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EXPERIMENTAL DETERMINATION OF THE KINETICS OF RELAXATION PROCESSES
DURING THE SHOCK COMPRESSION OF CONDENSING MEDIA

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Introduction

The compression of condensing media by shock waves can be accompanied by such relaxation processes as phase changes, chemical reactions, etc. For a quantitative analysis of the evolution of a shock wave one must know the dependence of the rate of the process on the parameters of state of a particle of the specimen at each time. Two methods of finding the kinetic data directly from the experimental results are discussed in the present report.

In [1, 2] it is shown that if a series of profiles of the pressure $p(t)$ or the mass velocity $u(t)$ are determined for different fixed coordinates in a specimen through which a one-dimensional compression wave propagates then from these data one can reconstruct the dependence of the variation of the pressure and the specific volume V for each layer of the specimen; from the experimental data one determines the trajectory of the change of state of fixed layers of the substance in the coordinates $p-V$, with each point of this trajectory corresponding to a certain (and known) time. Then if the equations of state of the initial and final products are known one can construct in the same coordinates a grid of curves of the compression of mixtures with different concentrations of the final product. One thereby determines the concentration of the final product at each point of the trajectory of the change of state; i.e., for a chosen layer one determines the law of variation of the concentration of the initial and final products with time. By varying the parameters of the shock wave introduced into the specimen one can obtain a set of kinetic curves and from them choose a single empirical dependence of the rate of the relaxation process on the parameters of state of a particle of the substance.

A set of trajectories of the change of state provides complete information on the process, but to construct them one must make a large number of measurements with increased demands on the accuracy. At the same time, there are reasons to assume that in a number of cases (such as in the analysis of the propagation of shock waves in explosive materials); it is sufficient to know the initial rate of the process. In the present report it is shown that the initial behavior of the trajectory of the change of state can be determined from the experimentally measured profiles of the pressure or the mass velocity and from the law of variation of the

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velocity of the shock wave front. Since these measurements are compatible, one test can be enough for the determination of the initial rate of a process at a given pressure of shock compression.

§1. Construction of Trajectories of the Change of State

The methods of recording the law of variation of the mass velocity [3] and the pressure [4] at internal cross sections of a specimen under the conditions of its compression by a shock wave have been developed sufficiently fully by now. These experimental methods yield information pertaining to a fixed Lagrangian coordinate, and therefore it is convenient to conduct the further analysis of the results of Lagrangian coordinates.

The Lagrangian form of the equations of one-dimensional motion is

$$(\partial V/\partial t)_h = V_0(\partial u/\partial h)_t; \quad (1.1)$$

$$V_0(\partial p/\partial h)_t = -(\partial u/\partial t)_h, \quad (1.2)$$

where h is the Lagrangian coordinate of the particle (the initial distance from the surface through which the wave is introduced into the specimen to the particle under consideration); V_0 is the initial specific volume of the substance. The equations of motion do not include any information of the material and they are applicable everywhere except for points of discontinuity of the main variables, where the laws of conservation for the shock wave must be used. The equations of motion make it possible to calculate the variation in the specific volume $V(t)$ and the mass velocity $u(t)$ for different Lagrangian coordinates if the pressure field $p(h, t)$ is known from experiments or to calculate $p(t)$ and $V(t)$ if the mass-velocity field $u(h, t)$ is known. The missing variables [$u(h, t)$ and $V(h, t)$ when $p(h, t)$ is given from experiment or $p(h, t)$ and $V(h, t)$ when $u(h, t)$ is given] are obtained either by direct integration of Eqs. (1.1) and (1.2), as suggested in [1], or by integration of the transformed equations of motion with the introduction of two phase velocities, as suggested in [2]. In the latter case the equations of motion are transformed so that there is only one independent variable t in the explicit form

$$\begin{aligned} \left(\frac{\partial V}{\partial t}\right)_h + \frac{V_0}{c_u} \left(\frac{\partial u}{\partial t}\right)_h &= 0, & c_u &= -\left(\frac{\partial u}{\partial t}\right)_h \left/\left(\frac{\partial u}{\partial h}\right)_t\right.; \\ \left(\frac{\partial u}{\partial t}\right)_h - \frac{V_0}{c_p} \left(\frac{\partial p}{\partial t}\right)_h &= 0, & c_p &= -\left(\frac{\partial p}{\partial t}\right)_h \left/\left(\frac{\partial p}{\partial h}\right)_t\right. \end{aligned} \quad (1.3)$$

From an examination of the total differentials of $u(h, t)$ and $p(h, t)$ it is clear that c_u and c_p are the phase velocities of the propagation of fixed levels of mass velocity and pressure, respectively. The quantities c_u and c_p are determined not only by the properties of the medium but also by the character of the wave process.

The method of two phase velocities is most convenient for the determination of the trajectory of the change of state from a series of experimental pressure profiles. In this case the calculating system can be the following.

1. The trajectory of the shock wave front and lines of constant pressure (isobars) are constructed in the coordinates $t-h$ from the experimental data. Since there is always some scatter of the experimental data due to measurement errors, it is necessary to smooth them. It is preferable to smooth the experimental data by constructing auxiliary functions with necessarily small gradients, such as by the construction of $p(h)$ along trajectories shifted by a fixed time interval relative to the trajectory of the shock wave front.

2. By differentiation of the isobars the function $c_p(p)$ is determined for different Lagrangian coordinates.

3. Equation (1.3) is integrated using the functions $c_p(p)$ and the profiles $p(t)$. As a result, the mass-velocity profiles $u(t)$ are determined.

4. The procedure is repeated for the calculation of the profiles $V(t)$ from the series of profiles $u(t)$.

5. The profiles $p(t)$ and $V(t)$ for the same coordinate yield the values of p and V at different times. The trajectory of the change of state is thereby determined.

The accuracy of the calculation of trajectories of the change of state by the method of two phase velocities is reduced considerably in the case when the experimental pressure

profiles have smooth maxima produced by a relaxation process, as has been observed for glass [5] and explosive materials [3]. As the maximum is approached the quantities $(\partial p/\partial \tau)_h$ and c_p approach zero, and in the process the error of their determination and to an even greater degree the error of the determination of $(\partial u/\partial \tau)_h$ grow. The method of construction of trajectories of the change of state through the direct integration of Eqs. (1.1) and (1.2) is more acceptable in this case. Here the volume of the intermediate calculations increases owing to the necessity of a transition from the experimental profiles of $p(\tau)$ or $u(\tau)$ to the functions $p(h)$ and $u(h)$ for different times.

§2. Determination of Kinetic Functions

It is simplest to calculate the concentrations of the initial and final products along the trajectory of the change of state on the assumption of additivity [6] for a specific volume of their mixture. In this case it is automatically assumed that the time of existence of the possible intermediate products is negligibly small and heat exchange between the initial and final products is unimportant because of the high rate of the process characteristic of experiments with shock waves. In this case the fraction of the final product by weight is

$$\alpha = [V(p) - V_1(p)]/[V_2(p) - V_1(p)], \quad (2.1)$$

where V is the specific volume of the mixture; V_1 and V_2 are the specific volumes of the initial and final products. Since a relaxation process is accompanied by a relatively smooth pressure variation, with the assumptions made above the quantity $V_1(p)$ should be taken on the isentrope of the initial substance passing through the point on its shock adiabat corresponding to the initial state behind the shock wave front. The position of the isentrope of the final product is determined in the following way when the equation of state is known. After the shock compression of the initial substance and completion of the relaxation process at a constant pressure the change in internal energy $E - E_0$ and the specific volume V are connected by the equation

$$E(p) - E_0 - Q = (1/2)p(V_0 - V_1) - p(V - V_1), \quad (2.2)$$

where Q is the heat of the process. On the other hand,

$$E(p) - E_0 - Q = E_2(p) - E_0 + \int_{V_2(p)}^{V(p)} \frac{\partial E}{\partial V} dV, \quad (2.3)$$

where $E_2(p)$ and $V_2(p)$ are the internal energy and specific volume at the given p on some known (reference) curve of compressibility of the final product. Equating the right sides of (2.2) and (2.3), we obtain the expression for the determination of V on the isentrope of the final product.

Usually there is not complete information on the equations of state of the initial and final products, and it is necessary to make one or another assumption for the determination of the isentropes. If the specimen is monolithic then a good enough approximation for the region of moderate pressures of shock compression is the assumption that the shock adiabat and the relief isentrope coincide in the coordinates $p-u$ [7]. In this case the calculating equation for the determination of the isentrope has the form

$$dV_1 = -(\rho_1 c_1)^{-2} dp = -\rho_0^{-2} (c_0^2 + 4bp/\rho_0)^{-1} dp,$$

where ρ is the density; c is the velocity of sound; c_0 and b the coefficients in the expression for the shock adiabat in the form $U = c_0 + bu$ (U is the velocity of the shock wave).

Explosive materials are one of the most interesting subjects for kinematic studies. In a rather wide pressure range the isentropes of the explosion products are described well by a polytropic equation with a constant polytrope exponent n [8]. Then the quantity $\partial E/\partial V$ is determined by the equation [8]

$$\frac{\partial E}{\partial V} = \left(1 + \frac{\partial \ln D}{\partial \ln \rho_0}\right) / p \left(n - 1 - 2 \frac{\partial \ln D}{\partial \ln \rho_0}\right),$$

where D is the detonation velocity.

§3. Determination of Initial Rate of the Process

The method described above for the determination of the kinetics of relaxation processes requires the conducting of a large number of measurements with increased demands on the accuracy. Below we will show how to find the initial rate of a process with the minimum number of measurements.

Time differentiation of the quantities p and u along the trajectory of the shock wave front, with allowance for the equations of motion (1.1) and (1.2), gives

$$\frac{dp}{dt} = \left(\frac{\partial p}{\partial t}\right)_h + U \left(\frac{\partial p}{\partial h}\right)_t = \left(\frac{\partial p}{\partial t}\right)_h - \rho_0 U \left(\frac{\partial u}{\partial t}\right)_h; \quad (3.1)$$

$$\frac{du}{dt} = \left(\frac{\partial u}{\partial t}\right)_h + U \left(\frac{\partial u}{\partial h}\right)_t = \left(\frac{\partial u}{\partial t}\right)_h + \rho_0 U \left(\frac{\partial V}{\partial t}\right)_h. \quad (3.2)$$

Eliminating the quantity $(\partial u/\partial t)_h$ from (3.1) and (3.2), we obtain

$$\frac{dp}{dt} + \rho_0 U \frac{du}{dt} = \left(\frac{\partial p}{\partial t}\right)_h + \rho_0^2 U^2 \left(\frac{\partial V}{\partial t}\right)_h. \quad (3.3)$$

The time derivative of the specific volume of the mixture of initial and final products determined from (2.1) is

$$\frac{\partial V}{\partial t} = (V_2 - V_1) \frac{\partial \alpha}{\partial t} + \alpha \frac{\partial V_2}{\partial p} \frac{\partial p}{\partial t} + (1 - \alpha) \frac{\partial V_1}{\partial p} \frac{\partial p}{\partial t}. \quad (3.4)$$

Directly behind the shock wave front $\alpha = 0$. Then (3.4) gives

$$\left(\frac{\partial \alpha}{\partial t}\right)_h = \left(\frac{\partial V}{\partial t} - \frac{\partial V_1}{\partial p} \frac{\partial p}{\partial t}\right) / (V_2 - V_1). \quad (3.5)$$

Substitution of the quantity $\partial V/\partial t$ from (3.3) into (3.5) gives the expression for the initial rate of the process behind the shock wave front:

$$\left(\frac{\partial \alpha}{\partial t}\right)_h = \frac{\frac{dp}{dt} + \rho_0 U \frac{du}{dt} - \left(\frac{\partial p}{\partial t}\right)_h \left(1 + \frac{\partial V_1}{\partial p} \rho_0^2 U^2\right)}{(V_2 - V_1) \rho_0^2 U^2}. \quad (3.6)$$

The derivative $\partial V_1/\partial p$ is the derivative along the isentrope of the initial substance, so that $\partial V_1/\partial p = -\rho_0^{-2} \alpha_1^{-2}$, where α_1 is the Lagrangian velocity of sound. If the shock adiabat of the initial substance is assigned in the form $U = c_0 + bu$, then

$$\frac{dp}{dt} = \rho_0 b^{-1} (2U - c_0) \frac{dU}{dt}, \quad \frac{du}{dt} = b^{-1} \frac{dU}{dt}. \quad (3.7)$$

The use of (3.6) in (3.7) gives a simple equation for finding the initial rate of the process:

$$\left(\frac{\partial \alpha}{\partial t}\right)_h = \frac{\rho_0 (3U - c_0) \frac{dU}{dt} - b (1 - U^2 \alpha_1^{-2})}{b (V_2 - V_1) \rho_0^2 U^2}. \quad (3.8)$$

Thus, for an experimental determination of the initial rate of the process it is sufficient to record the pressure profile $p(t)$ (using a manganin pickup [4], for example) and the time dependence of the velocity of the shock wave front (using rheostatic pickups [3], for example). Since these measurements are compatible, one test can be enough for a determination of the initial rate of the process.

For a determination of the initial rate of the process for the mass-velocity profiles the quantity $(\partial p/\partial t)_h$ in (3.6) is replaced by $(\partial u/\partial t)_h$ with the help of (3.1). As a result, one obtains

$$\left(\frac{\partial \alpha}{\partial t}\right)_h = \frac{\frac{dU}{dt} (a_1^2 + 2U^2 - c_0 U) - b \left(\frac{\partial u}{\partial t}\right)_h (a_1^2 - U^2)}{\rho_0 b U a_1^2 (V_2 - V_1)}. \quad (3.9)$$

In [9] in a study of the damping of elastic forerunners an expression close to (3.9) connecting the damping of an elastic forerunner and the quantity $\partial u/\partial t$ behind its front with the velocity of plastic deformation was obtained in a somewhat different way.

In [10] differentiation of the parameters of state in a chemically reacting mixture along the trajectory of the shock wave front was used to predict the damping or acceleration of the shock wave. However, the final equation obtained in this case contains a derivative with respect to the coordinate — a quantity not directly measured — which considerably reduces the practical value of the result. Equation (3.8) can also be used to predict the acceleration of the shock wave. For this (3.8) is converted to the form

$$A dU/dt = B (V_2 - V_1) (\partial \alpha/\partial t)_h + (\partial p/\partial t)_h, \quad (3.10)$$

$$A = \frac{\rho_0 (3U - c_0) a_1^2}{b (a_1^2 - U^2)}, \quad B = \frac{\rho_0^2 U^2 a_1^2}{a_1^2 - U^2}.$$

Since $\alpha_1 > U > c_0$ for all materials in which a shock wave is possible, the quantities A and B are positive. Then it follows from (3.10) that when $V_2(p) > V_1(p)$ the shock wave will accelerate if $(\partial p / \partial t)_h > -B(V_2 - V_1) \partial \alpha / \partial t$ and die out if $(\partial p / \partial t)_h < -B(V_2 - V_1) \partial \alpha / \partial t$. When $V_2(p) < V_1(p)$ the shock wave will die out if $(\partial p / \partial t)_h < B(V_1 - V_2) \partial \alpha / \partial t$ and accelerate if $(\partial p / \partial t)_h > B(V_1 - V_2) \partial \alpha / \partial t$.

One can see that in both cases the wave will be steady if the change in state behind its front corresponds to the Michelson straight line $p = \rho_0^2 U^2 (V_0 - V)$.

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DYNAMIC COMPRESSIBILITY, ELECTRICAL CONDUCTIVITY, AND SOUND VELOCITY BEHIND A SHOCK FRONT IN KAPROLON

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In the present article we summarize the results of an experimental study of the properties of shock-compressed Kaprolon at pressures up to 570 kbar. The initial density of the samples was 1.14 g/cm^3 , and the initial resistivity was $2.0 \cdot 10^{14} \Omega \cdot \text{cm}$. The dynamic compressibility of the Kaprolon was investigated by the reflection method using an electrical contact technique [1] to measure the shock velocity in the samples. The experimental results characterizing the shock compressibility of Kaprolon are given in Table 1, in which u_{bar} is the particle velocity in the barrier, D and u are the shock and particle velocity in the Kaprolon, p is the pressure at the shock front, and $\delta = V_0/V$ is the compression ratio. Starting with $u \approx 1.5 \text{ km/sec}$, the dependence D(u) is practically linear for Kaprolon and can be described by the relation

$$D = 3.30 + 1.32u.$$

For $u < 1.5 \text{ km/sec}$ the curve of D versus u exhibits a curvature similar to that observed earlier for Teflon (Soviet Fluoroplast-4) [2].

The electrical conductivity of shock-compressed Kaprolon was measured by a procedure similar to one used earlier [3], which permits one to trace the evolution of processes taking place in the substance after passage of a shock through it. The measurement configuration

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