# MECHANICS OF RUBBERLIKE SOLIDS

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**Abstract** In this paper we discuss: (i) the large deformation stress-strain response of rubberlike solids based on experimental observations, including both elastic and inelastic behaviour of particle-filled and unfilled rubber, (ii) the mathematical modelling of this behaviour through its phenomenological treatment using elasticity theory and extensions of the theory to account for inelastic responses such as the Mullins effect, stress softening and hysteretic stress-strain cycling, (iii) an introduction to the analysis of the coupling of mechanical and magnetic effects in so-called magneto-sensitive elastomers, which are being used as 'active' components in various applications where the mechanical properties of the material are changed rapidly when a suitable magnetic field is applied.

**Keywords:** Rubber elasticity, rubber inelasticity, Mullins effect, stress softening, large deformations, magneto-sensitive elastomers, magnetoelasticity

## 1. Introduction

In this paper we provide an account of the mechanics of rubberlike solids, including both elastic and inelastic behaviours. Additionally, we discuss the influence of magnetic fields on magneto-sensitive elastomers. Because of limited space the account is largely descriptive but includes detailed references to the literature to enable the reader to follow up both the experimental and theoretical aspects of the subject. We begin, in Sec. 2, with an overview of the large deformation stress-strain response of rubberlike solids based on experimental observations. First, experimental results that characterize the *elastic* behaviour of rubber are described. This is followed by illustrations of how the behaviour departs from the purely elastic. When subjected to cyclic loading many elastomers exhibit a *stress softening* phenomenon widely known as the *Mullins effect* (Mullins [32]). The Mullins effect is closely related to the fatigue of elastomeric parts used in engineering applications. A detailed qualitative and quantitative understanding of the Mullins effect is thus a necessary step towards the scientific evaluation of the *life* of a rubber product.

Next, in Sec. 3, we discuss briefly some theoretical approaches to the modelling of the inelastic behaviour of rubberlike solids.

The final section of the paper, Sec. 4, is motivated by recent renewed interest in the subject of electromagnetic continua generated by the development of so-called ‘smart’ materials. These are used, for example, in devices for controlling the damping characteristics of vibration absorbers. In particular, we are concerned here with elastomers containing a distribution of ferrous particles embedded within their bulk so that they respond to the application of magnetic fields by changing significantly their stress-strain behaviour. We outline recent theoretical work concerning the coupling of mechanical and magnetic effects in these so-called *magneto-sensitive (MS) elastomers*.

## 2. Description of Experimental Results

Figure 1(a) shows the characteristic sigmoid-shaped curve associated with the stress-stretch behaviour of rubber. Specifically, this is for loading of unfilled vulcanized natural rubber in simple tension based on data of Treloar [47], which correspond to the solid circles. Similar behaviour is found for other standard experimental tests such as pure shear and equibiaxial tension (see, for example, Treloar [48] and Ogden [36–38] and the recent collection of papers [49]). In pure shear and equibiaxial tension, however, the largest stretch achievable is generally less than for simple tension. The general characteristics of the results shown in Fig. 1(a) are also evident in many synthetic rubbers, but in some cases the largest stretch obtained can be much larger than for natural rubber (as much as 10–15 in simple tension for example).

Figure 1(a) shows only the loading curves. In general, the loading curve is not re-traced exactly on unloading but for natural rubber (and for many synthetic rubbers) there is only a small stress softening or hysteresis effect if the strains are not too large, and the materials are therefore modelled as perfectly elastic. Generally, the tests are conducted at a relatively low strain rate so that the curves can be regarded as corresponding essentially to quasi-static behaviour.

A different picture is evident, however, when unloading is accounted for. Figure 1(b) shows the typical simple tension behaviour of a specimen of natural rubber filled by 1 % by volume of carbon black particles. From its virgin state the material is loaded to a stretch $\lambda = 3$, unloaded, then loaded again to the same stretch in several loading-unloading cycles until

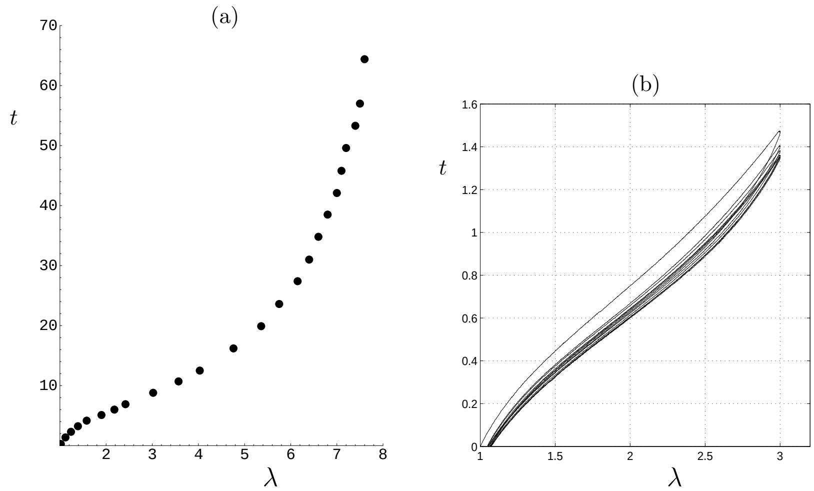


*Figure 1.* Simple tension data, with nominal stress $t$ vs stretch $\lambda$: (a) data of Treloar [47] – units of $t$, kg cm$^{-2}$; (b) from Dorfmann and Ogden [11] – units of $t$, N mm$^{-2}$.

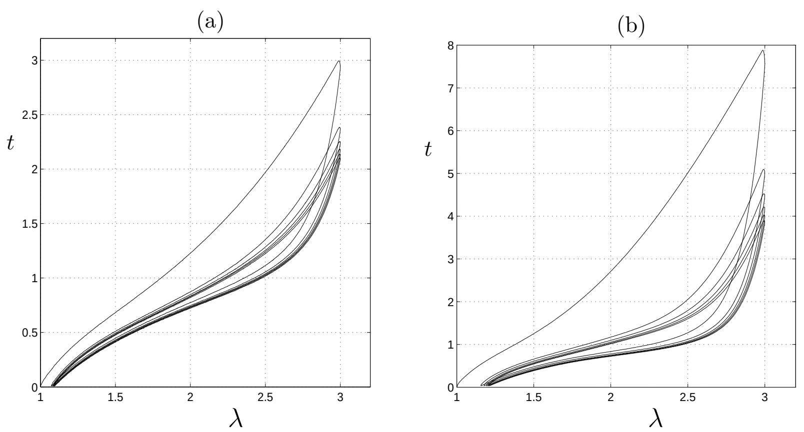


*Figure 2.* Cycles of nominal stress $t$ (units N mm$^{-2}$) vs stretch $\lambda$: rubber compound with (a) 20% and (b) 60% by volume of carbon black filler.

a repeatable pattern is established (typically after 5 or 6 cycles). Since the filler content is small the behaviour approximates that of an unfilled rubber. In this case there is little difference between the loading and unloading curves, particularly after the initial loading.

Figure 2 shows the corresponding picture for filled natural rubber with (a) 20 % and (b) 60 % by volume of carbon black filler. Here, the residual strain, which is also evident in Fig. 1(b), is much more noticeable. Moreover, there is a very marked stress softening, particularly on the first loading-unloading cycle, i.e. the stress on unloading is less than that on loading at the same level of stretch. This effect, associated with

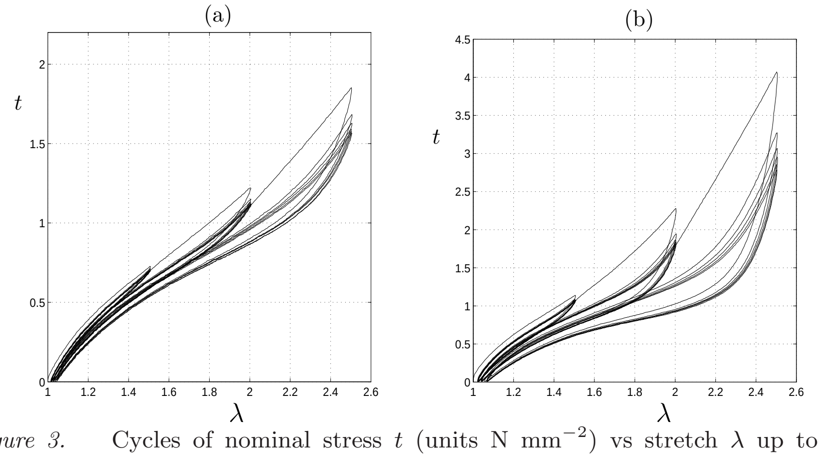


*Figure 3.* Cycles of nominal stress $t$ (units N mm$^{-2}$) vs stretch $\lambda$ up to $\lambda = 1.5, 2, 2.5$: rubber compound with (a) 20 % and (b) 60 % by volume of carbon black filler.

deformation from the virgin state and known as the *Mullins effect*, increases as the percentage of filler increases. The first cycle essentially provides a pre-conditioning of the material and was examined in detail by Mullins [32, 33] and Mullins and Tobin [34] and in several other papers. However, the effect had apparently been recorded in the literature much earlier by Bouasse and Carrière [3]. Figure 3 shows data for the same two materials as in Fig. 2 but with the stretch cycled first up $\lambda = 1.5$, then to $\lambda = 2$ and finally to $\lambda = 2.5$. These curves show clearly how the pre-conditioning, stress softening and residual strain effects depend on the maximum stretch achieved and on the proportion of filler.

The degree of stress softening and magnitude of the residual strain depend very much on the maximum stretch achieved and on the particular material under consideration. For example, for a liquid silicone rubber filled with silica particles, there is a very significant stress softening, but the residual strain is very small [35]. Moreover, the reloading curves are very close to the unloading curves. This pattern is very close to what may be described as the *idealized Mullins effect*, for which residual strain is absent and the reloading curves re-trace the unloading curves exactly. It is this idealized description that has formed the basis of most of the modelling in the literature.

As indicated above, the Mullins effect is associated with deformation from the virgin state of the material. Once the material has been deformed there is then (at least in the idealized situation) no further stress softening generated provided the material is not strained beyond the maximum value of the stretch already achieved. This observation is the basis for pre-conditioning the material: in pre-conditioning the material is strained to a level beyond that expected to be achieved in service conditions so that the Mullins effect is no longer operative. This pre-conditioning is evident in Figs. 1(b) and 2.

For further discussion of many of the aspects of material behaviour considered briefly here we refer to the three volumes of proceedings from the series of conferences on constitutive models for rubber [8, 2, 6] and the volume [49]. These include the effects of time dependence (creep, relaxation and recovery, for example), frequency dependence, hysteresis and viscoelasticity.

## 3. Theoretical Approaches

As far as the *elasticity* of rubber is concerned there are some fundamental assumptions that are adopted in the phenomenological theory: the material is (a) hyperelastic, (b) isotropic, and (c) incompressible. *Hyperelasticity* means that the properties of the material are described in terms of a strain-energy function. *Isotropy* (relative to a stress-free configuration) is a very good approximation in most circumstances and is almost invariably used by practitioners. *Incompressibility* is an idealization, justified by the fact that the shear modulus of the material is very much smaller than the bulk modulus (typically the ratio is of order $10^{-4}$) and volume changes can be neglected except in extreme situations where the hydrostatic stress is very large. For the most part isotropy and incompressibility are assumed in practical applications.

The strain energy is a function of the deformation gradient $\mathbf{F}$ (relative to some fixed reference configuration), and is written $W(\mathbf{F})$ per unit volume. This theory is well established and we omit details here. Suitable references are the review articles [37, 38, 4] and the volume [49]. Here we focus on models for the Mullins effect.

Many phenomenological theories aimed at modelling the Mullins effect have been proposed in the literature. Some of these are based on the two-phase micro-structural model introduced by Mullins and Tobin [34] and developed more recently by, for example, Johnson and Beatty [25, 26]. Another class of models is based on the introduction of a damage (or stress-softening) variable to describe the internal damage in the bulk material associated with the Mullins effect (see, for example, Gurtin and Francis [18], De Souza Neto et al. [7], Beatty and Krishnaswamy [1] and Zúñiga and Beatty [50] and references cited therein). Ogden and Roxburgh [42, 43] have developed a model based on a theory of *pseudo-elasticity* in which a pseudo-energy function is introduced as in standard nonlinear elasticity theory except that an additional scalar variable (the damage variable) is incorporated. All of these models share the common feature that the virgin material response is determined by a standard strain-energy function or a stress constitutive function for a perfectly elastic isotropic material. A connection between this theory and that of materials with multiple natural configurations due to Rajagopal and Wineman [44] has also been noted [20].

A model of the Mullins effect based on micro-mechanical continuum damage theory was proposed by Govindjee and Simo [17]. The constitutive equation for this model involves the integration of the inverse Langevin function over a history-dependent domain in the phase space of the rubber network. Recently Marckmann et al. [30] have proposed another model using the inverse Langevin function in the description of network alteration associated with the Mullins effect. The non-Gaussian statistics allow one to take into account the limiting stretch of the chains composing the polymeric network. In the Marckmann et al. [30] model the Mullins effect is explained by reference to an alteration of the average chain length driven by the previous maximum stretch in a given deformation. The network alteration idea has been investigated recently by Horgan et al [20] on the basis of the phenomenological model proposed by Gent [16] for rubberlike elasticity. This is a very simple strain-energy that accounts for the limiting chain extensibility of rubberlike polymers (for further discussion we refer to Horgan and Saccomandi [21–23]). The use of the Gent model allows one to bypass the computational problems associated with the inverse Langevin function and to provide a simpler and deeper interpretation of the network alteration phenomenon. Further references are contained in the paper by Horgan et al. [20].

An important feature of the effect of stress softening and any associated residual strains is the change in material symmetry generated. This has received very little attention in the literature thus far, although there is a recent discussion by Horgan et al. [20]. An extensive review of different aspects of rubber mechanics and thermomechanics, from the molecular to the phenomenological level, is provided in the volume edited by Saccomandi and Ogden [49], which contains many pointers to the literature.

In order to capture the essence of the Mullins effect with a relatively simple theory that requires only a slight modification of the theory of elasticity discussed so far, we use the theory of *pseudo-elasticity* based on the papers by Ogden and Roxburgh [42, 43] and Ogden [41]. We refer to these papers for full details of the theoretical development since only a brief summary is provided below. We consider the deformation of a continuous body which in its initial (virgin) stress-free configuration occupies the region $\mathcal{B}_r$ and in the deformed configuration $\mathcal{B}$. The deformation gradient relating $\mathcal{B}$ to $\mathcal{B}_r$ is again denoted by $\mathbf{F}$.

In *pseudo-elasticity* the strain-energy function $W(\mathbf{F})$ of elasticity theory is modified by incorporating an additional variable, here denoted $\eta$, into the function and we write $W = W(\mathbf{F}, \eta)$. The variable $\eta$ is referred to as a *damage variable* or *softening variable*. In the simplest case it is taken to be a scalar quantity, but more generally it may be a tensor variable, for example. It provides a means of changing the form of the energy function during the deformation process and hence changing the description of the material properties. In a deformation process in which $\eta$ changes, the overall response of the material is not elastic and we refer to $W(\mathbf{F}, \eta)$ as a *pseudo-energy function*. The variable $\eta$ may be active or inactive and may be switched from active to inactive (or conversely) according to some suitable criterion, thereby inducing a change in the material properties. This change may be either continuous or discontinuous.

A suitable means of determining the dependence of $\eta$ on $\mathbf{F}$ is the equation

$$\frac{\partial W}{\partial \eta}(\mathbf{F}, \eta) = 0. \tag{1}$$

For an incompressible material the nominal stress tensor then has the form

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{F}, \eta) - p\mathbf{F}^{-1}, \quad \det \mathbf{F} = 1, \tag{2}$$

where the right-hand side is evaluated for $\eta$ given implicitly by 1. The nominal stress is given by 2 whether $\eta$ is active or inactive. In the latter case $\eta \equiv 1$.

We may regard equation 1 as a field equation, which, in the absence of body forces, is coupled with the equilibrium equation in the form

$$\mathrm{Div}\,\mathbf{S} = \mathbf{0} \quad \text{in } \mathcal{B}_r, \tag{3}$$

where Div denotes the divergence operator in $\mathcal{B}_r$.

There is considerable flexibility in the choice of specific models, in particular the dependence of $W$ on $\eta$, subject to appropriate objectivity restrictions. A criterion for switching $\eta$ on or off is also needed. Such considerations depend on the application in question. For the stress softening associated with the Mullins effect, for example, $\eta$ is taken to be inactive during loading and to switch on during unloading (with loading and unloading being defined relative to the energy expended on a loading path). Moreover, the material is assumed to be isotropic. The theory gives a good fit to data, as demonstrated in the original paper by Ogden and Roxburgh [42], and has been extended to allow for partial unloading-reloading and residual strains [9, 11]. We refer to the above cited papers for further details and additional references.

## 4. Magneto–Sensitive Elastomers

The equations for a continuum deforming in the presence of an electromagnetic field are well established, as exemplified by the work of Hutter and van de Ven [24] and Maugin [31]. Here we are concerned with the static situation for materials that respond to a magnetic field. Specifically, such materials are elastomers with a distribution of ferrous particles embedded within their bulk. Background details of the theory of magnetoelasticity can be found in the work of Brown [5], while the book by Kovetz [29] contains a readable account of some aspects of the theory. In Kovetz the magnetic induction vector $\mathbf{B}$ was taken as the basic variable, which was also the case with the work of the present authors (Dorfmann and Ogden [10, 12]). Alternative formulations based on the use of the magnetic field vector $\mathbf{H}$ or the magnetization vector $\mathbf{M}$ have been discussed by Steigmann [46], while Kankanala and Triantafyllidis [28] base their recent analysis on use of $\mathbf{M}$ as an independent variable. A treatment of universal relations for magnetoelastic solids is contained in a paper by Dorfmann et al. [15].

A refined version of the theory that enables boundary-value problems to be formulated in a very simple form has been provided recently by Dorfmann and Ogden [13] (see also [14]). References to the (limited) literature are given in these papers. The theory is applied to some simple prototype boundary-value problems of practical interest, notably the axial shear and combined extension and torsion of MS material contained within a circular cylindrical tube in the presence of either an axial magnetic field (uniform) or an azimuthal field. The influence of the magnetic field strength on the various shear stiffnesses of the material has been evaluated. Data for the considered material is rather limited, but the paper by Jolly et al. [27], for example, shows how the shear modulus of the material stiffens with the application of a magnetic field and how this depends on the volume fraction of magnetic filler particles.

We consider a magnetoelastic body that is initially in an unstressed configuration, not subject to mechanical loads or magnetic fields. Let the region in three-dimensional Euclidean space occupied by the body in this configuration be denoted $\mathcal{B}_r$. Suppose next that the material is subject to a magnetic field, denoted $\mathbf{H}$, and an associated magnetic induction vector $\mathbf{B}$ and magnetization vector $\mathbf{M}$, together with mechanical boundary tractions, so that the deformation gradient is $\mathbf{F}$.

These (Eulerian) vector fields satisfy the standard relation and the Maxwell field equations given by

$$\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M}), \quad \mathrm{div}\mathbf{B} = 0, \quad \mathrm{curl}\,\mathbf{H} = \mathbf{0}, \tag{4}$$

where $\mu_0$ is the vacuum permeability.

In respect of the reference configuration $\mathcal{B}_r$ the Lagrangian counterparts of $\mathbf{B}$ and $\mathbf{H}$ are denoted $\mathbf{B}_l$ and $\mathbf{H}_l$, respectively, and (for an incompressible material) they are related to $\mathbf{B}$ and $\mathbf{H}$ by

$$\mathbf{B} = \mathbf{F}\mathbf{B}_l, \quad \mathbf{H} = \mathbf{F}^{-T}\mathbf{H}_l, \tag{5}$$

respectively (see, for example, [31, 12, 46]). Standard identities ensure that the pair of Eq. $(4)_{2,3}$ is entirely equivalent to the pair

$$\mathrm{Div}\mathbf{B}_l = 0, \quad \mathrm{Curl}\,\mathbf{H}_l = \mathbf{0}, \tag{6}$$

provided the deformation is suitably regular. Similarly to $\mathbf{H}_l$, a Lagrangian form of $\mathbf{M}$, denoted $\mathbf{M}_l$, is defined by $\mathbf{M}_l = \mathbf{F}^T\mathbf{M}$.

The equilibrium equation may be written in the equivalent forms

$$\mathrm{div}\tau = \mathbf{0}, \quad \mathrm{Div}\mathbf{T} = \mathbf{0}, \tag{7}$$

in the absence of mechanical body forces, where $\boldsymbol{\tau}$ is the total stress tensor and $\mathbf{T}$ is the total nominal stress tensor related to $\boldsymbol{\tau}$ by $\mathbf{T} = J\mathbf{F}^{-1}\boldsymbol{\tau}$. Balance of angular momentum requires that $\boldsymbol{\tau}$ is symmetric.

The constitutive law for a magnetoelastic solid may be expressed in various different but equivalent forms. For an incompressible magnetoelastic solid, for example, a simple description of the constitutive properties of the material is provided through an amended energy function, denoted $\Omega$ and defined per unit volume, given by

$$\Omega = \rho\Phi + \frac{1}{2}\mu_0^{-1}\mathbf{B}_l \cdot (\mathbf{c}\mathbf{B}_l), \tag{8}$$

where $\rho$ is the mass density, $\mathbf{c} = \mathbf{F}^T\mathbf{F}$ is the right Cauchy-Green tensor and $\Phi$ is the free energy of the material (per unit mass). With objectivity invoked, we take $\Omega$ to depend on $\mathbf{B}_l$ and $\mathbf{c}$ as independent variables. The total stress $\mathbf{T}$ and the magnetic field $\mathbf{H}_l$ are then given by

$$\mathbf{T} = \frac{\partial\Omega}{\partial\mathbf{F}} - p\mathbf{F}^{-1}, \quad \mathbf{H}_l = \frac{\partial\Omega}{\partial\mathbf{B}_l}, \tag{9}$$

where $p$ is a Lagrange multiplier associated with the incompressibility constraint. Corresponding equations may also be given in which the independent magnetic variable $\mathbf{B}_l$ is replaced by $\mathbf{H}_l$ [13].

Material symmetry has been discussed in detail in the papers cited above, and, in particular, constitutive laws specified in various forms for *isotropic* magnetoelastic solids. The material symmetry considerations are similar to those that arise for a transversely isotropic elastic material for which there is a preferred direction in the reference configuration analogous to $\mathbf{B}_l$. For relevant discussion of material symmetry for anisotropic elastic solids we refer to Spencer [45], Holzapfel [19] and Ogden [40], for example. For discussion of material symmetry in the magnetoelastic context we refer to Steigmann [46].

Important differences between the formulations based on use of $\mathbf{B}_l$ and $\mathbf{H}_l$ in respect of their application to particular boundary-value problems have been discussed in Dorfmann and Ogden [13, 14].

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