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**A note on Toupin's functional for dielectric materials (\*\*)**

Dedicated to Prof. T. Manacorda, on the occasion of his 70<sup>th</sup> birthday

**1 - Introduction**

In 1956 Toupin [14] proposed a variational principle for elastic dielectrics, from which he derived the Maxwell equations along with Cauchy's equation for stresses, under the form of the Euler-Lagrange equations. However, a remarkable feature of Toupin's procedure is that the Maxwell's stress tensor can be also defined as in the classical theory. The physical meaning of Toupin's variational principle is very simple: it states that the sum of electrostatic energy and thermomechanical energy of the system, composed by the body and the space surrounding it, is a minimum. Clearly the method is not new since Mossotti [9], before Toupin, had already proposed equilibrium equations for dielectric bodies, but Toupin gave a more general approach from the macroscopic point of view.

In 1971 Mindlin [7] [8] re-proposed Toupin's principle introducing the gradient of polarization among the independent variables of the energy functional, recovering in this way the piezoelectric effects in centrosymmetric and isotropic materials, which are not explained by the classical theory. More recently Maugin [6], derived the field equations for a dielectric continuum using the principle of the virtual power.

In this note the connection is shown between the classical form of the electrostatic energy, as given by Stratton [13] and Becker [1], and the Toupin's functional.

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In this respect, an energy density is assumed to exist in the entire space. Having distinguished the global electric field  $\mathbf{E}$  from the field  $\mathbf{E}_0$  which is due a to conductor in absence of dielectrics, it is possible to characterize a functional for dielectric bodies. Such a functional depends on the polarization and on the difference of these fields  $\mathbf{E}$  and  $\mathbf{E}_0$ . It will be called *electric free enthalpy of dielectrics*.

## 2 - The electrostatic energy

Let  $\mathbf{E}$  and  $\mathbf{D}$  be the electric field and the displacement field respectively; both defined in the Euclidean space  $\mathbb{R}^3$ . Let  $V$  and  $C$  be two bounded and disjointed open sets, of which  $V$  represents the region occupied by a dielectric and  $C$  is occupied by a conductor.  $BV$  and  $BC$  denote their boundaries respectively and let  $\mathbf{n}$  be the unit normal vector to  $BV$  and  $\mathbf{v}$  the unit normal vector to  $BC$ . Both these boundaries are assumed regular enough to apply the divergence theorem. The following expression for the electrostatic energy [13], [1], [2] is costumarily given

$$(2.1) \quad \mathcal{E} = \int_{\mathbb{R}^3} \int_0^{\mathbf{D}} \mathbf{E} \cdot \delta \mathbf{D} \, dV$$

and its first variation with respect to  $\mathbf{D}$  is denoted by

$$(2.2) \quad \delta \mathcal{E} = \int_{\mathbb{R}^3} \mathbf{E} \cdot \delta \mathbf{D} \, dV.$$

The quantity  $\mathcal{E}$  is evaluated with respect to a reference configuration in which the dielectric body is free of stresses and polarization, moreover, the electric fields are absent in the entire space. It is assumed that the energy density in the space has the form  $f(\mathbf{D}, T)$ , where  $\mathbf{D}$  is the electric displacement and  $T$  is the absolute temperature which is supposed to vanish identically out of  $V$  and  $C$ . The temperature  $T$  is assumed to be uniform and constant. The field  $\mathbf{D}$  must satisfy the following conditions:

$$(2.3)_1 \quad \operatorname{div} \mathbf{D} = 0 \quad \text{in } \mathbb{R}^3 - C \quad [\mathbf{D}] \cdot \mathbf{n} = 0 \quad \text{across } BV$$

$$(2.3)_2 \quad [\mathbf{D}] \cdot \mathbf{v} = -\mathbf{D} \cdot \mathbf{v}^- \quad \text{across } BC \quad \mathbf{D} \equiv 0 \quad \text{in } \bar{C}$$

$$(2.3)_3 \quad \int_{BC} -\mathbf{D}^- \cdot \mathbf{v} \, ds = q \quad |\mathbf{D}|_\infty \sim \frac{1}{R^2}$$

where brackets denote the discontinuities of the quantity included and  $q$  represents the electric free charge.

In addition,  $f$  is required to be restricted as follows

$$(2.4) \quad |f| \leq k_1 |\mathbf{D}|^2 \quad |f_{,D}| \leq k_2 |\mathbf{D}| \quad k_1, k_2 \in \mathbb{R}^+ \text{ as } R \rightarrow \infty.$$

Also the field  $\delta \mathbf{D}$  must satisfy the conditions (2.3) but  $\delta q$  at  $BC$  is required not to vanish. Let us consider the quantity

$$(2.5) \quad \mathcal{F} = \int_{\mathbb{R}^3} f(\mathbf{D}) dV$$

and its first variation  $\delta \mathcal{F}$ .

According to Landau [4], the isothermal electrostatic equilibrium is ensured by the variational principle

$$(2.6) \quad \int_{\mathbb{R}^3} f_{,D} \cdot \delta \mathbf{D} dV - \int_{\mathbb{R}^3} \phi \delta(\operatorname{div} \mathbf{D}) dV + \phi_0 \int_{BC} \delta([\mathbf{D} \cdot \mathbf{v}]) dS = 0$$

where  $\phi$  and  $\phi_0$  are Lagrange multipliers which satisfy the following conditions:

$$(2.7) \quad \phi_0 = \text{const} \quad \phi \in \mathcal{C}^0(\mathbb{R}^3) \cap \mathcal{C}^2(\mathbb{R}^3 - BC - BV)$$

$$(2.8) \quad \max_{S_R} |\phi| \rightarrow 0 \quad R \rightarrow \infty$$

$$(2.9) \quad S_R = \{\text{Spherical surface, of radius } R \text{ and external unit normal } \mathbf{N}\}.$$

Let us develop equation (2.6) and integrate on the exterior domain  $\mathbb{R}^3 - C$ .

Taking into account (2.3)<sub>2</sub> and (2.3)<sub>3</sub> and then applying the Gauss theorem we obtain

$$(2.10) \quad \int_{\mathbb{R}^3 - C} \{f_{,D} + \nabla \phi\} \cdot \delta \mathbf{D} dV - \int_{BC} \phi \delta \mathbf{D} \cdot \mathbf{v} dS + \int_{BC} \phi_0 \delta \mathbf{D} \cdot \mathbf{v} dS - \int_{S_R} \phi \delta \mathbf{D} \cdot \mathbf{N} dS = 0.$$

Recalling the equations (2.3)<sub>3</sub> and (2.9), the last surface integral of (2.10) vanishes, as  $R \rightarrow \infty$ ; hence equation (2.10) becomes

$$(2.11) \quad \int_{\mathbb{R}^3 - C} (f_{,D} + \nabla \phi) \cdot \delta \mathbf{D} dV + \int_{BC} (\phi_0 - \phi) \delta \mathbf{D} \cdot \mathbf{v} dS = 0.$$

Consequently it follows that, by the arbitrary choice of  $\delta \mathbf{D}$

$$(2.12) \quad f_{,D} = -\nabla\phi \quad \phi = \phi_0 \quad \text{on } BC.$$

Since these properties characterize the electric potential, one can interpret  $f_{,D}$  as the electric field  $\mathbf{E}$  and write

$$(2.13) \quad \delta \mathcal{F} = \int_{\mathbb{R}^3} \mathbf{E} \cdot \delta \mathbf{D} \, dV.$$

Hence, at the isothermal equilibrium,  $f$  represents the electric energy density of the space (including dielectrics) with respect to the changing of the electric charges on the conductor.

### 3 - A functional depending on the electric field

Let the energy density be represented by  $\tilde{f}(\mathbf{E}, T)$ ,  $\mathbf{E}$  being the electric field and  $T$  the temperature; the latter is assumed to be constant and uniform as required in 2.

We also assume that  $\mathbf{E}$  and  $\delta \mathbf{E}$  satisfy the following conditions:

$$(3.1)_1 \quad \mathbf{E} = -\nabla\phi \quad \phi \in \mathcal{C}^0(\mathbb{R}^3) \cap \mathcal{C}^2(\mathbb{R}^3 - BC - BV) \cap \{\text{const in } C\}$$

$$(3.1)_2 \quad \left[ \frac{\partial\phi}{\partial n} \right] \neq 0 \quad \text{across } BV \quad \left[ \frac{\partial\phi}{\partial \nu} \right] \neq 0 \quad \text{across } BC$$

$$(3.1)_3 \quad \phi|_{\infty} \sim \frac{1}{R} \quad |\nabla\phi|_{\infty} \sim \frac{1}{R^2} \quad \delta\phi \equiv 0 \quad \text{on } BC.$$

$\tilde{f}$  must be restricted by the conditions

$$(3.1a) \quad |\tilde{f}| \leq C_1 |\mathbf{E}|^2 \quad |\tilde{f}_{,E}| \leq C_2 |\mathbf{E}| \quad C_1, C_2 \in \mathbb{R}^+ \quad \text{as } R \rightarrow \infty$$

$$(3.1b) \quad [\tilde{f}_{,E} \cdot \mathbf{n}] = 0 \quad \text{across } BV \quad [\tilde{f}_{,E} \cdot \boldsymbol{\nu}] = (-\tilde{f}_{,E})^- \cdot \boldsymbol{\nu} \quad \text{across } BC$$

$$(3.1c) \quad \tilde{f}_{,E} \equiv 0 \quad \text{in } \tilde{C}.$$

Let  $\tilde{\mathcal{F}}$  be defined as

$$(3.2) \quad \tilde{\mathcal{F}} = \int_{\mathbb{R}^3} \tilde{f}(\mathbf{E}) \, dV$$

and its first variation  $\delta\tilde{\mathcal{F}}$  as

$$(3.3) \quad \delta\tilde{\mathcal{F}} = \int_{\mathbb{R}^3} \tilde{f}_{,E} \cdot \delta\mathbf{E} \, dV.$$

At the equilibrium,  $\delta\tilde{\mathcal{F}}$  is required to vanish

$$(3.4) \quad \int_{\mathbb{R}^3} \tilde{f}_{,E} \cdot \delta\mathbf{E} \, dV = 0.$$

By developing the integrand of equation (3.4), we obtain

$$(3.5) \quad \tilde{f}_{,E} \cdot \delta\mathbf{E} = -\tilde{f}_{,E} \cdot \partial(\nabla\phi) = -\operatorname{div}(\tilde{f}_{,E} \delta\phi) + \delta\phi \operatorname{div} \tilde{f}_{,E}.$$

Then integrating (3.5) in  $\mathbb{R}^3 - C$ , applying the Gauss theorem and taking into account (3.1b)<sub>1</sub>, we obtain

$$(3.6) \quad - \int_{S_R} \tilde{f}_{,E} \cdot \mathbf{N} \delta\phi \, dS - \int_{BC} \tilde{f}_{,E} \cdot \mathbf{v} \delta\phi \, dS + \int_{\mathbb{R}^3 - C} \delta\phi \operatorname{div}(\tilde{f}_{,E}) \, dV = 0.$$

The first surface integral of (3.6) vanishes as  $R \rightarrow \infty$ , while the second surface integral vanishes by virtue of (3.1b)<sub>2</sub>. Hence equation (3.6) reduces to

$$(3.7) \quad \int_{\mathbb{R}^3 - C} \delta\phi \operatorname{div}(\tilde{f}_{,E}) \, dV = 0$$

and, for being  $\delta\phi$  arbitrary, the following equation is derived by (3.7), taking into account (3.1c)

$$(3.8) \quad \operatorname{div} \tilde{f}_{,E} = 0 \quad \text{in} \quad \mathbb{R}^3 - BC - BV.$$

We interpret the quantity  $-\tilde{f}_{,E}$  as the electric displacement  $\mathbf{D}$  and the expression (3.3) becomes

$$(3.10) \quad \delta\tilde{\mathcal{F}} = - \int_{\mathbb{R}^3} \mathbf{D} \cdot \delta\mathbf{E} \, dV.$$

Hence, at the isothermal equilibrium,  $\tilde{f}$  represents the electric energy density with respect to the changing of the electric potential, while the conductor is held to a fixed potential.

Remark. Since the electric potential does not vary on the surface of the conductor (by (3.1b)), the increment in the electric energy expressed by (3.10) is due to a variation of the electric charge on this surface; this charge must be supplied by an «external» source, which is understood to be connected to the conductor. Though the system is not energetically closed, such a situation reflects the experimental condition in which dielectrics usually work.

#### 4 - The electric enthalpy

Let us now consider the difference between the increment of field energy due to a conductor held to a constant potential and the increment of energy due to an isolated and pre-charged conductor. With reference to (3.10) and to (2.13) this difference is given by

$$(4.1) \quad \int_{\mathbb{R}^3} \{-\mathbf{D} \cdot \delta \mathbf{E} - \mathbf{E} \cdot \delta \mathbf{D}\} dV = - \int_{\mathbb{R}^3} \delta(\mathbf{E} \cdot \mathbf{D}) dV.$$

Hence

$$(4.2) \quad \delta \tilde{\mathcal{F}} = \delta \mathcal{F} - \int_{\mathbb{R}^3} \delta(\mathbf{E} \cdot \mathbf{D}) dV.$$

As this stage, it is possible to define the quantity

$$(4.3) \quad \hat{f}(\mathbf{E}, \mathbf{D}) = f(\mathbf{D}) - \mathbf{E} \cdot \mathbf{D}$$

which we call *the electric free enthalpy* in analogy to the thermodynamic enthalpy defined for fluids.

It is worth noticing that

$$(4.4) \quad \delta \hat{f}(\mathbf{E}, \mathbf{D}(\mathbf{E})) \equiv \delta \tilde{f}(\mathbf{E})$$

holds, with the following additional conditions

$$(4.5) \quad \mathbf{D} = -\tilde{f}_{,E} \quad \mathbf{E} = f_{,D}.$$

The field  $\mathbf{D}$  defined in  $\mathbb{R}^3$ , takes into account the properties of the dielectric (i.e. polarization) as well as those of the conductor (i.e. the electric free charge). In order to point out the specific role of the polarization charges, we introduce the

field  $\mathbf{P}$ , identically vanishing out of  $V$ . On assuming

$$(4.6) \quad \mathbf{D} = \begin{cases} \varepsilon_0 \mathbf{E} + \mathbf{P} & \text{in } V \\ \varepsilon_0 \mathbf{E} & \text{in } \mathbb{R}^3 - V \end{cases}$$

we rewrite equations (2.3) as follows

$$(4.7) \quad \varepsilon_0 \operatorname{div} \mathbf{E} = \begin{cases} -\operatorname{div} \mathbf{P} & \text{in } V \\ 0 & \text{in } \mathbb{R}^3 - V \end{cases}$$

$$\varepsilon_0 [\mathbf{E}] \cdot \mathbf{n} = -[\mathbf{P} \cdot \mathbf{n}] \quad \text{across } BV$$

and the expression  $-\mathbf{D} \cdot \delta \mathbf{E}$  becomes

$$(4.8) \quad -\mathbf{D} \cdot \delta \mathbf{E} = -\left\{ \delta \left( \frac{\varepsilon_0 \mathbf{E}^2}{2} \right) + \mathbf{P} \cdot \delta \mathbf{E} \right\}.$$

With reference to (4.7) we interpret the quantity  $-(\operatorname{div} \mathbf{P})$  as a polarization charge volume density and  $-[\mathbf{P} \cdot \mathbf{n}]$  as a polarization charge surface density. Hence we are able to compute the elementary work performed by the electric field in order to produce a distribution of the polarization charge in  $V$  and at  $BV$ . This work will be given by

$$(4.9) \quad \delta \mathcal{L} = - \int_V (\operatorname{div} \mathbf{P}) \delta \phi \, dV - \int_{BV} [\mathbf{P} \cdot \mathbf{n}] \delta \phi \, dS.$$

Bearing in mind that  $[\mathbf{P}] \cdot \mathbf{n} = (\mathbf{P}^+ - \mathbf{P}^-) \cdot \mathbf{n}$  and that  $\mathbf{P}^+ \equiv 0$  and manipulating the expression (4.9) as before, we obtain

$$(4.10) \quad \delta \mathcal{L} = \int_{BV} \{ -\mathbf{P}^+(-\mathbf{n}) - \mathbf{P}^- \cdot \mathbf{n} \} \cdot \delta \phi \, dS - \int_{BV} [\mathbf{P} \cdot \mathbf{n}] \delta \phi \, dS + \int_V \mathbf{P} \cdot \nabla(\delta \phi) \, \delta V$$

$$= - \int_V \mathbf{P} \cdot \delta \mathbf{E} \, dV.$$

Let us consider now the difference between the work  $\delta \mathcal{L}$  and the increment of energy of the pre-charged conductor. It is expressed by

$$(4.11) \quad -\delta \mathcal{L} - \mathbf{E} \cdot \delta \mathbf{D} = -\delta \left\{ \varepsilon_0 \frac{\mathbf{E}^2}{2} + \mathbf{P} \cdot \mathbf{E} \right\}$$

or equivalently

$$(4.12) \quad -\delta \mathcal{L} - \mathbf{E} \cdot \delta \mathbf{D} = -\delta \left\{ \varepsilon_0 \frac{\mathbf{E}^2}{2} - \mathbf{E} \cdot \mathbf{D} \right\}.$$

If we put

$$(4.13) \quad h(\mathbf{E}, \mathbf{P}) = -\left( \varepsilon_0 \frac{\mathbf{E}^2}{2} + \mathbf{P} \cdot \mathbf{E} \right) + w(\mathbf{P})$$

with  $\mathbf{E}$  satisfying the relations (3.1), we obtain

$$(4.14) \quad \delta h|_{\mathbf{P}=\text{const}} \equiv \delta \tilde{f}$$

hence,  $h$  has to be considered as an extension of  $\tilde{f}$ , as well as of  $\hat{f}$ , according to (4.4). Now let us define  $H$  as

$$(4.15) \quad H = \int_{\mathbb{R}^3} h \, dV$$

and its variation  $\delta H$  as

$$(4.16) \quad \delta H = \int_{\mathbb{R}^3} \delta h(\mathbf{E}, \mathbf{P}) \, dV.$$

We assume that at equilibrium

$$(4.17) \quad \delta H = 0.$$

However, before developing the eq. (4.17), we should examine closely the electric field  $\mathbf{E}$ .  $\mathbf{E}$  has to be considered as a combined field in the sense that it is due to the free charges of the conductor and to the polarization charge of the dielectric. Thus it can be written in the form

$$(4.18) \quad \begin{aligned} \mathbf{E} &= \mathbf{E}_0 + \mathbf{E}_1 & \mathbf{D} &= \varepsilon_0 \mathbf{E} + \mathbf{P} \\ \mathbf{E}_0 &= -\nabla \phi_0 & \mathbf{E}_1 &= -\nabla \phi_1 & \phi &= \phi_0 + \phi_1 \end{aligned}$$

where  $\mathbf{E}_0$  represents the «external» field due to the free charges on the conductor, while in the absence of the dielectric.



$\mathbf{E}_0$  and  $\delta\mathbf{E}_0$  satisfy the following conditions

$$(4.19) \quad \begin{aligned} \phi_0 \in \mathcal{C}^0(\mathbb{R}^3) \cap \mathcal{C}^2(\mathbb{R}^3 - BC) \cap \{\text{const in } C\} & \quad \delta \int_{\mathbb{R}^3} \left(\frac{\nabla\phi_0}{2}\right)^2 dV = 0 \\ \delta\phi_0 \equiv 0 \quad \text{on } BC & \quad \phi_0|_{\infty} \rightarrow 0 & \quad \left|\frac{\partial\phi}{\partial R}\right|_{\infty} \sim \frac{1}{R^2}. \end{aligned}$$

By (4.19)<sub>2</sub> the field equation

$$(4.20) \quad \Delta\phi_0 = 0 \quad \text{in } \mathbb{R}^3 - BC$$

and the condition

$$(4.20a) \quad [\mathbf{E}_0 \cdot \mathbf{v}] \neq 0 \quad \text{across } BC$$

are derived.

It is worth noting that eqs. (4.19) and (4.20) express the physical situation in which the potential  $\phi_0$ , generated by a given charge  $q_0 = -\varepsilon_0 \int_{BC} \left(\frac{\partial\phi_0}{\partial n}\right)^- ds$ , is held constant along with the charge itself.

The variational principle (4.17) modifies as follows

$$(4.21) \quad \delta H - \lambda \delta \int_{\mathbb{R}^3} \frac{\mathbf{E}_0^2}{2} dV = 0$$

where  $\lambda$  is a Lagrange multiplier.

By developing (4.21), integrating inside and outside  $V$  and applying the Gauss theorem, an expression is obtained in which  $\delta\phi_0$  and  $\delta\phi_1$  vary independently. Henceforth, the result is

$$(4.22) \quad \begin{aligned} \int_V \{w_{,P} - (\mathbf{E}_0 + \mathbf{E}_1)\} \cdot \delta\mathbf{P} dV - \int_{\mathbb{R}^3} (\delta\phi_1 + \delta\phi_0) \operatorname{div}\{\varepsilon_0 \mathbf{E}_1 + \mathbf{P}\} dV \\ + \int_{BC} \{-\varepsilon_0(\mathbf{E}_0 + \mathbf{E}_1)\}^- \cdot \mathbf{v}(\delta\phi_1 + \delta\phi_0) dS + \int_{BV} [\varepsilon_0 \mathbf{E}_1 + \mathbf{P}] \cdot \mathbf{n}(\delta\phi_1 + \delta\phi_0) dS \\ + \int_{S_R} \varepsilon_0(\mathbf{E}_0 + \mathbf{E}_1) \cdot \mathbf{N}(\delta\phi_1 + \delta\phi_0) dS = 0. \end{aligned}$$

By the conditions (3.1b)<sub>2</sub> and (4.19)<sub>3</sub> the surface integral on  $BC$  vanishes; so does the surface integral on  $S_R$  as  $R \rightarrow \infty$ , because of (3.1)<sub>5,6</sub> and (4.19)<sub>4</sub>. Hence

(4.22) reduces to

$$(4.23) \quad \int_V \{w_{,P} - (\mathbf{E}_0 + \mathbf{E}_1)\} \cdot \delta \mathbf{P} \, dV + \int_{\mathbb{R}^3} (\delta \phi_1 + \delta \phi_0) \operatorname{div} \{ \varepsilon_0 \mathbf{E}_1 + \mathbf{P} \} \, dV \\ + \int_{BV} [\varepsilon_0 \mathbf{E}_1 + \mathbf{P}] \cdot \mathbf{n} (\delta \phi_1 + \delta \phi_0) \, dS = 0.$$

Acknowledging that  $\delta \mathbf{P}$ ,  $\delta \phi_0$  and  $\delta \phi_1$  are arbitrary, the following Euler-Lagrange equations hold

$$(4.24) \quad \begin{array}{ll} w_{,P} - \mathbf{E}_0 - \mathbf{E}_1 = 0 & \text{in } V \\ -\operatorname{div} P & \text{in } V \\ \varepsilon_0 \operatorname{div} \mathbf{E}_1 = & \\ 0 & \text{in } \mathbb{R}^3 - V \\ \varepsilon_0 [\mathbf{E}_1] \cdot \mathbf{n} = \mathbf{P}^- \cdot \mathbf{n} & \text{across } BV. \end{array}$$

The principle which is given by (4.17) can be stated in a different, though equivalent way

$$(4.25) \quad \delta H_1 = \int_V \mathbf{E}_0 \cdot \delta \mathbf{P} \, dV$$

where

$$H_1 = \int_{\mathbb{R}^3} h_1(\mathbf{E}_1, \mathbf{P}) \, dV$$

Notice that the variational principle expressed by (4.25) does not require  $\mathbf{E}_0$  to be characterized by (4.18) and (4.19).

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### Abstract

*Starting from the classical expression of the electrostatic energy it is possible to define a new functional called the electric free enthalpy for dielectric bodies. Toupin's variational principle can be derived from this enthalpy.*

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