



General form of the Mie–Grüneisen equation of state

Formulation générale de l'équation d'état de Mie–Grüneisen

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ABSTRACT

The Mie–Grüneisen equation of state is defined in an incomplete form $P(V, E)$ which does not allow access to temperature and entropy. We show here how we can extend it to a complete equation of state in the $S(V, E)$ form by providing an additional independent function which defines the heat capacity variations and then gives access to all the thermodynamic properties.

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RÉSUMÉ

L'équation d'état de Mie–Grüneisen est définie par la formulation incomplète $P(V, E)$ qui ne permet pas d'accéder à la température et à l'entropie. Nous présentons ici son extension dans le cas général $S(V, E)$, en fournissant une fonction indépendante qui définit les variations de la chaleur spécifique, et permet ainsi l'accès à toutes les grandeurs thermodynamiques.

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L'équation d'état de Mie–Grüneisen se présente sous la formulation incomplète $P(V, E)$ qui ne permet pas d'accéder à la température et à l'entropie. Pour la compléter, on lui adjoint souvent une l'expression de l'énergie interne sous la forme $E(V, T)$. Pour assurer la cohérence thermodynamique, celle-ci doit vérifier une équation aux dérivées partielles extrêmement contraignante. Nous présentons ici son extension dans le cas général sous la forme $S(V, E)$ qui vérifie par construction les conditions de compatibilité thermodynamique, et donne l'accès à toutes les grandeurs thermodynamiques. Cette extension est définie à partir d'une fonction adimensionnelle indépendante qui peut restituer les modèles connus pour la chaleur spécifique.

La formulation complète d'une équation d'état de type Mie–Grüneisen se compose donc de trois ingrédients indépendants : (1) un potentiel de référence qui ne dépend que du volume, généralement choisi sur une isentrope ; (2) la température de Debye qui ne dépend aussi que du volume et dont dérive le coefficient de Grüneisen ; et (3) une fonction adimensionnelle d'une variable qui dépend des deux fonctions précédentes et de l'énergie interne. Cette fonction définit l'entropie et la chaleur spécifique. Sa transformée de Legendre conduit à la formulation de l'énergie libre $F(V, T)$.

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Les différentes constantes d'intégrations introduites peuvent être choisies, par exemple pour restituer un état thermodynamique de référence.

Les résultats obtenus ici ont des conséquences particulièrement intéressantes pour l'étude des ondes de choc ou hautes pressions dynamiques avec changements de phase multiples, des alliages sous fortes pressions, et pour leur utilisation dans les codes hydrodynamiques.

1. Introduction

One century ago, Mie [1] and Grüneisen [2] developed a theory for solids which concluded that the pressure could be considered as a linear function of internal energy. The name Mie–Grüneisen is now associated to an equation of state which verifies this assumption. Starting from the study of crystals, they obtained that the internal energy of a solid is the sum of its potential energy at zero temperature resulting from interactions of atoms at rest, and a thermal vibrational energy $E_T(V, T)$ increasing with temperature

$$E = E_K(V) + E_T(V, T) \quad (1)$$

The electronic contribution was neglected. The related pressure is also the sum of two such terms:

$$P = P_K(V) + P_T(V, T) \quad (2)$$

The potential energy is also called the cold contribution or the reference potential. The cold pressure term is the derivative of the potential energy versus volume:

$$P_K(V) = -\frac{dE_K(V)}{dV} \quad (3)$$

The thermal pressure $P_T(V, T)$ is proportional to the energy $E_T(V, T)$. Their ratio does not depend on temperature but on the relationship of the vibration frequency ν versus volume. The Grüneisen coefficient Γ was introduced:

$$\Gamma(V) = -\frac{d \ln(\nu)}{d \ln(V)} \quad (4)$$

From the virial theorem, Grüneisen obtained:

$$P_T(V, T) = \frac{\Gamma(V)}{V} E_T(V, T) \quad (5)$$

from which we can deduce the equation which is today the definition of a Mie–Grüneisen equation of state:

$$P(V, E) = P_K(V) + \frac{\Gamma(V)}{V} (E - E_K(V)) \quad (6)$$

$E_K(V)$ is generally taken on the isotherm $T = 0$ K or on an isentrope $S = S_K$.

The Grüneisen coefficient, at the macroscopic level, is defined from (4) or thermodynamic considerations by:

$$\Gamma(V) = V \left(\frac{\partial P}{\partial E} \right)_V = - \left(\frac{\partial \log(T)}{\partial \log(V)} \right)_S \quad (7)$$

The linear assumption is also a result from experiments. Experimental measurements of the heat capacity at constant volume C_V

$$C_V = \left(\frac{\partial E}{\partial T} \right)_V \quad (8)$$

the coefficient of thermal expansion α

$$\alpha = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_P \quad (9)$$

and the compressibility coefficient K_T :

$$K_T = -V \left(\frac{\partial P}{\partial V} \right)_T \quad (10)$$

lead to the conclusion that the Grüneisen coefficient

$$\Gamma = \frac{\alpha \cdot V \cdot K_T}{C_V} \quad (11)$$

does not depend on temperature, then only on volume.

Eq. (6) is also valid for many other states of matter (ideal gas, Van der Waals, ...).

The $P(V, E)$ form is especially useful in fluid mechanics and shock wave physics where the conservation equations involve pressure, volume, internal energy and not explicitly temperature. But accurate equations of state of materials are often obtained from statistical thermodynamics in the (V, T) form deduced from the free energy. Then, generally we add an energetic equation of state $E(V, T)$ which must verify:

$$\left(\frac{\partial E}{\partial V}\right)_T = -P + T\left(\frac{\partial P}{\partial T}\right)_V = -P + T\frac{\Gamma(V)}{V}\left(\frac{\partial E}{\partial T}\right)_V \tag{12}$$

It is difficult to find families of internal energies, with a physical behavior, which solve this differential equation. Moreover, it is constrained by integration constants. This problem should be described in a better form.

Another problem is raised when phase transitions with triple points are taken into account. The (V, T) form cannot be used since the internal energy cannot be determined in triple points without further information.

In former studies, we have developed some complete $S(V, E)$ models using further assumptions and valid for restricted applications [3–7]. In the present study, we describe the general complete form of the Mie–Grüneisen equation of state with no more assumption. We will assume here that temperature, entropy and internal energy are continuous and monotone functions of each other.

2. Properties

Definition (6) has several interesting consequences:

- P is a linear function of E ,
- $\left(\frac{\partial \Gamma}{\partial E}\right)_V = 0$ then the Grüneisen coefficient Γ depends only on volume:

$$\Gamma = \Gamma(V) \tag{13}$$

- $\left(\frac{\partial C_V}{\partial V}\right)_S = 0$ then the specific heat C_V is a function of entropy:

$$C_V = C_V(S) \tag{14}$$

- the temperature is the product of a function of volume by a function of entropy:

$$T(V, S) = \theta(V)k(S) \tag{15}$$

The two first properties are obviously deduced from the definition. The third one is a consequence of a general thermodynamic property. From the equality of crossed derivatives

$$\frac{\partial}{\partial S}\left[\left(\frac{\partial T}{\partial V}\right)_S\right]_V - \frac{\partial}{\partial V}\left[\left(\frac{\partial T}{\partial S}\right)_V\right]_S = 0 \tag{16}$$

and noticing that

$$\left(\frac{\partial T}{\partial V}\right)_S = -\frac{\Gamma T}{V} \tag{17}$$

and

$$\left(\frac{\partial T}{\partial S}\right)_V = \frac{T}{C_V} \tag{18}$$

we deduce:

$$\left(\frac{\partial \Gamma}{\partial S}\right)_V + \left(\frac{\partial(1/C_V)}{\partial \log(V)}\right)_S = 0 \tag{19}$$

In our particular case, the Grüneisen coefficient does not depend on entropy, then the heat capacity does not depend on volume. This means that the heat capacity is a function of entropy.

Let us introduce a function $\Theta(V)$ which corresponds to the well-known Debye temperature [8]:

$$\Theta(V) = \Theta_0 \cdot \exp\left[-\int_{V_0}^V \frac{\Gamma(v)}{v} dv\right] \tag{20}$$

where Θ_0 is a constant of the material. By derivation, we deduce the Grüneisen coefficient $\Gamma(V)$:

$$\Gamma(V) = -\frac{d \ln(\Theta(V))}{d \ln(V)} = -V \frac{\Theta'(V)}{\Theta(V)} \tag{21}$$

The linear relationship between P and E yields that C_V depends only on entropy. Hence, from the thermodynamic relations:

$$\left(\frac{\partial \ln(T)}{\partial V}\right)_S = -\frac{\Gamma(V)}{V} \quad (22)$$

$$\left(\frac{\partial \ln(T)}{\partial S}\right)_V = \frac{1}{C_V} = \frac{1}{C_V(S)} \quad (23)$$

we deduce, by integration [9]:

$$\ln\left(\frac{T}{T_0(V_0, S_0)}\right) = -\int_{V_0}^V \frac{\Gamma(v)}{v} dv + \int_{S_0}^S \frac{ds}{C_V(s)} \quad (24)$$

We will not detail here the values of integration constants. We just keep them when there are useful. They will be determined later to match a reference state.

Then, we obtain our fourth property:

$$T(V, S) = \Theta(V)k(S) \quad (25)$$

3. Integration from the incomplete form to the complete form

From the thermodynamic relation

$$T(V, S) = \left(\frac{\partial E}{\partial S}\right)_V \quad (26)$$

the integration of (25) provides:

$$E - E_K(V) = \Theta(V)[K(S) - K(S_0)] \quad (27)$$

It is useful to take here the integration constant $E_K(V)$ on the reference isentrope S_0 . Notice that any function $E_0(V) = E_K(V) + \alpha \cdot \Theta(V)$ is also valid. Then:

$$S(V, E) = K^{(-1)}\left(\frac{E - E_0(V)}{\Theta(V)} + K(S_0)\right) \quad (28)$$

Let us introduce now the dimensionless variable $u(V, E)$:

$$u = \frac{E - E_K(V)}{C_{Vr} \cdot \Theta(V)} + u_r \quad (29)$$

and a dimensionless function $\Psi(u)$ (obtained from inversion of $K(S)$) which defines the entropy by:

$$S = S(u) = S(V, E) = S_r + C_{Vr}\Psi(u) \quad (30)$$

where S_r , C_{Vr} and u_r are parameters.

From (29), we deduce its partial derivatives:

$$\left(\frac{\partial u}{\partial E}\right)_V = \frac{1}{C_{Vr}\Theta(V)} \quad (31)$$

$$\left(\frac{\partial u}{\partial V}\right)_E = \frac{P_K(V) + \frac{\Gamma(V)}{V}(E - E_K(V))}{C_{Vr}\Theta(V)} \quad (32)$$

We obtain pressure and temperature from the first derivatives of (30):

$$\left(\frac{\partial S}{\partial E}\right)_V = \frac{1}{T} = C_{Vr}\Psi'(u)\left(\frac{\partial u}{\partial E}\right)_V \quad (33)$$

$$\left(\frac{\partial S}{\partial V}\right)_E = \frac{P}{T} = C_{Vr}\Psi'(u)\left(\frac{\partial u}{\partial V}\right)_E \quad (34)$$

$$T(V, E) = \frac{\Theta(V)}{\Psi'(u)} \quad (35)$$

$$P(V, E) = P_K(V) + \frac{\Gamma(V)}{V}[E - E_K(V)] \quad (36)$$

and

$$\left(\frac{\partial T}{\partial E}\right)_V = \frac{1}{C_V} = -\frac{\Psi''(u)}{C_{Vr}\Psi'(u)^2} \tag{37}$$

hence

$$C_V = C_V(u) = -C_{Vr} \frac{\Psi'(u)^2}{\Psi''(u)} \tag{38}$$

4. Reference state

The reference state is often a problem with the Mie–Grüneisen EoS. We propose here a method to adjust the integration constants to a reference state. These constants can also be adjusted to correspond to other requirements. Notice that we can modify the thermal function $\Psi(u)$ to $\Psi(u + \alpha)$ to fit a given temperature without changing the $P(V, E)$ relationship.

Let us introduce $\epsilon_K(V)$ such as $\epsilon_K(V_0) = 0$, $\epsilon'_K(V_0) = 0$, E_{K0} and P_{K0} :

$$E_K(V) = E_{K0} + \epsilon_K(V) \tag{39}$$

$$P_K(V) = P_{K0} - \epsilon'_K(V) \tag{40}$$

It is useful to take $P_{K0} = 0$ without further problems.

If we choose a reference state for which, $T_0, P_0, V_0, E_0, S_0, C_{V0}, \Theta_0$ and Γ_0 are given, we can deduce all the constants of the model.

$$P_0 = P(V_0, E_0) = \Gamma_0 \rho_0 (E_0 - E_{K0}) \tag{41}$$

then

$$E_{K0} = E_0 - \frac{P_0 V_0}{\Gamma_0} \tag{42}$$

We need to introduce u_0 :

$$u_0 = \frac{P_0 V_0}{\Gamma_0 C_{Vr} \Theta_0} + u_r \tag{43}$$

Then, S_r, C_{Vr}, u_0 and u_r are solutions of a system of four equations.

First we calculate u_0 from:

$$\Psi'(u_0) = \frac{\Theta_0}{T_0} \tag{44}$$

Then, we obtain C_{Vr} from (38) applied to $u = u_0$, then:

$$C_{Vr} = -C_{V0} \frac{\Psi''(u_0)}{\Psi'(u_0)^2} \tag{45}$$

We deduce u_r from (43):

$$u_r = u_0 - \frac{P_0 V_0}{\Gamma_0 C_{Vr} \Theta_0} \tag{46}$$

and S_r from (30) applied to $u = u_0$, then:

$$S_r = S_0 - C_{Vr} \Psi(u_0) \tag{47}$$

5. Link with the (V, T) form

The free energy is:

$$F(V, E) = E - T(V, E) \cdot S(V, E) = E - C_{Vr} \cdot \Theta(V) \frac{\Psi(u)}{\Psi'(u)} - T S_r \tag{48}$$

We introduce the dimensionless variable x [8]:

$$x = \frac{\Theta(V)}{T} = \Psi'(u) \tag{49}$$

Hence, its partial derivatives:

$$\left(\frac{\partial x}{\partial T}\right)_V = -\frac{x}{T} \quad (50)$$

$$\left(\frac{\partial x}{\partial V}\right)_T = \frac{\Theta'(V)}{T} = -\frac{\Gamma(V)}{V}x \quad (51)$$

We introduce $\Phi(x)$ and we define the free energy by:

$$F(V, T) = E_K(V) + C_{Vr}T[\Phi(x) - u_r x] - TS_r \quad (52)$$

where C_{Vr} , u_r and S_r are the parameters defined above.

We deduce pressure and entropy from partial first derivatives:

$$-\left(\frac{\partial F}{\partial V}\right)_T = P(V, T) = P_K(V) + C_{Vr} \cdot T \frac{\Gamma(V)}{V} x(\Phi'(x) - u_r) \quad (53)$$

$$-\left(\frac{\partial F}{\partial T}\right)_V = S(V, T) = S_r + C_{Vr}(x\Phi'(x) - \Phi(x)) \quad (54)$$

and from them the internal energy:

$$E(V, T) = E_K(V) + C_{Vr}Tx(\Phi'(x) - u_r) = E_K(V) + C_{Vr}\Theta(V)(\Phi'(x) - u_r) \quad (55)$$

and

$$C_V(V, T) = C_V(x) = -C_{Vr} \cdot x^2 \Phi''(x) \quad (56)$$

5.1. Reference state

We first calculate x_0 from

$$x_0 = \frac{\Theta_0}{T_0} \quad (57)$$

We deduce C_{Vr} from (56):

$$C_{Vr} = -\frac{C_{V0}}{x_0^2 \Phi''(x_0)} \quad (58)$$

We obtain E_{K0} from (42), and u_r from $P(V_0, T_0)$ and (53):

$$u_r = \Phi'(x_0) - \frac{P_0 V_0}{\Gamma_0 C_{Vr} \Theta_0} \quad (59)$$

Then S_r from (54):

$$S_r = S_0 - C_{Vr}(x_0 \Phi'(x_0) - \Phi(x_0)) \quad (60)$$

Notice that the reference state is the same for $S(V, E)$ and $F(V, T)$.

5.2. Fundamental property

Notice that we can switch from $\Psi(x)$ to $\Phi(u)$ by a Legendre transform:

$$\Psi'(u) = x = \frac{\Theta(V)}{T} \quad (61)$$

$$\Phi(x) = u \cdot x - \Psi(u) \quad (62)$$

$$\Phi'(x) = u = \frac{E - E_K(V)}{C_{Vr} \cdot \Theta(V)} + u_r \quad (63)$$

$$\Psi(u) = u \cdot x - \Phi(x) \quad (64)$$

6. Particular cases

6.1. Grüneisen coefficient models

In the case of ideal gas, it is constant and:

$$\Gamma = \gamma - 1 = \Gamma_0 \tag{65}$$

where γ is the polytropic coefficient. Then

$$\theta(V) = \theta_0 \left(\frac{V_0}{V} \right)^{\Gamma_0} \tag{66}$$

For solids, an usual assumption is that the following ratio is constant:

$$\frac{\Gamma(V)}{V} = \frac{\alpha \cdot K_T}{C_V} = \Gamma_0 \rho_0 \tag{67}$$

then:

$$\theta(V) = \theta_0 \cdot \exp[\Gamma_0(1 - \rho_0 V)] \tag{68}$$

Some models assume that the Grüneisen coefficient is deduced from the reference potential:

$$\Gamma(V) = \frac{1}{3}(\lambda - 1) - \frac{V}{2} \frac{\frac{d^2[P_K(V)V^{\frac{2(\lambda+1)}{3}}]}{dV^2}}{\frac{d[P_K(V)V^{(2/3)(\lambda+1)}]}{dV}} \tag{69}$$

These models are associated respectively to particular values for λ :

- Slater [10] for $\lambda = -1$,
- Dugdale–MacDonald [11] for $\lambda = 0$,
- the free volume theory of Vashchenko–Zubarev [12] for $\lambda = +1$.

We obtain by integration:

$$\theta(V) = \frac{\theta_0}{\sqrt{-P'_K(V_0)}} \left(\frac{V}{V_0} \right)^{2/3} \left[-V^{-\frac{2(\lambda+1)}{3}} \frac{d}{dV} (P_K(V) \cdot V^{\frac{2(\lambda+1)}{3}}) \right]^{1/2} \tag{70}$$

6.2. Reference potential models

The reference potentials $E_K(V)$ proposed by Murnaghan [13], Birch [14], Vinet [15,16], or Heuzé [7] are defined from a reference density, and for this density, by the isothermal bulk modulus K_0 and its derivative versus pressure K'_0 :

$$K_T = -V \left(\frac{\partial P}{\partial V} \right)_T \tag{71}$$

$$K_0 = -V \left(\frac{\partial P}{\partial V} \right)_{T=T_0, V=V_0} \tag{72}$$

$$N_0 = K'_0 = \left(\frac{dK_T}{dP} \right)_{P=0} = -1 - \left[\frac{\left(\frac{\partial^2 P}{\partial V^2} \right)_T}{\left(\frac{\partial P}{\partial V} \right)_T} \right]_{T=T_0, V=V_0} \tag{73}$$

Some other potentials have also two parameters which can be identified to the compressibility and its derivative for comparison. Many other potentials exist in the literature [17] which can be used here.

6.3. Heat capacity models

Constant heat capacity, Einstein and Debye models are often used for the heat capacity. The Debye model, defined by an integral, requires further developments to obtain its Legendre transform.

6.3.1. Constant heat capacity

In (V, E) form:

$$\Psi(u) = \ln(u) \quad (74)$$

In (V, T) form:

$$\Phi(x) = \ln\left(\frac{x}{x_0}\right) \quad (75)$$

6.3.2. Einstein model

In (V, E) form:

$$\Psi(u) = (u + 1) \cdot \ln(u + 1) - u \cdot \ln(u) \quad (76)$$

In (V, T) form, we have:

$$\Phi(x) = \ln(1 - e^{-x}) \quad (77)$$

For $C_{Vr} = 3R$, we deduce the well-known Einstein heat capacity:

$$C_V(x) = 3R \frac{x^2 e^x}{(e^x - 1)^2} \quad (78)$$

7. Examples

We provide here a comparison for Al, Cu and Pb between our thermal functions for the constant heat capacity (74) and Einstein heat capacity (76).

We have calculated the parameters of our model in the reference state: $E_0 = 0, S_0 = 0, P_0 = 0, T_0 = 298.13$ K. The physical data in this state for the three materials are taken from [18,19] and gathered in Table 1. In this state, the values of our parameters do not depend on the choice of the potential nor the Grüneisen model provided that $\Gamma(V_0) = \Gamma_0$. Notice also that $u_r = u_0$, and for constant heat capacity $C_{Vr} = C_{V0}$. The values of the parameters are collected in Table 1.

Table 1
Parameters u_r, C_{Vr} , and S_r of our model for Al, Cu and Pb deduced from physical data provided by [18,19].

		Al	Cu	Pb
	ρ_0 (kg/m ³)	2710	8930	11340
[18]	C_{V0} (J/kg/K)	896	382	129
	Γ_0	2.09	1.98	2.46
[19]	θ_0 (K)	428	343	105
	$u_r = u_0$	0.696565	0.869184	2.83933
Constant C_V	C_{Vr} (J/kg/K)	896	382	129
	S_r (J/kg/K)	323.988	53.5567	-134.62
	$u_r = u_0$	0.312282	0.463008	2.36862
Einstein	C_{Vr} (J/kg/K)	1060.86	426.04	130.339
	S_r (J/kg/K)	-763.908	-389.055	-267.028

8. Conclusion

We have extended the incomplete $P(V, E)$ Mie–Grüneisen equation of state to the complete forms $S(V, E)$ and $F(V, T)$. The initial incomplete form was defined from two functions of volume: a reference potential and the Grüneisen coefficient. Our extension consists in adding a thermal function whose second derivative provides the heat capacity. The thermal functions $\Psi(u)$ and $\Phi(x)$ associated respectively to $P(V, E)$ and $F(V, T)$ are Legendre transform from each other. For both forms $S(V, E)$ and $F(V, T)$, we have defined integration constants linked to a given reference state. These three functions are independent and many different combinations of published models are possible. This is especially useful to choose which elementary model in each of the three families better fits experiments or ab initio calculations. These results have particularly interesting consequences for the studies of shock waves with multiple phase transitions, alloys at high pressures, and in planetology, and more generally when wide ranges of pressures and temperatures are explored. It is the case, for instance, for ice when skyships cross icy clouds or when meteorites impact Mars, in powerful laser shot experiments, or for materials of the Earth mantle or other planets.

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