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Article

Thermodynamics of Composition Graded Thermoelastic Solids

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Abstract: We propose a thermodynamic model describing the thermoelastic behavior of composition graded materials. The compatibility of the model with second law of thermodynamics is explored by applying a generalized Coleman-Noll procedure. We calculate the speeds of coupled first- and second-sound pulses, propagating either through nonequilibrium or through equilibrium states. We characterize several different types of perturbations, depending on the value of the material coefficients. Under the assumption that the deformation of the body can produce changes in its stoichiometry, altering locally the material composition, the possibility of propagation of pure stoichiometric waves is pointed out. Thermoelastic perturbations generated by the coupling of stoichiometric and thermal effects are analyzed as well.

Keywords: Composition graded materials; thermoelastic solids; generalized Coleman-Noll procedure; thermoelastic-wave propagation

1. Introduction

Functionally graded systems are new materials whose behavior varies along some given directions inside the system [1,2]. Such a property can be used to optimize their performances in several applications [3]. For example, at nanoscale graded alloys of the type $\text{Si}_c\text{Ge}_{1-c}$, with the stoichiometric variable $c \in [0; 1]$ changing along a given direction, are used in the design of thermal diodes [4–8], or to enhance the efficiency of thermoelectric energy conversion [9,10].

The study of elastic and thermal properties of composition graded systems is a promising field of research, since applied mechanical stresses may influence their thermal conductivity, providing so a wider degree of control of their thermal behavior [6,11,12].

In the present paper we focus on the thermodynamics of composition graded thermoelastic systems, by postulating a system of balance equations modeling their behavior and investigating its compatibility with second law of thermodynamics [13]. A set of thermodynamic restrictions, giving necessary and sufficient conditions for the thermodynamic compatibility, is derived by applying a generalized Coleman-Noll procedure for the exploitation of the entropy inequality [14–16].

Furthermore, we investigate the propagation of thermoelastic waves. We derive the necessary and sufficient conditions for the propagation of the acceleration waves, and analyze them in different situations. In such a way we get useful information on what could be obtained from experimental thermoelastic-perturbation measurements. The outline of the paper is the following.

In Section 2, we postulate the model equations.

In Section 3, we prove the compatibility of the equations above with the second law of thermodynamics, and derive the form of the Cauchy stress tensor and of the specific entropy.

In Section 4, we calculate the speeds of propagation of thermoelastic pulses both in equilibrium states, and in nonequilibrium states.

In Section 5, a discussion of the obtained results is given, and possible developments of the theory are analyzed.

2. The Model Equations

In nanosystems nonlinear effects influence the propagation of thermomechanical disturbances [17–20]. Then, new mathematical models beyond the classical thermoelasticity [17,21–24] are necessary. In fact, at nanoscale even small differences of temperature or displacement may produce strong gradients, in such a way that linear constitutive equations are no longer valid. Furthermore, when a transient thermal pulse is applied to one end of a system, it arrives at the opposite end in a very short time, i.e., before of reaching its equilibrium value. Thus, when studying thermoelastic nanosystems, a nonlinear heat-transport equation allowing propagation of thermomechanical disturbances with a finite speed is also needed. Due to the considerations above, here we assume that:

- the heat-transport equation couples thermal and elastic effects;
- the thermal conductivity and the relaxation time depend on the deformation too;
- the stoichiometric variable $c \in [0, 1]$ and its gradient enter the state space;
- the variable c can change with time, due to the changes of volume produced by the deformation.

In practice, we regard c as a scalar internal variable, whose dependency on the points of the system is known when the process starts, but changes as a consequence of thermoelastic deformation. As usual for internal variables, the kinetic equation giving the evolution of c is defined on the state space.

In the following, we will focus on small deformations, so that our model equations will remain within the frame of linear thermoelasticity, although, due to the nonlinear dependency of the constitutive equations on the strain and on the heat flux, the full model is nonlinear.

Due to the considerations above, in the present research we develop a mathematical model which lays on the following set of equations

$$\rho \ddot{u}_i - T_{ij,j} = 0, \quad (1a)$$

$$\rho \dot{\varepsilon} - T_{ij} \dot{E}_{ij} + q_{i,i} = 0, \quad (1b)$$

$$\tau_q (1 + a_\tau E_{hk} E_{hk}) \dot{q}_i + q_i + \bar{\kappa} (1 + a_\kappa E_{hk} E_{hk}) \varepsilon_{,i} - \frac{2\tau_Q}{\rho \varepsilon} q_{j,i} q_j - \tau_E \dot{E}_{ij} q_j - a_c c_{,i} = 0, \quad (1c)$$

$$\dot{c} = H(\Sigma), \quad (1d)$$

wherein:

- the indices i, j, h, k , range from 1 to 3;
- u_i is the local displacement vector with respect to a reference material configuration;
- T_{ij} is the Cauchy stress tensor;
- ρ is the mass density;
- ε is the internal energy per unit mass;
- q_i is the local heat flux;
- E_{ij} is the strain tensor (i.e., $E_{ij} = \frac{u_{i,j} + u_{j,i}}{2}$);
- \bar{c} is the specific heat, related to the internal energy per unit of mass by the relation $\varepsilon = \bar{c}\vartheta$, where $\vartheta \equiv \frac{\partial \varepsilon}{\partial s}$ is the absolute temperature;
- κ is the thermal conductivity;
- τ_q is a material parameter, depending on ε and c , such that $\tau_q (1 + a_\tau E_{hk} E_{hk})$ represents the total relaxation time of the heat flux (i.e., the time elapsed between the application of a difference of temperature and the appearance of a heat flux);
- τ_E is the relaxation time of the strain (i.e., the time elapsed between the application of a stress and the appearance of a deformation);
- τ_Q is the relaxation time of the heat carriers, i.e., the quantity $\frac{\ell}{\bar{v}}$, with ℓ as the mean free path and \bar{v} as the mean speed of the heat carriers (phonons, electrons, holes);
- a_τ and a_κ and a_c are material functions depending on ε and c ;

- Σ denotes the set of the elements of the state space, which will be specified better below.

We note that in Equation (1c) the strain influences both the total relaxation time, and the heat conductivity, according to the results in [25,26]. We also note that we are supposing $\tau_q \neq \tau_Q$. Indeed, at nanoscale size effects on the relaxation of the heat flux become as more important as the characteristic dimension decreases. Consequently, in the present approach, the total relaxation time of the heat flux (namely, τ_q) does not coincide with the relaxation time of the heat carriers (namely, τ_Q) [27,28]. At macroscopic scale, instead, where the size effects are absent, τ_q reduces to τ_Q , and the strain does not influence the evolution equation of the heat flux, so that $\tau_E = a_\tau = a_\kappa = 0$. Then, Equation (1c) reduces to

$$\tau_Q \dot{q}_i + q_i + \frac{\kappa}{\bar{c}} \varepsilon_{,i} - \frac{2\tau_Q}{\rho \bar{c}} q_{j,i} q_j = 0, \quad (2)$$

which is the nonlinear Maxwell-Cattaneo equation derived in [29,30].

3. Thermodynamic compatibility

In the present section we postulate the following state space

$$\Sigma \equiv \{ \varepsilon, \varepsilon_{,k}, c, c_{,k}, q_h, q_{h,k}, E_{hk} \}, \quad (3)$$

and investigate the compatibility of Equation (1) with the second law of thermodynamics. In order to better specify the theoretical framework of the present theory, we observe that the heat flux, whose evolution is ruled by Equation (1c), enters the state space. We observe also that the gradients of the independent thermodynamic variables belong to Σ . Such considerations allow us to conclude that our model is within the frame of Extended Irreversible Thermodynamics (EIT), a weakly nonlocal thermodynamic theory in which the dissipative fluxes have the rank of independent thermodynamic variables [13,17,21,22,31]. According to the basic tenets of EIT, the gradients of the basic unknown fields can enter the state space.

Second law of thermodynamics imposes that the local rate of entropy production

$$\sigma^{(s)} = \rho \dot{s} + J_{i,i}^{(s)}, \quad (4)$$

wherein s means the local entropy per unit mass, and $J_i^{(s)}$ is the entropy flux, has to be nonnegative for arbitrary thermodynamic processes [13]. When the dependency of s and J_i on the state variables has been made explicit, the inequality (4) reads

$$\rho \left(\frac{\partial s}{\partial \varepsilon} \dot{\varepsilon} + \frac{\partial s}{\partial \varepsilon_{,i}} \dot{\varepsilon}_{,i} + \frac{\partial s}{\partial q_i} \dot{q}_i + \frac{\partial s}{\partial q_{j,i}} \dot{q}_{j,i} + \frac{\partial s}{\partial E_{ij}} \dot{E}_{ij} + \frac{\partial s}{\partial c} \dot{c} + \frac{\partial s}{\partial c_{,i}} \dot{c}_{,i} \right) + \frac{\partial J_i}{\partial \varepsilon} \varepsilon_{,i} + \frac{\partial J_i}{\partial \varepsilon_{,j}} \varepsilon_{,ji} + \frac{\partial J_i}{\partial q_j} q_{j,i} + \frac{\partial J_i}{\partial q_{j,k}} q_{j,ki} + \frac{\partial J_i}{\partial E_{hk}} E_{hk,i} + \frac{\partial J_i}{\partial c} c_{,i} + \frac{\partial J_i}{\partial c_{,k}} c_{,ki} \geq 0. \quad (5)$$

The inequality (5) will be exploited by a generalization of the classical Coleman-Noll procedure [14–16], which consists in substituting in (5) the evolution Equations (1b)–(1d), and their gradients, up to the order of the gradients of ε , q_i and c entering the state space (see also [32,33] for more details on the generalized exploitation procedures of the entropy inequality). Doing that, we consider of the first order of magnitude the gradients of the independent thermodynamic variables. Then, in a first order

approximation, we neglect the terms containing the products of such gradients as, for instance, the term $q_h E_{hk,i} q_{k,i}$. In this way, we obtain the following inequality

$$\begin{aligned} & \left(\frac{\partial s}{\partial q_i} \frac{\tau_E q_j}{\tau_q A_q} + \frac{\partial s}{\partial q_{i,k}} \tau_E q_{j,k} - \frac{\partial s}{\partial \varepsilon} T_{ij} + \varrho \frac{\partial s}{\partial E_{ij}} \right) \dot{E}_{ij} + \\ & \left(\frac{\partial s}{\partial c_{,k}} \frac{\partial H}{\partial \varepsilon_{,i}} - \frac{\partial s}{\partial q_{i,k}} \frac{\kappa A_K}{\bar{c} \tau_q A_q} - \frac{\partial J_i}{\partial \varepsilon_{,k}} \right) \varepsilon_{,ki} + \\ & \left(\frac{\partial s}{\partial c_{,k}} \frac{\partial H}{\partial q_{j,i}} + \frac{\partial s}{\partial q_{i,k}} \frac{2\tau_Q q_j}{\tau_q \varrho \varepsilon A_q} - \frac{\partial s}{\partial \varepsilon_{,i}} \delta_{jk} + \frac{\partial J_i}{\partial q_{j,k}} \right) q_{j,ki} + \\ & \left(\frac{\partial J_i}{\partial E_{hk}} + \frac{\partial s}{\partial c_{,i}} \frac{\partial H}{\partial E_{hk}} \right) E_{hk,i} + \\ & \left(\frac{\partial s}{\partial q_{i,k}} \tau_E q_j - \frac{\partial s}{\partial \varepsilon_{,i}} T_{jk} \right) \dot{E}_{ij,k} + \\ & \left(\frac{\partial s}{\partial c_{,k}} \frac{\partial H}{\partial c_{,i}} + \frac{\partial s}{\partial q_{i,k}} a_c + \frac{\partial J_i}{\partial c_{,k}} \right) c_{,ki} + f(\Sigma) \geq 0, \end{aligned} \quad (6)$$

wherein $A_K \equiv 1 + a_\kappa E_{hk} E_{hk}$, $A_q \equiv 1 + a_\tau E_{hk} E_{hk}$, δ_{jk} is the identity tensor, and $f(\Sigma)$ is a scalar-valued function defined on the state space whose expression is omitted for the sake of concision.

Theorem 1. *The inequality (6) is satisfied whatever the thermodynamic process is if, and only if, the following thermodynamic restrictions hold*

$$\left\langle \frac{\partial s}{\partial q_i} \frac{\tau_E q_j}{\tau_q A_q} + \frac{\partial s}{\partial q_{i,k}} \tau_E q_{j,k} - \frac{\partial s}{\partial \varepsilon} T_{ij} + \varrho \frac{\partial s}{\partial E_{ij}} \right\rangle = 0, \quad (7a)$$

$$\left\langle \frac{\partial s}{\partial c_{,k}} \frac{\partial H}{\partial \varepsilon_{,i}} - \frac{\partial s}{\partial q_{i,k}} \frac{\kappa A_K}{\bar{c} \tau_q A_q} - \frac{\partial J_i}{\partial \varepsilon_{,k}} \right\rangle = 0, \quad (7b)$$

$$\left\langle \frac{\partial s}{\partial c_{,k}} \frac{\partial H}{\partial q_{j,i}} + \frac{\partial s}{\partial q_{i,k}} \frac{2\tau_Q q_j}{\tau_q \varrho \varepsilon A_q} - \frac{\partial s}{\partial \varepsilon_{,i}} \delta_{jk} + \frac{\partial J_i}{\partial q_{j,k}} \right\rangle_{(ki)} = 0, \quad (7c)$$

$$\left\langle \frac{\partial J_i}{\partial E_{hk}} + \frac{\partial s}{\partial c_{,i}} \frac{\partial H}{\partial E_{hk}} \right\rangle_{(hk)} = 0, \quad (7d)$$

$$\left\langle \frac{\partial s}{\partial q_{i,k}} \tau_E q_j - \frac{\partial s}{\partial \varepsilon_{,i}} T_{jk} \right\rangle_{(ij)} = 0, \quad (7e)$$

$$\left\langle \frac{\partial s}{\partial c_{,k}} \frac{\partial H}{\partial c_{,i}} + \frac{\partial s}{\partial q_{i,k}} a_c + \frac{\partial J_i}{\partial c_{,k}} \right\rangle = 0, \quad (7f)$$

$$f(\Sigma) \geq 0, \quad (7g)$$

wherein the symbol $\langle F \rangle$ denotes the symmetric part of the tensor function F , while the symbol $\langle F \rangle_{(ab\dots)}$ denotes the symmetric part of F with respect to the indicated indices.

Proof. Preliminarily, let us evaluate the inequality (6) in a fixed point \bar{P} of the body, at a fixed instant \bar{t} . Then, taking into account that, in a fixed point, the values of a function and those of its derivative are independent quantities, Equation (6) can be regarded as an algebraic inequality which is linear with respect to the elements of the set

$$\mathcal{H}(\bar{P}, \bar{t}) \equiv \left\{ \varepsilon_{,ki}(\bar{P}, \bar{t}), c_{,ki}(\bar{P}, \bar{t}), q_{j,ki}(\bar{P}, \bar{t}), E_{hk,i}(\bar{P}, \bar{t}), \dot{E}_{hk}(\bar{P}, \bar{t}), \dot{E}_{hk,i}(\bar{P}, \bar{t}) \right\}, \quad (8)$$

called set of the higher derivatives in (\bar{P}, \bar{f}) . Such derivatives are independent of the quantities in the brackets, which, instead, depend on the elements of

$$\Sigma(\bar{P}, \bar{f}) \equiv \{\varepsilon(\bar{P}, \bar{f}), \varepsilon_{,k}(\bar{P}, \bar{f}), c(\bar{P}, \bar{f}), c_{,k}(\bar{P}, \bar{f}), q_h(\bar{P}, \bar{f}), q_{h,k}(\bar{P}, \bar{f}), E_{hk}(\bar{P}, \bar{f})\}.$$

Moreover, since the inequality (6) must hold for arbitrary thermodynamic processes, the elements of $\mathcal{H}(\bar{P}, \bar{f})$ can have arbitrary sign and, consequently, the same is true for each of the linear terms entering such inequality. This implies that the inequality can be violated for some values of the elements of $\mathcal{H}(\bar{P}, \bar{f})$, unless all the linear terms in it vanish. This happens if, and only if, the thermodynamic restrictions (7) hold in (\bar{P}, \bar{f}) , once the symmetry of the elements of $\mathcal{H}(\bar{P}, \bar{f})$ is taken into account. On the other hand, since the point (\bar{P}, \bar{f}) is arbitrary, the thermodynamic restrictions (7) ensue. \square

Remark 1. Inequality (7g) is also called *reduced-entropy inequality*, since it represents the effective local rate of entropy production, once the restrictions (7a)–(7f) have been satisfied.

Remark 2. The classical Coleman-Noll procedure [14] consists in substituting in Equation (5) the sole evolution Equations (1b)–(1d). Thus, the set of the higher derivatives becomes

$$\mathcal{H}(\bar{P}, \bar{f}) \equiv \{\varepsilon_{,ki}(\bar{P}, \bar{f}), c_{,ki}(\bar{P}, \bar{f}), q_{j,ki}(\bar{P}, \bar{f}), E_{hk,i}(\bar{P}, \bar{f}), \dot{E}_{hk}(\bar{P}, \bar{f}), \dot{E}_{hk,i}(\bar{P}, \bar{f}), \dot{\varepsilon}_{,i}(\bar{P}, \bar{f}), \dot{c}_{,k}(\bar{P}, \bar{f}), \dot{q}_{i,k}(\bar{P}, \bar{f})\},$$

and the set of coefficients which must vanish is different with respect to the previous one. In particular, according to such a procedure, the coefficients of the quantities $\dot{\varepsilon}_{,i}$, $\dot{c}_{,k}$, and $\dot{q}_{i,k}$ must be zero, preventing so the dependency of s on $\varepsilon_{,i}$, $c_{,k}$, and $q_{i,k}$.

It is expected that such a methodology implies more severe restrictions on the constitutive quantities, because it requires that the entropy inequality is satisfied under the constraints (1b)–(1d), independently of the form of the evolution equations of the quantities $\dot{\varepsilon}_{,i}$, $\dot{c}_{,k}$, and $\dot{q}_{i,k}$. It is very efficient in those physical situations in which it is expected that the partial derivatives of s with respect to the gradients of the unknown fields vanish. However, for some systems as, for instance, Korteweg fluids [15,16], such derivatives should not vanish, so that the generalized method seems to be more suited for the exploitation of the entropy inequality. Below we analyze some cases for the system at hand, in which s should depend on the gradients. The generalized Coleman-Noll method reduces to the classical one if the form of the gradients of the unknown fields is not taken into account.

Corollary 1. If the inequality (5) is restricted by the sole constraints (1b)–(1d), the following thermodynamic restrictions hold

$$s = s(\varepsilon, c, q_i, E_{ij}), \quad (9a)$$

$$\left\langle \frac{\partial s}{\partial q_i} \frac{\tau_E q_j}{\tau_q A_q} - \frac{\partial s}{\partial \varepsilon} T_{ij} + \varrho \frac{\partial s}{\partial E_{ij}} \right\rangle_{(ij)} = 0, \quad (9b)$$

$$\left\langle \frac{\partial J_i}{\partial \varepsilon_{,k}} \right\rangle = 0, \quad (9c)$$

$$\left\langle \frac{\partial J_i}{\partial q_{j,k}} \right\rangle_{(ki)} = 0, \quad (9d)$$

$$\left\langle \frac{\partial J_i}{\partial E_{hk}} \right\rangle_{(hk)} = 0, \quad (9e)$$

$$\left\langle \frac{\partial J_i}{\partial c_{,k}} \right\rangle = 0, \quad (9f)$$

$$g(\Sigma) \geq 0, \quad (9g)$$

wherein g is a new scalar-valued function which can be obtained by f once the quantities $\frac{\partial s}{\partial \varepsilon_i}$, $\frac{\partial s}{\partial q_{i,k}}$, and $\frac{\partial s}{\partial c_k}$ have been put equal to zero.

Proof. To prove the Corollary it is enough to observe that, without the substitution of the gradient extensions of Equations (1b)–(1d) into the entropy inequality (5), the terms $\frac{\partial s}{\partial \varepsilon_i} \dot{\varepsilon}_i$, $\frac{\partial s}{\partial q_{i,k}} \dot{q}_{i,k}$, and $\frac{\partial s}{\partial c_k} \dot{c}_k$ would not couple with any other term. This would imply the following thermodynamic restrictions

$$\frac{\partial s}{\partial \varepsilon_i} = 0, \quad (10a)$$

$$\frac{\partial s}{\partial q_{i,k}} = 0, \quad (10b)$$

$$\frac{\partial s}{\partial c_k} c_k = 0, \quad (10c)$$

i.e., $s = s(\varepsilon, c, q_i, E_{ij})$. The coupling of Equations (7) and (10), yields Equation (9). \square

Corollary 2. In order to satisfy the system of restrictions (9) it is sufficient that the following set of relations is true

$$s = s(\varepsilon, c, q_i, E_{ij}), \quad (11a)$$

$$\frac{\partial s}{\partial q_i} \tau_E q_j - \frac{\partial s}{\partial \varepsilon} T_{ij} + \varrho \frac{\partial s}{\partial E_{ij}} = 0, \quad (11b)$$

$$J_i = J_i(\varepsilon, c, q_i), \quad (11c)$$

$$h(\Sigma) \geq 0, \quad (11d)$$

where h is a new function which can be obtained by g once the quantities $\frac{\partial J_i}{\partial \varepsilon_k}$, $\frac{\partial J_i}{\partial q_{j,k}}$, $\frac{\partial J_i}{\partial E_{hk}}$, and $\frac{\partial J_i}{\partial c_k}$ have been put equal to zero.

Proof. It is evident that if Equation (11b) is true, then the restriction (9b) is satisfied. Moreover, if $\frac{\partial J_i}{\partial \varepsilon_k} = 0$, $\frac{\partial J_i}{\partial q_{j,k}} = 0$, and $\frac{\partial J_i}{\partial E_{hk}} = 0$ and $\frac{\partial J_i}{\partial c_k} = 0$, i.e., if Equation (11c) is true, then the restrictions (9c)–(9f) hold, while function g in Equation (9g) reduces to function h in Equation (11d). \square

Corollary 3. The thermodynamic restrictions (11a)–(11c) are satisfied by the following constitutive equations for the specific entropy, the Cauchy stress and the entropy flux

$$s = s_0(\varepsilon, \bar{c}) + \frac{s_1 \bar{c}}{2\varepsilon} q_i^2 + \frac{\bar{c}}{\rho \varepsilon} \left[\frac{\lambda}{2} E_{ii}^2 + \mu E_{ij}^2 - (3\lambda + 2\mu) \frac{\alpha}{\bar{c}} \varepsilon E_{ij} \delta_{ij} \right], \quad (12a)$$

$$T_{ij} = \frac{s_1 \tau_E q_i q_j}{\tau_q A_q} + \lambda E_{hh} \delta_{ij} + 2\mu E_{ij} - (3\lambda + 2\mu) \frac{\alpha}{\bar{c}} \varepsilon \delta_{ij}, \quad (12b)$$

$$J_i = \frac{\bar{c}}{\varepsilon} q_i, \quad (12c)$$

wherein s_0 and s_1 are material functions depending on ε and c , λ and μ are the Lamé coefficients, and α is the coefficient of thermal expansion.

Proof. It is evident that the constitutive Equation (12a) satisfies the restriction (11a), and the constitutive Equation (12c) satisfies the restriction (11c). Moreover, by Equation (12a) we get

$$\frac{\partial s}{\partial q_i} = \frac{s_1 \bar{c}}{\varepsilon} q_i, \quad (13a)$$

$$\frac{\partial s}{\partial E_{ij}} = \frac{\bar{c}}{\rho \varepsilon} \left[\lambda E_{hh} \delta_{ij} + 2\mu E_{ij} - (3\lambda + 2\mu) \frac{\alpha}{\bar{c}} \varepsilon \delta_{ij} \right]. \quad (13b)$$

By Equations (13a) and (13b) it follows immediately that the restriction (11b) is satisfied, once the thermodynamic relation $\frac{\partial s}{\partial \varepsilon} = \frac{1}{\vartheta} = \frac{\bar{c}}{\varepsilon}$ is taken into account. \square

Remark 3. Equation (12) yields the classical expression of the entropy flux postulated by Coleman and Noll in their celebrated paper on the thermodynamics of viscoelastic heat conducting solids [14], while Equations (12a) and (12b) generalize the constitutive equations for s and T_{ij} obtained in classical linear thermoelasticity [34]. Both the constitutive Equations (12a) and (12b) are local, as expected, and represent a very particular case of the more general constitutive equations compatible with the set of thermodynamic restrictions in Equation (7).

Remark 4. It is important observing that the material coefficients λ , μ , and α , entering Equations (12a) and (12b), depend, in general, on ε and c .

Remark 5. Due to the presence in s of the term $\frac{s_1 \bar{c}}{2\varepsilon} q_i^2$, in nonequilibrium situations the temperature $\vartheta = \frac{\partial s}{\partial \varepsilon}$ depends on the heat flux too.

One could wonder if, for the system at hand, the application of the generalized exploitation procedure is necessary or not. To answer that question we observe preliminarily that, since function H depends on E_{ij} , the gradient of c is not constant in time, as in the case of rigid heat conductors, but can change as a consequence of the deformation, as it can be easily seen by taking the gradient of Equation (1d). This variable gradient contribute to the variation of the heat flux too, according to Equation (1c), and this produces further thermal dissipation. Thus, both s and J_i are expected to contain terms which depend on E_{ij} and $c_{,i}$. Moreover, by Equation (1c) it follows that the temperature varies along the medium even in the absence of heat flux, so that a dependency of s and J_i on $\varepsilon_{,i}$ is expected as well. The propositions below clarify the conditions under which such dependencies are possible. To this end we generalize Equation (12c) as follows

$$J_i = \frac{\bar{c}}{\varepsilon} q_i + J_0(\varepsilon, c, E_{ij}) \varepsilon_{,i} + J_1(\varepsilon, c, E_{ij}) c_{,i}, \quad (14)$$

and investigate its compatibility with second law of thermodynamics.

Corollary 4. The constitutive equation of J_i may depend on E_{ij} if, and only if, s depends on $c_{,i}$ and H depends on E_{ij} .

Proof. To prove this proposition, we observe that if either s is independent of $c_{,i}$, or H is independent of E_{ij} , then the thermodynamic restriction (7d) implies

$\left\langle \frac{\partial J_i}{\partial E_{hk}} \right\rangle_{(hk)} = 0$. On the other hand, since the strain tensor E_{hk} is symmetric, this relation is equivalent to $\frac{\partial J_i}{\partial E_{hk}} = 0$, i.e., J_i is independent of E_{hk} . \square

Corollary 5. The constitutive equation of J_i may be linear in $\varepsilon_{,k}$ (as in Equation (14)) if, and only if, either s depends on $c_{,i}$ and H depends on $\varepsilon_{,i}$, or s depends on $q_{i,k}$.

Proof. To prove this proposition, it is enough to observe that if s is independent of $c_{,i}$, H is independent of $\varepsilon_{,i}$, and s is independent of $q_{i,k}$, then the thermodynamic restriction (7b) implies $\left\langle \frac{\partial J_i}{\partial \varepsilon_{,k}} \right\rangle = 0$ or, equivalently, $\frac{1}{2} \left(\frac{\partial J_i}{\partial \varepsilon_{,k}} + \frac{\partial J_k}{\partial \varepsilon_{,i}} \right) = 0$. It yields $J_0 \delta_{ik} = 0$, once Equation (14) is taken into account. \square

Corollary 6. *The constitutive equation of J_i may be linear in $c_{,k}$ (as in Equation (14)) if, and only if, either s depends on $c_{,k}$ and H depends on $c_{,i}$, or s depends on $q_{i,k}$.*

Proof. To prove this proposition, it is enough to observe that if s is independent of $c_{,k}$ or H is independent of $\varepsilon_{,i}$, and s is independent of $q_{i,k}$, then the thermodynamic restriction (7f) implies $\left\langle \frac{\partial J_i}{\partial c_{,k}} \right\rangle = 0$ or, equivalently, $\frac{1}{2} \left(\frac{\partial J_i}{\partial c_{,k}} + \frac{\partial J_k}{\partial c_{,i}} \right) = 0$. It yields $J_1 \delta_{ik} = 0$, once Equation (14) is taken into account. \square

The nonlocal terms entering Equation (14) may be important at nanometric scale, where nonlocal effects are evident. One should note that the additional terms $J_0(\vartheta, c, E_{ij})\varepsilon_{,i} + J_1(\vartheta, c, E_{ij})c_{,i}$ in Equation (14) may be regarded as the entropy extraflux proposed in [35].

4. Thermoelastic-wave propagation

In this section we study the propagation of thermoelastic waves along a graded material. Our analysis will be pursued under the following hypotheses:

- the constitutive equation (12b) for the Cauchy stress holds;
- all the material coefficients are constant;
- function H is linear in the gradients, i.e.,
- $H = H_0(\varepsilon, E_{hk}, c) + h_k \varepsilon_{,k} + m_k c_{,k} + N_{kj} q_{k,j}$;
- $\frac{\partial (s_1 \tau_E / \tau_q A_q)}{\partial x_j}$ is negligible; wherein h_k , m_k and N_{kj} depend on the set (ε, E_{hk}, c) .

Under the hypotheses above, the system of equations (1) writes as

$$\rho \ddot{u}_i - \frac{s_1 \tau_E q_{i,j} q_j}{\tau_q A_q} - \frac{s_1 \tau_E q_i q_{j,j}}{\tau_q A_q} - \lambda E_{hh,i} - 2\mu E_{ij,j} + \bar{b} \varepsilon_{,i} = 0, \quad (15a)$$

$$\rho \dot{\varepsilon} - \frac{s_1 \tau_E q_i q_j}{\tau_q A_q} \dot{E}_{ij} - \lambda E_{hh} \dot{E}_{ii} - 2\mu E_{ij} \dot{E}_{ij} + \bar{b} \varepsilon \dot{E}_{ii} + q_{i,i} = 0, \quad (15b)$$

$$\tau_q A_q \dot{q}_i + q_i + \bar{\kappa} A_K \varepsilon_{,i} - \frac{2\tau_Q}{\rho \varepsilon} q_{j,i} q_j - \tau_E \dot{E}_{ij} q_j - a_c c_{,i} = 0, \quad (15c)$$

$$\dot{c} = H_0(\varepsilon, E_{hk}, c) + h_k \varepsilon_{,k} + m_k c_{,k} + N_{kj} q_{k,j}, \quad (15d)$$

with $\bar{b} = (3\lambda + 2\mu)\alpha/\bar{c}$, and $\bar{\kappa} = \kappa/\bar{c}$.

The nonlinear system of equations above allows the existence of nonregular solutions.

Definition 1. *An acceleration wave is a traveling surface \mathcal{S} across which a solution $\{u_i, \varepsilon, q_i, c\}$ of Equation (15) is continuous, but its first- and higher-order derivatives suffer jump discontinuities [23,36].*

Remark 6. *It is worth observing that Equation (15) hold in the points of the two regions behind and ahead \mathcal{S} , while on \mathcal{S} those equations must be written in terms of the jumps of the discontinuous fields across the wavefront. Such jumps are given by the differences*

$$\begin{aligned} \delta \ddot{u}_i &= \ddot{u}_i^- - \ddot{u}_i^+, & \delta u_{i,j} &= u_{i,j}^- - u_{i,j}^+, & \delta u_{ijk} &= u_{ijk}^- - u_{ijk}^+, \\ \delta \varepsilon_{,j} &= \varepsilon_{,j}^- - \varepsilon_{,j}^+, & \delta q_{i,j} &= q_{i,j}^- - q_{i,j}^+, & \delta c_{,j} &= c_{,j}^- - c_{,j}^+, \end{aligned} \quad (16)$$

where the upscript $+$ denotes the value of the corresponding fields in the region which S is about to enter, and the upscript $-$ denotes the same value in the region which S is about to leave.

For the sake of simplicity, herein we consider a one dimensional system. Then, Equation (15) write as

$$\varrho \ddot{u} - (\lambda + 2\mu)u_{,xx} + \bar{b}\varepsilon_{,x} - 2\frac{s_1\tau_E}{\tau_q A_q}qq_{,x} = 0, \quad (17a)$$

$$- \frac{s_1\tau_E q^2}{\tau_q A_q} \dot{u}_{,x} - (\lambda + 2\mu)u_{,x}\dot{u}_{,x} + \bar{b}\varepsilon\dot{u}_{,x} + \varrho\dot{\varepsilon} + q_{,x} = 0, \quad (17b)$$

$$- \tau_E q \dot{u}_{,x} + \bar{\kappa} A_K \varepsilon_{,x} + \tau_q A_q \dot{q} + q - \frac{2\tau_Q}{\varrho\varepsilon} qq_{,x} - a_c c_{,x} = 0, \quad (17c)$$

$$H_0 + h\varepsilon_{,x} + Nq_{,x} - \dot{c} + mc_{,x} = 0. \quad (17d)$$

In order to determine the jumps of the discontinuous fields across S , we first observe that in the one dimensional case the sole component of the strain tensor is $E = u_{,x}$. Here and in the following we suppose that the fields $u_{,x}$, ε , q , c are continuous across S but their space and time derivatives suffer jump discontinuities. Meantime, we suppose that across S the time derivatives of the displacement u are discontinuous too. Hence, we introduce the following notation

$$\delta \frac{\partial^2 u}{\partial x^2} = \delta \frac{\partial(\partial u / \partial x)}{\partial x} = \delta \frac{\partial E}{\partial x} \equiv \delta E, \quad \delta \frac{\partial \varepsilon}{\partial x} \equiv \delta \varepsilon, \quad \delta \frac{\partial q}{\partial x} \equiv \delta q, \quad \delta \frac{\partial c}{\partial x} \equiv \delta c. \quad (18)$$

Moreover, being $u_{,x}$ continuous across S , by the classical Hadamard identities, (see Ref. [36] and Equations (54) and (55) therein), we can write

$$\delta \frac{\partial^2 u}{\partial t^2} = U^2 \delta \frac{\partial^2 u}{\partial x^2} = U^2 \delta E, \quad (19)$$

wherein U is the speed of propagation of thermomechanical disturbances. Then, the system (17) yields

$$[\varrho U^2 - (\lambda + 2\mu)] \delta E + \bar{b} \delta \varepsilon - 2\frac{s_1\tau_E}{\tau_q A_q} q \delta q = 0, \quad (20a)$$

$$U \left[\frac{s_1\tau_E q^2}{\tau_q A_q} + (\lambda + 2\mu)E - \bar{b}\varepsilon \right] \delta E - \varrho U \delta \varepsilon + \delta q = 0, \quad (20b)$$

$$U \tau_E q \delta E + \bar{\kappa} A_K \delta \varepsilon - \left[U \tau_q A_q + \frac{2\tau_Q}{\varrho\varepsilon} q \right] \delta q - a_c \delta c = 0, \quad (20c)$$

$$h \delta \varepsilon + N \delta q + (U + m) \delta c = 0, \quad (20d)$$

wherein it must be understood that the coefficients of the unknown jumps are evaluated in the region which S is about to enter. Equation (19) represent a linear and homogeneous algebraic system in the unknown quantities δE , $\delta \varepsilon$, δq and δc , which provides, in principle, the values of the jumps we are looking for. The following statement is straightforward.

Theorem 2. The system (20) admits nontrivial solution if, and only if, the following condition is fulfilled

$$\det \begin{bmatrix} [\varrho U^2 - (\lambda + 2\mu)] & \bar{b} & -2\frac{s_1\tau_E}{\tau_q A_q} q & 0 \\ U \left[\frac{s_1\tau_E q^2}{\tau_q A_q} + (\lambda + 2\mu)E - \bar{b}\varepsilon \right] & -\varrho U & 1 & 0 \\ U \tau_E q & \bar{\kappa} A_K & -\left(U \tau_q A_q + \frac{2\tau_Q}{\varrho\varepsilon} q \right) & -a_c \\ 0 & h & N & (U + m) \end{bmatrix} = 0 \quad (21)$$

Proof. The proof immediately follows by the observation that the system (20) admits non trivial solutions if, and only if, the matrix of the coefficients is singular, i.e., if and only if Equation (21) holds. \square

Suppose now that the following additional conditions hold

$$\tau_E = \tau_Q = N = \bar{b} = 0. \quad (22)$$

Remark 7. The conditions (22) mean that the Cauchy stress coincides with that of classical linear elasticity, in the absence of thermal effects. Moreover, nonlocal effects for the heat flux do not influence the evolution equations for the heat flux (Equation (1c)) and for c (Equation (1d)).

Under the hypotheses (22) the system (20) becomes

$$[\varrho U^2 - (\lambda + 2\mu)] \delta E = 0, \quad (23a)$$

$$U(\lambda + 2\mu)E \delta E - \varrho U \delta \varepsilon + \delta q = 0, \quad (23b)$$

$$\bar{\kappa} A_K \delta \varepsilon - U \tau_q A_q \delta q - a_c \delta c = 0, \quad (23c)$$

$$h \delta \varepsilon + (U + m) \delta c = 0. \quad (23d)$$

Theorem 3. The system (23) admits nontrivial solution if, and only if, the following equation is fulfilled

$$\left[\varrho U^2 - (\lambda + 2\mu) \right] \left[\varrho U^2 (U \tau_q A_q + m \tau_q A_q) - ((U + m) \bar{\kappa} A_K + h a_c) \right] = 0. \quad (24)$$

Proof. To prove the theorem we observe that the system (23) admits non trivial solutions if, and only if,

$$\det \begin{bmatrix} [\varrho U^2 - (\lambda + 2\mu)] & 0 & 0 & 0 \\ U(\lambda + 2\mu)E & -\varrho U & 1 & 0 \\ 0 & \bar{\kappa} A_K & -U \tau_q A_q & -a_c \\ 0 & h & 0 & (U + m) \end{bmatrix} = 0, \quad (25)$$

i.e, if, and only if, Equation (24) holds. \square

Remark 8. The compatibility condition (24) is satisfied if either two elastic waves propagate with speeds

$$U = \pm \sqrt{\frac{\lambda + 2\mu}{\varrho}}, \quad (26)$$

or two thermoelastic waves propagate with speeds

$$U = -m, \quad U = -m - \frac{h a_c}{\bar{\kappa} A_K}. \quad (27)$$

Remark 9. If all the relaxation times in Equation (1c) vanish, then the generalized Fourier law

$$q_i = -\bar{\kappa} A_K \varepsilon_{,i} - a_c c_{,i}, \quad (28)$$

holds. Thus, being $q_i = q_i(\varepsilon_{,i}, c_{,i}, E_{hk})$, we are in the realm of Rational Thermodynamics, i.e., the thermodynamic theory in which heat flux and stress tensor are assigned through suitable constitutive equations [13,37].

Corollary 7. Under the validity of the generalized Fourier law (28), if

$$N = \bar{b} = 0, \quad (29)$$

then the following speeds of propagation are possible

$$U = \pm \sqrt{\frac{\lambda + 2\mu}{\varrho}}, \quad U = -m - \frac{ha_c}{\bar{\kappa}A_K}. \quad (30)$$

Proof. The proof follows by the observation that, if $\tau_q = 0$, Equation (24) reduces to

$$\left[\varrho U^2 - (\lambda + 2\mu) \right] [(U + m)\bar{\kappa}A_K + ha_c] = 0. \quad (31)$$

□

Remark 10. The first speed in Equation (30) is of elastic type, while the second one, due to the combined presence of $\bar{\kappa}$ and A_K , corresponds to a thermo-mechanical wave. If $a_c = 0$, then the evolution equation of the heat flux does not appear in the wave speeds, and the constant speed $U = -m$ corresponds to a stoichiometric wave, i.e., to a transport of energy due to the variable composition.

Corollary 8. Under the hypotheses (22), if

$$a_c = m = 0, \quad A_q \simeq 1, \quad A_K \simeq 1, \quad (32)$$

then it is possible the propagation of elastic or thermal perturbations with speeds

$$U = \pm \sqrt{\frac{\lambda + 2\mu}{\varrho}}, \quad U = \pm \sqrt{\frac{\bar{\kappa}}{\varrho\tau_q}} \quad (33)$$

Proof. We first observe that the hypotheses (22) imply Equation (24). On the other hand, if the conditions (32) hold, then Equation (24) reduces to

$$\left[\varrho U^2 - (\lambda + 2\mu) \right] \left[U^2 \varrho \tau_q - \bar{\kappa} \right] = 0, \quad (34)$$

which proves the assertion. □

Remark 11. Under the hypotheses of Corollary 8 the system behaves as a purely elastic body (no thermoelastic coupling), with a Cattaneo type evolution equation for the heat flux. Thus, the presence of purely elastic and purely thermal waves (second sound) is expected.

Definition 2. An acceleration wave is said to propagate in a state of thermal equilibrium if in the region in which the wave is about to enter

$$\dot{\varepsilon} = \varepsilon_{,x} = q = 0. \quad (35)$$

In such a case, the system (20) becomes

$$[\varrho U^2 - (\lambda + 2\mu)] \delta E + \bar{b} \delta \varepsilon = 0, \quad (36a)$$

$$U[(\lambda + 2\mu)E - \bar{b}\varepsilon] \delta E - \varrho U \delta \varepsilon + \delta q = 0, \quad (36b)$$

$$\bar{\kappa}A_K \delta \varepsilon - U\tau_q A_q \delta q - a_c \delta c = 0, \quad (36c)$$

$$h \delta \varepsilon + N \delta q + (U + m) \delta c = 0. \quad (36d)$$

Theorem 4. The system (36) admits nontrivial solution if, and only if, the following condition is fulfilled

$$\det \begin{bmatrix} [\varrho U^2 - (\lambda + 2\mu)] & \bar{b} & 0 & 0 \\ U[(\lambda + 2\mu)E - \bar{b}\varepsilon] & -\varrho U & 1 & 0 \\ 0 & \bar{\kappa} A_K & -U\tau_q A_q & -a_c \\ 0 & h & N & (U + m) \end{bmatrix} = 0. \quad (37)$$

Proof. The proof immediately follows by the observation that the system (36) admits non trivial solutions if, and only if, the matrix of the coefficients is singular, i.e., if and only if Equation (37) holds. \square

Corollary 9. If $m = 0$, the system (34) admits nontrivial solution if, and only if, the following condition is fulfilled

$$[\varrho U^2 - (\lambda + 2\mu)] [\varrho U^3 \tau_q A_q - U(\varrho N a_c + \bar{\kappa} A_K) + h a_c] - \bar{b} U [(\lambda + 2\mu)E - \bar{b}\varepsilon] (-U^2 \tau_q A_q + N a_c) = 0. \quad (38)$$

Proof. To prove this statement we observe that Equation (38) follows by Equation (37) once the condition $m = 0$ is taken into account. \square

Remark 12. Equation (38) is satisfied if two elastic of speed

$$U = \pm \sqrt{\frac{\lambda + 2\mu}{\varrho}}, \quad (39)$$

propagate together with two thermoelastic waves with speed

$$U = \pm \sqrt{\frac{N a_c}{\tau_q A_q}}. \quad (40)$$

Moreover, if the third-grade equation

$$\varrho U^3 \tau_q A_q - U(\varrho N a_c + \bar{\kappa} A_K) + h a_c = 0, \quad (41)$$

admits real solutions, then Equation (38) is also satisfied if thermoelastic waves with speed given by such solutions propagate together with two thermoelastic waves with speed given by Equation (40).

5. Discussion

In Equation (1) we have proposed a mathematical model describing the nonlinear thermoelastic behavior of composition graded materials. The compatibility of the aforementioned model with second law of thermodynamics has been investigated by applying a generalized Coleman-Noll procedure.

We have determined the speeds of propagation of coupled first- and second-sound pulses, propagating either in nonequilibrium states, or in equilibrium states, under different physical conditions. We found that several different types of thermomechanical perturbations may propagate, depending on the value of the material coefficients characterizing the system.

The basic assumption underlying the present work is that the deformation of the body can produce changes in the stoichiometry, altering locally the material composition. An interesting consequence of such hypothesis is the possibility of propagation of pure stoichiometric waves, with speed $U = -m$, where m measures the nonlocality of \dot{c} with respect to c itself (see Equation (15d)). The coupling of thermal and stoichiometric effects has been taken into account through the term $-a_c c_{,i}$ in the evolution equation of the heat flux (see Equation (1d)). Such a coupling produces thermoelastic waves with speed

$$U = -m - \frac{ha_c}{\bar{\kappa}A_K}. \quad (42)$$

for propagation in nonequilibrium states, and

$$U = \pm \sqrt{\frac{Na_c}{\tau_q A_q}}. \quad (43)$$

for propagation in equilibrium states. Furthermore, it is capable to produce additional thermoelastic waves with speed given by the real solutions of Equation (41)- It should be noted that if $a_c = 0$, beside the solution $U = 0$, Equation (41) yields

$$U = \pm \sqrt{\frac{\bar{\kappa}A_K}{\varrho\tau_q A_q}}, \quad (44)$$

which corresponds to heat waves with strain-dependent thermal conductivity. Finally, the model is capable to reproduce the propagation of pure elastic waves and pure thermal waves, in the absence of coupling (see Equation (33)).

Such a rich behavior has been obtained under the hypothesis $\tau_Q = \tau_E = 0$. In future researches we aim at considering the most general case, which we expect to offer a lot of possibilities of nonlinear thermoelastic coupling.

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