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On a possible methodology for identifying the initiation of damage of a class of polymeric materials

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In this paper, we provide a possible methodology for identifying the initiation of damage in a class of polymeric solids. Unlike most approaches to damage that introduce a damage parameter, which might be a scalar, vector or tensor, that depends on the stress or strain (that requires knowledge of an appropriate reference configuration in which the body was stress free and/or without any strain), we exploit knowledge of the fact that damage is invariably a consequence of the inhomogeneity of the body that makes the body locally 'weak' and the fact that the material properties of a body invariably depend on the density, among other variables that can be defined in the current configuration, of the body. This allows us to use density, for a class of polymeric materials, as a means to identify incipient damage in the body. The calculations that are carried out for the biaxial stretch of an inhomogeneous multi-network polymeric solid bears out the appropriateness of the thesis that the density of the body can be used to forecast the occurrence of damage, with the predictions of the theory agreeing well with experimental results. The study also suggests a meaningful damage criterion for the class of bodies being considered.

1. Introduction

What constitutes damage of a body, and modelling of the initiation and progression of damage in a body, has been one of the most important questions confronting a scientist interested in understanding

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the underlying mechanisms that lead to the deterioration of a body with respect to its desired performance characteristics. A simple identification within the context of mechanical issues of what constitutes damage would be the degradation and the deterioration in the performance of the body during a process that it is subject to, that is, the inability of the body to continue performing in a certain expected manner. However, it is possible that while one aspect of the body's behaviour is worsening, its response with regard to another aspect of its response characteristics might be improving. For instance, while the load carrying capacity might decrease, the body's electrical or thermal conductivity could possibly increase. Thus, in talking about a body undergoing damage, we ought to clearly delineate which aspect of its behaviour or property is deteriorating. The kind of damage that one is interested in might also depend on the class of materials under consideration and thus the criterion for defining damage might change considerably from one class of materials to another. Polymers and metals need not suffer the same type of damage as the physical mechanisms underlying their response characteristics are different. For instance, while metals usually expand when heated large classes of polymers contract. Thus, one cannot treat a very large class of materials under the same umbrella as far as damage is concerned. This paper is concerned with a certain class of damage suffered by polymers owing to degradation in their load carrying capacity that is a consequence of network scission.

Even within the context of a specific body, damage/failure criteria can depend on the class of processes that a body is subject to. Thus, for instance, while a decrease in density and the attendant inability of a body to withstand loads might be appropriate when considering the tension of certain bodies, such a criterion based on density might be inappropriate in the same body undergoing compression wherein the density might increase but the body might yet fail owing to a totally different mode to failure. Thus, a single damage criterion cannot be used even in the case of a specific class of bodies. It is also important to bear in mind that a criterion based on changes in density will not be appropriate if one is concerned with small deformations as density changes during such deformations will be small. Here, we are primarily interested in problems wherein damage occurs owing to a decrease in the density of the polymer which is undergoing sufficiently large deformations that allows for sufficiently large changes in the density and volume of the body under consideration.

Damage is often times modelled by assuming rather ad hoc internal variables, which could be scalar, vector or tensor, which often times do not necessarily have meaningful physical significance. The initiation and the progression of the damage are also invariably prescribed in an ad hoc manner in terms of these damage variables. Parameters that qualify and quantify damage, especially within the context of a mechanical perspective, depend on the stress, strain or other kinematical variables through their invariants and herein lies a serious shortcoming. Given a body in a particular configuration, we do not know if it is stress free or if it has undergone a permanent set, that is whether it is strained with respect to some other unstrained or unstressed state. That is, what is perceived by an experimentalist as the initial configuration of a body could be in a stressed or strained state with respect to some previous configuration. Thus, if a damage parameter depends on the stress or strain, the fact that a body in question might already be in a stressed or strained state makes such a parameter an useless measure. On the other hand, if damage were to depend on quantities (properties of the body) that are unambiguously determined from information concerning its value purely in the current state, then such a measure of damage would be a meaningful measure. In this study, we provide a possible methodology to identify the onset of damage for a class of polymeric materials by assuming that damage depends on the current density of the body. However, before discussing this methodology, we shall discuss another aspect concerning damage that is often given the short shrift in practically all studies devoted to damage.

A rather strange circumstance with regard to most of the models currently in vogue is that they assume that the body is homogeneous and the causative agent for damage being the stress or the strain. This not true to the physics of the situation. *Damage invariably occurs owing to local inhomogeneities in a body*. The body is weaker at certain locations owing to some defect that exists at the microscopic level. Thus, the proper study of damage necessarily requires one to model the body under consideration as an inhomogeneous body. This is the point of view adopted in this paper. Once one adopts such a perspective, the study of damage in bodies, at least for a class bodies, becomes quite simple.

It would be appropriate at this juncture to mention that there have been several studies of multi-network polymers wherein one allows for scission and reformation of network junctions (see [1–8]). One could view scission as damage and the formation of new networks as a possible healing process. These studies, however, concern the mixture of two networks, each with different material properties that can allow for a deterioration in load carrying capacity, but the damage is not due to the density being lower in a small neighbourhood belonging to the body, namely damage as a consequence of inhomogeneity. Of course, one could generalize those multi-network approaches to include the possibility of local density variations. Using the multi-network approach and scission and reformation as the basis for damage, Rajagopal *et al.* [9] and Puglisi & Saccomandi [10] studied damage in rubber-like materials. Another way in which damage has been modelled that is similar in spirit to multi-network theory is when one considers the body undergoing damage as comprising two ‘phases’: a soft phase and a hard phase. Using such an approach, Tommasi *et al.* [11] carried out an interesting study of the damage and self-healing of spider silks. They took into account the breaking of the hydrogen bonds in the material that leads to the softening of the material. However, such an approach yet requires one to specify an activation threshold and a transition threshold. That is, one needs to specify the strain at which the hydrogen bond breaks and this is an ad hoc specification as assuming that the breaking occurs at a different strain will lead to different results. In fact, a similar approach of a mixture of soft and hard phases was used much earlier by Kratochvil *et al.* [12] to study the elastic–plastic response of materials where one could think of the onset of plasticity as damage but once again one has to specify when the plastic response is to be activated by a specification of an activation strain. The important aspect that one has to bear in mind is that the initiation of damage in all the above-mentioned approaches depends on the specification of a strain that requires one to know a configuration in which the body was strain free and from which an absolute measure of strain can be based. Unfortunately, one is usually presented with a specific specimen in a specific configuration and one does not know the history of the body and whether it has already undergone a permanent set. Unlike these studies, the present one however does not depend on the specification of knowing a stress free or strain free state from which thresholds for the strain or stress can be given, the damage being merely a consequence of the inhomogeneity of the body and properties of the body in the current state.

In this paper, we shall be concerned with damage that occurs in polymers owing to the scission of the network junctions with attendant changes in the density of the material. Such a situation can be observed in the stretching of polymeric bodies where locally the density falls to such an extent that the body ultimately bursts. In fact, in many situations, such as the inflation of a balloon, one can actually see a local region that undergoes a significant change in the density and ultimately bursts. Yet another example of the precipitous fall in the local density followed by a significant increase in the local volume that leads to a catastrophic burst that is observed in aneurysms. Similarly, there are a variety of problems involving structures where there is a dramatic fall in the density, triggering the onset of the collapse of the structure.

Phenomenon such as scission of the networks is entropy producing and dissipative.[1] However, as long as the dissipation associated with the scission is insignificant, we can ignore it, and model such bodies as elastic. However, when sufficient number of the network junctions break and the body is ‘damaged’, the body invariably ceases to be ‘elastic’ in its original sense. The body might continue to respond from a new natural state, the damaged state, as an elastic body, in

[1]Most rubber-like materials and polymers are not elastic. While under certain circumstances they can be approximated as elastic bodies, there is an erroneous opinion that filled rubber and rubber-like materials can be described, while they undergo large deformation, as elastic bodies. This has led to numerous incorrect attempts to describe a phenomenon such as the Mullins effect within the context of elasticity. Much of the experimental data on rubber-like materials is restricted to merely loading and one ought not to construe that such bodies are elastic as it only by unloading the body one recognizes the extent of inelasticity of the body.

an approximate sense, until further significant damage occurs. It is necessary to recognize that a body that is undergoing damage cannot be modelled as the same elastic body prior to the damage, after it has been damaged.

In fact, most constitutive relations are valid only for a small range of processes that a body is subject to. In a certain range, the body could respond in a manner that can be described by a constitutive relation for elastic bodies that however on further deformation beyond the range wherein it is elastic could behave inelastically. That is, there is a domain of process classes where a particular model is valid. This point cannot be overemphasized.

The thesis of the paper is that damage in a large class of materials can be described in an elegant and simple manner without resorting to unnecessary internal variables such as damage parameters. If the material properties depend on the density and if they change in such a manner that the load carrying capacity diminishes, so that it is unable to perform as intended, we can consider it as being damaged. The damage criterion which this study suggests, and which seems eminently reasonable from a physical standpoint is the assignment of the initiation of damage when the derivative of the norm of the current value of the stress with respect to the density, at the point of interest starts decreasing. Interestingly, this criterion is in keeping with experimental results as explained in detail later. In this study, we find that a slight increase in the far-field nominal stretch leads to a dramatic increase in the determinant of the deformation gradient at the location where the density is initially lower to start with, which in virtue of the conservation of mass implies a significant change in the density that we find leads to a steep drop in the load carrying capacity. Once the body is damaged it will not respond along the same response curve (elastically). But we do carry our simulation after damage as though the body yet responds elastically as we do not carry any unloading of the body. We plan to develop a more complete ala plasticiy. We specifically consider the density of the body in the reference configuration to be a function of the particles in the reference configuration, and we allow it to vary so that in a very small region within the body, say a circular domain whose radius is one-thousandth of a characteristic length associated with the body, the density is approximately 50% or 30% of the far-field density.

We consider two classes of models, first a generalization of a compressible neo-Hookean model and second a generalization of a model owing to Gent [13]. We note in both classes of models, the damage criterion makes good sense, and the values of stretch when damage occurs agrees well with the experiments of Gent [13]. We then subject the body, in our case, a layer, to biaxial stretch. We note that as the body is deformed, the current density of the body decreases, disproportionately larger in the small region that was initially less dense, than the rest of the body, with a tremendous increase in the local volume (that is the determinant of the deformation gradient) incipient to the bursting at the location.

2. Preliminaries

Let $\mathbf{x}$ denote the current position of a particle in the deformed body which is at $\mathbf{X}$ in a reference configuration, which is the configuration at which the body was given to the observer. Let $\mathbf{x} = \boldsymbol{\chi}(\mathbf{X}, t)$ denote the motion[2] of a particle and let us denote the displacement by

$$\mathbf{u} := \mathbf{x} - \mathbf{X}. \tag{2.1}$$

The displacement gradients $\partial\mathbf{u}/\partial\mathbf{X}$ and $\partial\mathbf{u}/\partial\mathbf{x}$ are given by

$$\frac{\partial \mathbf{u}}{\partial \mathbf{X}} = \nabla_{\mathbf{X}}\mathbf{u} = \mathbf{F} - \mathbf{I} \tag{2.2}$$

and

$$\frac{\partial \mathbf{u}}{\partial \mathbf{x}} = \nabla_{\mathbf{x}}\mathbf{u} = \mathbf{I} - \mathbf{F}^{-1}, \tag{2.3}$$

[2]The motion of particle depends on the choice of the reference configuration and we represent it as $\mathbf{x} = \boldsymbol{\chi}_{\kappa_R}(\mathbf{X}, t)$. We shall however dispense with the suffix κ_R as it is rather cumbersome. In the definition of a Cauchy elastic body given through equation (2.8) that follows, the function as well as all the arguments should have suffix κ_R.

where $\mathbf{F}$ is the deformation gradient defined through

$$\mathbf{F} = \frac{\partial \boldsymbol{\chi}}{\partial \mathbf{X}}. \tag{2.4}$$

The Cauchy–Green tensors[3] $\mathbf{B}$ and $\mathbf{C}$ are defined through

$$\mathbf{B} := \mathbf{F}\mathbf{F}^{\mathrm{T}} \quad \text{and} \quad \mathbf{C} := \mathbf{F}^{\mathrm{T}}\mathbf{F}. \tag{2.5}$$

The unimodular tensors $\bar{\bar{\mathbf{C}}}$ and $\bar{\bar{\mathbf{B}}}$ are defined through

$$\bar{\bar{\mathbf{C}}} := \det \mathbf{F}^{-2/3}\mathbf{C} \quad \text{and} \quad \bar{\bar{\mathbf{B}}} := \det \mathbf{F}^{-2/3}\mathbf{B}. \tag{2.6}$$

and their corresponding first principal invariants of are $I_1 = \mathrm{tr}\,\bar{\bar{\mathbf{C}}} = \mathrm{tr}\,\bar{\bar{\mathbf{B}}}$. The Green–St. Venant strain $\mathbf{E}$ and the Almansi–Hamel strain $\mathbf{e}$ are defined through

$$\mathbf{E} := \tfrac{1}{2}(\mathbf{C} - \mathbf{I}) \quad \text{and} \quad \mathbf{e} := \tfrac{1}{2}(\mathbf{I} - \mathbf{B}^{-1}). \tag{2.7}$$

A body is said to be Cauchy elastic ([15,16]) if the Cauchy stress $\mathbf{T}$ in the material can be expressed as

$$\mathbf{T} = \mathbf{T}(\rho, \mathbf{F}, \mathbf{X}). \tag{2.8}$$

We shall denote by the $\|\mathbf{T}\|$ the usual Frobenius trace norm for tensors. We shall later use the variation of this norm with the density to introduce the notion of damage. The most general compressible homogeneous isotropic Cauchy elastic body (see [15–17]) is defined through

$$\mathbf{T} = \delta_1\mathbf{I} + \delta_2\mathbf{B} + \delta_3\mathbf{B}^2, \tag{2.9}$$

where $\delta_i, i = 1, 2, 3$ depend on $\mathbf{X}$,ρ,tr$\mathbf{B}$,tr$\mathbf{B}^2$ and tr$\mathbf{B}^3$.

One of the bodies that we consider is a generalization of the compressible neo-Hookean body that is a special subcase of (2.9) and takes the following form

$$\mathbf{T} = K_0(\mathbf{X})\frac{(\det \mathbf{F} - 1)}{(\det \mathbf{F})^3}\mathbf{I} + \mu_0(\mathbf{X})\frac{1}{\det \mathbf{F}}\mathrm{dev}\,\bar{\bar{\mathbf{B}}}, \tag{2.10}$$

where $\mu_0(\mathbf{X})/\det \mathbf{F}$ and $K_0(\mathbf{X})/\det \mathbf{F}^3$ are the shear and bulk moduli,[4] which decrease with increase in $\det \mathbf{F}$.

Before solving the equations, we proceed by choosing an appropriate length scale to non-dimensionalize the constitutive equation. The main reason for non-dimensionalizing is to study in a systematic fashion the effect of the various parameters that affect the response of the body. The equations are non-dimensionalized using a characteristic length scale L (the length of the layer under consideration) and modulus K_1: $u = L\bar{u}$, $v = L\bar{v}$, $w = L\bar{w}$ and $\mathbf{T} = K_1\bar{\mathbf{T}}$. The final non-dimensionalized equation is given by

$$\bar{\mathbf{T}} = \frac{K_0(\bar{\mathbf{X}})}{K_1}\frac{(\det \bar{\mathbf{F}} - 1)}{(\det \bar{\mathbf{F}})^3}\mathbf{I} + \frac{\mu_0(\bar{\mathbf{X}})}{K_1}\frac{1}{\det \bar{\mathbf{F}}}\,\mathrm{dev}\,\bar{\mathbf{B}}. \tag{2.11}$$

The material parameters used to describe the above model are given in table 1.

The second model that we consider is a generalization of a model due to Gent, which is a Green elastic body. Truesdell [14] introduces a Green-elastic material [18,19] in the following manner: the response of a body is perfectly elastic if for coordinates X^α, referred to a certain natural state, there

[3]Various measures of deformation have been introduced, the interested reader can find a discussion of the same in Truesdell [14].

[4]By operating the trace operator on both sides of equation (2.10), it is easy to see that $\text{Mean normal stress}/\det \mathbf{F} - 1 = K/(\det \mathbf{F})^3$.

Table 1. Material parameters.

parameters	functions
ρ_0	smoothened Heaviside function (figure 4)
μ_0	$10^2\rho_0$
K_0	$100(1-\exp(-8\rho_0))\mu_0$
K_1	10^7

exists a stored energy Σ of the form

$$\Sigma = \Sigma(X^\alpha, G_{\alpha\beta}, g_{ij}, \rho, X^\alpha_{,i}), \tag{2.12}$$

where $G_{\alpha\beta}$ and g_{ij} are respectively the appropriate metric tensors and the stored energy Σ is such that

$$\frac{\rho}{\rho_R}\dot{\Sigma} = T^i_j D^j_i, \tag{2.13}$$

where ρ_R is the density in the reference configuration. The above, in direct notation, would read

$$\Sigma = \Sigma(\rho, \mathbf{F}, \mathbf{X}), \tag{2.14}$$

with

$$\frac{\rho}{\rho_R}\dot{\Sigma} = \mathbf{T}\cdot\mathbf{D}. \tag{2.15}$$

Accordingly, we shall assume that the stored energy associated with a compressible, inhomogeneous isotropic Green elastic material to be

$$\Sigma = -\frac{\mu_0(\mathbf{X})}{(\det\mathbf{F})^{n(\mathbf{X})}}\frac{I_m(\mathbf{X})-3}{2}\log\left(1-\left(\frac{I_1-3}{I_m(\mathbf{X})-3}\right)\right) + \frac{K_0(\mathbf{X})}{(\det\mathbf{F})^{n(\mathbf{X})}}\left(I_3^{\nu(\mathbf{X})} + \frac{1}{I_3^{\nu(\mathbf{X})}} - 2\right), \tag{2.16}$$

where $\mu_0(\mathbf{X})$ and $K_0(\mathbf{X})$ are the shear and bulk moduli, I_m is the stretch limit, ν is the volumetric exponent and n is exponent related to the extent of degradation of the material modulus.

It is a fairly common practice to partition the stored energy into a distortional and a volumetric part [20,21]. Gent [13] introduced a stored energy function for rubber-like materials wherein a finite stretch limit of cross-linked polymeric materials are taken into account. The rubber was assumed to be a homogeneous, isotropic, incompressible Green elastic body. Our stored energy cannot be split into a purely distortional and volumetric part as the $\det\mathbf{F}$ appears in both the terms, that is there is a nonlinear coupling between both the distortional and dilatational deformations. The first term of equation (2.16) has the same structure as that introduced by Gent except for the fact that it is inhomogeneous, and the modulus is made to decrease with an increase in $\det\mathbf{F}$. Note that this term becomes infinitely large as I_1 approaches the stretch limit I_m. The second term becomes infinitely large as $(\det\mathbf{F})\to 0$. Further, the stored energy is 0 when the deformation gradient is $\mathbf{I}$.

The Cauchy stress of the body takes the form

$$\begin{aligned}\mathbf{T} = {} & \frac{\mu_0(\mathbf{X})}{(\det\mathbf{F})^{n(\mathbf{X})+1}}\frac{1}{(1-((I_1-3)/(I_m(\mathbf{X})-3)))}\left[\bar{\bar{\mathbf{B}}} - \frac{1}{3}\operatorname{tr}(\bar{\bar{\mathbf{B}}})\mathbf{I}\right] \\ & + \frac{n(\mathbf{X})\mu_0(\mathbf{X})}{2(\det\mathbf{F})^{n(\mathbf{X})+1}}(I_m(\mathbf{X})-3)\log\left(1-\left(\frac{I_1-3}{I_m(\mathbf{X})-3}\right)\right)\mathbf{I} \\ & + \frac{K_0(\mathbf{X})\nu(\mathbf{X})}{(\det\mathbf{F})^{n(\mathbf{X})}}\left[I_3^{\nu(\mathbf{X})-1} - \frac{1}{I_3^{\nu(\mathbf{X})+1}}\right]\mathbf{I} - \frac{n(\mathbf{X})K_0(\mathbf{X})}{(\det\mathbf{F})^{n(\mathbf{X})+1}}\left[I_3^{\nu(\mathbf{X})} + \frac{1}{I_3^{\nu(\mathbf{X})}} - 2\right]\mathbf{I}.\end{aligned}$$

Note that the Cauchy stress vanishes when the deformation gradient is $\mathbf{I}$. Similar to the earlier model, we non-dimensionalize the above expression using the characteristic length scale L and

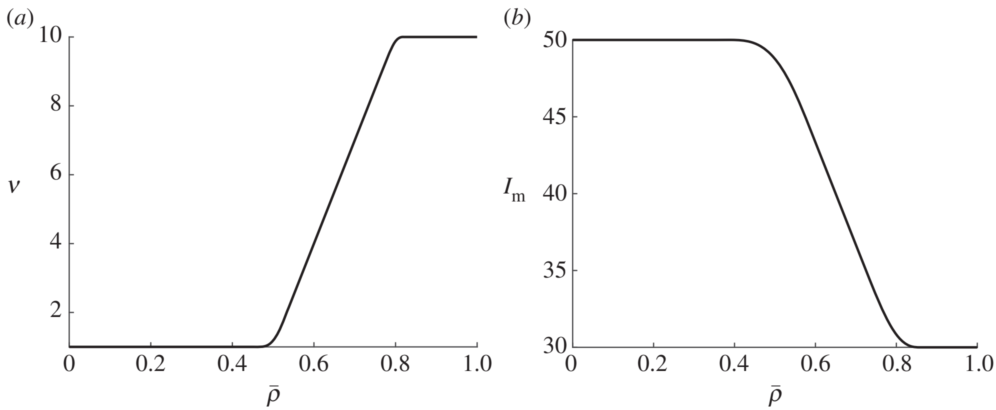

Figure 1. Material parameters variation. (*a*) Variation of ν and (*b*) Variation of I_m.

Table 2. Material parameters.

parameters	functions
ρ_0	smoothened Heaviside function (figure 4)
μ_0	$10^2\rho_0$
K_0	$10^4\rho_0$
ν	smoothened piecewise linear function (figure 1*a*)
I_m	smoothened piecewise linear function (figure 1*b*)
K_1	10^7
n	0.5

modulus K_1: $u = L\bar{u}$, $v = L\bar{v}$, $w = L\bar{w}$ and $\mathbf{T} = K_1\bar{\mathbf{T}}$.

$$\bar{\mathbf{T}} = \alpha_1[\bar{\mathbf{B}} - \tfrac{1}{3}\,\mathrm{tr}(\bar{\mathbf{B}})\mathbf{I}] + [\alpha_2 + \alpha_3 - \alpha_4]\,\mathbf{I}, \tag{2.17}$$

where

$$\alpha_1 = \frac{\mu_0(\bar{\mathbf{X}})}{K_1(\det\bar{\mathbf{F}})^{n(\bar{\mathbf{X}})+1}}\frac{1}{(1-((I_1-3)/(I_m(\bar{\mathbf{X}})-3)))}$$

$$\alpha_2 = \frac{n(\bar{\mathbf{X}})\mu_0(\bar{\mathbf{X}})}{2K_1(\det\bar{\mathbf{F}})^{n(\bar{\mathbf{X}})+1}}(I_m(\bar{\mathbf{X}})-3)\log\left(1-\left(\frac{I_1-3}{I_m(\bar{\mathbf{X}})-3}\right)\right)$$

$$\alpha_3 = \frac{K_0(\bar{\mathbf{X}})\nu(\bar{\mathbf{X}})}{K_1(\det\bar{\mathbf{F}})^{n(\bar{\mathbf{X}})}}\left[I_3^{\nu(\bar{\mathbf{X}})-1} - \frac{1}{I_3^{\nu(\bar{\mathbf{X}})+1}}\right]$$

and

$$\alpha_4 = \frac{K_0(\bar{\mathbf{X}})n(\bar{\mathbf{X}})}{K_1(\det\bar{\mathbf{F}})^{n(\bar{\mathbf{X}})+1}}\left[I_3^{\nu(\bar{\mathbf{X}})} + \frac{1}{I_3^{\nu(\bar{\mathbf{X}})}} - 2\right]$$

The material parameters used in this study are shown in table 2.

Table 3. Material parameters for three-dimensional simulation.

parameters	functions
ρ_0	smoothened Heaviside function (figure 4)
μ_0	$10^3\rho_0$
K_0	μ_0
K_1	10^6

3. Biaxial stretch of an inhomogeneous elastic sheet of finite thickness: three-dimensional solution

Let us consider a square plate of side L and thickness $0.0001L$ in a three-dimensional Cartesian coordinate system. The reference density ($\bar{\rho}_0$) is constant over a large region and it is allowed to vary in small circular region at the centre of the square plate (see figure 4 where $\bar{\rho}_{\min} = 0.5$), i.e. the inhomogeneity is modelled by a smoothened Heaviside function. Because this region is circular, we use the symmetry condition to consider only one quarter of the plate and the whole thickness for the further analysis. The initial density profile variation along the y-axis can be seen in figure 4.

The non-dimensionalized X, Y and Z component of displacements are assumed to be $\bar{u}(X, Y, Z)$, $\bar{v}(X, Y, Z)$ and $\bar{w}(X, Y, Z)$. The neo-Hookean plate is subjected to a biaxial stretch of 2. The boundary condition are as follows

$$\bar{u}(1, Y, Z) = 1 \quad \forall\, 0 \le Y \le 1 \;\&\; 0 \le Z \le 0.0001;$$
$$\bar{v}(X, 1, Z) = 1 \quad \forall\, 0 \le X \le 1 \;\&\; 0 \le Z \le 0.0001;$$
$$\bar{\mathbf{P}}.\mathbf{N} = 0 \quad \forall\, 0 \le X \le 1 \;\&\; 0 \le Y \le 1 \;\&\; Z = 0, 0.0001,$$

and the symmetry conditions are

$$\bar{u}(0, Y, Z) = 0 \quad \forall\, 0 < Y < 1 \;\&\; 0 < Z < 0.0001;$$
$$\bar{v}(X, 0, Z) = 0, \quad \forall\, 0 < X < 1 \;\&\; 0 < Z < 0.0001,$$

where $\bar{\mathbf{P}}$ is the first Piola–Kirchoff stress. The three-dimensional equilibrium equations, in the absence of body forces, are

$$\frac{\partial \bar{P}_{XX}}{\partial X} + \frac{\partial \bar{P}_{XY}}{\partial Y} + \frac{\partial \bar{P}_{XZ}}{\partial Z} = 0, \tag{3.1}$$

$$\frac{\partial \bar{P}_{YX}}{X} + \frac{\partial \bar{P}_{YY}}{\partial Y} + \frac{\partial \bar{P}_{YZ}}{\partial Z} = 0 \tag{3.2}$$

and

$$\frac{\partial \bar{P}_{ZX}}{\partial X} + \frac{\partial \bar{P}_{ZY}}{\partial Y} + \frac{\partial \bar{P}_{ZZ}}{\partial Z} = 0. \tag{3.3}$$

The constitutive relation for the Cauchy stress given in equation (2.10) and the relation between Cauchy stress and first Piola stress is given by

$$\bar{\mathbf{P}} = (\det \bar{\mathbf{F}})\bar{\mathbf{T}}\bar{\mathbf{F}}^{-\mathrm{T}}. \tag{3.4}$$

We use equation (2.10) and equations (3.1)–(3.4) to solve for the displacements components. The material parameters used for the three-dimensional analysis are shown in table 3. Figure 2 shows the displacement ('$\bar{w}$') in the 'Z'-direction along the thickness at the centre of the plate. It can be observed that the '$\bar{w}$' varies almost linearly with 'Z'. The above-mentioned numerical solution suggests a very useful simplification of the problem that will make it much more amenable to numerical analysis. We note that the displacement '$\bar{w}$' varies nearly linearly with

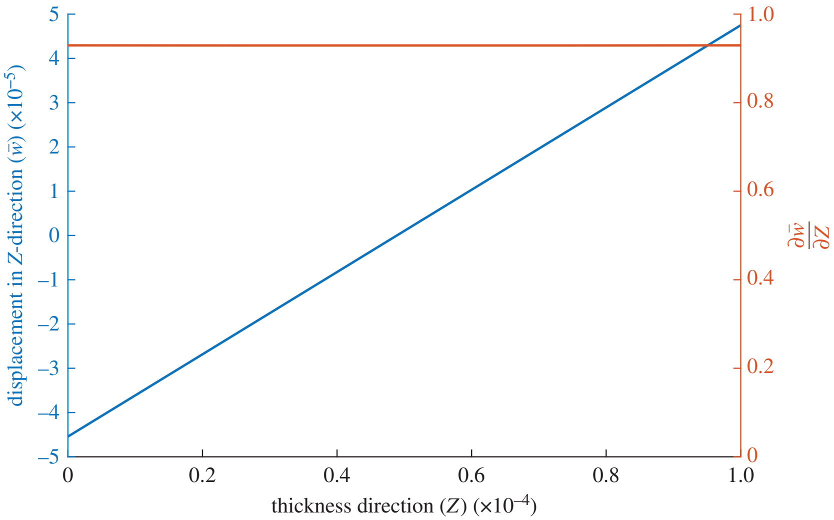

Figure 2. Displacement in the Z-direction at the centre of the plate through the whole thickness. (Online version in colour.)

the Z-coordinate suggesting the assumption that $\bar{w}(X, Y, Z) = Z\phi(X, Y)$ will reduce the three-dimensional problem into a two-dimensional problem, thereby reducing the complexity of the problem significantly. In virtue of the savings in memory and time, we now proceed to study in detail a two-dimensional approximation of the problem.

4. Biaxial stretch of an inhomogeneous elastic sheet: two-dimensional solution

Let us consider a thin square plate of side L. The density profile in the reference configuration is assumed to have a structure similar to that in the three-dimensional study.

(a) Governing equations

In virtue of the result for the three-dimensional solution that suggests a linear relationship between the displacement in the Z-direction and the Z-coordinate, for a specimen that is a sufficiently thin sheet, the non-dimensionalized X, Y and Z component of displacements are assumed to be $\bar{u}(X, Y)$, $\bar{v}(X, Y)$ and $\bar{w}(X, Y, Z) = Z\phi(X, Y)$. It is also worth observing that Varley & Cumberbatch [22] also appealed to the same assumption concerning the displacement '$\bar{w}$' to study large deformations of harmonic materials (the study by Varley & Cumberbatch [22]) is recondite as there are no bodies that can be described through the constitutive model for harmonic materials, it seems that the choice of the form of the constitutive relation was chosen, so that the problem could reduce to a well-known partial differential equation). The deformation gradient is given by

$$\mathbf{F} = \mathbf{I} + \frac{\partial \mathbf{u}(\mathbf{X})}{\partial \mathbf{X}}, \tag{4.1}$$

i.e.

$$\mathbf{F} = \begin{bmatrix} 1 + \dfrac{\partial \bar{u}}{\partial X} & \dfrac{\partial \bar{u}}{\partial Y} & 0 \\ \dfrac{\partial \bar{v}}{\partial X} & 1 + \dfrac{\partial \bar{v}}{\partial Y} & 0 \\ Z\dfrac{\partial \phi}{\partial X} & Z\dfrac{\partial \phi}{\partial Y} & 1 + \phi \end{bmatrix}. \tag{4.2}$$

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Using the linear dependence for the Z displacement, Cauchy stress components $\bar{T}_{XZ}$ and $\bar{T}_{YZ}$ for the generalized Gent body are as follows

$$\bar{T}_{XZ} = \frac{\beta_1 Z(\partial\phi/\partial X + \partial\bar{u}/\partial X(\partial\phi/\partial X) + \partial\bar{u}/\partial Y(\partial\phi/\partial Y))}{(\phi+1)((\partial\bar{u}/\partial X+1)\partial\bar{v}/\partial Y + \partial\bar{u}/\partial X - \partial\bar{u}/\partial Y(\partial\bar{v}/\partial X)+1)} \tag{4.3}$$

and

$$\bar{T}_{YZ} = \frac{\beta_1 Z(\partial\phi/\partial Y + \partial\bar{v}/\partial X(\partial\phi/\partial X) + \partial\bar{v}/\partial Y(\partial\phi/\partial Y))}{(\phi+1)((\partial\bar{v}/\partial Y+1)\partial\bar{u}/\partial X + \partial\bar{v}/\partial Y - \partial\bar{u}/\partial Y(\partial\bar{v}/\partial X)+1)}, \tag{4.4}$$

and for a neo-Hookean body, expression (4.4) for the shear stresses take the form

$$\bar{T}_{XZ} = \frac{\alpha_2 Z(\partial\phi/\partial X + \partial\bar{u}/\partial X(\partial\phi/\partial X) + \partial\bar{u}/\partial Y(\partial\phi/\partial Y))}{(\phi+1)((\partial\bar{u}/\partial X+1)\partial\bar{v}/\partial Y + \partial\bar{u}/\partial X - \partial\bar{u}/\partial Y(\partial\bar{v}/\partial X)+1)} \tag{4.5}$$

and

$$\bar{T}_{YZ} = \frac{\alpha_2 Z(\partial\phi/\partial Y + \partial\bar{v}/\partial X(\partial\phi/\partial X) + \partial\bar{v}/\partial Y(\partial\phi/\partial Y))}{(\phi+1)((\partial\bar{v}/\partial Y+1)\partial\bar{u}/\partial X + \partial\bar{v}/\partial Y - \partial\bar{u}/\partial Y(\partial\bar{v}/\partial X)+1)}. \tag{4.6}$$

The other components of Cauchy stress, namely $\bar{T}_{XX}$, $\bar{T}_{YY}$ and $\bar{T}_{XY}$, do not show any dependence on Z. Therefore, by selecting an appropriately small thickness for the plate, the components $Z(\partial\phi/\partial X)$, $Z(\partial\phi/\partial Y)$, $\bar{T}_{XZ}$ and $\bar{T}_{YZ}$ will be negligible, and from this perspective, we can drop these terms. This results in the deformation gradient being only a function of 'X' and 'Y', i.e.

$$\mathbf{F} = \begin{bmatrix} 1+\dfrac{\partial\bar{u}}{\partial X} & \dfrac{\partial\bar{u}}{\partial Y} & 0 \\ \dfrac{\partial\bar{v}}{\partial X} & 1+\dfrac{\partial\bar{v}}{\partial Y} & 0 \\ 0 & 0 & 1+\phi \end{bmatrix}. \tag{4.7}$$

The plate is biaxially stretched and the boundary conditions are given by

$$\bar{u}(1,Y) = u_1 \quad \forall\, 0 < Y < 1$$
$$\bar{v}(X,1) = u_1 \quad \forall\, 0 < X < 1$$

and the symmetry conditions are

$$\bar{u}(0,Y) = 0 \quad \forall\, 0 < Y < 1$$
$$\bar{v}(X,0) = 0 \quad \forall\, 0 < X < 1.$$

Because **F** is only a function of the coordinates X and Y, the traction-free condition on the top and the bottom of the sheet reduces to $\bar{P}_{ZZ} = 0$. As large deformations of the polymeric solid are involved and it is necessary to make a distinction between the Lagrangian and Eulerian formulations. It would be numerically convenient to use a Lagrangian formulation because it does not involve the evolving geometry and hence we use it in this study. The non-dimensional governing equation is given by

$$\mathrm{Div}\,\bar{\mathbf{P}} = 0,$$

where $\bar{\mathbf{P}}$ is the first Piola–Kirchhoff stress tensor. The relation between the Cauchy stress and first Piola–Kirchhoff tensor is given by

$$\bar{\mathbf{P}} = (\det\bar{\mathbf{F}})\bar{\mathbf{T}}\bar{\mathbf{F}}^{-\mathrm{T}}.$$

Using the above the relation, the Piola Kirchhoff stress components for the generalized Gent model are as follows

$$\bar{P}_{XX} = \beta_1 + \beta_1 \frac{\partial \bar{u}}{\partial X} + \left(\frac{\partial \bar{v}}{\partial Y} + 1 \right) (\phi + 1)(\beta_3 - \beta_1 \beta_2), \tag{4.8}$$

$$\bar{P}_{YY} = \beta_1 + \beta_1 \frac{\partial \bar{v}}{\partial Y} + \left(\frac{\partial \bar{u}}{\partial X} + 1 \right) (\phi + 1)(\beta_3 - \beta_1 \beta_2), \tag{4.9}$$

$$\bar{P}_{XY} = \beta_1 \frac{\partial \bar{u}}{\partial Y} - \frac{\partial \bar{v}}{\partial X}(\phi + 1)(\beta_3 - \beta_1 \beta_2), \tag{4.10}$$

$$\bar{P}_{YX} = \beta_1 \frac{\partial \bar{v}}{\partial X} - \frac{\partial \bar{u}}{\partial Y}(\phi + 1)(\beta_3 - \beta_1 \beta_2) \tag{4.11}$$

and

$$\bar{P}_{ZZ} = (\beta_3 - \beta_1 \beta_2) \left(\frac{\partial \bar{u}}{\partial X} + \frac{\partial \bar{v}}{\partial Y} + \frac{\partial \bar{u}}{\partial X}\frac{\partial \bar{v}}{\partial Y} - \frac{\partial \bar{u}}{\partial Y}\frac{\partial \bar{v}}{\partial X} + 1 \right) + \beta_1(\phi + 1), \tag{4.12}$$

where

$$\beta_1 = \frac{\mu_0(\bar{\mathbf{X}})}{K_1(\det \bar{\mathbf{F}})^{n(\bar{\mathbf{X}})}} (\det \bar{\mathbf{F}})^{-2/3} \frac{1}{(1 - ((\mathrm{tr}(\bar{\mathbf{B}}) - 3)/(I_m(\bar{\mathbf{X}}) - 3)))},$$

$$\beta_2 = \frac{1}{3} \frac{\mathrm{tr}(\bar{\mathbf{B}})}{\det \bar{\mathbf{F}}},$$

$$\beta_3 = \frac{n\mu_0(\bar{\mathbf{X}})(I_m(\bar{\mathbf{X}}) - 3)}{2K_1(\det \bar{\mathbf{F}})^{n(\bar{\mathbf{X}})}} \log \left(1 - \left(\frac{\mathrm{tr}(\bar{\mathbf{B}}) - 3}{I_m(\bar{\mathbf{X}}) - 3} \right) \right) + \frac{K_0(\bar{\mathbf{X}})\nu(\bar{\mathbf{X}})}{K_1(\det \bar{\mathbf{F}})^{n(\bar{\mathbf{X}})}} \left[I_3^{\nu(\bar{\mathbf{X}})} - \frac{1}{I_3^{\nu(\bar{\mathbf{X}})}} \right]$$
$$- \frac{K_0(\bar{\mathbf{X}})n(\bar{\mathbf{X}})}{K_1(\det \bar{\mathbf{F}})^{n(\bar{\mathbf{X}})}} \left[I_3^{\nu(\bar{\mathbf{X}})} + \frac{1}{I_3^{\nu(\bar{\mathbf{X}})}} - 2 \right].$$

The material parameters for the generalized Gent model are shown in table 2.

Similarly, the Piola stress components for the generalized neo-Hookean model are as follows

$$\bar{P}_{XX} = \alpha_2 + \alpha_2 \frac{\partial \bar{u}}{\partial X} + \alpha_1 \left(\frac{\partial \bar{v}}{\partial Y} + 1 \right) (\phi + 1), \tag{4.13}$$

$$\bar{P}_{YY} = \alpha_2 + \alpha_2 \frac{\partial \bar{v}}{\partial Y} + \alpha_1 \left(\frac{\partial \bar{u}}{\partial X} + 1 \right) (\phi + 1), \tag{4.14}$$

$$\bar{P}_{XY} = \alpha_2 \frac{\partial \bar{u}}{\partial Y} - \alpha_1 \frac{\partial \bar{v}}{\partial X} - \alpha_1 \phi \frac{\partial \bar{v}}{\partial X}, \tag{4.15}$$

$$\bar{P}_{YX} = \alpha_2 \frac{\partial \bar{v}}{\partial X} - \alpha_1 \frac{\partial \bar{u}}{\partial Y} - \alpha_1 \phi \frac{\partial \bar{u}}{\partial Y} \tag{4.16}$$

and

$$\bar{P}_{ZZ} = \alpha_2(\phi + 1) + \alpha_1 \left(\frac{\partial \bar{u}}{\partial X} + \frac{\partial \bar{v}}{\partial Y} + \frac{\partial \bar{u}}{\partial X}\frac{\partial \bar{v}}{\partial Y} - \frac{\partial \bar{u}}{\partial Y}\frac{\partial \bar{v}}{\partial X} + 1 \right), \tag{4.17}$$

where

$$\alpha_1 = \frac{K_0(\det \bar{\mathbf{F}} - 1)}{K_1 \det \bar{\mathbf{F}}} - \frac{\mu_0 \det \bar{\mathbf{F}}}{3K_1} \mathrm{tr}\, \bar{\mathbf{B}},$$

$$\alpha_2 = \frac{\mu_0}{K_1} \det \bar{\mathbf{F}}.$$

The material parameters for the generalized neo-Hookean model are shown in table 1.

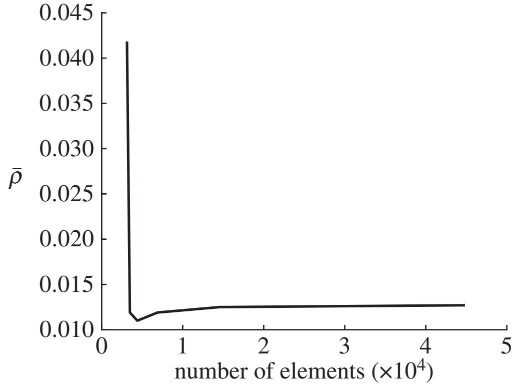

Figure 3. Normalized current density at a stretch of 3.9 for different mesh sizes at the centre of the plate, where initial reference density is the lowest.

The equilibrium equations, in the absence of body forces, are

$$\frac{\partial \bar{P}_{XX}}{\partial X} + \frac{\partial \bar{P}_{XY}}{\partial Y} = 0, \tag{4.18}$$

$$\frac{\partial \bar{P}_{YX}}{\partial X} + \frac{\partial \bar{P}_{YY}}{\partial Y} = 0 \tag{4.19}$$

and

$$\bar{P}_{ZZ} = 0, \tag{4.20}$$

and the balance of mass is given by

$$\rho = \rho_0 \det \bar{\mathbf{F}}. \tag{4.21}$$

(b) Numerical method

The equation-based modelling in COMSOL Multiphysics® was used to solve the above-mentioned set of partial differential equations using the finite-element method. As the study involves large deformations of the plate, the displacement boundary condition was applied in step increments, and the solution of the previous step was fed as the initial guess for the next step. The number of elements was fixed based on a grid independence study. The relative tolerance was of the order of 10^{-6}. Very fine triangular elements were provided in the region where there were initial density variations. Of the different quantities such as stresses, displacement and current density, the current density ($\bar{\rho}$) was the one, which showed sensitivity to the number of elements. Hence, we show here the variation of $\bar{\rho}$ with the number of elements in figure 3. The mesh size of 2.5×10^{-6} in the inner region corresponds to 44 038 elements (figure 3), and this was chosen for further analysis in this study.

5. Damage initiation criterion and results

The effect of inhomogeneity on the response of the polymeric sheet was studied using a smoothened Heaviside function for two specific inhomogeneities, one wherein the normalized densities in the central region and outside the central region were chosen to be 0.3 and 1, respectively, with a sharp change in a narrow domain (figure 4), whereas in the other case the density is 0.5 in the centre region, and 1 outside the circular region of inhomogeneity.

In both these cases, we find that the region that is less dense in the reference (undeformed) configuration, deteriorates (in the sense that the density and hence the load carrying capacity drops) very much more severely than the material in the rest of the body, indicating that failure will occur first in this region of initially lower density. For example, the percentage reduction

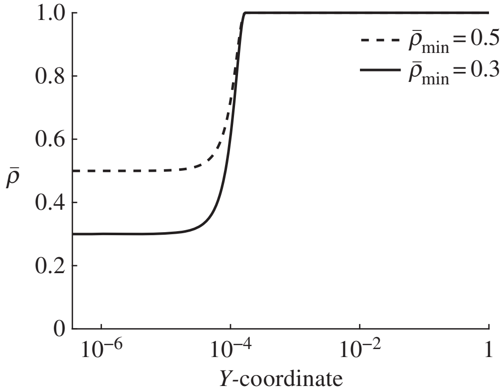

Figure 4. Initial variation of $\bar{\rho}$ with the Y-coordinate for the two cases considered in this study.

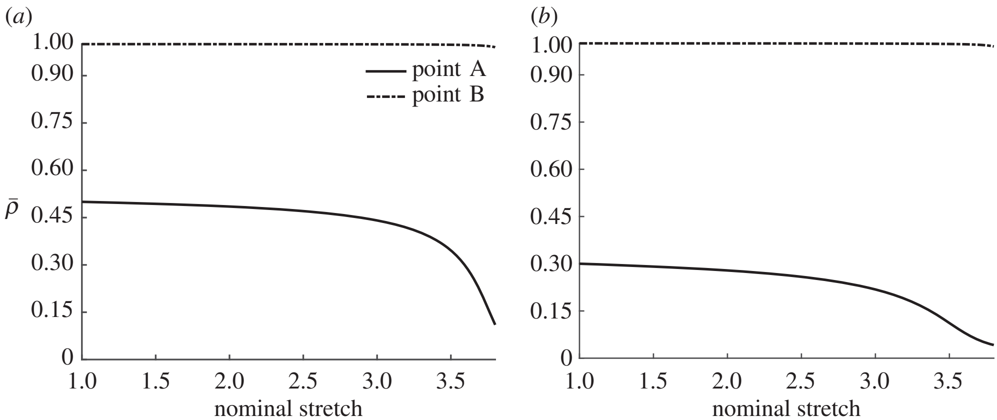

Figure 5. Density variation with nominal stretch for the generalized Gent model. (*a*) $\bar{\rho}$ at the centre is 50% of farfield value and (*b*) $\bar{\rho}$ at the centre is 30% of the farfield value.

in density is above 95% in the central region, whereas away from the central circular region of initially lower density, it is less than 1% at a stretch of 3.9 for the generalization of the Gent model (table 4). The drop in the density and drastic increase in the local volume which are observed are clear indicators of the body's impending failure. Let us first consider the case of the generalization of the Gent model. Figure 5*a* displays the variation of the density with the stretch in the case of the central inner region which is 50% of the far-field value. We note that at point B (figure 5*a*), which is representative of the far-field, the density does not change significantly implying that the integrity of the material is maintained. The manner in which the norm of the stress changes with the density is portrayed in figure 6. We note as expected, as the density decreases (which corresponds to the stretch increasing), initially the norm of the stress increases. However, we note that a critical value of the density the norm of the stress starts to decrease with an increase in stretch. It is our conjecture that it is at this point that the material gets damaged as the load carrying capacity decreases with increasing stretch (decreasing density) at this point. *Thus, we postulate the criterion for initiation of damage to be the deformation corresponding to which the derivative of the norm of the stress decreases with respect to density*. The stretch at which damage is initiated according to this criterion agrees quite well with the experiments of Gent wherein damage initiates at stretches around 3.5. Although the body has been stretched beyond this value of stretch and calculations have been carried out beyond this critical value for the density, as though the body is yet elastic, as damage

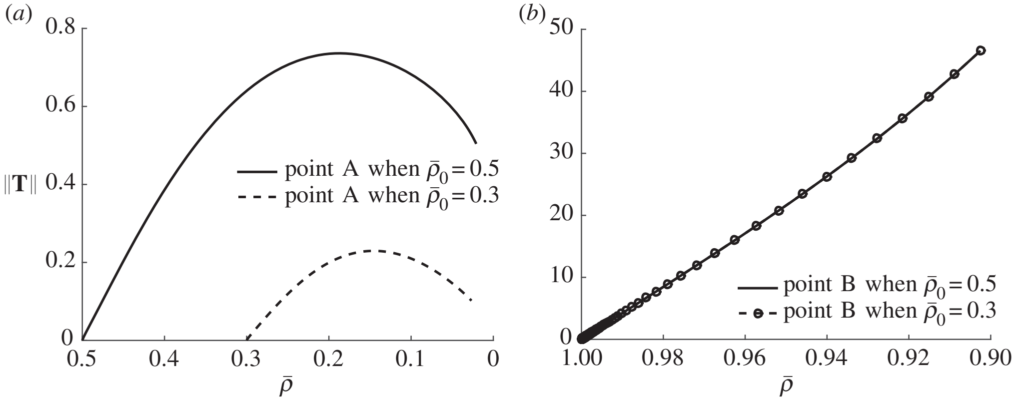

Figure 6. Norm of stress tensor with stretch for generalized Gent model. (*a*) Centre point variation and (*b*) Farfield point variation.

Table 4. Percentage reduction in density for the generalized Gent model at a stretch of 3.9.

Y-coordinate	percentage reduction in density	*Y*-coordinate	percentage reduction in density
0	95.7	5.71×10^{-5}	45.4
7.50×10^{-6}	94.9	6.07×10^{-5}	40.1
1.93×10^{-5}	90.2	6.43×10^{-5}	35.1
2.61×10^{-5}	85.2	6.82×10^{-5}	30.0
3.14×10^{-5}	80.1	7.21×10^{-5}	25.2
3.57×10^{-5}	75.3	7.71×10^{-5}	20.0
4.00×10^{-5}	70.0	8.29×10^{-5}	15.2
4.36×10^{-5}	65.2	8.96×10^{-5}	10.8
4.71×10^{-5}	60.1	1.02×10^{-4}	5.9
5.04×10^{-5}	55.4	1	0.98

has been initiated, one needs to use a different model when the derivative of the norm of the stress with respect to density starts decreasing. That is, unloading a real body after the peak value in both the curves depicted in figure 6*a*. On unloading a real body after the peak values depicted in figure 6, the material will not go back along the same original loading curve as an elastic material will but respond by traversing a different path to a new stress free state. That is, our calculations have been carried out under the assumption that the material is elastic and hence on unloading the material, it would follow the same loading curve. Our aim in this study is just to highlight the use of the inhomogeneity of the body with the density as a possible indicator for damage rather than develop a model for a body that is undergoing damage in that a constitutive relation is also provided for the response of the body after damage has been initiated. This point needs to be emphasized. We note that at point A after a stretch of approximately 3.5 the density starts to decrease significantly. We note that our damage criterion, namely the derivative of the norm of the stress with respect to the density, occurs when the density is 0.185, a nearly 65% drop in the density, at a stretch of approximately 3.73 and the value of the determinant of $\bar{\mathbf{F}}$ which is a measure of the volume change being 2.7 (figure 6*a*). We also note that the density in the far-field falls by less than 5%, and the material is undamaged. We reiterate that the value of the stretch

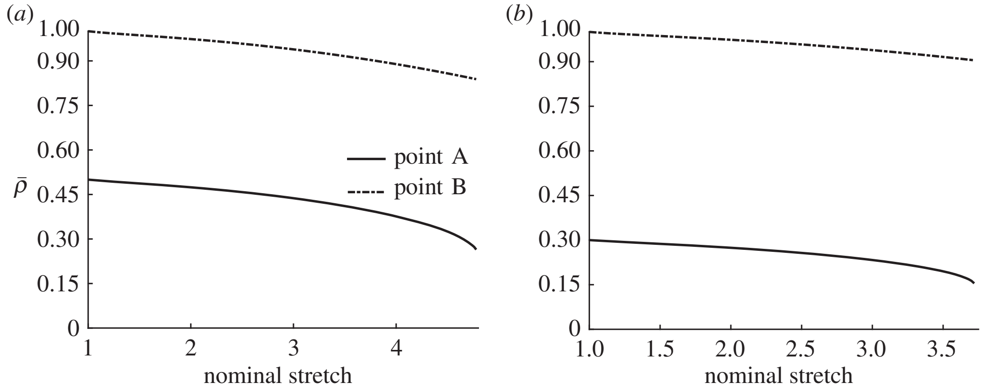

Figure 7. Density variation with nominal stretch for generalized neo-Hookean model. (*a*) $\bar{\rho}$ at the centre is 50% of farfield and (*b*) $\bar{\rho}$ at the centre is 30% of farfield.

at which damage occurs in the region of lower density, namely 3.73 is in keeping with what is observed in experiments [23]. Also the volumetric strain in the far-field is of order 10^{-4} similar to what has been observed in experiments (see [20]). Moreover, the value of the stretch where the norm of the stress starts to decrease and the density starts to undergo a precipitous drop is approximately the same. In the case of an inhomogeneity that is merely 30% of the far-field value, we see from figure 5*b* that the density once again decreases by less than 5% in the farfield (point A) while the damage criterion implies that damage has occurred when a density of 0.135 is reached, a decrease of around 52%, corresponding to a stretch of 3.385 and the determinant of the deformation gradient of 2.06 (figure 6*a*,*b*). Once again, the value of the stretch and the volumetric strain in the far-field when damage occurs are consistent with experimental observations. As before, the value of the stretch where the norm of the stress starts to decrease, and the density starts to undergo a precipitous drop is approximately the same.

Next, let us consider the biaxial stretching of a generalized compressible neo-Hookean body. The results in this case for both types of inhomogeneity are similar to that in the previous case, the only difference being in the values. In the case of the inhomogeneity wherein the density is 50% of the far-field value, the density reduces to a value of 0.335, a drop of 33%, at a stretch of 4.43, the determinant of the deformation gradient being 1.5, whereas in the far-field, the density falls by less than 10% (figures 7 and 8). In the case of an initially inhomogeneity wherein the density is 30% of the far-field value, the density decreases to 0.2, once again a drop of 33%, at a stretch of 3.45, the determinant of the deformation being 1.5, while in the far-field the density falls by around 10% (figures 7 and 8).

The state of normal, shear, radial and circumferential stress for a generalized Gent and a generalized compressible neo-Hookean, body with inhomogeneity in the density that is 30% of the far-field value, are shown in figures 9 and 10. The circular arc shown in figures is the region above which the normalized density is 1. We note that the maximum value of the tensile stress as well as the shear stress occur in the transitional region where the density falls from the far-field value to that of the lower level at the centre. Furthermore, the circumferential stress depicted in figure 9*d* indicates an annular circular region of maximum stress that coincides with the region in the undeformed state wherein the density changes significantly from the value in the far-field to the lower value in the central circular domain. The radial stresses shown in 9*c* are axisymmetric, because the inhomogeneity is circular and stretch is equi-biaxial. Similar behaviour is observed in the generalized neo-Hookean model. Because there is no stretch limit in the generalized neo-Hookean body unlike that for the generalized Gent model, the magnitude of stress difference between the centre and far-field is much less than that compared with the generalized Gent model (figures 9 and 10).

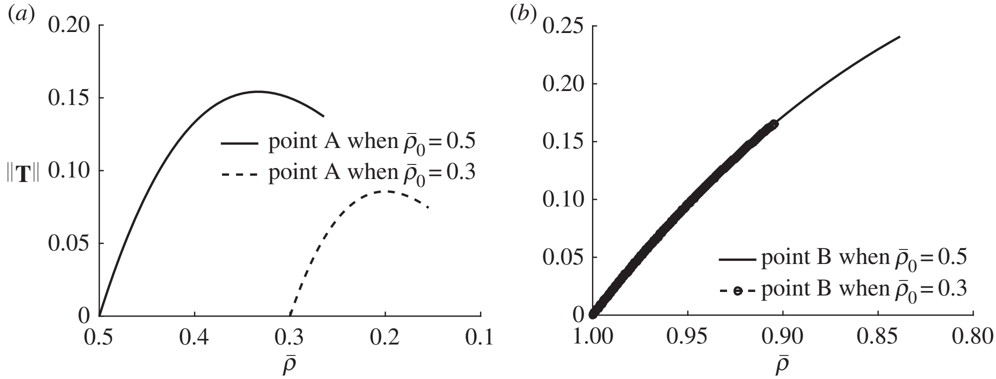

Figure 8. Norm of stress tensor with stretch for generalized neo-Hookean model. (*a*) Centre point variation and (*b*) Farfield point variation.

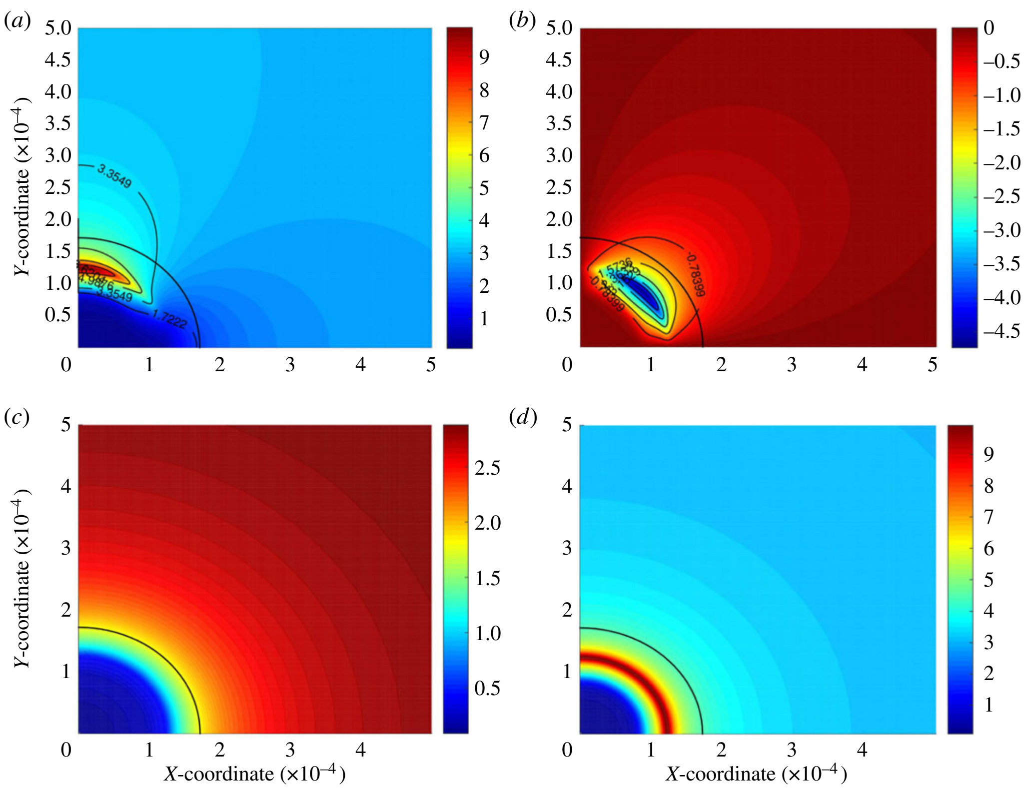

Figure 9. Generalized Gent model. (*a*) Normal stress component, T_{xx}, (*b*) shear stress component, T_{xy}, (*c*) radial stress and (*d*) circumferential stress. (Online version in colour.)

6. Conclusion

It is usual to prescribe the initiation of damage in a body by resorting to ad hoc internal variables, which could be scalar, vector or tensor in character, that most often do not have meaningful physical significance. These parameters that qualify and quantify damage, within a mechanical context, are assumed to depend on the stress, strain or other kinematical variables. Such a procedure is fraught with difficulties in virtue of the fact that for given a body in a particular configuration, one does not know if it is stress free or if it has undergone permanent set in that it

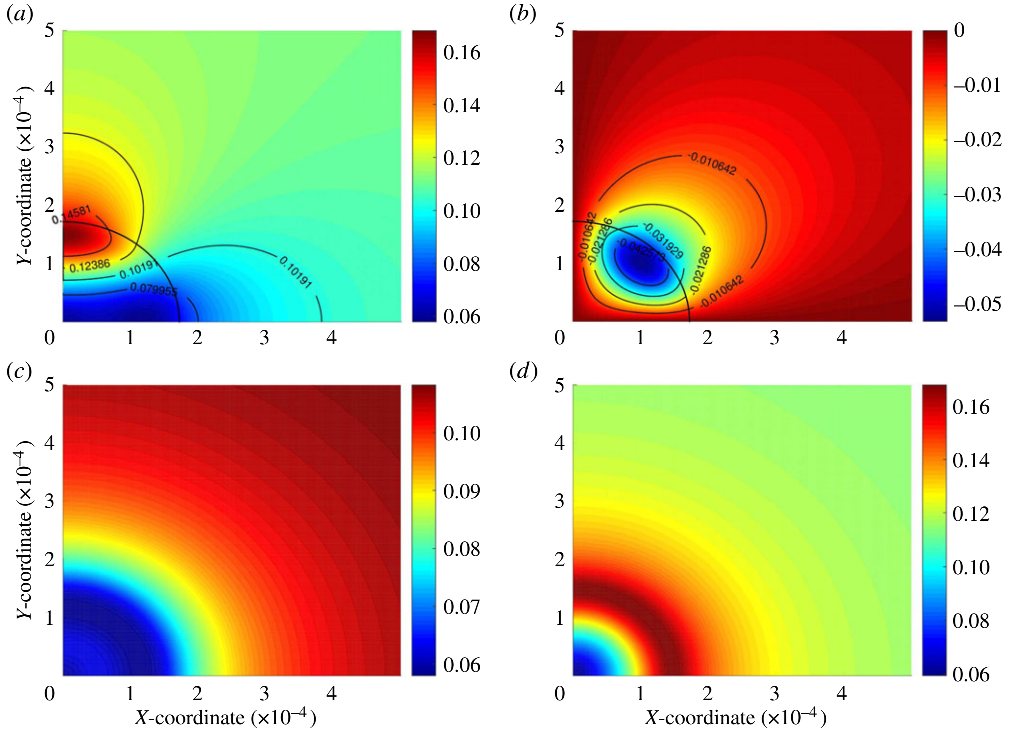

Figure 10. Generalized compressible neo-Hookean model. (*a*) Normal stress component, T_{xx}, (*b*) shear stress component, T_{xy}, (*c*) radial stress and (*d*) circumferential stress. (Online version in colour.)

is strained with respect to some other previous configuration. Thus, a damage parameter that depends on measures such as the stress or strain, or for that matter any quantity that could depend on knowledge of a previous configuration that the body might have taken would be inappropriate. To be a meaningful criterion for defining the initiation of damage in a material, one needs to use quantities whose values are completely determined by the current configuration of the body. One such quantity is the density of the body that is completely determined in its current configuration. It also seems a very meaningful quantity for defining the initiation of damage in a class of materials as damage in such materials seems to be concomitant with the density in the body decreasing precipitously at local inhomogeneities. In this paper, we have investigated the possibility of using 'damage' as the parameter for defining the initiation of damage. We define damage to be initiated when the derivative of the norm of the stress with respect to the density becomes negative. Such a criterion implies that the load carrying capacity of the body starts to decrease at that location. We find that in the biaxial stretching of polymeric sheets such a criterion agrees well with the experiments of Gent.

Authors' contributions. P.A. was involved in the development of the governing equations and carried out all the numerical computations, Dr K.K. helped in the formulation of the boundary value problems and the development of the governing equations. Prof. K.R.R. suggested the problem and developed the theoretical framework within which to study it and supervised the solution of the problem. All the authors were involved in the write up and presentation of the problem.

Competing interests. We declare we have no competing interests.

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