

RHEOLOGICAL THEORY OF ANISOTROPIC MEDIA

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Résumé

The tensorial formulation of the mathematical theory of anisotropic rheological bodies is presented taking into account thermal phenomena. The present theory includes, in special cases, those obtained by the author,¹⁾ Oshida,²⁾ Sips,³⁾ Frenkel⁴⁾ and Kneser,⁵⁾ respectively. The expression of macroscopic internal energy of the media is somewhat different from the one given by Meixner.⁶⁾ In an isothermal process, the effect of initial stress of solid-like body is also considered being based on Sakadi's formalism.⁷⁾

I. Preliminaries and Notations

In this paper the author presents the generalized theory of anisotropic media with rheological character, so as to include author's theory¹⁾ in 1953. The anisotropic thermal conduction and expansion are also taken into account.

Notations

x_i : rectangular coordinates, ($i = 1, 2, 3$)

ξ_i : components of displacement, being infinitesimal quantities of first order, ($i = 1, 2, 3$)

$u_i = \frac{\partial \xi_i}{\partial t}$: components of velocity, ($i = 1, 2, 3$)

$\sigma_{ij} = \frac{1}{2} \left(\frac{\partial \xi_j}{\partial x_i} + \frac{\partial \xi_i}{\partial x_j} \right)$: components of strain tensor, being of first order, ($i, j = 1, 2, 3$)

$\bar{\omega}_{ij} = \frac{1}{2} \left(\frac{\partial \xi_j}{\partial x_i} - \frac{\partial \xi_i}{\partial x_j} \right)$: components of rotation of displacement, being also of first order, ($i, j = 1, 2, 3$)

A_{ij} : components of stress tensor, being free from initial stress A_{ij}^0 ; and being first order infinitesimal quantities for solid-like body, and not for liquid-like one.

$A_{ij}^0(\mathbf{x}')$: components of initial stress of finite magnitude at initial coordinate point \mathbf{x}' , components being x'_i , ($i, j = 1, 2, 3$)

t : time,

$\rho^0 + \rho$: density,

ρ^0 : density at initial state,

$p^0 + p$: pressure for solid-like body,

p^0 : pressure at initial state for liquid-like body,

$T^0 + T$: temperature,

T^0 : initial temperature,

ρ, p , and T are all infinitesimal quantities of first order,

U : internal energy per unit mass in the macroscopic sense,

q_i : components of heat current vector, $= -\kappa_{ij} \frac{\partial T}{\partial x_j}$,

κ_{ij} : coefficients of thermal conduction, ($i, j = 1, 2, 3$)

k^0 : static volume modulus for liquid-like body,

$a_{ijkl}^{(r)}$: partial coefficients of elasticity, ($i, j, k, l = 1, 2, 3$; and $r = 1, 2, \dots, R$)

$a_{ijkl}^{(r)l}$: partial coefficients of viscosity, $= \tau_i^{(r)} a_{ijkl}^{(r)}$, ($i, j, k, l = 1, 2, 3$; and $r = 1, 2, \dots, R$)

$\alpha_{ij}^{(n)}$: coefficients corresponding to partial thermal expansion, ($i, j = 1, 2, 3$; and $n = 1, 2, \dots, N$)

$f^{(r)}$, $g^{(m)}$, $\psi^{(l)}$, $\phi^{(s)}$, $\bar{\psi}^{(m)}$, $\bar{\phi}^{(k)}$: hysteresis functions, ($r = 1, 2, \dots, R$; $n = 1, 2, \dots, N$; $l = 1, 2, \dots, L$; $m = 1, 2, \dots, M$; $k = 1, 2, \dots, K$)

$\tau_1^{(r)}$, $\tau_2^{(m)}$, $\tau_3^{(l)}$, $\tau_4^{(s)}$, $\tau_5^{(m)}$, and $\tau_6^{(k)}$: relaxation times,

c^0 : static specific heat at constant volume,

$c^{(m)}$: partial specific heat at constant volume, ($m = 1, 2, \dots, M$)

ω : circular frequency,

v : complex velocity of wave,

n_j^0 : direction cosines of the outward normal to the boundary of the unstrained state for solid-like body. ($j = 1, 2, 3$)

For the abbreviation we shall write the differential operator with respect to the time t as follows:

$$D \equiv \frac{d}{dt} \equiv \frac{\partial}{\partial t} + \frac{\partial \xi_i}{\partial t} \frac{\partial}{\partial x_i} = \frac{\partial}{\partial t}.$$

As usual in the tensorial notations, one should sum up over double indices.

II. Equations of State

In rheological media such as visco-elastic bodies, if the volume dilatation is produced suddenly and kept constant (i.e. kept in an isostrained state), normal stress decays after a sufficiently long time and finally vanishes. While, in liquids, pressure also decays after a sufficiently long time, and does not vanish but finally approaches to a constant value, still remaining finite. This is the main difference between solid-like (visco-elastic) and liquid-like materials. In the following we shall be chiefly concerned with the equations of state for liquid-like and solid-like bodies, respectively.

The equations of state for an anisotropic liquid-like body are written, in linear approximation, as follows:

$$A_{ij}(t) = \{ -p^0 + k^0 \sigma_{kk}(t) \} \cdot \delta_{ij} + \sum_{r=1}^R a_{ijkl}^{(r)*} \sigma_{kl}(t) - \sum_{n=1}^N \alpha_{ij}^{(n)*} T(t), \quad (1)$$

$$\left. \begin{aligned} a_{ijkl}^{(r)*} \sigma_{kl}(t) &= a_{ijkl}^{(r)} \left\{ \sigma_{kl}(t) - \int_0^\infty f^{(r)}(t') \cdot \sigma_{kl}(t-t') dt' \right\}, \\ \alpha_{ij}^{(n)*} T(t) &= \alpha_{ij}^{(n)} \left\{ T(t) - \int_0^\infty g^{(n)}(t') \cdot T(t-t') dt' \right\}, \end{aligned} \right\} \quad (2)$$

$$\left. \begin{aligned} a_{ijkl}^{(r)} &= a_{klij}^{(r)} = a_{ikjl}^{(r)}, \\ \alpha_{ij}^{(n)} &= \alpha_{ji}^{(n)}, \\ (i, j, k, l) &= 1, 2, 3 \end{aligned} \right\} \quad (3)$$

with initial pressure p^0 .

The second terms in (1) accompanied with hysteresis functions come from the elastic relaxation of the media, while the last express the relaxation of thermal expansion.

While, we can write, in analogous form, the equations of state for solid-like body with a slight modification of the rheological coefficients $a_{ijkl}^{(r)}$ and $\alpha_{ij}^{(n)}$.

$$A_{ij}(t) = \sum_{r=1}^R a_{ijkl}^{(r)*} \sigma_{kl}(t) - \sum_{n=1}^N \alpha_{ij}^{(n)*} T(t). \quad (1')$$

The summations in (1) and (1') can be replaced by Stieltjes integrations when the distribution of hysteresis functions is discrete in some region and continuous in the other.

Equations (1) are reduced to (4) in an isotropic body,

$$\begin{aligned} A_{ij}(t) &= \{ -p^0 + k^0 \sigma_{kk}(t) \} \cdot \delta_{ij} + \left\{ \sum_{l=1}^L \lambda^{(l)*} - \frac{2}{3} \sum_{s=1}^S \mu^{(s)*} \right\} \sigma_{kk}(t) \cdot \delta_{ij} \\ &+ 2 \sum_{s=1}^S \mu^{(s)*} \sigma_{ij}(t) - \sum_{n=1}^N \alpha^{(n)*} T(t) \cdot \delta_{ij}, \\ \lambda^{(l)*} \sigma_{kk}(t) &= \lambda^{(l)} \left\{ \sigma_{kk}(t) - \int_0^{\infty} \phi^{(l)}(t') \sigma_{kk}(t-t') dt' \right\}, \\ \mu^{(s)*} \sigma_{ij}(t) &= \mu^{(s)} \left\{ \sigma_{ij}(t) - \int_0^{\infty} \phi^{(s)}(t') \sigma_{ij}(t-t') dt' \right\}, \\ \alpha^{(n)*} T(t) &= \alpha^{(n)} \left\{ T(t) - \int_0^{\infty} g^{(n)}(t') T(t-t') dt' \right\}, \\ \alpha_{ij}^{(n)} &= \alpha^{(n)} \delta_{ij}, \end{aligned} \quad (4)$$

with partial coefficients corresponding to Lamé's constants $\lambda^{(l)}$ and $\mu^{(s)}$, and partial coefficients corresponding to isotropic thermal expansion $\alpha^{(n)}$.

The expression (4) was already presented by the author¹⁾ in 1953, and, in case $\alpha^{(n)} = 0$, corresponds to the expression given by Oshida²⁾ and Sips.³⁾ Further, if we put $L=1$ and $S=2$, (4) is proved to be equivalent to the equation of state presented by Frenkel-Obratzov.⁴⁾

In a solid-like body, equations (1') are reduced to (4') for the isotropic case,

$$\begin{aligned} A_{ij}(t) &= \left\{ \sum_{l=1}^L \lambda^{(l)*} - \frac{2}{3} \sum_{s=1}^S \mu^{(s)*} \right\} \sigma_{kk}(t) \cdot \delta_{ij} \\ &+ 2 \sum_{s=1}^S \mu^{(s)*} \sigma_{ij}(t) - \sum_{n=1}^N \alpha^{(n)*} T(t) \cdot \delta_{ij}, \end{aligned} \quad (4')$$

with a slight modification of λ^* , μ^* , and α^* .

For the sake of simplicity, we shall consider in (1), (1'), (4), and (4') the Maxwellian relaxations:

$$\left. \begin{aligned} f^{(r)} &= \frac{1}{\tau_1^{(r)}} e^{-t'/\tau_1^{(r)}}, \\ g^{(n)} &= \frac{1}{\tau_2^{(n)}} e^{-t'/\tau_2^{(n)}}, \\ \phi^{(l)}(t') &= \frac{1}{\tau_3^{(l)}} e^{-t'/\tau_3^{(l)}}, \\ \phi^{(s)}(t') &= \frac{1}{\tau_4^{(s)}} e^{-t'/\tau_4^{(s)}}, \end{aligned} \right\} \quad (5)$$

with τ 's all positive.

Then, in linear approximation, all the operators with superscript *, can be written symbolically by the integro-differential operators:

$$\left. \begin{aligned} a_{ijkl}^{(r)*} &= \frac{a_{ijkl}^{(r)} \tau_1^{(r)} D}{1 + \tau_1^{(r)} D} = \frac{a_{ijkl}^{(r)'} D}{1 + \tau_1^{(r)} D}, \\ \alpha_{ij}^{(n)*} &= \frac{\alpha_{ij}^{(n)} \tau_2^{(n)} D}{1 + \tau_2^{(n)} D}, \\ \lambda^{(l)*} &= \frac{\lambda^{(l)} \tau_3^{(l)} D}{1 + \tau_3^{(l)} D} = \frac{\lambda^{(l)'} D}{1 + \tau_3^{(l)} D}, \\ \mu^{(s)*} &= \frac{\mu^{(s)} \tau_4^{(s)} D}{1 + \tau_4^{(s)} D} = \frac{\mu^{(s)'} D}{1 + \tau_4^{(s)} D}, \end{aligned} \right\} \quad (6)$$

with $D = \frac{\partial}{\partial t}$, where $a_{ijkl}^{(r)'}$, $\lambda^{(l)'}$, and $\mu^{(s)'}$, correspond to the partial coefficients of viscosity.

The equations of motion for anisotropic media are written under the consideration of (1) or (1'):

$$\rho^0 \frac{\partial^2 \xi_i}{\partial t^2} = \frac{\partial A_{ij}}{\partial x_j} + \rho^0 X_i, \quad (7)$$

with external body force X_i . ($i=1, 2, 3$)

The change of density, if necessary, is given by the equation of continuity.

III. Conservation of Energy

The energy equation for rheological media inside the volume enclosed by any surface F is

$$\frac{dK}{dt} + \frac{d}{dt} \int \rho U dV = W_1 + W_2 + Q, \quad (8)$$

where the integration extends throughout the volume enclosed by F . K is the total kinetic energy, U the internal energy per unit mass in the macroscopic sense, W_1 the rate at which work is done on the media by the surface traction at F , W_2 the rate at which work is done by the body force, and Q the rate at which heat is conducted across F into the media inside F .

The kinetic energy is given by

$$K = \frac{1}{2} \int \rho^0 u_i^2 dV, \quad (9)$$

By using Green's theorem,

$$W_1 = - \int A_{\nu j} u_j dF = \int \frac{\partial}{\partial x_j} (u_j A_{ij}) dV,$$

(ν is the inward normal to the surface F)

which after introduction of equations of motion (7) is reduced to

$$W_1 = \int A_{ij} \frac{\partial u_j}{\partial x_i} dV + \int \rho^0 \left(u_j \frac{\partial^2 \xi_j}{\partial t^2} - u_j X_j \right) dV. \quad (10)$$

When no body force exists,

$$X_i = 0 \quad \text{and} \quad W_2 = 0. \quad (11)$$

Heat inflow is

$$Q = - \int \frac{\partial q_i}{\partial x_i} dV = \int \frac{\partial}{\partial x_i} \left(\kappa_{ij} \frac{\partial T}{\partial x_j} \right) dV, \quad (12)$$

with the coefficients of thermal conduction $\kappa_{ij} = \kappa_{ji}$.

The combination of (8), (9), (10), (11), and (12), gives the energy equation in the media, in linear approximation,

$$\rho^0 \frac{\partial U}{\partial t} = A_{ij} \frac{\partial \sigma_{ij}}{\partial t} + \kappa_{ij} \frac{\partial^2 T}{\partial x_i \partial x_j}. \quad (13)$$

Here arises a question. What is the expression of the internal energy, then? The answer is as follows. After we have taken σ_{ij} and T as independent variables of thermodynamical state as already done in (1) and (1'), we suppose that it is natural to take

$$dU = dU_\sigma + dU_T = \left(\frac{\partial U}{\partial T} \right)_\sigma dT + \left(\frac{\partial U}{\partial \sigma_{ij}} \right)_T d\sigma_{ij}, \quad (14)$$

where suffixes σ and T outside the brackets mean the isostrained and the isothermal states respectively.

Then, as usual in *thermostatistics*, we can interpret that $\left(\frac{\partial U}{\partial T} \right)_\sigma$ is specific heat at constant volume, which we shall designate as c , and we shall write $\left(\frac{\partial U}{\partial \sigma_{ij}} \right)_T$ as b_{ij} , which measures the heat effect of deformation of the body at isothermal state. In rheological media, however, we shall rather put instead of c and b_{ij} ,

$$c^* = c^0 + \sum_{m=1}^M c^{(m)*}, \quad c^{(m)*} T = c^{(m)} \left\{ T(t) - \int_0^{\infty} \bar{\psi}^{(m)}(t') T(t-t') dt' \right\}, \quad (15)$$

$$b_{ij}^* = \epsilon_{ij}^0 + \sum_{k=1}^K \epsilon_{ij}^{(k)*}, \quad \epsilon_{ji}^{(k)*} \sigma_{ij} = \epsilon_{ij}^{(k)} \left\{ \sigma_{ij}(t) - \int_0^{\infty} \bar{\phi}^{(k)}(t') \sigma_{ij}(t-t') dt' \right\}, \quad (16)$$

as presented in the previous paper.¹⁾

Now, we shall consider the meaning of (15) and (16), taking merely their first terms. If the media undergo the infinitesimal deformation adiabatically as we see from (14), the expression $U_{\text{adiabatic}} = c^0 T + \epsilon_{ij}^0 \sigma_{ij}$ is the work done by the surface force and is equal to $\frac{\partial}{\partial x_j} \left(A_{ij} \frac{\partial \xi_i}{\partial t} \right) = \{ -p^0 \delta_{ij} + (\text{first order terms}) \} \cdot \frac{\partial \sigma_{ij}}{\partial t} + (\text{second order terms})$, the right-hand side of which is a small infinitesimal quantity of first order for liquids, and is of second order for visco-elastic bodies, according to the finite value of p^0 or the vanishing value of p^0 .

In those cases, if we put $U_{\text{adiabatic}} + \frac{p^0}{\rho^0} \frac{\partial \sigma_{kk}}{\partial t} + (\text{second order terms}) = 0$, then the change of temperature is obtained as

$$T = \frac{-1}{c^0} \left\{ \epsilon_{ij}^0 \sigma_{ij} + \frac{p^0}{\rho^0} \frac{\partial \sigma_{kk}}{\partial t} + (\text{second order terms}) \right\}.$$

On the other hand, if the media change their state isothermally always under the quasistatic process, the change in free energy can be expressed as mechanical work and is $\delta U - T \delta S$, S being the entropy of the system. Thus, when p^0 is finite, this quantity is of first order. Accordingly the entropy term (bounded energy) is also of first order.

While, when p^0 vanishes, $\delta U - T \delta S$ is of second order and $U_{\text{isothermal}} = \epsilon_{ij}^0 \sigma_{ij}$ is of first order. Accordingly, the bounded energy is of first order.

If we take the Maxwellian relaxational process in (15), relaxation times being $\tau_5^{(m)}$, and put $c_{\infty} = c^0 + \sum_{m=1}^M c^{(m)}$ with $M=1$, then the expression of internal energy is shown to be formally equivalent to the final expression presented by Kneser.⁵⁾

IV. Applications and Discussions

The simultaneous equations (7) and (13) with (1), (14), (15), and (16); or with (1'), (14), (15), and (16), are generalized fundamental equations of motion and of conservation of energy for anisotropic rheological media. These, when simplified, can be applicable to the theories of surface rheology of mono-molecular layer of macromolecules such as presented by Tachibana-Inokuchi,⁶⁾ Oka,⁹⁾ Oka-Satô¹⁰⁾ etc. and also may have some contributions to acoustic (ultrasonic) birefringence in high polymer solutions and highpolymeric substances. The rheology of solid crystal-like materials is also involved in the present theory, *mutatis mutandis*.

When the process occurring in the media is isothermal, i.e. $\delta T = 0$, (7) with (1) can be simplified, and (13) shall be omitted. For the sake of simplicity we shall take the Maxwellian relaxational process and consider the progressing sinusoidal wave with circular frequency ω . Then, as is well known, owing to the symmetric property of the matrix $a_{ij}^{(r)}$, we see that there exist three waves with different velocities, whose planes of oscillation are perpendicular to each other. Moreover

these waves show the dispersion and the dissipation because of the existence of the complex elastic moduli :

$$a_{ijkl}^{(r)*} = \frac{i\omega a_{ijkl}^{(r)'}}{1 + i\omega\tau_1^{(r)}}.$$

When the initial stress of finite magnitude is distributed in the solid-like media, we have the equations of motion in the isothermal process according to Sakadi's formalism⁷⁾

$$\begin{aligned} \rho^0 \frac{\partial^2 \xi_i}{\partial t^2} = & -\sigma_{jk} \frac{\partial A_{ij}^0}{\partial x_k} + \frac{\partial \tilde{\omega}_{jk}}{\partial x_k} A_{ij}^0 + \frac{\partial \tilde{\omega}_{ji}}{\partial x_k} A_{kj}^0 + \frac{\partial A_{ij}'}{\partial x_j} \\ & + \rho^0 (X_i' - \sigma_{kk} X_i^0 + \tilde{\omega}_{ij} X_j^0), \end{aligned} \quad (17)$$

where

$$A_{ij}' = A_{ij} \text{ in } (1'),$$

$X_i^0(\mathbf{x}')$: body force per unit mass at initial coordinate point \mathbf{x}' ,

$X_i(\mathbf{x})$: body force at \mathbf{x} in a strained state,

At the reference state in equilibrium with initial stress,

$$\rho_{(\mathbf{x})}^0 X_i^0(\mathbf{x}) + \frac{\partial A_{ij}^0(\mathbf{x})}{\partial x_j} = 0. \quad (18)$$

Here the initial stress is expressed as $A_{ij}^0(\mathbf{x}')$ at initial coordinate point \mathbf{x}' , $\rho_{(\mathbf{x}')}$ being the initial density.

Boundary conditions state

$$F_i - F_i^0 = -\sigma_{kj} n_k^0 A_{ij} + \sigma_{kl} n_k^0 n_l^0 F_i^0 + \tilde{\omega}_{ki} F_k^0 + n_j^0 A_{ij}', \quad (19)$$

with initial conditions at the equilibrium state :

$$F_i^0 = n_j^0 A_{ij}', \quad (20)$$

where n_j^0 are direction cosines of the outward normal to the boundary of the unstrained state; F_i^0 and F_i surface tractions at initial state and at strained state, respectively.

Owing to the existence of the initial stress, in some cases progressing waves such as surface waves actually damp, and moreover the fugitive character of rheological stress A_{ij}' , makes the damping more rapid. Equations (17) must be quite useful in the case of the practical problems occurring in many engineering fields.

As a special case, with isotropic stress (4'), we have from (17), by using complex Lamé's moduli $\lambda^{(s)*}$ and $\mu^{(s)*}$, complex velocities of three waves for infinite solid-like media :

$$\begin{aligned} v_1 &= \sqrt{\frac{\mu_1}{\rho^0}}, & \text{with } \mu_1 &= \sum_{s=1}^8 \mu^{(s)*} - \frac{1}{2} (B_{11}^0 - B_{33}^0), \\ v_2 &= \sqrt{\frac{\mu_2}{\rho^0}}, & \text{with } \mu_2 &= \sum_{s=1}^8 \mu^{(s)*} - \frac{1}{2} (B_{22}^0 - B_{33}^0), \end{aligned}$$

and

$$v_3 = \sqrt{\frac{\lambda_1}{\rho^0}}, \quad \text{with } \lambda_1 = \sum_{l=1}^L \lambda^{(l)*} + 2 \sum_{s=1}^S \mu^{(s)*},$$

if we take $B_{12}^0 = 0$ after suitable transformation, B_{ij}^0 being the transformed initial stress, and X_i^0 and X_i being put equal to zero.

Starting from (7) with (1'), or with (1), no temperature terms and no body force being taken into account, we can see that the three waves, one purely longitudinal and two purely transverse, are obtained after suitable orthogonal transformation. Calculation of the transformed elements of the matrix $a_{ijkl}^{(r)*}$ can be quite analogously carried out as in the case of elastic waves in crystals.^{(11) (12)} Once the velocities of these three waves are determined, we can obtain specific heat of anisotropic media, when they are solid-like or liquid-like, according to Debye's theory and Oomori's treatment⁽¹³⁾ of the dissipative wave.

In the analogous line of consideration the surface energy of liquids can be also calculated by using the relaxationally elastic surface waves, following to the method presented by Frenkel.⁽¹⁴⁾

When no initial stress exists, the behaviour of the mechanical and thermodynamical system hitherto considered, is completely described by the system of equations (7) and (13), with (1), (14), (15), and (16); or with (1'), (14), (15), and (16). This macroscopic system, however, is not itself *closed* in the sense of thermostatics. Here arises a question how we can consider the system in itself closed from the view-point of irreversible theory of thermodynamics. Actually the terms of heat conduction, fugitive specific heat, bulk and shearing viscosities, and thermal expansion of fugitive nature, are present in our fundamental equations, so the dissipation of energy and the entropy production do actually occur in our *open system*. But, if we take some *subsystems* characterised by some extra *inner variables*, including those such as chemical potentials and some measures of excited states of constituent molecules and atoms, we may set up a *closed system* as a whole. Meixner⁽⁶⁾ considered affinities in the expression of internal energy and arrived at the hysteresis functions in the stress-strain relation after eliminating the linearly decaying inner variables. He, however, did not propose any equation involving thermal conduction. So, the expressions of internal energy and of energy equation in the present paper are different from his in these points.

Taking into consideration some subsystems with a sufficient number of inner variables and of energy equations, and those of entropy production in the individual subsystems, we may be in the goal of the closed system in the sense of irreversible theory of thermodynamics, after eliminating a sufficient number of inner variables, which do not fall under our direct observation. Thus, (7) and (13) with (1), (14), (15), and (16), although still remained in the scope of an open system, should be reconsidered, if necessary, from the view-point of a consistent irreversible theory of thermodynamics.

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