Shock compression of porous iron, copper, and tungsten, and their equation of state in the terapascal pressure range

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Shock waves were used to determine experimentally the compressibility of porous iron and tungsten samples in the terapascal pressure range. The maximum pressures of 2.2 and 1.4 TPa were reached for tungsten ($\rho_{00} = 6.27$ g/cm^3) and iron ($\rho_{00} = 2.24$ g/cm^3) as a result of interaction of these metals with very strong shock waves. A comparison of the parameters of a standard substance and of the two investigated metals yielded the compressibility of these metals. New experimental data were obtained for iron and tungsten by absolute methods at pressures up to 700 GPa and for copper up to 250 GPa. The complete experimental data, throughout the full P–ρ–E range investigated by dynamic methods, were described by the equations of state.

INTRODUCTION

From the practical point of view the interest in a strongly nonideal dense plasma is primarily due to various energy projects requiring the knowledge of the plasma properties in a wide range of parameters in the pressure–temperature (P–T) diagram. Experiments on porous metals provide information on little-investigated and difficult-to-study ranges corresponding to densities close to normal and temperatures of a few to tens of electron volts.

The simplest method of covering this range is the use of shock waves for the determination of the compressibility of substances. However, even the experimental data obtained by this method on the high-pressure behavior of substances, particularly metal elements, are clearly insufficient. For example, studies carried out by dynamic methods have not yet exceeded the range reported in Refs. 1 and 2, where pressures of 0.3–0.5 TPa have been applied to metals with initial densities 3–4 times lower than the crystallographic values.

The next major step in the scale of pressures in devices employing explosive substances, based on acceleration of strikers by the explosion products, is so difficult that it is a very daunting task to obtain quantitative results with the precision expected of such experiments. Therefore, in recent years the attention of investigators has turned to studies of the compressibility of substances by a comparative method[3–10] using very strong shock waves. In this case a standard material is a substance which satisfies the following conditions: the adiabat must have been determined reliably (by calculation and experimental methods) throughout the range of pressures required for investigation; the adiabat plotted in terms of the P–u coordinates (i.e., the coordinates of the pressure and the mass velocity of substance) should lie as close as possible to the adiabat of the investigated substance; the standard material should have a high compressibility, because this ensures smaller errors in the determination of the compression parameters of the investigated material (Ref. 3).[1)]

In a series of investigations of the comparative compressibility[3–10] the attention has been concentrated on shock adiabats of continuous substances ($\rho_0 = \rho_{\text{cryst}}$). The first and essentially the only paper reporting the results of dynamic measurements on porous substances was a brief report[11] of the results of a study of porous copper ($m = \rho_0/\rho_{00} = 3.4$) at pressures up to ~ 2 TPa.

In this relative method for the determination of the compressibility the positions of shock adiabats of metals and some compounds have been determined quite reliably in spite of the absence of direct experimental data, at pressures 1–6 TPa, on the shock adiabat of a standard substance in the form of a screen (with the precision required for our purpose). This is demonstrated by a fully satisfactory agreement between the values of the compressibility of metals, silica, and other compounds,[3–5,8–13] obtained by different groups of investigators in the Soviet Union and in the United States, who used different standard materials (Pb, Fe, and Al in the Soviet Union and Mo in the USA) and found their adiabats using different calculation models. This is supported also by the first measurements of the absolute compressibility of aluminum[13,14] and molybdenum,[8] which—though not yet very accurate—are not in conflict with the interpolation dependences adopted for standard metals.

Therefore, there are grounds for assuming that the shock compression curves of lead, iron, and aluminum used in the Soviet Union as the standard adiabats[3–5,11,12] do represent their real positions at pressures $P = 1$–6 TPa and can be used to determine the compressibility of porous substances.

We shall report new data obtained using both absolute and relative (comparative) measurement methods.

EXPERIMENTAL RESULTS

We prepared samples by compacting powders of metals until the required initial density ρ_{00} was reached. We used powders of iron, copper, and tungsten of 98.7, 99.7, and 99.6% purity, respectively. The dimensions of the samples used in the absolute measurements were as follows: the diameter was 12–16 mm and the thickness was 3–5 mm. In some of the copper experiments we used much larger samples with a diameter up to 250 mm and a thickness up to 80 mm.

Special methodological investigations established variation of the grain size within the range 1–100 μm, which did not affect the velocity of a shock wave, at least above pressures of 5 GPa. The positions of the shock adiabats were independent of the thickness of the investigated sample. Taken together, these data demonstrated that the size of the samples did not affect the results of the experiments and that the propagation of a shock wave in the process of compression of porous materials was a steady-state process. Table I

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TABLE I.

Investigated material	Screen material, u (km/s)	Porosity, $m=\rho_0/\rho_{00}$	Shock compression parameters				
			D, km/s	u, km/s	P, GPa	$m\sigma$	σ
Fe, ρ_0=7.85 g/cm^3	Al, 2.72	2.45	5.24	3.13	52.5	2.479	1.012
	Al, 2.72	1.78	5.47	2.72	65.6	1.994	1.120
	Al, 1.74	1.81	3.85	1.95	32.6	2.027	1.120
	Al, 1.63	1.88	3.57	1.75	26.1	1.961	1.043
	Al, 1.63	1.83	3.44	1.70	25.1	1.977	1.080
	Al, 1.63	3.01	3.21	2.05	17.2	2.767	0.920
	Al, 2.82	2.49	5.46	3.33	57.3	2.563	1.030
	Al, 2.41	1.83	6.32	3.15	85.1	1.993	1.089
	Fe, 4.15	1.72	9.66	5.13	226.0	2.132	1.239
	Fe, 4.15	1.83	9.51	5.22	213.0	2.216	1.211
	Fe, 4.15	2.21	9.28	5.52	182.0	2.468	1.117
	Fe, 4.15	2.99	9.46	5.46	146.0	2.642	0.884
	Fe, 7.67	1.82	14.85	9.10	583.0	2.584	1.420
	Fe, 2.90	2.90	15.87	10.75	462.0	3.099	1.069
W, ρ_0=19.35 g/cm^3	Fe, 7.67	3.03	13.19	9.22	773.0	3.322	1.097
Cu, ρ_0=8.93 g/cm^3	Al, 7.17	3.00	11.02	7.58	249.0	3.203	1.068
	Al, 4.12	3.10	7.20	4.78	99.0	2.975	0.960
	Fe, 4.15	1.57	9.31	4.83	256.0	2.080	1.325
	Fe, 4.15	2.00	9.28	5.21	216.0	2.280	1.140
	Fe, 4.15	3.00	8.90	5.81	154.0	2.880	0.960
	Fe, 4.15	4.00	8.96	6.15	123.0	3.190	0.800
	Fe, 7.67	1.50	14.16	8.77	739.0	2.627	1.751
	Fe, 7.67	2.02	14.36	9.56	607.0	2.992	1.481
	Fe, 7.67	4.00	14.40	11.23	361.0	4.542	1.136

gives the experimental results of our determinations of the compressibility. This table includes the data taken from Ref. 2, reworked using refined initial parameters for the screens and investigated samples.

The procedure used in determination of the relative compressibility was similar to that described in Ref. 11. An aluminum screen 100 mm thick was located in the path of a strong shock wave and cylindrical samples (with diameter 250 mm and width 80 mm) were placed on this screen. The transit time of a shock wave across the screen and samples was recorded using electric-contact sensors. We ensured symmetry of the shock wave for a period of $\sim 0.2\,\mu$s in a field of linear dimensions ~ 1000 mm. The shock wave was practically undamped: the corrections in the course of conversion from the average to the instantaneous velocities at the screen–sample interface were at most 300 m/s. The initial density of the samples was close to that of powders and was selected to ensure ease of filling of cylindrical containers with powders.

The measured relative compressibility values are listed in Table II. The data for copper were taken from Ref. 11. In determining the metal compression parameters we used the equations of shock adiabats (for screens) in the form $D = c_0 + a_1 u + a_2 u^2$ with coefficients given in Table III. In plotting the P–u diagrams we made a correction to allow for a small difference between the positions of isentropes (double compression adiabats) and the results obtained by mirror reflection of the dynamic adiabat of aluminum.

EQUATION OF STATE

In describing the experimental results in a wide range of states it is customary to use quite complicated multiparameter equations[2,16,17] based, as a rule, on representation of the energy E and pressure P in the form of cold (corresponding to the elastic interaction between atoms) and thermal components. The latter are divided into nuclear and electronic terms, and the equations themselves contain a particular set of constants determined from the experimental results. It is usual to use the cold components (for which experimental results are not available) as the initial approxi-

TABLE II.

Investigated material	Screen material Al, u (km/s)	Porosity, $m=\rho_0/\rho_{00}$	Shock compression parameters				
			D, km/s	u, km/s	P, GPa	$m\sigma$	σ
Fe, ρ_0=7.85 g/cm^3	19.98	3.52	28.62	21.54	1.375	4.042	1.148
	14.72	3.26	22.35	15.52	0.835	3.272	1.004
W, ρ_0=19.35 g/cm^3	20.35	3.09	21.55	16.51	2.230	4.276	1.384
Cu, ρ_0=8.93 g/cm^3	24.65	4.00	36.00	26.10	2.098	3.636	0.909
	21.29	4.00	31.10	22.80	1.583	3.747	0.937
	19.85	4.00	28.71	21.40	1.372	3.927	0.982
	17.09	3.10	24.34	17.25	1.209	3.433	1.107

TABLE III.

	ρ_0, g/cm^3	c_0, km/s	a_1, (km/s)$^{-1}$	a_2, (km/s)$^{-1}$	Range of u, km/s	Ref.
Al	2.71	5.333	1.356	–	<5.8	[15]
Al	2.71	5.28	1.218	–	>11	[12]
Fe	7.85	3.664	1.79	−0.0337	<7.27	[15]
Fe	7.85	5.445	1.30	–	$7,27<u<25$	[3, 5, 10]
Cu	8.93	3.899	1.52	−0.009	<15	[15]
Cu	8.93	5.924	1.25	–	$15<u<25$	[4, 5, 10]
W	19.35	4.02	1.22	–	<15	[15, 9]

mation and to allow for the thermal factors using the Grüneisen coefficient

$$\gamma = V(\partial P/\partial E)_V.$$

In the present study we shall describe the experimental results using a P–V–E equation of state of the following type:

$$P = P^\Gamma + \xi_z^\Gamma (V - V^\Gamma), \tag{1}$$

$$PV = P^\Gamma V^\Gamma + z(E - E^\Gamma). \tag{2}$$

The index Γ in these equations represents quantities on the reference (isentrope or shock adiabat) curve, the equation of which is assumed to be known; z is a constant. Equation (2) is a series expansion (through first order in the energy E) of the product PV for the reference curve. This expansion is taken along connecting lines described by Eq. (1). The slope of these lines is $\xi_z^\Gamma (\equiv dP/dV)$ and it is found from the condition that $\partial(PV)/\partial E = z$ along the connecting lines at the points where these lines intersect the reference curve. Transformations yield the following expression for ξ_z^Γ:

$$\xi_z^\Gamma = \rho^\Gamma (c^\Gamma)^2 \left[z - (z+1) \frac{\gamma^\Gamma P^\Gamma}{\rho^\Gamma (c^\Gamma)^2} \right] \Big/ (\gamma^\Gamma - z), \tag{3}$$

where ρ is the density and c is the velocity of sound.

The equation for the straight connecting lines with the slope ξ_z^Γ is given by Eq. (1). Since these lines are chosen, by definition, so that $\partial(PV)/\partial E = z$ is satisfied along these lines near the reference curve, it follows that expansion of the product PV as a series in terms of E (up to the first order term) yields Eq. (2). It should be pointed out that the choice of the connecting lines satisfying the condition $\partial(PV)/\partial E = z$ near the reference curve is not unique; they are chosen to be straight lines for the sake of simplicity. It is clear from Eqs. (1) and (2) that if $\xi_z^\Gamma > 0$, then in the limit $V \gg V^\Gamma$ the equation of state is

$$PV = zE. \tag{4}$$

This equation gives the maximum compression on the shock adiabats $h = 1 + 2/z$ or the slopes $D'_\infty = 1 + z/2$. If we select $z = 0.5$, we obtain the maximum slope $D'_\infty \approx 1.25$. For $z \approx 0.5$, it follows from Eq. (3) that under normal conditions ($P = 0$, $\rho = \rho_0$) we find that $\xi_z^\Gamma > 0$ for substances with $\gamma_0 > z$ (this is usually satisfied for metals when $\gamma_0 \approx 2$). Therefore, a connecting line passing through the point representing a normal state has a positive slope. It follows that representation of the connecting lines by Eq. (1) is valid if ξ_z^Γ is a nondecreasing function of the pressure on the reference curve, since otherwise the connecting lines intersect so that the procedure using two variables and the equation of state to find the third variable becomes ambiguous for straight connecting lines. The form of ξ_z^Γ is not known *a priori*. In some extreme cases such as an isentrope passing through the state $P = 0$ and $\rho = \rho_0$, which can be selected as the reference curve, the quantity ξ_z^Γ behaves as follows. Under normal conditions when $\gamma = \gamma_0(\rho_0/\rho)^n$ (n is a constant) the derivative obeys $d\xi_z^\Gamma/dP^\Gamma > 0$ provided n satisfies the following inequality:

$$n > \frac{(z-\gamma_0)[z(K'+1) - (z+1)\gamma_0]}{\gamma_0 z}, \tag{5}$$

where

$$K^\Gamma = \rho^\Gamma (c^\Gamma)^2, \quad K' = dK^\Gamma/dP^\Gamma.$$

Using the results of Ref. 18 and assuming that $z = 0.5$ we find that the inequality of Eq. (5) is satisfied for all the sample metals that do not undergo phase transitions. In the limit of high compressions the substitution of $\rho^\Gamma (c^\Gamma)^2/P^\Gamma \to 5/3$ and $\gamma^\Gamma \to 1/2$ (Ref. 19) into Eq. (3) yields a positive and increasing function if z lies within the interval $3/7 < z < 0.5$. Therefore, in two extreme cases if $z \approx 0.5$, the quantity ξ_z^Γ is positive and increases with the compression. Hence, we can assume that this behavior of ξ_z^Γ applies to the whole isentrope. However, isentropes are usually quite complex and are selected on the basis of the shock compression data. Therefore, it is preferable to select the reference curves to be the shock adiabats of continuous substances, since these have been investigated in greatest detail. Since the adiabats expressed in terms of the D–u coordinates are easiest to represent (usually linearly or quadratically), we shall select ξ_z^Γ (in terms of these coordinates) to be a function which increases with compression in accordance with the following equation:

$$\xi_z^\Gamma = \frac{z\rho_0^2 c_0}{\gamma_0 - z} D. \tag{6}$$

This agrees with the value of ξ_z^Γ under normal conditions. Equations (1), (2), and (6) subject to the laws of conservation under shock compression conditions give the simple form of the equation of state, which is used later in the calculations. Shock adiabats in the form $D = c_0 + a_1 u + a_2 u^2$ were used as the reference curves of iron, copper, and tungsten. The values of the relevant coefficients are given in Table III. In the case of iron and copper the shock adiabats for the lower range were taken from Ref. 15, whereas at higher velocities the D–u relationship was selected on the basis of agreement with the data of Refs. 5 and 10.

Since the experimental point obtained in Ref. 9 deviated slightly from the general tendency revealed in the laboratory measurements ($a_1 = 1.252$ according to Ref. 15), we used a linear D–u relationship for tungsten. We adopted the follow-

ing values of γ: $\gamma_{0\,\mathrm{Fe}} = 2.5$, $\gamma_{0\,\mathrm{Cu}} = 2.0$, $\gamma_{0\,\mathrm{W}} = 1.64$.

In constructing the equation of state of iron we ignored the phase transition occurring at low pressures (≈ 13 GPa). Consequently, the selection of $\gamma_{0\,\mathrm{Fe}}$ was based on the estimates given in Ref. 20 for the high-density phase. The selection of z was related to the condition that the shock adiabats of continuous and porous samples plotted in the form of the D–u dependences should be parallel and should be governed by the equation $z = 2(D'_\infty - 1)$. The following values of z were adopted: $z_{\mathrm{Fe}} = z_{\mathrm{Cu}} = 0.5$, $z_{\mathrm{W}} = 0.44$. Equations (6) and (3) can be used to calculate the velocity of sound, which was measured earlier[21] for copper and iron, since γ was determined (see Ref. 22) from the compression derivatives of the pressure on the shock adiabat and on the isentrope (S) using the following expression:

$$\gamma^{\Gamma} = \frac{2\sigma[(\partial P/\partial\sigma)_{\Gamma} - (\partial P/\partial\sigma)_{S}]}{\sigma(\sigma-1)(\partial P/\partial\sigma)_{\Gamma} - P}. \qquad (7)$$

Substituting Eq. (7) into Eq. (3) and using Eq. (6), the constants γ_0 and z, and the selected D–u dependences we found the velocities of sound, which were within $\approx 5\%$ to the experimental values. It should be pointed out that substitution of Eq. (7) into Eq. (3) and the use of the experimentally determined velocities of sound[21] enabled us to calculate directly ξ_z^{Γ}. However, this yielded nonmonotonic dependences of γ^{Γ} and ξ_z^{Γ}. Clearly, these nonmonotonic depen-

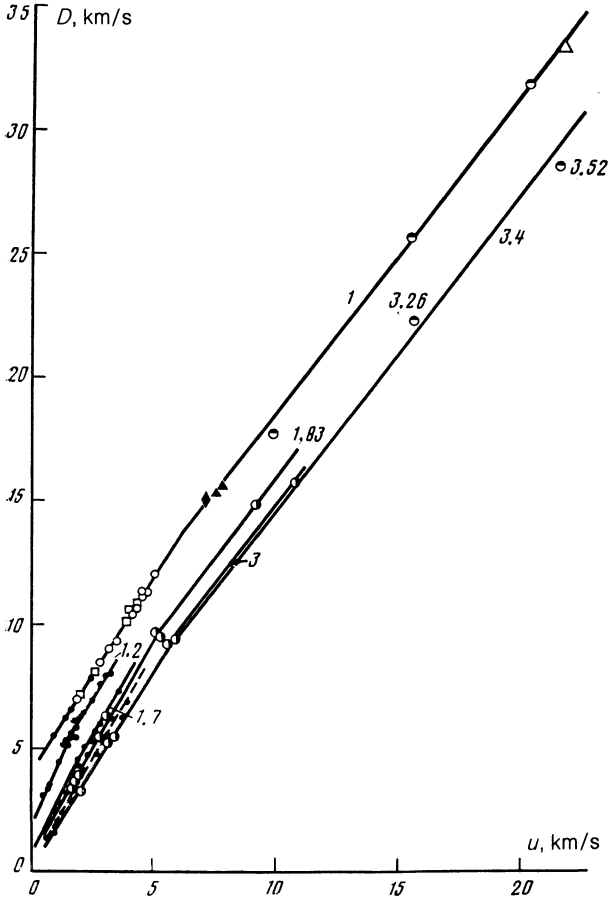

FIG. 1. D–u diagram of iron. Measurements made by absolute methods: ◑) Table I; ◆) Ref. 16; ▲) Ref. 23; ○) Refs. 15 and 24; □) Ref. 25; ●) Ref. 26. Comparative measurements: ◓) Refs. 3 and 5 and Table II; △) Ref. 10. The numbers alongside the curves in Figs. 1–6 give the porosity.

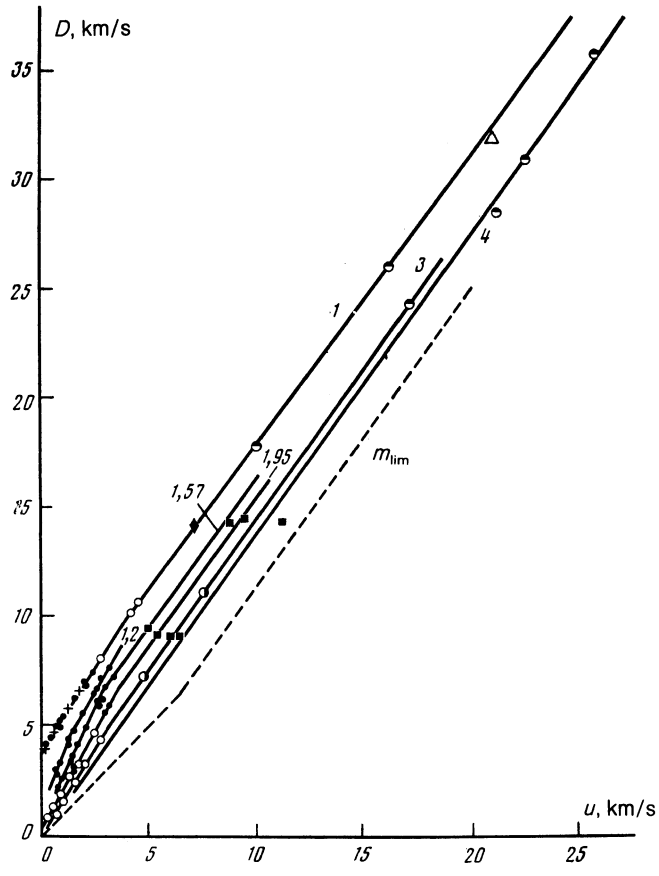

FIG. 2. D–u diagram of copper. Measurements by absolute methods: ◑) Table I; ■) Ref. 2; +) Ref. 15; ○) Refs. 24 and 27; ◆) Ref. 16; ●) Ref. 26. Comparative measurements: ◓) Refs. 4, 5, and 11; △) Ref. 10.

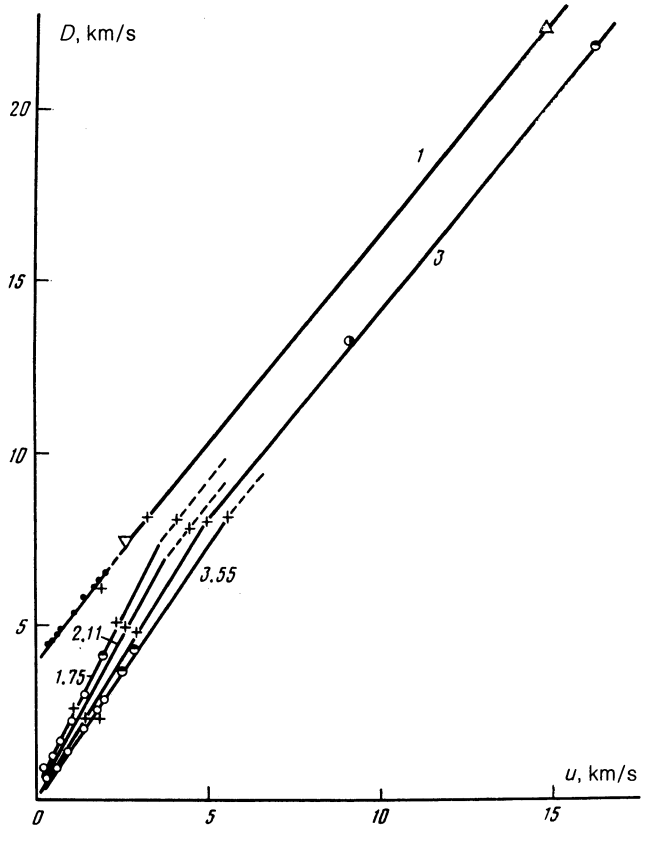

FIG. 3. D–u diagram of tungsten. Measurements by absolute methods: ◑) Table I; +) Ref. 1; ▽) Ref. 28; ●) Ref. 20; ○) Ref. 27. Comparative measurements; ◓) Table II; △) Ref. 9.

dences were obtained because Eq. (7) contains a difference between two quantities and this difference is comparable with the errors in each of the quantities. A description of our calculation of the shock adiabats of porous samples is given in the next section.

DISCUSSION OF RESULTS

The shock adiabats of normal- and low-density ($m \geqslant 1$) samples of iron, copper, and tungsten are plotted in Figs. 1–3, respectively (experimental data, D–u coordinates) and in Figs. 4–6 (comparison of the calculated and experimental values, P–σ coordinates, where $\sigma = \rho/\rho_0$ represents the relative compression). First of all, we note a satisfactory agreement between the results obtained by different authors both in the range of measurements carried out by the absolute methods and in the terapascal range of relative measurements. As pointed out already, such an agreement is a confirmation of the correctness of the results obtained throughout the investigated range of pressures.

It follows from the D–u dependences that, for any value of m used in the present study, the experimental data for metals can be represented by segments of straight lines with different slopes D'_u and parabolic connecting lines in a certain range of D_{cr} typical of each metal. The initial parts (in the range $D < D_{cr}$) obtained for all the adiabats corresponding to $1.7 < m < m_p <$ (m_p is a value of the porosity close to

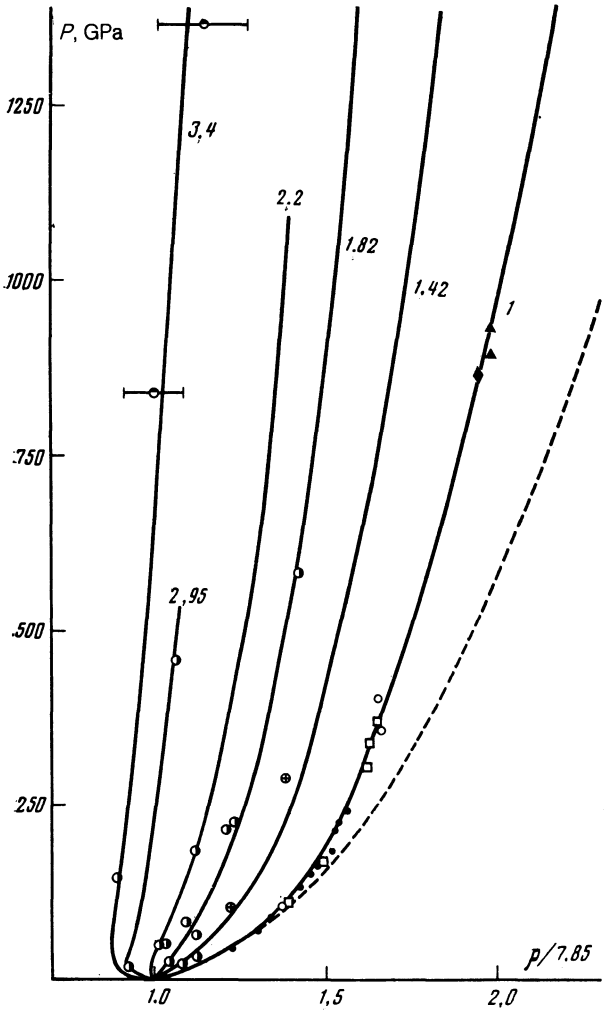

FIG. 4. P–σ diagram of iron. The continuous curves are the shock adiabats (calculated from the equation of state) and the dashed curve is the isentrope beginning from the initial state P_{S_0}. The experimental points are labeled in the same way as in Fig. 1; the data represented by ⊕ were taken from Ref. 29.

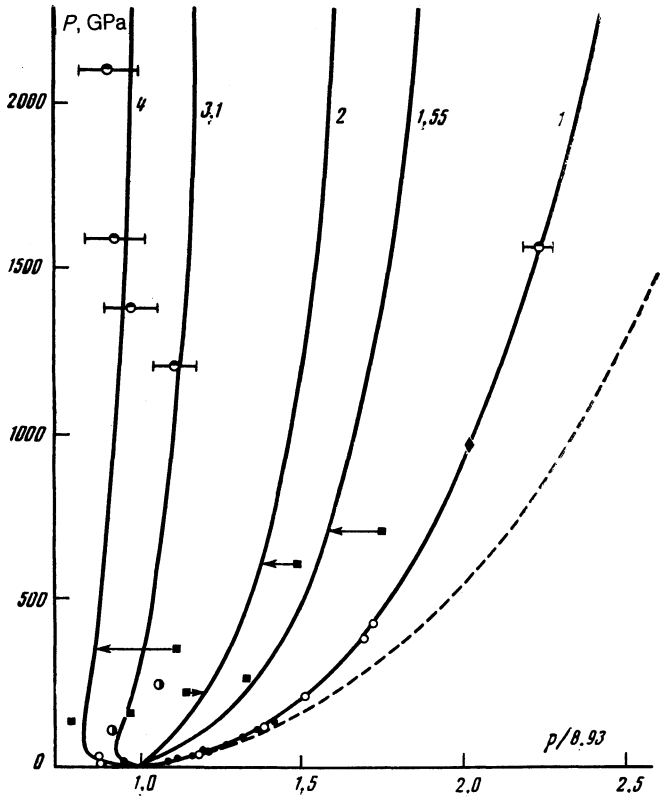

FIG. 5. P–σ diagram of copper. The notation is the same as in Figs. 2 and 4.

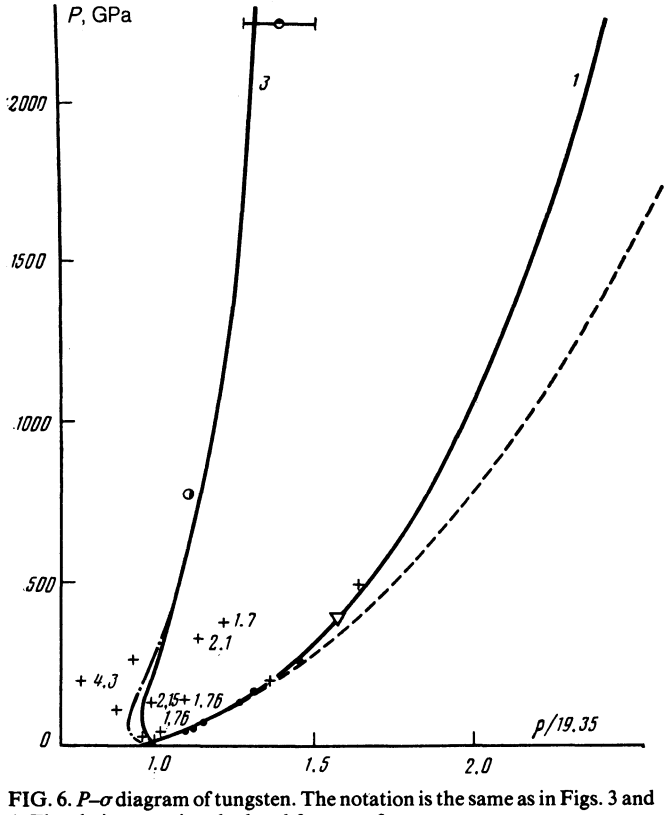

FIG. 6. P–σ diagram of tungsten. The notation is the same as in Figs. 3 and 4. The chain curve is calculated for $\gamma_0 = 3$.

that of a powder), represent a fan of straight lines intersecting near a point with the coordinates $u = 0$ and $D \geqslant 0$. These lines have quite different derivatives D'_u. For example, in the case of copper, which was investigated most thoroughly, a reduction in m from values corresponding to the powder density ($m = 4$) to $m \approx 1.2$ increases the slope of the adiabats from $D'_u \approx 1.3$ (powder density) to $D'_u \approx 2.3$ ($m \approx 1.2$).

The second portions of all the adiabats were approximately parallel lines with slopes close to the limit. It should be pointed out that in the range $m > 5$ the copper adiabat has clearly an opposite kink, i.e., in the first region where $D < D_{cr}$ holds the slope is less, whereas in the range $D > D_{cr}$ it is equal to its limiting value. The minimum slope in the first region (where $D \approx u$) tends to unity. Figure 2 shows the position of the postulated shock adiabat of ultraporous copper. A similar situation occurs (but for different values of m) in the case of the other metals. Estimates of the effective Grüneisen coefficient from the shock adiabats of porous samples at the maximum pressures reached in our experiments (in this range the electronic components predominate so that $\gamma_{eff} \approx \gamma_{el}$) demonstrate that in the case of the investigated metals the values of γ_{eff} amount to 0.5 for tungsten, but are 0.7–0.9 for copper and iron. This conclusion is in conflict with the reports[2,16] of different values of γ_{el} for simple ($\gamma_{el} \sim 0.5$ for copper) and transition ($\gamma_{el} \sim 1.0$ for both nickel and iron) metals.

A comparison of the calculated shock adiabats of iron, copper, and tungsten with the experimental results is made in Figs. 4–6 using the P–σ coordinates. In the case of iron and copper the agreement between the calculations and experiments is good throughout the investigated range. There is no such agreement in the case of tungsten. Although the highest points ($m = 3$) are described satisfactorily, at low pressures the difference between the calculations and experiments is in some cases 10% with respect of the compression. The difference in this range can be reduced if we assume that $z_W > 0.44$, but this results in deterioration of the description of the experimental results obtained at higher pressures. Bearing in mind the simple form of our equation and the wide range of the P–V–E states which are being described by this equation, we can regard the calculated results as on the whole being in full agreement with the experiments.

[1)] In investigations carried out in the range $P > 10$ TPa the adiabat of a standard substance must also be continuous, i.e., it should not exhibit any significant oscillations.[6]

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