

ON MAGNETIC RELAXATION EQUATION FOR ISOTROPIC MAGNETIZABLE REACTING FLUID MIXTURES*

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*Dedicated by Prof. Lidia Palese and Dr. Arcangelo Labianca to Prof.
Liliana Restuccia on the occasion of her 70th anniversary*

DOI 10.56082/annalsarscimath.2025.3.409

Abstract

In a previous paper, a linear theory for magnetic relaxation phenomena in reacting anisotropic fluid mixtures was developed, within the framework of the irreversible processes thermodynamics with internal variables. In that model, the total magnetization was split into two irreversible contributions, introducing one of them as an internal variable. In this paper, the same approach is applied to isotropic and perfect isotropic magnetizable reacting fluid mixtures. In the latter case, it is shown that in the phenomenological equations no cross effects arise between magnetic relaxation and other irreversible phenomena, since fluxes and thermodynamic forces of different tensorial character do not couple (Curie's principle). For perfect isotropic mixtures, we also derive the equations of state and the magnetic relaxation equation, which, unlike in the anisotropic case, do not contain contributions from temperature, concentrations, and their time derivatives. Special cases are treated. The results have applications in nuclear resonance, biology, medicine, and other fields.

*Accepted for publication on December 12, 2025

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Keywords: continuum thermodynamics, classical irreversible thermodynamics, reacting fluid mixtures with magnetic relaxation, internal variables, isotropic media.

MSC: 76A99, 80A17.

1 Introduction

In previous studies [9], [10], [11], [34], [35], [36], [37], [38], [39], [16] linear theories for magnetic relaxation phenomena in magnetizable continuous media have been developed, within the framework of the thermodynamics of irreversible processes [2], [12], [7], [44], [23], [24], [25], [26], [22], [8], with internal variables [27], [19], [20], [1]. In [9], Kluitenberg, under linear approximation, assumed that the total specific magnetization could be expressed as the sum of one reversible part $\mathbf{m}^{(0)}$ and one irreversible part $\mathbf{m}^{(1)}$, and derived for isotropic media the classical Snoek equation describing magnetic relaxation phenomena (see [43]). Later, in [10], by considering the total specific magnetization as composed of two irreversible contributions, Kluitenberg obtained a more general relaxation equation. In [35], the theory was extended further: assuming that an arbitrary number n of microscopic phenomena contribute to the total specific magnetization, which can then be split into $n + 1$ irreversible parts, $\mathbf{m} = \mathbf{m}^{(0)} + \sum_{k=1}^n \mathbf{m}^{(k)}$, Kluitenberg together with one of the authors (L. R.) derived a generalized Snoek equation. This formulation took the form of a linear relation among the magnetic field, its first n time derivatives, the total magnetization field, and the first $n + 1$ time derivatives of this field. The results of [35] were subsequently reviewed and discussed in [36] and [37].

In [34] mixtures of n reacting fluid components, heat conducting and presenting magnetic and dielectric relaxation, were described within the same thermodynamic framework and the phenomenological equations were derived in the anisotropic and in the isotropic case. In [38], the focus shifted to anisotropic reacting fluid mixtures with magnetic relaxation. The irreversible microscopic phenomena responsible for relaxation were described by assuming that the total specific magnetization consists of two irreversible parts, $\mathbf{m}^{(0)}$ and $\mathbf{m}^{(1)}$, and in the linear case the corresponding magnetic relaxation equation was derived. In [39] the heat conduction equation and the dissipation function were derived for anisotropic magnetizable media with relaxation, under the assumption of two irreversible contributions. In [16] the heat conduction equation and the heat dissipation function for anisotropic

mixtures composed of n reacting fluid components with magnetic relaxation were carried out.

In [29], [30], [31], [32], [33] analogous studies for dielectric relaxation phenomena in polarizable media with internal variables were performed by using the same methods of the classical thermodynamics of irreversible processes with internal variables (see [38]).

In the present paper, we derive a magnetic relaxation equation for perfect isotropic reacting fluid mixtures. The paper is organized as follows. In Sections 2, 3, and 6 we review the model previously developed by the authors in [38] for anisotropic reacting fluid mixtures, presenting the governing equations describing all the processes involved, the entropy balance, the phenomenological equations, and the equations of state. In Sections 4 and 5 we derive the entropy balance equation and the phenomenological equations for the cases in which the reacting fluid mixtures are isotropic, i.e. their properties are invariant under all orthogonal rotations of the reference frame, and perfect isotropic, meaning invariant under both orthogonal rotations and inversions of the reference axes. In the latter case, it is shown that no cross effects arise between magnetic relaxation and other irreversible phenomena, since fluxes and thermodynamic forces of different tensorial character do not couple (Curie's principle). Sections 7, 8, and 9 are devoted to the perfect isotropic case, where we obtain the equations of state, the constitutive equations, and the magnetic relaxation equation, where unlike in the anisotropic case, no contributions arise from the temperature, the concentrations of the n fluid components, and their first time derivatives. We also derive expressions for the the field $\mathbf{B}^{(1)}$, conjugated to the partial specific magnetization $\mathbf{m}^{(1)}$, the specific internal energy, and the specific entropy. In Section 10, special perfect isotropic magnetizable fluid systems are treated. The results obtained have applications in medicine, biology, and nuclear resonance, where different molecular species, characterized by distinct magnetic susceptibilities and relaxation times, contribute to the total magnetization. Finally, we recall that a continuum phenomenological theory with internal variables for magnetizable media with relaxation phenomena, composed of multiple ionic species, was developed by Maugin in [17, 18] to explain the internal mechanisms of such media.

2 Recalling the model for magnetizable reacting fluid mixtures

In this Section, we present the model for magnetizable reacting fluid mixtures, consisting of n reacting chemical components having magnetic relaxation, given in [38], within the framework of classical irreversible thermodynamics with internal variables. The standard Cartesian tensor notation in a rectangular coordinate system is used and the equations governing the behavior of these media are considered in a current configuration $\mathcal{K}_t$ (see [2], [34]).

The conservation of mass is expressed by

$$\frac{d\varrho}{dt} + \varrho \nabla \cdot \mathbf{v} = 0, \quad (1)$$

where ϱ and $\mathbf{v}$ are the total mass density and the barycentric velocity, respectively, defined by

$$\varrho = \sum_{k=1}^n \varrho^{(k)}, \quad \mathbf{v} = \frac{1}{\varrho} \sum_{k=1}^n \varrho^{(k)} \mathbf{v}^{(k)}, \quad (2)$$

with $\varrho^{(k)}$ and $\mathbf{v}^{(k)}$ the mass density and the velocity of the k -th chemical component, respectively. Furthermore, the symbol " $\nabla \cdot$ " denotes the divergence operator, $\frac{d}{dt}$ is material time derivative, defined by $\frac{d}{dt} = \frac{\partial}{\partial t} + v_i \frac{\partial}{\partial x_i}$, with $\frac{\partial}{\partial t}$ the time partial derivative and $\frac{\partial}{\partial x_i}$ the spatial partial derivative, and Einstein convention for repeated indices is used.

Let us introduce the mass fraction $c^{(k)}$ and the diffusion flux $\mathbf{J}_{(diff)}^{(k)}$ of the k -th substance, with respect to the barycentric motion, by the following definitions, see also (2),

$$c^{(k)} = \frac{\varrho^{(k)}}{\varrho}, \quad \mathbf{J}_{(diff)}^{(k)} = \varrho^{(k)}(\mathbf{v}^{(k)} - \mathbf{v}) \quad (k = 1, 2, \dots, n),$$

$$\text{with } \sum_{k=1}^n c^{(k)} = 1, \quad \sum_{k=1}^n \mathbf{J}_{(diff)}^{(k)} = \mathbf{0}, \quad (3)$$

being only $n - 1$ of the n diffusion fluxes independent.

The balance equations for the mass fractions $c^{(k)}$ are given by [2]

$$\varrho \frac{dc^{(k)}}{dt} = -\nabla \cdot \mathbf{J}_{(diff)}^{(k)} + \sum_{h=1}^r \nu^{(kh)} J_{(chem)}^{(h)} \quad (k = 1, 2, \dots, n), \quad (4)$$

where the quantity $\nu^{(kh)}$ divided by the molecular mass $\mathcal{M}^{(k)}$ of the k -th fluid component is proportional to the stoichiometric coefficient with which the k -th component appears in the h -th chemical reaction, $J_{(chem)}^{(h)}$ is the chemical reaction rate of the h -th chemical reaction and $\nu^{(kh)} J_{(chem)}^{(h)}$ is the production of k -th component per unit volume and per unit time by the h -th chemical reaction,

Maxwell's equations in Galilean approximation (in the rationalized Gauss system) keep the form

$$\begin{cases} \nabla \times \mathbf{H} - \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} = \frac{1}{c} \mathbf{I}, \\ \nabla \cdot \mathbf{E} = \varrho^{(el)}, \\ \nabla \times \mathbf{E} + \frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} = \mathbf{0}, \\ \nabla \cdot \mathbf{B} = 0, \end{cases} \quad (5)$$

where c is the light velocity, $\mathbf{E}$, $\mathbf{B}$ and $\mathbf{H}$ are the electric field, the magnetic field and the magnetic displacement field, respectively, $\varrho^{(el)}$ and $\mathbf{I}$ are the electric charge density per unit volume and the total electric current density, respectively, given by

$$\varrho^{(el)} = \varrho e = \sum_{k=1}^n \varrho^{(k)} e^{(k)}, \quad \mathbf{I} = \sum_{k=1}^n \varrho^{(k)} e^{(k)} \mathbf{v}^{(k)}, \quad (6)$$

satisfying the following *charge conservation law*

$$\frac{\partial \varrho^{(el)}}{\partial t} = -\nabla \cdot \mathbf{I}, \quad \text{where } \mathbf{I} = \varrho^{(el)} \mathbf{v} + \sum_{k=1}^n e^{(k)} \mathbf{J}_{(diff)}^{(k)}. \quad (7)$$

In (7) $e^{(k)}$ is the charge per unit of mass of the k -th fluid component, $\varrho^{(el)} \mathbf{v}$ is the convection electric current and the conduction current, $\mathbf{j}^{(el)}$, due to the relative motion of the charges of the n fluid components, is given by

$$\mathbf{j}^{(el)} = \sum_{k=1}^n e^{(k)} \mathbf{J}_{(diff)}^{(k)}. \quad (8)$$

Furthermore, let us introduce the magnetization $\mathbf{M}$ and the specific magnetization $\mathbf{m}$, that are axial vectors, defined by

$$\mathbf{M} = \mathbf{B} - \mathbf{H}, \quad \mathbf{m} = \frac{1}{\varrho} \mathbf{M}. \quad (9)$$

For the media under consideration the polarization vector, defined by $\mathbf{P} = \mathbf{D} - \mathbf{E}$, with $\mathbf{D}$ the electric displacement field, is null, then $\mathbf{D} = \mathbf{E}$ in equations (5)₁ and (5)₂.

The *first law of thermodynamics* for magnetizable fluid mixtures in an electromagnetic field, in Galilean approximation, when we assume that the velocity of the medium under consideration is small compared with the velocity of the light, is given by [2]

$$\varrho \frac{du}{dt} = -\nabla \cdot \mathbf{J}^{(q)} + \tau_{\alpha\beta} \frac{d\varepsilon_{\alpha\beta}}{dt} + \mathbf{j}^{(el)} \cdot \mathbf{E} + \varrho \mathbf{B} \cdot \frac{d\mathbf{m}}{dt}, \quad (10)$$

where u is the specific internal energy of the system, $\mathbf{J}^{(q)}$ is the heat flux density, $\tau_{\alpha\beta}$ is the symmetric mechanical stress tensor, $\frac{d\varepsilon_{\alpha\beta}}{dt}$ is the small strain rate tensor, given by

$$\frac{d\varepsilon_{\alpha\beta}}{dt} = \frac{1}{2} \left(\frac{\partial v_\alpha}{\partial x_\beta} + \frac{\partial v_\beta}{\partial x_\alpha} \right) \quad (\alpha, \beta = 1, 2, 3), \quad (11)$$

and the heat source is neglected. For the relativistic formulation of equation (10) see [14] and [15]. On the right hand side of equation (10) the contributions describe the heat supply, the work done by mechanical stress, the Joule heat and the work done by the magnetic field to change the magnetization, respectively.

The *equation of motion* of a magnetizable medium in an electromagnetic field is given by (see [2], [35])

$$\varrho \frac{d\mathbf{v}}{dt} = -\nabla \cdot \boldsymbol{\tau} + \rho^{(el)} \mathbf{E} + \frac{1}{2} \mathbf{j}^{(el)} \times \mathbf{B} + (\nabla \mathbf{B}) \cdot \mathbf{M} - \frac{1}{c} \frac{d(\mathbf{M} \times \mathbf{E})}{dt}, \quad (12)$$

where the force per unit of mass is neglected and the velocity field $\mathbf{v}$ is defined by $\mathbf{v} = \frac{d\mathbf{u}}{dt}$, with $\mathbf{u}$ the displacement field.

3 Phenomenological equations and entropy production for anisotropic reacting fluid mixtures with magnetic relaxation

In this Section we give a review of some results obtained in [38] for anisotropic reacting fluid mixtures with magnetic relaxation (see also the model for reacting fluid mixtures with magnetic and dielectric relaxation in [34]). Let us assume that the total specific magnetization $\mathbf{m}$ is composed of two irreversible parts $\mathbf{m}^{(0)}$ and $\mathbf{m}^{(1)}$, describing the magnetic relaxation phenomena

due to irreversible microscopic phenomena within the considered media, i.e. $\mathbf{m} = \mathbf{m}^{(0)} + \mathbf{m}^{(1)}$. Let us introduce $\mathbf{m}^{(1)}$ in the thermodynamic state space as internal variable to describe these phenomena. Thus, we assume that the specific entropy s (i.e. the entropy per unit of mass) is a function of the specific internal energy u , the strain tensor $\varepsilon_{\alpha\beta}$, the specific magnetization $\mathbf{m}^{(1)}$, the concentrations $c^{(k)}$ of the n fluid components ($k = 1, 2, \dots, n$)

$$s = s(u, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}). \quad (13)$$

We shall define the equilibrium temperature T , the equilibrium stress tensor $\tau_{\alpha\beta}^{(eq)}$, the equilibrium magnetic field $\mathbf{B}^{(eq)}$, the thermodynamic affinity $\mathbf{B}^{(1)}$, conjugate to the internal variable $\mathbf{m}^{(1)}$, and the thermodynamic or chemical potential $\mu^{(k)}$ of the fluid component k , respectively, by

$$\left\{ \begin{array}{l} T^{-1} = \frac{\partial}{\partial u} s(u, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}), \\ \tau_{\alpha\beta}^{(eq)} = \varrho T \frac{\partial}{\partial \varepsilon_{\alpha\beta}} s(u, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}), \\ \mathbf{B}^{(eq)} = -T \frac{\partial}{\partial \mathbf{m}} s(u, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}), \\ \mathbf{B}^{(1)} = T \frac{\partial}{\partial \mathbf{m}^{(1)}} s(u, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}), \\ \mu^{(k)} = -T \frac{\partial}{\partial c^{(k)}} s(u, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}) \quad (k = 1, \dots, n). \end{array} \right. \quad (14)$$

Let us expand the specific entropy (13) into Taylor's series with respect to a local equilibrium state, taking into account very small deviations with respect to this state and confining our consideration to the linear terms, we obtain the differential of the entropy s in a point of the thermodynamical phase space (see [28] and [21]), called Gibbs relation, given by

$$Tds = du - \frac{1}{\varrho} \tau_{\alpha\beta}^{(eq)} d\varepsilon_{\alpha\beta} - \mathbf{B}^{(eq)} \cdot d\mathbf{m} + \mathbf{B}^{(1)} \cdot d\mathbf{m}^{(1)} - \sum_{k=1}^n \mu^{(k)} dc^{(k)}, \quad (15)$$

where we have used equations (14). Thus, the time derivative of the entropy s in the considered point of the thermodynamical phase space takes the form

$$\frac{ds}{dt} = \frac{du}{dt} - \frac{1}{\varrho} \tau_{\alpha\beta}^{(eq)} \frac{d\varepsilon_{\alpha\beta}}{dt} - \mathbf{B}^{(eq)} \cdot \frac{d\mathbf{m}}{dt} + \mathbf{B}^{(1)} \cdot \frac{d\mathbf{m}^{(1)}}{dt} - \sum_{k=1}^n \mu^{(k)} \frac{dc^{(k)}}{dt}. \quad (16)$$

Multiplying both sides of equation (4) by $\frac{\mu^{(k)}}{T}$, summing over k and introducing the chemical affinity of each reaction h -th, $A^{(h)} = -\sum_{k=1}^n \mu^{(k)} \nu^{(kh)}$

$$(h = 1, 2, \dots, r), \text{ we have } \quad \frac{\varrho}{T} \sum_{k=1}^n \mu^{(k)} \frac{dc^{(k)}}{dt} = -\nabla \cdot \left(\frac{1}{T} \sum_{k=1}^n \mu^{(k)} \mathbf{J}_{(diff)}^{(k)} \right) + \sum_{k=1}^n \mathbf{J}_{(diff)}^{(k)} \cdot \nabla \left(\frac{\mu^{(k)}}{T} \right) - \frac{1}{T} \sum_{h=1}^r A^{(h)} J_{(chem)}^{(h)}. \quad (17)$$

Using (10), (16) and (17), we obtain the following form for the entropy balance equation

$$\begin{aligned} \varrho \frac{ds}{dt} = & -\nabla \cdot \mathbf{J}^{(s)} + \frac{1}{T} \left(-\frac{1}{T} \mathbf{J}^{(q)} \cdot \nabla T + \tau_{\alpha\beta}^{(vi)} \frac{d\varepsilon_{\alpha\beta}}{dt} + \varrho \mathbf{B}^{(ir)} \cdot \frac{d\mathbf{m}}{dt} \right. \\ & \left. + \varrho \mathbf{B}^{(1)} \cdot \frac{d\mathbf{m}^{(1)}}{dt} + \sum_{h=1}^r A^{(h)} J_{(chem)}^{(h)} \right) - \sum_{k=1}^n \mathbf{J}_{(diff)}^{(k)} \cdot \nabla \left(\frac{\mu^{(k)}}{T} \right) + \frac{1}{T} \mathbf{j}^{(el)} \cdot \mathbf{E}, \end{aligned} \quad (18)$$

where $\mathbf{J}^{(s)}$, $\tau_{\alpha\beta}^{(vi)}$ and $\mathbf{B}^{(ir)}$ are the entropy flux, the viscous stress tensor and the irreversible magnetic field, respectively, defined by

$$\mathbf{J}^{(s)} = \frac{1}{T} \left(\mathbf{J}^{(q)} - \sum_{k=1}^n \mu^{(k)} \mathbf{J}_{(diff)}^{(k)} \right), \quad (19)$$

$$\tau_{\alpha\beta}^{(vi)} = \tau_{\alpha\beta} - \tau_{\alpha\beta}^{(eq)} \quad (\alpha, \beta = 1, 2, 3), \quad \mathbf{B}^{(ir)} = \mathbf{B} - \mathbf{B}^{(eq)}. \quad (20)$$

Defining the thermodynamic force $\chi^{(k)}$, conjugate to the diffusion flux $\mathbf{J}_{(diff)}^{(k)}$, by

$$\chi^{(k)} = - \left[T \nabla \left(\frac{\mu^{(k)}}{T} \right) - e^{(k)} \mathbf{E} \right] \quad (k = 1, \dots, n), \quad (21)$$

by virtue of equation (8) we have

$$\begin{aligned} & - \sum_{k=1}^n \mathbf{J}_{(diff)}^{(k)} \cdot \nabla \left(\frac{\mu^{(k)}}{T} \right) + \frac{1}{T} \mathbf{j}^{(el)} \cdot \mathbf{E} \\ & = \frac{1}{T} \sum_{k=1}^n \mathbf{J}_{(diff)}^{(k)} \cdot \chi^{(k)} = \frac{1}{T} \sum_{k=1}^{n-1} \mathbf{J}_{(diff)}^{(k)} \cdot (\chi^{(k)} - \chi^{(n)}) = \frac{1}{T} \sum_{k=1}^{n-1} \mathbf{J}_{(diff)}^{(k)} \cdot \mathbf{X}^{(k)}, \end{aligned} \quad (22)$$

where we have used relations (3) and defined $\mathbf{X}^{(k)} = \chi^{(k)} - \chi^{(n)}$.

Using (22), the entropy balance equation (18) can be written in the following form

$$\varrho \frac{ds}{dt} = -\nabla \cdot \mathbf{J}^{(s)} + \sigma^{(s)}, \quad (23)$$

where $\sigma^{(s)}$ is the entropy production per unit volume and per unit time, given by

$$\begin{aligned} \sigma^{(s)} = & \frac{1}{T} \left(\varrho \mathbf{B}^{(ir)} \cdot \frac{d\mathbf{m}}{dt} + \varrho \mathbf{B}^{(1)} \cdot \frac{d\mathbf{m}^{(1)}}{dt} - \frac{1}{T} \mathbf{J}^{(q)} \cdot \nabla T \right. \\ & \left. + \sum_{k=1}^{n-1} \mathbf{J}_{(diff)}^{(k)} \mathbf{X}^{(k)} + \sum_{h=1}^r A^{(h)} J_{(chem)}^{(h)} + \tau_{\alpha\beta}^{(vi)} \frac{d\varepsilon_{\alpha\beta}}{dt} \right). \end{aligned} \quad (24)$$

In the expression (24) $\sigma^{(s)}$ is the entropy production defined non-negative quantity by the *second law of thermodynamics*: $\sigma^{(s)} \geq 0$.

$\sigma^{(s)} = 0$ when the system is in *thermodynamic equilibrium*, i.e. when there are not irreversible processes inside the system. In thermodynamic equilibrium the sources (the body force, the heat source, the external entropy production) are null, the velocity of the medium is constant or null, the fluxes of physical quantities, the gradients and the time derivatives of the fields are null, "the internal variables become dependent on the variables of the equilibrium subspace, but they are not determined by them, i.e., the equilibrium subspace is many-valued" [24].

From (24) it is seen that the entropy production $\sigma^{(s)}$ is a bilinear form composed of a sum of six terms, where each term is a product of two factors, of which one is a flux and the other is the thermodynamic force or affinity, conjugate to the flux

According the usual procedure of non-equilibrium thermodynamics [2], [12], we have for magnetizable reacting fluid mixtures the following phenomenological equations, in which the irreversible fluxes $B_{\alpha}^{(ir)}$, $\varrho \frac{dm_{\alpha}^{(1)}}{dt}$, $J_{\alpha}^{(q)}$, $J_{(diff)\alpha}^{(j)}$ ($j = 1, 2, \dots, n-1$), $J_{(chem)}^{(l)}$ ($l = 1, 2, \dots, r$), $\tau_{\alpha\beta}^{(vi)}$ are *linear functions* of the thermodynamic forces $\frac{dm_{\beta}}{dt}$, $B_{\beta}^{(1)}$, $-\frac{1}{T} \frac{\partial T}{\partial x_{\beta}}$, $X_{\beta}^{(k)}$ ($k = 1, 2, \dots, n-1$), $A^{(h)}$ ($h = 1, 2, \dots, r$), $\frac{d\varepsilon_{\beta\gamma}}{dt}$ (see [38])

$$B_{\alpha}^{(ir)} = \varrho L_{(M)\alpha\beta}^{(0,0)} \frac{dm_{\beta}}{dt} + L_{(M)\alpha\beta}^{(0,1)} B_{\beta}^{(1)} - \frac{1}{T} L_{(M)\alpha\beta}^{(0,q)} \frac{\partial T}{\partial x_{\beta}} + \sum_{k=1}^{n-1} L_{(MD)\alpha\beta}^{(0,k)} X_{\beta}^{(k)}$$

$$+ \sum_{h=1}^r L_{(MC)\alpha}^{(0,h)} A^{(h)} + L_{(M)\alpha\beta\gamma}^{(0,vi)} \frac{d\varepsilon_{\beta\gamma}}{dt}, \quad (25)$$

$$\begin{aligned} \varrho \frac{dm_{\alpha}^{(1)}}{dt} = & \varrho L_{(M)\alpha\beta}^{(1,0)} \frac{dm_{\beta}}{dt} + L_{(M)\alpha\beta}^{(1,1)} B_{\beta}^{(1)} - \frac{1}{T} L_{(M)\alpha\beta}^{(1,q)} \frac{\partial T}{\partial x_{\beta}} + \sum_{k=1}^{n-1} L_{(MD)\alpha\beta}^{(1,k)} X_{\beta}^{(k)} \\ & + \sum_{h=1}^r L_{(MC)\alpha}^{(1,h)} A^{(h)} + L_{(M)\alpha\beta\gamma}^{(1,vi)} \frac{d\varepsilon_{\beta\gamma}}{dt}, \end{aligned} \quad (26)$$

$$\begin{aligned} J_{\alpha}^{(q)} = & \varrho L_{(M)\alpha\beta}^{(q,0)} \frac{dm_{\beta}}{dt} + L_{(M)\alpha\beta}^{(q,1)} B_{\beta}^{(1)} - \frac{1}{T} L_{\alpha\beta}^{(q,q)} \frac{\partial T}{\partial x_{\beta}} + \sum_{k=1}^{n-1} L_{(D)\alpha\beta}^{(q,k)} X_{\beta}^{(k)} \\ & + \sum_{h=1}^r L_{(C)\alpha}^{(q,h)} A^{(h)} + L_{\alpha\beta\gamma}^{(q,vi)} \frac{d\varepsilon_{\beta\gamma}}{dt}, \end{aligned} \quad (27)$$

$$\begin{aligned} J_{(diff)\alpha}^{(j)} = & \varrho L_{(DM)\alpha\beta}^{(j,0)} \frac{dm_{\beta}}{dt} + L_{(DM)\alpha\beta}^{(j,1)} B_{\beta}^{(1)} - \frac{1}{T} L_{(D)\alpha\beta}^{(j,q)} \frac{\partial T}{\partial x_{\beta}} + \sum_{k=1}^{n-1} L_{(DD)\alpha\beta}^{(j,k)} X_{\beta}^{(k)} \\ & + \sum_{h=1}^r L_{(DC)\alpha}^{(j,h)} A^{(h)} + L_{(D)\alpha\beta\gamma}^{(j,vi)} \frac{d\varepsilon_{\beta\gamma}}{dt} \quad (j = 1, 2, \dots, n-1), \end{aligned} \quad (28)$$

$$\begin{aligned} J_{(chem)}^{(l)} = & \varrho L_{(CM)\beta}^{(l,0)} \frac{dm_{\beta}}{dt} + L_{(CM)\beta}^{(l,1)} B_{\beta}^{(1)} - \frac{1}{T} L_{(C)\beta}^{(l,q)} \frac{\partial T}{\partial x_{\beta}} + \sum_{k=1}^{n-1} L_{(CD)\beta}^{(l,k)} X_{\beta}^{(k)} \\ & + \sum_{h=1}^r L_{(CC)}^{(l,h)} A^{(h)} + L_{(C)\beta\gamma}^{(l,vi)} \frac{d\varepsilon_{\beta\gamma}}{dt} \quad (l = 1, 2, \dots, r), \end{aligned} \quad (29)$$

$$\begin{aligned} \tau_{\alpha\beta}^{(vi)} = & \varrho L_{(M)\alpha\beta\gamma}^{(vi,0)} \frac{dm_{\gamma}}{dt} + L_{(M)\alpha\beta\gamma}^{(vi,1)} B_{\gamma}^{(1)} - \frac{1}{T} L_{\alpha\beta\gamma}^{(vi,q)} \frac{\partial T}{\partial x_{\gamma}} + \sum_{k=1}^{n-1} L_{(D)\alpha\beta\gamma}^{(vi,k)} X_{\gamma}^{(k)} \\ & + \sum_{h=1}^r L_{(C)\alpha\beta}^{(vi,h)} A^{(h)} + L_{\alpha\beta\gamma\delta}^{(vi,vi)} \frac{d\varepsilon_{\gamma\delta}}{dt}. \end{aligned} \quad (30)$$

In principle, all irreversible phenomena described by (25)-(30) can influence each other. For instance, the third, fourth, fifth and sixth term on the right-hand sides of (26) describe the influences of heat flux, diffusion fluxes, chemical reactions and viscous flow on magnetic relaxation. These phenomena are called cross effects. Equations (25) and (26) describe irreversible

changes in the magnetization, the phenomenological equations (27)-(29) describe the irreversible processes of heat flux, diffusion fluxes and chemical reactions. Equation (30) is a generalization of Newton's law for viscous fluid flow. The quantities $L_{(MM)\alpha\beta}^{(0,0)}$, $L_{(MM)\alpha\beta}^{(0,1)}$, $L_{(M)\alpha\beta}^{(0,q)}$, etc. which occur in (25)-(30) are called phenomenological tensors.

Because of the symmetry of $\varepsilon_{\alpha\beta}$, $\tau_{\alpha\beta}^{(eq)}$ and the viscous stress $\tau_{\alpha\beta}^{(vi)}$, one has the following symmetry relations (see [38])

$$\begin{aligned}
 L_{(M)\alpha\beta\gamma}^{(1,vi)} &= L_{(M)\alpha\gamma\beta}^{(1,vi)}, & L_{(M)\alpha\beta\gamma}^{(vi,1)} &= L_{(M)\beta\alpha\gamma}^{(vi,1)}, \\
 L_{\alpha\beta\gamma}^{(q,vi)} &= L_{\alpha\gamma\beta}^{(q,vi)}, & L_{\alpha\beta\gamma}^{(vi,q)} &= L_{\beta\alpha\gamma}^{(vi,q)}, \\
 L_{(M)\alpha\beta\gamma}^{(0,vi)} &= L_{(M)\alpha\gamma\beta}^{(0,vi)}, & L_{(M)\alpha\beta\gamma}^{(vi,0)} &= L_{(M)\beta\alpha\gamma}^{(vi,0)}, \\
 L_{(C)\alpha\beta}^{(l,vi)} &= L_{(C)\beta\alpha}^{(l,vi)}, & L_{(C)\alpha\beta}^{(vi,h)} &= L_{(C)\beta\alpha}^{(vi,h)} \quad (l, h = 1, 2, \dots, r), \\
 L_{(D)\alpha\beta\gamma}^{(j,vi)} &= L_{(D)\alpha\gamma\beta}^{(j,vi)}, & L_{(D)\alpha\beta\gamma}^{(vi,k)} &= L_{(D)\beta\alpha\gamma}^{(vi,k)} \quad (j, k = 1, 2, \dots, n-1), \\
 L_{\alpha\beta\gamma\delta}^{(vi,vi)} &= L_{\alpha\beta\delta\gamma}^{(vi,vi)} = L_{\beta\alpha\gamma\delta}^{(vi,vi)} = L_{\beta\alpha\delta\gamma}^{(vi,vi)}.
 \end{aligned} \tag{31}$$

In (24), from the microscopic point of view, the macroscopic physical quantities $\varrho \frac{d\mathbf{m}}{dt}$, $\varrho \frac{d\mathbf{m}^{(1)}}{dt}$, $A^{(h)}$ ($h = 1, 2, \dots, r$), $T^{-1} \text{grad} T$, $\mathbf{X}^{(k)}$ ($k = 1, 2, \dots, n-1$) and $\tau_{\alpha\beta}^{(vi)}$, are called even, because they are even functions of the microscopic particle velocities constituting the considered fluid system at the mesoscopic level, while $\mathbf{B}^{(ir)}$, $\mathbf{B}^{(1)}$, $\mathbf{J}^{(q)}$, $\mathbf{J}_{(diff)}^{(k)}$, ($k = 1, 2, \dots, r$), $\mathbf{J}^{(l)}$, ($l = 1, 2, \dots, r$), $\frac{d\varepsilon_{\alpha\beta}}{dt}$ are called odd, because they are odd functions of these velocities. From a macroscopic point of view, we distinguish the macroscopic quantities in even and odd when they are even or odd functions under time reversal, respectively. Hence, according to the usual procedure of non-equilibrium thermodynamics [2], [12] we have the following Onsager-Casimir reciprocity relations for the cross effects, which occur in (25)-(30) (see [38])

$$\begin{aligned}
 L_{(M)\alpha\beta}^{(0,0)} &= L_{(M)\beta\alpha}^{(0,0)}, & L_{(M)\alpha\beta}^{(1,1)} &= L_{(M)\beta\alpha}^{(1,1)}, & L_{\alpha\beta}^{(q,q)} &= L_{\beta\alpha}^{(q,q)}, \\
 L_{(M)\alpha\beta}^{(0,1)} &= -L_{(M)\beta\alpha}^{(1,0)}, & L_{(M)\alpha\beta}^{(0,q)} &= L_{(M)\beta\alpha}^{(q,0)}, & L_{\alpha\beta\gamma}^{(q,vi)} &= -L_{\beta\gamma\alpha}^{(vi,q)},
 \end{aligned}$$

$$\begin{aligned}
L_{(MD)\alpha\beta}^{(0,k)} &= L_{(DM)\beta\alpha}^{(k,0)}, & L_{(DD)\alpha\beta}^{(j,k)} &= L_{(DD)\beta\alpha}^{(k,j)} \quad (j, k = 1, 2, \dots, n-1), \\
L_{(MC)\alpha}^{(0,h)} &= L_{(CM)\alpha}^{(h,0)}, & L_{(DC)\alpha}^{(j,h)} &= L_{(CD)\alpha}^{(h,j)} \quad (j = 1, \dots, n-1), \quad (h = 1, \dots, r), \\
L_{(MD)\alpha\beta}^{(1,k)} &= -L_{(MD)\beta\alpha}^{(k,1)}, & L_{(D)\alpha\beta\gamma}^{(k,vi)} &= -L_{(D)\beta\gamma\alpha}^{(vi,k)} \quad (k = 1, 2, \dots, n-1), \\
L_{(M)\alpha\beta}^{(1,q)} &= -L_{(M)\beta\alpha}^{(q,1)}, & L_{(D)\alpha\beta}^{(q,k)} &= L_{(D)\beta\alpha}^{(k,q)} \quad (k = 1, 2, \dots, n-1), \\
L_{(MC)\alpha}^{(1,h)} &= -L_{(CM)\alpha}^{(h,1)}, & L_{(CC)}^{(l,h)} &= L_{(CC)}^{(h,l)} \quad (l, h = 1, 2, \dots, r), \\
L_{(C)\alpha}^{(q,h)} &= L_{(C)\alpha}^{(h,q)}, & L_{(M)\alpha\beta\gamma}^{(0,vi)} &= -L_{(M)\beta\gamma\alpha}^{(vi,0)}, \\
L_{(C)\alpha\beta}^{(h,vi)} &= -L_{(C)\alpha\beta}^{(vi,h)} \quad (h = 1, 2, \dots, r), & L_{(M)\alpha\beta\gamma}^{(1,vi)} &= L_{(M)\beta\gamma\alpha}^{(vi,1)}, & L_{\alpha\beta\gamma\delta}^{(vi,vi)} &= L_{\gamma\delta\alpha\beta}^{(vi,vi)}.
\end{aligned} \tag{32}$$

Onsager-Casimir reciprocity relations reduce the number of independent components of the phenomenological tensors. Relation (31)₁₁, by virtue of Onsager relations (32)₃, gives

$$L_{\alpha\beta\gamma\delta}^{(vi,vi)} = L_{\alpha\beta\delta\gamma}^{(vi,vi)} = L_{\beta\alpha\gamma\delta}^{(vi,vi)} = L_{\beta\alpha\delta\gamma}^{(vi,vi)} = L_{\gamma\delta\alpha\beta}^{(vi,vi)} = L_{\gamma\delta\beta\alpha}^{(vi,vi)} = L_{\delta\gamma\alpha\beta}^{(vi,vi)} = L_{\delta\gamma\beta\alpha}^{(vi,vi)}. \tag{33}$$

In [16], from (24) and the phenomenological equations, using Onsager-Casimir relations, the entropy production $\sigma^{(s)}$ was obtained, having a bilinear form in the components of the time derivative of the total specific magnetization vector, the components of the thermodynamic force conjugate to the partial specific magnetization, $\mathbf{B}^{(1)}$, the components of the k -th thermodynamic force $\mathbf{X}^{(k)}$, conjugate to the k -th diffusion flux $\mathbf{J}_{(diff)}^{(k)}$ ($k = 1, 2, \dots, n-1$), the chemical affinity of h -th each reaction A^h ($h = 1, 2, \dots, r$), the components of the temperature gradient and the components of the time derivative of the strain tensor

$$\begin{aligned}
\sigma^{(s)} &= T^{-1} \left(\varrho^2 L_{(M)\alpha\beta}^{(0,0)} \frac{dm_\beta}{dt} \frac{dm_\alpha}{dt} - 2\varrho \frac{1}{T} L_{(M)\alpha\beta}^{(0,q)} \frac{\partial T}{\partial x_\beta} \frac{dm_\alpha}{dt} \right. \\
&+ 2\varrho \sum_{k=1}^{n-1} L_{(MD)\alpha\beta}^{(0,k)} X_\beta^{(k)} \frac{dm_\alpha}{dt} + 2\varrho \sum_{h=1}^r L_{(MC)\alpha}^{(0,h)} A^{(h)} \frac{dm_\alpha}{dt} + L_{(M)\alpha\beta}^{(1,1)} B_\beta^{(1)} B_\alpha^{(1)} \\
&\left. + 2L_{(M)\alpha\beta\gamma}^{(1,vi)} \frac{d\varepsilon_{\beta\gamma}}{dt} B_\alpha^{(1)} - \frac{2}{T} \sum_{k=1}^{n-1} L_{(D)\alpha\beta}^{(q,k)} X_\beta^{(k)} \frac{\partial T}{\partial x_\alpha} - \frac{2}{T} \sum_{h=1}^r L_{(C)\alpha}^{(q,h)} A^{(h)} \frac{\partial T}{\partial x_\alpha} \right)
\end{aligned}$$

$$\begin{aligned}
& + \frac{1}{T^2} L_{\alpha\beta}^{(q,q)} \frac{\partial T}{\partial x_\alpha} \frac{\partial T}{\partial x_\beta} + 2 \sum_{j=1}^{n-1} \sum_{h=1}^r L_{(DC)\alpha}^{(j,h)} A^{(h)} X_\alpha^{(j)} + \sum_{j,k=1}^{n-1} L_{(DD)\alpha\beta}^{(j,k)} X_\beta^{(k)} X_\alpha^{(j)} \\
& + \sum_{l,h=1}^r L_{(CC)}^{(l,h)} A^{(h)} A^{(l)} + L_{\alpha\beta\gamma\delta}^{(vi,vi)} \frac{d\varepsilon_{\gamma\delta}}{dt} \frac{d\varepsilon_{\alpha\beta}}{dt} \Bigg). \quad (34)
\end{aligned}$$

The entropy production is a positive definite quadratic form, i.e.

$$\sigma^{(s)} \geq 0. \quad (35)$$

From the positive definite character of the entropy production several inequalities may be derived for the components of the phenomenological coefficients, resulting from the fact that all the elements of the main diagonal of the matrix associated to the quadratic form (34) must be non-negative and all principal minors of this matrix must be non-negative. For instance, we have

$$L_{(M)\alpha\alpha}^{(0,0)} \geq 0, \quad L_{(M)\alpha\alpha}^{(1,1)} \geq 0, \quad L_{\alpha\alpha}^{(q,q)} \geq 0, \quad L_{\alpha\alpha}^{(k,k)} \geq 0 \quad (k = 1, 2, \dots, n-1), \quad L_{\alpha\beta\alpha\beta}^{(vi,vi)} \geq 0. \quad (36)$$

Also, from the fifth of the inequalities we obtain by virtue of symmetry and Onsager-Casimir relations

$$L_{\alpha\beta\beta\alpha}^{(vi,vi)} \geq 0, \quad L_{\beta\alpha\alpha\beta}^{(vi,vi)} \geq 0, \quad L_{\beta\alpha\beta\alpha}^{(vi,vi)} \geq 0. \quad (37)$$

4 Phenomenological equations and entropy production for systems, which are isotropic (with respect to all the rotations of the frame of axes)

In this Section, taking into account the model presented in Sections 2 we work out the phenomenological equations and the entropy production for magnetizable reacting fluid mixtures, supposed isotropic (having symmetry properties such that under orthogonal transformations are invariant) with respect to all rotations of the frame of axes. The existence of spatial symmetry properties in a material system may simplify the form of the phenomenological equations in such way that the Cartesian components of the fluxes do not depend on all Cartesian components of the thermodynamic forces. This statement is called Curie symmetry principle [2], [12].

Let us observe that in the phenomenological equations, $\varrho \frac{d\mathbf{m}}{dt}$, $\varrho \frac{d\mathbf{m}^{(1)}}{dt}$, $\mathbf{B}^{(ir)}$ and $\mathbf{B}^{(1)}$ are *pseudo vectors* (or *axial vectors*) while $T^{-1} \text{grad} T$, $\mathbf{X}^{(k)}$

$(k = 1, 2, \dots, n-1)$ and $\tau_{\alpha\beta}^{(vi)} \mathbf{J}^{(q)}, \mathbf{J}_{(diff)}^{(k)} (k = 1, 2, \dots, n-1), \frac{d\varepsilon_{\alpha\beta}}{dt}$ are polar tensors and $J^{(l)}, A^{(l)} (l = 1, 2, \dots, r)$ are scalar quantities.

From the phenomenological equations (25)-(30) we see that the phenomenological coefficients

$$\begin{aligned} &L_{(M)\alpha\beta}^{(0,q)}, L_{(MD)\alpha\beta}^{(0,k)}, L_{(MC)\alpha}^{(0,h)}, L_{(M)\alpha\beta\gamma}^{(0,vi)} (k = 1, 2, \dots, n-1), (h = 1, 2, \dots, r), \\ &L_{(M)\alpha\beta}^{(1,q)}, L_{(MD)\alpha\beta}^{(1,k)}, L_{(MC)\alpha}^{(1,h)}, L_{(M)\alpha\beta\gamma}^{(1,vi)} (k = 1, 2, \dots, n-1), (h = 1, 2, \dots, r), \\ &L_{(M)\alpha\beta}^{(q,0)}, L_{(M)\alpha\beta}^{(q,1)}, L_{(DM)\alpha\beta}^{(j,0)}, L_{(DM)\alpha\beta}^{(j,1)} (k = 1, 2, \dots, n-1), \\ &L_{(CM)\beta}^{(l,0)}, L_{(CM)\beta}^{(l,1)}, L_{(M)\alpha\beta\gamma}^{(vi,0)}, L_{(M)\alpha\beta\gamma}^{(vi,1)} (l = 1, 2, \dots, r) \end{aligned}$$

are pseudo tensors of different order. For instance $L_{(M)\alpha\beta\gamma}^{(1,vi)}$ is a pseudo tensor of third order connected with the influence of the viscous flow on the magnetic relaxation; $L_{(M)\alpha\beta}^{(1,q)}$ is a pseudo tensor of second order connected with the influence of heat flux on magnetic relaxation, $L_{(MC)\beta}^{(0,l)} (l = 1, 2, \dots, r)$ are pseudo tensors of first order connected with the influence the chemical reaction rate of the l -th reaction on the magnetic relaxation; $L_{(M)\alpha\beta\gamma}^{(0,vi)}$ and $L_{(M)\alpha\beta\gamma}^{(1,vi)}$ are pseudo tensors of third order, connected with the influence of the viscous flow on the magnetic relaxation.

Also, from (25)-(30) we see that the phenomenological coefficients

$$\begin{aligned} &L_{(M)\alpha\beta}^{(0,0)}, L_{(M)\alpha\beta}^{(0,1)}, L_{(M)\alpha\beta}^{(1,0)}, L_{(M)\alpha\beta}^{(1,1)}, L_{\alpha\beta}^{(q,q)}, L_{(D)\alpha\beta}^{(q,k)} (k = 1, 2, \dots, n-1), \\ &L_{(C)\alpha}^{(q,h)}, L_{\alpha\beta\gamma}^{(q,vi)}, L_{(D)\alpha\beta}^{(j,q)}, L_{(DD)\alpha\beta}^{(j,k)}, L_{(DC)\alpha}^{(j,h)} (j, k = 1, \dots, n-1), (h = 1, \dots, r), \\ &L_{(M)\alpha\beta\gamma}^{(j,vi)}, L_{(C)\alpha}^{(l,q)}, L_{(CD)\alpha}^{(l,k)}, L_{(CC)\alpha}^{(l,h)} (j, k = 1, 2, \dots, n-1), (l, h = 1, 2, \dots, r) \\ &L_{(C)\alpha\beta\gamma}^{(l,vi)}, L_{\alpha\beta\gamma}^{(vi,q)}, L_{\alpha\beta\gamma}^{(vi,k)}, L_{\alpha\beta\gamma}^{(vi,h)}, L_{\alpha\beta\gamma}^{(vi,vi)} (l, h = 1, 2, \dots, r) \end{aligned}$$

are polar tensors of different order. For instance, $L_{\alpha\beta}^{(q,q)}$ is the heat conductivity polar tensor of order two; $L_{(DD)\alpha\beta}^{(j,k)} (j, k = 1, 2, \dots, n-1)$, are polar tensors of order two connected with the diffusion flux of the k -th fluid component; $L_{(CC)}^{(l,h)}$ are scalars connected with the chemical affinity of the reaction $h (l, h = 1, 2, \dots, r)$; $L_{\alpha\beta\gamma}^{(vi,q)}$, is a polar tensor of order three connected with the influence of the heat flux on the stress tensor, $L_{\alpha\beta\gamma\delta}^{(vi,vi)}$ is the viscosity polar tensor of order four; $L_{(MM)\alpha\beta}^{(0,0)}$ and $L_{(MM)\alpha\beta}^{(0,1)}$ are polar tensors of second order connected with the magnetic relaxation,

In this isotropic case (see [6], and [5] for a more in-depth study in the case of isotropic porous media) we have that the *tensors of order four* $L_{\alpha\beta\zeta\gamma}$ must have the form

$$L_{\alpha\beta\zeta\gamma} = L_1\delta_{\alpha\beta}\delta_{\zeta\gamma} + L_2\delta_{\alpha\zeta}\delta_{\beta\gamma} + L_3\delta_{\alpha\gamma}\delta_{\beta\zeta}, \quad (38)$$

with L_i ($i = 1, 2, 3$) scalars.

In the case when (see (33))

$$L_{\alpha\beta\zeta\gamma} = L_{\beta\alpha\zeta\gamma} \quad (39)$$

we have

$$L_{\beta\alpha\zeta\gamma} = L_1\delta_{\beta\alpha}\delta_{\zeta\gamma} + L_2\delta_{\beta\zeta}\delta_{\alpha\gamma} + L_3\delta_{\beta\gamma}\delta_{\alpha\zeta}. \quad (40)$$

Adding equations (38) and (40), using (39) and multiplying by 1/2, we obtain

$$L_{\alpha\beta\zeta\gamma} = A_1\delta_{\alpha\beta}\delta_{\zeta\gamma} + A_2(\delta_{\alpha\zeta}\delta_{\beta\gamma} + \delta_{\alpha\gamma}\delta_{\beta\zeta}), \quad (41)$$

where

$$A_1 = L_1, \quad A_2 = (L_2 + L_3)/2 \quad (42)$$

are scalars.

The *polar or pseudo tensors of order three* must have the form

$$L_{\alpha\beta\gamma} = L\epsilon_{\alpha\beta\gamma}, \quad (43)$$

where $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita symbol and L is a scalar but, because of the symmetry properties of $L_{\alpha\beta\gamma}$, one has in our case

$$L_{\alpha\beta\gamma} = 0. \quad (44)$$

The *polar or pseudo tensors of order two* $L_{\alpha\beta}$ must have the form

$$L_{\alpha\beta} = L\delta_{\alpha\beta}, \quad (45)$$

where L is a scalar. If $L_{\alpha\beta}$ has zero trace, it is null, i.e. $L_{\alpha\beta} = 0$.

The polar and pseudo vectors L_α vanish, i.e.

$$L_\alpha = 0. \quad (46)$$

Let us introduce the deviator $\tilde{A}_{\alpha\beta}$ and the scalar part A of an arbitrary tensor field of order two $A_{\alpha\beta}$ defined by

$$\tilde{A}_{\alpha\beta} = A_{\alpha\beta} - \frac{1}{3}A_{\gamma\gamma}\delta_{\alpha\beta} \quad \text{and} \quad A = \frac{1}{3}A_{\gamma\gamma}. \quad (47)$$

Hence,

$$A_{\alpha\beta} = \tilde{A}_{\alpha\beta} + A\delta_{\alpha\beta} \quad \text{and} \quad \tilde{A}_{\gamma\gamma} = 0. \quad (48)$$

Moreover, if $A_{\alpha\beta}$ is symmetric also $\tilde{A}_{\alpha\beta}$ is symmetric.

Using the symmetry relations (31) and the above properties of isotropy, the phenomenological equations (25)-(30) take the following form

$$B_{\alpha}^{(ir)} = \varrho L_{(M)}^{(0,0)} \frac{dm_{\alpha}}{dt} + L_{(M)}^{(0,1)} B_{\alpha}^{(1)} - \frac{1}{T} L_{(M)}^{(0,q)} \frac{\partial T}{\partial x_{\alpha}} + \sum_{k=1}^{n-1} L_{(MD)}^{(0,k)} X_{\alpha}^{(k)}, \quad (49)$$

$$\varrho \frac{dm_{\alpha}^{(1)}}{dt} = \varrho L_{(M)}^{(1,0)} \frac{dm_{\alpha}}{dt} + L_{(M)}^{(1,1)} B_{\alpha}^{(1)} - \frac{1}{T} L_{(M)}^{(1,q)} \frac{\partial T}{\partial x_{\alpha}} + \sum_{k=1}^{n-1} L_{(MD)}^{(1,k)} X_{\alpha}^{(k)}, \quad (50)$$

$$J_{\alpha}^{(q)} = \varrho L_{(M)}^{(q,0)} \frac{dm_{\alpha}}{dt} + L_{(M)}^{(q,1)} B_{\alpha}^{(1)} - \frac{1}{T} L_{(M)}^{(q,q)} \frac{\partial T}{\partial x_{\alpha}} + \sum_{k=1}^{n-1} L_{(D)}^{(q,k)} X_{\alpha}^{(k)}, \quad (51)$$

$$J_{(diff)\alpha}^{(j)} = \varrho L_{(DM)}^{(j,0)} \frac{dm_{\alpha}}{dt} + L_{(DM)}^{(j,1)} B_{\alpha}^{(1)} - \frac{1}{T} L_{(D)}^{(j,q)} \frac{\partial T}{\partial x_{\alpha}} + \sum_{k=1}^{n-1} L_{(DD)}^{(j,k)} X_{\alpha}^{(k)}. \quad (52)$$

$$J_{(chem)}^{(l)} = \sum_{h=1}^r L_{(CC)}^{(l,h)} A^{(h)} + 3L_{(C)}^{(l,vi)} \frac{d\varepsilon}{dt} \quad (l = 1, 2, \dots, r), \quad (53)$$

$$\tau_{\alpha\beta} = \sum_{h=1}^r L_{(C)}^{(vi,h)} A^{(h)} \delta_{\alpha\beta} + \eta_s \frac{d\tilde{\epsilon}_{\alpha\beta}}{dt} + \eta_v \frac{d\epsilon}{dt} \delta_{\alpha\beta}. \quad (54)$$

In the Appendix we demonstrate the phenomenological equation (54) for $\tau_{\alpha\beta}$.

From (24) and the phenomenological equations (49)-(54), using the Onsager-Casimir relations in the isotropic case and the relations (41), (43)-(46) (or equivalently from (24) calculated in the isotropic case), we obtain the entropy production $\sigma^{(s)}$ having the following form

$$\begin{aligned} \sigma^{(s)} = T^{-1} & \left[\varrho^2 L_{(M)}^{(0,0)} \left(\frac{d\mathbf{m}}{dt} \right)^2 + L_{(M)}^{(1,1)} \left(\mathbf{B}^{(1)} \right)^2 - 2\varrho \frac{1}{T} L_{(M)}^{(0,q)} (\nabla T) \cdot \frac{d\mathbf{m}}{dt} \right. \\ & \left. + 2\varrho \sum_{k=1}^{n-1} L_{(MD)}^{(0,k)} \mathbf{X}^{(k)} \cdot \frac{d\mathbf{m}}{dt} - \frac{2}{T} \sum_{k=1}^{n-1} L_{(D)}^{(q,k)} \mathbf{X}^{(k)} \cdot \nabla T + \frac{1}{T^2} L_{(q,q)} (\nabla T)^2 \right] \end{aligned}$$

$$+ \sum_{j,k=1}^{n-1} L_{(DD)}^{(j,k)} \mathbf{X}^{(k)} \cdot \mathbf{X}^{(j)} + \sum_{l,h=1}^r L_{(CC)}^{(l,h)} A^{(h)} A^{(l)} + \eta_s \left(\frac{d\tilde{\epsilon}_{\alpha\beta}}{dt} \right)^2 + 3\eta_v \left(\frac{d\epsilon}{dt} \right)^2 \geq 0. \quad (55)$$

From (55) it is seen that the entropy production is a bilinear form in the components of the time derivative of the total specific magnetization vector, the components of the thermodynamic force conjugate to the partial specific magnetization vector, the components of the thermodynamic forces conjugate to the diffusion fluxes of matter, the chemical affinities, the components of the temperature gradient, the components of the time derivative of the deviator of the strain tensor and the time derivative of the scalar part of this tensor. The entropy production is a positive definite quadratic form, thus several inequalities may be derived for the components of the phenomenological coefficients, resulting from the fact that all the elements of the main diagonal of the matrix associated to the quadratic form (55) must be non-negative and all principal minors of this matrix must be non-negative (see [3], [4] and [40], as examples in the case of three-dimensional isotropic and anisotropic rigid media and isotropic magnetizable media).

For instance, we have

$$L_{(M)}^{(0,0)} \geq 0, L_{(M)}^{(1,1)} \geq 0, L^{(q,q)} \geq 0, L^{(k,k)} \geq 0 \quad (k = 1, 2, \dots, n-1), \eta_s \geq 0, \eta_v \geq 0. \quad (56)$$

5 Phenomenological equations and entropy production for perfect isotropic systems

In this Section, taking into consideration the model presented in Sections 2, we work out the phenomenological equations and the entropy production for magnetizable reacting fluid mixtures, supposed perfect isotropic, i.e. having symmetry properties such that under orthogonal transformations are invariant with respect to all rotations and inversions of the frame axes, The existence of these spatial symmetry properties simplifies the form of the phenomenological equations and of the entropy production and for example it can be shown that fluxes and thermodynamic forces of different tensorial character (polar or pseudo) do not couple (*Principle of Curie symmetry*), see [2], [12].

In the perfect isotropic case (see [6], and [5] for a more in-depth study, where isotropic and perfect isotropic porous media are analyzed) we have that:

the *tensors of order four* $L_{\alpha\beta\zeta\gamma}$ must have the form (41).
The *polar tensors of order three* vanish, i.e.

$$L_{\alpha\beta\gamma} = 0. \quad (57)$$

The *pseudo tensors of order three* keep the form

$$L_{\alpha\beta\gamma} = L\epsilon_{\alpha\beta\gamma}, \quad (58)$$

but, because of the symmetry properties of $L_{\alpha\beta\gamma}$, they are null

$$L_{\alpha\beta\gamma} = 0. \quad (59)$$

The *polar tensors of order two* $L_{\alpha\beta}$ take the form

$$L_{\alpha\beta} = L\delta_{\alpha\beta}, \quad (60)$$

which reduces to

$$L_{\alpha\beta} = 0, \quad (61)$$

if $L_{\alpha\beta}$ has zero trace.

The *pseudo tensors of order two* vanish, i.e.

$$L_{\alpha\beta} = 0. \quad (62)$$

The *polar and pseudo vectors* L_α vanish, i.e.

$$L_\alpha = 0. \quad (63)$$

Then, in the perfect isotropic case taking into account of (41), (57)-(63) we obtain

$$B_\alpha^{(ir)} = \varrho L_{(M)}^{(0,0)} \frac{dm_\alpha}{dt} + L_{(M)}^{(0,1)} B_\alpha^{(1)}, \quad (64)$$

$$\varrho \frac{dm_\alpha^{(1)}}{dt} = \varrho L_{(M)}^{(1,0)} \frac{dm_\alpha}{dt} + L_{(M)}^{(1,1)} B_\alpha^{(1)}, \quad (65)$$

$$J_\alpha^{(q)} = -\frac{1}{T} L_{(D)}^{(q,q)} \frac{\partial T}{\partial x_\alpha} + \sum_{k=1}^{n-1} L_{(D)}^{(q,k)} X_\alpha^{(k)} \quad (66)$$

$$J_{(diff)\alpha}^{(j)} = -\frac{1}{T} L_{(D)}^{(j,q)} \frac{\partial T}{\partial x_\alpha} + \sum_{k=1}^{n-1} L_{(DD)}^{(j,k)} X_\alpha^{(k)} \quad (j = 1, 2, \dots, n-1), \quad (67)$$

$$J_{(chem)}^{(l)} = \sum_{h=1}^r L_{(CC)}^{(l,h)} A^{(h)} + 3L_{(C)}^{(l,vi)} \frac{d\varepsilon}{dt} \quad (l = 1, 2, \dots, r), \quad (68)$$

$$\tau_{\alpha\beta}^{(vi)} = \sum_{h=1}^r L_{(C)}^{(vi,h)} A^{(h)} \delta_{\alpha\beta} + \eta_v \frac{d\epsilon}{dt} \delta_{\alpha\beta} + \eta_s \frac{d\tilde{\epsilon}_{\alpha\beta}}{dt}. \quad (69)$$

From (24) and the phenomenological equations (64)-(69), using the Onsager-Casimir relations in the perfect isotropic case and the relations (41), (57)-(63) (or equivalently from (24) calculated in the perfect isotropic case) we derive the entropy production $\sigma^{(s)}$ having the following form

$$\begin{aligned} \sigma^{(s)} = T^{-1} & \left[\varrho^2 L_{(M)}^{(0,0)} \left(\frac{d\mathbf{m}}{dt} \right)^2 + L_{(M)}^{(1,1)} \left(\mathbf{B}^{(1)} \right)^2 - \frac{2}{T} \sum_{k=1}^{n-1} L_{(D)}^{(q,k)} \mathbf{X}^{(k)} \cdot \nabla T \right. \\ & + \sum_{j,k=1}^{n-1} L_{(DD)}^{(j,k)} \mathbf{X}^{(k)} \cdot \mathbf{X}^{(j)} + \sum_{l,h=1}^r L_{(CC)}^{(l,h)} A^{(h)} A^{(l)} + \frac{1}{T^2} L^{(q,q)} (\nabla T)^2 \\ & \left. + \eta_s \left(\frac{d\tilde{\epsilon}_{\alpha\beta}}{dt} \right)^2 + 3\eta_v \left(\frac{d\epsilon}{dt} \right)^2 \right] \geq 0. \quad (70) \end{aligned}$$

From (70) it is seen that there are no contributions due to the cross effects among magnetic relaxation phenomena and the other irreversible phenomena, because fluxes and thermodynamic forces of different tensorial character (polar or axial) do not couple. In (70) the terms with $\left(\frac{d\mathbf{m}}{dt} \right)^2$ and $\left(\mathbf{B}^{(1)} \right)^2$ are connected with entropy production due to magnetic relaxation, the terms with $(\nabla T)^2$, $\mathbf{X}^{(k)} \cdot \mathbf{X}^{(j)}$ ($k, j = 1, 2, \dots, n-1$), $A^{(h)} A^{(l)}$ ($l, h = 1, 2, \dots, n-1$), $\left(\frac{d\tilde{\epsilon}_{\alpha\beta}}{dt} \right)^2$, $\left(\frac{d\epsilon}{dt} \right)^2$ are due to the temperature gradient, the diffusion fluxes of matter, the chemical affinities and the viscous flow, respectively, The terms $\mathbf{X}^{(k)} \cdot \nabla T$ arise from the cross effects among the diffusion of mass and the temperature gradient. It is seen from (70) that because of the non-negative character of the entropy production the phenomenological coefficients satisfy inequalities as (56).

6 Linear equations of state for anisotropic fluid systems

In this Section we recall some results obtained in the paper [38], while providing some insight. In order to obtain the linear equations of state for

anisotropic reacting fluid mixtures with magnetic relaxation we use as thermodynamic potential, the specific free energy f defined by

$$f = u - Ts. \quad (71)$$

Using the Gibbs relation (15), we obtain the following expression for the differential of f ,

$$df = -sdT + v\tau_{\alpha\beta}^{(eq)} d\varepsilon_{\alpha\beta} + \mathbf{B}^{(eq)} \cdot d\mathbf{m} - \mathbf{B}^{(1)} \cdot d\mathbf{m}^{(1)} + \sum_{k=1}^n \mu^{(k)} dc^{(k)}. \quad (72)$$

Therefore, the following definitions are valid

$$s = -\frac{\partial}{\partial T} f \left(T, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)} \right), \quad (73)$$

$$\tau_{\alpha\beta}^{(eq)} = \varrho \frac{\partial}{\partial \varepsilon_{\alpha\beta}} f \left(T, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)} \right), \quad (74)$$

$$\mathbf{B}^{(eq)} = \frac{\partial}{\partial \mathbf{m}} f \left(T, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)} \right), \quad (75)$$

$$\mathbf{B}^{(1)} = -\frac{\partial}{\partial \mathbf{m}^{(1)}} f \left(T, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)} \right), \quad (76)$$

$$\mu^{(k)} = \frac{\partial}{\partial c^{(k)}} f \left(T, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)} \right) \quad (k = 1, 2, \dots, n). \quad (77)$$

We consider a reference state of the medium (indicated by the symbol " $_{(0)}$ ") and we also require that this reference state is a state of thermodynamic equilibrium. We assume that in this state there is an uniform temperature, that has an arbitrary (but fixed) value $T_{(0)}$, the concentrations $c^{(k)}$ ($k = 1, 2, \dots, n$) of the n fluid components have fixed values $c_{(0)}^{(k)}$ and the values of the strain tensor $\varepsilon_{(0)\alpha\beta}$, of $\mathbf{m}_{(0)}$ and $\mathbf{m}_{(0)}^{(1)}$ are null. Then, we assume that the mechanical stress tensor $\tau_{\alpha\beta}$, the magnetic field $\mathbf{B}$ and the fields $\mathbf{B}^{(1)}$ and $\mu^{(k)}$ vanish in this state, i.e. we assume that

$$\tau_{\alpha\beta}^{(eq)} \left(T_{(0)}, \varepsilon_{(0)\alpha\beta}, \mathbf{m}_{(0)}, \mathbf{m}_{(0)}^{(1)}, c_{(0)}^{(1)}, \dots, c_{(0)}^{(n)} \right) = 0, \quad (78)$$

$$\mathbf{B}^{(eq)} \left(T_{(0)}, \varepsilon_{(0)\alpha\beta}, \mathbf{m}_{(0)}, \mathbf{m}_{(0)}^{(1)}, c_{(0)}^{(1)}, \dots, c_{(0)}^{(n)} \right) = \mathbf{0}, \quad (79)$$

$$\mathbf{B}^{(1)} \left(T_{(0)}, \varepsilon_{(0)\alpha\beta}, \mathbf{m}_{(0)}, \mathbf{m}_{(0)}^{(1)}, c_{(0)}^{(1)}, \dots, c_{(0)}^{(n)} \right) = \mathbf{0}, \quad (80)$$

$$\mu^{(k)} \left(T_{(0)}, \varepsilon_{(0)\alpha\beta}, \mathbf{m}_{(0)}, \mathbf{m}_{(0)}^{(1)}, c_{(0)}^{(1)}, \dots, c_{(0)}^{(n)} \right) = 0 \quad (k = 1, \dots, n), \quad (81)$$

if

$$T = T_{(0)}, \quad c^{(k)} = c_{(0)}^{(k)} \quad (k = 1, 2, \dots, n), \quad \varepsilon_{(0)\alpha\beta} = 0, \quad \mathbf{m}_{(0)} = \mathbf{0}, \quad \mathbf{m}_{(0)}^{(1)} = \mathbf{0}.$$

Let us expand the free energy f into Taylor's series with respect to the considered reference state and we consider very small deviations with respect to this state. We postulate the following form for the specific free energy f of an anisotropic reacting fluid mixtures

$$f = f^{(1)} + f^{(2)}, \quad (82)$$

where

$$\begin{aligned} f^{(1)} = v_{(0)} & \left[\frac{1}{2} a_{\alpha\beta\gamma\zeta} \varepsilon_{\alpha\beta} \varepsilon_{\gamma\zeta} + a_{\alpha\beta} \varepsilon_{\alpha\beta} (T - T_{(0)}) \right. \\ & + \sum_{k=1}^n b_{(cT)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) (T - T_0) + \sum_{k=1}^n b_{(c\varepsilon)\alpha\beta}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) \varepsilon_{\alpha\beta} \\ & \left. + \frac{1}{2} \sum_{i,k=1}^n b_{(cc)}^{(i,k)} \left(c^{(i)} - c_{(0)}^{(i)} \right) \left(c^{(k)} - c_{(0)}^{(k)} \right) \right] - \varphi(T) \end{aligned} \quad (83)$$

and

$$\begin{aligned} f^{(2)} = \frac{1}{2} \varrho_{(0)} & \left[a_{(M)\alpha\beta}^{(0,0)} m_{\alpha} \left(m_{\beta} - 2m_{\beta}^{(1)} \right) + a_{(M)\alpha\beta}^{(1,1)} m_{\alpha}^{(1)} m_{\beta}^{(1)} \right] \\ & + \left(a_{(M)\alpha}^{(0)} m_{\alpha} - a_{(M)\alpha}^{(1)} m_{\alpha}^{(1)} \right) (T - T_{(0)}) \\ & + \sum_{k=1}^n \left(b_{(M)\alpha}^{(0,k)} m_{\alpha} + b_{(M)\alpha}^{(1,k)} m_{\alpha}^{(1)} \right) \left(c^{(k)} - c_{(0)}^{(k)} \right). \end{aligned} \quad (84)$$

In (83) $\varphi(T)$ is some function of the temperature, $v_{(0)}$ is the specific volume in the reference state, defined by $v_{(0)} = \frac{1}{\varrho_{(0)}}$, that in the following will be indicated by $v = \frac{1}{\varrho}$ and supposed constant. Furthermore, in

(83)-(84) the scalar material coefficients $b_{(cT)}^{(k)}, b_{(cc)}^{(k,i)}$ ($i, k = 1, 2, \dots, n$) and the vector material coefficients $a_{(M)\alpha}^{(0)}, a_{(M)\alpha}^{(1)}, b_{(M)\alpha}^{(0,k)}, b_{(M)\alpha}^{(1,k)}$ ($k = 1, 2, \dots, n$) are constant, the material coefficients tensor of order four and order two $a_{\alpha\beta\gamma\zeta}, a_{\alpha\beta}, b_{(c\varepsilon)\alpha\beta}^{(k)}, a_{(M)\alpha\beta}^{(0,0)}, a_{(M)\alpha\beta}^{(1,1)}$ are constant and satisfy the following symmetry relations

$$a_{\alpha\beta\gamma\zeta} = a_{\beta\alpha\gamma\zeta} = a_{\alpha\beta\zeta\gamma} = a_{\beta\alpha\zeta\gamma} = a_{\gamma\zeta\alpha\beta} = a_{\gamma\zeta\beta\alpha} = a_{\zeta\gamma\alpha\beta} = a_{\zeta\gamma\beta\alpha}, \quad (85)$$

$$a_{\alpha\beta} = a_{\beta\alpha}, \quad b_{\alpha\beta}^{(k)}, b_{(c\varepsilon)\alpha\beta}^{(k)} = b_{(c\varepsilon)\beta\alpha}^{(k)}, \quad a_{(M)\alpha\beta}^{(0,0)} = a_{(M)\beta\alpha}^{(0,0)}, \quad a_{(M)\alpha\beta}^{(1,1)} = a_{(M)\beta\alpha}^{(1,1)}. \quad (86)$$

Also, we have

$$b_{(cc)}^{(i,k)} = b_{(cc)}^{(k,i)} \quad (i, k = 1, 2, \dots, n). \quad (87)$$

All these constants are determined by the physical properties of the medium in the reference state. As usually in thermodynamics, in (83) and in (84) we have named the second partial derivatives of free energy with respect to the considered independent variables using the names of the material coefficients, measurable by experiments, supposed constant and coming from their physical interpretation. These coefficients have well-known physical meanings. For instance,

$$\begin{aligned} \frac{\partial^2 f}{\partial T \partial \varepsilon_{\alpha\beta}} &= \rho^{-1} a_{\alpha\beta}, & \frac{\partial^2 f}{\partial T \partial m_{\alpha}} &= a_{(M)\alpha}^{(0)}, \\ \frac{\partial^2 f}{\partial \varepsilon_{\alpha\beta} \partial \varepsilon_{\gamma\delta}} &= \rho^{-1} a_{\alpha\beta\gamma\delta}, & \frac{\partial^2 f}{\partial m_{\alpha} \partial m_{\beta}} &= \rho a_{(M)\alpha\beta}^{(0,0)}. \end{aligned} \quad (88)$$

Furthermore, the physical dimensions of the present physical quantities have been taken into account, the introduction of the minus sign comes from physical reasons and the symmetry relations (85), (86) and (87), satisfied by some phenomenological coefficients, are coming by the invariance properties of the derivation of free energy with respect to the independent variables (taking into account also the symmetry of the small strain tensor $\varepsilon_{\alpha\beta}$). The symmetry relations reduces the number of independent components of these phenomenological tensors. From (74)-(77) and (82)-(84) we have the following equations of state for the equilibrium stress tensor $\tau_{\alpha\beta}^{(eq)}$, the equilibrium magnetic field $\mathbf{B}^{(eq)}$, and the fields $\mathbf{B}^{(1)}$ and $\mu^{(k)}$

$$\tau_{\alpha\beta}^{(eq)} = a_{\alpha\beta\gamma\zeta} \varepsilon_{\gamma\zeta} + a_{\alpha\beta} (T - T_{(0)}) + \sum_{k=1}^n b_{(c\varepsilon)\alpha\beta}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right), \quad (89)$$

$$B_{\alpha}^{(eq)} = a_{(M)\alpha\beta}^{(0,0)} \left(M_{\beta} - M_{\beta}^{(1)} \right) + a_{(M)\alpha}^{(0)} (T - T_{(0)}) + \sum_{k=1}^n b_{(M)\alpha}^{(0,k)} \left(c^{(k)} - c_{(0)}^{(k)} \right), \quad (90)$$

$$B_{\alpha}^{(1)} = a_{(M)\alpha\beta}^{(0,0)} M_{\beta} - a_{(M)\alpha\beta}^{(1,1)} M_{\beta}^{(1)} + a_{(M)\alpha}^{(1)} (T - T_{(0)}) + \sum_{k=1}^n b_{(M)\alpha}^{(1,k)} \left(c^{(k)} - c_{(0)}^{(k)} \right), \quad (91)$$

$$\begin{aligned} \mu^{(k)} = v & \left[b^{(k)}(cT)(T - T_0) + b_{(c\varepsilon)\alpha\beta}^{(k)} \varepsilon_{\alpha\beta} + \sum_{i=1}^n b^{(i,k)}(cc) \left(c^{(i)} - c_{(0)}^{(i)} \right) \right. \\ & \left. + b_{(M)\alpha}^{(0,k)} M_{\alpha} - b_{(M)\alpha}^{(1,k)} M_{\alpha}^{(1)} \right] \quad (k = 1, 2, \dots, n), \end{aligned} \quad (92)$$

where, we have defined the field $\mathbf{M}^{(1)}$ as

$$\mathbf{M}^{(1)} = \varrho \mathbf{m}^{(1)}. \quad (93)$$

7 Linear equations of state and constitutive equations for perfect isotropic systems

In this Section we derive the linear equations of state and the constitutive equations for perfect isotropic magnetizable reacting fluid mixtures, using the model developed in [38]. From (82), (83) and (84), using relations (41) and (57)-(63), we have the following form for the free energy

$$f = f^{(1)} + f^{(2)}, \quad (94)$$

where

$$\begin{aligned} f^{(1)} = v & \left[\frac{1}{2} \left(a^{(1)} \tilde{\varepsilon}_{\alpha\beta} + a^{(2)} \varepsilon \delta_{\alpha\beta} \right) \varepsilon_{\alpha\beta} + 3a^{(3)} \varepsilon (T - T_{(0)}) \right. \\ & + \sum_{k=1}^n b_{(cT)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) (T - T_0) + \sum_{k=1}^n b_{(c\varepsilon)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) \varepsilon_{\alpha\alpha} \\ & \left. + \frac{1}{2} \sum_{i,k=1}^n b_{(cc)}^{(i,k)} \left(c^{(i)} - c_{(0)}^{(i)} \right) \left(c^{(k)} - c_{(0)}^{(k)} \right) \right] - \varphi(T) \end{aligned} \quad (95)$$

and

$$f^{(2)} = \frac{1}{2} \varrho \left[a_{(M)}^{(0,0)} (\mathbf{m})^2 - 2a_{(M)}^{(0,0)} \mathbf{m} \cdot \mathbf{m}^{(1)} + a_{(M)}^{(1,1)} (\mathbf{m}^{(1)})^2 \right], \quad (96)$$

where $\varphi(T)$ as in (95) is some function of the temperature and v is the specific volume in the reference state, defined by $v = \frac{1}{\varrho}$, with ϱ constant. Furthermore, the material constants have taken their corresponding form valid in the case of perfect isotropic systems, keeping the same name, except the phenomenological constants $a_{\alpha\beta}$, which have been replaced by $a^{(3)}$, and the constants $a_{\alpha\beta\gamma\zeta}$, which have been replaced by the two constants $a^{(1)}$ and $a^{(2)}$ (see relation (41)). From (74)-(77) and (94)-(96), or equivalently from (89)-(92) considered in the perfect isotropic case, we derive the following equations of state for the equilibrium stress tensor $\tau_{\alpha\beta}^{(eq)}$, the equilibrium magnetic field $B^{(eq)}$ and the fields $B^{(1)}$ and $\mu^{(k)}$

$$\tau_{\alpha\beta}^{(eq)} = a^{(1)} \tilde{\varepsilon}_{\alpha\beta} + a^{(2)} \varepsilon + a^{(3)} (T - T_{(0)}) + \sum_{k=1}^n b_{(c\varepsilon)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) \delta_{\alpha\beta}. \quad (97)$$

$$B_{\alpha}^{(eq)} = a_{(M)}^{(0,0)} \left(M_{\alpha} - M_{\alpha}^{(1)} \right), \quad (98)$$

$$B_{\alpha}^{(1)} = a_{(M)}^{(0,0)} M_{\alpha} - a_{(M)}^{(1,1)} M_{\alpha}^{(1)}, \quad (99)$$

$$\mu^{(k)} = v \left[b_{(cT)}^{(k)} (T - T_0) + 3b_{(c\varepsilon)}^{(k)} \varepsilon + \sum_{i=1}^n b_{(cc)}^{(i,k)} \left(c^{(i)} - c_{(0)}^{(i)} \right) \right]. \quad (100)$$

Thus, we obtain the following constitutive equations for B_{α} , $\frac{dM_{\alpha}^{(1)}}{dt}$ and $\tau_{\alpha\beta}$ (see equations (64), (65) and (69))

$$B_{\alpha} = B_{\alpha}^{(eq)} + L_{(M)}^{(0,0)} \frac{dM_{\alpha}}{dt} + L_{(M)}^{(0,1)} B_{\alpha}^{(1)}, \quad (101)$$

$$\frac{dM_{\alpha}^{(1)}}{dt} = L_{(M)}^{(1,0)} \frac{dM_{\alpha}}{dt} + L_{(M)}^{(1,1)} B_{\alpha}^{(1)}, \quad (102)$$

$$\begin{aligned} \tau_{\alpha\beta} &= a^{(1)} \tilde{\varepsilon}_{\alpha\beta} + a^{(2)} \varepsilon \delta_{\alpha\beta} + a^{(3)} (T - T_{(0)}) \delta_{\alpha\beta} \\ &+ \sum_{k=1}^n b_{(c\varepsilon)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) \delta_{\alpha\beta} + \eta_v \frac{d\varepsilon}{dt} \delta_{\alpha\beta} + \eta_s \frac{d\tilde{\varepsilon}_{\alpha\beta}}{dt}, \end{aligned} \quad (103)$$

Constitutive equations for $J_{\alpha}^{(q)}$, $J_{(diff)\alpha}^{(j)}$ ($j = 1, 2, \dots, n-1$), $J_{(chem)}^{(l)}$ ($l = 1, 2, \dots, r$) are given by the phenomenological equations (66)-(68).

8 Magnetic relaxation equation for perfect isotropic magnetizable reacting fluid mixtures

In this Section we derive the magnetic relaxation equation for perfect isotropic magnetizable reacting fluid mixtures. Taking into account (98), (99), equations (101) and (102) may be written, respectively, in the form

$$c^{(1)} M_\alpha^{(1)} = Q_{(0,0)\alpha}^{(1)}, \quad (104)$$

$$\frac{dM_\alpha^{(1)}}{dt} + h M_\alpha^{(1)} = Q_{(1,0)\alpha}, \quad (105)$$

where

$$c^{(1)} = a_{(M)}^{(0,0)} + L_{(M)}^{(0,1)} a_{(M)}^{(1,1)}, \quad (106)$$

$$h = L_{(M)}^{(1,1)} a_{(M)}^{(1,1)}, \quad (107)$$

$$Q_{(0,0)\alpha}^{(1)} = a_{(M)}^{(0,0)} \left(1 + L_{(M)}^{(0,1)} \right) M_\alpha + L_{(M)}^{(0,0)} \frac{dM_\alpha}{dt} - B_\alpha, \quad (108)$$

$$Q_{(1,0)\alpha} = L_{(M)}^{(1,1)} a_{(M)}^{(0,0)} M_\alpha + L_{(M)}^{(1,0)} \frac{dM_\alpha}{dt}. \quad (109)$$

Multiplying equation (104) by $(c^{(1)})^{-1}$ the partial magnetization field $\mathbf{M}^{(1)}$ is given by

$$M_\alpha^{(1)} = \left(c^{(1)} \right)^{-1} Q_{(0,0)\alpha}^{(1)}. \quad (110)$$

We calculate the time derivative of (110), having the form

$$\frac{dM_\alpha^{(1)}}{dt} = \left(c^{(1)} \right)^{-1} \frac{dQ_{(0,0)\alpha}^{(1)}}{dt}. \quad (111)$$

Multiplying (105) by $c^{(1)}$ and with the aid of (110) and (111) we obtain

$$c^{(1)} h M_\alpha^{(1)} = c^{(1)} Q_{(1,0)\alpha} - \frac{dQ_{(0,0)\alpha}^{(1)}}{dt}, \quad (112)$$

from which we derive the following magnetic relaxation equation

$$\chi_{(BM)}^{(0)} B_\alpha + \frac{dB_\alpha}{dt} = \chi_{(MB)}^{(0)} M_\alpha + \chi_{(MB)}^{(1)} \frac{dM_\alpha}{dt} + \chi_{(MB)}^{(2)} \frac{d^2 M_\alpha}{dt^2}, \quad (113)$$

where

$$\chi_{(BM)}^{(0)} = c^{(1)} h \left(c^{(1)} \right)^{-1}, \quad (114)$$

$$\chi_{(MB)}^{(0)} = c^{(1)} \left[h(c^{(1)})^{-1} \left(a_{(M)}^{(0,0)} + L_{(M)}^{(0,1)} a_{(M)}^{(0,0)} \right) - L_{(M)}^{(1,1)} a_{(M)}^{(0,0)} \right], \quad (115)$$

$$\chi_{(MB)\alpha\beta}^{(1)} = c^{(1)} \left[h(c^{(1)})^{-1} L_{(M)}^{(0,0)} - L_{(M)}^{(1,0)} \right] + a_{(M)}^{(0,0)} + L_{(M)}^{(0,1)} a_{(M)}^{(0,0)}, \quad (116)$$

$$\chi_{(MB)}^{(2)} = L_{(M)}^{(0,0)}. \quad (117)$$

To derive equation (113) we have assumed that the material coefficients are constant and the total magnetization field and the magnetic field have time derivatives of sufficiently high order. From (113) it is seen that the linearization of the theory leads to a magnetic relaxation equation for the perfect isotropic considered fluid system having the form of a linear relation among the components of the magnetic field, the components of the total magnetization, the first time derivative of the components of the magnetic field, of the total magnetization field, and the second time derivative of this last field. The magnetic relaxation equation (113) does not include the contributions due to the temperature, the concentrations of the n fluid components, and the first time derivatives of the temperature and of the concentrations of the n fluid components, which, however, are present if the considered fluid system is anisotropic (see [38]). Finally, we notice that the relaxation equation (113) could also be derived from the magnetic relaxation equation obtained in [38] for anisotropic reacting fluid mixtures, using relations (41), (57)-(63).

9 Derivation of the field $\mathbf{B}^{(1)}$, the entropy s and the internal energy u in the perfect isotropic case

In this Section we derive the field $\mathbf{B}^{(1)}$, the entropy s and the internal energy u for the considered fluid system in the case when it is perfect isotropic. Using (98)-(101), (108) and (110) we see that the field $\mathbf{B}^{(1)}$ can be written as follows

$$B_{\alpha}^{(1)} = a_{(M)}^{(0,0)} M_{\alpha} - a_{(M)}^{(1,1)} \left(c^{(1)} \right)^{-1} \left[\left(a_{(M)}^{(0,0)} + L_{(M)}^{(0,1)} a_{(M)}^{(0,0)} \right) M_{\alpha} + L_{(M)}^{(0,0)} \frac{dM_{\alpha}}{dt} - B_{\alpha} \right], \quad (118)$$

where $c^{(1)}$ is given by (106).

We have

$$B_{\alpha}^{(1)} = D_{(M)}^{(1)} M_{\alpha} + D_{(M)}^{(2)} \frac{dM_{\alpha}}{dt} + D_{(M)}^{(3)} B_{\alpha}, \quad (119)$$

$$D_{(M)}^{(1)} = \left(c^{(1)}\right)^{-1} \left[a_{(M)}^{(0,0)} c^{(1)} - a_{(M)}^{(1,1)} \left(a_{(M)}^{(0,0)} + L_{(M)}^{(0,1)} a_{(M)}^{(0,0)} \right) \right], \quad (120)$$

$$D_{(M)}^{(2)} = - \left(c^{(1)}\right)^{-1} a_{(M)}^{(1,1)} L_{(M)}^{(0,0)}, \quad (121)$$

$$D_{(M)}^{(3)} = \left(c^{(1)}\right)^{-1} a_{(M)}^{(1,1)}. \quad (122)$$

The partial magnetization field $\mathbf{M}^{(1)}$ is given by the relation (110)

To derive the *internal energy* u , we use equations (71) and (94)-(96), thus we have

$$u = f + Ts = f^{(1)} + f^{(2)} + Ts. \quad (123)$$

From (73) we have the following form for the *specific entropy* s

$$s = - \sum_{k=1}^n b_{(cT)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) - 3va^{(3)}\varepsilon + \frac{d\varphi}{dT}. \quad (124)$$

Then, we obtain

$$\begin{aligned} u = & \frac{1}{2} \varrho \left[a_{(M)}^{(0,0)} (\mathbf{m})^2 - 2a_{(M)}^{(0,0)} \mathbf{m} \cdot \mathbf{m}^{(1)} + a_{(M)}^{(1,1)} (\mathbf{m}^{(1)})^2 \right] \\ & + v \left[\frac{1}{2} a^{(1)} (\tilde{\varepsilon}_{\alpha\beta})^2 + \frac{3}{2} a^{(2)} (\varepsilon)^2 - 3a^{(3)} T_{(0)} \varepsilon \right] + T \frac{d\varphi}{dT} - \varphi(T) \\ & - v T_{(0)} \sum_{k=1}^n b_{(cT)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) + v \sum_{k=1}^n b_{(c\varepsilon)}^{(k)} \left(c^{(k)} - c_{(0)}^{(k)} \right) \varepsilon_{\alpha\alpha} \\ & + \frac{v}{2} \sum_{i,k=1}^n b_{(cc)}^{(i,k)} \left(c^{(i)} - c_{(0)}^{(i)} \right) \left(c^{(k)} - c_{(0)}^{(k)} \right). \end{aligned} \quad (125)$$

The specific heat at constant deformation $c(\varepsilon)$ can be defined by

$$c(\varepsilon) = \frac{\partial u}{\partial T} \left(T, \varepsilon_{\alpha\beta}, \mathbf{m}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)} \right). \quad (126)$$

From (125) and (126) we have

$$c(\varepsilon) = T \frac{d^2 \varphi}{dT^2}. \quad (127)$$

If $c(\varepsilon)$ is constant one obtains the result

$$\varphi = c(\varepsilon)T \log \frac{T}{T_{(0)}} + s_{(0)}T - c(\varepsilon)(T - T_{(0)}) - u_{(0)}, \quad (128)$$

where $s_{(0)}$ and $u_{(0)}$ are the specific entropy and the specific energy in the reference state, respectively. Equation (128) was derived in [13] but was proved in full detail in [16]. Finally, we notice that equations (119), (124) and (125) could also be derived from the analogous equations obtained in [38] for anisotropic reacting fluid mixtures, using relations (41), (57)-(63).

10 Special perfect isotropic cases: Snoek case and De Groot-Mazur case

If we assume that the specific entropy s is a constitutive function of the state space $C = C(u, \varepsilon_{\alpha\beta}, \mathbf{m}^{(0)}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)})$, i.e.

$$s = s(u, \varepsilon_{\alpha\beta}, \mathbf{m}^{(0)}, \mathbf{m}^{(1)}, c^{(1)}, \dots, c^{(n)}), \quad (129)$$

equation (24) for $\sigma^{(s)}$ assumes the following form

$$\begin{aligned} \sigma^{(s)} = & \frac{1}{T} \left[-\frac{1}{T} \mathbf{J}^{(q)} \cdot \nabla T + \tau_{\alpha\beta}^{(vi)} \frac{d\varepsilon_{\alpha\beta}}{dt} + \varrho \mathbf{B}^{(ir)} \cdot \frac{d\mathbf{m}^{(0)}}{dt} \right. \\ & \left. + \varrho (\mathbf{B}^{(1)} + \mathbf{B}^{(ir)}) \cdot \frac{d\mathbf{m}^{(1)}}{dt} + \sum_{h=1}^r A^{(h)} J_{(chem)}^{(h)} + \sum_{k=1}^{n-1} \mathbf{J}_{(diff)}^{(k)} \cdot \mathbf{X}^{(k)} \right]. \quad (130) \end{aligned}$$

It is seen from (130) that if the magnetic field $\mathbf{B}$ equals the equilibrium magnetic field $\mathbf{B}^{(eq)}$, $\mathbf{B}^{(ir)}$ vanishes, only the specific partial magnetization $\mathbf{m}^{(1)}$ contributes to the entropy production, i. e. changes in $\mathbf{m}^{(0)}$ become reversible, and we obtain the *Snoek case*.

In this case the total magnetization vector $\mathbf{M}$ is composed of two parts $\mathbf{M} = \mathbf{M}^{(0)} + \mathbf{M}^{(1)}$, where the contribution $\mathbf{M}^{(0)}$, defined by $\mathbf{M}^{(0)} = \varrho \mathbf{m}^{(0)}$, has reversible character and the contribution $\mathbf{M}^{(1)}$ has irreversible character. Furthermore, when $\mathbf{B}^{(ir)}$ vanishes (see (20)₂) from (101) and (102) we have

$$L_{(M)\alpha\beta}^{(0,0)} = 0, \quad \text{and} \quad L_{(M)\alpha\beta}^{(0,1)} = -L_{(M)\beta\alpha}^{(1,0)} = 0. \quad (131)$$

Thus, the magnetic relaxation equation (113) becomes

$$\chi_{(BM)}^{(0)} B_\alpha + \frac{dB_\alpha}{dt} = \chi_{(MB)}^{(0)} M_\alpha + \chi_{(MB)}^{(1)} \frac{dM_\alpha}{dt}, \quad (132)$$

where

$$\chi_{(BM)}^{(0)} = L_{(M)}^{(1,1)} a_{(M)}^{(1,1)}, \quad (133)$$

$$\chi_{(MB)}^{(0)} = L_{(M)}^{(1,1)} \left[a_{(M)}^{(0,0)} a_{(M)}^{(1,1)} - \left(a_{(M)}^{(0,0)} \right)^2 \right], \quad (134)$$

$$\chi_{(MB)}^{(1)} = a_{(M)}^{(0,0)}. \quad (135)$$

Equation (132), in the case where we have only an one component system without chemical reactions, reduces to *Snoek equation* for perfect isotropic systems.

Also, from (130) we can obtain *De Groot-Mazur case*, when $\mathbf{m} = \mathbf{m}^{(0)}$, i.e. there is no internal variable $\mathbf{m}^{(1)}$, and $\mathbf{m}^{(0)}$ is reversible. In this case

$$L_{(M)}^{(1,1)} = 0, \quad \text{and} \quad L_{(M)}^{(1,0)} = -L_{(M)}^{(0,1)} = 0 \quad (136)$$

Equations (101) and (102) become

$$B_\alpha^{(ir)} = L_{(M)\alpha\beta}^{(0,0)} \frac{dM_\beta}{dt} \quad (137)$$

and

$$\frac{dM_\alpha^{(1)}}{dt} = 0. \quad (138)$$

It is seen that $\mathbf{M}^{(1)}$ is constant and it can be supposed that $\mathbf{M}^{(1)} = 0$.

Equation (113) reduces to a generalized *De Groot-Mazur* magnetic relaxation equation (see [9]- [11])

$$B_\alpha = \chi_{(MB)}^{(1)} M_\alpha + \chi_{(MB)}^{(2)} \frac{dM_\alpha}{dt}, \quad (139)$$

where

$$\chi_{(MB)}^{(1)} = a_{(M)}^{(0,0)}, \quad (140)$$

$$\chi_{(MB)}^{(2)} = L_{(M)}^{(0,0)}, \quad (141)$$

The magnetic relaxation equations (132) and (139) could have been derived from the analogous equations obtained in [38] for anisotropic reacting fluid mixtures, using relations (41), (57)-(63).

11 Conclusions

In this paper, we have described the behavior of isotropic and perfect isotropic magnetizable reacting fluid mixtures, within the framework of classical irreversible thermodynamics with internal variables. We have assumed that the irreversible microscopic phenomena responsible for magnetic relaxation can be modeled by splitting the total specific magnetization into two irreversible contributions and introducing one of them as an internal variable in the thermodynamic state vector. Under the assumptions of negligible body forces and heat sources, constant mass density and phenomenological coefficients, we have derived, within the linear approximation, the phenomenological equations and the entropy production for both isotropic and perfect isotropic systems. For perfect isotropic mixtures, we have obtained the equations of state, the constitutive equations, the magnetic relaxation equation, the field $\mathbf{B}^{(1)}$ conjugated to the internal variable $\mathbf{m}^{(1)}$, as well as the specific internal energy and the specific entropy. Together with the governing equations, these results provide to describe the considered systems. A forthcoming paper will address the first law of thermodynamics and the heat conduction equation in the perfect isotropic case, thereby completing the theoretical framework. The approach adopted here was developed by the authors for anisotropic reacting fluid mixtures with magnetic relaxation in [16, 38], where the phenomenological equations, the magnetic relaxation equation and the heat equation with the dissipation function were derived. In contrast to the anisotropic case, perfect isotropic systems exhibit no cross effects between magnetic relaxation and other irreversible processes, since fluxes and thermodynamic forces of different tensorial character do not couple. Likewise, in the magnetic relaxation equation for perfect isotropic systems, no contributions arise from the temperature, the concentrations of the fluid components, and their time derivatives, terms that are instead present in anisotropic media. Since 1973 (see [9]), several linear theories for magnetizable media with magnetic relaxation have been developed within the same framework of classical irreversible thermodynamics with internal variables, which are a powerful tool for describing complex materials. In some approaches, such as the one used here, the internal variables are introduced directly, while in others they can be identified through appropriate methods as in [41] and [42], or even eliminated depending on the chosen formalism as in [4]. The results obtained here have applications in nuclear magnetic resonance, biological systems, and medical diagnostics where multiple species with distinct susceptibilities and relaxation times contribute to the total magnetization. Finally, we remark that, in order to obtain a complete de-

scription of a material system in the isotropic or perfect isotropic cases and to derive phenomenological equations that include all the contributions of the fields involved in the problem under consideration, it is necessary to start from the anisotropic case and then to use the isotropy or perfect isotropy relations for the phenomenological and material coefficients. The same applies to the equations of state.

Appendix

In this Appendix we demonstrate the phenomenological equation (54) for $\tau_{\alpha\beta}^{(vi)}$ valid in the case in which the fluid system is isotropic respect to all rotations of the frame of axes (and also when it is perfect isotropic respect to all rotations and inversions of the frame of axes). From the phenomenological equation (54), using relations (41), (43)-(45), we have

$$\begin{aligned}\tau_{\alpha\beta}^{(vi,vi)} &= \sum_{h=1}^r L_{(C)}^{(vi,h)} A^{(h)} \delta_{\alpha\beta} + [A_1 \delta_{\alpha\beta} \delta_{\zeta\gamma} + A_2 (\delta_{\alpha\zeta} \delta_{\beta\gamma} + \delta_{\alpha\gamma} \delta_{\beta\zeta})] \frac{d\epsilon_{\zeta\gamma}}{dt} \\ &= \sum_{h=1}^r L_{(C)}^{(vi,h)} A^{(h)} \delta_{\alpha\beta} + A_1 \delta_{\alpha\beta} \frac{d\epsilon_{\gamma\gamma}}{dt} + A_2 [\delta_{\alpha\zeta} \delta_{\beta\gamma} + \delta_{\alpha\gamma} \delta_{\beta\zeta}] \frac{d\epsilon_{\zeta\gamma}}{dt} \\ &= \sum_{h=1}^r L_{(C)}^{(vi,h)} A^{(h)} \delta_{\alpha\beta} + (3A_1 + 2A_2) \frac{d\epsilon}{dt} \delta_{\alpha\beta} + 2A_2 \frac{d\tilde{\epsilon}_{\alpha\beta}}{dt},\end{aligned}\quad (142)$$

where $\frac{d\epsilon}{dt}$ and $\frac{d\tilde{\epsilon}_{\alpha\beta}}{dt}$ the scalar part and the deviatoric part, respectively, of the symmetric tensor field of order two $\frac{d\epsilon_{\alpha\beta}}{dt}$. Furthermore, because $\frac{d\epsilon_{\alpha\beta}}{dt}$ is symmetric also its deviatoric part is symmetric.

If we assume

$$\eta_v = (3A_1 + 2A_2) \quad \text{and} \quad \eta_s = 2A_2,$$

with η_v and η_s the bulk viscosity and the shear viscosity, respectively, of the medium under consideration, we obtain

$$\tau_{\alpha\beta}^{(vi)} = \sum_{h=1}^r L_{(C)}^{(vi,h)} A^{(h)} \delta_{\alpha\beta} + \eta_v \frac{d\epsilon}{dt} \delta_{\alpha\beta} + \eta_s \frac{d\tilde{\epsilon}_{\alpha\beta}}{dt}.$$

From the definitions (11) we have

$$\frac{d\epsilon}{dt} = \frac{1}{3} \text{div} \mathbf{v} \quad \text{and} \quad \frac{d\tilde{\epsilon}_{\alpha\beta}}{dt} = \frac{1}{2} \left(\frac{\partial v_\alpha}{\partial x_\beta} + \frac{\partial v_\beta}{\partial x_\alpha} - \frac{2}{3} \delta_{\alpha\beta} \text{div} \mathbf{v} \right). \quad (143)$$

Acknowledgements. A.L. acknowledges the support of the National Group of Mathematical Physics, GNFM-INdAM. L.R. acknowledges the hospitality of Bari University.

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