Entropy Analysis for a Nonlinear Fluid with a Nonlinear Heat Flux Vector

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Abstract: Flowing media in both industrial and natural processes are often characterized as assemblages of densely packed granular materials. Typically, the constitutive relations for the stress tensor and heat flux vector are fundamentally nonlinear. Moreover, these equations are coupled through the Clausius–Duhem inequality. However, the consequences of this coupling are rarely studied. Here we address this issue by obtaining constraints imposed by the Clausius–Duhem inequality on the constitutive relations for both the stress tensor and the heat flux vector in which the volume fraction gradient plays an important role. A crucial result of the analysis is the restriction on the dependency of phenomenological coefficients appearing in the constitutive equations on the model objective functions.

Keywords: heat flux vector; nonlinear heat conduction; granular materials; nonlinear fluids; continuum mechanics

1. Introduction

Complex or nonlinear fluids such as coal slurries, polymers, and drilling fluids are fundamental to many industrial processes. They are also essential agents in natural disasters such as mudslides, avalanches, and pancake ice in high latitude regions. Flowing granular materials such as extractions of materials from mines, agricultural grain products, dry chemicals, sand, etc. can also be considered as nonlinear fluids (see [1–3]). Typically these materials are composed of many constituents; thus, they should theoretically be treated as multicomponent (or multiphase) materials. However, in many engineering applications, it is possible to treat these fluids as suspensions and model them as nonlinear—and sometimes non-homogeneous—single-component fluids. This avoids the necessity of specifying numerous phenomenological coefficients to characterize constituent interactions. The price of this simplification is nonlinear constitutive equations. Nonlinearity of these equations is compounded when thermodynamic considerations arise, as in fluidized bed combustion and drying of particulate media. An early example is Gudhe et al. [4], who studied the role of viscous dissipation in the flow of granular materials.

Many nonlinear fluids, including granular materials, exhibit nonlinear effects such as the normal stress differences when sheared. This was first observed by Reynolds [5] for wet sand, and he named this phenomenon dilatancy. It is possible to treat flowing dense granular materials as a continuum, indicating that the material properties may be expressed by continuous functions. Alternatively, the kinetic theory of gases [6] can be used. It is noteworthy that a thermodynamic analysis of the constitutive equations is rarely performed for many of these models.
One of the first thermo-mechanical continuum models developed for granular materials was by Goodman and Cowin [7,8]. This work was later modified and improved upon by others [9,10]. The theory of Goodman and Cowin is applicable to situations where the stress levels are smaller than 10 psi. In addition, they assumed that the pneumatic effects can be neglected; that is, the effects of the interstitial gas were ignored. They assumed that the material properties of the ensemble are continuous functions of position and postulated the existence of new concepts such as “balance of equilibrated force” or “balance of equilibrated inertia”, etc. They also introduced a new form of the entropy inequality. Based on the restrictions imposed by the Clausius-Duhem inequality, the principle of material frame-indifference, incompressibility of the grains, and the assumption that the constitutive parameters depend on the temperature gradient, velocity gradient, and gradient of the volume fraction, the Goodman and Cowin theory becomes so constrained that there are few solutions of interest. In this paper, we obtain restrictions imposed by the Clausius-Duhem inequality on the nonlinear stress tensor proposed by Rajagopal and Massoudi [12], and the nonlinear heat flux vector derived by Massoudi [13]. In most other thermomechanical applications in granular materials, only the traditional linear Fourier’s law is used (see [8]). The application of the entropy law to thermomechanical and physical processes is an active area of research (see [14–17]).

The next section reviews the governing equations for granular materials. This includes the energy equation and a statement of the entropy inequality equation appropriate for this study. Section 3 discusses the constitutive equations used in the analysis. For the stress tensor, we use the equation derived by Rajagopal and Massoudi [12], and for the heat flux vector we use the constitutive equations derived by Massoudi [18,19] and consider the consequences of the entropy inequality (the Clausius-Duhem inequality) on the material properties. Section 4 develops the constraints imposed by the entropy inequality. The report concludes with a summary of our results and a commentary on further work in this area.

2. The Governing Equations

Through the homogenization process, we can think of the granular media as a single phase continuum [20]. If there are no chemical and electromagnetic effects, the basic governing equations are the conservation of mass:

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \mathbf{v}) = 0$$  \hspace{1cm} (1)

the balance of linear momentum:

$$\rho \frac{d \mathbf{v}}{dt} = \text{div} \mathbf{T} + \rho \mathbf{b}$$  \hspace{1cm} (2)

and the energy equation:

$$\rho \frac{d \varepsilon}{dt} = \mathbf{T} \cdot \mathbf{L} - \text{div} \mathbf{q} + \rho r.$$  \hspace{1cm} (3)

In these equations, \( \frac{d}{dt} \) is the material or total time derivative given by

$$\frac{d(.)}{dt} = \frac{\partial(.)}{\partial t} + \mathbf{v} \cdot [\nabla(.)]$$  \hspace{1cm} (4)

\( \mathbf{v} \) is the velocity vector, \( \rho \) is the density, \( \mathbf{T} \) is the Cauchy stress tensor, \( \mathbf{b} \) is the body force, \( \varepsilon \) is the specific internal energy, \( \mathbf{q} \) is the heat flux vector, \( r \) is the radiant heating, and \( \mathbf{L} \) is the velocity gradient.
The equation for the angular momentum (in the absence of couple stresses) indicates that the stress is symmetric.

The entropy equation, in a general form, is given by [21]:

\[
\rho \frac{d\eta}{dt} + \text{div} \, \phi - \rho s \geq 0, \tag{5}
\]

where \( \eta(x,t) \) is the specific entropy density, \( \phi(x,t) \) is the entropy flux, and \( s \) is the (external) entropy supply density. Specifying \( \phi = \frac{q}{\theta} \) and \( s = \frac{r}{\theta} \), where \( \theta \) is the absolute temperature, reduces (5) to the standard Clausius–Duhem inequality

\[
\rho \frac{d\eta}{dt} + \text{div} \, \frac{q}{\theta} - \rho \frac{r}{\theta} \geq 0. \tag{6}
\]

Using the mass and energy conservation Equations (1) and (3), an alternate form of the entropy Equation (5) is

\[
\rho \frac{d\eta}{dt} = -\text{div} \, \phi + \rho s + \dot{\gamma}_s \tag{7}
\]

where

\[
\dot{\gamma}_s = -\frac{1}{\theta^2} \nabla \theta \cdot \mathbf{q} + \frac{1}{\theta} \text{tr}(\mathbf{T}) \geq 0. \tag{8}
\]

See [22] for details.

In Coleman and Noll’s [23] approach, this inequality must be satisfied for all admissible processes, regardless of the materials and corresponding constitutive equations. Numerous papers have applied this idea in developing constitutive relations. See, for example, [2,8,24–27]. Equation (8) is appropriate for obtaining restrictions on the constitutive parameters. Constitutive relations are required for \( T \), \( q \), \( \varepsilon \), and \( r \).

3. The Constitutive Equations

The basic issue is whether a single constitutive relation for \( T \) and a single constitutive relation \( q \) are sufficient to describe the behavior of granular materials and situations encountered in many engineering applications. To study granular materials, we need to consider several things. For example, granular materials can take the shape of the container; they cannot be liquid, since they can be piled into heaps, and they will not expand to fill the vessel as a gas would.

We view granular materials as assemblages of densely packed and flowing solid particles. We treat this as a fluid-like material with variable density. In this case, the volume fraction \( \phi(x,t) \) is an important parameter. Even though there are distinct solid particles with a certain diameter or shape, the volume fraction homogenizes the assemblage. See Collins [28] for a discussion of the homogenization of granular media microstructure.

The function \( \phi(x,t) \) is a kinematical variable where \( 0 \leq \phi(x,t) \leq \phi_{\text{max}} \leq 1 \) in a granular system. In practice, \( \phi \) has a maximum value, generally designated as the maximum packing fraction. The density, \( \rho \), is related to the particle density \( \rho_s \) and \( \phi \) through \( \rho(x,t) = \phi(x,t) \rho_s \), where \( \rho_s \) is the density of the individual particle which is assumed to be constant. We need to mention that through this process of homogenization a great deal of information about the (micro-)structure of the granular materials is lost: the size of the particles, the shape, texture, patterns, surface conditions, etc. (see [29–34]). That is, we do not consider the size of the particles or the (relative) scale of the problem which may enter the formulation through concepts such as representative elemental volume (REV). We treat the material as a continuum. Clearly, this is one of the limitations of the classical continuum theories (which we have used in this paper). To overcome some of these deficiencies, more advanced continuum theories such as micromechanical theories [35], micropolar theories [36], or fabric tensor approach [37,38], etc. can be used. Alternatively, one can use statistical or numerical simulations [39–41].
Here we use the stress relation proposed by Rajagopal and Massoudi [12], where \( T \) is a function of the gradient of the density (or volume fraction) and \( D \) is the symmetric part of the velocity gradient \( [D = 1/2(\nabla v + (\nabla v)^T)] \). Employing the language of matrix notation [42], this is written as

\[
T = [\beta_0(\phi) + \beta_1(\phi)\nabla \phi \cdot \nabla \phi + \beta_2(\phi) tr D][1 + \beta_3(\phi)D + \beta_4(\phi) \nabla \phi \otimes \nabla \phi + \beta_5(\phi) D^2],
\]

where

\[
\beta_0 = -f \phi, \quad \beta_1(\phi) = \beta_1^*(1 + \phi + \phi^2), \quad \beta_2(\phi) = \beta_2^*(\phi + \phi^2)
\]

\[
\beta_3(\phi) = \beta_3^*(\phi + \phi^2), \quad \beta_4(\phi) = \beta_4^*(1 + \phi + \phi^2), \quad \beta_5(\phi) = \beta_5^*(\phi + \phi^2)
\]

(10)

In (9), the \( \cdot \) denotes the inner product, \( \otimes \) denotes the tensor (or the outer) product of two vectors, and \( \nabla \) is the gradient operator. Equation (10) can be viewed as a Taylor series approximation for the material parameters. Of course, the \( \beta_i \) moduli are functions of appropriate tensor invariants identified below. Rajagopal and Massoudi [12] regarded \( \beta_0 \) and \( \beta_2 \) as analogous to the pressure and second viscosity, respectively, of a compressible fluid and \( \beta_3 \) as the viscosity. They also interpreted \( \beta_1 \) and \( \beta_4 \) as material properties related to how the granular materials are distributed. Rajagopal et al. [43] established criteria for the existence of solutions when \( \beta_1 + \beta_4 > 0 \). Additionally, \( \beta_5 \) accounts for the second-order effects, sometimes referred to as the cross-viscosity in the Reiner–Rivlin model; see [44–46]. Note that in this scheme, the stress vanishes as \( \phi \to 0 \). Later, Rajagopal and Massoudi [12] and Rajagopal et al. [43] indicate that

\[
\begin{align*}
\beta_0 &< 0 \\
\beta_1 + \beta_4 &> 0
\end{align*}
\]

(11)

where the first condition implies that compression causes densification of the material and the second condition is necessary if solutions are to exist. Note that (9) has certain similarities to the stress tensor for the Korteweg fluids, where density gradient plays an important role (see [47,48]). Additional details along with some applications of this model are given in [13,20,49].

For \( q \), we follow Massoudi [18,19]. From mechanics-based arguments, he suggested that the heat flux vector depends not only on the temperature gradient, but also on the motion and the density (or volume fraction) gradient. He derived the following equation:

\[
q = a_1 n + a_2 m + a_3 Dn + a_4 Dm + a_5 D^2 n + a_6 D^2 m.
\]

(12)

Here \( n = \nabla \theta \) is the temperature gradient and \( m \) is the gradient of the volume fraction. The moduli \( a_1 - a_6 \) and \( \beta_0 - \beta_5 \) are functions of the 14 scalar invariants. These are

\[
\phi, \theta, m \cdot m, n \cdot n, m \cdot n, tr D, tr D^2, tr D^3, n \cdot Dn, n \cdot D^2 n, m \cdot Dm, m \cdot D^2 m, n \cdot Dm, n \cdot D^2 m.
\]

(13)

There are several versions of the invariants of \( tr D, tr D^2, \) and \( tr D^3 \); [42,50]. Later, it will be convenient to express these invariants in terms of the principal values of \( D \). That is,

\[
\begin{align*}
tr D &= D_1 + D_2 + D_3 \\
tr D^2 &= D_1^2 + D_2^2 + D_3^2 \\
tr D^3 &= D_1^3 + D_2^3 + D_3^3
\end{align*}
\]

(14)

Massoudi [18,19] set \( a_1 = -k \), where \( k \) is the thermal conductivity, \( m = \nabla \phi \), and \( n = \nabla \theta \). Equation (12) can then be re-written as:

\[
q = (a_1 1 + a_3 D + a_5 D^2) \cdot \nabla \theta + (a_2 1 + a_4 D + a_6 D^2) \cdot \nabla \phi.
\]

(15)
A notable feature of (15) is that it predicts a heat flux caused by the volume fraction gradient. Similar expressions were obtained by Jaric and Golubovic [51].

Yang et al. [20] used (9) and (15) to study the heat transfer in granular materials. However, they did not consider the Clausius–Duhem inequality. Using this inequality and the additional restriction that \( \beta_5 = 0 \), Massoudi and Kirwan [52] showed that non-negative entropy production required nonlinear constraints between the moduli and appropriate gradients of the constitutive functions. Apparently there is an elementary connection between the existence of solutions to (1)–(3) and (6). Recently, Yang and Massoudi (2017) [53] have studied flow and heat transfer of a nonlinear fluid, modeled by (9) and (12), on an inclined plane. They assumed that the (material) coefficients appearing in these constitutive relations are functions of the volume fraction, and they used different thermal boundary conditions, including radiation boundary condition, at the free surface. Their results indicate that the temperature profiles show strong non-linearity due to the nonlinear effects of volume fraction.

As observed above in both (9) and (15), volume fraction gradients produce fluxes of heat and momentum in the absence of thermal and velocity gradients. However, these equations do not provide any information on the magnitudes and signs of these fluxes relative to those of the traditional Fourier heat flux and non-Newtonian fluid models. To address this matter, we appeal to constraints imposed by (8). Here we follow the tradition of Coleman and Noll [23] and perform a second law analysis of the nonlinear stress and heat flux vector constitutive equations (9), (15) to obtain such constraints.

4. Entropy Analysis

Coleman and Noll [23] define an admissible process as a thermodynamic process that is compatible with the constitutive relations under consideration. The second law of thermodynamics (or the Clausius–Duhem inequality) determines which thermodynamic processes are admissible. Hence the Clausius–Duhem inequality provides restrictions on the constitutive relations for all admissible processes. We use the same procedure to find sufficient conditions for the material coefficients in the Cauchy stress tensor (9) and heat flux (15) constitutive equations for admissible processes.

Putting (9) and (15) in (8) gives

\[
\dot{\gamma}_s = -\frac{1}{\theta^2} \left[ a_1 (\nabla \theta) \cdot \nabla \theta + \nabla \theta \cdot (a_3 D + a_5 D^2) \cdot \nabla \theta + a_2 \nabla \theta \cdot \nabla \phi + \nabla \theta \cdot (a_4 D + a_6 D^2) \cdot \nabla \phi \right] \\
+ \frac{1}{\theta} [\beta_0 + \beta_1 \nabla \phi \cdot \nabla \phi + \beta_2 tr D] tr D + \beta_3 tr D^2 + \beta_4 tr ((\nabla \phi \otimes \nabla \phi) D) + \beta_5 tr (D^3) \geq 0.
\]

(16)

Second law analysis of linear theories is straightforward. This inevitability produces constraints on the signs of phenomenological coefficients. However, as noted by [52], second law constraints for nonlinear theories produce nonlinear relations involving both phenomenological coefficients and related gradients. Evidently, second law constraints for nonlinear models impact the stability of solutions, as they set limits on gradients of the field variables. Apparently, this facet of nonlinear model solutions has not been widely studied.

Equation (16) involves traces of tensor products and inner products of a tensor and two vectors. Moreover, the constitutive coefficients \( \beta_j \) and \( a_j \) are functions of the 14 invariants listed in (13).

The calculation of constraints imposed by the second law is simplified by the use of these invariants.

We now consider inequality constraints arising from admissible thermodynamic processes. First, consider only the case of heat conduction; i.e., \( \nabla \phi = D = 0 \). Then, (16) reduces to

\[
\dot{\gamma}_s = -\frac{1}{\theta^2} a_1 |\nabla \theta|^2 \geq 0.
\]

(17)

This is simply the well-known constraint anticipated by [18,19], that the thermal conductivity \( a_1 \leq 0 \). In the general case, the thermal conductivity is a second-order tensor. In that case, Wang [54] showed that \( a_1 = \lambda_0 I + \lambda_1 D + \lambda_2 D^2 \) with the \( \lambda_i \) functions of the parameters listed in (13).
Next, consider second law constraints for the special case where $\nabla \theta = \nabla \phi = 0$. Then, (16) reduces to the dissipation of a non-Newtonian fluid:

$$\gamma_s = \theta^{-1} \left[ (\beta_0 + \beta_2 \text{tr} \mathbf{D}) \text{tr} \mathbf{D} + \beta_3 \text{tr} \mathbf{D}^2 + \beta_5 \text{tr} \mathbf{D}^3 \right] \geq 0. \quad (18)$$

Each term on the right hand side (RHS) of (18) can be rendered non-negative by requiring

$$\begin{align*}
\beta_0 &= \beta_{00} (\text{tr} \mathbf{D})^p \\
\beta_2 &= \beta_{20} \\
\beta_3 &= \beta_{30} (\text{tr} \mathbf{D}^2)^q \\
\beta_5 &= \beta_{50} (\text{tr} \mathbf{D}^3)^r.
\end{align*} \quad (19)$$

Here $p$ and $r$ are odd integers, but not 0, and the coefficients $\beta_{ij}$ are non-negative functions of the remaining invariants listed in (13). Navier–Stokes fluids arise as a special case when $\beta_5 = 0$ and $q = 0$. From (14), one readily obtains the identity

$$\text{tr} \mathbf{D}^2 \equiv \frac{1}{3} \left[ (\text{tr} \mathbf{D})^2 + (D_1 - D_2)^2 + (D_2 - D_3)^2 + (D_3 - D_1)^2 \right].$$

Then, (18) is expressed as

$$\theta^{-1} \left\{ -\beta_{00} \text{tr} (\mathbf{D})^{p+1} + \left[ \beta_{20} + \left( \frac{\beta_{30}}{3} \right) \right] (\text{tr} \mathbf{D})^2 + \left( \frac{\beta_{30}}{3} \right) \left[ (D_1 - D_2)^2 + (D_2 - D_3)^2 + (D_3 - D_1)^2 \right] \right\} \geq 0. \quad (20)$$

Clearly, both $\beta_{30} \geq 0$ and $3\beta_{20} + \beta_{30} \geq 0$, a well established result.

It has now been established that (17) and (18) hold for two admissible processes that involve just heat and mechanical energy. This is a simple extension of classical results to granular materials but with no interactions between the volume fraction, temperature, and velocity fields. Inclusion of other admissible processes should not impact these basic inequalities. However, the constitutive model discussed here is highly nonlinear, and it could be argued that the gradients of $\phi$ and $\theta$ could conspire with $\mathbf{D}$ to produce significant sinks for entropy production; i.e., the remaining terms in (16) could be negative. Nevertheless, even in this most extreme situation, the second law still rules. If these processes are sinks for entropy production, their combined total effect must be less than the production of entropy by purely thermal and mechanical processes.

Recall that in linear theories of non-equilibrium thermodynamics, the second law constraint requires that the matrix of phenomenological coefficients be positive semi-definite. See [14] for a recent discussion of this approach and an application to a mixture of two fluids at different temperatures. Physically, this means that the entropy production associated with the primary forces is larger than that associated with the cross-gradient mechanisms. Apparently, this notion holds for nonlinear theories as well.

Moreover, nonlinear theories such as those developed here permit the possibility that every term in (16) is non-negative. This is achieved by specifying

$$\begin{align*}
a_2 &= -a_{20} (\nabla \theta \cdot \nabla \phi)^{p_1} \\
a_3 &= -a_{30} (\nabla \theta \cdot \mathbf{D} \nabla \theta)^{p_3} \\
a_4 &= -a_{40} (\nabla \theta \cdot \mathbf{D} \nabla \phi)^{p_4} \\
a_5 &= -a_{50} (\nabla \theta \cdot \mathbf{D}^2 \nabla \phi)^{p_5} \\
a_6 &= -a_{60} (\nabla \theta \cdot \mathbf{D} \nabla \phi)^{p_6} \\
\beta_1 &= \beta_{10} (\nabla \phi \cdot \nabla \phi)^{p_1} \\
\beta_4 &= \beta_{40} (\nabla \phi \cdot \mathbf{D} \nabla \phi)^{p_2}.
\end{align*} \quad (21)$$
Here, each $p_i$ is an odd integer. Of course, the coefficients on the RHS of (21) are functions of the model invariants. Equations (21) require these functions to be non-negative. Then, each term in (16) is positive and the inequality is satisfied identically.

5. Discussion

In this paper, we have used the Clausius–Duhem inequality to obtain certain restrictions on the constitutive relations first derived by Rajagopal and Massoudi (1990) [12] (for the stress tensor) and Massoudi (2006a,b) [18,19] (for the heat flux vector). These constitutive relations are properly frame-indifferent models (see [48,55]). Using the second law, we established constraints on the constitutive equations for a fluid model that includes nonlinear processes arising from flow, heat flux, and the redistribution of the particles. This extends an earlier analysis by Massoudi and Kirwan [52]. Here we specifically show that the second law constraint is satisfied when the phenomenological coefficients are appropriate functions of the gradients of the field variables. Hopefully, the analysis here will be useful in experiments to determine values of phenomenological parameters.

We anticipate that these results will provide a useful extension for certain nonlinear models used in non-Newtonian fluid mechanics; for example, the Reiner–Rivlin or the second-grade fluids. For example, the stress tensor model used in this paper can be extended to have a structure similar to the generalized second-grade fluid model which has been used for modelling the creep of ice and the flow of coal slurries, etc., that can describe shear thinning/thickening and exhibit normal stress effects (see [56–60]).

As an example of the theory developed here, consider (15). With no second law constraints, this equation predicts that a heat flux can be generated even in the absence of a temperature gradient. That is, if $\nabla \theta = 0$, (15) reduces to

$$q = (a_2 \mathbf{1} + a_4 \mathbf{D} + a_6 \mathbf{D}^2) \nabla \phi.$$  

(22)

However, in (21) we showed that a sufficient condition imposed by the second law is that $a_2, a_4, \text{ and } a_6$ should include terms involving $\nabla \theta$:

$$a_2 = -a_{20}(\nabla \phi \cdot \nabla \theta)^{p_2}$$

$$a_4 = -a_{40}(\nabla \theta \cdot \mathbf{D} \nabla \phi)^{p_4}$$

$$a_6 = -a_{60}(\nabla \theta \cdot \mathbf{D}^2 \nabla \phi)^{p_6}.$$  

(23)

Clearly, these coefficients are 0 when $\nabla \theta = 0$. Yang and Massoudi [53] applied this condition with $p_2 = p_4 = p_6 = 1$ to their model. Consequently, in that study, heat was not generated by the gradient of the volume fraction unless there was also a temperature gradient.

An important consequence of the analysis presented here is that for nonlinear materials the second law constrains the magnitudes of the gradients of the field variables. This suggests that thermodynamic considerations will play an important role in the stability of solutions to specific problems. Ramifications of this have yet to be explored. Finally, since our models for the stress tensor and the heat flux vector are nonlinear, it is important to do stability analyses and consider the uniqueness and existence of solutions. This is left for future studies (see [61–63]).

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