EQUATIONS OF STATE OF CONDENSED MEDIA

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We discuss a method for constructing the complete equation of state of a condensed medium thermodynamically, this method being based on the utilization of experimental data on shock compression and on the second law of thermodynamics. Determination of the accuracy of the solution so obtained, so far as its dependence on experimental error connected with the dynamical experiments and initial conditions was concerned, was carried out by means of statistical tests (Monte Carlo). The method is free from restricting assumptions as to the properties and nature of the medium under investigation. In the present paper, equations of state for W, Cu, and LiF are constructed. The results obtained are compared with experimental results and with equations of state based on models.

At the present time the theoretical calculation of thermodynamic properties of matter in the condensed state meets with serious difficulties owing to the presence of strong interparticle interactions in the quantum mechanical many-body problem [1]. This circumstance promotes the use of experimental, and primarily of dynamical methods of investigation [2], these being based on ways of obtaining the required states through compression and irreversible heating of the material in the front of a strong shock wave. A characteristic feature of dynamical experiments, connected with the recording of the kinematic parameters of shock wave propagation, is the fact that such experiments give immediate information that concerns only the caloric equation of state $E = E(P, V)$, from which the very important thermodynamic parameter — the temperature T — is absent, it being necessary for the calculation of this parameter to resort to models to represent the properties and nature of the material under investigation.

All semiempirical methods of constructing an equation of state on the results of dynamical experiments that are exploited at present are based on the division of the total pressure and the internal energy into "thermal" and "cold" components, with subsequent deductions based on models of theoretical representations concerning the nature of the functional dependence of the thermal factors on the specific volume and the temperature. At the same time the experiments in question are employed as a check on the constants and functions entering the assumed equation of state, purely theoretical expressions often being employed as terms in the latter. The criterion for the merit of an equation of state constructed in this way is the agreement between the Hugoniot adiabat of the substance under study as measured and as calculated by theoretical considerations. However, it is possible for a situation to arise when different equations of state, which give a sufficiently good description of the original experimental material, lead to different values of the Grüneisen coefficient and of the temperature of a shock-compressed medium [3]. Moreover, in the framework of equations of state based on models it is rather difficult to take account of polymorphic transitions, melting [4], possible departures of the compression curves from smoothness, filling of electron energy bands induced by anomalies, and phase transitions of an electron type [5]. To take account of experimental information relating to these effects would require the introduction of new terms and a considerable modernization of known models of equations of state.

A thermodynamically complete equation of state can, however, be constructed on the basis of experimental data alone, without the introduction of theroretical assumptions concerning the properties and the nature of the medium under investigation [6]. The thermodynamic universality of the method under discussion enabled us to construct equations of the state for media with very different physical properties: metals of the transitional (W) and the main (Cu) groups of the periodic system and an ionic crystal (LiF).

TABLE 1. Parameters of the Equations of State (1.4)

	W		Cu		LiF	
e_{00}	−.297987230↑	2	−.489321103↑	1	.326508990↑	3
e_{10}	.102046666↑	4	−.131722742↑	3	−.253883236↑	4
e_{20}	−.102685511↑	5	.282661005↑	4	.645766095↑	4
e_{30}	.325694626↑	5	−.113090465↑	5	−.536077139↑	4
e_{01}	.223170240↑	0	.156770568↑	0	−.230157248↑	1
e_{11}	−.656993225↑	1	−.426786389↑	1	.976449861↑	1
e_{21}	.552261155↑	2	.329967175↑	2	−.802928927↑	1
e_{02}	−.564755165↑	−3	−.220433040↑	−3	.725437439↑	1
e_{12}	.954237483↑	−2	.431656107↑	−2	−.122096372↑	−1
e_{03}	.386205157↑	−6	−.560081364↑	−7	−.947216102↑	−5

Note: −.2979837230   2 = −29.79837230

TABLE 2

$P \cdot 10^{-12}$, dyn/cm^2	σ	$C \cdot 10^{-5}$, cm/sec	$\left(\frac{dP}{dV}\right)_s \cdot 10^{-12}$, g^2/cm$^4 \cdot$sec^2	$\left(\frac{dE}{dV}\right)_P \cdot 10^{-10}$, erg/cm^3	$\left(\frac{dE}{dP}\right)_V$, $\frac{cm^3}{g}$	γ
			Data of [14]			
1.31	1.065	5.61	1.039	264	0.025	1.64
0.31	1.017	4.79	0.84	210	0.027	1.87
2.62	0.928	—	—	—	—	1.13
0.91	0.871	—	—	—	—	1.26
		Results of calculations according to the equation of state (1.4)				
1.31	1.065	5.75	1.465	257.3	0.0269	1.79
0.31	1.017	4.73	0.852	206.3	0.0279	1.83
2.62	0.928	—	—	—	—	0.814
0.91	0.871	—	—	—	—	1.576

1. General Introduction to the Method for Constructing an Equation of State. The measurement of two of the five parameters characterizing the propagation of a steady shock through a material under study enables one, through use of general conservation laws, to determine the internal energy E as a function of the pressure P and the specific volume V in each particular experiment [2]. By carrying out suitable measurements with various initial conditions and intensities of the shock wave one can determine the caloric equation of state E = E (P, V) in a region of the PV-plane that is covered by Hugoniot adiabats [2].

Measurements of the initial states of the material can be made by performing shock-wave experiments at various temperatures T_0 and densities that are related to T_0 by the law of thermal expansion [7, 8]. Characteristic of this procedure is the fact that the parameter domain that is accessible to experimental determination is widened only insignificantly. Far more effective for this purpose is the compression of a substance by a shock wave, the substance being in the form of a loosely dispersed powder, as this makes it possible to increase the entropy jump greatly and thus considerably to extend the region of the PV-plane that is accessible to experiment [9]. An additional extension of the region in question can be accomplished both by use of incident and reflected shocks and by the recording of the unloading adiabats of a shock-compressed substance [2]. In the latter case the parameters of the medium (distinguished by dashes) in the centered rarefaction wave are related to the parameters behind the shock front by Riemann's integrals

$$V' - V = \int_{P'}^{P} \left(\frac{du}{dP}\right)^2 dP, \qquad E - E' = \int_{P'}^{P} P \left(\frac{du}{dP}\right)^2 dP$$

which are calculated according to the experimentally known functional relationship between the particle velocity and the pressure u = u (P).

Thus a dynamical experiment enables one to determine an equation of state directly only in a caloric form: E = E (P, V). In this form the equation of state can be used for hydrodynamic calculations of adiabatic flows, since in this case it is not necessary to introduce the temperature (through an equation of state) in order to close the system of hydrodynamical equations.
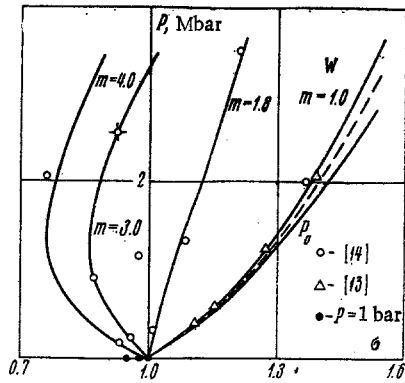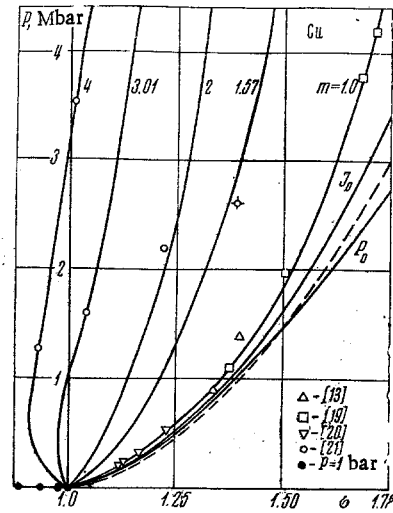
Fig. 1

Fig. 2

The internal energy, however, is not a thermodynamic potential with respect to P−V variables, and for the construction of the complete thermodynamics of the system under study the supplementary depen-- dence T = T(P, V) is required. Measurement of the temperature together with other parameters of shock compression in an overwhelming number of cases does not seem to be possible because of the small length of the free path of light radiation in condensed media, since light radiation of a shock-compressed medium is absorbed in front of the shock wave and is not sufficient for registration. Even in optic- ally isotropic and transparent media − ionic crystals − the equilibrium temperature can be measured only in a comparatively narrow pressure range (0.5-1.0 Mbar), outside of which light radiation has an essentially nonequilibrium character [10].

The shock-compression temperature T = T (P, V) can be determined directly from the caloric equation of state E = E (P, V) without introducing restrictive assumptions as to the properties, nature, and phase constitution of the material under investigation [9, 6]. Taking account of the dependence E = E (P, V), as known from experiment, we easily obtain, on the basis of the second law of thermodynamics:

$$\left[P + \left(\frac{\partial E}{\partial V}\right)_P\right]\frac{\partial T}{\partial P} - \left(\frac{\partial E}{\partial P}\right)_V\frac{\partial T}{\partial V} = T \tag{1.1}$$

The solution of this linear inhomogeneous partial differential equation with variable (known from ex- periment) coefficients is the functional dependence T = T (P, V). The solution of (1.1) is constructed from the solution of the characteristic system of ordinary differential equations

$$\frac{dP}{dV} = -\frac{P + (\partial E/\partial V)_P}{(\partial E/\partial P)_V} \tag{1.2}$$

$$\frac{dT}{dV} = -\frac{T}{(\partial E/\partial P)_V} \tag{1.3}$$

Equation (1.1) or Eqs. (1.2), (1.3) are supplemented by the boundary conditions: the temperature must be given in a region either where it is possible to make a reliable calculation of the equation of state by the method of statistical physics or where the temperature is known from experiment. For condensed media the latter case is preferable. In calculations in the present paper the temperature is prescribed as a func- tion of the specific volume on the isobar: P = 1 atm, where experimental data on the thermal expansion V_0' = $V_0'(T_0')$ of the substance is used.

Supposing that N experimental points $\{ E_i \pm \Delta E_i, P_i, V_i \}_{i=1}^N$, randomly distributed over the PV- plane, are known, we construct a caloric equation of state in the form of a power polynomial
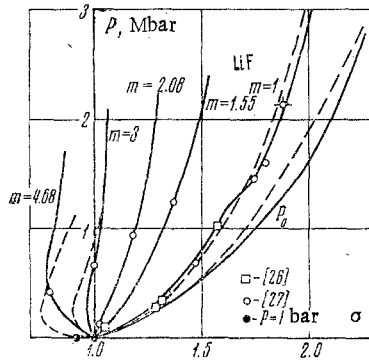
$$E^q(P, V) = \sum\sum_{k+l\leqslant q} e_{kl}V^k P^l \tag{1.4}$$
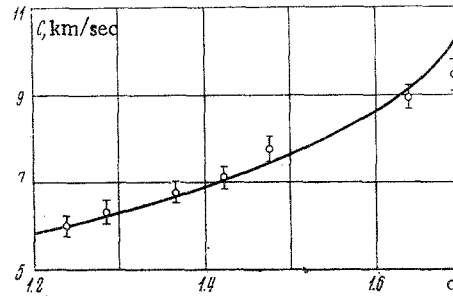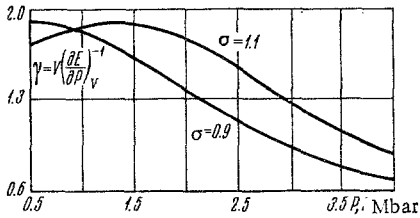
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Fig. 3

Fig. 4

Fig. 5

The coefficients e_{kl} (see Table 1, where P is expressed in 10^{10} dyn/cm^2, V in cm^3/g, and E in 10^{10} ergs/g) were found from the condition for a minimum $\partial S/\partial e_{kl} = 0$ of an element of the best approximation S

$$S = \sum_{i=1}^{N} \omega_i{}^2 [E_i - E^q(P_i, V_i)]^2 \qquad (1.5)$$

which leads to the requirement that Gauss' system of normal equations be solved. Difficulties, familiar in the one-dimensional analogue of the problem, that are connected with the poor conditionality of the matrix of the system of normal equations can be removed by conversion to orthogonal Chebyshev polynomials [6].

The degree q of the polynomial in (1.4) is chosen by an analysis of the experimental data, using the statistical criterion of Fisher's relationship [11].

An equation of state in the form (1.4) can be utilized immediately in hydrodynamic calculations. Shock adiabats of the material under study are determined by numerical solution of the Rankine—Hugoniot equation

$$E^q(P, V) - E_0(P, V) = {}^1\!/_2 (P + P_0)(V_0 - V) \qquad (1.6)$$

where the subscript 0 refers to the state in front of the shock front and E^q (P, V) is taken in conformity with (1.4). The characteristic equation (1.2) determines adiabatic compressibility and consequently the sound speed $C^2 = -V^2 (\partial P/\partial V)_S$ in a shock-compressed medium. Numerical integration of Eq. (1.2) enables one to find the isentropic unloading curves. Using (1.4), one can calculate the right-hand sides of the system (1.2), (1.3) and integrate it throughout the region for which relation (1.4) holds.

The dependence of the accuracy of the results obtained on errors in the original data is determined by the Monte Carlo method through a modeling of the random structure of the measuring process by a computer [6]. A uniform estimate of the accuracy of the solution for a given configuration of the distribution of experimental points in the PV-plane and of a specific level of experimental error was made.

2. Results of the Construction of Equations of State. In view of the thermodynamical universality of the method under discussion it is possible to construct the equation of state of media having very different physical properties by a single procedure. These media were simple (Cu) and transitional (W) metals and an ionic crystal (LiF). Nickel has been discussed earlier in [12].

In the construction of the equations of state of the materials under discussion here we used experimental data on the shock compression of solid and porous samples (see points in Figs. 1, 2, 3). Moreover, points corresponding to the solid and liquid phases on the atmospheric pressure isobar were used. In calculating the parameters of these points, the internal energy of the solid and liquid phases and also the latent heat of melting at P = 1 bar were assigned for tungsten in accordance with [15, 17, 34], for copper [22-24], and for LiF [28]. For the determination of the bulk thermal expansion curves (initial data for the temperature calculations) the data of [15, 17] for tungsten, of [16, 24, 25] for copper, and of [28] for LiF were employed. In view of the complete lack of experimental data on liquid tungsten, the single point used here in the calculation of the internal energy in the domain of the liquid phase [its weight in the construction (1.5) was $\omega = 0.5$] was based on the theoretical data of [17]. The value $\Delta\rho_m/\rho_m = 0.02$ was taken as the
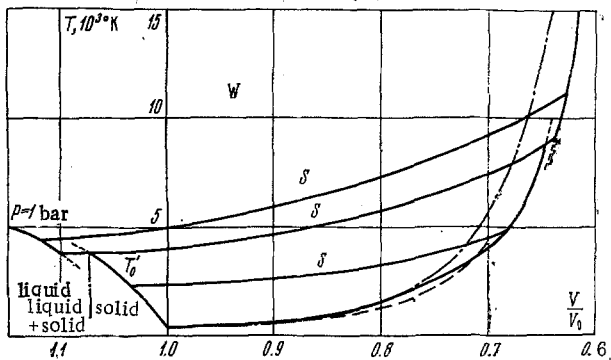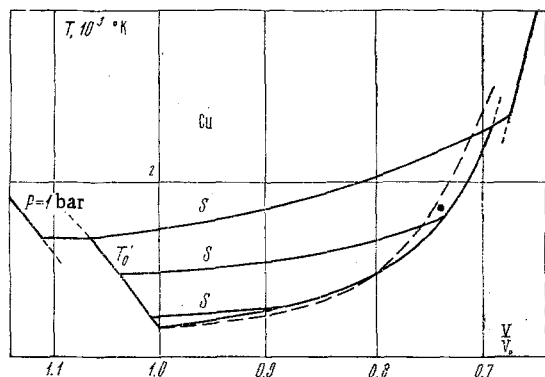
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Fig. 6

Fig. 7

change in density upon melting, taking account of the presence of a correlation between this quantity and the type of crystal lattice [18]. Parameters of the equations of state (1.4) that were constructed are given in Table 1.

Statistical analysis showed that for the description of available experimental material it is sufficient to take q = 3 in (1.4) for W, Cu, and LiF. Results of calculations of the Hugoniot adiabats for various initial porosities m $=\rho_0*/\rho_0$ (ρ_0* being the density of the compact material) are shown in Figs. 1-3. The constructed shock adiabats of W, Cu, and LiF have an "anomalous" form [2] with an alternating derivative (dP/dσ) along the Hugoniot adiabat which, in the framework of the model equations of state, is connected with the dependence of the Grüneisen coefficient γ on the temperature [14]. The equation of state obtained for lithium fluoride describes (see the kink in the shock adiabat m =1 on Fig. 3) a polymorphic transition to the denser crystalline modification (of the type CsCl [27]); the dashed curve in Fig. 3 is a plot of the shock adiabat of [27]. The merit of the constructed equations of state is determined by the correspondence between calculated and experimental measurements of states of shock compression. The manner in which the calculated Hugoniot adiabat passes through experimental points that were not used in the construction of (1.4) (these "control" points are indicated by crosses in Figs. 1, 3) determines the interpolation properties of the equations of state we have obtained. In both cases the deviation in the degree of compression (at the specified pressure) did not exceed 1-2%, which is obviously within the limits of experimental error [2].

Measurements of the sound speed in shock-compressed tungsten [14] and copper [29] provide an additional possibility for the comparison of these quantities with results of the calculation of isentropic compressibility according to Eq. (1.2). Results of such a comparison for Cu are shown in Fig. 4 [solid curve, calculated according to (1.2), (1.4)], and for W in Table 2 where, moreover, a comparison is made between derivatives of the internal energy found in [14] and the Grüneisen coefficient γ and results of calculation of these quantities according to the equation of state (1.4).

An important characteristic of matter, used in all model equations of state, is the cold compression curve $P_0(V)$, the dependence of the pressure on the specific volume along the isotherm T =0°K. The method of extracting this dependence from results of a dynamical experiment is based on the numerical solution of a differential equation for P_0, which is derived from model representations of the dependence of the frequency spectrum of harmonic vibrations on the specific volume. Another way of determining $P_0(V)$ is to prescribe this curve in the form of a polynomial [3]

$$P_0(V) = \sum_{i=1}^{7} a_i \left(\frac{V_{0k}}{V} \right)^{i/3+1} \qquad (2.1)$$

where the coefficients a_i are found from characteristics of the substance under normal conditions, the data of dynamical experiments, and the Thomas−Fermi−Dirac theory. Taking account of the fact that the isotherm T =0° K coincides with an isentrope, we can obtain the cold compression curve according to this approach by integrating (1.2) for T_0 =0° K. The $P_0(V)$ obtained in this way are shown in Figs. 1, 2, 3 (dashed curves), where they are compared with the cold compression curves constructed according to (2.1). Satisfactory agreement between these curves is evident. It is significant that the zero isentrope for copper (Fig. 2), constructed in this way, does not exceed the upper bound of the zero isentrope J_0 obtained by measurements of the sound speed [29]. We remark that the method of constructing the cold compression curve under consideration presupposes an extrapolation of the equation of state (1.4) out of the region where experimental information is specified, and for that reason the curve is an approximation in the high-pressure domain.
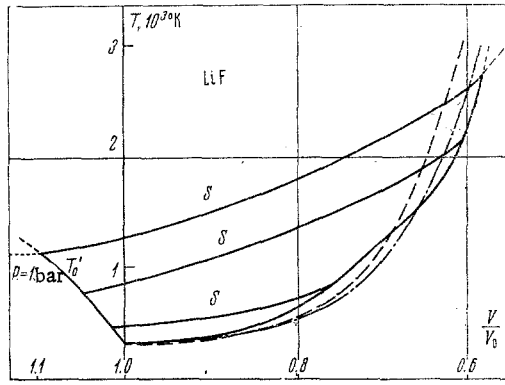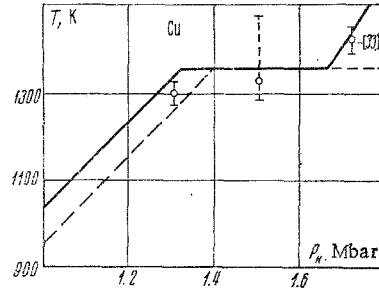
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Fig. 8   Fig. 9

The basic premise underlying an equation of state in the Mie–Grüneisen form is the assumption that the Grüneisen coefficient $\gamma = V (\partial E / \partial P) v^{-1}$ is independent of the temperature [2]. This assumption breaks down in the domains of high pressures and low densities. As an illustration of this fact, calculations of γ were made for various pressures (temperatures) on the isochors: $\sigma = 1.1$ and $\sigma = 0.9$, using the equation of state constructed for W (Fig. 5). It is evident that, in contrast to the assumption of the Mie–Grüneisen theory ($\gamma = $const for V = const), the coefficient γ decreases with increasing pressure, tending in the limit to the value 2/3 that is characteristic for an ideal gas. The small increase in γ during the initial portion of the isochor $\sigma = 1.1$ is apparently connected with experimental error. A similar increase in γ on the adiabat m = 1.8 was noted in [14].

Temperature calculations were made by simultaneous integration of Eqs. (1.2) and (1.3) by computer. The initial temperature T_0' was determined by feeding the curve of thermal expansion at P = 1 bar (see Figs. 6, 7, 8) into the computer as the initial value of the specific volume $V_0' > V_0 *$ on the atmospheric pressure isobar. With these initial conditions the characteristic system (1.2), (1.3) was integrated by Adams' numerical scheme up to the intersection of the isentrope with the shock adiabat for solid matter in the PV-plane. In the TV-plane a few isentropes, distinguished by the index s, are shown in Figs. 6, 7, 8. In Fig. 6 the calculated temperature on the shock adiabat for tungsten (solid curve) is compared with the calculation according to the Mie–Grüneisen theory [13] (dashed curve) and the calculations of [30] (dot-dash curve). In Fig. 7 the temperature obtained for Cu is compared with the Mie–Grüneisen theory [13] (dashed curve) and the "free" volume theory [19] (point). In Fig. 8 the temperature for LiF is compared with the results of [10] (dashed curve) and of [26] (dot-dash curve). It is evident that at moderate compressions there is good agreement between the calculated temperature and results of calculations according to the theories cited. The lack of smoothness of the plot of the temperature for LiF in Fig. 8 is related to the polymorphic transition mentioned above.

The characteristic "step" in the temperature on the shock adiabat m = 1 (Fig. 6, 7) is caused by melting of the material behind the shock front. The magnitude of the change in the specific volume with melting on the Hugoniot adiabat corresponds qualitatively with experimental results for ionic crystals [31].

Experimental [31] and theoretical [4] studies of the melting of matter under shock compression indicate that for the entropy change along the melting curve dS/dT > 0. The lack of detailed experimental data in the area of the melting curve results in this curve's coinciding with an isentrope in the calculations under discussion. The actual temperature step is therefore located somewhat higher than those depicted in Figs. 7 and 6. * For pressures that exceed by an order of magnitude the value corresponding to the intersection of the shock adiabat with the Lindemann–Gil'varr melting curve in the phase diagram [10] the calculations under discussion yield undistorted results for the melting effect in view of the fact that the solution of Eq. (1.1) is constructed by the method of characteristics and the temperature on the isentrope does not depend on the solution in the neighboring region through which the isentrope in question does not pass.

Figure 9 shows the temperature of copper after isentropic expansion (unloading) of the shock-compressed material from the state on the Hugoniot adiabat with m = 1 to the state at atmospheric pressure as a function of the pressure behind the shock front. Relation (1.2) was used for the construction of the

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*The character of the kink in the isentropes at their intersection with the two-phase region results in a shift of the step in Figs. 7 and 6 into the region of higher temperatures [32].
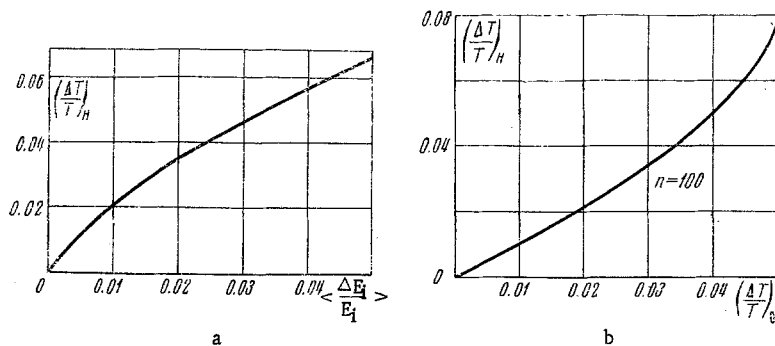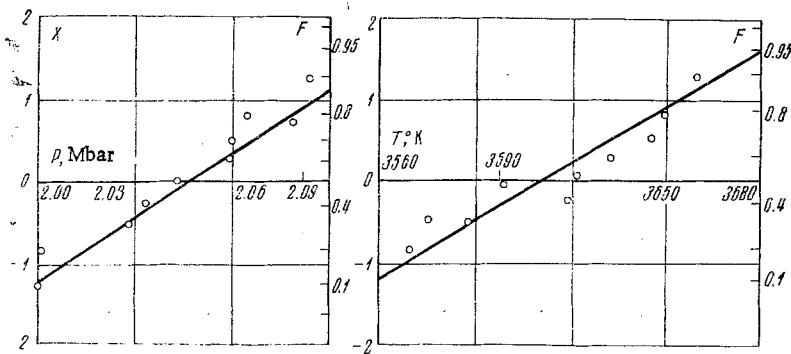
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Fig. 10

Fig. 11

isentrope. The temperature dependence of the density on the isobar P =1 bar was taken from experiments on thermal expansion. Represented on the same diagram are the data of [13] (dashed curve) and results of Tyler's experiments [33] that measured the temperature in the unloading wave in shock-compressed copper by photoelectric means.

The agreement between theoretical and experimental results on temperature measurements in the unloading wave is an indirect confirmation of the correctness of the method of calculating the temperature of optically dense media that has been adopted here. Experiment [33] and the present calculations indicate, moreover, that there is thermodynamic equilibrium during the expansion of shock-compressed copper from a state on the Hugoniot adiabat to normal conditions (P =1 bar).

As an example, Fig. 10 shows results of the determination of the accuracy of the solution for W by the method of statistical sampling (Monte Carlo). Under the assumption of equal accuracy of the measurements various values of the relative error $\langle \Delta E_i/E_i \rangle$ were given the dynamical experiments sequentially. In each case the size n of the sample used for estimating the error of the calculations was equal to 100. The dependence of the error in the temperature determination for a certain characteristic point of the shock adiabat m =1 on the average error $\langle \Delta E_i/E_i \rangle$ of the dynamical experiments is plotted in Fig. 10a. Gaussian distribution formulas were used for the calculation of the mathematical expectation and the variance of the random variables (temperatures). The normality of the distribution law of a random variable is verified in the probability text [11] (Fig. 11). The dependence of the error in temperature determination $(\Delta T/T)_H$ on the error in the initial data at P =1 bar is represented in Fig. 10b. A certain (very slight) growth along the isentrope is connected with the divergence of the fan of characteristics in the TV-plane. A similar statistical analysis of the accuracy of the temperature determinations for Cu and LiF was also made. On the basis of estimates made by the Monte Carlo method we can conclude that, on taking account of the accuracy and character of the distribution of experimental data in the PV-plane, the error in the determination of the temperature of a condensed medium on the adiabat with m =1 is of the order of 5-8%. This is confirmed independently by a comparison with temperature calculations according to model theories.

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