Freakley and Payne, 1978)

Because of time effects the return stress - strain curve never coincides with the outward curve. So there is a loss of energy, usually referred to as hysteresis, which appears as heat [2].

2.4 Stress softening

When new samples of (filled) rubber vulcanizates are stretched to a point P and then allowed to retract, subsequent extensions to the same strain require a lower force.

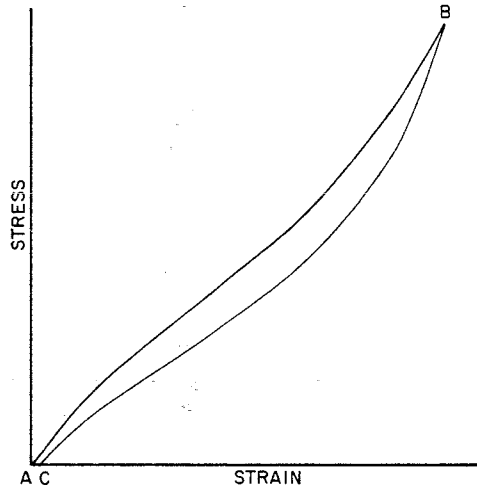


Figure 2.2: Hysteresis loop (Freakley and Payne, 1978)

In other words the stiffness at all points up to the prestressing limit P is reduced. This effect is normally referred to as stress softening or the ‘Mullins effect’ after its first investigator [2]. Most of this softening occurs during the first deformation and after some stretching cycles a steady state is reached. The Mullins effect has been mainly investigated with the use of standard tensile extension and recovery tests. The change in the form of the stress-strain curves with respect to a number of cycles to a constant maximum stress or strain is denoted. A typical plot for a tensile specimen subjected to a number of cycles to constant deformation is shown in figure 2.3.

The dependence of the magnitude of stress softening on the filler content has been investigated by Mullins and Tobin [5]. They have shown that the stress-strain behaviour of a carbon black filled rubber can be expressed in the stress-strain behaviour of a pure gum vulcanizate by use of the strain amplification factor. This strain amplification factor X describes the ratio of the average strain in the rubber phase to the measured overall strain. The use of this factor involves the assumption that the effective strain in the rubber phase of a filled vulcanizate at any stress is equal to the strain in the base vulcanizate without filler at the same stress.

Harwood, Mullins and Payne have shown, by comparing stress softening in both gum and filler loaded vulcanizate, that the softening process is mainly due to the rubber phase alone. Differences between the stress-strain hysteresis loops of gum and filler loaded vulcanizates are caused by an increase in the effective strain in the rubber phase resulting from the presence of a filler. Possible sources for the mechanism of stress softening in gum vulcanizates are [6]:

1. Breaking and remaking of crosslinks during extension.

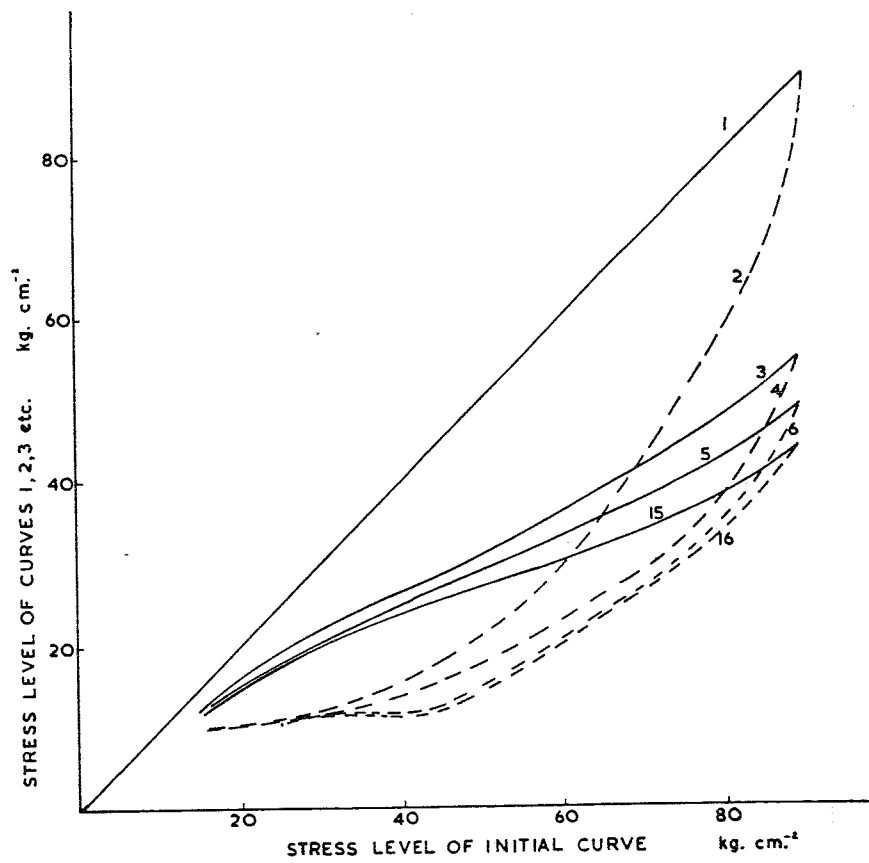


Figure 2.3: Comparison of stress levels for repeated cycling of a gum natural rubber vulcanisite (Harwood, Mullins and Payne, 1965)

2. Residual load orientation of network chains persisting after recovery.
3. Breaking of network chains.

2.5 The glass-rubber transition

The rubber-like state depends on the possibility of random thermal motion of chain elements by rotation about single bonds in the chain backbone. On lowering the temperature a point will be reached at which rotation will no longer take place at an appreciable rate. In this state the material ceases to behave as rubber and becomes hard and rigid like glass [1].

At this so-called glass transition, rubbers exhibit rapidly increasing modulus and viscosity through several orders of magnitude within a small temperature range. There are however no discontinuous changes in volume, entropy or enthalpy. Characteristic of rubbers is that the glass transition temperature T_g is below room temperature, usually below -50°C [4].

The importance of T_g in rheology is that it plays the role of a characteristic or normalizing temperature. Basically the greater the value of $T - T_g$, the lower the value of the modulus. This idea is partly quantitatively formulated by the universal WLF-equation given by [7]:

$$\log\left(\frac{\eta}{\eta_g}\right) = \frac{-17.4(T - T_g)}{51.6 + (T - T_g)} \quad (2.5)$$

where: η = viscosity of the polymer.

η_g = viscosity at T_g .

2.6 Crystalline Transitions

Certain types of rubbers, including natural rubber, which have a very regular backbone structure in the sense of identical character of most of the structural units, gradually crystallize when they are maintained at a low temperature (e.g. 0°C or lower). This is a gradual hardening, often referred to as freezing, due to the tendency of parts of neighbouring molecules to line up in parallel. The latter permits stronger intermolecular attractions which bind the molecule segment into a more or less rigid 'crystallite'. Because of the reinforcing effect of the crystallite, crystalline rubber is considerably harder (by a factor of 100) and less extensible than amorphous rubber, though it is still flexible, owing to the residual amorphous (rubber-like) component of the structure. Vulcanisation reduces the crystallization but does not eliminate it.

Those rubbers which show the phenomenon just described also exhibit a somewhat similar effect when they are stretched. In the so-called stress-induced crystallization a new phase is formed with greatly increased modulus and memory. The

crystallized phase exhibits elastic-plastic rather than visco-elastic properties. Further crystallization occurs first in the region of highest stress. Thus stress-induced crystallization acts both to increase modulus and resistance to deformation and it prevents the propagation of defects [1] [3] [4].

2.7 Silicone rubbers

In this section the properties and applications of silicone rubbers are considered. An overview of the properties of other elastomers is given in reference [8].

The importance of silicone rubber lies in its ability to serve under severe environments where nothing else will function or in its ability to function more reliably in less stringent environments. Silicone rubber made its first appearance on the market in the mid 1940's. A review of the development of silicone elastomers is given by Warrick [9]. Silicone rubber began as a polydimethylsiloxane elastomer vulcanized by benzylperoxide. Today silicone rubber is a line of products of widely different compositions and vulcanized by many different systems. Detailed information is given by Ranney [10]. The most common used silicone elastomer is principally based on polydimethylsiloxane [11].

Because of their unique properties silicone rubbers are being used in an increasing number of applications. These properties include:

- Extreme weather and high thermal stability due to the strong silicon-oxygen bond ($-Si-O-Si-O-$) that makes up the backbone of silicone polymers.
- High flexibility at low temperatures due to low intermolecular forces.
- Good release properties.
- Incompatibility with most organic compounds.
- Excellent isolator.
- High surface tension.
- High permeability to gases.
- Hydrophobicity.
- Good biocompatibility.

However due to the poor mechanical properties of silicone elastomers, many of its initial potential applications have suffered in terms of performance.

Silicone elastomers exhibit moderate resistance to hydrocarbon liquids and oils. This can be overcome by replacing one methyl group on each silicon atom with a trifluoropropyl group ($-CH_2-CH_2-CF_3$). The result is a fluorosilicone polymer

that has superior fuel resistance along with higher specific gravity and lower gas permeability than dimethyl silicone elastomers. The other properties are similar. Nowadays silicone rubber parts are used in the automotive, electronic, diary, medical, construction, and aerospace industries [11] [12].

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Chapter 3

Constitutive equations for rubber materials

3.1 Introduction

A constitutive equation describes the behaviour of a specific material under influence of external forces. Initial investigations have shown that the behaviour of rubber dampers cannot be modelled with simple linear viscoelastic models. Therefore it is necessary to apply nonlinear viscoelastic constitutive equations.

In section 3.2 the theory of linear viscoelasticity is treated. This provides a basis for understanding the discussion of nonlinear behaviour contained in the remainder of this chapter. There are three possible methods of modelling nonlinear material behaviour [1]:

1. Empirical function fitting.
2. Rational analysis based on a particular theory.
3. A combination of the above two.

The behaviour of nonlinear materials is very complex and therefore the first method is not likely to be successful, except for very simple situations and the second method is too complicated. Thus the last approach has been adapted by the majority of the authors.

3.2 Linear theory of viscoelasticity

In this section some of the main results of the classical theory of linear viscoelasticity are summarized. The mechanical behaviour of a material is linear when twice the load produces twice the response or in other words when the deformation process does not influence the material properties. Most polymers exhibit linear behaviour

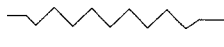
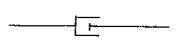
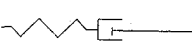
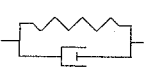
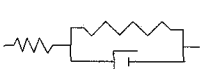
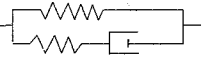
Model	Representation	Constitutive equation
Hooke (elastic)		$\sigma = E\epsilon$
Newton (viscous)		$\sigma = \eta\dot{\epsilon}$
Maxwell (viscoelastic)		$\frac{\sigma}{\eta} + \frac{\dot{\sigma}}{E} = \dot{\epsilon}$
Voight (viscoelastic)		$\sigma = E\epsilon + \eta\dot{\epsilon}$
Standard linear solid		$(1 + \frac{E_1}{E_2})\sigma + \frac{\eta}{E_2}\dot{\sigma} = E_1\epsilon + \eta\dot{\epsilon}$
Pointing Thomson		$\frac{\sigma(\eta+E_1)}{\eta} = \frac{(E_1+E_2)\eta\dot{\epsilon}+E_1E_2\epsilon}{\eta}$

Table 3.1: Simple spring and dashpot models

in a much broader area than most metals and therefore the linear theory is more often applied for polymers [2].

In the early development of viscoelasticity theory, much use was made of the modelling of materials by networks made up of springs and dashpots. The spring represents the elastic component of the material. All energy put into the spring is equal to the energy the spring gives up when the load is removed. This assumption gives the spring 100% efficiency. The concept of an ideal linear elastic spring is often referred to as Hookean. The dashpot represents the viscous component of the material. All energy put into a viscous system is lost, generally in the form of heat. The concept of an ideal viscous material is sometimes referred to as a Newtonian fluid [3]. Since viscoelastic materials combine elastic and viscous effects it is possible to model them approximately by combining the elastic and viscous components in various parallel and series arrangements. Some simple combinations and their constitutive equations are given in table 3.1. In the constitutive equations in table 3.1 σ represents the stress, ϵ represents the strain, E is the elastic modulus and η is the viscosity.

A more general mechanical model is obtained by combination of an arbitrary number of elements. These models are referred to as canonical models. Thus for

example by parallel arrangement of 'n' Maxwell elements, each of which has a certain relaxation time $\theta_i = \frac{\eta_i}{E_i}$, one obtains the generalized Maxwell model [6].

The mechanical models discussed in the foregoing subsection represented the one-dimensional response behaviour of a linear viscoelastic material. A linear infinitesimal theory of viscoelasticity for three-dimensional behaviour is given by Lockett [4]. The stress components σ_{ij} are supposed to depend on the strain components $\epsilon_{ij}(\tau)$ at the same material point, to which that point was subjected at all times τ up to and including the time t . Mathematically this can be expressed in the form:

$$\sigma_{ij}(t) = \mathcal{F}_{ij}^t \{ \epsilon_{kl}(\tau) \} \quad (3.1)$$

where \mathcal{F}_{ij} are functionals. In the linear theory, the above equation can be written as:

$$\sigma_{ij}(t) = \int_{-\infty}^t c_{ijkl}(t - \tau) \dot{\epsilon}_{kl} d\tau \quad (3.2)$$

where c_{ijkl} are stress relaxation functions. They must be determined either by experiment or from the physics of the material structure. For an isotropic incompressible material the above equation simplifies into:

$$\boldsymbol{\sigma}(t) = -p\mathbf{I} + 2 \int_{-\infty}^t \mu(t - \tau) \dot{\boldsymbol{\epsilon}}(\tau) d\tau \quad (3.3)$$

where: μ is a stress relaxation function.

p is the pressure.

$\boldsymbol{\sigma}$ is the stress tensor.

\mathbf{I} is the unity tensor.

$\boldsymbol{\epsilon}$ is the classical linear strain tensor, defined by:

$$\boldsymbol{\epsilon} = \frac{1}{2}(\vec{\nabla}_0 \vec{u} + (\vec{\nabla}_0 \vec{u})^c) \quad (3.4)$$

In the above equation \vec{u} denotes the displacement vector.

3.3 Nonlinear elasticity

3.3.1 Introduction

In this section the theory of nonlinear elasticity is considered. This theory is of interest because the behaviour of viscoelastic solids at the limits of the time and frequency range is essentially elastic. At short times or at long times and low frequencies, memory effects become negligible and the material can be considered elastic.

3.3.2 Statistical theory

The retractive force in stretched rubber is essentially due to entropy changes, as discussed in section 2.2. Therefore it is possible to develop general expressions for the deformation of rubber units based upon the molecular structure of rubber. The derivation of these general expressions is often referred to as statistical theory or as kinetic theory. This theory, among others, is derived by Treloar [7]. Based on the original theory of Kuhn, Treloar makes use of the following assumptions:

1. The network contains N chains per unit volume, a chain being defined as the segment of the molecule between successive points of cross-linkage.
2. The mean end to end distance for the whole assembly of chains is the same as for a corresponding set of free chains.
3. There is no change of volume on deformation.
4. The junction points between chains move on deformation as if they were imbedded in an elastic continuum. As a result the components of length of each chain change in the same ratio as the corresponding dimensions of the bulk rubber. (Affine deformation).
5. The individual network can be described by Gaussian statistics. This means that the entropy of the network is the sum of the entropies of the individual chains.

With the use of these assumptions it can be shown that the expression for the free energy of deformation W is equal to:

$$W = \frac{G}{2} (\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3) \quad (3.5)$$

where:

$$G = NkT = \frac{\rho RT}{M_c} \quad (3.6)$$

where: λ_i is the principal extension ratio.

(The three extension ratios are defined along three mutually perpendicular axes).

T is the absolute temperature.

N is the number of molecular chains per unit volume.

M_c is the mean chain molecular weight.

ρ is the density.

k is Boltzman's constant.

R is the gasconstant.

From this general expression the stresses can be calculated. For example in simple extension or compression, where simple means that a rectangular block retains a rectangular form, the extension ratio's are given by:

$$\lambda_1 = \lambda \quad \text{and} \quad \lambda_2 = \lambda_3 = \lambda^{-\frac{1}{2}} \quad (3.7)$$

Assuming the unstrained block to be in the form of a unit cube, it then follows that:

$$dW = Fdl = \sigma d\lambda \quad (3.8)$$

where: dl is the change in length.

F is the force.

σ is the stress (force per unit area of unstrained section).

Thus:

$$\sigma = G\left(\lambda - \frac{1}{\lambda^2}\right) \quad (3.9)$$

Equation (3.9) predicts that, at low strains, the stress in a rubber is linear proportional to the strain (Hookean). The slope of equation (3.9) at the origin is $3G$ for a pure rubber. By definition the Young's modulus is $E = \frac{\sigma}{\epsilon}$, so the Young's modulus is three times the shear modulus G . The addition of particulate fillers reduces Poisson's ratio, the material becomes less incompressible and E becomes bigger than $3G$. At higher strains, however, the behaviour becomes nonlinear as described by equation (3.9). The latter is sometimes referred to as Neo-Hookean material behaviour. Comparison with experiments show that the statistical theory in extension is only valid until about 30% strain, above which it is necessary to use either non-Gaussian theories or phenomenological theories (See figure 3.1) [8].

3.3.3 Non-Gaussian theory

The statistical theory described in the previous subsection is based on network chains that obey Gaussian statistics. The non-Gaussian statistical treatment takes into account the finite extensibility of the network chains.

The simplest model is the three-chain model. This model is based on the assumption that the network can be replaced by three independent sets of chains parallel to the axes of a rectangular coordinate system. For a simple extension, with the assumption that the unstrained block is in the form of a unit cube, it can be shown that:

$$\sigma = \frac{NkT}{3} n^{\frac{1}{2}} \left\{ \mathcal{L}^{-1}\left(\frac{\lambda}{n^{\frac{1}{2}}}\right) - \lambda^{-\frac{3}{2}} \mathcal{L}^{-1}\left(\frac{1}{\lambda^{\frac{1}{2}} n^{\frac{1}{2}}}\right) \right\} \quad (3.10)$$

where n is the number of random links and \mathcal{L}^{-1} is the inverse Langevin function.

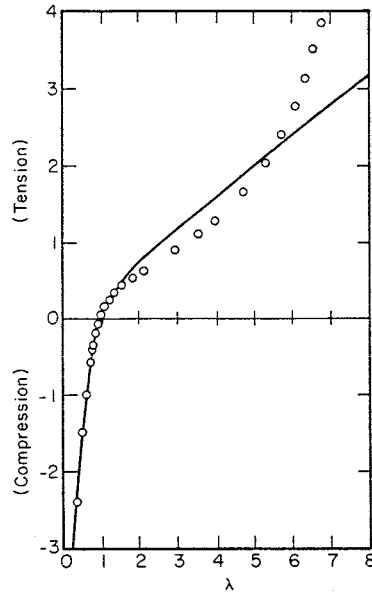


Figure 3.1: Comparison of uniaxial extension and compression data of natural rubber (circle) with Gaussian theory (line) (Eirich, 1978)

The four chain (tetrahedral) model is a more realistic representation of the conditions existing in an actual network. The model considers an elementary ‘cell’ of the network consisting of four chains radiating outwards from a common junction point P. It takes into account a number of factors which do not arise in the Gaussian theory. The most important of these factors are:

1. The non-Gaussian character of the individual chains.
2. The non-affine displacement of the mean or equilibrium position of the central junction point.
3. The variation of the extent of fluctuations of the central junction point with strain.

Calculations of the entropy of deformation which takes into account all of these factors can only be carried out by numerical methods of computation. This has been done by Treloar [7]. A typical result is shown in figure 3.2

3.3.4 Phenomenological theories

In parallel with the development of the statistical theory of rubber elasticity, the so-called phenomenological theory is derived. This theory aims at providing a mathematical framework for all materials which are capable of large elastic deformations, and is not concerned with molecular structure. Work in this field was initiated by Mooney and continued by Rivlin [9].

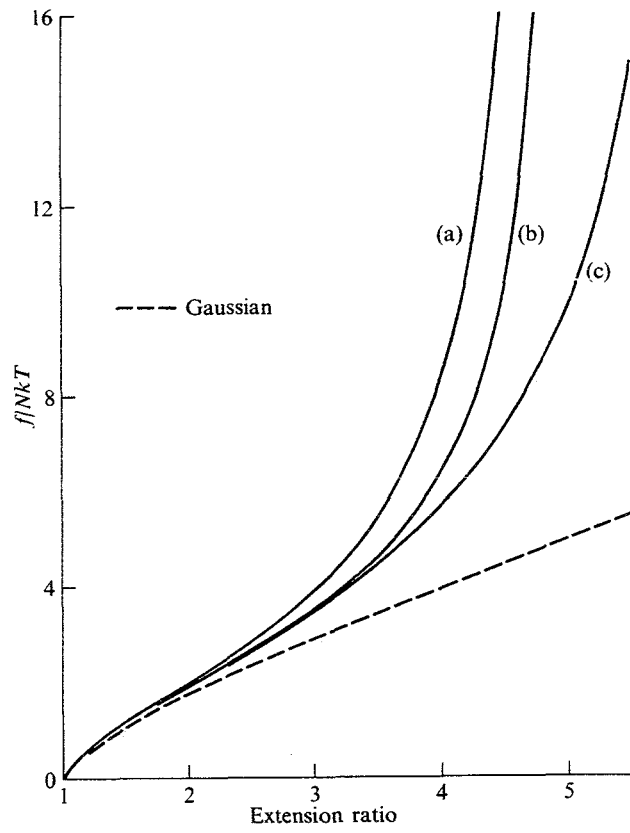


Figure 3.2: Non-Gaussian force extension curves for $n=25$. (a) 3-chain model; (b) tetrahedral model, affine displacement; (c) tetrahedral model, non-affine displacement. (Treloar, 1975)

From symmetry considerations it can be shown that the strain energy function for an isotropic material is given by:

$$W = \sum_{i,j,k=0}^{\infty} c_{ijk} (I_1 - 3)^i (I_2 - 3)^j (I_3 - 1)^k \quad (3.11)$$

where c_{ijk} are constants and I_i are strain invariants. The latter are defined by:

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2 \quad (3.12)$$

$$I_2 = \lambda_1^2 \lambda_2^2 + \lambda_2^2 \lambda_3^2 + \lambda_3^2 \lambda_1^2 \quad (3.13)$$

$$I_3 = \lambda_1^2 \lambda_2^2 \lambda_3^2 \quad (3.14)$$

The third strain invariant is equal to unity for an incompressible material. Note that the first term in equation (3.11) ($i=1, j=0, k=0$) is equal to the statistical theory (equation (3.5)). Another form of interest of equation (3.11) is:

$$W = C_{100}(I_1 - 3) + C_{010}(I_2 - 3) \quad (3.15)$$

For uniaxial extension of an incompressible material equation (3.15) becomes:

$$\sigma = (2C_1 + \frac{2C_2}{\lambda})(\lambda - \lambda^{-2}) \quad (3.16)$$

This equation is known as the Mooney-Rivlin equation, and is valid up to $\lambda = 2.5$, but does not describe uniaxial compression data ($\lambda < 1$) well with the same constants [8] [10].

Rivlin assumed that the strain energy is a polynomial function of the principal strain invariant. Alternatively the strain energy function W can be cast in terms of the principal extension ratio's. Ogden, Valanis-Landel, Peng and Peng-Landel material models belong to this category. The Ogden material model for incompressible materials can be expressed as [11]:

$$W = \sum_{j=1}^m \frac{c_j}{b_j} [(\lambda_1)^{b_j} + (\lambda_2)^{b_j} + (\lambda_3)^{b_j} - 3] \quad (3.17)$$

where c_j and b_j are material coefficients. The principal Cauchy stresses are given by:

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i} - p \quad (i = 1, 2, 3) \quad (3.18)$$

where p is an arbitrary hydrostatic pressure introduced because of the incompressibility constraint. The tensor notation of the Ogden material model has been derived by Rivlin and Sawyers [15] and is given by:

$$\boldsymbol{\sigma} = \sum_{i=1}^3 \frac{\lambda_i \frac{\partial W}{\partial \lambda_i}}{2\lambda_i^4 - I_1 \lambda_i^2 + \lambda_i^{-2}} [(\lambda_i^2 - I_1) \mathbf{B} + \mathbf{B}^2] - p \mathbf{I} \quad (3.19)$$

where \mathbf{B} is the left Cauchy-Green tensor, defined by:

$$\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^c \quad (3.20)$$

When the coefficients of the Ogden model are chosen correctly, this formulation describes the mechanical representation of hyperelastic materials accurately for large ranges of deformations [14]. For the special case of uniaxial tension the stress-strain equation yields:

$$f = \sum_j c_j [\lambda^{b_j-1} - \lambda^{-(1+\frac{1}{2}b_j)}] \quad (3.21)$$

where f is the force per unit original area.

The Peng material model states that the free energy of deformation W takes the form:

$$W = w(\lambda_1) + w(\lambda_2) + w(\lambda_3) \quad (3.22)$$

subject to the incompressibility constraint: $\lambda_1 \lambda_2 \lambda_3 = 1$. This form was originally proposed by Valanis and Landel. A detailed expression for the functions $w(\lambda_i)$ can be found in reference [16].

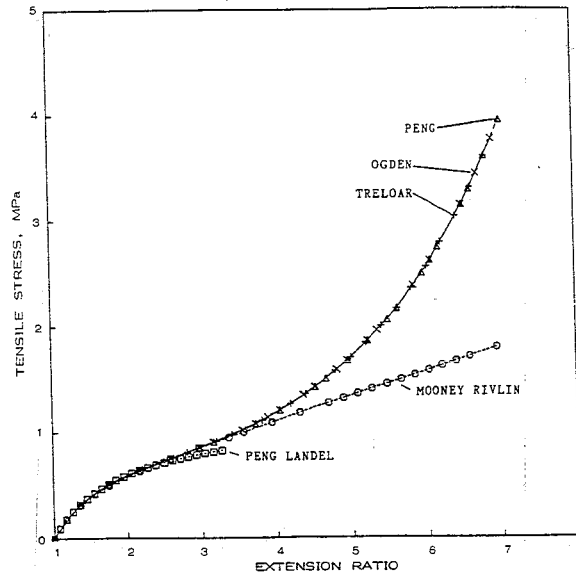


Figure 3.3: numerical data compared with experimental Treloar data in a simple extension test. (Finney and Kumar, 1988)

The Peng-Landel material model expresses W for an incompressible material as:

$$W = \sum_{i=0}^3 c[\lambda_i - 1 - \ln \lambda_i - \frac{1}{6}(\ln \lambda_i)^2 + \frac{1}{18}(\ln \lambda_i)^3 - \frac{1}{216}(\ln \lambda_i)^4] \quad (3.23)$$

where c is the initial tensile modulus. If the material is compressible the initial bulk modulus is also needed.

Finney and Kumar describe methods to develop material constants for Mooney-Rivlin, Ogden, Peng and Peng-Landel material models [18]. In figures 3.3, 3.4 and 3.5 the numerical data from the material models are compared with experimental results.

3.4 Integral constitutive equations for nonlinear viscoelastic material behaviour

3.4.1 Green-Rivlin theory

The equations below presume that there is an initial configuration for which the material is isotropic, homogenous, unstressed and unstrained. Furthermore the material is assumed to be simple, i.e. the stress σ_{ij} at a given particle at time t depends only upon the history of the deformation gradient tensor at that particle. This deformation tensor \mathbf{F} is defined by:

$$\mathbf{F} = (\vec{\nabla}_0 \vec{x}) \quad ; \quad d\vec{x} = \mathbf{F} \cdot d\vec{x}_0 \quad (3.24)$$

where: $d\vec{x}$ = the distance vector between two neighbouring material points in

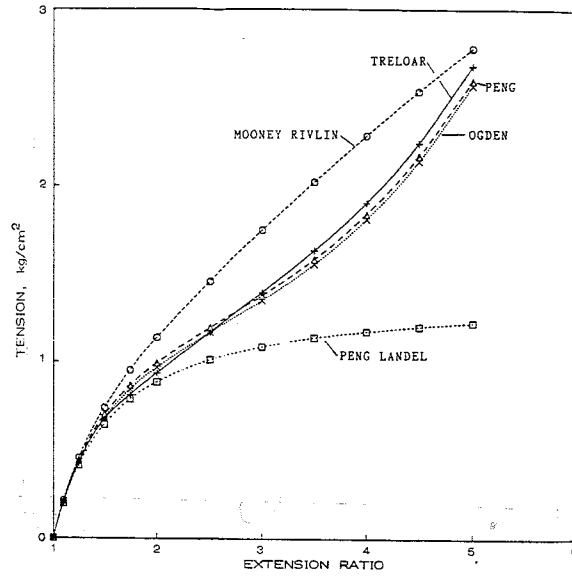


Figure 3.4: numerical data compared with experimental Treloar data in a pure shear test. (Finney and Kumar, 1988)

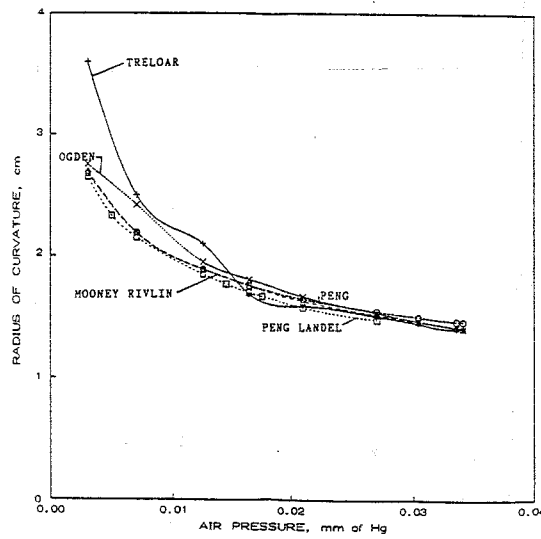


Figure 3.5: numerical data compared with experimental Treloar data in a two-dimensional extension test. (Finney and Kumar, 1988)

the current configuration.

$d\vec{x}_0$ = the distance vector between these points in the reference configuration.

The polar decomposition theorem allows \mathbf{F} to be written as:

$$\mathbf{F} = \mathbf{R} \cdot \mathbf{U} \quad (3.25)$$

where \mathbf{R} is an orthogonal tensor and \mathbf{U} denotes a positive definite symmetric tensor. The decomposition of \mathbf{F} expresses the idea that the deformation may be considered as a pure stretch \mathbf{U} followed by a rigid rotation \mathbf{R} .

A restriction on the manner in which the stress can depend on the deformation tensor arises through the principle of ‘objectivity’ or ‘frame indifference’. This principle states that the mechanical properties of a material depend only on the material itself and not on the observer. With use of the above assumptions, the constitutive equation can be written as:

$$\mathbf{Q}(t) = \mathcal{F}_{ij}^t \{ \mathbf{P}(\tau) \} \quad (3.26)$$

where \mathcal{F} is an isotropic functional and \mathbf{Q} and \mathbf{P} can be interpreted either as:

$$\mathbf{Q} = \boldsymbol{\Sigma} = \mathbf{R}^c \cdot \boldsymbol{\sigma} \cdot \mathbf{R}, \quad \mathbf{P} = \mathbf{E} \quad (\text{stress relaxation formulation}) \quad (3.27)$$

$$\mathbf{Q} = \mathbf{E}, \quad \mathbf{P} = \boldsymbol{\Sigma} = \mathbf{R}^c \cdot \boldsymbol{\sigma} \cdot \mathbf{R} \quad (\text{creep formulation}) \quad (3.28)$$

in which $\boldsymbol{\Sigma}$ is the stress tensor, evaluated in a coordinate system which is rotated with the rigid-body-rotation component of the deformation and \mathbf{E} is the Green-Lagrange strain tensor defined by:

$$\mathbf{E} = \frac{1}{2}(\mathbf{U}^2 - \mathbf{I}) = \frac{1}{2}(\mathbf{F}^c \cdot \mathbf{F} - \mathbf{I}) \quad (3.29)$$

The constitutive functional can be expressed as the sum of a series of multiple integrals, as was done by Green and Rivlin [1] [31] [20]:

$$\begin{aligned} \mathbf{Q}(t) = & \int_{-\infty}^t \{ \mathbf{I} \psi_1 T_1 + \psi_2 \mathbf{M}_1 \} d\tau_{(1)} + \\ & \int \int_{-\infty}^t \{ \mathbf{I} \psi_3 T_1 T_2 + \mathbf{I} \psi_4 T_{12} + \psi_5 T_1 \mathbf{M}_2 + \psi_6 \mathbf{M}_1 \cdot \mathbf{M}_2 \} d\tau_{(2)} + \\ & \int \int \int_{-\infty}^t \{ \mathbf{I} \psi_7 T_{123} + \mathbf{I} \psi_8 T_1 T_{23} + \psi_9 T_1 T_2 \mathbf{M}_3 + \psi_{10} T_{12} \mathbf{M}_3 + \\ & \psi_{11} T_1 \mathbf{M}_2 \cdot \mathbf{M}_3 + \psi_{12} \mathbf{M}_1 \cdot \mathbf{M}_2 \cdot \mathbf{M}_3 \} d\tau_{(3)} + \dots \end{aligned} \quad (3.30)$$

where:

$$\begin{aligned} d\tau_{(n)} &= d\tau_1 d\tau_2 \dots d\tau_n ; \quad \mathbf{M}_\alpha = \dot{\mathbf{P}}(\tau_\alpha) \\ T_\alpha &= \text{tr } \mathbf{M}_\alpha ; \quad T_{\alpha\beta} = \text{tr } \mathbf{M}_\alpha \mathbf{M}_\beta ; \quad T_{\alpha\beta\gamma} = \text{tr } \mathbf{M}_\alpha \mathbf{M}_\beta \mathbf{M}_\gamma \end{aligned} \quad (3.31)$$

Depending on the choice of the definitions (3.27) and (3.28), the material functions ψ_α are stress-relaxation or creep functions of various order. For an aging material, the material functions ψ_α are functions of t and of the integration variables τ_α . For a hereditary or non-ageing material, ψ_1 and ψ_2 are functions of $t - \tau_1$, $\psi_3 \dots \psi_6$ are functions of $t - \tau_1$ and $t - \tau_2$, $\psi_7 \dots \psi_{12}$ are functions of $t - \tau_1$, $t - \tau_2$ and $t - \tau_3$. It has been shown by Lockett that the following symmetries may be assumed without loss of generality [21]:

$$\begin{aligned} \psi_3, \psi_4, \psi_9, \psi_{10}, &= s(1, 2) \\ \psi_7 &= s(1, 2, 3) \\ \psi_8, \psi_{11} &= s(2, 3) \\ \psi_{12} &= s(1, 3) \end{aligned} \quad (3.32)$$

where $s(i, j)$ denotes symmetry with respect to the interchange of the i^{th} and j^{th} arguments.

A constitutive equation for an incompressible Green-Rivlin material has been obtained by Pipkin [4], [20]. In addition to the constraint of no volume change, Pipkin assumes that as a reaction to this constraint, there arises an arbitrary hydrostatic pressure. Equation (3.30) then has the form:

$$\begin{aligned} \mathbf{Q}(t) &= \int_{-\infty}^t \psi_2 \mathbf{M}_1 d\tau_{(1)} + \\ &\quad \int \int_{-\infty}^t \psi_6 \mathbf{M}_1 \mathbf{M}_2 d\tau_{(2)} + \\ &\quad \int \int \int_{-\infty}^t \{ \psi_{10} T_{12} \mathbf{M}_3 + \psi_{12} \mathbf{M}_1 \mathbf{M}_2 \mathbf{M}_3 \} d\tau_{(3)} \end{aligned} \quad (3.33)$$

where

$$\mathbf{Q} = \mathbf{R}^C \cdot \mathbf{s} \cdot \mathbf{R} + p \mathbf{I} \quad (3.34)$$

$$\mathbf{s} = \boldsymbol{\sigma} - \frac{1}{3} \mathbf{I} \text{tr } \boldsymbol{\sigma} \quad (3.35)$$

\mathbf{s} is the deviatoric stress tensor and p is a hydrostatic pressure.

Lockett describes an experimental method to determine the material functions ψ_1, \dots, ψ_{12} which appear in the third order approximation of the 3D constitutive equations (3.30), (3.33) [21]. However the number of tests required, indicates that

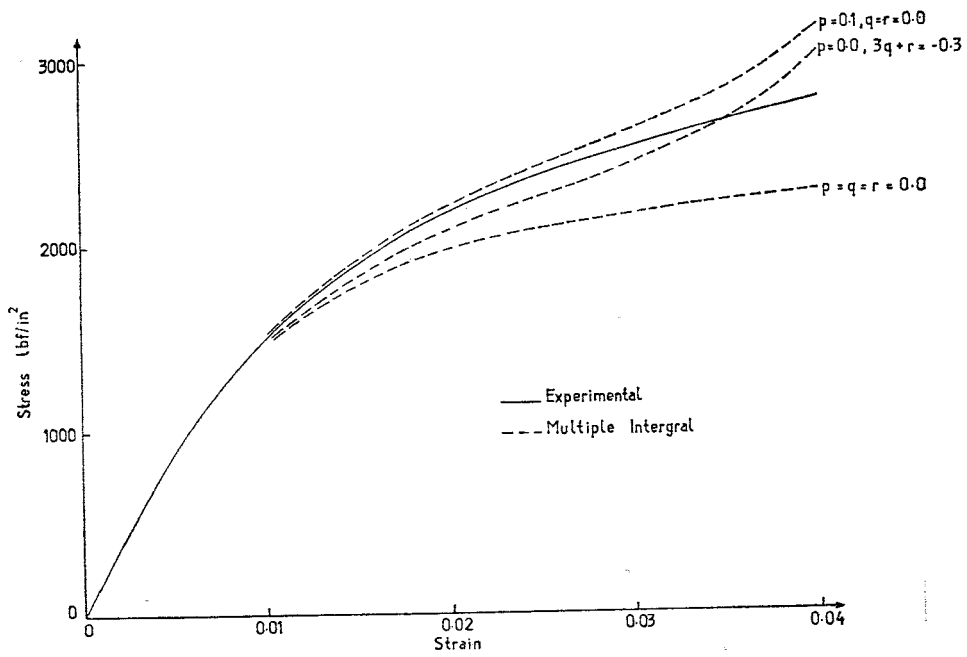


Figure 3.6: Predictions for constant strain rate loading using the multiple integral theory for polypropylene (Smart and Williams, 1972)

these equations may be too complex for routine practical application. Furthermore third order equations like (3.30) and (3.33) are restricted to small strain or stress rates and to nonlinear materials which do not differ much from linear materials.

Because of the difficulty to obtain the required data, several workers have tried to model the material functions. These material functions are also referred to as time kernel functions in literature. A short review is given by Smart and Williams [22]. For a one-dimensional deformation the multiple integral theory can be written correct to the third order as:

$$\begin{aligned}\sigma(t) = & \int_0^t J_1(t - \tau_1) \frac{d\epsilon}{d\tau_1} + \\ & \int \int_0^t J_2(t - \tau_1, t - \tau_2) \frac{d\epsilon}{d\tau_1} \frac{d\epsilon}{d\tau_2} d\tau_1 d\tau_2 + \\ & \int \int \int_0^t J_3(t - \tau_1, t - \tau_2, t - \tau_3) \frac{d\epsilon}{d\tau_1} \frac{d\epsilon}{d\tau_2} \frac{d\epsilon}{d\tau_3} d\tau_1 d\tau_2 d\tau_3\end{aligned}\quad (3.36)$$

The relaxation stress is then given by:

$$\sigma(t) = a(\epsilon t^{-l} + \alpha \epsilon^2 t^{-m} + \beta \epsilon^3 t^{-n}) \quad (3.37)$$

Smart and Williams [23] propose general kernel functions of the form:

$$\left. \begin{aligned}J_1(t - \tau_1) &= a(t - \tau_1)^{-l} \\ J_2(t - \tau_1, t - \tau_2) &= a\alpha(t - \tau_1)^{p-m}(t - \tau_2)^{-p} \\ J_3(t - \tau_1, t - \tau_2, t - \tau_3) &= a\beta(t - \tau_1)^{q-n}(t - \tau_2)^r(t - \tau_3)^s\end{aligned}\right\} \quad (3.38)$$

with $q+r+s=0$ and the restriction $\tau_1 \geq \tau_2 \geq \tau_3$. The parameters a, α, β, l, m and n are fitted to equation (3.37) and the remaining unknown parameters are determined by experiments. In figure 3.6 the results of the use of the kernel functions proposed by Smart and Williams is shown.

3.4.2 The theory of Pipkin and Rogers

Pipkin and Rogers proposed a multiple integral constitutive equation that is nonlinear even in its first term approximation [24]. Their motivation was that the first integral term should model single step tests exactly, the second term should extend the representation to two-step tests. The higher order terms of the series continue this process of error and refinement. The n -th term is determined directly by the difference, if any is observed, between experimental results from n step histories and the prediction obtained from $(n-1)$ step data. For the one-dimensional case, the stress is given by:

$$\sigma(t) = \sum_{n=1}^{\infty} S_n(t) \quad (3.39)$$

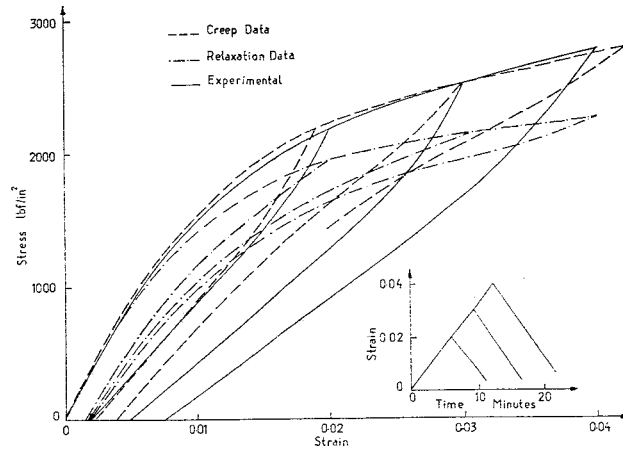


Figure 3.7: Predictions for constant strain rate loading using the Pipkin-Rogers theory for polypropylene (Smart and Williams, 1972)

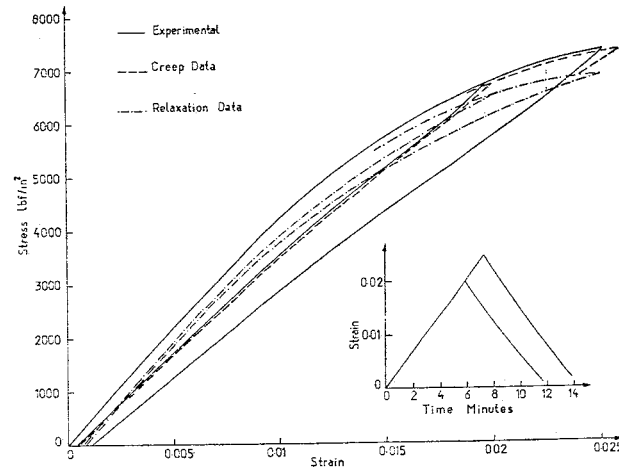


Figure 3.8: Predictions for constant strain rate loading using the Pipkin-Rogers theory for polyvinylchloride (pvc). (Smart and Williams, 1972)

$$S_n(t) = \frac{1}{n!} \int_{-\infty}^t \dots \int_{-\infty}^t d_{\epsilon(\tau_1)} \dots d_{\epsilon(\tau_n)} R_n[\epsilon(\tau_1), t - \tau_1; \dots; \epsilon(\tau_n), t - \tau_n] \quad (3.40)$$

$$d_{\epsilon} R[\epsilon(\tau), t - \tau] = \frac{\partial R[\epsilon(t), t - \tau]}{\partial \epsilon} \dot{\epsilon}(\tau) d\tau \quad (3.41)$$

where $R_n(\epsilon, t)$ is the (non-linear) stress relaxation function. Pipkin and Rogers generalized this one-dimensional result to three-dimensional tensor relations by replacing σ by σ_{ij} , ϵ by ϵ_{ij} , S_n by $S_{ij(n)}$ and R_n by $R_{ij(n)}$. Furthermore they replace the initial strain condition $\epsilon = 0$ by $\epsilon_{ij} = \delta_{ij}$.

Smart and Williams compared the theoretical results of the first terms of the multiple integral expansion for the one-dimensional case with experimental data for PVC. They used both a stress relaxation formulation:

$$\sigma(t) = \int_{\tau=-\infty}^t d_{\epsilon} R[\epsilon(\tau), t - \tau] \quad (3.42)$$

as a creep formulation:

$$\epsilon(t) = \int_{\tau=-\infty}^t d_{\sigma} C[\sigma(\tau), t - \tau] \quad (3.43)$$

where $C(\sigma, t)$ is the creep strain after a stress of σ has been applied for time t . The results are shown in figures 3.7 and 3.8. The predictions of stress responses from strain inputs are rather poor. The errors are smaller for PVC than for polypropylene because PVC exhibits a smaller nonlinearity and viscosity than polypropylene.

3.4.3 The Schapery thermodynamic theory

Schapery developed stress-strain equations for nonlinear, initially isotropic, viscoelastic materials with constant temperature by extending Biot's linear thermodynamic theory [25]. The basic assumption underlying Schapery's theory is that a viscoelastic material behaves thermodynamically as a linear viscoelastic system but with nonlinear measures of strain and with modified stresses. This leads to the following set of constitutive equations for three-dimensional nonlinear viscoelastic

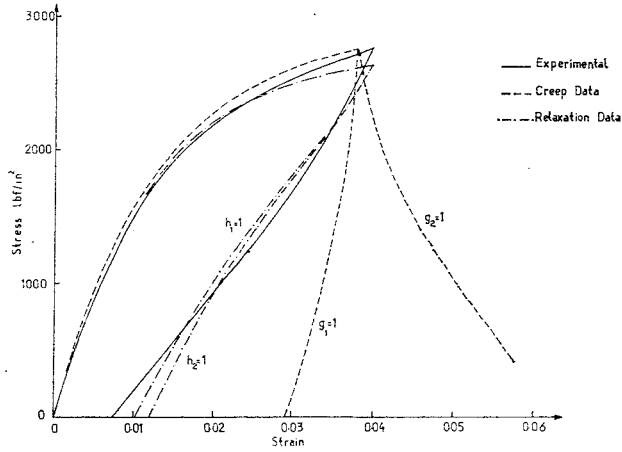


Figure 3.9: Predictions for constant strain rate loading using the Schapery theory for polypropylene (Smart and Williams, 1972)

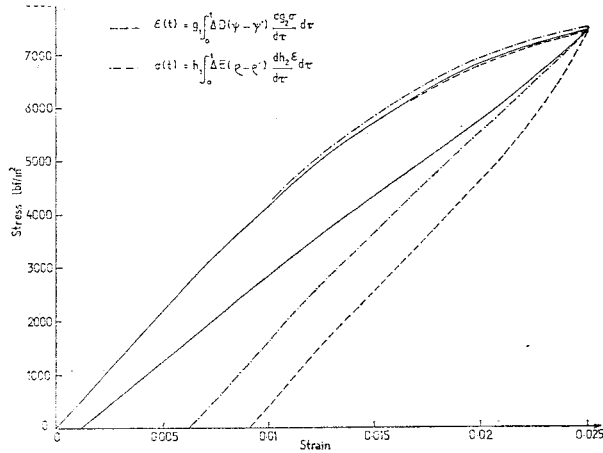


Figure 3.10: Predictions for constant strain rate loading using the Schapery theory for polyvinylchloride (pvc). (Smart and Williams, 1972)

behaviour:

$$\left. \begin{aligned}
\sigma_{xx} &= \frac{\partial \Delta W_e}{\partial \epsilon_{xx}} + \frac{2}{3} \int_0^t G(\rho - \rho') \frac{d}{d\tau} (2\epsilon_{xx} - \epsilon_{yy} - \epsilon_{zz}) d\tau + \int_0^t K(\rho - \rho') \frac{de}{d\tau} d\tau \\
\sigma_{yy} &= \frac{\partial \Delta W_e}{\partial \epsilon_{yy}} + \frac{2}{3} \int_0^t G(\rho - \rho') \frac{d}{d\tau} (2\epsilon_{yy} - \epsilon_{xx} - \epsilon_{zz}) d\tau + \int_0^t K(\rho - \rho') \frac{de}{d\tau} d\tau \\
\sigma_{zz} &= \frac{\partial \Delta W_e}{\partial \epsilon_{zz}} + \frac{2}{3} \int_0^t G(\rho - \rho') \frac{d}{d\tau} (2\epsilon_{zz} - \epsilon_{xx} - \epsilon_{yy}) d\tau + \int_0^t K(\rho - \rho') \frac{de}{d\tau} d\tau \\
\sigma_{xy} &= \frac{\partial \Delta W_e}{\partial \epsilon_{xy}} + \int_0^t G(\rho - \rho') \frac{d\epsilon_{xy}}{d\tau} \\
\sigma_{xz} &= \frac{\partial \Delta W_e}{\partial \epsilon_{xz}} + \int_0^t G(\rho - \rho') \frac{d\epsilon_{xz}}{d\tau} \\
\sigma_{yz} &= \frac{\partial \Delta W_e}{\partial \epsilon_{yz}} + \int_0^t G(\rho - \rho') \frac{d\epsilon_{yz}}{d\tau}
\end{aligned} \right\} (3.44)$$

$$\rho = \int_0^t \frac{dt}{a_\epsilon}; \quad \rho' = \int_0^\tau \frac{dt}{a_\epsilon} \quad (3.45)$$

where: σ_{ij} are the Cauchy stress components referred to a Cartesian base.

ϵ_{ij} are the strain components referred to a Cartesian base.

$e = \epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}$.

ΔW_e is the equilibrium free energy.

a_ϵ is the strain shift factor.

ρ is a strain reduced time.

G is the shear relaxation modulus.

K is the bulk relaxation modulus.

However it is often difficult to apply these constitutive equations to specific problems and to simple configurations which are used in material property evaluation. Normally it will be easier to derive the constitutive relations for each application separately [25].

The constitutive equations for uniaxial loading (as well for the stress in terms of the strain history as for the strain in terms of the stress history) are given by Smart and Williams [23]. They showed that the one-dimensional Schapery thermodynamic theory modelled loading behaviour of polypropylene and PVC very well using both a stress history and a strain history input. In unloading the predictions for a strain input analysis were superior to those for a stress input. See figures 3.9 and 3.10. The disadvantage of the Schapery model is that it is complicated to determine the shift factors and modulus.

Henriksen [26] and also Rooyackers [27] derived finite element models of the Schapery thermodynamic theory. Henriksen extended the one-dimensional single integral constitutive equation of Schapery (for the strain in terms of the stress history) to general three-dimensional behaviour by assuming that each strain component is a linear function of the stress operators. This approach is not correct

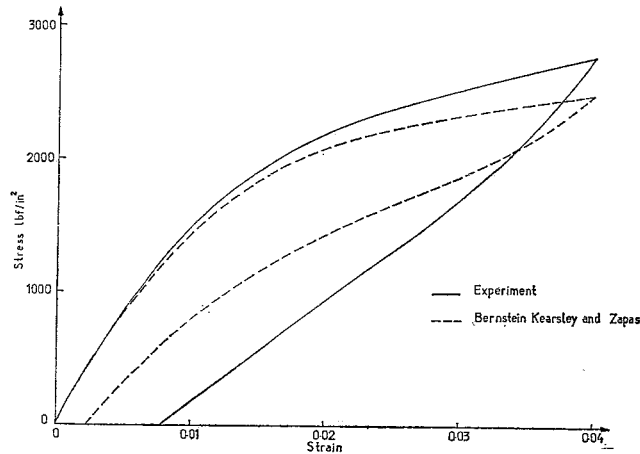


Figure 3.11: Predictions for constant strain rate loading using the BKZ-fluid theory for polypropylene (Smart and Williams, 1972)

according to Rooyackers. Rooyackers derived the three-dimensional formulation of the Schapery model from irreversible thermodynamics and implemented these equations for isotropic material behaviour in a finite element package.

3.4.4 The Bernstein, Kearsley and Zapas theories

Bernstein, Kearsley and Zapas [4] developed two forms of single integral constitutive equations. They observed that at low temperatures and for short times, it is difficult to distinguish the behaviour of a viscoelastic material from that of an elastic solid. In this case it is possible to choose a constitutive equation which is defined with respect to a preferred configuration. When the material is observed for sufficiently long times and at sufficiently high temperatures, flow behaviour is more significant and a fluid theory can be applied (no preferred configuration).

Bernstein, Kearsley and Zapas expressed the free energy for an incompressible solid in terms of the strain invariants I_1 and I_2 commonly used in the theory of incompressible nonlinear elasticity (see equations 3.12 and 3.13):

$$W = \left(\frac{1}{2}M + B\right)(I_1 - 3) + \frac{1}{4}\left(\frac{1}{2}A + B\right)(I_1 - 3)^2 - \frac{1}{2}B(I_2 - 3) \quad (3.46)$$

Where M is a constant and A and B are material functions. Note that if $A + 2B = 0$ the long term behaviour is that of a Mooney material, whereas if $A = B = 0$ it is Neo-Hookean. In a simple extension stress relaxation test, equation 3.46 can be

written as:

$$\frac{\sigma(t)}{(\lambda^2 - \lambda^{-1})} = (\lambda^2 - 1)\left[\frac{1}{2}A(t) + B(t)\right] + \lambda^{-1}[A(t) + B(t)] + M - A(t) \quad (3.47)$$

The BKZ theory for an incompressible elastic fluid can be written as:

$$\boldsymbol{\sigma}(t) = -p\mathbf{I} + \int_{-\infty}^t \left\{ [M(t-\tau) + \frac{1}{2}a(t-\tau)(\text{tr}\mathbf{B}_\tau - 3) - b(t-\tau)]\mathbf{B}_\tau + b(t-\tau)\mathbf{B}_\tau^2 \right\} d\tau \quad (3.48)$$

Where $\mathbf{B}_\tau = \mathbf{F}_\tau \mathbf{F}_\tau^C$ is the left Cauchy-Green strain measure, corresponding to:

$$\mathbf{F}_\tau = (\vec{\nabla}_\tau \vec{x})^c \quad (3.49)$$

and M , a and b are material functions. For simple extension stress-relaxation tests it then follows that:

$$\begin{aligned} \frac{\sigma(t)}{(\lambda^2 - \lambda^{-1})} &= (\lambda^2 - 1)\left[\frac{1}{2}A(t) + B(t)\right] + \lambda^{-1}[A(t) + B(t)] \\ &+ M(t) - A(t) \end{aligned} \quad (3.50)$$

Equations (3.47) and (3.50) are similar in form, however $M(t)$, $A(t)$ and $B(t)$ are not the same in the two expressions.

Lockett [4] describes a method to experimentally determine the material functions in the BKZ theories. All measurements indicate that the fluid theory is more appropriate to describe nonlinear viscoelastic material behaviour. Smart and Williams [23] compared the BKZ theory for an elastic fluid with the constant strain rate loading and unloading of PVC and polypropylene. Especially during unloading the data from the constitutive equations show poor agreement with the experimental results, see figure 3.11.

3.4.5 The theory of Bloch, Chang and Tschoegl

Bloch, Chang and Tschoegl [28] observed that the effects of time and strain are separable at moderate strains in as well uniaxial as multiaxial stress relaxation or creep curves obtained on soft rubberlike materials. If the effects of time and strain are uncoupled then time-shift invariance (in the sense of the Boltzmann superposition principle) is preserved. This means that the ratio of stress (or strain) at time t to that at an arbitrary reference time t_r in a stress relaxation experiment (or a creep experiment) is independent of strain (or stress). Deformations to which the above applies are called moderately large deformations. In such a deformation, nonlinear viscoelastic behaviour results only from time-independent stress-strain nonlinearity, which can be accounted for by choosing an appropriate strain measure.

Bloch, Chang and Tschoegl proposed the following two models:

1. The solid model, which is designed for polymeric solids such as crosslinked rubberlike materials.
2. The liquid model, which is designed for polymeric liquids such as polymeric melts.

Both models contain a single time dependent material function (the relaxation modulus of linear viscoelasticity) and the material constant n which characterizes the strain measure. The time dependent material function and the material constant n have to be determined by experiment (response to a step of strain in simple tension).

The solid model for an isotropic incompressible rubberlike material is given by:

$$\bar{\sigma}_\alpha(t) = -p + \frac{2}{3n} \int_0^t E(t-u) \frac{d\lambda_\alpha^n(u)}{du} du \quad \alpha = 1, 2, 3 \quad (3.51)$$

where: $\bar{\sigma}_\alpha(t)$ is the principal true stress in the α -direction.

p is an unspecified hydrostatic pressure.

n is a material constant characterizing the strain measure.

$E(t)$ is the small deformation tensile relaxation modulus.

$\lambda_\alpha(u)$ is the principal stretch ratio in the α -direction at the past time u .

In an isothermal deformation $\lambda_\alpha(u)$ is defined as:

$$\lambda_\alpha(u) = \frac{L_\alpha(u)}{L_{\alpha 0}} \quad (3.52)$$

where: $L_\alpha(u)$ is the length of the specimen in α -direction at time u .

$L_{\alpha 0}$ is the length of the specimen in the same direction at zero stress.

The liquid model yields the equation:

$$\bar{\sigma}_\alpha(t) = -p + \frac{2}{3n} \int_0^t E(t-u) d\left[\frac{\lambda_\alpha(t)}{\lambda_\alpha(u)}\right]^n du \quad \alpha = 1, 2, 3 \quad (3.53)$$

for the principal true stress component in the homogenous deformation of an incompressible material.

In order to show the validity of the solid model experiments were done on a dicumyl-peroxide-cured styrene butadiene rubber (SBR), lightly plastized with silicone oil (1.5%) [29]. The small deformation tensile relaxation modulus $E(t)$ was determined at the reference temperature (23°C) with the use of time-temperature superposition from a relaxation curve in the linear region of SBR. To determine $E(t)$ the following equation was used:

$$E(t) = E_e + \frac{E}{[1 + (\frac{t}{\tau})^k]} \quad (3.54)$$

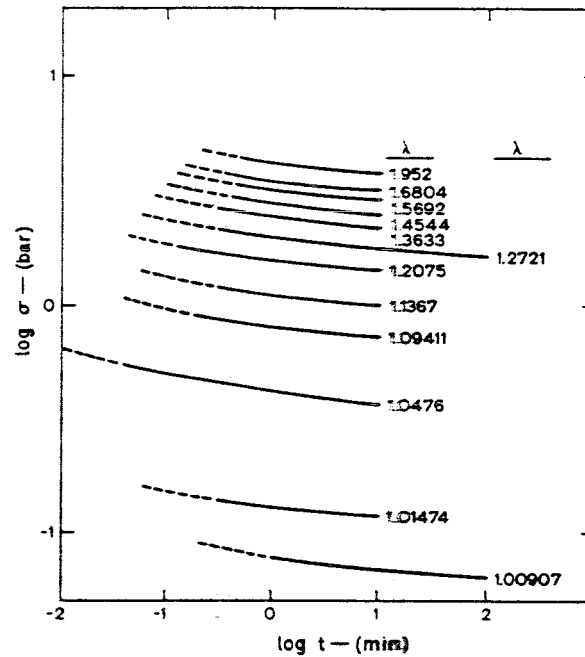


Figure 3.12: Response of SBR at 23°C to a step of strain at various stretch ratios as a function of time. (Bloch, Chang and Tschoegl, 1978)

The coefficients in equation (3.54) were found by nonlinear least squares fitting. The strain parameter n was obtained in relaxation tests by imposing strains which are outside the linear true stress-strain range. Such curves are shown in figure 3.12. The curves are parallel which means that time shift invariance is preserved. At a certain, so-called isochronal, time t_r (in this case $t_r = 10$ min.) the stress σ_r is cross plotted as a function of λ_r as shown in figure 3.13. The parameter n is then obtained from nonlinear least square fit to the data.

For the stretch ratio as a ramp function of time the numerical results (solid lines) of the solid model are compared with the experimental results (solid circles) in figure 3.14. A more severe test is the prediction of the response to the removal or reversal of the strain. The results of the solid model (solid lines) and the experimental data (solid circles) are shown in figure 3.15. From these figures it is shown that the solid model represents the behaviour of SBR very well for deformations until about 150% strain in simple extension.

3.4.6 An empirical model of the Mullins effect

The stresses observed during the return of a stretched rubber to an unstrained state are less than the corresponding stresses during the extension. This phenomenon is

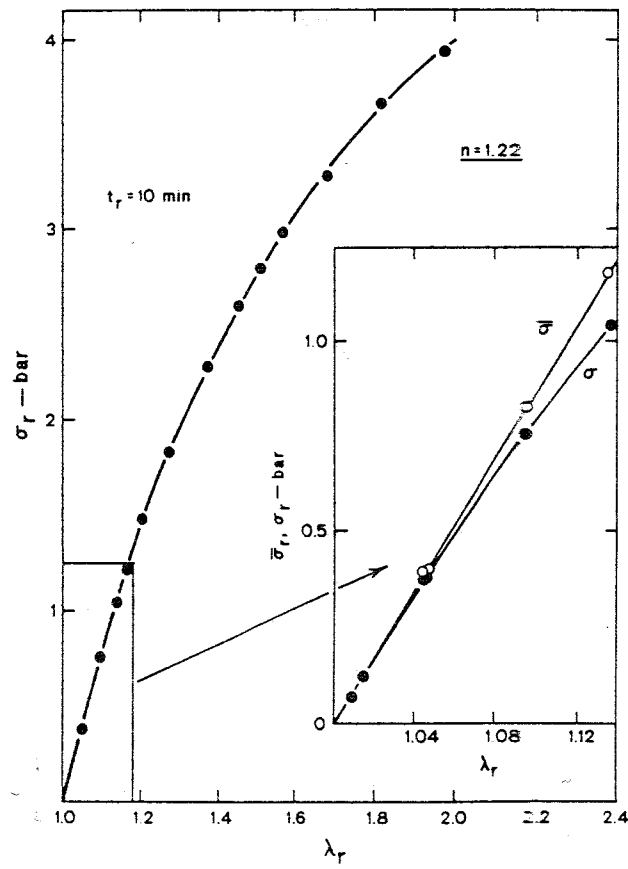


Figure 3.13: Isochronal stress-strain curve for SBR at 23°C. (Bloch, Chang and Tschögl, 1978)

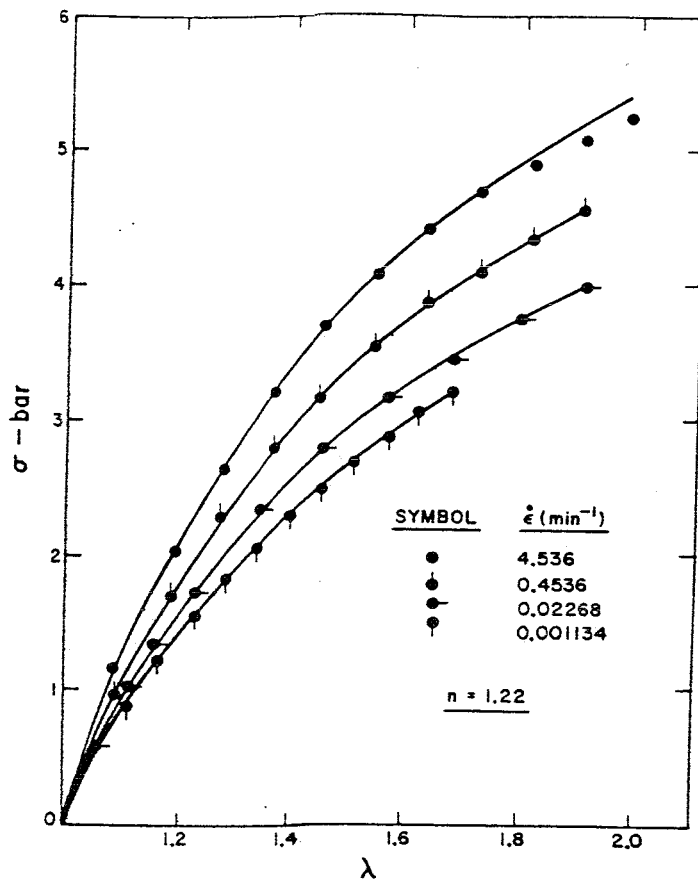


Figure 3.14: Response of SBR at 23°C to a ramp of strain at various rates of extension. (Bloch, Chang and Tschoegl, 1978)

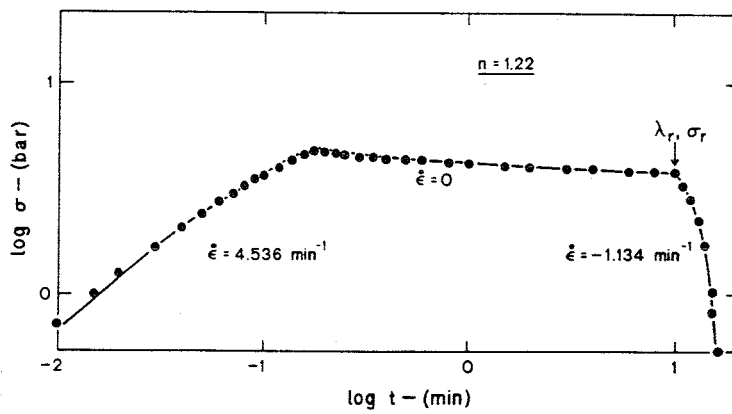


Figure 3.15: Response of SBR at 23°C to a trapezoidal strain as a function of time. (Bloch, Chang and Tschoegl, 1978)

known as the Mullins effect. (See also section 2.4).

The Boltzmann equation for linear viscoelastic material behaviour is given by:

$$\sigma(t) = \int_0^t E(t - \tau) \dot{\epsilon} d\tau \quad (3.55)$$

where $E(t - \tau)$ is the stress relaxation function. Beyond the maximum strain for which the above equation is valid, the following constitutive equation can be employed:

$$\sigma(t) = \int_0^t E(t - \tau, \epsilon(t - \tau)) \dot{\epsilon}(\tau) d\tau \quad (3.56)$$

If time-shift invariance is preserved, then the effects of time and strain can be uncoupled:

$$\sigma(t) = \int_0^t E(t - \tau) g(\epsilon) \dot{\epsilon} d\tau \quad (3.57)$$

where $g(\epsilon)$ is the so-called damping function. The presence of this damping function reflects the non-linearity. The term $g(\epsilon) \dot{\epsilon}$ can be considered as a new strain measure, which is related to the stress σ . By application of the Mooney-Rivlin equation (3.15) the following damping function has been suggested:

$$g(\epsilon) = 1 + \frac{C_2}{C_1} \lambda^{-1} \quad (3.58)$$

which by custom is normalized to unity at $\epsilon = 0$.

Roland [30] shows that this equation, which assumes reversible damping, fails to describe the Mullins effect. He then introduces irreversibility into the damping function by assuming that the damping function during extension is given by equation (3.58), and after reversal of the strain by:

$$g(\epsilon) = 1 + \frac{C_2}{C_1} \lambda_{\max} \quad (3.59)$$

where λ_{\max} is the extension ratio at reversal. He shows that this assumption improves the agreement between the experimental and calculated results.

The assumption of irreversibility is based on the notion that in reversing deformations of polymeric liquids the stresses after reversal of the strain are less than the values calculated assuming linear viscoelasticity. Although this decrease in modulus is caused by a different physical effect than for rubber networks, it is similarly accounted for in the constitutive equation by introduction of a damping function. It has been suggested that this damping function for polymeric fluids is not reversible; that means that when the strain is reversed the damping function does not increase back to unity. This implies that the structure breakdown is not recovered upon strain reversal. The Mullins effect in rubber has a similar origin to that underlying the irreversibility of the damping function for polymeric liquids. But unlike in polymeric liquids sufficiently slow deformation of networks is fully reversible. A consequence of this is that in the damping function for rubber time dependence should be included.

3.5 Differential and rate type models for non-linear viscoelastic material behaviour.

3.5.1 Introduction

Constitutive equations of the rate or differential type found in literature are commonly used to describe the behaviour of non-linear viscoelastic fluids rather than that of nonlinear viscoelastic solids. It is necessary that the derivatives which occur in these equations are objective quantities. The principle of objectivity or material frame indifference states that the stress state in a body may not change when the body is subjected to a rigid body rotation or translation. It has been shown that partial derivatives and material derivatives do not satisfy this requirement [4].

Objective time derivatives of the strain measure can be written in the form of a set of kinematic tensors as was done by Rivlin and Ericksen [31]:

$$\mathbf{A}_1 = 2\mathbf{D} \quad (3.60)$$

$$\mathbf{A}_{n+1} = \dot{\mathbf{A}}_n + \mathbf{A}_n \mathbf{L}^c + (\mathbf{A}_n \mathbf{L}^c)^c, \quad n = 1, 2, \dots \quad (3.61)$$

where $(\dot{})$ denotes the material time derivative. \mathbf{L} is the velocity gradient tensor, defined by:

$$\mathbf{L} = (\vec{\nabla} \vec{v})^c \quad (3.62)$$

and \mathbf{D} is the rate of deformation tensor, defined by:

$$\mathbf{D} = \frac{1}{2}(\mathbf{L}^c + \mathbf{L}) \quad (3.63)$$

In which $\vec{\nabla}$ is the gradient operator with respect to the current configuration of the body, and \vec{v} is the velocity vector. Eringen showed that for a restricted class of materials, the so-called materials of the rate type, the constitutive equation can be expressed in the form [32]:

$$\mathcal{F}\{\boldsymbol{\sigma}, \overset{(1)}{\boldsymbol{\sigma}}, \overset{(2)}{\boldsymbol{\sigma}}, \dots, \mathbf{A}_1, \mathbf{A}_2, \dots; \mathbf{B}\} = \mathbf{O} \quad (3.64)$$

where: \mathcal{F} is a tensor function.

$\boldsymbol{\sigma}$ is the stress tensor.

$\overset{(n)}{\boldsymbol{\sigma}}$ is a frame indifferent time derivative of the stress tensor.

\mathbf{B} is the left Cauchy Green tensor.

A more restricted class of materials is that for which \mathcal{F} does not depend on the stress rates, equation (3.64) then reduces to:

$$\boldsymbol{\sigma} = \mathcal{G}\{\mathbf{A}_1, \mathbf{A}_2, \dots; \mathbf{B}\} \quad (3.65)$$

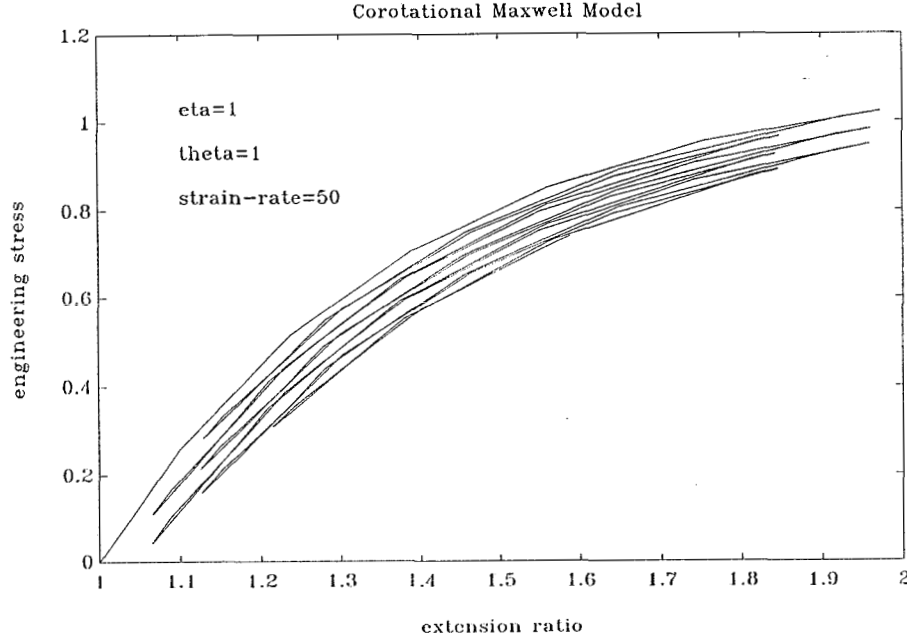


Figure 3.16: Hysteresis loops in uniaxial extension, calculated with the use of the corotational Maxwell model

where \mathcal{G} is a tensor function. These materials are called materials of differential type or Rivlin-Ericksen materials. When the dependence on \mathbf{B} is omitted from (3.65), the equation describes the behaviour of Rivlin-Ericksen fluids. On the other hand in the case of a purely elastic material $\boldsymbol{\sigma}$ depends only on the left Cauchy Green tensor \mathbf{B} and the constitutive equation can be expressed in the form:

$$\boldsymbol{\sigma} = \alpha_0 \mathbf{I} + \alpha_1 \mathbf{B} + \alpha_2 \mathbf{B}^2 \quad (3.66)$$

where α_i are functions of the three invariants of \mathbf{B} and may be determined from the strain energy function W [33].

3.5.2 Maxwell material models

Maxwellian materials belong to the class of materials of rate type. The simple linear one dimensional Maxwell equation (see table 3.1) is not frame invariant, due to the non-objectivity of the material time derivative. It is necessary that an objective time derivative is used. Olroyd recognized that infinitely many objective time derivatives can be constructed [34]. Objectivity requires that:

$$\dot{\mathbf{S}} - \boldsymbol{\Omega} \cdot \mathbf{S} - \mathbf{S} \cdot \boldsymbol{\Omega}^c - f(\mathbf{S}, \mathbf{D}) = \mathbf{O} \quad (3.67)$$

where: f is an objective tensor function.

\mathbf{S} is an arbitrary second order tensor.

$\boldsymbol{\Omega}$ is the rate of rotation tensor, $\boldsymbol{\Omega} = \frac{1}{2}(\mathbf{L} - \mathbf{L}^c)$.

\mathbf{L} is the velocity gradient tensor defined by equation (3.62).
 \mathbf{D} is the rate of deformation tensor defined by equation (3.63)

In the simplest case the corotational or Jauman rate defined by:

$$\overset{\circ}{\mathbf{S}} = \dot{\mathbf{S}} - \boldsymbol{\Omega} \cdot \mathbf{S} - \mathbf{S} \cdot \boldsymbol{\Omega}^c \quad (3.68)$$

is obtained. The corresponding Maxwell equation, called the corotational equation for an isotropic viscoelastic material is given by:

$$\left. \begin{aligned} \boldsymbol{\sigma} &= -p\mathbf{I} + \boldsymbol{\tau} \\ \overset{\circ}{\boldsymbol{\tau}} + \frac{1}{\theta}\boldsymbol{\tau} &= \frac{2\eta}{\theta}\mathbf{D} \end{aligned} \right\} \quad (3.69)$$

where: $\boldsymbol{\sigma}$ is the Cauchy stress tensor.

p is an isotropic constant.

\mathbf{I} is the unity tensor.

$\boldsymbol{\tau}$ is the extra stress tensor.

$\overset{\circ}{\boldsymbol{\tau}}$ is the Jauman rate of the extra stress tensor.

θ is a time constant.

η is a viscosity.

Another objective derivative which has received considerable attention in the theoretical literature is the upper-convected or Truesdell rate defined by:

$$\overset{\nabla}{\mathbf{S}} = \dot{\mathbf{S}} - \mathbf{L} \cdot \mathbf{S} - \mathbf{S} \cdot \mathbf{L}^c \quad (3.70)$$

The corresponding Maxwell equation for an isotropic viscoelastic material is given by:

$$\left. \begin{aligned} \boldsymbol{\sigma} &= -p\mathbf{I} + \boldsymbol{\tau} \\ \overset{\nabla}{\boldsymbol{\tau}} + \frac{1}{\theta}\boldsymbol{\tau} &= \frac{2\eta}{\theta}\mathbf{D} \end{aligned} \right\} \quad (3.71)$$

This equation is called the upperconvected Maxwell equation. It's intergral equivalent is called the Lodge equation and is given by:

$$\boldsymbol{\sigma} = - \int_{-\infty}^t \frac{\eta}{\theta^2} e^{\frac{(\tau-t)}{\theta}} \mathbf{B}(t, \tau) d\tau \quad (3.72)$$

where \mathbf{B} denotes the left Cauchy-Green tensor. The Lodge equation results from the molecular model of Green and Tobolsky. This so-called temporary network model is based on the notion that polymer chains that are not chemically cross-linked interact with each other to form a transient network. In this 'liquid' network strands break free from crosslinks and reform new ones at equal rates [34].

In figures 3.16, 3.17 and 3.18 the numerical results of the corotational, the upper-convected Maxwell model and the generalized (2 relaxation times and 2 viscosities) upperconvected Maxwell model for hysteresis loops in uniaxial extension are shown.

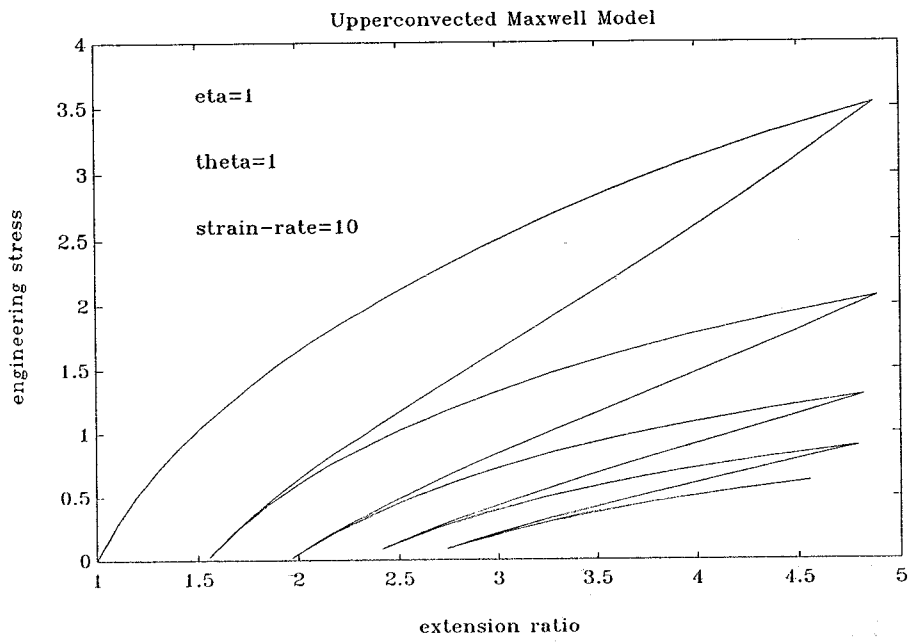


Figure 3.17: Hysteresis loops in uniaxial extension, calculated with the use of the upperconvected Maxwell model

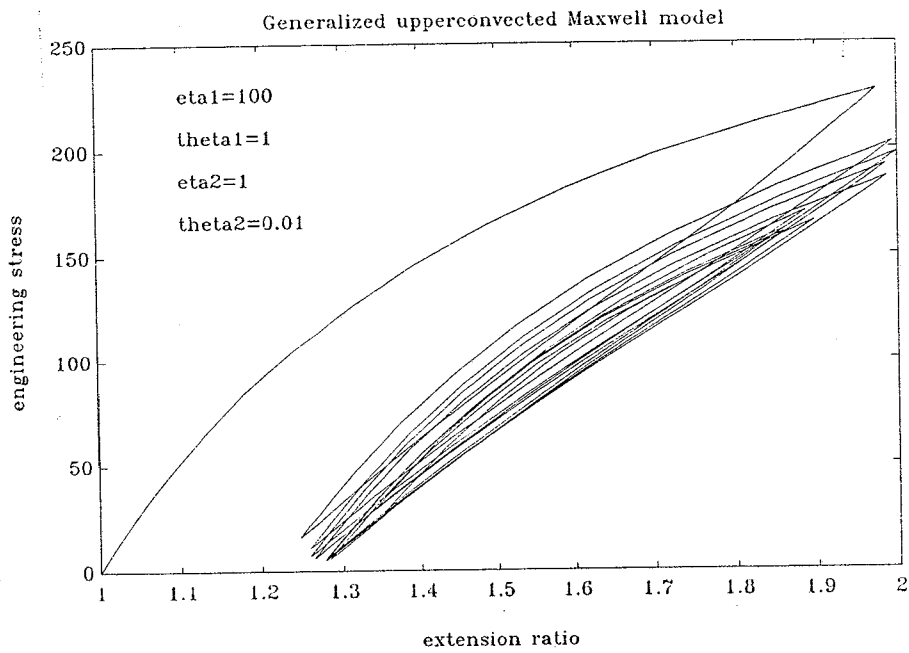


Figure 3.18: Hysteresis loops in uniaxial extension, calculated with the use of the generalized upperconvected Maxwell model

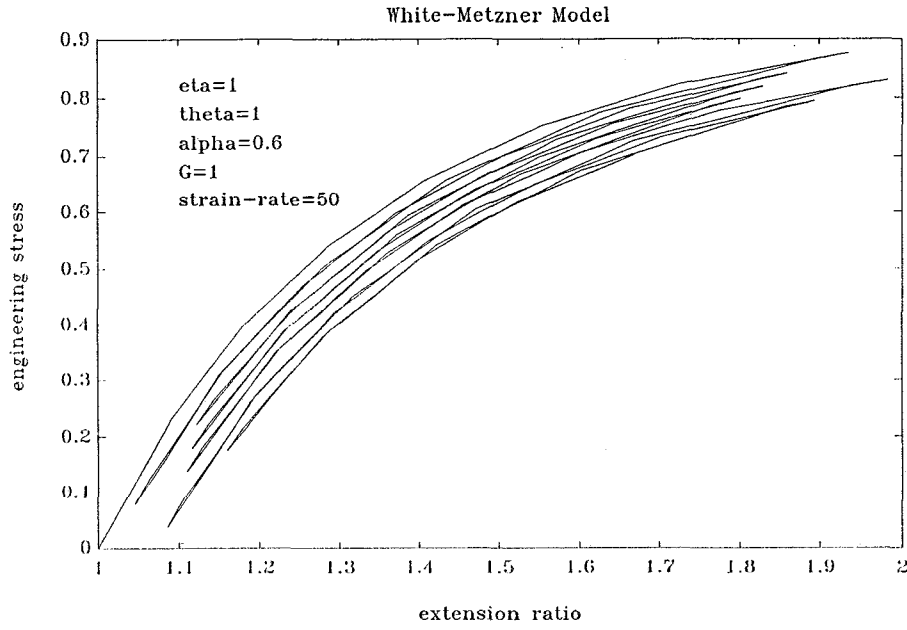


Figure 3.19: Hysteresis loops in uniaxial extension, calculated with the use of the White-Metzner equation

3.5.3 The White-Metzner equation

White and Metzner introduced a modification of the upperconvected Maxwell (UCM) differential equation that allows relaxation that, like non-affine motion, is faster than any deformation. Therefore their constitutive equation describes shear thinning and strain softening. The White-Metzner (WM) constitutive equation is given by:

$$\left. \begin{aligned} \boldsymbol{\sigma} &= -p\mathbf{I} + \boldsymbol{\tau} \\ \theta(I_D) \overset{\nabla}{\boldsymbol{\tau}} + \boldsymbol{\tau} &= 2\eta(I_D)\mathbf{D} \end{aligned} \right\} \quad (3.73)$$

Where θ and η depend on the rate of strain:

$$I_D = 2\mathbf{D} : \mathbf{D} \quad (3.74)$$

and

$$\eta(I_D) = G\theta(I_D) \quad (3.75)$$

where G is a modulus. One form for $\theta(I_D)$ proposed by Ide and White is [37]:

$$\theta(I_D) = \frac{\theta_0}{1 + \alpha\theta_0(2\mathbf{D} : \mathbf{D})^{\frac{1}{2}}} \quad (3.76)$$

where α is an empirical parameter.

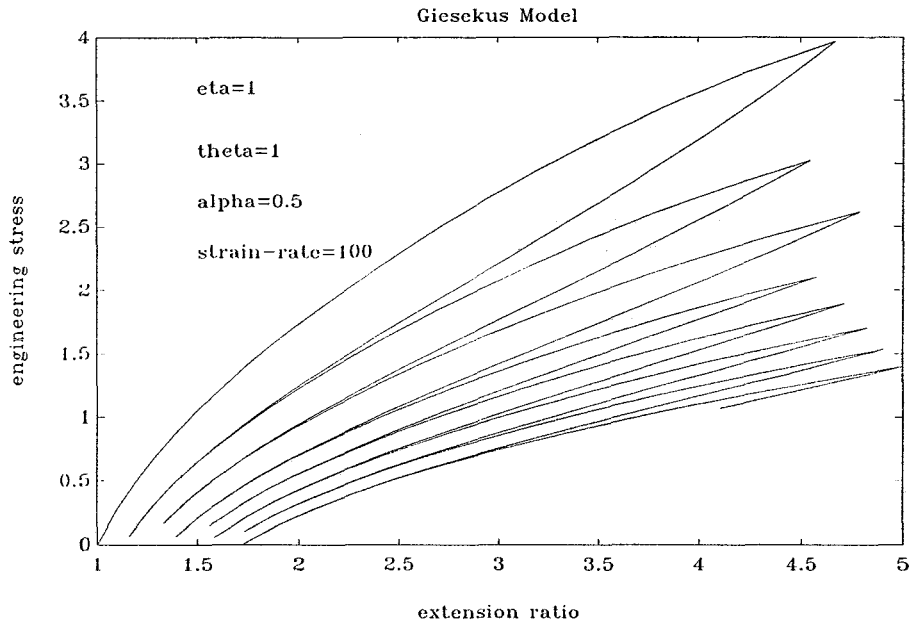


Figure 3.20: Hysteresis loops in uniaxial extension, calculated with the use of the Giesekus equation

3.5.4 The Giesekus model

The Giesekus model is based on a kinetic theory of closely packed polymer chains and on a series of simplifications. The extra stress tensor for the Giesekus model is given by:

$$\overset{\nabla}{\tau} + \frac{\alpha}{\eta} \tau^2 + \frac{1}{\theta} \tau = \frac{2\eta}{\theta} \mathbf{D} \quad (3.77)$$

where α is an empirical constant of proportionality. α lies in the range from 0 to 1. When $\alpha = 0$ the upperconvected Maxwell equation (UCM) is recovered. When $\alpha = 1$ the Giesekus model predicts extensional stresses and steady state shear identical to those of the corotational Maxwell equation (CRM). Thus as α is increased from 0 to 1 strain softening increases from that of the UCM equation (no strain softening) to that of the CRM equation [34] [35]. For $\alpha = 0.5$ the results of the Giesekus model are shown in figure 3.21.

3.5.5 The Leonov model

Leonov's constitutive equation in its simplest form is similar to the Giesekus model, but is derived from a thermodynamic rather than a molecular approach. The basic assumption underlying his theory is given by the principle of relaxibility in the small. This principle states that each infinitesimal material volume can be elastically relaxed from the current, deformed state, to an (imaginary) stress free configuration.

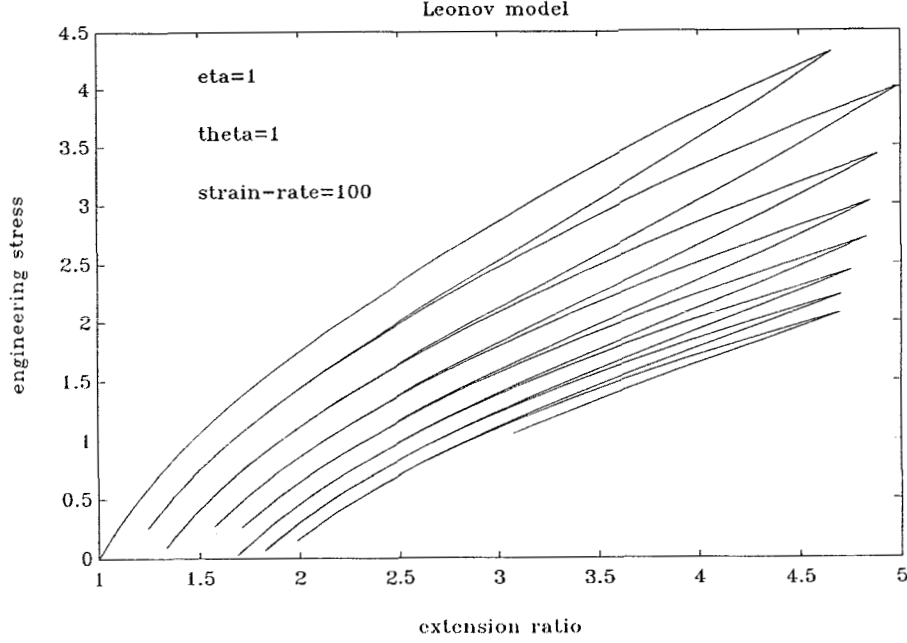


Figure 3.21: Hysteresis loops in uniaxial extension, calculated with the use of the Leonov equation

This implies that the deformation tensor \mathbf{F} can be multiplicatively decomposed in an elastic (\mathbf{F}_e) and a plastic (\mathbf{F}_p) part.

$$\mathbf{F} = \mathbf{F}_e \cdot \mathbf{F}_p \quad (3.78)$$

It follows logically that the elastic left Cauchy Green tensor is given by:

$$\mathbf{B}_e = \mathbf{F}_e \cdot \mathbf{F}_e^c \quad (3.79)$$

and the rate of deformation tensor by:

$$\mathbf{D} = \mathbf{D}_e + \mathbf{D}_p \quad (3.80)$$

where \mathbf{D}_e is the elastic rate of deformation tensor and \mathbf{D}_p is the irreversible rate of deformation tensor [38]. Furthermore the stress tensor can be split as follows:

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_e + \boldsymbol{\sigma}_p \quad (3.81)$$

where for isothermal behaviour $\boldsymbol{\sigma}_p = 0$.

For an incompressible isotropic medium of the Maxwellian type, Leonov [39, 40] relates the stress to the elastic strain:

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_e = -p\mathbf{I} + 2 \frac{\partial W}{\partial I_1} \mathbf{B}_e - 2 \frac{\partial W}{\partial I_2} \mathbf{B}_e^{-1} \quad (3.82)$$

where W is the free energy of deformation and I_1 and I_2 are the invariants of \mathbf{B}_e . In the simplest case W is given by the statistical theory of rubber elasticity:

$$W = \frac{\eta}{2\theta}(I_1 - 3) \quad (3.83)$$

Then:

$$\boldsymbol{\sigma} = -p\mathbf{I} + \frac{\eta}{\theta}\mathbf{B}_e \quad (3.84)$$

The elastic strain is related to the deformation history through the relation:

$$\overset{\nabla}{\mathbf{B}}_e + \mathbf{B}_e \cdot \mathbf{D}_p + \mathbf{D}_p \cdot \mathbf{B}_e = \mathbf{O} \quad (3.85)$$

The incompressibility constraint can be written as:

$$\text{tr}(\mathbf{D}) = 0 \quad (3.86)$$

Leonov assumes that:

$$\mathbf{D}_p = 2 \frac{\partial \psi}{\partial I_1} (\mathbf{B}_e - \frac{1}{3} I_1 \mathbf{I}) - 2 \frac{\partial \psi}{\partial I_2} (\mathbf{B}_e - \frac{1}{3} I_2 \mathbf{I}) \quad (3.87)$$

where $\psi = \frac{1}{8\theta}(I_1 + I_2 - 6)$ is called the nonequilibrium potential by Leonov. Then, equation (3.87) reduces to:

$$\mathbf{D}_p = \frac{1}{4\theta} [\mathbf{B}_e - \mathbf{B}_e^{-1} + \frac{1}{3}(I_2 - I_1)\mathbf{I}] \quad (3.88)$$

By substitution of equation (3.88) into equation (3.85), the simplest Leonov equation is obtained.

$$\left. \begin{aligned} \boldsymbol{\tau} &= \frac{\eta}{\theta} \mathbf{B}_e \\ \overset{\nabla}{\mathbf{B}}_e + \frac{1}{2\theta} [\mathbf{B}_e \cdot \mathbf{B}_e - \mathbf{I} - \frac{1}{3}(\text{tr} \mathbf{B}_e - \text{tr}(\mathbf{B}_e^{-1}))\mathbf{B}_e] &= \mathbf{O} \end{aligned} \right\} \quad (3.89)$$

3.5.6 The Larson equation

Larson proposed a differential model which obeys time strain separability. The model is given by:

$$\overset{\nabla}{\boldsymbol{\tau}} + \frac{2\alpha\theta}{3\eta} \mathbf{D} : \boldsymbol{\tau} (\boldsymbol{\tau} + \frac{\eta}{\theta} \mathbf{I}) + \frac{1}{\theta} \boldsymbol{\tau} = \frac{2\eta}{\theta} \mathbf{D} \quad (3.90)$$

The adjustable parameter α describes the strain softening character of the material. For $\alpha = 0$ the UCM is retrieved.

This model gives a reasonable description of experimental data in step-shear, steady uniaxial extension and step biaxial extension of a high-density polyethylene (HDPE) for $\alpha = 0.35$. For this value of α , the results of the use of the Larson equation in uniaxial extension are shown in figure 3.22. However as with the Giesekus equation the uniaxial data shows less strain softening than the shear data, i.o.w. the value of α that would best fit the uniaxial data is less than that which would give the best fit to the shear data [34] [35].

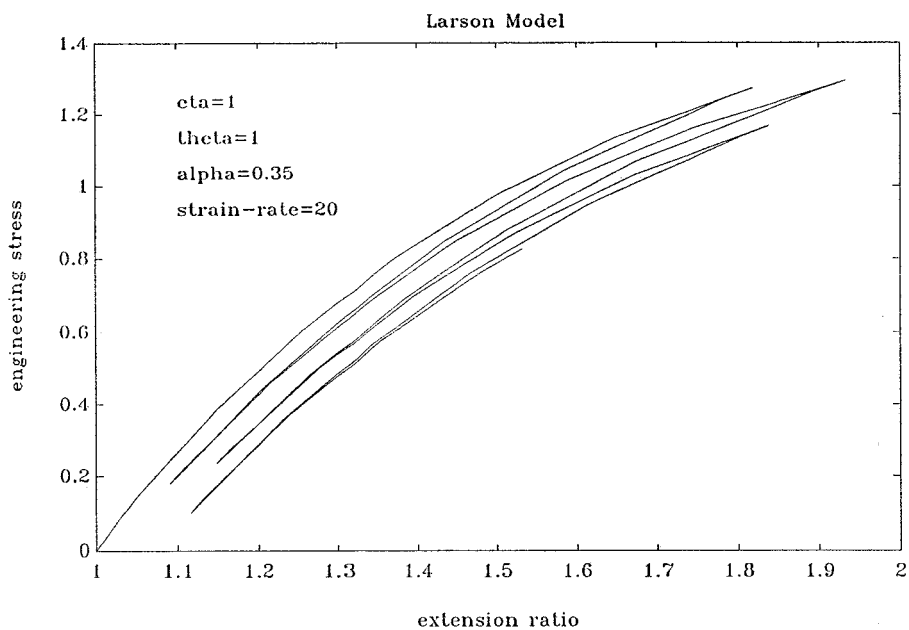


Figure 3.22: Hysteresis loops in uniaxial extension, calculated with the use of the Larson equation

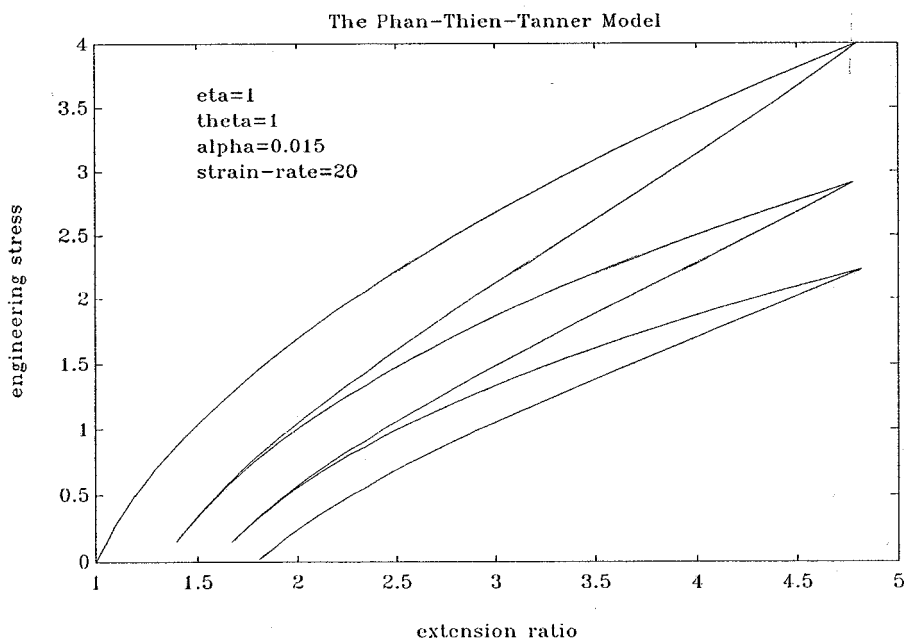


Figure 3.23: Hysteresis loops in uniaxial extension, calculated with the use of the Phan-Thien-Tanner model

3.5.7 The Phan-Thien-Tanner model

Phan-Thien and Tanner derived a constitutive equation from a Lodge-Yamamoto type of network theory for polymeric fluids. Phan-Thien and Tanner allowed the network junctions to have a certain slip and assumed that the rate of creation and destruction of the junctions depend on the instantaneous elastic energy of the network. Their constitutive equation can be described by the following set of equations [42]:

$$\left. \begin{aligned} \tau &= \sum_i \tau_i \\ \lambda_i \dot{\tau}_i + \tau_i \left(1 + \frac{\alpha}{G_i} \text{tr} \tau_i\right) &= \frac{2G_i \lambda_i}{1-\xi} \mathbf{D} \end{aligned} \right\} \quad (3.91)$$

where λ_i denotes the relaxation spectrum, G_i denotes the rigidity modulus spectrum and α, ξ are adjustable parameters.

When ξ is set to zero the motion is assumed to be affine. One version of the Phan-Thien-Tanner (PTT) model with $\xi = 0$ is given by:

$$\dot{\tau} + \frac{1}{\theta} \exp\left(\frac{\alpha\theta}{\eta} \text{tr}(\tau)\right) \tau = \frac{2\eta}{\theta} \mathbf{D} \quad (3.92)$$

This version gives reasonable good agreement with experimental data in step shear, steady uniaxial extension and step biaxial extension of HDPE [35]. With the use of this version hysteresis loops in uniaxial extension are computed. The results are shown in figure 3.23.

3.5.8 Non-linear Kelvin-Voight solid

The fundamental idea underlying the Kelvin-Voight solid is that the stress is dependent on the strain and deformation rate tensors. The one-dimensional linear theory for a Kelvin-Voight solid is given by (see also table 3.1):

$$\sigma = E\epsilon + \eta\dot{\epsilon} \quad (3.93)$$

A generalization of this one-dimensional linear theory to a non-linear three-dimensional theory is given by a constitutive equation of the form:

$$\sigma_{ij} = \mathcal{F}_{ij}(\mathbf{F}_{ij}, \dot{\mathbf{F}}_{ij}, \theta, X_i) \quad (3.94)$$

where: σ_{ij} are the components of the stress tensor.

\mathcal{F}_{ij} is a tensor function.

\mathbf{F}_{ij} are the components of the deformation gradient tensor.

X_i are the material or Lagrangian coordinates.

$\theta = \theta(X_i, t)$ is the temperature.

and simialar expressions can be derived for the heat vector \vec{q} , the internal energy E and the entropy S . An explicit expression for the stress is obtained by Eringen. However this expression is too complicated for most applications [43]. An approximation can be found in reference [44]

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Chapter 4

Measurement techniques

4.1 Introduction

A great variety of tests to determine the properties of rubber can be found in literature, see e.g. [1]. In the case of rubbers designed for suspensions it is important to have a good knowledge of the dynamic stress-strain properties. Non-linearities in this dynamic stress-strain behaviour arises from two sources [2]:

1. Modes of deformations and geometries which give rise to a non-linear stress-strain curve.
2. Inherent non-linearities due to material behaviour which manifest themselves in all modes of deformation.

The non-linearities due to 1 are dependent upon the amplitude of vibration, frequency of vibration and temperature. The non-linearities due to 2 are dependent upon the amplitude and number of cycles. Furthermore the effect of prestrain is not to be neglected [6]. Therefore all test conditions should be specified and dynamic measurements shall be made only after at least six cycles of stress have been applied to the test pieces. This is to permit internal structural rearrangements to reach near equilibrium, so that the results are reproducible. General requirements for dynamic testing of elastomers are given in ISO 2856 [7].

4.2 Static test methods

4.2.1 Hardness

The most common standard method of characterizing a rubber compound is to measure its hardness. The term hardness means resistance to deformation. Hardness is measured by indentation of a rigid body into the rubber material. It is an approximate measurement which is related to the modulus of elasticity [1]. However for highly non-linear rubbers this relation is not valid [8].

In indentation tests as Shore or IRHD tests the loading procedure and uncertainties in measuring delay are major causes of dispersion. An instrument that takes into account the relaxation of rubber is the so-called dynalyser. This instrument is contrary to other indentation tests based on deformation control. The deformation will be the same for soft and hard rubber, the force variation is measured. The dynalyser is unique in the sense that it also provides frequency dependent relations. This dynamic data is calculated from the relaxation behaviour measured by the indentation test. The calculation is based on two principles [3]:

1. The correspondence principle which states that the stress and strain relations in a visco-elastic medium are similar to those existing in an elastic medium, provided that the strain is applied instantaneously and that a time-dependent modulus is introduced instead of Young's modulus.
2. The superposition principle of Boltzman expressing the linearity of the material.

4.2.2 Tensile stress/strain properties

Probably the most common test on rubber, after hardness is that of tensile properties. The test piece is in the shape of either a dumb-bell or a ring. The latter avoids difficulties of gripping. The test piece is stretched at a constant rate of elongation until it breaks [4]. The quantities commonly measured are [5]:

- Tensile strength, the tensile stress at break.
- Elongation at break, the difference between the final and initial lengths expressed as a percentage of the latter.
- Modulus, tensile stress at a specified elongation. This value is determined during the test. Use of Young's modulus of a rubber at a unspecified strain can lead to significant errors in predicted stiffness.

As rubber is seldom used under conditions where the breaking stress is approached, and test methods more relevant to service conditions are being developed, there will be a tendency to attach less importance to the properties at the breaking point [1].

4.2.3 Tension set and compression set

In both a tension set test as a compression set test a rubber test piece is deformed, respectively in tension or compression, for a specified time, then allowed to recover for a specified time and the residual strain (set) is measured. In both tests the applied deformation may be under a constant stress or to a constant strain. The latter method is being more common [1].

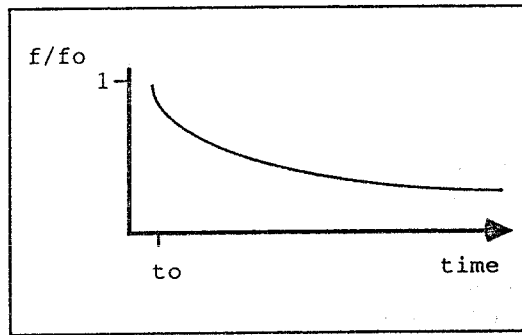


Figure 4.1: A typical stress relaxation normalized to time t_0 (Björk, Dickman and Stenberg, 1989)

4.3 Dynamic and related test methods

4.3.1 Creep and relaxation tests

Measurements of either creep or stress relaxation can be made in tension, compression or shear. They cover the frequency range from approximately 1 Hz to very low frequencies and are very effective in revealing the non-linear visco-elasticity. For creep measurements a constant load is applied and the gradual increase in strain is measured. For stress relaxation tests the rubber is held at a constant strain and the load needed to maintain this strain is measured. A typical stress relaxation curve is given in figure 4.1 At a so-called iso-chronal time the stress is crossplotted as a function of the extension ratio and a so-called isochronal stress-strain curve is obtained (e.g. figure 3.13).

When the logarithm of the stresses, obtained from stress relaxation tests at various stretch ratio's, are plotted against the logarithm of the time, for most rubbers a similar figure as figure 3.12 is obtained. Time-shift invariance is preserved if the curves are parallel. This means that the effects of time and strain are factorable, so that the iso-chronal stress-strain curve may be written as a modified Hooke's law with a time dependent modulus $S = E(t)f(\epsilon)$, where $f(\epsilon)$ is an appropriate function of the strain [9].

Dynamic stress relaxation is measured by compressing or extending a rubber test piece by a prestrain S and then superimposing a sinusoidally varying strain with amplitude A . The deformation thus varies between a maximum length and a minimum length. The force is constantly being measured and is recorded at its minimum and maximum values. These values respectively occur after the minimum or maximum values of the deformation due to the viscoelastic nature of rubber materials. The results are presented in a diagram by two lines, one for the maximum and one for the minimum values [10].

4.3.2 Forced vibration

In this test the material specimen is forced to execute homogenous vibrations at some frequency ω . Frequencies lower than approximately 10^{-4} Hz are difficult to achieve and due to the neglect of inertia effects this method cannot be used at frequencies in excess of about 10^3 Hz. It is immaterial whether stress or strain is the applied quantity provided that the other is measured. When a sinusoidal displacement is applied to a linear viscoelastic material, the response is a sinusoidal stress having the same frequency but a phase difference δ . The stress and strain cycles are represented by the real parts of [19]:

$$\sigma(t) = \sigma_0 \exp[i(\omega t + \delta)] \quad (4.1)$$

and

$$\epsilon(t) = \epsilon_0 \exp(i\omega t) \quad (4.2)$$

respectively, where $\omega = 2\pi f$. The complex modulus is defined by the stress to strain ratio:

$$M^* = \frac{\sigma_0}{\epsilon_0} \exp(i\delta) \quad (4.3)$$

This can be written into a real component (M') and an imaginary component (M'') such that:

$$M^* = M' + iM'' \Rightarrow M' = \frac{\sigma_0}{\epsilon_0} \cos \delta \quad \text{and} \quad M'' = \frac{\sigma_0}{\epsilon_0} \sin \delta \quad (4.4)$$

M' is called the storage modulus and is proportional to the maximum energy stored per cycle whilst M'' is called the loss modulus and is proportional to the energy dissipated per cycle. The loss factor ($\tan \delta$) is defined by:

$$\tan \delta = \frac{M''}{M'} \quad (4.5)$$

Determination of the moduli of rubber is most conveniently carried out under a simple shear deformation. The complex shear modulus is defined by (in analogy with equation (4.3)):

$$G^* = \frac{\tau(t)}{\gamma(t)} \quad (4.6)$$

where $\tau(t)$ is the dynamic shear stress and $\gamma(t)$ is the dynamic shear strain. Displaying the stress-strain loop for one circle of deformation (assuming linear viscoelasticity) results in the Lissajou figure of figure 4.2. From this figure δ and the moduli can be obtained [2]:

$$\sin \delta = \frac{B_\gamma}{A_\gamma} \quad (4.7)$$

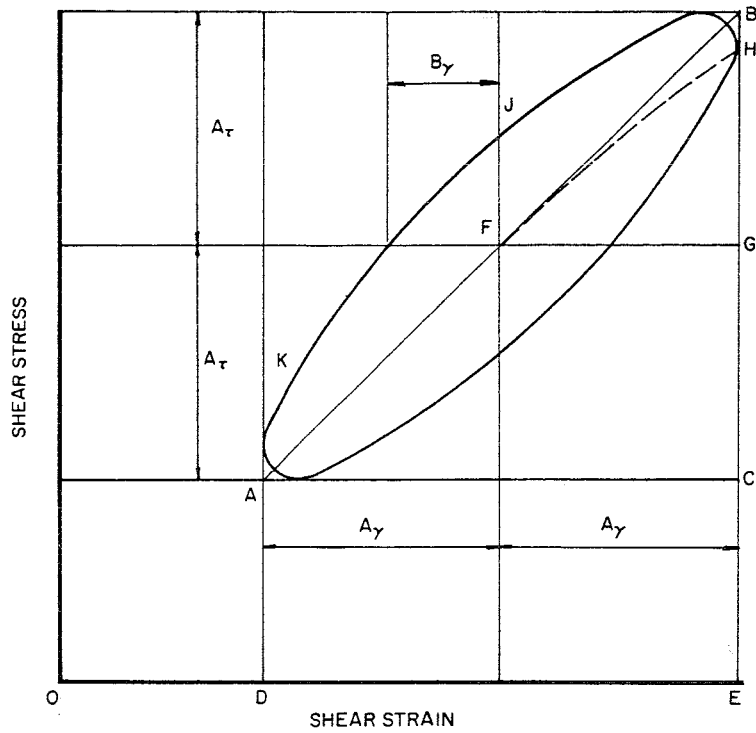


Figure 4.2: Hysteresis loop in dynamic stress-strain cycles (linear viscoelasticity). (Freakley and Payne, 1978)

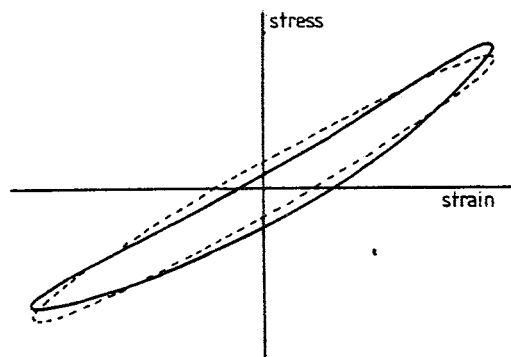


Figure 4.3: Hysteresis loop for NIR in compression, solid line: non-linear, dashed line: linear (from fundamentals). (Harris, 1987)

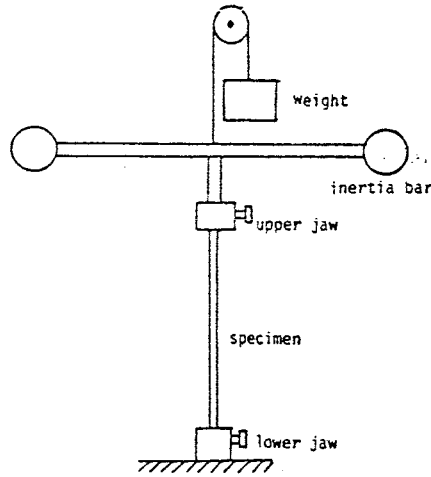


Figure 4.4: Torsion pendulum (Worth, 1985)

and

$$G' = \frac{BC}{AC} \cos \delta, \quad G'' = \frac{BC}{AC} \sin \delta \quad (4.8)$$

However rubbers commonly exhibit non-linear material behaviour. Input of a sinusoidal strain can lead to a stress output in which higher harmonics play a significant role [20]. The hysteresis loop is distorted from its linear elliptical form (see figure 4.3). According to Harris [21] the energy loss per cycle (the area of the ellipse) is the same for non-linear material behaviour as for linear material behaviour.

The moduli and loss tangent of rubber depend on the frequency, temperature and dynamic strain amplitude. Viscoelastic rubbers exhibit large loss in the frequency range of 1 Hz to 1 kHz and above. For shock absorption and vibration damping a large loss tangent is desirable above 10 Hz and creep should be minimal for time longer than 10 sec.

4.3.3 Torsion and bending pendulum

The torsion pendulum is shown in figure 4.4. The specimen undergoes a free torsional vibration, in which the amplitude of vibration is allowed to decay. The frequency range is of the order 10^{-1} -10 Hz. From the frequency and rate of decay of the vibration, the stiffness and damping constant of the specimen can be calculated. The inertia of the specimen is assumed to be negligible.

Because the torsion pendulum is measuring properties under shear, a bending pendulum has been designed in order to measure tensile properties [16]. Again the stiffness and damping constant are calculated from the rate of decay of a free vibration.

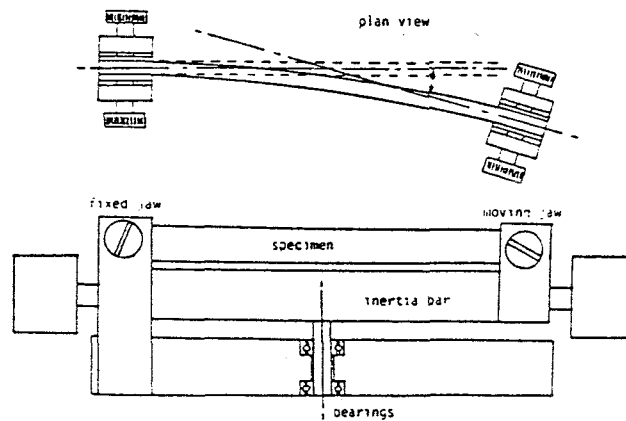


Figure 4.5: Bending pendulum (Worth, 1985)

4.3.4 Resonance measurements

A very popular resonance technique is the vibrating reed or cantilever. The frequency range is of the order 10^3 - 10^4 Hz. A small specimen is clamped at one end in a vibrating head. The amplitude of vibration, i.e. the resonance curve is measured as a function of the frequency, at fixed temperatures. From the equation of motion of the cantilever the resonance frequency f_r can be obtained. The loss tangent is then given by:

$$\tan \delta = \frac{\Delta f}{f_r} \quad (4.9)$$

where Δf is the band width, or the difference between those frequencies for which the amplitude of vibration has $\frac{1}{\sqrt{2}}$ its maximum value [11]

This resonance technique can be made more versatile by attaching to the sample elements of additional inertia and/or elastance with known properties [12].

4.3.5 Wave propagation

The velocity of propagation of waves travelling in viscoelastic materials depends upon the frequency (or frequency spectrum) of the excitation. When the frequency is sufficiently high (10^3 - 10^4 Hz) the waves length of the stress waves is of the same order of magnitude as the sample. Then it is necessary to consider methods employing travelling waves. Either longitudinal or transverse (shear) waves can be propagated.

An alternative to considering the propagation of continuous harmonic vibrations is to consider the propagation of stress pulses of arbitrary shape. Continuous waves can be generated at frequencies up to approximately 10^4 Hz, but pulse propagation

techniques must be used at higher frequencies. The upper limit is approximately 10^7 Hz. [11] [13].

4.3.6 Resilience tests

In the ball rebound resilience test a rigid ball is released at a fixed height h_0 , above a rubber test piece. During the collision, part of the kinetic energy of the ball is dissipated as heat in the rubber. As a result the ball returns to a lower height h_1 and after a second collision to a still lower height h_2 etc. The rebound resilience is defined as:

$$R = \frac{h_{n+1}}{h_n} \quad (4.10)$$

and is related to the loss tangent in the following way:

$$R \approx \exp(-\pi \tan \delta) \quad (4.11)$$

Instead of a free falling rigid ball, most standardized methods attach a striker to a pendulum, which falls from a definite angle and strikes the test piece surface when the pendulum is vertical [14]. This is the so-called pendulum rebound resilience test. The resilience measured from the pendulum rebound tester varies with specimen thickness. As the thickness increases, the contact time increases, while the equivalent impact frequency decreases. This combination results in a higher resilience, but a lower loss tangent [15].

4.3.7 Rotational rheometers

With rotational rheometers (such as the rheometric) the relaxation behaviour in shear and the dynamic moduli can be obtained. A sample of the material is held between two discs, steady rotation of one or both of the discs causes a shear strain in the material [14]. The discs can have different shapes. The most common shapes are cone-and-plate and parallel plate discs. The latter has the disadvantage that it causes non-homogenous strain in the sample, but the advantage that the sample preparation is simpler and that with the same rheometer larger strains can be obtained than with cone-plate discs. Soskey [17] derived a correction term to account for the non-homogeneity of the strain in the parallel disc rheometer.

4.3.8 Dynamic mechanical thermal analysis

Solid polymers and viscoelastic liquids can be characterized by dynamic mechanical thermal analysis (DMTA). Absolute modulus and damping are obtainable from DMTA measurements, provided proper attention is paid to clamping and optimising geometry. DMTA involves applying an oscillating mechanical strain and resolving the stress into real and imaginary components. This procedure essentially detects

all changes in the state of molecular motion as temperature is scanned. Different instruments are commercially available. A short review is given by Wetton [18].

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Chapter 5

Design of rubber dampers

5.1 Introduction

In this chapter the basic principles of design for vibration isolation and damping are outlined. First a short introduction in the theory of vibration isolation is given. In section 5.3 the effects of geometry and mode of loading are considered, and in section 5.4 some practical design details are given.

5.2 Theory of vibration isolation

The function of an isolator is to reduce the magnitude of motion transmitted from a vibrating foundation to the equipment, or to reduce the magnitude of force transmitted from the equipment to its foundation. The nature and degree of vibration isolation is influenced markedly by the characteristics of the damper [1]. The performance is usually given in terms of transmissibility. Transmissibility is defined as the ratio of the transmitted amplitude to the imposed amplitude of vibration or the ratio of the transmitted force to the externally imposed force. It is expressed in decibels and usually based on the consideration of a single degree of freedom spring/mass system. In this simple model shown in figure 5.1(a) K is a stiffness and C is a damping coefficient. K and C are constants and independent of frequency. From the differential equation of motion the transmissibility can be computed and is given for a steady state sinusoidal excitation by [2]:

$$T = \frac{\text{output}}{\text{input}} = \sqrt{\frac{1 + (2ur)^2}{(1 - r^2)^2 + 2(ur)^2}} \quad (5.1)$$

where r is the frequency ratio ($=f/f_n$) and u is the damping ratio ($=C/C_c$), with f_n the natural frequency and C_c the critical damping coefficient given by:

$$f_n = \frac{1}{2\pi} \sqrt{K/M} \quad (5.2)$$

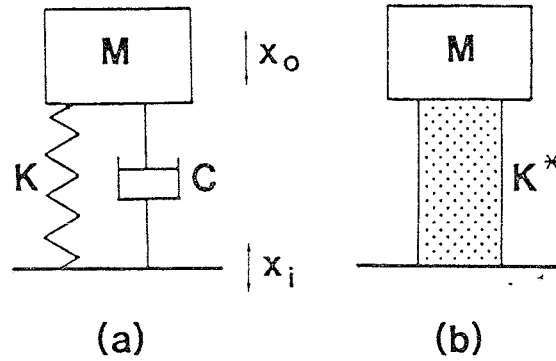


Figure 5.1: Single degree of freedom models for transmissibility; (a) with linear spring and viscous damper; (b) with complex stiffness for elastomers (Harris, Rubber chemistry and technology 62.)

$$C_c = 2\sqrt{KM} \quad (5.3)$$

In figure 5.2 the transmissibility predicted by equation (5.1) is shown. It can be seen that isolation is achieved for $r > \sqrt{2}$. When the damping is increased the peak transmissibility at f_n reduces, but this is at the expense of poorer isolation at higher frequencies.

The above approach illustrates the basic principles of vibration isolation, however it is not applicable to the analysis of elastomers where aswell K as C vary with the frequency. In order to evaluate the transmissibility iterative calculations are required. A more general representation of the dynamic properties of elastomers has been derived by Snowdon [3] and is shown in figure 5.1(b). The dynamic stiffness K^* can be splitted in an in and out of phase component (which both are allowed to depend on the frequency):

$$K^* = K' + iK'' \quad (5.4)$$

The transmissibility is then given by [2]:

$$T = \sqrt{\frac{1 + \tan^2 \delta}{(1 - r^2 K'_n / K')^2 + \tan^2 \delta}} \quad (5.5)$$

where $\tan \delta$ is the loss tangent defined by: $\tan \delta = \frac{K''}{K'}$, and K'_n is the in-phase stiffness at the natural frequency ($r=1$).

From the above relations it may be concluded that an effective antivibration mounting should confer [3]:

- A low natural frequency of vibration- of the order of one-third of the exciting frequency for adequate isolation.

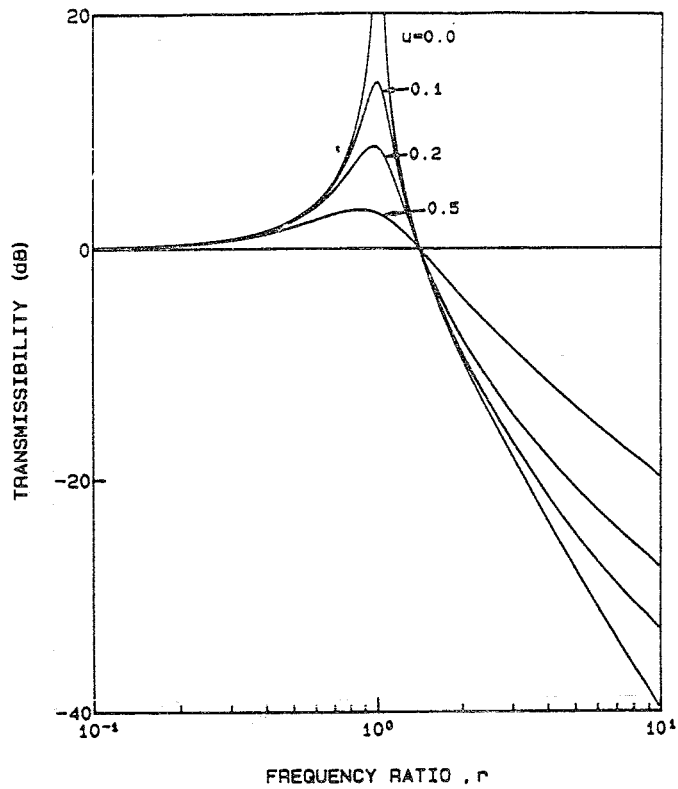


Figure 5.2: Transmissibility for single degree of freedom model with linear spring and viscous damper for various damping ratio's u (Harris, Rubber chemistry and technology 62.)

- A low transmissibility at resonance.
- A transmissibility that decreases rapidly with frequency at frequencies greater than f_n .

It can be shown that the simple linear system is limited in its ability to meet the above requirements. Snowdon has investigated methods of reducing the considerable transmissibility at high frequencies while retaining the low amplitude of vibration at resonance. The method of greatest interest involves mechanically combining a rubber of low damping characteristics in parallel with a high damping rubber in the same mounting.

5.3 Nonlinear geometrical effects

Nonlinearity in the force deflection behaviour of elastomeric components depends on the geometry and mode of loading. The three major modes of deformation of rubber dampers are shear, compression and torsion, the latter being treated as a form of shear. Commonly rubber dampers are used in shear, because large deflections in one direction can be obtained with good stability in other directions and because the characteristics of a shear mounting can be predicted with a greater ease [4]. The latter is due to the essential linear static stress/strain behaviour ($\tau = G\gamma$) up to approximately 80 % strain. Beyond this the introduction of tensile components becomes significant, resulting in a progressive stiffness. Design limits for permanent deformation do not normally exceed 50 % shear strain.

In compression the geometry of the rubber component becomes a governing factor in the derivation of expressions for the force-deformation characteristics. The shape factor S provides a relation between the stiffness of the construction and the elasticity modulus. S is defined as the ratio of one loaded surface to the force-free surface. Shape factor considerations have been used widely in the design of elastomeric components to exploit the ability to have vastly different stiffnesses in different modes of deformation in a single component [5].

For the compression of a solid section the stiffness increases with increasing the deflection. For the compression of a hollow section of material, there is the possibility of buckling of the sidewall to occur under certain conditions resulting in a rapid reduction in stiffness with increasing deflection [6]. As well as the reduction in stiffness that occurs with buckling, there is also an increase in the level of damping. This is a result of the change in mode of deformation that occurs from compression to bending in the cylinder walls [7]. Eventually on further deflection after buckling, material either side of the buckled region comes into contact and there is a final rapid increase in stiffness.

An interesting mounting that utilises the buckling column principle is shown in figure 5.3a. The unit is designed to provide very large deflections under comparatively small loads. In figure 5.3b the load in terms of tension, compression and shear

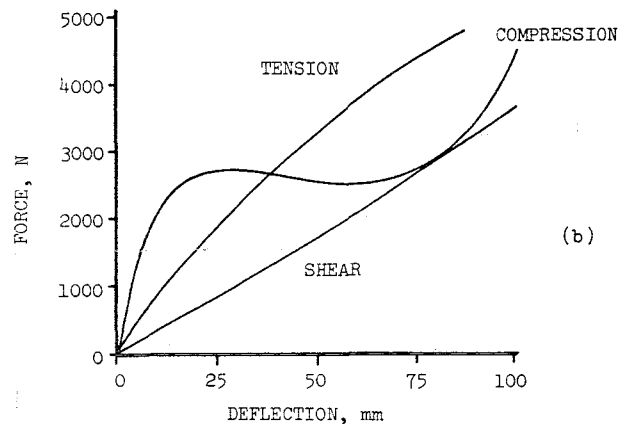
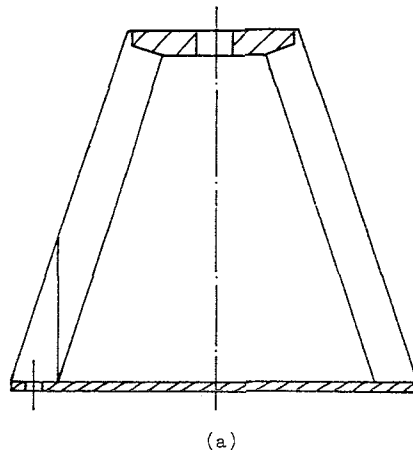


Figure 5.3: (a) Buckling type shock mounting and (b) force deflection characteristics (Freakley and Payne 1978)

as a function of deflection for this type of mounting is shown. The essential feature of this mounting is the large area beneath the stress-strain curve [3]. A significant attraction of this type of geometry is that being hollow, there is an internal cavity available which can be subjected to fluid pressure in the form of either gas or liquid. Varying the internal pressure then provides the basis for controlling the dynamic characteristics of the system [6].

5.4 Some practical design details

Ideally a rubber mounting should be so designed that stresses are uniform throughout the rubber. Probably this can seldom be achieved, but excessive stresses should be avoided. For instance if a rubber block, bonded to metal, is compressed, very high shear stress can develop at the edges, owing to the barreling of the rubber under compression, but this can be mitigated if the edge is radiused instead of meeting the metal at right angles. Sharp corners or edges in the rubber should be always avoided, and this also applies to internal cavities.

Another rather obvious requirement, though sometimes overlooked, is that since rubber is (almost) incompressible, it must be given space to expand in one direction

if compressed in another [8].

Most elastomers exhibit shrinkage. The volume changes approximately 0.047 % per °C or a total reduction of about 6 % from curing temperature to room temperature. Beside this shrinkage other partially compensating volume change effects exists and it is common practice to allow for about 5 % shrink after a rubber has being cured. Where an elastomer is bonded to a metal that cannot move, this small amount of schrinkage may cause a damaged bond, voids or unrelieved tensile stresses in parts of the body [3].

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