FINITE COMPRESSION OF SOLIDS—
SECOND ORDER THERMOELASTIC ANALYSIS

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Abstract—In order to adequately reproduce the thermo-mechanical behaviour of metals under finite volume reduction and large temperature increase, it is necessary to use the second order thermo-elastic analysis, i.e. to use the free energy function expanded up to the third degree in strain and temperature. Determination of the corresponding material constants is given and the resulting expressions are applied to the adiabatic compression of aluminum. The results are compared with those obtained from the first order theory.

1. INTRODUCTION

The behaviour of metals subjected to high pressures, such as those produced by explosive loadings, has long been a subject of special interest[1–5]. The volume reduction caused by pressures in the range of 10–30 GPa can be of the order of 25%. Such a large (finite) volume reduction is followed by a rise in temperature of about 100–200 K, depending on material and applied pressure. In this temperature range the thermal characteristics of materials (like the coefficients of thermal expansion and specific heat) are not constant and in order to reproduce their variation adequately, it is necessary to include the terms of higher order than second in the expression for the free energy. Also, large geometry (volume) changes demand the use of finite strain which therefore, together with the previous, leads to nonlinear thermo-elastic analysis. In this paper we consider its second order approximation. The determination of the material constants which appear in the expansion of the free energy function is done by using the experimental data and results from the solid state physics theory which accurately represents the thermal effects of many metals. On the basis of developed expressions, some specific results are obtained in the case of aluminum which is rapidly (adiabatically) compressed. The comparison with the results obtained by using the first order theory is also given.

2. PRELIMINARY THERMODYNAMIC ANALYSIS

We consider a homogeneous material which is subjected to high uniform pressure. Many tests have shown that such a material can sustain enormous pressures and still remain elastic. According to the first law of thermodynamics the increase of the specific internal energy (or internal energy per unit mass of the system) is produced by the corresponding work done and heat input to the system (per unit mass), i.e.

$$\mathrm{d}u = -\frac{1}{\rho V} p \,\mathrm{d}V + T \,\mathrm{d}\eta, \tag{2.1}$$

where $\mathrm{d}\omega = -(1/\rho) p \,(\mathrm{d}V/V)$ is the specific work associated with the change of volume $\mathrm{d}V$ (p is the pressure, ρ is the density and V is the volume), while the specific heat input is $\mathrm{d}q = T \,\mathrm{d}\eta$, where T is the absolute temperature and η is the specific entropy, since hydrostatic compression is elastic (recoverable). Introducing the Helmholtz free energy (per unit mass) $\psi = u - \eta T$, (2.1) can be rewritten as

$$\mathrm{d}\psi = -\frac{1}{\rho V} p \,\mathrm{d}V - \eta \,\mathrm{d}T, \tag{2.2}$$

which is the form of the first law of thermodynamics that is convenient in the subsequent analysis, where we use the Helmholtz free energy as a potential function. Indeed, since we are dealing with hydrostatic compression, which is purely (thermo-)elastic deformation, the free energy is a function of only two state variables, which completely define the state, the volume V and temperature T, i.e. $\psi = \psi(V, T)$. Introducing further the free energy per unit initial volume $\tilde{\psi} = \tilde{\psi}(V, T)$ then

$$\psi = \frac{1}{\rho_0} \tilde{\psi}(V, T), \tag{2.3}$$

where ρ_0 is the initial density of the body. Applying the differential on (2.3), we have

$$\mathrm{d}\psi = \frac{1}{\rho_0} \left(\frac{\partial \tilde{\psi}}{\partial V} \, \mathrm{d}V + \frac{\partial \tilde{\psi}}{\partial T} \, \mathrm{d}T \right), \tag{2.4}$$

so that comparison with (2.2) gives the usual expressions for the pressure and entropy:

$$p = -\frac{\rho}{\rho_0} V \frac{\partial \tilde{\psi}}{\partial V}, \quad \eta = -\frac{1}{\rho_0} \frac{\partial \tilde{\psi}}{\partial T}. \tag{2.5}$$

However, in dealing with finite volume compressions it is convenient to use the logarithmic strain $\varepsilon = \ln (V/V_0)$ as a measure of volume change[5, 6], and if we also introduce the relative (dimensionless) temperature change $\theta = (T - T_0)/T_0$, where V_0 and T_0 are the initial volume and temperature, then we have $\tilde{\psi} = \tilde{\psi}(V, T) \equiv \hat{\psi}(\varepsilon, \theta)$, and by the chain rule differentiation we obtain from (2.5):

$$p = -\mathrm{e}^{-\varepsilon} \frac{\partial \hat{\psi}}{\partial \varepsilon}, \quad \eta = -\frac{1}{\rho_0 T_0} \frac{\partial \hat{\psi}}{\partial \theta}. \tag{2.6}$$

The first of expressions (2.6) was utilized in [6] in establishing the isothermal pressure-volume relation at high pressures, where $\hat{\psi}$ (there interpreted as a strain energy) is approximated by a second and third order polynomial in strain ε. The results were compared with Murnaghan's results[2] in the case of Bridgman's experimental data for the metal sodium[1], and the differences were critically examined. In the next section we shall analyse and establish the polynomial representation (expression) for the free energy function which is suitable in the case of large volume and temperature changes.

3. EXPRESSION FOR THE FREE ENERGY FUNCTION AND DETERMINATION OF MATERIAL CONSTANTS

The classical linear thermoelasticity equations are obtained by taking the free energy function to be a polynomial up to the second degree in strains and temperature. In the case of hydrostatic loading this means that $\hat{\psi}$ is of the form

$$\hat{\psi}(\varepsilon, \theta) = a_0 + a_1\varepsilon + a_2\varepsilon^2 + b_1\varepsilon\theta + c_1\theta + c_2\theta^2, \tag{3.1}$$

where $a_0 - c_2$ are constants. The free energy expansion (3.1) leads to thermoelastic relations which are appropriate in the range of sufficiently small pressures and temperature increases. If we are, however, dealing with the pressures large enough to produce finite volume changes (say, 25%) and large temperature changes (say, 100–200 K, such as those produced by explosive loadings), thermo-mechanical behaviour of the solid is such that, for its accurate description, we also need to take the third order terms in strain and temperature in the expression for the free energy function, i.e.

$$\hat{\psi}(\varepsilon, \theta) = a_0 + a_1\varepsilon + a_2\varepsilon^2 + a_3\varepsilon^3 + b_1\varepsilon\theta + b_2\varepsilon^2\theta + b_3\varepsilon\theta^2 + c_1\theta + c_2\theta^2 + c_3\theta^3. \tag{3.2}$$

The corresponding expressions for the pressure and entropy follow on substituting (3.2) into (2.6):

$$p = -e^{-\varepsilon}(a_1 + 2a_2\varepsilon + 3a_3\varepsilon^2 + b_1\theta + 2b_2\varepsilon\theta + b_3\theta^2) \qquad (3.3)$$

$$\eta = -\frac{1}{\rho_0 T_0}(b_1\varepsilon + b_2\varepsilon^2 + 2b_3\varepsilon\theta + c_1 + 2c_2\theta + 3c_3\theta^2). \qquad (3.4)$$

Taking pressure and entropy to be zero at zero strain ($\varepsilon = 0$) and initial temperature ($T = T_0$, $\theta = 0$), the constants a_1 and c_1 are equal to zero. The constant a_0 in (3.2) is an arbitrary value of the initial free energy function, and can also be taken to be zero. The remaining seven constants can be determined as follows. First, consider an isothermal bulk test ($\theta = 0$) under small strain

$$\left(\varepsilon \cong -\frac{\Delta V}{V}, \ \Delta V = V_0 - V, \ e^{-\varepsilon} \to 1, \ \varepsilon^2 \to 0\right),$$

then

$$p = 2a_2\frac{\Delta V}{V_0} \equiv K_0\frac{\Delta V}{V_0}, \qquad (3.5)$$

and therefore $2a_2$ is equal to the initial bulk modulus of the material (K_0). To get the constant a_3 we need to consider the isothermal bulk test at large strain, in which case

$$p = -e^{-\varepsilon}(K_0\varepsilon + 2a_3\varepsilon^2). \qquad (3.6)$$

The constant a_3 now can be obtained by comparing expression (3.6) with the experimental data for pressure–volume relation at extreme pressures. For example, for sodium metal such data were obtained by Bridgman[1] and were used by Murnaghan[2] to determine the second order elastic constant in his theory which is based on Lagrangian strain, rather than logarithmic strain. In our case, i.e. with relation (3.6) as a pressure–volume relation, the second order elastic constant a_3 for sodium is determined in [6]. Determination of the second order elastic constants for some other materials (not necessarily under hydrostatic loading) is given, for example, in [7].

In order to determine the physical meaning and the values of the constants b_1–b_3 we consider the thermal expansion test at zero pressure ($p = 0$), in which case from (3.3) we have

$$K_0\varepsilon + 3a_3\varepsilon^2 + b_1\theta + 2b_2\varepsilon\theta + b_3\theta^2 = 0. \qquad (3.7)$$

However, the increase in temperature of the order 100–200 K (the temperature range of interest in our case) produces a volume expansion small enough that the ε^2 term in (3.7) can be neglected, and therefore

$$(K_0 + 2b_2\theta)\varepsilon = -b_1\theta - b_3\theta^2. \qquad (3.8)$$

The expression $(K_0 + 2b_2\theta)$ in (3.8) can be identified as a temperature dependent bulk modulus, $K(\theta) = K_0 + 2b_2\theta$. However, for most applications it is sufficient to take the bulk modulus of solids as independent of temperature[8], although it generally decreases somewhat with increasing temperature (for example, bulk modulus of copper decreases by about 3% with a temperature increase of 100 K, from 273 to 373 K[9]). If we therefore take bulk modulus to be temperature independent, we can take b_2 to be equal to zero, and therefore from (3.8)

$$\varepsilon = -\frac{b_1}{K_0}\theta - \frac{b_3}{K_0}\theta^2. \qquad (3.9)$$

On the other hand, an increase in temperature by dT produces the relative volume expansion

$$\frac{dV}{V} = \alpha(T)\, dT \tag{3.10}$$

where $\alpha = \alpha(T)$ is the temperature dependent coefficient of volume thermal expansion, so that integration from initial to final state gives

$$\varepsilon = \int_{T_0}^{T} \alpha(T)\, dT. \tag{3.11}$$

The temperature dependence of α can be further represented by a series expansion

$$\alpha(T) = \sum_{n=0}^{\infty} \alpha_n (T-T_0)^n, \tag{3.12}$$

so that integration of (3.11) gives

$$\varepsilon = \alpha_0(T-T_0) + \tfrac{1}{2}\alpha_1(T-T_0)^2 + \tfrac{1}{3}\alpha_2(T-T_0)^3 + \ldots. \tag{3.13}$$

If we retain the first two terms in the expansion of (3.12), (3.13) becomes

$$\varepsilon = \alpha_0 T_0 \theta + \tfrac{1}{2}\alpha_1 T_0^2 \theta^2. \tag{3.14}$$

Since the coefficients α_0 and α_1 can be determined experimentally (numerical values for various metals are given, for example, in [8]), the comparison of (3.9) and (3.14) gives

$$b_1 = -K_0 \alpha_0 T_0, \quad b_3 = -\tfrac{1}{2}K_0 \alpha_1 T_0^2. \tag{3.15}$$

Expressions (3.15) can also be obtained starting from the thermodynamic definition of the coefficient of volume thermal expansion[8, 9]

$$\alpha = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p \equiv \frac{1}{K}\left(\frac{\partial p}{\partial T}\right)_V = \frac{1}{KT_0}\left(\frac{\partial p}{\partial \theta}\right)_\varepsilon, \tag{3.16}$$

where $K = -(\partial p/\partial \varepsilon)_0$ is the bulk modulus. Since

$$\frac{1}{K}\left(\frac{\partial p}{\partial \theta}\right)_\varepsilon \cong -\frac{1}{K_0}(b_1 + 2b_3\theta),$$

substitution in (3.16) and comparison with the two term approximation of (3.12) gives again (3.15).

The remaining constants (c_2 and c_3) can be determined from the consideration of the specific heat at constant volume (c_ε). It is well known from solid state physics that the temperature dependence of the specific heat at constant volume for most metals is accurately represented by the Debye theory[9, 10], according to which

$$c_\varepsilon = 3R\left[4D\left(\frac{T_D}{T}\right) - \frac{3\dfrac{T_D}{T}}{e^{T_D/T}-1}\right], \tag{3.17}$$

where

$$D(z) = \frac{3}{z^3} \int_0^z \frac{x^3 \, dx}{e^x - 1}, \quad \left(z = \frac{T_D}{T}\right)$$

is Debye's function, and T_D is the Debye temperature characteristic for each solid. (For aluminum, for example, $T_D = 428$ K, for iron $T_D = 467$ K, while for other solids the values are given in [9].) At high temperatures ($T \gg T_D$), c_t asymptotically approaches the Dulong–Petit value of $c_t = 3R$, which is for most metals $3R = 24.9381$ J mol^{-1} K^{-1}. On the other hand, using our expression for the entropy (3.4) the specific heat c_t at constant volume (V_0) is

$$c_t = T\left(\frac{\partial \eta}{\partial T}\right)_t = -\frac{2}{\rho_0 T_0}[c_2 + (c_2 + 3c_3)\theta + 3c_3\theta^2]. \tag{3.18}$$

As suggested in [5], the constants c_2 and c_3 can be determined by fitting (3.18) to (3.17) in the temperature range of interest (in our case, for example, the temperature range is approx. 273–423 K). To do this we first calculate c_t according to (3.17) at two appropriately selected temperatures: $T_D/T = 1.4$ and $T_D/T = 1$. (These temperatures would approximately define the temperature range of interest for both aluminum and iron.) Using the tabular values of Debye's function[11], we find $D(1.4) = 0.570793$ and $D(1) = 0.674416$, so that $c_t(1.4) = 22.6555$ J mol^{-1} K^{-1} and $c_t(1) = 23.7341$ J mol^{-1} K^{-1}. Equating this with (3.18) at corresponding values of θ

$$\left(\theta = \frac{1}{1.4}\frac{T_D}{T_0} - 1, \quad \text{and} \quad \theta = \frac{T_D}{T_0} - 1\right)$$

we obtain two algebraic equations for c_2 and c_3, from which:

$$c_2 = -25.8384 \, \text{J mol}^{-1}\text{K}^{-1}\left(\frac{T_D}{T_0} - 0.5407\right)\frac{\rho_0 T_0}{\left(\frac{T_D}{T_0}\right)^2}, \quad c_3 = 4.6571 \, \text{J mol}^{-1}\text{K}^{-1}\frac{\rho_0 T_0}{\left(\frac{T_D}{T_0}\right)^2}. \tag{3.19}$$

The specific heat variation (3.18) with constants c_2 and c_3 determined by (3.19) is plotted, together with Debye's expression (3.17), in Fig. 1 and satisfactory agreement in the temperature range of interest is achieved.

It should be noticed that expression (3.18) gives the temperature variation of the
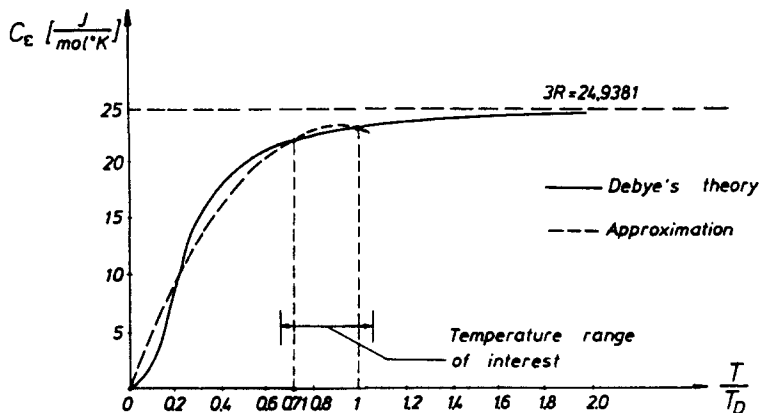

Fig. 1. The specific heat variation.

specific heat at constant volume V_0, i.e. $\varepsilon = 0$. The specific heat at constant volume $V(\varepsilon \neq 0)$ is

$$c_\varepsilon(\theta, \varepsilon) = -\frac{2}{\rho_0 T_0}[c_2 + (c_2 + 3c_3)\theta + 3c_3\theta^2] - \frac{2}{\rho_0 T_0}(1+\theta)b_3\varepsilon, \qquad (3.20)$$

which, of course, is the form of the Clausius relation[9]

$$c_v(T, v) = c_v(T, v_0) + T\int_{v_0}^{v}\left(\frac{\partial^2 p}{\partial T^2}\right)_v dv, \qquad (3.21)$$

where $v = 1/\rho$ is the specific volume. It is clear from (3.20) that for a given temperature (θ), c_ε decreases as ε grows (in absolute value). This is in agreement with (3.17) since, according to the Grüneisen equation, the volume dependence of the Debye temperature is

$$T_D = \text{constant} \cdot v^{-\gamma} \qquad (3.22)$$

where γ is the Grüneisen constant (for aluminum, for example, $\gamma = 2.1$[8]), so that

$$T_D(\varepsilon \neq 0) = e^{-\gamma\varepsilon} T_D(\varepsilon = 0), \qquad (3.23)$$

and therefore T_D increases with compression ε, i.e. for a given temperature c_ε also decreases with compression ε.

4. ADIABATIC LOADING AND SOME NUMERICAL RESULTS

Consider the case of rapidly applied pressure, so that there is no time for significant heat transfer to take place (but with inertia effects still to be neglected). The process then can be considered to be adiabatic. Since it is also elastic, with no dissipative effects, the entropy cannot change and is equal to its initial value ($\eta = 0$). Therefore, from (3.4)

$$b_1\varepsilon + b_2\varepsilon^2 + 2b_3\varepsilon\theta + 2c_2\theta + 3c_3\theta^2 = 0, \qquad (4.1)$$

so that the temperature increase corresponding to given volume strain ε is $\Delta T = \theta T_0$, where

$$\theta = -\frac{1}{3}\left(\frac{c_2}{c_3} + \frac{b_3}{c_3}\varepsilon\right) \pm \left[\frac{1}{9}\left(\frac{c_2}{c_3} + \frac{b_3}{c_3}\varepsilon\right)^2 - \left(\frac{b_1}{3c_3}\varepsilon + \frac{b_2}{3c_3}\varepsilon^2\right)\right]^{1/2}. \qquad (4.2)$$

Since for $\varepsilon = 0$ we have $\theta = 0$ and since from (3.19) c_2 is negative, we must take a minus sign in expression (4.2). To determine the adiabatically applied pressure which is required to produce a given amount of volume reduction (V/V_0), we first calculate θ from (4.2) and then with this value and with $\varepsilon = \ln (V/V_0)$ obtain the pressure from (3.3).

For example, let us consider the case of moderate volume reduction when we presumably can neglect the $a_3\varepsilon^3$ term in the free energy expansion (3.2), and the $3a_3\varepsilon^2$ term in the expression for the pressure (3.3). Reasonable agreement with the experimental pressure-volume data is still achieved, as indicated in [5], due to the presence of the $e^{-\varepsilon}$ term in the expression for the pressure. (Note that, although we are not retaining the ε^3 term in the expression for the free energy, ε is still a finite strain and $K_0 e^{-\varepsilon}\varepsilon$ is still capable of reproducing moderately large-finite strain behaviour. The use of powers of Lagrange strain, for example, does not provide such a close approximation to the geometric nonlinearity.) In this case then (3.3) becomes

$$p = -e^{-\varepsilon}(2a_2\varepsilon + b_1\theta + b_3\theta^2), \qquad (4.3)$$

where b_2 is taken to be zero, as discussed in the previous section.

In the case of aluminum, for example, with $K_0 = 70$ GPa, $\alpha_0 = 6.663 \times 10^{-5}$ K^{-1}, $\alpha_1 = 6.84 \times 10^{-8}$ K^{-2}, $\rho_0 = 2.768$ g cm^{-3}, $T_D = 428$ K[9,12] and taking $T_0 = 283$ K, we obtain from (3.5), (3.15) and (3.19): $a_2 = 35$ GPa, $b_1 = -1.32$ GPa, $b_3 = -0.192$ GPa, $c_2 = -0.139$ GPa and $c_3 = -0.059$ GPa. If we wish to calculate the temperature rise and the needed adiabatically applied pressure for the volume reduction of, say $V/V_0 = 0.85$, i.e. $\varepsilon = -0.1625$, we obtain first from (4.2) (with $b_2 = 0$) $\theta = 0.429$, which corresponds to a temperature increase $\Delta T = \theta T_0 = 121.4$ K. The required pressure is then from (4.3) $p = 14.09$ GPa. It should be mentioned that the inclusion of the $a_3\varepsilon^3$ term in the expression for free energy [which we have neglected in (4.3)] would make the value of pressure more accurate, but the temperature rise would still be the same, as calculated by (4.2), which does not depend on a_2 and a_3.

It is useful to compare these results with those obtained by using the free energy expansion with terms only up to the second degree, i.e.

$$\tilde{\psi} = a_2\varepsilon^2 + b_1\varepsilon\theta + c_2\theta^2. \tag{4.4}$$

In this case we have from (2.6):

$$p = -e^{-\varepsilon}(2a_2\varepsilon + b_1\theta) \tag{4.5}$$

$$\eta = -\frac{1}{\rho_0 T_0}(b_1\varepsilon + 2c_2\theta), \tag{4.6}$$

so that for the adiabatic compression:

$$\theta = -\frac{b_1}{2c_2}\varepsilon \tag{4.7}$$

$$p = -e^{-\varepsilon}\left(2a_2 - \frac{b_1^2}{2c_2}\right)\varepsilon. \tag{4.8}$$

The constants a_2 and b_1 again can be determined from $a_2 = \frac{1}{2}K_0$ and $b_1 = -K_0\alpha_0 T_0$, i.e. by using the values of bulk modulus and coefficient of volume thermal expansion at temperature $T = T_0$ (initial values). The constant c_2 can be determined by using the expression for the specific heat

$$c_\varepsilon = -\frac{2}{\rho_0 T_0}(1+\theta)c_2 \tag{4.9}$$

and the Debye value of c_ε at $T = T_0$ ($\theta = 0$), which is for aluminum $c_\varepsilon(T = T_0) \cong 22.6$ J mol^{-1}K^{-1}. In this way we obtain: $a_2 = 35$ GPa, $b_1 = -1.32$ GPa and $c_2 = -0.328$ GPa, so that a volume compression of $\varepsilon = -0.1625$ is followed by a temperature increase of $\Delta T = 92.5$ K, since from (4.7), $\theta = 0.327$. The corresponding pressure is from (4.8), $p = 13.89$ GPa. We see, therefore, that the first order theory (with free energy function of the second degree) predicts 28.9 K, i.e. a 23.8% smaller increase in temperature for a given volume reduction of $V = 0.85V_0$. On the other hand, the value for the required pressure is only 0.2 GPa, i.e. 1.4% smaller in the case of the first order theory. This is not surprising because the difference of 28.9 K in the increase of temperature requires a somewhat higher pressure (14.09 vs 13.89 GPa) to produce a given amount of compression.

Better agreement of the first order theory with the second order theory could be obtained if we use the mean values of the coefficient of volume thermal expansion α and specific heat c_ε in the interval of expected temperature increase. For example, if we take $\alpha = 7.347 \times 10^{-5}$K^{-1} and $c_\varepsilon = 23.17$ J mol^{-1} K^{-1} (which are the values approximately at $T = T_0 + 60$ K), we get $b_1 = -1.4555$ GPa and $c_2 = -0.2775$ GPa, so that $\theta = 0.426$, $\Delta T = 120.5$ K and $p = 14.11$ GPa. However, we cannot in advance know the amount of

temperature increase, and therefore cannot know the mean values of α and c_r (except, of course, by assuming them approximately), so that it is clearly necessary to use the second order theory to determine the true temperature increase and required pressure for a given amount of adiabatic compression.

5. DISCUSSION

It can be seen from this paper, that in order to adequately reproduce the thermo-mechanical behaviour of metals under finite volume reduction and large temperature increase, it is necessary to use the second order thermo-elastic analysis, i.e. to use the free energy function expanded up to the third degree. In particular, we have seen that in the case of adiabatically compressed aluminum, the first order theory predicts a 23.8% lower increase of temperature than the second order theory. The analysis performed in this paper was restricted to hydrostatic loading, when the solid can be described by an equation of state which includes just volume strain (or density) and temperature, as in the case of a fluid. However, inclusion of the nonhydrostatic loading, as it arises in explosive loadings, would clearly demand elastic–plastic theory, as is done in the analysis of the propagation of elastic–plastic waves at finite strain[4, 5]. However, at extremely high pressures (say, higher than 20 GPa), the effects of shear stresses can be neglected in comparison with the influence of mean pressure and this again leads to a fluid-like behaviour, i.e. hydrodynamic theory as discussed in the papers on the wave and shock propagation phenomena, such as [3, 13, 14].

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