Thermodynamic modeling of fully coupled finite-deformation thermo-electro-magneto-mechanical behavior for multifunctional applications

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ABSTRACT

In this paper, we model fully coupled thermo-electro-magneto-mechanical (TEMM) behavior in the finite-deformation regime by (i) developing for the first time a comprehensive catalogue of free energies, state variables, and state equations, and (ii) combining this catalogue with the first principles of nonlinear continuum electrodynamics. We develop our catalogue in a thermodynamically consistent manner, and circumvent the ambiguities and challenges inherent in nonlinear continuum electrodynamics, by connecting with classical equilibrium thermodynamics. We use its formalism as a blueprint for characterizing a fundamental energetic process, that is, one where internal energy is the characterizing potential, the independent variables are extensive, and the dependent variables are intensive. A key feature of identifying this fundamental energetic process is the resulting ability to transparently and rigorously introduce new free energies – many appearing in the finite-deformation TEMM literature for the first time – that employ any set of intensive or extensive quantities as independent variables. We also develop novel mathematical transformations that accommodate alternative electromagnetic work conjugates as independent variables.

Each thermodynamic potential in our comprehensive catalogue characterizes a particular thermo-electro-magneto-mechanical process. Each process, in turn, correlates with a particular experiment, the independent variables being controlled and the dependent variables being the measured responses. Our framework will thus enable the development of constitutive models for multifunctional materials under different experimental conditions. Additionally, the research presented herein can be used to convert targeted performance properties that are inherently nonlinear, three dimensional, and anisotropic into a "recipe" for multifunctional material design.

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1. Introduction

Multifunctional materials are a novel class of engineered materials whose distinguishing feature is their ability to provide structural integrity (e.g., strength, stiffness, rigidity) while simultaneously accomplishing one or more performance-based functions (e.g., sensing, actuation) (Christodoulou & Venables, 2003). They are often manufactured by layering a constituent set of materials into a composite, or by embedding particles or fibers in a matrix (Nemat-Nasser, Nemat-Nasser, Plaisted,

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Starr, & Amirkhizi, 2006; Salonitis, Pandremenos, Paralikas, & Chryssolouris, 2010). Multifunctional materials are generally tailored to optimally accomplish a particular set of tasks. For instance, conformal load-bearing antennas embedded in the structure of an aircraft are fabricated from materials that are capable of supporting aerodynamic loads in addition to radiating and receiving electromagnetic signals (Lee & Inman, 2009; You & Hwang, 2005). These and many other novel capabilities afforded by multifunctional materials (e.g., self healing, power generation, power storage, and vibration mitigation) promise to (i) improve existing actuating and sensing technologies by increasing efficiency and reducing weight, cost, energy consumption, and size, and (ii) expand the performance space of the next generation of intelligent systems, structures, and devices (Nemat-Nasser et al., 2006).

In order to optimally design new multifunctional materials, there is a need to develop rigorous mathematical models that can corroborate experimental data, be used to generate numerical simulations, and be employed in computational design software. A common practice in the mathematical modeling of material response is to *a priori* specialize the constitutive equations to a particular regime of behavior. For example, in the continuum modeling of smart materials, material response is frequently confined to low-signal, small-deformation ranges of operation; time dependence of the electromagnetic fields and material nonlinearity are often neglected; or only a subset of the full thermo-electro-magneto-mechanical (TEMM) coupling is accounted for.

These simplifying assumptions are used to generate mathematically tractable models that are well-suited for particular materials under particular operating conditions. However, this specialized approach is too restrictive to facilitate the design and characterization of the next generation of multifunctional materials and devices. As an example, for multifunctional devices such as load-bearing antenna systems that operate at high frequencies (up to MHz-GHz) (You & Hwang, 2005), the customary quasi-static electromagnetic field approximation breaks down. To model their behavior accurately, more advanced dynamic simulations of fully coupled TEMM behavior are required. Thus, we contend that enabling multifunctional materials to reach their full performance potential demands a modeling architecture that (a) is based as much as possible on first principles; (b) has the breadth to accommodate 3-D fully coupled thermo-electro-magneto-mechanical behavior, finite deformations, anisotropy, nonlinear constitutive response, and dynamic electromagnetic fields; and (c) has the flexibility to accommodate multifunctionality at various time scales, length scales, and compositional levels.

The foundation for this type of mathematical framework was presented, for instance, in Truesdell and Toupin (1960), Toupin (1963), Landau, Lifshitz, and Pitaevskii (1984), Penfield and Haus (1967), Tiersten (1971), Tiersten (1990), Hutter, van de Ven, and Ursescu (2006), Hutter (1979), Hutter (1977), Hutter and Pao (1974), Pao (1978), Pao and Hutter (1975), Maugin and Eringen (1977), Maugin (1988), Eringen and Maugin (1990), Green and Naghdi (1984) and Green and Naghdi (1995), to list a small but influential portion of the early work. These seminal models unify continuum mechanics with classical electrodynamics, and consist of fundamental laws or first principles (i.e., Maxwell's equations, conservation of charge, conservation of mass, balance of linear momentum, balance of angular momentum, and the first law of thermodynamics), coupling terms (i.e., electromagnetic interaction terms that couple the thermomechanical balance laws to Maxwell's equations), and boundary conditions. They describe the most general form of *fully coupled* thermo-electro-magneto-mechanical behavior in a deformable, polarizable, magnetizable continuum. Theoretial aspects of these general models have been revisited in recent years in Kovetz (2000), Ericksen (2007), Ericksen (2008), Steigmann (2009) and Maugin (2009), while practical applications to nonlinear electro-elastic solids, nonlinear magneto-elastic solids, and electro-rheological fluids have been pursued in Dorfmann and Ogden (2003, 2004a, 2004b, 2005a, 2005b, 2006), Steigmann (2004), Rajagopal and Ruzicka (2001), McMeeking and Landis (2005), McMeeking, Landis, and Jimenez (2007), Vu, Steinmann, and Possart (2007), Kankanala and Triantafyllidis (2004), Voltairas, Fotiadis, and Massalas (2003), Suo, Zhao, and Greene (2008), Zhao and Suo (2008), Zhu, Stoyanov, Kofod, and Suo (2010), Qin, Librescu, Hasanyan, and Ambur (2003), Qin, Hasanyan, Librescu, and Ambur (2003), Richards and Odegard (2010), Oates, Wang, and Sierakowski (2012).

However, certain aspects of constitutive modeling and material characterization, enabled through the principles of thermodynamics, remain unexplored in the finite-deformation TEMM literature. In particular:

(i) The constitutive models do not at the outset explicitly identify the intensive–extensive work conjugates in the second law, as is customary in classical thermodynamics (Callen, 1985). As the electromagnetic work conjugates differ from model to model in finite-deformation TEMM (more on this in Section 5), this leads to ambiguity in specifying the fundamental energetic relationship (i.e., the functional form of the internal energy). The fundamental energetic relationship is the starting point for proceeding transparently and rigorously with the formalism of Legendre transformations, which is used to define new free energies (Callen, 1985). Often, the fundamental energetic relationship is not explicitly specified, making the connection between the characterizing free energy in the constitutive model and the internal energy of the system opaque.

(ii) The constitutive models are not comprehensive in that they do not consider all possible combinations of independent variables and corresponding thermodynamic potentials. Notably absent from many finite-deformation TEMM theories are free energies that employ stress as an independent variable. From an experimental perspective, intensive quantities like stress and temperature are often more convenient to control than extensive quantities like deformation and entropy. Hence, it is important to identify all possible free energies to provide the experimentalist with optimal flexibility when characterizing novel multifunctional materials.

In this paper, our primary contributions are addressing these shortcomings and taking the necessary first steps toward creating a unified, comprehensive, and thermodynamically consistent model for designing and characterizing novel multifunctional materials with coupled thermo-electro-magneto-mechanical response. Importantly, our approach reconciles large-deformation Eulerian continuum mechanics and electrodynamics with the principles of classical thermodynamics. We remedy shortcoming (i) above by explicitly identifying the extensive and intensive quantities in our particular statement of the second law. As is customary in classical thermodynamics, the internal energy is defined as a function of the *extensive* quantities, and the thermodynamic state equations, derived from the second law of thermodynamics (Coleman & Noll, 1963), define the corresponding *intensive* quantities (Callen, 1985).

With this fundamental energetic relationship as our starting point, we remedy shortcoming (ii) by deriving free energies that utilize any or all of the *intensive* quantities as independent variables using Legendre transforms of the internal energy, defined in a manner consistent with classical thermodynamics (Callen, 1985). This formalism enables us to develop an exhaustive list of free energies, most of which are presented in the literature for the first time, with any combination of intensive or extensive independent variables. Another novel feature of our framework is the introduction of a secondary variable as an independent variable in a free energy. Secondary variables are physical quantities that are not a part of the aforementioned intensive–extensive conjugate pairs, but are accessible algebraically from them. For instance, electric displacement is a linear combination of electric field (intensive) and polarization (extensive). To rigorously enable the use of secondary quantities as independent variables, we develop novel Legendre-type transformations that define a new set of free energies in a thermodynamically consistent manner. The resulting constitutive framework is comprehensive in that it is capable of characterizing all TEMM processes.

This paper is structured as follows: In Section 2, we explicitly state the first principles (i.e., Maxwell's equations, conservation of mass, balance of linear momentum, balance of angular momentum, and the first law of thermodynamics) for a deformable, polarizable, magnetizable TEMM material. The thermomechanical balance laws presented in Section 2 include electromagnetic interaction terms (namely, the electromagnetic body force, body couple, and energy supply) that couple the thermomechanical balance laws to Maxwell's equations. These coupling terms are described and quantified in Section 3. In Section 4, using the formalism described earlier, we develop a comprehensive catalogue of free energies, state variables, and state equations that characterize fully coupled TEMM processes. Contact can be established with similar catalogues presented by other authors, e.g., Pérez-Fernández, Bravo-Castillero, Rodríguez-Ramos, and Sabina (2009) and Smith (2005, pp. 63–64) for the special case of thermo-electro-mechanical processes, and Soh and Liu (2005) for the special case of electro-magneto-mechanical processes, all within the regime of small deformations and linear material response.

In Section 5, energy potentials that utilize secondary electromagnetic quantities as independent variables are introduced using novel Legendre-type transformations of the internal energy, and contact is established with previous work by Green and Naghdi (1984). In Section 6, we extend the formalism of Sections 4 and 5 to TEMM processes employing entropy rather than energy as the characterizing thermodynamic potential. We deduce a set of Legendre-transformed entropic potentials, from which those in a previous paper by Rooney and Bechtel (2004) can be recovered as special cases. In Section 7, we highlight some potential applications of our TEMM framework.

2. The first principles for TEMM materials

In this section, the first principles (or fundamental laws) for a polarizable, magnetizable, deformable thermo-electro-magneto-mechanical (TEMM) material are presented. We first give the integral expressions, rather than starting directly with a set of pointwise equations, because the pointwise equations must be derivable from an integral set, and the assumptions for continuum models are imposed on the integral form (Green & Naghdi, 1991, 1995). An important feature of this progression is that it enables scientists and engineers to assess the physical relevance of a particular pointwise equation by determining if a corresponding integral statement exists.

In this paper, we adopt the following Eulerian integral statements[2] of the first principles (Hutter et al., 2006; Pao & Hutter, 1975):

Gauss's law for magnetism

$$\int_{\partial \mathcal{V}} \mathbf{b}^* \cdot \mathbf{n} da = 0, \tag{1a}$$

Faraday's law

$$\frac{d}{dt} \int_{\mathcal{S}} \mathbf{b}^* \cdot \mathbf{n} da = - \int_{\partial \mathcal{S}} \mathbf{e}^* \cdot \mathbf{l} dl, \tag{1b}$$

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[2] We employ as much as possible the convention that lowercase Greek letters denote scalars, bold lowercase Latin letters denote first-order tensors (vectors), and bold uppercase Latin letters denote second-order tensors (tensors). Accordingly, our notation for the various electromagnetic vector fields is lowercase, which departs from the uppercase notation appearing in much of the literature.

Gauss's law for electricity

$$\int_{\partial \mathcal{V}} \mathbf{d}^* \cdot \mathbf{n} da = \int_{\mathcal{V}} \sigma^* dv, \tag{1c}$$

Ampère–Maxwell law

$$\frac{d}{dt} \int_{\mathcal{S}} \mathbf{d}^* \cdot \mathbf{n} da + \int_{\mathcal{S}} \mathbf{j}^* \cdot \mathbf{n} da = \int_{\partial \mathcal{S}} \mathbf{h}^* \cdot \mathbf{l} dl, \tag{1d}$$

Conservation of mass

$$\frac{d}{dt} \int_{\mathcal{V}} \rho dv = 0, \tag{1e}$$

Balance of linear momentum

$$\frac{d}{dt} \int_{\mathcal{V}} \rho \mathbf{v} dv = \int_{\mathcal{V}} \rho(\mathbf{f}^m + \mathbf{f}^{em}) dv + \int_{\partial \mathcal{V}} \mathbf{t} da, \tag{1f}$$

Balance of angular momentum

$$\frac{d}{dt} \int_{\mathcal{V}} \mathbf{x} \times \rho \mathbf{v} dv = \int_{\mathcal{V}} \mathbf{x} \times \rho(\mathbf{f}^m + \mathbf{f}^{em}) dv + \int_{\mathcal{V}} \rho \mathbf{c}^{em} dv + \int_{\partial \mathcal{V}} \mathbf{x} \times \mathbf{t} da, \tag{1g}$$

First law of thermodynamics

$$\frac{d}{dt} \int_{\mathcal{V}} \rho(\varepsilon + \frac{1}{2} \mathbf{v} \cdot \mathbf{v}) dv = \int_{\mathcal{V}} \rho(\mathbf{f}^m + \mathbf{f}^{em}) \cdot \mathbf{v} dv + \int_{\partial \mathcal{V}} \mathbf{t} \cdot \mathbf{v} da + \int_{\mathcal{V}} \rho(r^t + r^{em}) dv - \int_{\partial \mathcal{V}} h da. \tag{1h}$$

These integral statements are global, i.e., valid on the entire body and all subsets, namely any open material volume $\mathcal{V}$ bounded by a closed material surface $\partial \mathcal{V}$, or any open material surface $\mathcal{S}$ bounded by a closed material curve $\partial \mathcal{S}$, all in the present configuration. In (1a)–(1h), $\mathbf{l}$, is the unit tangent along $\partial \mathcal{S}$; $\mathbf{n}$ is an outward unit normal; dv, da, and dl are volume, area, and line elements in the present configuration; $() \cdot ()$ denotes an inner product; $() \times ()$ denotes a vector cross product; and d/dt denotes the derivative of a function of a single variable t.

The thermo-electro-magneto-mechanical fields appearing in (1a)–(1h) are Eulerian, i.e., they are functions of the present position $\mathbf{x}$ of a continuum particle and time t. These fields include the present density ρ, velocity $\mathbf{v}$, mechanically and electromagnetically induced specific body forces $\mathbf{f}^m$ and $\mathbf{f}^{em}$, traction $\mathbf{t}$, electromagnetically induced specific body couple $\mathbf{c}^{em}$, specific internal energy ε, thermally and electromagnetically induced specific energy supply rates r^t and r^{em}, and heat flux h. Additionally, $\mathbf{e}^*, \mathbf{d}^*, \mathbf{h}^*, \mathbf{b}^*, \sigma^*$, and $\mathbf{j}^*$ are the effective electric field intensity, effective electric displacement, effective magnetic field intensity, effective magnetic induction, effective free charge density, and effective free current density. These *effective* fields are the electromagnetic fields acting on the deforming continuum as seen in its present configuration, measured with respect to a co-moving frame (Eringen & Maugin, 1990; Pao, 1978). The relations between the *effective* fields and the *standard* fields, the latter being measured with respect to a fixed frame, vary from model to model. As an example, the transformation equations of Minkowski[3] are

$$\mathbf{e}^* = \mathbf{e} + \mathbf{v} \times \mathbf{b}, \quad \mathbf{d}^* = \mathbf{d}, \quad \mathbf{h}^* = \mathbf{h} - \mathbf{v} \times \mathbf{d}, \quad \mathbf{b}^* = \mathbf{b}, \quad \sigma^* = \sigma, \quad \mathbf{j}^* = \mathbf{j} - \sigma \mathbf{v}, \tag{2}$$

where the effective electromagnetic fields are denoted by superscript stars, and the standard electromagnetic fields are unstarred. Other sets of transformations are given by the Chu, Lorentz, and Statistical formulations, each based on a different set of principles and postulates (Pao, 1978).

The transformations (2) describing the effective electromagnetic fields that appear in Maxwell's equations must be consistent with invariance requirements. We note that the Maxwell Eqs. (1a)–(1d) are invariant under an extended Lorentz transformation, whereas the thermomechanical balance laws (1e)–(1h) are invariant under a Galilean transformation, resulting in ambiguity when the two sets of equations are coupled. For the purpose of this work, we assume non-relativistic effects, i.e., $|\mathbf{v}| \ll c$. The Lorentz invariance of the Maxwell equations can then be approximated as Galilean invariance for certain special classes of non-relativistic motions (Rajagopal & Ruzicka, 2001).

The continuous, bounded nature of the integrands in (1a)–(1h) enables the transport, divergence, and Stokes's theorems, and the requirement that (1a)–(1h) be global, i.e., true for the entire body and all subsets, enables the localization theorem. Assuming that the traction $\mathbf{t}$ and heat flux h are dependent on surface geometry only through the outward unit normal $\mathbf{n}$, so that $\mathbf{t} = \mathbf{Tn}$ and $h = \mathbf{q} \cdot \mathbf{n}$, application of the transport, divergence, Stokes's, and localization theorems to the Eulerian integral Eqs. (1a)–(1h) leads to the Eulerian pointwise equations

$$div \; \mathbf{b}^* = 0, \tag{3a}$$

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[3] The Minkowski formulation is motivated by Einstein's special theory of relativity (Hutter et al., 2006; Pao, 1978). In this approximation, the effective fields are related to the standard fields through semi-relativistic inverse Lorentz transformations.

$$curl\ \mathbf{e}^* = -(\mathbf{b}^*)' - curl\ (\mathbf{b}^* \times \mathbf{v}), \tag{3b}$$

$$div\ \mathbf{d}^* = \sigma^*, \tag{3c}$$

$$curl\ \mathbf{h}^* = (\mathbf{d}^*)' + curl\ (\mathbf{d}^* \times \mathbf{v}) + \sigma^*\ \mathbf{v} + \mathbf{j}^*, \tag{3d}$$

$$\dot{\rho} + \rho\ div\ \mathbf{v} = 0, \tag{3e}$$

$$\rho\dot{\mathbf{v}} = \rho(\mathbf{f}^m + \mathbf{f}^{em}) + div\ \mathbf{T}, \tag{3f}$$

$$\rho\ \mathbf{\Gamma}^{em} + \mathbf{T} - \mathbf{T}^{\mathrm{T}} = \mathbf{0}, \tag{3g}$$

$$\rho\dot{\varepsilon} = \mathbf{T} \cdot \mathbf{L} + \rho(r^t + r^{em}) - div\ \mathbf{q}, \tag{3h}$$

where $\mathbf{T}$ is the Cauchy stress, $\mathbf{q}$ is the Eulerian heat flux vector, $\mathbf{L} = grad\ \mathbf{v} = \partial\mathbf{v}/\partial\mathbf{x}$, is the Eulerian velocity gradient, $\mathbf{\Gamma}^{em}$ is a skew tensor whose corresponding axial vector is $\mathbf{c}^{em}$, and

$$\mathbf{u}' = \frac{\partial\mathbf{u}}{\partial t}, \quad \dot{\mathbf{u}} = \mathbf{u}' + (\mathbf{v} \cdot grad)\mathbf{u}$$

denote the Eulerian and material time derivatives of an arbitrary vector $\mathbf{u}$. In (3a)–(3h), div () denotes the Eulerian divergence, $curl$ () denotes the Eulerian curl, and $\mathbf{T}^{\mathrm{T}}$ denotes the transpose of $\mathbf{T}$. We emphasize that the pointwise Eqs. (3a)–(3h) are valid statements of the first principles because they are progenies of the global Eqs. (1a)–(1h).

The pointwise first principles (3a)–(3h) are supplemented by two additional sets of equations. One set, discussed in Section 3, quantifies the electromagnetically induced body force $\mathbf{f}^{em}$, body couple $\mathbf{c}^{em}$, and energy supply rate r^{em}. A second set, discussed in Sections 4–6, consists of state equations that characterize a general TEMM process by relating the independent and dependent variables in the first-principle equations.

3. Modeling of the electromagnetically induced coupling terms

Electromagnetic forces in deformable continua can be modeled as inertial terms, incorporated into the constitutive response, or introduced through an electromagnetically induced body force, body couple, and energy supply (Eringen & Maugin, 1990; Hutter et al., 2006). In this paper, the lattermost approach is adopted, i.e., the thermomechanical Eqs. (3e)–(3h) are coupled to the Maxwell Eqs. (3a)–(3d) through the electromagnetic body force $\mathbf{f}^{em}$, body couple $\mathbf{c}^{em}$ (or, equivalently, $\mathbf{\Gamma}^{em}$), and energy supply rate r^{em}.

In principle, the continuum or macroscopic-scale coupling terms $\mathbf{f}^{em}$, $\mathbf{c}^{em}$, and r^{em} can be derived from the atomic or microscopic-scale electromagnetic problem by solving for the forces created by moving charges at the atomic level (Eringen & Maugin, 1990): Conceptually, knowledge of the position, velocity, and charge of each discrete particle enables the electromagnetic fields acting on each such particle to be calculated. With these atomic-scale electromagnetic fields known, the continuum electromagnetic fields $\mathbf{e}^*$, $\mathbf{d}^*$, $\mathbf{h}^*$, and $\mathbf{b}^*$ are deduced by statistically averaging the values of their discrete microscopic counterparts over an infinitesimal volume element (De Groot & Suttorp, 1972). Similarly, the continuum body force $\mathbf{f}^{em}$, body couple $\mathbf{c}^{em}$, and energy supply rate r^{em} are computed by statistically averaging the Lorentz forces (and accompanying couples and energies) acting on the charged particles. Thus, if the solution of the microscopic problem is known, the continuum coupling terms $\mathbf{f}^{em}$, $\mathbf{c}^{em}$, and r^{em} can be *deduced* using statistical averaging techniques.

In practice, however, the atomic details (e.g., position, velocity, and charge of each discrete particle) required to generate the solution of the microscopic problem are unavailable. Hence, the continuum-scale body force $\mathbf{f}^{em}$, body couple $\mathbf{c}^{em}$, and energy supply rate r^{em} modeling the effect of the atomic-scale forces, couples, and energies induced by moving charges must be *postulated*. These postulates are generally motivated by either atomic physics or empiricism. One such model for a polarizable, magnetizable, deformable continuum is coined the Maxwell–Minkowski formulation (Hutter et al., 2006):

$$\rho\mathbf{f}^{em} = \sigma^*\mathbf{e}^* + \mathbf{j}^* \times \mathbf{b}^* + (grad\ \mathbf{e}^*)^{\mathrm{T}}\mathbf{p}^* + \mu_o(grad\ \mathbf{h}^*)^{\mathrm{T}}\mathbf{m}^* + \mathbf{d}^* \times \mathbf{b}^* + \mathbf{d}^* \times \mathbf{b}^*, \tag{4a}$$

$$\rho\mathbf{\Gamma}^{em} = (\mathbf{e}^* \otimes \mathbf{p}^* - \mathbf{p}^* \otimes \mathbf{e}^*) + \mu_o(\mathbf{h}^* \otimes \mathbf{m}^* - \mathbf{m}^* \otimes \mathbf{h}^*), \tag{4b}$$

$$\rho r^{em} = \mathbf{j}^* \cdot \mathbf{e}^* + \rho\mathbf{e}^* \cdot \overline{\left(\frac{\dot{\mathbf{p}^*}}{\rho}\right)} + \rho\mu_o\mathbf{h}^* \cdot \overline{\left(\frac{\dot{\mathbf{m}^*}}{\rho}\right)}, \tag{4c}$$

where () $\otimes$ () denotes the dyadic product of two vectors,

$$\mathbf{u}^* = \dot{\mathbf{u}} + \mathbf{u}\ (div\ \mathbf{v}) - (grad\ \mathbf{v})\mathbf{u} = \mathbf{u}' + curl\ (\mathbf{u} \times \mathbf{v}) + \mathbf{v}\ (div\ \mathbf{u}) \tag{5}$$

is a convected rate of an arbitrary vector $\mathbf{u}$, and

$$\mathbf{p}^* = \mathbf{d}^* - \epsilon_o \mathbf{e}^*, \quad \mathbf{m}^* = \frac{1}{\mu_o}\mathbf{b}^* - \mathbf{h}^* \tag{6}$$

are the effective electric polarization and effective magnetization, with ϵ_o and μ_o the permittivity and permeability *in vacuo*.

The Maxwell–Minkowski model (4a)–(4c) has as special cases interaction theories describing forces exerted by electrostatic fields in polarizable solids (Maxwell, 1873) and magnetostatic fields in magnetizable solids (Brown, 1966). Use of (4a)–(4c) in balance of linear momentum (3f), balance of angular momentum (3g), and the first law of thermodynamics (3h) yields[4]

$$\rho\dot{\mathbf{v}} = \rho\mathbf{f}^m + \sigma^*\mathbf{e}^* + \mathbf{j}^* \times \mathbf{b}^* + (grad\ \mathbf{e}^*)^\mathsf{T}\mathbf{p}^* + \mu_o(grad\ \mathbf{h}^*)^\mathsf{T}\mathbf{m}^* + \mathbf{d}^* \times \mathbf{b}^* + \mathbf{d}^* \times \mathbf{b}^* + di\nu\ \mathbf{T}, \tag{7a}$$

$$\mathbf{T} - \mathbf{T}^\mathsf{T} = (\mathbf{p}^* \otimes \mathbf{e}^* - \mathbf{e}^* \otimes \mathbf{p}^*) + \mu_o(\mathbf{m}^* \otimes \mathbf{h}^* - \mathbf{h}^* \otimes \mathbf{m}^*), \tag{7b}$$

$$\dot{\varepsilon} = \frac{1}{\rho_R}\mathbf{P} \cdot \dot{\mathbf{F}} + \mathbf{e}^* \cdot \overline{\left(\frac{\mathbf{p}^*}{\rho}\right)} + \mu_o\mathbf{h}^* \cdot \overline{\left(\frac{\mathbf{m}^*}{\rho}\right)} + r^t + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho}di\nu\ \mathbf{q}, \tag{7c}$$

where

$$\mathbf{P} = J\mathbf{T}\mathbf{F}^{-\mathsf{T}} \tag{8}$$

is the first Piola–Kirchhoff stress, $J = \rho_R/\rho$ is the determinant of the deformation gradient $\mathbf{F} = Grad\ \mathbf{x} = \partial\mathbf{x}/\partial\mathbf{X}, \rho_R$ is the uniform reference density, and $\mathbf{X}$ is the reference position of a continuum particle.

4. State equations rendered by the second law of thermodynamics: energy formulations

In this section, we demonstrate how restrictions imposed by the second law of thermodynamics can be used to develop a catalogue of free energies and thermodynamic state equations for characterizing fully coupled thermo-electro-magneto-mechanical (TEMM) materials.

4.1. The Clausius–Duhem inequality

In this paper, as our particular statement of the second law, we adopt the Clausius–Duhem inequality (Coleman & Noll, 1963; Truesdell & Noll, 1992; Truesdell & Toupin, 1960)

$$\frac{d}{dt}\int_\mathcal{V}\rho\eta d\nu \geqslant \int_\mathcal{V}\rho\frac{r^t}{\theta}d\nu - \int_{\partial\mathcal{V}}\frac{h}{\theta}da, \tag{9}$$

whose pointwise version is

$$\rho\dot{\eta} - \rho\frac{r^t}{\theta} + di\nu\left(\frac{\mathbf{q}}{\theta}\right) \geqslant 0, \tag{10}$$

where θ is the absolute temperature, η is the specific entropy, and we have used $h = \mathbf{q} \cdot \mathbf{n}$. We note that alternative statements of the second law have been presented and investigated by numerous authors, e.g., Gurtin and Williams (1966), Liu and Müller (1972), Hutter (1975), Hutter (1977) and Green and Naghdi (1977). We also mention an approach to the thermodynamic treatment of continua recently set forth by Rajagopal & Srinivasa (see, for instance, Rajagopal & Srinivasa (1998) and Rajagopal & Srinivasa (2004)) that appeals to the maximization of the rate of entropy production.

Inequality (10) is then algebraically combined with the first law of thermodynamics (7c) to produce the reduced Clausius–Duhem inequality

$$-\dot{\varepsilon} + \frac{1}{\rho_R}\mathbf{P} \cdot \dot{\mathbf{F}} + \theta\dot{\eta} + \mathbf{e}^* \cdot \overline{\left(\frac{\mathbf{p}^*}{\rho}\right)} + \mu_o\mathbf{h}^* \cdot \overline{\left(\frac{\mathbf{m}^*}{\rho}\right)} + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\ \theta \geqslant 0. \tag{11}$$

Analogous to classical thermodynamics (Callen, 1985), this *fundamental* statement of the second law consists of contributions from *conjugate pairs* of thermal, electrical, magnetic, and mechanical quantities. Each of these conjugate pairs (or work conjugates) is the product of an *extensive* quantity in rate form ($\mathbf{F}, \eta, \mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$) and an *intensive* quantity in non-rate form ($\mathbf{P}, \theta, \mathbf{e}^*$, and $\mathbf{h}^*$). Note that when we use the adjective 'extensive', it is tacit that we mean an extensive quantity per unit

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[4] Recall that electromagnetic effects in the thermomechanical balance laws can be modeled as an electromagnetically induced body force, body couple, and energy supply, or, alternatively, incorporated into the constitutive response. The latter is accomplished by defining a total stress tensor $\tau = \mathbf{T} + \mathbf{T}^{em}$ that consists of contributions from the Cauchy stress tensor $\mathbf{T}$ and the Maxwell stress tensor $\mathbf{T}^{em}$. $\mathbf{T}^{em}$ is defined so that its skew symmetric part is $\rho\Gamma^{em}$ and its divergence is $\rho\mathbf{f}^{em}$. (For the Maxwell–Minkowski formulation (4a)–(4c), $\mathbf{T}^{em} = \mathbf{e}^* \otimes \mathbf{d}^* + \mathbf{h}^* \otimes \mathbf{b}^* - \frac{1}{2}(\epsilon_o\mathbf{e}^* \cdot \mathbf{e}^* + \mu_o\mathbf{h}^* \cdot \mathbf{h}^*)\mathbf{I}$, (Hutter et al., 2006).) By formulating the first principles and the constitutive equations in terms of the total stress tensor τ instead of the Cauchy stress tensor $\mathbf{T}$, explicit coupling between the electromagnetic fields and the thermomechanical fields is eliminated from Eqs. (7a)–(7c), and is instead accounted for in the constitutive equations for τ.

mass. The utility of the second law, and our identification of the extensive and intensive quantities, will become apparent shortly.

4.2. The fundamental or internal energy formulation

The pointwise field Eqs. (3a)–(3e) and (7a)–(7c) constitute the first principles of our model, true for all TEMM materials. For the purposes of developing the thermodynamic state equations (Callen, 1985; Moran, Shapiro, Boettner, & Bailey, 2010) that supplement these first principles and characterize particular TEMM materials, we conceptually divide the fields appearing in (3a)–(3e) and (7a)–(7c) into three groups

$$\{\mathbf{x}, \eta, \mathbf{p}^*, \mathbf{m}^*\}, \quad \{\mathbf{P}, \theta, \mathbf{e}^*, \mathbf{h}^*, \varepsilon, \mathbf{q}, \mathbf{j}^*\}, \quad \{\rho, \mathbf{f}^m, r^t, \sigma^*\}, \tag{12}$$

denoted the independent variables, dependent variables, and balancing terms, respectively.[5] (Note that the set of independent variables contains slots occupied by one mechanical, one thermal, one electrical, and one magnetic quantity, respectively, from left to right.) Hence, the fields $\mathbf{P}, \theta, \mathbf{e}^*, \mathbf{h}^*, \varepsilon, \mathbf{q}$, and $\mathbf{j}^*$ are determined from constitutive equations that, in general, depend on the history of the motion $\mathbf{x}$, specific entropy η, effective electric polarization $\mathbf{p}^*$, and effective magnetization $\mathbf{m}^*$, and possibly their rates or gradients. A group of quantities $\mathbf{x}, \eta, \mathbf{p}^*, \mathbf{m}^*, \mathbf{P}, \theta, \mathbf{e}^*, \mathbf{h}^*, \varepsilon, \mathbf{q}, \mathbf{j}^*, \rho, \mathbf{f}^m, r^t$, and σ^* that satisfy the governing Eqs. (3a)–(3e) and (7a)–(7c) for all space and time in the domain of interest describes a thermo-electro-magneto-mechanical (TEMM) process.

In this paper, we assume that the material response is path independent, reversible, and rate insensitive. This implies that the deformation, although it may be large, is elastic and fully recoverable, and the material only undergoes non-dissipative TEMM processes. It follows, then, that these non-dissipative TEMM processes can be approximated as quasi-static thermodynamic processes, which can be completely described through the principles of classical equilibrium thermodynamics[6] (Callen, 1985).

Analogous to classical equilibrium thermodynamics, our fundamental energy potential is the specific internal energy ε, which employs *extensive* quantities as its independent variables. Hence, for the fundamental formulation, the *natural* independent variables are the *extensive* quantities $\mathbf{F}, \eta, \mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$ appearing as rates in (11), and the *natural* dependent variables are the conjugate *intensive* quantities $\mathbf{P}, \theta, \mathbf{e}^*$, and $\mathbf{h}^*$. Thus, for thermodynamic consistency, and to respect the reversible elastic nature of the process, we adjust the division (12) so that the dependence of the response on the present position $\mathbf{x}$, polarization $\mathbf{p}^*$, and magnetization $\mathbf{m}^*$ is through the deformation gradient $\mathbf{F} = Grad\,\mathbf{x}, \mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$, respectively. Additionally, we demand that the response depends on these independent variables only through their values at the present time t, not their histories, rates, or gradients, i.e.,

$$\mathbf{P} = \breve{\mathbf{P}}\left(\mathbf{F}, \eta, \frac{\mathbf{p}^*}{\rho}, \frac{\mathbf{m}^*}{\rho}\right), \quad \theta = \breve{\theta}\left(\mathbf{F}, \eta, \frac{\mathbf{p}^*}{\rho}, \frac{\mathbf{m}^*}{\rho}\right), \quad \mathbf{e}^* = \breve{\mathbf{e}}^*\left(\mathbf{F}, \eta, \frac{\mathbf{p}^*}{\rho}, \frac{\mathbf{m}^*}{\rho}\right), \quad \mathbf{h}^* = \breve{\mathbf{h}}^*\left(\mathbf{F}, \eta, \frac{\mathbf{p}^*}{\rho}, \frac{\mathbf{m}^*}{\rho}\right), \quad \varepsilon = \breve{\varepsilon}\left(\mathbf{F}, \eta, \frac{\mathbf{p}^*}{\rho}, \frac{\mathbf{m}^*}{\rho}\right), \tag{13}$$

where the superscript breve is used to distinguish a function from its value.

In what follows, we demonstrate how restrictions imposed by the second law of thermodynamics (11) yield a set of state equations that provide the dependent variables $\mathbf{P}, \theta, \mathbf{e}^*$, and $\mathbf{h}^*$ (the intensive quantities) as partial derivatives of the specific internal energy ε (the fundamental thermodynamic potential) with respect to the independent variables $\mathbf{F}, \eta, \mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$ (the extensive quantities), respectively. (In order to obtain the *constitutive equations* that characterize a particular TEMM material, the *state equations* are amended to satisfy invariance, angular momentum, and material symmetry requirements, and the material constants (e.g., permeability, permittivity, Young's modulus, coefficient of thermal expansion) are determined experimentally).

Use of the chain rule on $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$ gives

$$\dot{\varepsilon} = \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}} \cdot \dot{\mathbf{F}} + \frac{\partial \breve{\varepsilon}}{\partial \eta} \dot{\eta} + \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}^*}{\rho}\right)} \cdot \overline{\left(\frac{\mathbf{p}^*}{\rho}\right)} + \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}^*}{\rho}\right)} \cdot \overline{\left(\frac{\mathbf{m}^*}{\rho}\right)} \tag{14}$$

and substitution of this result into the second law (11) leads to

$$\left(\frac{1}{\rho_R}\mathbf{P} - \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}}\right) \cdot \dot{\mathbf{F}} + \left(\theta - \frac{\partial \breve{\varepsilon}}{\partial \eta}\right)\dot{\eta} + \left(\mathbf{e}^* - \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}^*}{\rho}\right)}\right) \cdot \overline{\left(\frac{\mathbf{p}^*}{\rho}\right)} + \left(\mu_o\mathbf{h}^* - \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}^*}{\rho}\right)}\right) \cdot \overline{\left(\frac{\mathbf{m}^*}{\rho}\right)} + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0. \tag{15}$$

As is customary, we demand that the second law holds for all processes (Coleman & Noll, 1963). Since the coefficients of the rates ($\dot{\mathbf{F}}, \dot{\eta}$, etc.) in inequality (15) are independent of the rates themselves, and the rates may be varied independently and are arbitrary, it follows that the coefficients vanish, i.e.,

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[5] In this division, $\mathbf{b}^*, \mathbf{d}^*$, and $\mathbf{T}$ are relegated to *secondary dependent variables*, i.e., variables that can be calculated from the independent and *primary dependent variables* in (12) using the algebraic relationships (6). Also note that $\mathbf{v}, \mathbf{F}$, and $\mathbf{L}$ can be calculated from $\mathbf{x}$ using vector and tensor calculus.

[6] Although we derive our constitutive framework assuming that the system is in thermodynamic equilibrium (i.e., it only undergoes reversible processes), our framework can be used to model irreversible systems that operate in a regime close to equilibrium (Callen, 1985).

$$\mathbf{P} = \rho_R \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}}, \quad \theta = \frac{\partial \breve{\varepsilon}}{\partial \eta}, \quad \mathbf{e}^* = \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}^*}{\rho}\right)}, \quad \mathbf{h}^* = \frac{1}{\mu_o} \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}^*}{\rho}\right)}. \tag{16}$$

What remains of inequality (15), i.e., $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta} \mathbf{q} \cdot grad\, \theta \geqslant 0$, is called the residual dissipation inequality.[7] We collectively coin the set of independent variables $\{\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho\}$, the thermodynamic energy potential $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$, and the state Eqs. (16) the *fundamental formulation* or *internal energy formulation*.

4.3. The modified internal energy formulation

From an experimental point of view, it is more practical to control the electric polarization $\mathbf{p}^*$ and magnetization $\mathbf{m}^*$ than the "mixed" quantities $\mathbf{p}^*/\rho$ and $\mathbf{m}^*/\rho$. Hence, we modify the internal energy formulation to accommodate the use of $\mathbf{p}^*$ and $\mathbf{m}^*$ as the electromagnetic independent variables.

We proceed by using the chain rule

$$\overline{\left(\frac{\mathbf{p}^*}{\rho}\right)}^{\cdot} = \frac{1}{\rho} \dot{\mathbf{p}}^* + \frac{1}{\rho} \left(\mathbf{F}^{-T} \cdot \dot{\mathbf{F}}\right) \mathbf{p}^*, \quad \overline{\left(\frac{\mathbf{m}^*}{\rho}\right)}^{\cdot} = \frac{1}{\rho} \dot{\mathbf{m}}^* + \frac{1}{\rho} \left(\mathbf{F}^{-T} \cdot \dot{\mathbf{F}}\right) \mathbf{m}^* \tag{17}$$

to rewrite the fundamental form (11) of the Clausius–Duhem inequality:

$$-\dot{\varepsilon} + \left[\frac{1}{\rho_R} \mathbf{P} + \frac{1}{\rho} (\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^*) \mathbf{F}^{-T}\right] \cdot \dot{\mathbf{F}} + \theta \dot{\eta} + \frac{1}{\rho} \mathbf{e}^* \cdot \dot{\mathbf{p}}^* + \frac{\mu_o}{\rho} \mathbf{h}^* \cdot \dot{\mathbf{m}}^* + \frac{1}{\rho} \mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta} \mathbf{q} \cdot grad\, \theta \geqslant 0, \tag{18}$$

where we have used

$$\overline{\left(\frac{1}{\rho}\right)}^{\cdot} = \frac{1}{\rho} div\, \mathbf{v}, \quad div\, \mathbf{v} = tr\, \mathbf{L} = tr(\dot{\mathbf{F}} \mathbf{F}^{-1}) = \mathbf{F}^{-T} \cdot \dot{\mathbf{F}}. \tag{19}$$

In the modified form (18) of the second law, polarization $\mathbf{p}^*$ and magnetization $\mathbf{m}^*$ appear as rates (i.e., natural independent variables). Accordingly, the thermodynamic potential ε is a function of $\mathbf{F}, \eta, \mathbf{p}^*$, and $\mathbf{m}^*$, i.e., $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$; a superscript bar is used instead of a superscript breve to signify a different internal energy function with the same value. Use of the chain rule gives

$$\dot{\varepsilon} = \frac{\partial \bar{\varepsilon}}{\partial \mathbf{F}} \cdot \dot{\mathbf{F}} + \frac{\partial \bar{\varepsilon}}{\partial \eta} \dot{\eta} + \frac{\partial \bar{\varepsilon}}{\partial \mathbf{p}^*} \cdot \dot{\mathbf{p}}^* + \frac{\partial \bar{\varepsilon}}{\partial \mathbf{m}^*} \cdot \dot{\mathbf{m}}^* \tag{20}$$

and substitution of this result into (18) leads to

$$\left(\frac{1}{\rho_R} \mathbf{P} + \frac{1}{\rho} (\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^*) \mathbf{F}^{-T} - \frac{\partial \bar{\varepsilon}}{\partial \mathbf{F}}\right) \cdot \dot{\mathbf{F}} + \left(\theta - \frac{\partial \bar{\varepsilon}}{\partial \eta}\right) \dot{\eta} + \left(\frac{1}{\rho} \mathbf{e}^* - \frac{\partial \bar{\varepsilon}}{\partial \mathbf{p}^*}\right) \cdot \dot{\mathbf{p}}^* + \left(\frac{\mu_o}{\rho} \mathbf{h}^* - \frac{\partial \bar{\varepsilon}}{\partial \mathbf{m}^*}\right) \cdot \dot{\mathbf{m}}^* + \frac{1}{\rho} \mathbf{j}^* \cdot \mathbf{e}^*$$
$$- \frac{1}{\rho\theta} \mathbf{q} \cdot grad\, \theta \geqslant 0. \tag{21}$$

Since the coefficients of $\dot{\mathbf{F}}, \dot{\eta}, \dot{\mathbf{p}}^*$, and $\dot{\mathbf{m}}^*$ in inequality (21) are independent of the rates, and the rates may be varied independently and are arbitrary, it follows that the coefficients vanish, i.e.,

$$\mathbf{P} = \rho_R \frac{\partial \bar{\varepsilon}}{\partial \mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^*) \mathbf{F}^{-T}, \quad \theta = \frac{\partial \bar{\varepsilon}}{\partial \eta}, \quad \mathbf{e}^* = \rho \frac{\partial \bar{\varepsilon}}{\partial \mathbf{p}^*}, \quad \mathbf{h}^* = \frac{\rho}{\mu_o} \frac{\partial \bar{\varepsilon}}{\partial \mathbf{m}^*}. \tag{22}$$

We collectively coin the set of independent variables $\{\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*\}$, the thermodynamic energy potential $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$, and the state Eqs. (22) the *modified internal energy formulation*.

We emphasize that the second term on the right-hand side of Eq. (22)$_1$ is generally absent in the existing literature for coupled material behavior, e.g., Smith (2005), wherein material response is customarily modeled in the small-deformation regime. It can be shown that this term vanishes from our more general large-deformation constitutive framework (and other finite-deformation constitutive models, e.g., Hutter et al. (2006)) when the standard assumptions of small-deformation theory are invoked.

4.4. Other energetic formulations

In principle, knowledge of the internal energy $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$ or $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$ is necessary and sufficient to characterize any general TEMM process. (Said differently, specification of the internal energy alone determines $\mathbf{P}, \theta, \mathbf{e}^*$, and $\mathbf{h}^*$; refer to the state Eqs. (16) and (22).) However, process characterization is most straightforwardly accomplished

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[7] The residual dissipation inequality quantifies irreversibilities in a thermodynamic process, in this case Joule heating and heat conduction, both transport processes (Hutter et al., 2006). Accordingly, unlike the other dependent variables (see Eq. (16)), the conductive current $\mathbf{j}^*$ and heat flux $\mathbf{q}$ are not derivable from a thermodynamic potential.

Table 1
Energy potentials and associated independent variables.

Family 1	Family 2	Family 3	Family 4
$\breve{\varepsilon}$, or $\bar{\varepsilon}$	$E^{F\theta pm}$	$E^{P\eta pm}$	$E^{P\theta pm}$
$E^{F\eta em}$	$E^{F\theta em}$	$E^{P\eta em}$	$E^{P\theta em}$
$E^{F\eta ph}$	$E^{F\theta ph}$	$E^{P\eta ph}$	$E^{P\theta ph}$
$E^{F\eta eh}$	$E^{F\theta eh}$	$E^{P\eta eh}$	$E^{P\theta eh}$

when the independent and dependent variables synchronize with those one wishes to control and measure, respectively. In experiments, it is generally more convenient to control intensive quantities than extensive quantities. For example, temperature is easier to control than entropy or heat.

To change any or all of the independent variables from extensive to intensive, a new thermodynamic potential function is defined through a Legendre transformation of the internal energy (Callen, 1985). In this section, we investigate all possible Legendre transforms $E^{(a)(b)(c)(d)}$ of the internal energy, where the superscript letters (a), (b), (c), and (d) are placeholders for an appropriate mechanical, thermal, electrical, and magnetic independent variable, respectively. This compact notation denotes that the Legendre-transformed energy potential $E^{F\theta pm}$, for instance, is a function of deformation, temperature, effective electric polarization, and effective magnetization.

As shown in Table 1, we categorize the energy potentials into four families, each employing a common set of thermomechanical independent variables: Family 1, deformation and entropy (both extensive); Family 2, deformation (extensive) and temperature (intensive); Family 3, stress (intensive) and entropy (extensive); and Family 4, stress and temperature (both intensive). Either electric field intensity or electric polarization, and either magnetic field intensity or magnetization, complete the set of independent variables. Thus, within each family, there are four different energy potentials, each associated with one of the four possible sets of independent variables.

With the formulations of Sections 4.2 and 4.3 as templates, in the subsections that follow, we derive the state equations associated with some of the potentials listed in Table 1. These representative cases are intended to provide the reader with a sufficient blueprint to reproduce the full catalogue presented in Appendix A.

4.4.1. Family 1: entropy-deformation family

In Family 1, the common thermomechanical independent variables are deformation **F** and specific entropy η, both extensive quantities. Energy potentials that use deformation, entropy, and either or both of the intensive electromagnetic quantities as independent variables are introduced as Legendre transformations of the internal energy $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$.

Entropy-deformation-electric field-magnetic field formulation

In this formulation, we promote the intensive electromagnetic quantities $\mathbf{e}^*$ and $\mathbf{h}^*$ to independent variables, and concomitantly relegate $\mathbf{p}^*$ and $\mathbf{m}^*$ to dependent variables. The thermodynamic energy potential $E^{F\eta eh}$ is defined as the Legendre transformation of internal energy $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$ with respect to the electromagnetic variables, from $\mathbf{p}^*$ to $\mathbf{e}^*$ and $\mathbf{m}^*$ to $\mathbf{h}^*$,

$$E^{F\eta eh} = \varepsilon - \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^* - \frac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*, \tag{23}$$

from which

$$\dot{E}^{F\eta eh} = \dot{\varepsilon} - \frac{1}{\rho}\left[(\mathbf{e}^* \cdot \mathbf{p}^*)\mathbf{F}^{-T} \cdot \dot{\mathbf{F}} + \mathbf{e}^* \cdot \dot{\mathbf{p}}^* + \mathbf{p}^* \cdot \dot{\mathbf{e}}^*\right] - \frac{\mu_o}{\rho}\left[(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T} \cdot \dot{\mathbf{F}} + \mathbf{h}^* \cdot \dot{\mathbf{m}}^* + \mathbf{m}^* \cdot \dot{\mathbf{h}}^*\right] \tag{24}$$

follows, where we have used Eq. (19). Substituting (24) into (18) leads to

$$-\dot{E}^{F\eta eh} + \frac{1}{\rho_R}\mathbf{P} \cdot \dot{\mathbf{F}} + \theta\dot{\eta} - \frac{1}{\rho}\mathbf{p}^* \cdot \dot{\mathbf{e}}^* - \frac{\mu_o}{\rho}\mathbf{m}^* \cdot \dot{\mathbf{h}}^* + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0, \tag{25}$$

a statement of the second law of thermodynamics for this formulation.[8] Via the chain rule,

$$\dot{E}^{F\eta eh} = \frac{\partial E^{F\eta eh}}{\partial \mathbf{F}} \cdot \dot{\mathbf{F}} + \frac{\partial E^{F\eta eh}}{\partial \eta}\dot{\eta} + \frac{\partial E^{F\eta eh}}{\partial \mathbf{e}^*} \cdot \dot{\mathbf{e}}^* + \frac{\partial E^{F\eta eh}}{\partial \mathbf{h}^*} \cdot \dot{\mathbf{h}}^*, \tag{26}$$

and, upon subsequent use of (26) in (25), it follows that

$$\left(\frac{1}{\rho_R}\mathbf{P} - \frac{\partial E^{F\eta eh}}{\partial \mathbf{F}}\right) \cdot \dot{\mathbf{F}} + \left(\theta - \frac{\partial E^{F\eta eh}}{\partial \eta}\right)\dot{\eta} - \left(\frac{\mathbf{p}^*}{\rho} + \frac{\partial E^{F\eta eh}}{\partial \mathbf{e}^*}\right) \cdot \dot{\mathbf{e}}^* - \left(\frac{\mu_o}{\rho}\mathbf{m}^* + \frac{\partial E^{F\eta eh}}{\partial \mathbf{h}^*}\right) \cdot \dot{\mathbf{h}}^* + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0. \tag{27}$$

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[8] All formulations presented in this paper have a corresponding second law statement.

Since the coefficients of $\dot{\mathbf{F}}, \dot{\eta}, \dot{\mathbf{e}}^*$, and $\dot{\mathbf{h}}^*$ are independent of the rates, and the rates may be varied independently and are arbitrary, it follows that the coefficients vanish, i.e.,

$$\mathbf{P} = \rho_R \frac{\partial E^{F\eta eh}}{\partial \mathbf{F}}, \quad \theta = \frac{\partial E^{F\eta eh}}{\partial \eta}, \quad \mathbf{p}^* = -\rho \frac{\partial E^{F\eta eh}}{\partial \mathbf{e}^*}, \quad \mathbf{m}^* = -\frac{\rho}{\mu_o} \frac{\partial E^{F\eta eh}}{\partial \mathbf{h}^*}, \tag{28}$$

with $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta} \mathbf{q} \cdot grad\,\theta \geqslant 0$ the residual dissipation inequality.

4.4.2. Family 2: temperature-deformation family

In Family 2, we introduce Legendre-transformed energy potentials that use temperature, an intensive quantity, rather than specific entropy, an extensive quantity, as the thermal independent variable.

Temperature-deformation-electric field-magnetization formulation

In this formulation, $\mathbf{F}, \theta, \mathbf{e}^*$, and $\mathbf{m}^*$ are the independent variables. We define the energy potential $E^{F\theta em}$ as the Legendre transformation of internal energy $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$ with respect to the thermal and electrical variables, from η to θ and $\mathbf{p}^*$ to $\mathbf{e}^*$,

$$E^{F\theta em} = \varepsilon - \theta\eta - \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*, \tag{29}$$

whose rate form

$$\dot{E}^{F\theta em} = \dot{\varepsilon} - \theta\dot{\eta} - \eta\dot{\theta} - \frac{1}{\rho}\left[(\mathbf{e}^* \cdot \mathbf{p}^*)\mathbf{F}^{-T} \cdot \dot{\mathbf{F}} + \mathbf{e}^* \cdot \dot{\mathbf{p}}^* + \mathbf{p}^* \cdot \dot{\mathbf{e}}^*\right], \tag{30}$$

when inserted in (18), yields the second law statement

$$-\dot{E}^{F\theta em} + \left[\frac{1}{\rho_R}\mathbf{P} + \frac{\mu_o}{\rho}(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}\right] \cdot \dot{\mathbf{F}} - \eta\dot{\theta} - \frac{1}{\rho}\mathbf{p}^* \cdot \dot{\mathbf{e}}^* + \frac{\mu_o}{\rho}\mathbf{h}^* \cdot \dot{\mathbf{m}}^* + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0. \tag{31}$$

Use of the chain rule on $\dot{E}^{F\theta em}$ leads to the state equations

$$\mathbf{P} = \rho_R \frac{\partial E^{F\theta em}}{\partial \mathbf{F}} - \mu_o J(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}, \quad \eta = -\frac{\partial E^{F\theta em}}{\partial \theta}, \quad \mathbf{p}^* = -\rho \frac{\partial E^{F\theta em}}{\partial \mathbf{e}^*}, \quad \mathbf{h}^* = \frac{\rho}{\mu_o} \frac{\partial E^{F\theta em}}{\partial \mathbf{m}^*}. \tag{32}$$

4.4.3. Family 3: entropy-stress family

In Family 3, we introduce energy potentials that utilize the intensive quantity $\mathbf{P}$, rather than the extensive quantity $\mathbf{F}$, as the mechanical independent variable. We construct sets of state equations using the fundamental form of the Clausius–Duhem inequality (11), where $\mathbf{P}$ and $\mathbf{F}$ form an intensive–extensive conjugate pair and $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$.[9] In the formulation that follows, we promote the intensive quantities $\mathbf{P}$ and $\mathbf{h}^*$ to independent variables, and concomitantly relegate the extensive quantities $\mathbf{F}$ and $\mathbf{m}^*/\rho$ to dependent variables.

Entropy-stress-polarization-magnetic field formulation

In this formulation, the independent variables are $\mathbf{P}, \eta, \mathbf{p}^*/\rho$, and $\mathbf{h}^*$. $E^{P\eta ph}$, the thermodynamic energy potential,[10] is defined as the Legendre transformation of internal energy $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$ with respect to the mechanical and magnetic variables, from $\mathbf{F}$ to $\mathbf{P}$ and $\mathbf{m}^*/\rho$ to $\mathbf{h}^*$,

$$E^{P\eta ph} = \varepsilon - \frac{1}{\rho_R}\mathbf{P} \cdot \mathbf{F} - \mu_o \mathbf{h}^* \cdot \frac{\mathbf{m}^*}{\rho}, \tag{33}$$

or, in rate form,

$$\dot{E}^{P\eta ph} = \dot{\varepsilon} - \frac{1}{\rho_R}\mathbf{P} \cdot \dot{\mathbf{F}} - \frac{1}{\rho_R}\mathbf{F} \cdot \dot{\mathbf{P}} - \mu_o \frac{\mathbf{m}^*}{\rho} \cdot \dot{\mathbf{h}}^* - \mu_o \mathbf{h}^* \cdot \overline{\left(\frac{\mathbf{m}^*}{\rho}\right)}. \tag{34}$$

Inserting (34) in (11) yields the second law statement

$$-\dot{E}^{P\eta ph} - \frac{1}{\rho_R}\mathbf{F} \cdot \dot{\mathbf{P}} + \theta\dot{\eta} + \mathbf{e}^* \cdot \overline{\left(\frac{\mathbf{p}^*}{\rho}\right)} - \mu_o \frac{\mathbf{m}^*}{\rho} \cdot \dot{\mathbf{h}}^* + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0 \tag{35}$$

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[9] Note that $\mathbf{P}$ and $\mathbf{F}$ are not conjugate variables in the modified form of the Clausius–Duhem inequality (18), where $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$, necessitating our use of the fundamental form (11).

[10] For notational brevity, we do not distinguish $\mathbf{p}^*/\rho$ from $\mathbf{p}^*$ (nor $\mathbf{m}^*/\rho$ from $\mathbf{m}^*$) in the superscript of the energy potentials, as this difference can be straightforwardly ascertained from the corresponding state equations.

and application of the chain rule to $\dot{E}^{P\eta ph}$ leads to

$$-\left(\frac{1}{\rho_R}\mathbf{F}+\frac{\partial E^{P\eta ph}}{\partial \mathbf{P}}\right)\cdot\dot{\mathbf{P}}+\left(\theta-\frac{\partial E^{P\eta ph}}{\partial \eta}\right)\dot{\eta}+\left(\mathbf{e}^*-\frac{\partial E^{P\eta ph}}{\partial\left(\frac{\mathbf{p}^*}{\rho}\right)}\right)\cdot\overline{\left(\frac{\dot{\mathbf{p}}^*}{\rho}\right)}-\left(\frac{\mu_o}{\rho}\mathbf{m}^*+\frac{\partial E^{P\eta ph}}{\partial \mathbf{h}^*}\right)\cdot\dot{\mathbf{h}}^*$$

$$+\frac{1}{\rho}\mathbf{j}^*\cdot\mathbf{e}^*-\frac{1}{\rho\theta}\mathbf{q}\cdot grad\,\theta\geqslant 0,\tag{36}$$

from which the state equations

$$\mathbf{F}=-\rho_R\frac{\partial E^{P\eta ph}}{\partial\mathbf{P}},\quad\theta=\frac{\partial E^{P\eta ph}}{\partial\eta},\quad\mathbf{e}^*=\frac{\partial E^{P\eta ph}}{\partial\left(\frac{\mathbf{p}^*}{\rho}\right)},\quad\mathbf{m}^*=-\frac{\rho}{\mu_o}\frac{\partial E^{P\eta ph}}{\partial\mathbf{h}^*}\tag{37}$$

follow.

4.4.4. Family 4: temperature-stress family

In Family 4, we introduce Legendre-transformed energy potentials that use the intensive quantities stress and temperature, rather than the extensive quantities deformation and entropy, as the thermomechanical independent variables.

Temperature-stress-electric field-magnetic field formulation

The set of independent variables for this formulation is $\{\mathbf{P},\theta,\mathbf{e}^*,\mathbf{h}^*\}$. The corresponding thermodynamic energy potential $E^{P\theta eh}$ is defined as the Legendre transformation of internal energy $\varepsilon=\breve{\varepsilon}(\mathbf{F},\eta,\mathbf{p}^*/\rho,\mathbf{m}^*/\rho)$ with respect to the mechanical, thermal, electrical, and magnetic variables, from $\mathbf{F}$ to $\mathbf{P},\eta$ to $\theta,\mathbf{p}^*/\rho$ to $\mathbf{e}^*$, and $\mathbf{m}^*/\rho$ to $\mathbf{h}^*$,

$$E^{P\theta eh}=\varepsilon-\frac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F}-\theta\eta-\mathbf{e}^*\cdot\frac{\mathbf{p}^*}{\rho}-\mu_o\mathbf{h}^*\cdot\frac{\mathbf{m}^*}{\rho}.\tag{38}$$

Taking the rate of (38) gives

$$\dot{E}^{P\theta eh}=\dot{\varepsilon}-\frac{1}{\rho_R}\mathbf{P}\cdot\dot{\mathbf{F}}-\frac{1}{\rho_R}\mathbf{F}\cdot\dot{\mathbf{P}}-\theta\dot{\eta}-\eta\dot{\theta}-\mathbf{e}^*\cdot\overline{\left(\frac{\dot{\mathbf{p}}^*}{\rho}\right)}-\frac{\mathbf{p}^*}{\rho}\cdot\dot{\mathbf{e}}^*-\mu_o\mathbf{h}^*\cdot\overline{\left(\frac{\dot{\mathbf{m}}^*}{\rho}\right)}-\mu_o\frac{\mathbf{m}^*}{\rho}\cdot\dot{\mathbf{h}}^*\tag{39}$$

and substitution of this result into (11) yields

$$-\dot{E}^{P\theta eh}-\frac{1}{\rho_R}\mathbf{F}\cdot\dot{\mathbf{P}}-\eta\dot{\theta}-\frac{\mathbf{p}^*}{\rho}\cdot\dot{\mathbf{e}}^*-\mu_o\frac{\mathbf{m}^*}{\rho}\cdot\dot{\mathbf{h}}^*+\frac{1}{\rho}\mathbf{j}^*\cdot\mathbf{e}^*-\frac{1}{\rho\theta}\mathbf{q}\cdot grad\,\theta\geqslant 0.\tag{40}$$

Use of the chain rule on $\dot{E}^{P\theta eh}$ leads to the state equations

$$\mathbf{F}=-\rho_R\frac{\partial E^{P\theta eh}}{\partial\mathbf{P}},\quad\eta=-\frac{\partial E^{P\theta eh}}{\partial\theta},\quad\mathbf{p}^*=-\rho\frac{\partial E^{P\theta eh}}{\partial\mathbf{e}^*},\quad\mathbf{m}^*=-\frac{\rho}{\mu_o}\frac{\partial E^{P\theta eh}}{\partial\mathbf{h}^*}.\tag{41}$$

5. Energy formulations utilizing secondary independent variables

Recall that the internal energy $\varepsilon=\breve{\varepsilon}(\mathbf{F},\eta,\mathbf{p}^*/\rho,\mathbf{m}^*/\rho)$ is a function of the *extensive* quantities $\mathbf{F},\eta,\mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$. In Section 4, thermodynamic energy potentials employing one or more of the *intensive* quantities $\mathbf{P},\theta,\mathbf{e}^*$, and $\mathbf{h}^*$ as independent variables were introduced using Legendre transformations of ε. These *intensive–extensive conjugate pairs* or *work conjugates* (i.e., $\mathbf{P}$ and $\mathbf{F},\theta$ and $\eta,\mathbf{e}^*$ and $\mathbf{p}^*/\rho,\mathbf{h}^*$ and $\mathbf{m}^*/\rho$) appearing in the *fundamental form* of the second law of thermodynamics (11) are dictated by the choice (4c) of the electromagnetic energy r^{em}. It follows, then, that different choices of r^{em} (of which there are many options available in the literature; see, for instance, Eringen & Maugin (1990), Hutter et al. (2006), Pao & Hutter (1975) and Kovetz (2000)) lead to different sets of electromagnetic conjugate pairs in finite-deformation TEMM. For instance, the electric displacement $\mathbf{d}^*$ and magnetic induction $\mathbf{b}^*$, both extensive quantities, frequently appear as parts of a conjugate pair (see, for example, Green & Naghdi (1984), Kovetz (2000), Kankanala & Triantafyllidis (2004), Rajagopal & Ruzicka (2001) and Dorfmann & Ogden (2004a)).

The formalism of Legendre transforms presented in Section 4, however, does not enable us to use $\mathbf{d}^*$ and $\mathbf{b}^*$ as independent variables since they are not part of the electromagnetic work conjugates employed in this paper (i.e., $\mathbf{e}^*$ and $\mathbf{p}^*/\rho,\mathbf{h}^*$ and $\mathbf{m}^*/\rho$). We thus coin $\mathbf{d}^*$ and $\mathbf{b}^*$ *secondary* or *auxiliary quantities*. In this section, we overcome this limitation by, for the first time, presenting a mathematically rigorous procedure for introducing secondary electromagnetic quantities as independent variables.

5.1. Entropy-deformation-electric displacement-magnetic induction formulation

In this section, $\mathbf{F},\eta,\mathbf{d}^*$, and $\mathbf{b}^*$ are selected as the independent variables. However, as discussed above, the *secondary quantities* $\mathbf{d}^*$ and $\mathbf{b}^*$ cannot be introduced as independent variables through a conventional Legendre transformation of the internal energy. We circumvent this by positing a *Legendre-type transformation* of $\varepsilon=\bar{\varepsilon}(\mathbf{F},\eta,\mathbf{p}^*,\mathbf{m}^*)$, i.e.,

$$E^{F\eta db} = \varepsilon + \frac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^* + \frac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*, \tag{42}$$

whose rate form is

$$\dot{E}^{F\eta db} = \dot{\varepsilon} + \frac{1}{2\rho}\left(\epsilon_o\mathbf{e}^* \cdot \mathbf{e}^* + \mu_o\mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-\mathsf{T}} \cdot \dot{\mathbf{F}} + \frac{1}{\rho}\left(\epsilon_o\mathbf{e}^* \cdot \dot{\mathbf{e}}^* + \mu_o\mathbf{h}^* \cdot \dot{\mathbf{h}}^*\right), \tag{43}$$

where we have used (19). Note that the last two terms on the right-hand side of (42) represent electrical and magnetic energies, respectively, *in vacuo*. Substitution of (43) into (18), and subsequent use of the algebraic relationships in (6), leads to

$$-\dot{E}^{F\eta db} + \left[\frac{1}{\rho_R}\mathbf{P} + \frac{1}{\rho}\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \frac{1}{2}\epsilon_o\mathbf{e}^* \cdot \mathbf{e}^* - \frac{1}{2}\mu_o\mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-\mathsf{T}}\right] \cdot \dot{\mathbf{F}} + \theta\dot{\eta} + \frac{1}{\rho}\mathbf{e}^* \cdot \dot{\mathbf{d}}^* + \frac{1}{\rho}\mathbf{h}^* \cdot \dot{\mathbf{b}}^* + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^*$$
$$- \frac{1}{\rho\theta}\mathbf{q} \cdot grad\ \theta \geqslant 0, \tag{44}$$

the second law statement for this formulation. Note that $\mathbf{d}^*$ and $\mathbf{b}^*$ appear as rates in the second law inequality (44), i.e., as natural independent variables. Use of the chain rule on $\dot{E}^{F\eta db}$ leads to

$$\left[\frac{1}{\rho_R}\mathbf{P} - \frac{\partial E^{F\eta db}}{\partial \mathbf{F}} + \frac{1}{\rho}\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \frac{1}{2}\epsilon_o\mathbf{e}^* \cdot \mathbf{e}^* - \frac{1}{2}\mu_o\mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-\mathsf{T}}\right] \cdot \dot{\mathbf{F}} + \left(\theta - \frac{\partial E^{F\eta db}}{\partial \eta}\right)\dot{\eta} + \left(\frac{1}{\rho}\mathbf{e}^* - \frac{\partial E^{F\eta db}}{\partial \mathbf{d}^*}\right) \cdot \dot{\mathbf{d}}^*$$
$$+ \left(\frac{1}{\rho}\mathbf{h}^* - \frac{\partial E^{F\eta db}}{\partial \mathbf{b}^*}\right) \cdot \dot{\mathbf{b}}^* + \frac{1}{\rho}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta}\mathbf{q} \cdot grad\ \theta \geqslant 0, \tag{45}$$

from which the state equations

$$\mathbf{P} = \rho_R\frac{\partial E^{F\eta db}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \frac{1}{2}\epsilon_o\mathbf{e}^* \cdot \mathbf{e}^* - \frac{1}{2}\mu_o\mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-\mathsf{T}}, \quad \theta = \frac{\partial E^{F\eta db}}{\partial \eta}, \quad \mathbf{e}^* = \rho\frac{\partial E^{F\eta db}}{\partial \mathbf{d}^*}, \quad \mathbf{h}^* = \rho\frac{\partial E^{F\eta db}}{\partial \mathbf{b}^*} \tag{46}$$

follow.

We collectively coin the set of independent variables $\{\mathbf{F}, \eta, \mathbf{d}^*, \mathbf{b}^*\}$, the thermodynamic energy potential $E^{F\eta db}$, and the state equations (46) the entropy-deformation-electric displacement-magnetic induction formulation. This formulation makes contact with a formulation presented in Green and Naghdi (1984). This contact is significant as the two formulations were developed from different perspectives. In particular, the terms from the energy supply rate r^{em} that Green & Naghdi "transferred to be included in the internal energy" (Green & Naghdi, 1984, p. 184), although not explicitly identified by the authors, are precisely the last two terms on the right-hand side of our Legendre-type transformation (42). Green & Naghdi's "augmented" internal energy (although not symbolically differentiated from their original internal energy) is thus equivalent to our transformed energy potential $E^{F\eta db}$ defined in (42).

State equations for other formulations that employ either $\mathbf{d}^*$ or $\mathbf{b}^*$ as an independent variable are catalogued in Appendix A.

5.2. Generation of an infinite number of thermodynamic energy potentials

We extend the formalism of the previous sections by presenting a generalized energy potential

$$E^{(\cdot)(\cdot)(\cdot)(\cdot)} = \varepsilon - \alpha_1\frac{1}{\rho_R}\mathbf{P} \cdot \mathbf{F} - \alpha_2\theta\eta - \frac{1}{\rho}\mathbf{e}^* \cdot \left(\frac{\alpha_3}{2}\epsilon_o\mathbf{e}^* + \alpha_4\mathbf{p}^*\right) - \frac{\mu_o}{\rho}\mathbf{h}^* \cdot \left(\frac{\alpha_5}{2}\mathbf{h}^* + \alpha_6\mathbf{m}^*\right), \tag{47}$$

where the internal energy function takes its appropriate representation, either $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$ or $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$. The integers $\alpha_1, \alpha_2, \alpha_4$, and α_6 can either be 1 or 0. Integers α_3 and α_5 are *then* determined per the following restrictions:

   (i) When α_4 is 1, α_3 is 0.
  (ii) When α_4 is 0, α_3 is arbitrary. This introduces energy potentials with arbitrary linear combinations of $\mathbf{e}^*$ and $\mathbf{p}^*$ as the electrical independent variable.
 (iii) When α_6 is 1, α_5 is 0.
 (iv) When α_6 is 0, α_5 is arbitrary. This introduces energy potentials with arbitrary linear combinations of $\mathbf{h}^*$ and $\mathbf{m}^*$ as the magnetic independent variable.

Among the infinite number of energy potentials that can be constructed using this formalism are those catalogued in Appendix A. For instance, if we select $\alpha_1 = \alpha_2 = \alpha_4 = \alpha_6 = 0$, restrictions (ii) and (iv) dictate that α_3 and α_5 are arbitrary. Setting the arbitrary integers α_3 and α_5 to -1, and employing $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}^*, \mathbf{m}^*)$, we recover the special case (42). In this example, by setting the arbitrary integers α_3 and α_5 to -1, the special linear combinations $\mathbf{d}^* = \mathbf{p}^* + \epsilon_o\mathbf{e}^*$ and $\mathbf{b}^* = \mu_o(\mathbf{h}^* + \mathbf{m}^*)$ are introduced as the electrical and magnetic independent variables, respectively.

6. Entropy as a characterizing thermodynamic potential

In Sections 4 and 5, we developed a comprehensive thermodynamic framework that uses energetic potentials to characterize near-equilibrium fully coupled TEMM processes. In this section, we present an alternative thermodynamic framework, wherein we use entropy and Legendre transforms of entropy as the characterizing potentials for near-equilibrium fully coupled TEMM processes. Entropic potentials are often used to describe equilibrium states in statistical thermodynamics[11] (Callen, 1985; Landau & Lifshitz, 1980).

We proceed by rewriting the fundamental form of the Clausius–Duhem inequality (11) as:

$$\dot{\eta} + \frac{1}{\rho_R}\frac{\mathbf{P}}{\theta}\cdot\dot{\mathbf{F}} - \frac{1}{\theta}\dot{\varepsilon} + \frac{\mathbf{e}^*}{\theta}\cdot\overline{\left(\frac{\dot{\mathbf{p}^*}}{\rho}\right)} + \mu_o\frac{\mathbf{h}^*}{\theta}\cdot\overline{\left(\frac{\dot{\mathbf{m}^*}}{\rho}\right)} + \frac{1}{\rho\theta}\mathbf{j}^*\cdot\mathbf{e}^* - \frac{1}{\rho\theta^2}\mathbf{q}\cdot grad\,\theta \geqslant 0. \tag{48}$$

With entropy η as the thermodynamic potential, the natural independent variables, i.e., the extensive quantities, are the rate terms $\mathbf{F}, \varepsilon, \mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$. The corresponding dependent variables, i.e., the conjugate intensive quantities, are identified as the coefficients of these rate terms, namely $\mathbf{P}/\theta, 1/\theta, \mathbf{e}^*/\theta$, and $\mathbf{h}^*/\theta$. Thus, the *fundamental entropic relationship* (Callen, 1985), i.e., entropy as a function of *extensive variables,* is

$$\eta = \breve{\eta}\left(\mathbf{F}, \varepsilon, \frac{\mathbf{p}^*}{\rho}, \frac{\mathbf{m}^*}{\rho}\right). \tag{49}$$

In order to use polarization $\mathbf{p}^*$ and magnetization $\mathbf{m}^*$ as the electromagnetic independent variables instead of the extensive quantities $\mathbf{p}^*/\rho$ and $\mathbf{m}^*/\rho$, the Clausius–Duhem inequality (48) is rewritten using the chain rule (17):

$$\dot{\eta} + \frac{1}{\rho_R}\left[\frac{\mathbf{P}}{\theta} + J\left(\frac{\mathbf{e}^*}{\theta}\cdot\mathbf{p}^* + \mu_o\frac{\mathbf{h}^*}{\theta}\cdot\mathbf{m}^*\right)\mathbf{F}^{-\mathsf{T}}\right]\cdot\dot{\mathbf{F}} - \frac{1}{\theta}\dot{\varepsilon} + \frac{1}{\rho}\frac{\mathbf{e}^*}{\theta}\cdot\dot{\mathbf{p}}^* + \frac{\mu_o}{\rho}\frac{\mathbf{h}^*}{\theta}\cdot\dot{\mathbf{m}}^* + \frac{1}{\rho\theta}\mathbf{j}^*\cdot\mathbf{e}^* - \frac{1}{\rho\theta^2}\mathbf{q}\cdot grad\,\theta \geqslant 0. \tag{50}$$

Use of the chain rule on the thermodynamic potential $\eta = \bar{\eta}(\mathbf{F}, \varepsilon, \mathbf{p}^*, \mathbf{m}^*)$ gives

$$\dot{\eta} = \frac{\partial\bar{\eta}}{\partial\mathbf{F}}\cdot\dot{\mathbf{F}} + \frac{\partial\bar{\eta}}{\partial\varepsilon}\dot{\varepsilon} + \frac{\partial\bar{\eta}}{\partial\mathbf{p}^*}\cdot\dot{\mathbf{p}}^* + \frac{\partial\bar{\eta}}{\partial\mathbf{m}^*}\cdot\dot{\mathbf{m}}^*. \tag{51}$$

Substitution of this result into inequality (50) leads to the state equations for this formulation, i.e.,

$$\mathbf{P} = -\rho_R\theta\frac{\partial\bar{\eta}}{\partial\mathbf{F}} - J(\mathbf{e}^*\cdot\mathbf{p}^* + \mu_o\mathbf{h}^*\cdot\mathbf{m}^*)\mathbf{F}^{-\mathsf{T}}, \quad \frac{1}{\theta} = \frac{\partial\bar{\eta}}{\partial\varepsilon}, \quad \mathbf{e}^* = -\rho\theta\frac{\partial\bar{\eta}}{\partial\mathbf{p}^*}, \quad \mathbf{h}^* = -\frac{\rho\theta}{\mu_o}\frac{\partial\bar{\eta}}{\partial\mathbf{m}^*}. \tag{52}$$

We collectively coin the set of independent variables $\{\mathbf{F}, \varepsilon, \mathbf{p}^*, \mathbf{m}^*\}$, the thermodynamic entropy potential $\eta = \bar{\eta}(\mathbf{F}, \varepsilon, \mathbf{p}^*, \mathbf{m}^*)$, and the state Eqs. (52) the *modified entropic formulation.*

With this formulation as a template, we investigate other formulations involving alternative sets of independent variables. Similar to the energetic formulations presented in Section 4 (refer to Table 1 and Appendix A), the entropic formulations are categorized into four families, each with a common set of thermomechanical independent variables: Family 1 – $\mathbf{F}$ and ε (both extensive); Family 2 – $\mathbf{F}$ (extensive) and θ (intensive); Family 3 – $\mathbf{P}/\theta$ (intensive) and ε (extensive); and Family 4 – $\mathbf{P}/\theta$ and θ (both intensive). Either $\mathbf{e}^*/\theta$ (intensive) or $\mathbf{p}^*/\rho$ (extensive), and either $\mathbf{h}^*/\theta$ (intensive) or $\mathbf{m}^*/\rho$ (extensive), are selected as the electrical and magnetic independent variables, respectively. Thus, within each family, there are four different formulations, each associated with one of the four possible sets of independent variables. Also associated with each of these formulations is an entropy potential $\eta^{(a)(b)(c)(d)}$, where the superscripts (a), (b), (c), and (d) are placeholders for an appropriate mechanical, thermal, electrical, and magnetic independent variable, respectively (refer to Table 2).

We note that the intensive variables $\mathbf{P}/\theta, \mathbf{e}^*/\theta$, and $\mathbf{h}^*/\theta$ discussed above depend on temperature and are thus not purely mechanical, electrical, and magnetic, respectively. We later show that, in certain instances, this temperature dependence can be eliminated.

6.1. Legendre-transformed entropic potentials

Legendre-transformed entropic potentials for a purely thermomechanical process are known as *Massieu–Planck functions*[12] (Callen, 1985; Rooney & Bechtel, 2004). In this paper, the Massieu–Planck functions are extended to a fully coupled finite-deformation thermo-electro-magneto-mechanical process for the first time. Recall that for the fundamental entropic relationship, the extensive quantities $\mathbf{F}, \varepsilon, \mathbf{p}^*/\rho$, and $\mathbf{m}^*/\rho$ are the independent variables, and the conjugate intensive quantities $\mathbf{P}/\theta, 1/\theta, \mathbf{e}^*/\theta$, and $\mathbf{h}^*/\theta$ are the dependent variables. The first-order Legendre transformations required to change any of the extensive thermal, electrical, magnetic, or mechanical independent variables to their intensive counterparts are listed in Table 3.

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[11] Other applications, and the limitations of using entropy as a thermodynamic potential, are discussed in Landauer (1975).

[12] Legendre transforms of entropy are known as free entropies, a concept first introduced by Francois Massieu to characterize fluids (Massieu, 1869; Massieu, 1861).

Table 2
Entropic potentials and associated independent variables.

Family 1	Family 2	Family 3	Family 4
$\breve{\eta}$, or $\bar{\eta}$	$\eta^{F\theta pm}$	$\eta^{P\varepsilon pm}$	$\eta^{P\theta pm}$
$\eta^{F\varepsilon em}$	$\eta^{F\theta em}$	$\eta^{P\varepsilon em}$	$\eta^{P\theta em}$
$\eta^{F\varepsilon ph}$	$\eta^{F\theta ph}$	$\eta^{P\varepsilon ph}$	$\eta^{P\theta ph}$
$\eta^{F\varepsilon eh}$	$\eta^{F\theta eh}$	$\eta^{P\varepsilon eh}$	$\eta^{P\theta eh}$

In fact, any potential listed in Table 2 can be constructed from a combination of the first-order Legendre transformations listed in Table 3, and the corresponding state equations can be derived from second law restrictions.

6.2. State equations

In this section, we demonstrate the derivation of the state equations for some of the entropic potentials listed in Table 2. The full set of Legendre transformations and state equations is catalogued in Appendix B.

Internal energy-deformation-electric field-magnetization formulation

In this formulation, the independent variables are $\mathbf{F}, \varepsilon, \mathbf{e}^*/\theta$, and $\mathbf{m}^*$. The potential $\eta^{F\varepsilon em}$ is introduced as the Legendre transform of entropy $\eta = \bar{\eta}(\mathbf{F}, \varepsilon, \mathbf{p}^*, \mathbf{m}^*)$ with respect to the electrical variable, i.e.,

$$\eta^{F\varepsilon em} = \eta + \frac{1}{\rho}\mathbf{p}^* \cdot \frac{\mathbf{e}^*}{\theta}, \tag{53}$$

as shown in Table 3, whose rate form is

$$\dot{\eta}^{F\varepsilon em} = \dot{\eta} + \frac{1}{\rho}\left(\frac{\mathbf{e}^*}{\theta} \cdot \mathbf{p}^*\right)\mathbf{F}^{-T} \cdot \dot{\mathbf{F}} + \frac{1}{\rho}\frac{\mathbf{e}^*}{\theta} \cdot \dot{\mathbf{p}}^* + \frac{1}{\rho}\mathbf{p}^* \cdot \overline{\left(\frac{\mathbf{e}^*}{\theta}\right)}. \tag{54}$$

Upon substituting (54) into (50) and eliminating $\dot{\eta}$, the second law inequality for this formulation becomes

$$\dot{\eta}^{F\varepsilon em} + \frac{1}{\rho_R}\left[\frac{\mathbf{P}}{\theta} + J\left(\mu_o \frac{\mathbf{h}^*}{\theta} \cdot \mathbf{m}^*\right)\mathbf{F}^{-T}\right] \cdot \dot{\mathbf{F}} - \frac{1}{\theta}\dot{\varepsilon} - \frac{1}{\rho}\mathbf{p}^* \cdot \overline{\left(\frac{\mathbf{e}^*}{\theta}\right)} + \frac{\mu_o}{\rho}\frac{\mathbf{h}^*}{\theta} \cdot \dot{\mathbf{m}}^* + \frac{1}{\rho\theta}\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\rho\theta^2}\mathbf{q} \cdot grad\,\theta \geqslant 0. \tag{55}$$

Using

$$\dot{\eta}^{F\varepsilon em} = \frac{\partial \eta^{F\varepsilon em}}{\partial \mathbf{F}} \cdot \dot{\mathbf{F}} + \frac{\partial \eta^{F\varepsilon em}}{\partial \varepsilon}\dot{\varepsilon} + \frac{\partial \eta^{F\varepsilon em}}{\partial\left(\frac{\mathbf{e}^*}{\theta}\right)} \cdot \overline{\left(\frac{\mathbf{e}^*}{\theta}\right)} + \frac{\partial \eta^{F\varepsilon em}}{\partial \mathbf{m}^*} \cdot \dot{\mathbf{m}}^* \tag{56}$$

in inequality (55) leads to the state equations

$$\mathbf{P} = -\rho_R\theta\frac{\partial \eta^{F\varepsilon em}}{\partial \mathbf{F}} - J\left(\mu_o\mathbf{h}^* \cdot \mathbf{m}^*\right)\mathbf{F}^{-T}, \quad \frac{1}{\theta} = \frac{\partial \eta^{F\varepsilon em}}{\partial \varepsilon}, \quad \mathbf{p}^* = \rho\frac{\partial \eta^{F\varepsilon em}}{\partial\left(\frac{\mathbf{e}^*}{\theta}\right)}, \quad \mathbf{h}^* = -\frac{\rho\theta}{\mu_o}\frac{\partial \eta^{F\varepsilon em}}{\partial \mathbf{m}^*}. \tag{57}$$

For this particular formulation, the electrical independent variable $\mathbf{e}^*/\theta$ is dependent on temperature. This dependence cannot be eliminated for $\eta^{F\varepsilon em}$ or any of the other potentials belonging to Families 1 and 3. However, for the potentials belonging to Families 2 and 4, where the thermal independent variable is θ, it is possible to eliminate this dependence, as we demonstrate in the following example.

Temperature-stress-electric field-magnetic field formulation

In this formulation, the independent variables are all intensive quantities. The entropic potential $\eta^{P\theta eh}$ is defined as the Legendre transform of entropy $\eta = \breve{\eta}(\mathbf{F}, \varepsilon, \mathbf{p}^*/\rho, \mathbf{m}^*/\rho)$ with respect to all four independent variables, from extensive to intensive ($\mathbf{F}$ to $\mathbf{P}/\theta$, ε to $1/\theta$, $\mathbf{p}^*/\rho$ to $\mathbf{e}^*/\theta$, and $\mathbf{m}^*/\rho$ to $\mathbf{h}^*/\theta$), i.e.,

Table 3
First-order Legendre transformations.

IVs	Transformation equations
$\varepsilon \Rightarrow \dfrac{1}{\theta}$	$\eta^{F\theta pm} = \eta - \varepsilon\dfrac{1}{\theta}$
$\mathbf{F} \Rightarrow \dfrac{\mathbf{P}}{\theta}$	$\eta^{P\varepsilon pm} = \eta + \dfrac{1}{\rho_R}\mathbf{F} \cdot \dfrac{\mathbf{P}}{\theta}$
$\dfrac{\mathbf{p}^*}{\rho} \Rightarrow \dfrac{\mathbf{e}^*}{\theta}$	$\eta^{F\varepsilon em} = \eta + \dfrac{\mathbf{p}^*}{\rho} \cdot \dfrac{\mathbf{e}^*}{\theta}$
$\dfrac{\mathbf{m}^*}{\rho} \Rightarrow \dfrac{\mathbf{h}^*}{\theta}$	$\eta^{F\varepsilon ph} = \eta + \mu_o\dfrac{\mathbf{m}^*}{\rho} \cdot \dfrac{\mathbf{h}^*}{\theta}$

$$\eta^{P\theta eh} = \eta - \varepsilon\frac{1}{\theta} + \frac{1}{\rho_R}\mathbf{F}\cdot\frac{\mathbf{P}}{\theta} + \frac{\mathbf{p}^*}{\rho}\cdot\frac{\mathbf{e}^*}{\theta} + \mu_o\frac{\mathbf{m}^*}{\rho}\cdot\frac{\mathbf{h}^*}{\theta}, \tag{58}$$

following Table 3. The rate form of this equation is

$$\dot{\eta}^{P\theta eh} = \dot{\eta} - \frac{1}{\theta}\dot{\varepsilon} - \varepsilon\overline{\left(\frac{1}{\theta}\right)} + \frac{1}{\rho_R}\frac{\mathbf{P}}{\theta}\cdot\dot{\mathbf{F}} + \frac{1}{\rho_R}\mathbf{F}\cdot\overline{\left(\frac{\mathbf{P}}{\theta}\right)} + \frac{\mathbf{e}^*}{\theta}\cdot\overline{\left(\frac{\mathbf{p}^*}{\rho}\right)} + \frac{\mathbf{p}^*}{\rho}\cdot\overline{\left(\frac{\mathbf{e}^*}{\theta}\right)} + \mu_o\frac{\mathbf{h}^*}{\theta}\cdot\overline{\left(\frac{\mathbf{m}^*}{\rho}\right)} + \mu_o\frac{\mathbf{m}^*}{\rho}\cdot\overline{\left(\frac{\mathbf{h}^*}{\theta}\right)}. \tag{59}$$

The second law corresponding to this formulation is obtained by substituting the rate form (59) into inequality (48), i.e.,

$$\dot{\eta}^{P\theta eh} - \frac{1}{\rho_R}\mathbf{F}\cdot\overline{\left(\frac{\mathbf{P}}{\theta}\right)} + \varepsilon\overline{\left(\frac{1}{\theta}\right)} - \frac{\mathbf{p}^*}{\rho}\cdot\overline{\left(\frac{\mathbf{e}^*}{\theta}\right)} - \mu_o\frac{\mathbf{m}^*}{\rho}\cdot\overline{\left(\frac{\mathbf{h}^*}{\theta}\right)} + \frac{1}{\rho\theta}\mathbf{j}^*\cdot\mathbf{e}^* - \frac{1}{\rho\theta^2}\mathbf{q}\cdot grad\,\theta \geqslant 0. \tag{60}$$

The rate terms in inequality (60) are rewritten as

$$\overline{\left(\frac{1}{\theta}\right)} = -\frac{\dot{\theta}}{\theta^2}, \quad \overline{\left(\frac{\mathbf{P}}{\theta}\right)} = \frac{1}{\theta}\dot{\mathbf{P}} - \frac{\dot{\theta}}{\theta^2}\mathbf{P}, \quad \overline{\left(\frac{\mathbf{e}^*}{\theta}\right)} = \frac{1}{\theta}\dot{\mathbf{e}}^* - \frac{\dot{\theta}}{\theta^2}\mathbf{e}^*, \quad \overline{\left(\frac{\mathbf{h}^*}{\theta}\right)} = \frac{1}{\theta}\dot{\mathbf{h}}^* - \frac{\dot{\theta}}{\theta^2}\mathbf{h}^*, \tag{61}$$

which leads to

$$\dot{\eta}^{P\theta eh} - \frac{1}{\rho_R\theta}\mathbf{F}\cdot\dot{\mathbf{P}} - \frac{1}{\theta^2}\left(\varepsilon - \frac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \frac{1}{\rho}\mathbf{e}^*\cdot\mathbf{p}^* - \frac{\mu_o}{\rho}\mathbf{h}^*\cdot\mathbf{m}^*\right)\dot{\theta} - \frac{1}{\rho\theta}\mathbf{p}^*\cdot\dot{\mathbf{e}}^* - \frac{\mu_o}{\rho\theta}\mathbf{m}^*\cdot\dot{\mathbf{h}}^* + \frac{1}{\rho\theta}\mathbf{j}^*\cdot\mathbf{e}^* - \frac{1}{\rho\theta^2}\mathbf{q}\cdot grad\,\theta \geqslant 0, \tag{62}$$

enabling the use of $\mathbf{P}, \theta, \mathbf{e}^*$, and $\mathbf{h}^*$ as independent variables. The chain rule

$$\dot{\eta}^{P\theta eh} = \frac{\partial\eta^{P\theta eh}}{\partial\mathbf{P}}\cdot\dot{\mathbf{P}} + \frac{\partial\eta^{P\theta eh}}{\partial\theta}\dot{\theta} + \frac{\partial\eta^{P\theta eh}}{\partial\mathbf{e}^*}\cdot\dot{\mathbf{e}}^* + \frac{\partial\eta^{P\theta eh}}{\partial\mathbf{h}^*}\cdot\dot{\mathbf{h}}^* \tag{63}$$

is then used in inequality (62) to derive the state equations

$$\varepsilon = \theta^2\frac{\partial\eta^{P\theta eh}}{\partial\theta} + \frac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} + \frac{1}{\rho}\mathbf{e}^*\cdot\mathbf{p}^* + \frac{\mu_o}{\rho}\mathbf{m}^*\cdot\mathbf{h}^*, \quad \mathbf{F} = \rho_R\theta\frac{\partial\eta^{P\theta eh}}{\partial\mathbf{P}}, \quad \mathbf{p}^* = \rho\theta\frac{\partial\eta^{P\theta eh}}{\partial\mathbf{e}^*}, \quad \mathbf{m}^* = \frac{\rho\theta}{\mu_o}\frac{\partial\eta^{P\theta eh}}{\partial\mathbf{h}^*}. \tag{64}$$

We emphasize that operation (61) allowed the state Eqs. (64) to be written in terms of the purely mechanical ($\mathbf{P}$), thermal (θ), electrical ($\mathbf{e}^*$), and magnetic ($\mathbf{h}^*$) independent variables, which are more favorable for characterization than $\mathbf{P}/\theta, 1/\theta, \mathbf{e}^*/\theta$, and $\mathbf{h}^*/\theta$. This formalism can be applied to all potentials belonging to Families 2 and 4 (i.e., potentials with temperature θ as the thermal independent variable) in order to eliminate the temperature dependence of the mechanical, electrical, and magnetic independent variables.

7. Applications

We envision our mathematical framework serving as a rigorous starting point for enabling the design and characterization of novel multifunctional materials with fully coupled thermo-electro-magneto-mechanical response. In the following subsections, we elaborate on this vision.

7.1. Experimental characterization of multifunctional materials

Each thermo-electro-magneto-mechanical process in our catalogue correlates with a particular experiment, the independent variables being controlled and the dependent variables being the measured responses. In practice, a given experiment is most straightforwardly characterized by a thermodynamic potential that employs the quantities one wishes to control as its independent variables. For instance, in an experiment conducted on a viscous fluid at constant temperature and constant pressure (i.e., an isothermal, isobaric process), Gibbs free energy, which uses pressure and temperature as independent variables, is the most favorable thermodynamic potential for characterizing the process. Hence, the breadth of our catalogue of TEMM processes is intended to provide engineers, applied physicists, and material scientists with optimal flexibility in their approach to experimentally characterizing multifunctional materials and deducing their 3-D energy landscapes.

7.2. Irreversible TEMM material behavior

Irreversibilities such as mechanical hysteresis (e.g., viscoelasticity), electrical hysteresis (e.g., ferroelectricity), magnetic hysteresis (e.g., ferromagnetism), dependence on deformation history or loading history (e.g., plasticity), and viscous dissipation (e.g., Newtonian fluid dynamics) occur in many practically important physical processes. The state equations presented in our comprehensive catalogue, however, are derived under the auspices of classical equilibrium thermodynamics, which gov-

erns *reversible* TEMM processes or *near-equilibrium irreversible* TEMM processes (Callen, 1985). Nevertheless, the formalism and methodology presented in this paper can be straightforwardly extended to model *far-from-equilibrium irreversibilities* through the use of internal state variables (Coleman & Gurtin, 1967; Landis, 2002; Maugin & Muschik, 1994) that parameterize the *history* of a thermo-electro-magneto-mechanical process.

7.3. Constraints and stability in TEMM

In the context of thermomechanics, a constrained theory assumes *a priori* that only those processes satisfying a prescribed relation between certain thermal and mechanical quantities are allowable. A familiar example is that of an absolutely incompressible material, whose density cannot be changed by any means, be it thermal or mechanical. Constrained theories have two powerful advantages: the governing equations produced by a constrained theory are generally easier to solve than those produced by the full 'unconstrained' theory, and less experiments are necessary to characterize the material in the context of the constrained model (Rooney & Bechtel, 2004).

A modern approach for developing constrained theories is to model the constraint as a constitutive limit, i.e., a restriction on the constitutive behavior of the material as described by a thermodynamic potential (Bechtel, Rooney, & Forest, 2004). Although the formalism of constitutive limits has been used to model constraints such as incompressibility in thermomechanics (see, for example, Bechtel et al. (2004) and Bechtel, Rooney, & Wang (2004) for mechanically and absolutely incompressible Newtonian fluids, respectively, and Rooney & Bechtel (2004) for mechanically incompressible finite-deformational elastic solids), this approach remains largely unexplored in thermo-electro-magneto-mechanics, as do investigations into the stability of constrained TEMM theories. Stability of the thermodynamic equilibrium (Bechtel, Rooney, & Forest, 2005) is an important consideration when assessing the validity of a constrained theory: In finite-deformation elasticity, for instance, temperature-deformation constraints are found to have an unstable equilibrium, i.e., certain perturbations of the equilibrium state grow without bound (Rooney & Bechtel, 2004).

Our comprehensive catalogue of free energies and state equations represents the starting point for (i) the development of constrained TEMM theories via the formalism of constitutive limits and (ii) investigations into their stability.

7.4. Design of new multifunctional materials

The standard practice in the fabrication of new materials is to first manufacture the material, then experimentally determine its macroscopic-scale or continuum performance properties (e.g., permeability, permittivity, Young's modulus, coefficient of thermal expansion). Following this customary approach, our thermodynamic framework can be utilized to characterize novel multifunctional materials as described in the previous parts of this section. However, numerous experiments and costly trial-and-error are required to iteratively arrive at a design that optimally accomplishes the performance objectives required by a particular multifunctional application. We envision the research in this paper – in particular, our comprehensive catalogue of free energies and state equations – enabling the development of a new "inverse design" paradigm: Briefly, the targeted macroscopic-scale performance properties will be expressed in terms of material response functions relating a set of independent and dependent variables from the continuum model. Using the state equations, the material response functions will then be converted into conditions on the free energy. By integrating these conditions, an energy landscape will be constructed that provides a "recipe" for microscopic-scale material design and fabrication.

8. Summary

In this paper, we have developed a first-principles-based continuum thermodynamic framework for modeling fully coupled thermo-electro-magneto-mechanical (TEMM) processes in the finite-deformation regime. This framework was developed from a unified perspective, integrating fundamental principles from continuum mechanics, electrodynamics, and classical thermodynamics. As such, it represents a first step towards facilitating the design and characterization of novel multifunctional materials. In order to enable these multifunctional materials to reach their full performance potential, our modeling architecture was developed to accommodate (a) 3-D fully coupled thermo-electro-magneto-mechanical behavior, finite deformations, nonlinear constitutive response, dynamic electromagnetic fields, and anisotropy, and (b) multifunctionality at various time scales, length scales, and compositional levels.

Our primary contribution was the development of a *comprehensive* catalogue of free energies and state equations that enable the modeling and characterization of any general TEMM process. Central to the development of this comprehensive catalogue was the fundamental energetic process, which, *analogous to classical thermodynamics,* features extensive independent variables, intensive dependent variables, and specific internal energy as the characterizing thermodynamic potential. The fundamental energetic process was the thermodynamically consistent starting point from which all alternative energetic processes in this paper were derived. These alternative processes were classified as either (a) those whose free energies use any or all of the conjugate *intensive* quantities as independent variables, or (b) those whose free energies use one or more *secondary* electromagnetic quantities as independent variables. The former were introduced using Legendre transformations of the internal energy, the latter using novel Legendre-type transformations. This formalism was then extended to processes that employ entropy rather than energy as the characterizing thermodynamic potential. Most of the thermodynamic potentials appearing in our catalogue, both energetic and entropic, were introduced for the first time.

Acknowledgements

Appendix A. Energy formulations

(See Tables 4–9.)

Table 4
Fully coupled state equations for energy Family 1.

Residual Dissipation Inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$			
Energy Potential	State Equations		
$\breve{\varepsilon}$	$\mathbf{P} = \rho_R \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}}$		
	$\theta = \frac{\partial \breve{\varepsilon}}{\partial \eta}$	$\mathbf{e}^* = \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}^*}{\rho}\right)}$	$\mathbf{h}^* = \frac{1}{\mu_o}\frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}^*}{\rho}\right)}$
$\bar{\varepsilon}$	$\mathbf{P} = \rho_R \frac{\partial \bar{\varepsilon}}{\partial \mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial \bar{\varepsilon}}{\partial \eta}$	$\mathbf{e}^* = \rho \frac{\partial \bar{\varepsilon}}{\partial \mathbf{p}^*}$	$\mathbf{h}^* = \frac{\rho}{\mu_o}\frac{\partial \bar{\varepsilon}}{\partial \mathbf{m}^*}$
$E^{F\eta em} = \varepsilon - \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta em}}{\partial \mathbf{F}} - \mu_o J(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta em}}{\partial \eta}$	$\mathbf{p}^* = -\rho \frac{\partial E^{F\eta em}}{\partial \mathbf{e}^*}$	$\mathbf{h}^* = \frac{\rho}{\mu_o}\frac{\partial E^{F\eta em}}{\partial \mathbf{m}^*}$
$E^{F\eta ph} = \varepsilon - \frac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta ph}}{\partial \mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^*)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta ph}}{\partial \eta}$	$\mathbf{e}^* = \rho \frac{\partial E^{F\eta ph}}{\partial \mathbf{p}^*}$	$\mathbf{m}^* = -\frac{\rho}{\mu_o}\frac{\partial E^{F\eta ph}}{\partial \mathbf{h}^*}$
$E^{F\eta eh} = \varepsilon - \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^* - \frac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta eh}}{\partial \mathbf{F}}$		
	$\theta = \frac{\partial E^{F\eta eh}}{\partial \eta}$	$\mathbf{p}^* = -\rho \frac{\partial E^{F\eta eh}}{\partial \mathbf{e}^*}$	$\mathbf{m}^* = -\frac{\rho}{\mu_o}\frac{\partial E^{F\eta eh}}{\partial \mathbf{h}^*}$

Table 5
Fully coupled state equations for energy Family 1 (with secondary electromagnetic IVs).

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$			
Energy potential	State equations		
$E^{F\eta dm} = \varepsilon + \frac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta dm}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^* - \frac{1}{2}\epsilon_o \mathbf{e}^* \cdot \mathbf{e}^*\right)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta dm}}{\partial \eta}$	$\mathbf{e}^* = \rho \frac{\partial E^{F\eta dm}}{\partial \mathbf{d}^*}$	$\mathbf{h}^* = \frac{\rho}{\mu_o}\frac{\partial E^{F\eta dm}}{\partial \mathbf{m}^*}$
$E^{F\eta pb} = \varepsilon + \frac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta pb}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{p}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \frac{1}{2}\mu_o \mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta pb}}{\partial \eta}$	$\mathbf{e}^* = \rho \frac{\partial E^{F\eta pb}}{\partial \mathbf{p}^*}$	$\mathbf{h}^* = \rho \frac{\partial E^{F\eta pb}}{\partial \mathbf{b}^*}$
$E^{F\eta dh} = \varepsilon + \frac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^* - \frac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta dh}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* - \frac{1}{2}\epsilon_o \mathbf{e}^* \cdot \mathbf{e}^*\right)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta dh}}{\partial \eta}$	$\mathbf{e}^* = \rho \frac{\partial E^{F\eta dh}}{\partial \mathbf{d}^*}$	$\mathbf{m}^* = -\frac{\rho}{\mu_o}\frac{\partial E^{F\eta dh}}{\partial \mathbf{h}^*}$
$E^{F\eta eb} = \varepsilon - \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^* + \frac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta eb}}{\partial \mathbf{F}} - J\left(\mathbf{h}^* \cdot \mathbf{b}^* - \frac{1}{2}\mu_o \mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta eb}}{\partial \eta}$	$\mathbf{p}^* = -\rho \frac{\partial E^{F\eta eb}}{\partial \mathbf{e}^*}$	$\mathbf{h}^* = \rho \frac{\partial E^{F\eta eb}}{\partial \mathbf{b}^*}$
$E^{F\eta db} = \varepsilon + \frac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^* + \frac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*$	$\mathbf{P} = \rho_R \frac{\partial E^{F\eta db}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \frac{1}{2}\epsilon_o \mathbf{e}^* \cdot \mathbf{e}^* - \frac{1}{2}\mu_o \mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-T}$		
	$\theta = \frac{\partial E^{F\eta db}}{\partial \eta}$	$\mathbf{e}^* = \rho \frac{\partial E^{F\eta db}}{\partial \mathbf{d}^*}$	$\mathbf{h}^* = \rho \frac{\partial E^{F\eta db}}{\partial \mathbf{b}^*}$

Table 6
Fully coupled state equations for energy Family 2.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$		
Energy potential	**State equations**	
$E^{F\theta pm} = \varepsilon - \theta\eta$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta pm}}{\partial \mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta pm}}{\partial \theta} \qquad\qquad \mathbf{e}^* = \rho\dfrac{\partial E^{F\theta pm}}{\partial \mathbf{p}^*}$	$\mathbf{h}^* = \dfrac{\rho}{\mu_o}\dfrac{\partial E^{F\theta pm}}{\partial \mathbf{m}^*}$
$E^{F\theta em} = \varepsilon - \theta\eta - \dfrac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta em}}{\partial \mathbf{F}} - \mu_o J(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta em}}{\partial \theta} \qquad\qquad \mathbf{p}^* = -\rho\dfrac{\partial E^{F\theta em}}{\partial \mathbf{e}^*}$	$\mathbf{h}^* = \dfrac{\rho}{\mu_o}\dfrac{\partial E^{F\theta em}}{\partial \mathbf{m}^*}$
$E^{F\theta ph} = \varepsilon - \theta\eta - \dfrac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta ph}}{\partial \mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^*)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta ph}}{\partial \theta} \qquad\qquad \mathbf{e}^* = \rho\dfrac{\partial E^{F\theta ph}}{\partial \mathbf{p}^*}$	$\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{F\theta ph}}{\partial \mathbf{h}^*}$
$E^{F\theta eh} = \varepsilon - \theta\eta - \dfrac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^* - \dfrac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta eh}}{\partial \mathbf{F}}$	
	$\eta = -\dfrac{\partial E^{F\theta eh}}{\partial \theta} \qquad\qquad \mathbf{p}^* = -\rho\dfrac{\partial E^{F\theta eh}}{\partial \mathbf{e}^*}$	$\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{F\theta eh}}{\partial \mathbf{h}^*}$

Table 7
Fully coupled state equations for energy Family 2 (with secondary electromagnetic IVs).

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$		
Energy potential	**State equations**	
$E^{F\theta dm} = \varepsilon - \theta\eta + \dfrac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta dm}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mu_o \mathbf{h}^* \cdot \mathbf{m}^* - \dfrac{1}{2}\epsilon_o \mathbf{e}^* \cdot \mathbf{e}^*\right)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta dm}}{\partial \theta} \qquad\qquad \mathbf{e}^* = \rho\dfrac{\partial E^{F\theta dm}}{\partial \mathbf{d}^*}$	$\mathbf{h}^* = \dfrac{\rho}{\mu_o}\dfrac{\partial E^{F\theta dm}}{\partial \mathbf{m}^*}$
$E^{F\theta pb} = \varepsilon - \theta\eta + \dfrac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta pb}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{p}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \dfrac{1}{2}\mu_o \mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta pb}}{\partial \theta} \qquad\qquad \mathbf{e}^* = \rho\dfrac{\partial E^{F\theta pb}}{\partial \mathbf{p}^*}$	$\mathbf{h}^* = \rho\dfrac{\partial E^{F\theta pb}}{\partial \mathbf{b}^*}$
$E^{F\theta dh} = \varepsilon - \theta\eta + \dfrac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^* - \dfrac{\mu_o}{\rho}\mathbf{h}^* \cdot \mathbf{m}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta dh}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* - \dfrac{1}{2}\epsilon_o \mathbf{e}^* \cdot \mathbf{e}^*\right)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta dh}}{\partial \theta} \qquad\qquad \mathbf{e}^* = \rho\dfrac{\partial E^{F\theta dh}}{\partial \mathbf{d}^*}$	$\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{F\theta dh}}{\partial \mathbf{h}^*}$
$E^{F\theta eb} = \varepsilon - \theta\eta - \dfrac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^* + \dfrac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta eb}}{\partial \mathbf{F}} - J\left(\mathbf{h}^* \cdot \mathbf{b}^* - \dfrac{1}{2}\mu_o \mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta eb}}{\partial \theta} \qquad\qquad \mathbf{p}^* = -\rho\dfrac{\partial E^{F\theta eb}}{\partial \mathbf{e}^*}$	$\mathbf{h}^* = \rho\dfrac{\partial E^{F\theta eb}}{\partial \mathbf{b}^*}$
$E^{F\theta db} = \varepsilon - \theta\eta + \dfrac{\epsilon_o}{2\rho}\mathbf{e}^* \cdot \mathbf{e}^* + \dfrac{\mu_o}{2\rho}\mathbf{h}^* \cdot \mathbf{h}^*$	$\mathbf{P} = \rho_R \dfrac{\partial E^{F\theta db}}{\partial \mathbf{F}} - J\left(\mathbf{e}^* \cdot \mathbf{d}^* + \mathbf{h}^* \cdot \mathbf{b}^* - \dfrac{1}{2}\epsilon_o \mathbf{e}^* \cdot \mathbf{e}^* - \dfrac{1}{2}\mu_o \mathbf{h}^* \cdot \mathbf{h}^*\right)\mathbf{F}^{-T}$	
	$\eta = -\dfrac{\partial E^{F\theta db}}{\partial \theta} \qquad\qquad \mathbf{e}^* = \rho\dfrac{\partial E^{F\theta db}}{\partial \mathbf{d}^*}$	$\mathbf{h}^* = \rho\dfrac{\partial E^{F\theta db}}{\partial \mathbf{b}^*}$

Table 8
Fully coupled state equations for energy Family 3.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$

Energy potential	State equations	
$E^{P\eta pm} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\eta pm}}{\partial\mathbf{P}}$ $\mathbf{e}^* = \dfrac{\partial E^{P\eta pm}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\theta = \dfrac{\partial E^{P\eta pm}}{\partial\eta}$ $\mathbf{h}^* = \dfrac{1}{\mu_o}\dfrac{\partial E^{P\eta pm}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$E^{P\eta em} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \mathbf{e}^*\cdot\dfrac{\mathbf{p}^*}{\rho}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\eta em}}{\partial\mathbf{P}}$ $\mathbf{p}^* = -\rho\dfrac{\partial E^{P\eta em}}{\partial\mathbf{e}^*}$	$\theta = \dfrac{\partial E^{P\eta em}}{\partial\eta}$ $\mathbf{h}^* = \dfrac{1}{\mu_o}\dfrac{\partial E^{P\eta em}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$E^{P\eta ph} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \mu_o\mathbf{h}^*\cdot\dfrac{\mathbf{m}^*}{\rho}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\eta ph}}{\partial\mathbf{P}}$ $\mathbf{e}^* = \dfrac{\partial E^{P\eta ph}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\theta = \dfrac{\partial E^{P\eta ph}}{\partial\eta}$ $\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{P\eta ph}}{\partial\mathbf{h}^*}$
$E^{P\eta eh} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \mathbf{e}^*\cdot\dfrac{\mathbf{p}^*}{\rho} - \mu_o\mathbf{h}^*\cdot\dfrac{\mathbf{m}^*}{\rho}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\eta eh}}{\partial\mathbf{P}}$ $\mathbf{p}^* = -\rho\dfrac{\partial E^{P\eta eh}}{\partial\mathbf{e}^*}$	$\theta = \dfrac{\partial E^{P\eta eh}}{\partial\eta}$ $\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{P\eta eh}}{\partial\mathbf{h}^*}$

Table 9
Fully coupled state equations for energy Family 4.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \dfrac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$

Energy potential	State equations	
$E^{P\theta pm} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \theta\eta$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\theta pm}}{\partial\mathbf{P}}$ $\mathbf{e}^* = \dfrac{\partial E^{P\theta pm}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\eta = -\dfrac{\partial E^{P\theta pm}}{\partial\theta}$ $\mathbf{h}^* = \dfrac{1}{\mu_o}\dfrac{\partial E^{P\theta pm}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$E^{P\theta em} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \theta\eta - \mathbf{e}^*\cdot\dfrac{\mathbf{p}^*}{\rho}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\theta em}}{\partial\mathbf{P}}$ $\mathbf{p}^* = -\rho\dfrac{\partial E^{P\theta em}}{\partial\mathbf{e}^*}$	$\eta = -\dfrac{\partial E^{P\theta em}}{\partial\theta}$ $\mathbf{h}^* = \dfrac{1}{\mu_o}\dfrac{\partial E^{P\theta em}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$E^{P\theta ph} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \theta\eta - \mu_o\mathbf{h}^*\cdot\dfrac{\mathbf{m}^*}{\rho}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\theta ph}}{\partial\mathbf{P}}$ $\mathbf{e}^* = \dfrac{\partial E^{P\theta ph}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\eta = -\dfrac{\partial E^{P\theta ph}}{\partial\theta}$ $\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{P\theta ph}}{\partial\mathbf{h}^*}$
$E^{P\theta eh} = \varepsilon - \dfrac{1}{\rho_R}\mathbf{P}\cdot\mathbf{F} - \theta\eta - \mathbf{e}^*\cdot\dfrac{\mathbf{p}^*}{\rho} - \mu_o\mathbf{h}^*\cdot\dfrac{\mathbf{m}^*}{\rho}$	$\mathbf{F} = -\rho_R\dfrac{\partial E^{P\theta eh}}{\partial\mathbf{P}}$ $\mathbf{p}^* = -\rho\dfrac{\partial E^{P\theta eh}}{\partial\mathbf{e}^*}$	$\eta = -\dfrac{\partial E^{P\theta eh}}{\partial\theta}$ $\mathbf{m}^* = -\dfrac{\rho}{\mu_o}\dfrac{\partial E^{P\theta eh}}{\partial\mathbf{h}^*}$

Appendix B. Entropy formulations

(See Tables 10–13.)

Table 10
Fully coupled state equations for entropy Family 1.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$

Entropy potential	State equations		
$\breve{\eta}$	$\mathbf{P} = -\rho_R\theta\frac{\partial\breve{\eta}}{\partial\mathbf{F}}$		
	$\frac{1}{\theta} = \frac{\partial\breve{\eta}}{\partial\varepsilon}$	$\mathbf{e}^* = -\theta\frac{\partial\breve{\eta}}{\partial\left(\frac{\mathbf{p}^*}{\rho}\right)}$	$\mathbf{h}^* = -\frac{\theta}{\mu_o}\frac{\partial\breve{\eta}}{\partial\left(\frac{\mathbf{m}^*}{\rho}\right)}$
$\bar{\eta}$	$\mathbf{P} = -\rho_R\theta\frac{\partial\bar{\eta}}{\partial\mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$		
	$\frac{1}{\theta} = \frac{\partial\bar{\eta}}{\partial\varepsilon}$	$\mathbf{e}^* = -\rho\theta\frac{\partial\bar{\eta}}{\partial\mathbf{p}^*}$	$\mathbf{h}^* = -\frac{\rho\theta}{\mu_o}\frac{\partial\bar{\eta}}{\partial\mathbf{m}^*}$
$\eta^{F\varepsilon em} = \eta + \frac{1}{\rho}\frac{\mathbf{e}^*}{\theta} \cdot \mathbf{p}^*$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\varepsilon em}}{\partial\mathbf{F}} - J\mu_o(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$		
	$\frac{1}{\theta} = \frac{\partial\eta^{F\varepsilon em}}{\partial\varepsilon}$	$\mathbf{p}^* = \rho\frac{\partial\eta^{F\varepsilon em}}{\partial\left(\frac{\mathbf{e}^*}{\theta}\right)}$	$\mathbf{h}^* = -\frac{\rho\theta}{\mu_o}\frac{\partial\eta^{F\varepsilon em}}{\partial\mathbf{m}^*}$
$\eta^{F\varepsilon ph} = \eta + \frac{\mu_o}{\rho}\frac{\mathbf{h}^*}{\theta} \cdot \mathbf{m}^*$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\varepsilon ph}}{\partial\mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^*)\mathbf{F}^{-T}$		
	$\frac{1}{\theta} = \frac{\partial\eta^{F\varepsilon ph}}{\partial\varepsilon}$	$\mathbf{e}^* = -\rho\theta\frac{\partial\eta^{F\varepsilon ph}}{\partial\mathbf{p}^*}$	$\mathbf{m}^* = \frac{\rho}{\mu_o}\frac{\partial\eta^{F\varepsilon ph}}{\partial\left(\frac{\mathbf{h}^*}{\theta}\right)}$
$\eta^{F\varepsilon eh} = \eta + \frac{1}{\rho}\frac{\mathbf{e}^*}{\theta} \cdot \mathbf{p}^* + \frac{\mu_o}{\rho}\frac{\mathbf{h}^*}{\theta} \cdot \mathbf{m}^*$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\varepsilon eh}}{\partial\mathbf{F}}$		
	$\frac{1}{\theta} = \frac{\partial\eta^{F\varepsilon eh}}{\partial\varepsilon}$	$\mathbf{p}^* = \rho\frac{\partial\eta^{F\varepsilon eh}}{\partial\left(\frac{\mathbf{e}^*}{\theta}\right)}$	$\mathbf{m}^* = \frac{\rho}{\mu_o}\frac{\partial\eta^{F\varepsilon eh}}{\partial\left(\frac{\mathbf{h}^*}{\theta}\right)}$

Table 11
Fully coupled state equations for entropy Family 2.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$

Entropy potential	State equations	
$\eta^{F\theta pm} = \eta - \frac{\varepsilon}{\theta}$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\theta pm}}{\partial\mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^* + \mu_o\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$	$\mathbf{e}^* = -\rho\theta\frac{\partial\eta^{F\theta pm}}{\partial\mathbf{p}^*}$
	$\varepsilon = \theta^2\frac{\partial\eta^{F\theta pm}}{\partial\theta}$	$\mathbf{h}^* = -\frac{\rho\theta}{\mu_o}\frac{\partial\eta^{F\theta pm}}{\partial\mathbf{m}^*}$
$\eta^{F\theta em} = \eta - \frac{\varepsilon}{\theta} + \frac{\mathbf{p}^*}{\rho} \cdot \frac{\mathbf{e}^*}{\theta}$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\theta em}}{\partial\mathbf{F}} - J\mu_o(\mathbf{h}^* \cdot \mathbf{m}^*)\mathbf{F}^{-T}$	$\mathbf{p}^* = \rho\theta\frac{\partial\eta^{F\theta em}}{\partial\mathbf{e}^*}$
	$\varepsilon = \theta^2\frac{\partial\eta^{F\theta em}}{\partial\theta} + \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*$	$\mathbf{h}^* = -\frac{\rho\theta}{\mu_o}\frac{\partial\eta^{F\theta em}}{\partial\mathbf{m}^*}$
$\eta^{F\theta ph} = \eta - \frac{\varepsilon}{\theta} + \mu_o\frac{\mathbf{m}^*}{\rho} \cdot \frac{\mathbf{h}^*}{\theta}$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\theta ph}}{\partial\mathbf{F}} - J(\mathbf{e}^* \cdot \mathbf{p}^*)\mathbf{F}^{-T}$	$\mathbf{e}^* = -\rho\theta\frac{\partial\eta^{F\theta ph}}{\partial\mathbf{p}^*}$
	$\varepsilon = \theta^2\frac{\partial\eta^{F\theta ph}}{\partial\theta} + \frac{\mu_o}{\rho}\mathbf{m}^* \cdot \mathbf{h}^*$	$\mathbf{m}^* = \frac{\rho\theta}{\mu_o}\frac{\partial\eta^{F\theta ph}}{\partial\mathbf{h}^*}$
$\eta^{F\theta eh} = \eta - \frac{\varepsilon}{\theta} + \frac{\mathbf{p}^*}{\rho} \cdot \frac{\mathbf{e}^*}{\theta} + \mu_o\frac{\mathbf{m}^*}{\rho} \cdot \frac{\mathbf{h}^*}{\theta}$	$\mathbf{P} = -\rho_R\theta\frac{\partial\eta^{F\theta eh}}{\partial\mathbf{F}}$	$\mathbf{p}^* = \rho\theta\frac{\partial\eta^{F\theta eh}}{\partial\mathbf{e}^*}$
	$\varepsilon = \theta^2\frac{\partial\eta^{F\theta eh}}{\partial\theta} + \frac{\mu_o}{\rho}\mathbf{m}^* \cdot \mathbf{h}^* + \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*$	$\mathbf{m}^* = \frac{\rho\theta}{\mu_o}\frac{\partial\eta^{F\theta eh}}{\partial\mathbf{h}^*}$

Table 12
Fully coupled state equations for entropy Family 3.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$		
Entropy potential	State equations	
$\eta^{P\varepsilon pm} = \eta + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta}$	$\mathbf{F} = \rho_R \frac{\partial \eta^{P\varepsilon pm}}{\partial(\frac{\mathbf{P}}{\theta})}$ $\mathbf{e}^* = -\theta \frac{\partial \eta^{P\varepsilon pm}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\frac{1}{\theta} = \frac{\partial \eta^{P\varepsilon pm}}{\partial \varepsilon}$ $\mathbf{h}^* = -\frac{\theta}{\mu_o}\frac{\partial \eta^{P\varepsilon pm}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$\eta^{P\varepsilon em} = \eta + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta} + \frac{\mathbf{p}^*}{\rho} \cdot \frac{\mathbf{e}^*}{\theta}$	$\mathbf{F} = \rho_R \frac{\partial \eta^{P\varepsilon em}}{\partial(\frac{\mathbf{P}}{\theta})}$ $\mathbf{p}^* = \rho \frac{\partial \eta^{P\varepsilon em}}{\partial(\frac{\mathbf{e}^*}{\theta})}$	$\frac{1}{\theta} = \frac{\partial \eta^{P\varepsilon em}}{\partial \varepsilon}$ $\mathbf{h}^* = -\frac{\theta}{\mu_o}\frac{\partial \eta^{P\varepsilon em}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$\eta^{P\varepsilon ph} = \eta + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta} + \mu_o \frac{\mathbf{m}^*}{\rho} \cdot \frac{\mathbf{h}^*}{\theta}$	$\mathbf{F} = \rho_R \frac{\partial \eta^{P\varepsilon ph}}{\partial(\frac{\mathbf{P}}{\theta})}$ $\mathbf{e}^* = -\theta \frac{\partial \eta^{P\varepsilon ph}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\frac{1}{\theta} = \frac{\partial \eta^{P\varepsilon ph}}{\partial \varepsilon}$ $\mathbf{m}^* = \frac{\rho}{\mu_o}\frac{\partial \eta^{P\varepsilon ph}}{\partial(\frac{\mathbf{h}^*}{\theta})}$
$\eta^{P\varepsilon eh} = \eta + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta} + \frac{\mathbf{p}^*}{\rho} \cdot \frac{\mathbf{e}^*}{\theta} + \mu_o \frac{\mathbf{m}^*}{\rho} \cdot \frac{\mathbf{h}^*}{\theta}$	$\mathbf{F} = \rho_R \frac{\partial \eta^{P\varepsilon eh}}{\partial(\frac{\mathbf{P}}{\theta})}$ $\mathbf{p}^* = \rho \frac{\partial \eta^{P\varepsilon eh}}{\partial(\frac{\mathbf{e}^*}{\theta})}$	$\frac{1}{\theta} = \frac{\partial \eta^{P\varepsilon eh}}{\partial \varepsilon}$ $\mathbf{m}^* = \frac{\rho}{\mu_o}\frac{\partial \eta^{P\varepsilon eh}}{\partial(\frac{\mathbf{h}^*}{\theta})}$

Table 13
Fully coupled state equations for entropy Family 4.

Residual dissipation inequality: $\mathbf{j}^* \cdot \mathbf{e}^* - \frac{1}{\theta}\mathbf{q} \cdot grad\,\theta \geqslant 0$			
Entropy potential	State equations		
$\eta^{P\theta pm} = \eta - \frac{\varepsilon}{\theta} + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta}$	$\varepsilon = \theta^2 \frac{\partial \eta^{P\theta pm}}{\partial \theta} + \frac{1}{\rho_R}\mathbf{P} \cdot \mathbf{F}$ $\mathbf{F} = \rho_R\theta \frac{\partial \eta^{P\theta pm}}{\partial \mathbf{P}}$	$\mathbf{e}^* = -\theta \frac{\partial \eta^{P\theta pm}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\mathbf{h}^* = -\frac{\theta}{\mu_o}\frac{\partial \eta^{P\theta pm}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$\eta^{P\theta em} = \eta - \frac{\varepsilon}{\theta} + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta} + \frac{\mathbf{p}^*}{\rho} \cdot \frac{\mathbf{e}^*}{\theta}$	$\varepsilon = \theta^2 \frac{\partial \eta^{P\theta em}}{\partial \theta} + \frac{1}{\rho_R}\mathbf{P} \cdot \mathbf{F} + \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*$ $\mathbf{F} = \rho_R\theta \frac{\partial \eta^{P\theta em}}{\partial \mathbf{P}}$	$\mathbf{p}^* = \rho\theta \frac{\partial \eta^{P\theta em}}{\partial \mathbf{e}^*}$	$\mathbf{h}^* = -\frac{\theta}{\mu_o}\frac{\partial \eta^{P\theta em}}{\partial(\frac{\mathbf{m}^*}{\rho})}$
$\eta^{P\theta ph} = \eta - \frac{\varepsilon}{\theta} + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta} + \mu_o \frac{\mathbf{m}^*}{\rho} \cdot \frac{\mathbf{h}^*}{\theta}$	$\varepsilon = \theta^2 \frac{\partial \eta^{P\theta ph}}{\partial \theta} + \frac{1}{\rho_R}\mathbf{P} \cdot \mathbf{F} + \frac{\mu_o}{\rho}\mathbf{m}^* \cdot \mathbf{h}^*$ $\mathbf{F} = \rho_R\theta \frac{\partial \eta^{P\theta ph}}{\partial \mathbf{P}}$	$\mathbf{e}^* = -\theta \frac{\partial \eta^{P\theta ph}}{\partial(\frac{\mathbf{p}^*}{\rho})}$	$\mathbf{m}^* = \frac{\rho\theta}{\mu_o}\frac{\partial \eta^{P\theta ph}}{\partial \mathbf{h}^*}$
$\eta^{P\theta eh} = \eta - \frac{\varepsilon}{\theta} + \frac{1}{\rho_R}\mathbf{F} \cdot \frac{\mathbf{P}}{\theta} + \frac{\mathbf{p}^*}{\rho} \cdot \frac{\mathbf{e}^*}{\theta} + \mu_o \frac{\mathbf{m}^*}{\rho} \cdot \frac{\mathbf{h}^*}{\theta}$	$\varepsilon = \theta^2 \frac{\partial \eta^{P\theta eh}}{\partial \theta} + \frac{1}{\rho_R}\mathbf{P} \cdot \mathbf{F} + \frac{\mu_o}{\rho}\mathbf{m}^* \cdot \mathbf{h}^* + \frac{1}{\rho}\mathbf{e}^* \cdot \mathbf{p}^*$ $\mathbf{F} = \rho_R\theta \frac{\partial \eta^{P\theta eh}}{\partial \mathbf{P}}$	$\mathbf{p}^* = \rho\theta \frac{\partial \eta^{P\theta eh}}{\partial \mathbf{e}^*}$	$\mathbf{m}^* = \frac{\rho\theta}{\mu_o}\frac{\partial \eta^{P\theta eh}}{\partial \mathbf{h}^*}$

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