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Thermodynamics of Viscodielectric Materials

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1. Introduction

Extended Irreversible Thermodynamics (EIT) [1,2] has proved to be a useful tool to analyze the non-equilibrium behaviour of complex materials close to the linear regime. The axiomatic formulation of the EIT ([1], p.54) furnishes a formal structure that can be used as the basis of the study of different type of systems, i.e., dielectric/magnetic[3-5] and in particular viscoelastic materials [6-8]. Basically EIT extends the scope of the Classical Irreversible Thermodynamics (CIT) as developed by Prigogine, Onsager and more recently by De Groot and Mazur [9] among others. The basic idea is to formulate a generalized entropy function that depends not only on the conserved classical variables, but also on the dissipative fluxes. This implies abandoning the local equilibrium hypothesis. In the particular case of viscoelastic materials, the dissipative flux is the viscous component of the stress tensor.

It is important to note that owing to the memory exhibited by viscoelastic materials the mechanical and dielectric response of these substances to mechanical and electric perturbation fields applied at time t not only depends on the actual fields at t but also on the history of the materials in the range $-\infty < \vartheta \le t$. In fact the phenomenological theory of mechanical relaxations is nearly the counterpart of dielectric relaxations [10] in such a way that in the general case both phenomena can be considered coupled processes. To account for the interactions between mechanical and dielectric compliance in the continuum it is convenient to describe adequately such a coupling in terms of the polarization vector and the stress tensor. However, since the symmetric part of the stress tensor only accounts for the translational hydrodynamic and taking into account that polarization arises from rotation of dipoles, in what follows that part will not be considered because does not contribute to the dielectric relaxation. It will be also assumed that the relative velocity of the medium with respect to dipoles rotation is negligible. In this way only the antisymmetric part of the stress tensor produces an effective polarization under an electric field. This is the specific way to locally relate the dipole rotation with the mechanical friction.

2. Balance equations

The linear momentum equation in local form can be written

$$div\boldsymbol{\sigma} + \rho\mathbf{b} = \rho\ddot{\mathbf{u}} \tag{1}$$

where **b** is the volume force and **u** is the displacement; bold symbols are used for tensors of order ≥ 1.

The flux or current equation for polarization charges can be written as

$$\frac{d\rho}{dt} + div\mathbf{J} = 0, \quad \mathbf{J} = \frac{d\mathbf{P}}{dt} \tag{2}$$

where **P** is the polarization vector.

In local form the more convenient expression for the energy balance, excluding radiation and other thermal effects is

$$\mathbf{E} \cdot \dot{\mathbf{P}} + \mathbf{Q}grad\mathbf{v} = \rho\dot{u} \tag{3}$$

where **E** is the electric field, u is the internal energy per unit of mass, ρ is the mass density and **Q** is the sum of the mechanical stress tensor $\boldsymbol{\sigma}$ and the Maxwell stress tensor **T**.

In absence of magnetic fields or induced magnetization the Maxwell stress tensor is expressed by the following equation

$$\mathbf{T} = \mathbf{DE} - \frac{1}{2}\varepsilon_0 E^2 \mathbf{I} \tag{4}$$

where **D** is the dielectric displacement, ε_0 the vacuum permittivity and **I** represents the unit tensor.

The tensor **Q** can be decomposed in the corresponding symmetric and antisymmetric parts as follows

$$\mathbf{Q}^s = \frac{1}{2}(\mathbf{Q} + \mathbf{Q}^T), \quad \mathbf{Q}^a = \frac{1}{2}(\mathbf{Q} - \mathbf{Q}^T) \tag{5}$$

where super index T indicates transposition operation.

The gradient of velocity tensor can be decomposed in a similar way, *i.e.*,

$$grad\mathbf{v} = \mathbf{L} = \mathbf{D} + \mathbf{W} \tag{6}$$

On account that the inner product of a symmetric tensor by an antisymmetric tensor is nil, the following expression holds

$$\mathbf{QL} = \mathbf{Q}^s\mathbf{D} + \mathbf{Q}^a\mathbf{W} \tag{7}$$

However, owing to the fact that the symmetric part of the total stress tensor is not coupled with dipoles rotation and **W** is associated to the angular rotation vector

$$\boldsymbol{\omega} = \frac{1}{2}\nabla \times \mathbf{v} \tag{8}$$

the two vectors $\boldsymbol{\omega}$ and **W** are related in the usual way, *i.e.*,

$$\varepsilon_{pqi}\omega_i = \frac{1}{2}\varepsilon_{pqi}\varepsilon_{ijk}v_{k,j} = \frac{1}{2}(\delta_{pj}\delta_{qk} - \delta_{pk}\delta_{qj})v_{k,j} = \frac{1}{2}(v_{q,p} - v_{p,q}) = W_{qp} \tag{9}$$

where the index notation was used, comma indicates derivation and ε_{ijk} is the alternating tensor. As a consequence, one can write

$$\mathbf{Q^a W} = \mathbf{Q^a \varepsilon \omega} \tag{10}$$

Then eq. (3) can be rewritten in the following way

$$\rho \frac{du}{dt} = \mathbf{E\dot{P}} + \mathbf{Q^a \varepsilon \omega} \tag{11}$$

The conservation of the angular moment has to be considered, *i.e.*,

$$I\dot{\boldsymbol{\omega}} = \boldsymbol{\varepsilon} \mathbf{Q^a} \tag{12}$$

where I is the mean inertia moment of the rotating dipoles.

In general, the entropy production per unit of time is the sum of the entropy flux from the exterior plus the internally generated entropy. Accordingly, one can write

$$\rho \frac{ds}{dt} = -div\mathbf{J}_S + \sigma_s \tag{13}$$

where $\mathbf{J}_s$ is the entropy flux and σ_s the entropy production per unit of volume and time.

3. Entropy equation

According to the usual methodology of the EIT, it will assumed that there exists a regular enough function, called generalized entropy defined over a set of variables

$$\eta = \hat{\eta}(u, \rho_p, \mathbf{J}, \mathbf{Q}) \tag{14}$$

This function is such that the corresponding generalized Gibbs equation can be written as

$$\rho \frac{d\eta}{dt} = \frac{\rho}{T} \frac{du}{dt} + \frac{\phi}{T} \frac{d\rho_p}{dt} + \alpha_1 \mathbf{J} \frac{d\mathbf{J}}{dt} + \alpha_2 \mathbf{Q^a} \frac{d\mathbf{Q^a}}{dt} \tag{15}$$

where ρ_p is the polarization density and ϕ is the electrical potential. In principle ϕ and the coefficients α_i should be functions of the conserved variables, u, ρ_p, but here for simplicity will be considered constant quantities.

Then, the following equation will be assumed for the extended entropy flux,

$$\mathbf{J}_\eta = \mu_1 \mathbf{J} + \mu_2' \mathbf{JQ^a} \tag{16}$$

where μ_i is subject to the same restrictions as α_i.

By using eqs. (14) and (15) the entropy production is given by

$$\begin{aligned} \sigma_\eta &= \rho \frac{d\eta}{dt} + div\mathbf{J}_\eta = \\ &= \frac{\rho}{T} \frac{du}{dt} + \frac{\phi}{T} \frac{d\rho_p}{dt} + \alpha_1 \mathbf{J} \frac{d\mathbf{J}}{dt} + \alpha_2 \mathbf{Q^a} + \mu_1 div\mathbf{J} + \mathbf{J} grad\mu_1 + \mu_2' \mathbf{J} div\mathbf{Q^a} + \mu_2' \mathbf{Q^a} (grad\mathbf{J})^a \end{aligned} \tag{17}$$

According to eqs. (1), (2) and (11) and after grouping terms, eq. (17) becomes

$$\sigma_{\eta} = \dot{\mathbf{P}}\left(\frac{\mathbf{E}}{T} + \alpha_1\ddot{\mathbf{P}} - \frac{1}{\varepsilon_0\chi T}\mathbf{P} + \mu_2'div\mathbf{Q}^a\right) + \mathbf{Q}^a\left(\frac{1}{T}\boldsymbol{\varepsilon}\boldsymbol{\omega} + \alpha_2\dot{\mathbf{Q}}^a + \mu_2'(grad\mathbf{J})^a\right) \tag{18}$$

Notice that in the development of eq. (18) the following expressions were used

$$\mu_1 = \frac{\phi}{T}, \quad grad\phi = -\frac{\mathbf{P}}{\chi\varepsilon_0} \tag{19}$$

where χ is the dielectric susceptibility. Eq. (18) has the form of a linear combination of products of two forces (terms between brackets) by the corresponding fluxes, $\dot{\mathbf{P}}$ and $\mathbf{Q}^a$.

According to the methodology of CIT the forces can be expressed as linear functions of current fluxes and their spatial and temporal derivatives (entire or fractional) in order to account for the polarization inhomogeneity. In this way,

$$\frac{\mathbf{E}}{T} + \alpha_1\ddot{\mathbf{P}} - \frac{1}{\varepsilon_0\chi T}\mathbf{P} + \mu_2'div\mathbf{Q}^a =$$

$$= \xi_1\left(\frac{d^{\alpha}\mathbf{P}}{dt^{\alpha}} + \frac{d^{\beta}\mathbf{P}}{dt^{\beta}}\right) + \xi_2 div\,(grad\mathbf{P})^a + \xi_3 div\left(grad\left(\frac{d^{\alpha}\mathbf{P}}{dt^{\alpha}} + \frac{d^{\beta}\mathbf{P}}{dt^{\beta}}\right)\right)^a + \xi_3'div\mathbf{Q}^a$$

$$\frac{1}{T}\boldsymbol{\varepsilon}\boldsymbol{\omega} + \alpha_2\dot{\mathbf{Q}}^a + \mu_2'(grad\dot{\mathbf{P}})^a = \tag{20}$$

$$= \xi_4\mathbf{Q}^a + \xi_5(grad\mathbf{P})^a + \xi_6\left(grad\left(\frac{d^{\alpha}\mathbf{P}}{dt^{\alpha}} + \frac{d^{\beta}\mathbf{P}}{dt^{\beta}}\right)\right)^a$$

where fractional derivatives have been introduced. By grouping terms eq. (20) can be rewritten as

$$\frac{\mathbf{E}}{T} + \alpha_1\ddot{\mathbf{P}} - \frac{1}{\varepsilon_0\chi T}\mathbf{P} + \mu_2 div\mathbf{Q}^a =$$

$$= \xi_1\left(\frac{d^{\alpha}\mathbf{P}}{dt^{\alpha}} + \frac{d^{\beta}\mathbf{P}}{dt^{\beta}}\right) + \xi_2 div(grad\mathbf{P})^a + \xi_3 div\left(grad\left(\frac{d^{\alpha}\mathbf{P}}{dt^{\alpha}} + \frac{d^{\beta}\mathbf{P}}{dt^{\beta}}\right)\right)^a$$

$$\frac{1}{T}\boldsymbol{\varepsilon}\boldsymbol{\omega} + \alpha_2\dot{\mathbf{Q}}^a + \mu_2'(grad\dot{\mathbf{P}})^a = \tag{21}$$

$$= \xi_4\mathbf{Q}^a + \xi_5(grad\mathbf{P})^a + \xi_6\left(grad\left(\frac{d^{\alpha}\mathbf{P}}{dt^{\alpha}} + \frac{d^{\beta}\mathbf{P}}{dt^{\beta}}\right)\right)^a$$

where $\mu_2 = \mu_2' - \xi_3'$.

Eq. (21b) contains the angular velocity $\boldsymbol{\omega}$ which can be eliminated in favour of $\mathbf{Q}^a$ by using (12). According to eq. (12) and premultiplying by the alternating tensor $\boldsymbol{\varepsilon}$, one obtains

$$\boldsymbol{\varepsilon}\dot{\boldsymbol{\omega}} = \frac{\boldsymbol{\varepsilon}\boldsymbol{\varepsilon}\mathbf{Q}^a}{I} \tag{22}$$

According to eq. (9), and taking into account the antisymmetric character of $\mathbf{Q}^a$, the following relationship holds

$$\varepsilon_{pqi}\varepsilon_{ijk}Q_{hl}^{a} = \left(\delta_{ih}\delta_{jl} - \delta_{il}\delta_{hj}\right)Q_{hl}^{a} = Q_{ij}^{a} - Q_{ji}^{a} = 2Q_{ij}^{a} \tag{23}$$

and eq. (22) can be written as

$$\boldsymbol{\varepsilon}\dot{\boldsymbol{\omega}} = \frac{2\mathbf{Q}^{a}}{I} \tag{24}$$

In principle eq. (21) and (24) should provide the solution to our problem. However, in order to interpret the results, it is more convenient to consider some particular cases. To start with let we assume $\alpha = 1$; after omission of the fractional derivatives of order β, eq. (21) can be written as

$$\frac{\mathbf{E}}{T} + \alpha_{1}\ddot{\mathbf{P}} - \frac{1}{\varepsilon_{0}\chi T}\mathbf{P} + \mu_{2}div\mathbf{Q}^{a} = \xi_{1}\dot{\mathbf{P}} + \xi_{2}div(grad\mathbf{P})^{a} + \xi_{3}div(grad\dot{\mathbf{P}})^{a}$$
$$\frac{1}{T}\boldsymbol{\varepsilon}\boldsymbol{\omega} + \alpha_{2}\dot{\mathbf{Q}}^{a} + \mu_{3}(grad\dot{\mathbf{P}})^{a} = \xi_{4}\mathbf{Q}^{a} + \xi_{5}(grad\mathbf{P})^{a} \tag{25}$$

where $\mu_{3} = \mu_{2}' - \mu_{6}$.

From eq. (24) and after taking derivatives, eq. (25b) becomes

$$\frac{2}{IT}\mathbf{Q}^{a} + \alpha_{2}\ddot{\mathbf{Q}}^{a} + \mu_{3}(grad\ddot{\mathbf{P}})^{a} = \xi_{4}\dot{\mathbf{Q}}^{a} + \xi_{5}(grad\dot{\mathbf{P}})^{a} \tag{26}$$

The second term of the right hand side of eq. (25a) can be written as

$$div(grad\mathbf{P})^{a} = \frac{1}{2}\left(\Delta\mathbf{P} - graddiv\mathbf{P}\right) \tag{27}$$

For a unidirectional propagation wave with vector wave $\mathbf{k} = (k,0,0)$ under an electrical field transversal to the propagation, i.e., $\mathbf{E} = (0,E_{2},E_{3})$, the components for the dielectric susceptibility χ_{ij} tensor are

$$\chi_{ij} = 0, \quad k_{i}k_{j} = 0, \quad \forall i \neq j; \quad \chi_{11} = 0 \tag{28}$$

Notice that the constitutive relationship between polarization and electric field is given by

$$P_{i} = \varepsilon_{0}\chi_{ij}E_{j} \tag{29}$$

Accordingly, eq. (27) can be written as

$$\mathbf{k}\cdot\left(\mathbf{k}\mathbf{P}^{a}\right) = \frac{1}{2}\left(k^{2}\mathbf{P} - \mathbf{k}\left(\mathbf{k}\cdot\mathbf{P}^{a}\right)\right) \tag{30}$$

This expression in conjunction with $(graddiv\mathbf{P})_{i} = k_{j}k_{i}P_{i} = 0$, leads eq. (27) to

$$div(grad\mathbf{P})^{a} = \frac{1}{2}\Delta\mathbf{P} \tag{31}$$

Elimination of $\mathbf{Q}^{a}$ between eq. (25b) and (26), and considering eq. (21), the following expression is obtained after multiplying by $\varepsilon_{0}\chi IT^{2}\mu_{2}$,

$$\mathbf{P} + \dot{\mathbf{P}}T\left(2\xi_1\varepsilon_0\chi - \xi_4 I\right) + \ddot{\mathbf{P}}T(\alpha_2 I - \xi_1\xi_4\varepsilon_0\chi IT - 2\alpha_1\varepsilon_0\chi) + \dddot{\mathbf{P}}\varepsilon_0\chi IT^2(\alpha_2\xi_1 + \alpha_1\xi_4) -$$
$$-\dddot{\mathbf{P}}\alpha_1\alpha_2\varepsilon_0\chi IT^2 + \frac{1}{2}\Delta\mathbf{P}(2\xi_2\varepsilon_0\chi T) + \frac{1}{2}\Delta\dot{\mathbf{P}}\varepsilon_0\chi T(2\xi_3 - \xi_5\mu_2 IT - \xi_2\xi_4 IT) +$$
$$+\frac{1}{2}\Delta\ddot{\mathbf{P}}\varepsilon_0\chi T^2 I(\alpha_1\xi_2 + \mu_2\mu_3 - \xi_3\xi_4) + \frac{1}{2}\Delta\dddot{\mathbf{P}}(\alpha_2\xi_3\varepsilon_0\chi IT^2) -$$
$$-\varepsilon_0\chi(2\mathbf{E} - \xi_4 IT\dot{\mathbf{E}} + \alpha_2 IT\ddot{\mathbf{E}}) = 0 \tag{32}$$

If electromechanical coupling is absent, *i.e.*, $\mu_2 = 0$ one obtains

$$\frac{\mathbf{E}}{T} + \alpha_1\ddot{\mathbf{P}} - \frac{1}{\varepsilon_0\chi T}\mathbf{P} = \xi_1\dot{\mathbf{P}} + \xi_2 div(grad\mathbf{P})^a + \xi_3 div(grad\dot{\mathbf{P}})^a \tag{33}$$

Rearrangement of eq. (33) in conjunction with eq. (31) leads to

$$\mathbf{P} + \tau\dot{\mathbf{P}} + \lambda_1\ddot{\mathbf{P}} = \varepsilon_0\chi\mathbf{E} + \frac{1}{2}D_1\Delta\mathbf{P} + \frac{1}{2}D_2\Delta\dot{\mathbf{P}} \tag{34}$$

where

$$\tau = \varepsilon_0\chi T\xi_1, \quad \lambda_1 = -\alpha_1\varepsilon_0\chi T, \quad D_1 = -\varepsilon_0\chi T\xi_2, \quad D_2 = -\varepsilon_0\chi T\xi_3 \tag{35}$$

Equation (34) is a telegrapher type equation for the polarization propagation. The velocity of the propagation *c* is given by

$$c^2 = \frac{D_1}{2\lambda_1} \tag{36}$$

If $D_1 = D_2 = 0$, eq. (34), becomes a Debye type equation with inertial effects, *i.e.*, the so-called Rocard equation is obtained. Finally, if $\lambda_1 = 0$, the classical Debye equation is recovered.

4. Complex dielectric permittivity

In this section, the linear response to alternating electrical fields will be considered. For this purpose the Laplace transform will be used. By substituting eq. (24) in the temporal derivative of eq. (21b) one obtains

$$\frac{2\mathbf{Q}^a}{IT} + \alpha_2\ddot{\mathbf{Q}}^a + \mu_2(grad\ddot{\mathbf{P}})^a = \xi_4\dot{\mathbf{Q}}^a + \xi_5(grad\dot{\mathbf{P}})^a + \xi_6 grad\left(\frac{d^{\alpha+1}\mathbf{P}}{dt^\alpha} + \frac{d^{\beta+1}\dot{\mathbf{P}}}{dt^\beta}\right)^a \tag{37}$$

In order to simplify the calculations and without losing significant generality, it will be assumed $\xi_6 = 0$. In an analogous way, taking $\xi_3 = 0$, eqs. (33) and (37) can be written as

$$\frac{\mathbf{E}}{T} + \alpha_1\ddot{\mathbf{P}} - \frac{1}{\varepsilon_0\chi T}\mathbf{P} + \mu_2 div\mathbf{Q}^a = \xi_1\left(\frac{d^\alpha\mathbf{P}}{dt^\alpha} + \frac{d^\beta\mathbf{P}}{dt^\beta}\right) + \xi_2 div(grad\mathbf{P})^a$$

$$\frac{2}{IT}\mathbf{Q}^a + \alpha_2\ddot{\mathbf{Q}}^a + \mu_2(grad\ddot{\mathbf{P}})^a = \xi_4\dot{\mathbf{Q}}^a + \xi_5(grad\dot{\mathbf{P}})^a \tag{38}$$

The two first derivatives of eq. (38a) and the divergence of (38b) are used to eliminate $\mathbf{Q}^a$. Then the Laplace transform of the resulting expression, in conjunction with eqs. (28) and (31), lead to

$$\chi^* = \chi\left[\left(1+\lambda_1 s^2 + \tau_D(s^\alpha + s^\beta)\right) - \frac{1}{2}\frac{k^2}{\xi_1}\left(\xi_2 + \frac{\mu_2\xi_5 s(1+\lambda_2 s)}{\xi_4 s(1+\lambda_3 s) - \frac{2}{IT}}\right)\right]^{-1} \tag{39}$$

where

$$\tau_D = \xi_1\varepsilon_0\chi T, \quad \lambda_1 = -\frac{\alpha_1}{\xi_1}, \quad \lambda_2 = -\frac{\mu_3}{\xi_5}, \quad \lambda_3 = -\frac{\alpha_2}{\xi_4} \tag{40}$$

Taking $\chi = \chi_0 - \chi_\infty$, adding the instantaneous component of the polarization χ_∞, and using, $s = i\omega$, eq. (39) becomes

$$\chi^* = \chi_\infty + \frac{\chi_0 - \chi_\infty}{\left(1-\lambda_1\omega^2 + \tau_D((i\omega)^\alpha + (i\omega)^\beta)\right) - \frac{1}{2}\frac{k^2}{\xi_1}\left(\xi_2 + \frac{i\mu_2\xi_5\omega(1+i\lambda_2\omega)}{i\xi_4\omega(1+i\lambda_3\omega) - \frac{2}{IT}}\right)} \tag{41}$$

Taking into account that

$$\chi^* = \varepsilon_r^* - 1, \chi_0 = \varepsilon_0 - 1, \chi_\infty = \varepsilon_\infty - 1 \tag{42}$$

Eq. (41) can be expressed in terms of the permittivity by

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{\left(1-\lambda_1\omega^2 + \tau_D((i\omega)^\alpha + (i\omega)^\beta)\right) - \frac{1}{2}\frac{k^2}{\xi_1}\left(\xi_2 + \frac{i\mu_2\xi_5\omega(1+i\lambda_2\omega)}{i\xi_4\omega(1+i\lambda_3\omega) - \frac{2}{IT}}\right)} \tag{43}$$

Equation (43) is the equation for the asymmetric absorption and dispersion of a viscodielectric material in a heterogeneous medium, with inertial viscoelastic coupling effects. The heterogeneity of the medium is taken into account through the wave vector k and the viscoelastic coupling is accounted for by means of the coefficient μ_2. The asymmetry is present through the different behaviour at low and high frequencies of both sides of the corresponding transition peak. This asymmetry has been introduced in the model by means of the degrees α and β of the fractional derivatives. Dielectric and mechanical inertial effects are expressed through λ_1 and I, respectively.

It should be noted that in the present formulation it is compulsory to take into account inertial effects because if $I = 0$ in eq. (43), the viscoelastic-dielectric coupling disappears. To avoid this problem it should be necessary to introduce in eq. (24) a term corresponding to the spin transport.

It is noteworthy that, strictly speaking, the molecule "sees" the local field, i.e., the field screened by the dipolar cloud surrounding the molecule. However in the preceding analysis the distinction between the applied and local field has not been considered, thus making equivalent macroscopic and microscopic responses. This a common practice in the analysis of the dielectric relaxation.

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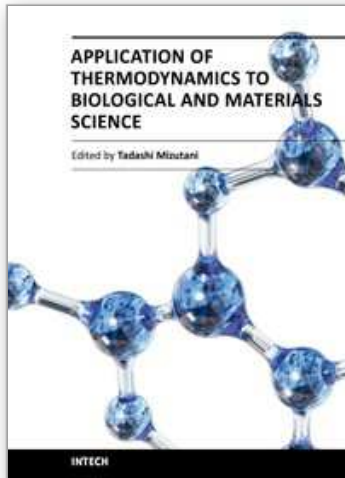
Application of Thermodynamics to Biological and Materials Science

Edited by Prof. Mizutani Tadashi

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