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ON THE MODELING OF ENTROPY PRODUCING PROCESSES[*]

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Abstract. A general thermodynamic framework is presented for the study of the response of bodies undergoing entropy producing processes. In general, in such processes the natural configuration of a body, i.e., the configuration that the body would take on the removal of all external stimuli, changes. The fact that material symmetry of the body in these various natural configurations could be different allows one to model the response of bodies that cannot be described by traditional models that are in place. It is assumed that the processes take place in a manner such that the rate at which entropy is produced is maximized. Knowing how the material stores energy, produces entropy, conducts heat, absorbs or emits radiation, etc., allows one to determine the constitutive equation for the stress and other relevant quantities. The fact that the body's natural configuration changes and the form for the stress response from the natural configuration changes, leads to a lot of challenges with regard to the development of analytical as well as numerical methods for the study of the response of bodies.

Key words. Rate of entropy production, internal energy, Helmholtz potential, rate of dissipation, second law of thermodynamics.

AMS subject classifications. 80A17, 74C10, 74C15, 74C20, 76A05, 76A10, 76A15.

1. Introduction.

The response of bodies to external stimuli is characterized by the many ways in which bodies store energy, how they release this energy that is stored, the various ways in which they produce entropy, how they conduct heat, how they emit and absorb radiation, the structures for their latent heat and latent energy (the difference in the internal energy associated with the different phases of the body), how much of the working that is supplied is converted into heat, and in general other pertinent information with regard to the electromagnetic response of bodies. For instance, a particular body might be able to store the energy that is supplied to the body in such a manner that all of it can be recovered in a purely mechanical process (such bodies are usually called elastic), however crystalline bodies with dislocations are capable of storing energy due to the rearrangement of the dislocation structure, that cannot be recovered in a purely mechanical process, the energy being recovered in a thermodynamic process such as annealing. The external stimuli are not restricted to mechanical and thermal quantities, a body is also stimulated by electrical and magnetic fields or an active chemical environment; here we shall restrict ourselves to purely mechanical and thermal stimuli.

The configuration that a body takes in the absence of external stimuli is called a natural configuration. Given a set of external stimuli, it is possible that different natural configurations might be achieved in the manner in which the external stimuli are removed. The natural configuration achieved might be different based on whether the external loading is removed instantaneously or very slowly. In an elastic body the manner in which the external load is removed is irrelevant and one attains the same stress free configuration. It is however important to recognize that a traction free configuration does not necessarily lead to a stress free configuration. A simple example for the same is the case of an elastic hemisphere; it could be traction free in a stress free state or traction free in an everted stressed state. This leads to the important fact that all equilibrium solutions for elastic bodies cannot be obtained by requiring that the stored energy be a global minimum.

As the body is undergoing a thermodynamic process, the underlying natural configuration can change, that is the configuration that the body attains on the removal of external

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stimuli, at different actual configurations of the body undergoing a thermodynamic process, could change. An elastic material is a body that has one natural configuration and thus, on the removal of external loading from any natural configuration, the body will return to the same natural configuration. However, an inelastic body, such as the a metal that has been deformed so that it has “yielded”, has an infinity of natural configurations, and a body that is undergoing solid to solid phase transition has a finite number of natural configurations; see [16] and [18, 19] for a detailed discussion of the role of natural configurations in thermomechanics. The question then arises as to whether there is some rational means for determining how the natural configuration changes. It turns out that natural configurations change whenever entropy production takes place (thus the reason for the natural configuration not changing for an elastic body, though it is possible that one can construct a mathematical theory wherein the natural configurations do not change even when entropy is produced). Eckart [4] seems to have been the first to recognize the important role that natural configurations play in specifying the response of materials. For instance, classical plasticity can be viewed as infinity of response functions from infinity of evolving natural configurations. His work is amongst the most important studies in the thermomechanics of entropy producing processes and it is unfortunate that this seminal work has been largely ignored until recently. While Eckart’s work made a significant advance on the state of entropy producing processes and the notion of natural configurations, he did not recognize the role of the changing material symmetry associated with these natural configurations and other related issues; see Rajagopal [16] for a discussion of the same.

We shall find that requiring the thermodynamic processes to proceed according to the rate of entropy production being maximized, leads naturally to determining the manner in which the natural configurations evolve. As we shall see, appealing to this idea has been successful in developing models to describe a disparate class of material responses.

Suppose that the current configuration of the body is denoted by κ_t, and further suppose that on the removal of the external stimuli the body attains the configuration $\kappa_{p(t)}$, the preferred natural configuration of the body amongst the several natural configurations that are available to the body. As mentioned earlier, different natural configurations can be attained based on the class of allowable thermodynamic processes (it depends on the way in which the external stimuli are removed, for instance if the external stimuli are removed instantaneously the process under consideration would be adiabatic, while if they are removed slowly the process would be isothermal). Also, a body might have one, a finite number or infinity of natural configurations associated with it while undergoing a thermodynamic process. Twinning and solid to solid phase transitions are examples of a body having a finite number of natural configurations.

Given a body, we have to decide on the set of properties that define the state of each material point that belongs to the body. For instance, the deformation gradient, temperature, stress, velocity gradient, the various temporal and spatial derivatives of the above quantities as well as several other quantities could qualify to define the state of a particle. An important point to bear in mind is that the set of natural configurations that a body can attain will also be a part of the specification of the state variables. Constitutive relations are in their most general form implicit relationships between the various state variables. Once the state space associated with the body can be defined, we can discuss the processes that take the particle from one state to another. These processes cannot be arbitrary; they have to be such that in addition to the balance equations the second law of thermodynamics is met. The second law requires the rate of entropy production to be non-negative. The second law is enforced in a variety of ways; see Thomson [21], Clausius [2], and Planck [14]. However, at times more stringent restrictions than the second law are enforced, the reasons for the same being that the second law allows for too many possible candidates for the constitutive relations. One such more stringent requirement is the one introduced by Onsager; see Onsager [12], Prigogine [13], Glansdorff and Prigogine [5]. Onsager's requirement, which is often referred to as Onsager's Principle, does not have the same universality as the second law and is expected to hold for only special materials in special processes. Rajagopal and Srinivasa, in a series of recent papers, require that the rate of entropy production be maximal to choose a subset of constitutive relations from those that are non-negative. While Ziegler [23] had earlier appealed to such a requirement, he did not use it to obtain constitutive relations in the manner of Rajagopal and Srinivasa; see Rajagopal and Srinivasa [18] for a discussion of the differences between their approach and that of Ziegler. It might seem like the requirement demanded by Rajagopal and Srinivasa contradicts Onsager's Principle since they demand that the rate of entropy production is maximal while the latter demands that it be minimal in equilibrium. There is no contradiction whatsoever between the demands of Rajagopal and Srinivasa and that of Onsager as they refer to totally different circumstances.

2. Kinematics and basic equations. An abstract body B is a set that has a topological and measure theoretic structure (a detailed treatment of what is meant by a body can be found in Truesdell [22]). Let κ be a reference placer that maps the abstract body onto its configuration $\kappa(B)$ in a three dimensional Euclidean space. Let $\kappa_t(B)$ denote the configuration of the body B, at time t. By the motion χ of the body, we mean a one to one mapping at each instant of time t, that associates a particle $\mathbf{X} \in \kappa(B)$ with a particle $\mathbf{x} \in \kappa_t(B)$, i.e.,

$$\mathbf{x} = \chi_\kappa(\mathbf{X}, t).$$

Properties associated with the body can be defined on the basis of reference configuration (usually referred to as a Lagrangian representation, though it was introduced by Euler), the current configuration (usually referred to as Eulerian representation, though it was used earlier by D'Alembert and Bernoulli), or for that matter any other possible configuration the body can be placed in, i.e., a property φ can be defined through

$$\varphi = \varphi_\kappa(\mathbf{X}, t) = \varphi_{\kappa_t}(\mathbf{x}, t).$$

We shall use the following notation to represent the derivatives based on referential and current configurations:

$$\nabla \varphi = \frac{\partial \varphi_\kappa}{\partial \mathbf{X}}, \quad \operatorname{grad} \varphi = \frac{\partial \varphi_{\kappa_t}}{\partial \mathbf{x}}, \quad \frac{d\varphi}{dt} = \frac{\partial \varphi_\kappa}{\partial t}, \quad \frac{\partial \varphi}{\partial t} = \frac{\partial \varphi_{\kappa_t}}{\partial t}.$$

The gradient of the motion (usually called the deformation gradient) is defined through

$$\mathbf{F}_\kappa = \frac{\partial \chi_\kappa}{\partial \mathbf{X}}, \tag{2.1}$$

and the velocity $\mathbf{v}$ is defined through

$$\mathbf{v}(\mathbf{X}, t) = \frac{\partial \chi_\kappa(\mathbf{X}, t)}{\partial t} \implies \mathbf{v}(\mathbf{x}, t) = \mathbf{v}(\chi_\kappa^{-1}(\mathbf{x}, t), t). \tag{2.2}$$

Consequently,

$$\frac{d\varphi}{dt} = \frac{\partial \varphi}{\partial t} + \mathbf{v} \cdot \operatorname{grad} \varphi.$$

We also define the Cauchy-Green stretch tensors $\mathbf{B}_\kappa$ and $\mathbf{C}_\kappa$ through

$$\mathbf{B}_\kappa = \mathbf{F}_\kappa \mathbf{F}_\kappa^T, \qquad \mathbf{C}_\kappa = \mathbf{F}_\kappa^T \mathbf{F}_\kappa,$$

and the velocity gradient $\mathbf{L}$ and its symmetric part $\mathbf{D}$ through[1]

$$\mathbf{L} = \operatorname{grad}\mathbf{v}, \qquad \mathbf{D} = \frac{1}{2}\left(\mathbf{L} + \mathbf{L}^T\right).$$

It immediately follows from (2.1) and (2.2) that

$$\mathbf{L} = \frac{d\mathbf{F}_\kappa}{dt}\mathbf{F}_\kappa^{-1}. \tag{2.3}$$

Analogously to the above setting, we introduce the deformation gradient $\mathbf{F}_{\kappa_{p(t)}}$ as the measure of the deformation between $\kappa_{p(t)}(B)$ and $\kappa_t(B)$, and we set

$$\mathbf{B}_{\kappa_{p(t)}} = \mathbf{F}_{\kappa_{p(t)}}\mathbf{F}^T_{\kappa_{p(t)}}, \qquad \mathbf{C}_{\kappa_{p(t)}} = \mathbf{F}^T_{\kappa_{p(t)}}\mathbf{F}_{\kappa_{p(t)}}.$$

Finally, we use $\mathbf{G}$ to denote the measure of deformation between $\kappa(B)$ and $\kappa_{p(t)}(B)$. Obviously, $\mathbf{F}_\kappa = \mathbf{F}_{\kappa_{p(t)}}\mathbf{G}$. Motivated by (2.3), we define

$$\mathbf{L}_{\kappa_{p(t)}} = \frac{d\mathbf{G}}{dt}\mathbf{G}^{-1}$$

and we also set

$$\mathbf{D}_{\kappa_{p(t)}} = \frac{1}{2}\left(\mathbf{L}_{\kappa_{p(t)}} + \mathbf{L}^T_{\kappa_{p(t)}}\right).$$

The minimal kinematical definitions provided above suffice for our discussions here.

Any process undergone by the body has to meet the balance of mass, linear and angular momentum, and energy which are given by

$$\begin{aligned} \frac{d\rho}{dt} &= -\rho \operatorname{div}\mathbf{v}, \\ \rho\frac{d\mathbf{v}}{dt} &= \operatorname{div}\mathbf{T} + \rho\mathbf{b}, \\ \mathbf{T}^T &= \mathbf{T}, \\ \rho\frac{d\varepsilon}{dt} &= \mathbf{T}\cdot\mathbf{L} - \operatorname{div}\mathbf{q} + \rho r, \end{aligned} \tag{2.4}$$

where ρ denotes the density, $\mathbf{T}$ the Cauchy stress, $\mathbf{b}$ the specific body force, ε the specific internal energy, $\mathbf{q}$ the heat flux and r the radiant heating.

3. On the modeling of entropy producing process. Finally, we record the second law of thermodynamics which the body has to meet in every process[2]

$$\rho\frac{d\eta}{dt} + \operatorname{div}\left(\frac{\mathbf{q}}{\theta}\right) = \frac{\rho r}{\theta} + \rho\xi, \quad \xi \geq 0,$$

where η is the entropy, θ the temperature, $\mathbf{q}$ the heat flux, and ξ is the rate of entropy production. The second law expressed as above is different from the usual expression, where

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[1] It would be more appropriate to refer to $\mathbf{L}$ and $\mathbf{D}$ as $\mathbf{L}_{\kappa_t}$ and $\mathbf{D}_{\kappa_t}$, but we have dropped the suffix κ_t from which these measurements are made for the sake of convenience of notation.

[2] The second law, unlike the other laws that have been postulated, is inviolate. For instance, during radioactivity the balance of mass is violated. The balance laws, as stated, hold for classical mechanics and have to be restated for processes such as radioactivity, etc.

the last term in the above equation is omitted and the equality is replaced by an inequality. The above approach was used by Green and Nagdhi [6] and more recently by Rajagopal and Srinivasa (see the review articles by Rajagopal and Srinivasa [18, 19] for a discussion of its use in a variety of applications) to study the thermodynamic response of bodies, and we shall find it convenient to use the second law in the above form. It is common practice in continuum thermodynamics to obtain restrictions on the constitutive response functions by allowing the body to undergo arbitrary thermodynamic processes. Such a procedure assumes that the forms assumed for the constitutive response functions are valid in all these arbitrary processes, but this is not the case as the type of response being modeled might not be possible in all arbitrary processes. Thus, one would have to ensure that the process the body is subject to is compatible with the assumed form for the response function.

On combining the balance of energy with the above equation we obtain

$$\mathbf{T}\cdot\mathbf{L}-\rho\frac{d\varepsilon}{dt}+\rho\theta\frac{d\eta}{dt}-\frac{\mathbf{q}\cdot\operatorname{grad}\theta}{\theta}=\rho\theta\xi=:\zeta\geq 0, \tag{3.1}$$

where we refer to ζ as the rate of dissipation. Usually the rate of dissipation refers to the product of the density, temperature and the rate of entropy production associated with working being converted to heat, i.e., energy in its thermal form, but here we shall use it to mean the product of the density, temperature and the rate of entropy production associated with all forms of entropy production.

On introducing the specific Helmholtz potential

$$\psi=\varepsilon-\theta\eta,$$

we can rewrite the above equation as

$$\mathbf{T}\cdot\mathbf{L}-\rho\frac{d\psi}{dt}-\rho\eta\frac{d\theta}{dt}-\frac{\mathbf{q}\cdot\operatorname{grad}\theta}{\theta}=\zeta. \tag{3.2}$$

If one further assumes that the rate of entropy production ζ can be expressed additively (it is not necessary to make this assumption and one can deal with more complicated forms of entropy production) as

$$\zeta=\zeta_c+\zeta_d,$$

where ζ_c is the rate of entropy production due to conduction and ζ_d the rate of entropy production due for the various other entropy producing processes, and if we further assume that

$$\zeta_c=-\frac{\mathbf{q}\cdot\operatorname{grad}\theta}{\theta}\geq 0,$$

then we are left with (using also (2.4))

$$\mathbf{T}\cdot\mathbf{D}-\rho\frac{d\psi}{dt}-\rho\eta\frac{d\theta}{dt}=\zeta_d\geq 0.$$

Now, depending on the problem under consideration we will have to assume appropriate forms for the specific Helmholtz potential ψ, the rate of entropy production ζ_d, and the entropy η. It is worth observing a definite advantage of the above approach, compared to the usual assumption for the constitutive relation for the stress, namely that of making assumptions concerning two scalar functions instead of six scalar functions for the components of the stress. Let us consider, for the sake of illustration, isothermal[3] processes. In this case, equation (3.2) will further simplify to

$$\mathbf{T}\cdot\mathbf{D}-\rho\frac{d\psi}{dt}=\zeta_d\geq 0. \tag{3.3}$$

It is important to bear in mind that the specific Helmholtz potential was introduced to simplify the mathematical manipulations. Thus, in general, one has to prescribe constitutive relations for the specific internal energy and the specific entropy.

The crux of the idea is to maximize the rate of entropy production subject to the second law expressed in the form (3.3) (or in the more general case (3.1)) enforced as a constraint. If any other constraints such as those of incompressibility are to be enforced, then one has to incorporate them into the function which is being maximized by introducing the appropriate Lagrange multipliers. A few words concerning the rationale for the maximization are warranted. In an isolated system, the entropy of the system tends to a maximum and the system attains equilibrium. The quickest way for the system to attain the maximal value of entropy is to undergo processes that produce the maximal rate of entropy. Though this might seem a reasonable expectation for closed (isolated) systems we shall also require this to open systems. It is important to bear in mind that the above requirement of the maximal rate of entropy production is not a “principle” of thermodynamics, only a plausible and reasonable assumption. Ziegler [23] appealed to such an assumption, but from a different perspective; see Rajagopal and Srinivasa [18] for a detailed discussion of the distinction between Ziegler’s approach and the one advocated here.

It is also important to discuss another idea in thermodynamics that is used a great deal, especially when dealing with linear constitutive relations, namely the Onsager’s “principle”. This “principle” requires that the rate of entropy production be minimal. This requirement seems to contradict the requirement advocated earlier, but it is not so. The rate of entropy production is a Liapunov function. The procedure of maximizing the rate of entropy production picks one (or more) amongst numerous possible contenders for the choice of the rate of entropy production. Once a choice for the rate of entropy production is made, as it is a Liapunov function, it decreases to a minimal value with respect to time, and it is to this minimality Onsager’s “principle” refers to; see Onsager [12], de Groot and Mazur [3], Glansdorff and Prigogine [5]. A detailed discussion of the differences between the requirement of maximal rate of entropy production, for the choice of the rate of entropy production function, and Onsager’s “principle” can be found in the paper by Rajagopal and Srinivasa [20]. In fact, Rajagopal and Srinivasa [20] also provide a way for generalizing Onsager’s “principle” to non-linear phenomenological laws. For the special case of the rate of entropy production being quadratic in an appropriate variable, one obtains linear phenomenological relations such as Fourier’s law of heat conduction, Darcy’s law or Fick’s laws. The linear phenomenological relations satisfy Onsager’s relations and, as the rate of entropy productions is quadratic and a Liapunov function, it attains a minimum with time. However, when the rate of entropy production is not quadratic, one does not obtain linear phenomenological relations, and one cannot apply Onsagers’s relations. Rajagopal and Srinivasa [20] show how one can deal with a rate of entropy production that is not quadratic and leads to non-linear phenomenological relations. We shall not discuss these issues in detail here, but refer the interested reader to the paper by Rajagopal and Srinivasa [20], where all the relevant issues are discussed at length.

In the case of isothermal processes, we are then required to maximize the rate of entropy production ζ_d subject to the constraint (3.3). Thus, we maximize the function

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[3] A fully thermodynamic theory that allows for phase transformations, conduction, radiation, etc., can be found in the papers by Kannan et al. [7], Rao and Rajagopal [15] and Rajagopal and Srinivasa [19].

$$\Phi = \zeta_d + \lambda_1 \left(\zeta_d - \mathbf{T}\cdot\mathbf{D} + \rho \frac{d\psi}{dt} \right).$$

If in addition the body under consideration is incompressible, then we have to also take into consideration that

$$\mathbf{I}\cdot\mathbf{D} = \operatorname{div}\mathbf{v} = 0,$$

and thus we have to maximize

$$\Phi = \zeta_d + \lambda_1 \left(\zeta_d - \mathbf{T}\cdot\mathbf{D} + \rho \frac{d\psi}{dt} \right) + \lambda_2 \, \mathbf{I}\cdot\mathbf{D}.$$

3.1. An example: viscoelastic fluids. In order to illustrate the efficacy of the idea we apply our ideas to viscoelastic fluids. Suppose the specific Helmholtz potential and the rate of entropy production are given by

$$\psi = \frac{\mu}{2}\left(\mathbf{I}\cdot\mathbf{B}_{\kappa_{p(t)}} - 3\right),$$

and

$$\zeta_d = \eta \mathbf{D}\cdot\mathbf{B}_{\kappa_{p(t)}}\mathbf{D},$$

where μ and η are constants.

A straightforward application of the procedure (see Rajagopal and Srinivasa [17] for details) leads to the following representation for the Cauchy stress for the fluid

$$\mathbf{T} = -p\mathbf{I} + \mu \mathbf{B}_{\kappa_{p(t)}},$$

where

$$-\frac{1}{2}\left[\frac{d}{dt}\mathbf{B}_{\kappa_{p(t)}} - \mathbf{L}\mathbf{B}_{\kappa_{p(t)}} - \mathbf{B}_{\kappa_{p(t)}}\mathbf{L}^T\right] = \frac{\mu}{\eta}\left[\mathbf{B}_{\kappa_{p(t)}} - \lambda\mathbf{I}\right]$$

and

$$\lambda = \frac{3}{\left(\mathbf{I}\cdot\mathbf{B}^{-1}_{\kappa_{p(t)}}\right)}.$$

The above model is a generalization of the non-linear Maxwell model. Such a fluid stores energy like a non-linear neo-Hookean solid and dissipates it like a viscous fluid that takes into account the change in the underlying natural configuration. If one assumes that the elastic response is that of a linearized elastic solid, then one obtains the three dimensional generalization of the one dimensional model developed by Maxwell [11] in his seminal paper on the viscoelastic response of bodies. Different choices for the stored energy and the rate of entropy production will lead to different models for viscoelastic fluids. The reader can find how one could obtain models for different rate type viscoelastic fluids in Rajagopal and Srinivasa [17].

The above procedure does not apply to merely viscoelastic fluids. Appropriate choices for the stored energy and the rate of entropy production lead to all the known models for the inelastic response of solids in addition to leading to new useful models, including models within the context of finite deformations. An important point to observe is that the methodology automatically provides the "yield condition" and there is no need to make additional *ad hoc* assumptions for the "yield condition".

It is also worth mentioning that the procedure has been used to develop implicit constitutive models. For example, Málek and Rajagopal [8] develop models for incompressible fluids wherein the viscosity depends on the pressure (the mean normal stress) and the symmetric part of the velocity gradient (the model being a special implicit fluid model).

The above thermodynamic framework is able to describe a plethora of diverse phenomena of materials: viscoelasticity, traditional inelastic response, twinning, solid to solid transitions in shape memory alloys (see Rajagopal and Srinivasa [18, 19]), crystallization of polymers (Rao and Rajagopal [15]), shape memory polymers (Barot et al. [1]), granular materials (Málek and Rajagopal [9]), mixtures (Málek and Rajagopal [10]), etc. The procedure seems to have a lot of promise, but it is important to recognize that the procedure is not universal; there are probably responses of bodies that cannot be described within the purview of the above thermodynamic framework. However, the fact that it yields most of the known forms of materials response provides a certain amount of assurance as to its applicability.

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