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Study of phase change in materials under high pressure

V. P. Singh[a] and D. V. Renuka
Centre for Aeronautical System Studies and Analyses, Bangalore-560 075, India

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Propagation of converging shock in metals and radioactive materials is studied. Variation of pressure in different metals and radioactive materials has been calculated numerically as the shock converges to its center. © *1999 American Institute of Physics.* [S0021-8979(99)06421-X]

INTRODUCTION

The study of laws governing the propagation of shock waves through condensed media such as metals is of great theoretical and practical importance. Behavior of metals under high pressure has been analyzed by many authors[1–4] by taking into account, the general Hugoniot relationship between shock velocity and particle velocity of the material. This linear equation fails when we analyze the material undergoing a crystallographic phase change under high pressure. Thus, knowledge of the thermodynamic properties of the materials is necessary to study their behavior under high pressure. Although, no appreciable[2] difficulties are encountered in calculating the thermodynamic properties of gases, a theoretical description of the thermodynamic properties of solids and liquids at the high pressure generated by very strong shocks presents a very complex problem.

In the present article, we have taken thermodynamic properties of the materials into account and attained the variation of material parameters under shock loading. The basic feature distinguishing the condensed material from the gaseous state and determining the behavior of solids and liquids compressed by shock waves is the strong interaction between atoms or molecules of the medium. A very strong compression of a condensed medium generates a colossal internal pressure even in the absence of heating due only to the repulsive forces between the atoms. The material is also very strongly heated by shock waves and this results in the appearance of a pressure associated with the thermal motion of atoms. This pressure is referred to as "thermal pressure" (P_T) in contrast to the elastic or "cold pressure" (P_c) caused by the repulsive forces. In principle, as the shock strength tends to infinity, the relative importance of thermal pressure increases and in the limit, the "excitation pressure" (P_e) becomes small in comparison with the thermal pressure. However, for shock waves with pressures of the order of millions of atmospheres, these two pressures are of comparable magnitude. The elastic pressure is dominant in weaker shock waves with pressures of the order of thousands of atmospheres and below.

FORMULATION OF THE PROBLEM

Let us consider a solid metallic sphere as the medium for shock wave propagation and assume that a spherical converging shock wave is induced by the detonation of an explosive pad at the surface of the metallic sphere concentric with the solid sphere.[1] If "U" is the shock velocity and "u_2" is the particle velocity, ρ_2, P, and E the density, pressure, and internal energy of the material behind the shock front which is at a distance from the center of the sphere and ρ_1, $P_0(=0)$, and E_o the values ahead of it, then the equations of conservation of mass, momentum and energy of the medium are given by

$$(u_2-U)\rho_2=-U\rho_1, \tag{1}$$

$$P+\rho_2(U-u_2)^2=-\rho_1 U^2, \tag{2}$$

$$P/\rho_2+E+1/2(U-u_2)^2=-E_0+1/2U^2. \tag{3}$$

In order to solve Eqs. (1)–(3) in terms of one of the parameter say (ρ_2/ρ_1), we require one more equation, i.e., the equation of state of the material. In the present problem, for introducing thermodynamic properties of the material into the equation, the pressure and energy terms are to be modified. Other than the compressed pressure and energy terms, total pressure and energy should include the thermal terms also.

The atoms of a material are set into motion by heating. A definite energy and pressure are connected with the thermal motion of the atoms. At temperatures of the order of tens and thousands of degrees and above the thermal excitation of the electrons plays an important role. Hence, the total energy and pressure can be represented as a sum of their elastic and thermal contributions. The thermal contribution, in turn can be broken up into two parts. One part corresponding to the thermal motion of the atoms and another part corresponding to the thermal excitation of the electrons with excitation energy E_e and excitation pressure P_e^3. The specific internal energy and pressure of a solid can then be written as

$$E=E_c+E_T+E_e, \tag{4}$$

$$P=P_c+P_T+P_e, \tag{5}$$

where P_c represents the cold pressure of the compressed material which is given by

$$P_c=A(S)\left[\left(\frac{\rho_2}{\rho_1}\right)^n-1\right], \tag{6}$$

where A and n are constant parameters for a material. Equation (6) for P_c can also be expressed in polynomial form as

[a]Electronic mail: drvpsingh@hotmail.com

TABLE I. Data used in this article.

Material	a km/s	b	Γ_0	Γ at boundary	n	β erg/g deg^2	ρ g/cm^3	E_0 10^8 erg/g
Al	5.328	1.338	2.0	1.35	4.352	500	2.785	16.1
SS	4.569	1.49	2.17	1.49	4.96	540.6	7.896	12.9
Fe.	3.574	1.92	1.69	1.56	6.68	541	7.85	12.85
Mo	5.124	1.233	1.520	1.39	3.932	495	10.206	7.2
Ir	3.916	1.457	1.97	1.19	4.828	380.8	22.484	3.89
Pa	3.948	1.588	2.26	1.57	5.352	469.6	11.991	6.838
U–Mo	2.565	1.531	2.03	1.42	5.124	406.7	18.45	3.367

$$P_c = nA\left(\frac{\rho_2}{\rho_1}-1\right)\left[1+\frac{(n-1)(n-2)}{2}\left(\frac{\rho_2}{\rho_1}-1\right) + \frac{(n-1)(n-2)}{6}\left(\frac{\rho_2}{\rho_1}-1\right)^2+\ldots\right]. \tag{6a}$$

The Hugoniot form of the equation of state is written as

$U = a + bu_2 + cu_2^2$,

where a, b, and c can be related with ρ and n as.[5]

$$a=\sqrt{An/\rho_1},\quad b=\frac{n+1}{4},\quad c=-\frac{b}{6a}(b-2), \tag{7}$$

where a is sound velocity in the compressed material. The temperature dependence of the thermal pressure is written as

$$P_T=\Gamma(V)\frac{E_T}{V}, \tag{8}$$

where $E_T=3Nk(T-T_0)+E_0$, and the quantity Γ characterizing the ratio of the thermal pressure to the thermal energy of the lattice is called the Gruneisen coefficient[4] and Γ is calculated from Eq. (6a) using the relation

$$\Gamma(V)=-\frac{2}{3}-\frac{V}{2}\frac{(d^2Pc/dV^2)}{(dPc/dV)}, \tag{9}$$

where V is the specific volume of the material ($=1/\rho_2$). E_o is the thermal energy at room temperature and is given by

$$E_o=\int_o^{T_o} C_V(T)dT. \tag{10}$$

The excitation pressure is

$$P_e=\frac{1}{2}\frac{E_e}{V}, \tag{11}$$

where

$$E_e=\frac{1}{2}\beta_0\left(\frac{\rho_1}{\rho_2}\right)^{1/2}T^2 \quad\text{and}\quad \beta_0=21.179\frac{k^2 m_e}{h^2}N_e^{1/3}\left(\frac{1}{\rho_1}\right)^{2/3}, \tag{12}$$

where m_e is the electron mass k is the Boltzmann constant, h is the Planck's constant, and N_e is the number of free electrons per unit volume of the metal.

Thus, by using Eqs. (8) and (11) in Eq. (5), the equation of state of material can be written as

$$P=P_c(V)+\Gamma(v)C_vT\rho_2+1/2E_e\rho_2. \tag{13}$$

SOLUTION OF THE PROBLEM

Equations (1) to (3) and (13) are solved to get the pressure, temperature, shock velocity, and particle velocity of the material behind the shock front and are expressed in terms of $\delta(=\rho_2/\rho_1)$

$$T=\frac{-C_v\left[\Gamma+1+\frac{\Gamma\delta}{2}\right]-\sqrt{C_v\left[\Gamma+1+\frac{\Gamma\delta}{2}\right]^2-x(\delta)}}{2*Y(\delta)}$$

$$U=\sqrt{\frac{\sqrt{\delta\beta_0T^2}}{4}+\Gamma\delta C_VT+\frac{X}{\rho_1}+\frac{1}{\delta}} \tag{14}$$

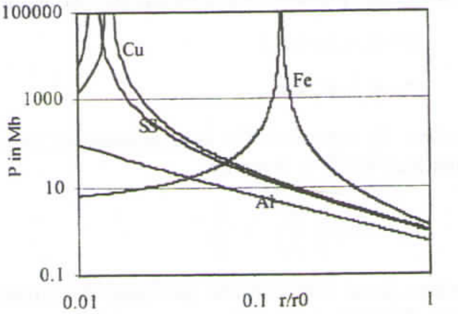

FIG. 1. P vs r/r_0 with equation $U=a+bu$.

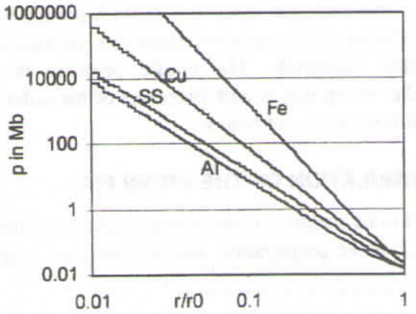

FIG. 2. P vs r/r_0 with revised calculation.

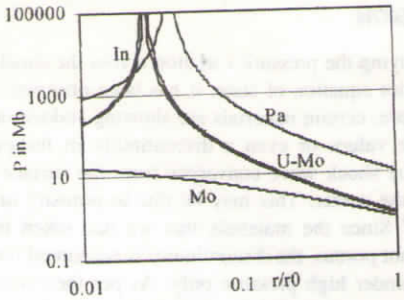

FIG. 3. P vs r/r_0 with equation $U=a+bu$ for radioactive materials.

$$u_2=\frac{U(\delta-1)}{\delta}$$

$$P=A(\delta^n-1)+\rho_1\Gamma\delta(C_v(T-T_0)+E_0)+\frac{\rho_1\delta^{1/2}}{4}\beta_0T^2,$$

where

$$x(\delta)=4*\left(\frac{3\beta_0}{4\delta^{1/2}}+\frac{1}{8}\beta_0\delta^{1/2}\right)*\left(\frac{y_1(\delta)}{\delta\rho_1}+E_c-C_vT_0-\frac{1}{2\delta^2}+\frac{y_1(\delta)}{2\rho_1}+\frac{1}{2\delta}\right)$$

$$Y(\delta)=2*\left(\frac{3\beta_0}{4\delta^{1/2}}+\frac{1}{8}\beta_0\delta^{1/2}\right)$$

$$y_1(\delta)=\rho_1\delta\Gamma\epsilon_0-\rho_1\delta\Gamma C_vT_0+A(\delta^n-1)$$

$$X=\rho_1\delta\Gamma E_0-\rho_1\delta\Gamma C_VT_0+A(\delta^n-1).$$

Equation (14) lists the four conditions relating five unknowns, P, U, u_2, T, and ρ_2. Our aim is to determine the function relating the above five terms with the distance "r" of the shock front from the center of the sphere. Transmitted pressure due to the initial compression of the metal by the shock transmitted from explosive to material at its outer boundary is calculated using the equation[6]

$$\frac{P_2}{P_D}=\frac{2\rho_1U}{\rho_DU_D+\rho_1U}, \tag{15}$$

where P_D and U_D denote the pressure and velocity of the detonation wave and ρ_D represents the initial density of the explosive.

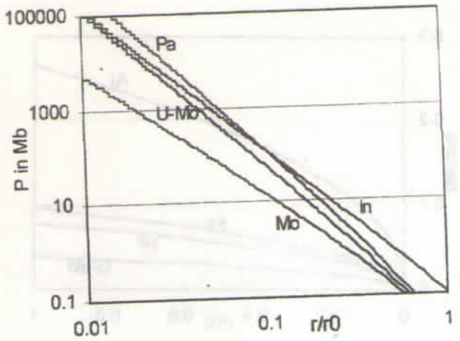

FIG. 4. P vs r/r_0 with revised calculation for radioactive materials.

Variation of δ as the shock moves from the surface of metallic sphere to its apex is calculated by using energy hypothesis by T. Y. Thomas[7] as

$$\left(\frac{r_o}{r}\right)^3=\frac{\delta\,E(\delta)}{\delta_i\,E(\delta_i)}, \tag{16}$$

where index "i" denotes, values at the explosive-material boundary. Energy $E(\delta)$ is given by

$$E(\delta)=\int_v P_c(V)dV+3NK(T-T_0)+E_0+\frac{1}{2}\beta_0\delta^{-1/2}T^2. \tag{17}$$

Knowing δ as a function of r/r_0, other parameters are evaluated from Eq. (14).

RESULTS AND DISCUSSION

In this article, we have taken eight materials, of which five are radioactive in nature. Some materials are undergoing phase change and some are not undergoing phase change at high pressure. Existence of change in the normal behavior of materials under high pressure is established from the pressure-distance curve drawn using the Hugoniot equation of state.[1] Data used in this article is given in Table I.

Figure 1 shows the curves of Al, Fe, Cu, and stainless steel (Type 404). In this, Al shows a continuous pressure curve whereas the other three (Fe, SS, and Cu) show a discontinuity in the pressure curve which amounts to a phase change in the materials under high pressure.[3] Behavior of radioactive materials is shown in Fig. 2. Molybdenium (Mo), Palladium (Pa), U–Mo alloy, and Iridium (Ir) are taken and

TABLE II. Comparison table showing pressure values at few points.

	Pressure in Mb							
	Aluminum		Iron		Stainless steel		Copper	
r/r_0	With old equation	With revised equation	With old equation	With revised equation	With old equation	With revised equation	With old equation	With revised equation
0.9	0.618	0.5234	1.552	1.025	1.161	1.256	0.947	0.872
0.5	1.165	1.06	4.1754	4.43	2.1347	2.285	2.2354	3.085
0.1	6.957	7.352	[a]	164.825	18.834	51.235	22.2448	74.82

[a]Pressure discontinuity region.

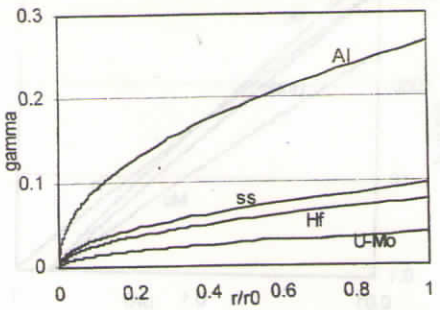

FIG. 5. Γ vs r/r_0 for different materials.

as it is seen Mo is not undergoing any change under high pressure whereas the rest of the materials are showing a discontinuity in their respective curves.

Now, to study those materials that are showing discontinuity, modified equations are used which are obtained by including thermodynamic properties of the material in the earlier equations [Eqs. (14), (4), and (5)]. Figures 3 and 4 show the curves of the respective material earlier shown in Figs. 1 and 2. Materials showing discontinuity in Figs. 1 and 2 give a smooth variation in the pressure value in Figs. 3 and 4 thereby giving a complete picture of the pressure change while the shock wave converges towards the center. At low pressure, both sets of equations give similar values of pressure, then the time discontinuity starts, and pressure values are changed. Table II gives a comparative study of pressure values for different materials at few points as shock converges to the center. Figure 5 gives the variation in Γ value with the distance r/r_0.

CONCLUSION

In studying the pressure variation across the shock using the Hugoniot equation of state, it has been observed that at high pressure, certain materials are showing sudden increase in pressure values or even a discontinuity in the pressure curve as the shock wave converges from the surface of the sphere to the center. This may be due to porosity or phase transition.[3] Since the materials that we had taken into account are not porous, the discontinuity is accounted for phase transition under high pressure only. As per the revised calculations, pressure variations have been evaluated, without any discontinuity even at high pressure. Pressure values calculated with both sets of equations of state at a few points are listed in Table II for a comparative analysis.

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