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Evaporation of shock-compressed lead in release waves

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We determined experimentally the rates of expansion of lead samples in air following compression by strong shock waves of amplitude 0.4–3.4 Mbar. The strong increase of the expansion rate at pressures exceeding 1.32 Mbar attest to evaporate of the lead in the release wave. The results are used to determine the shock-compression entropy and to refine the equation of state of the liquid phase of lead at high pressures and temperatures.

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1. INTRODUCTION

The solution of many problems of applied physics calls for the study of high-temperature evaporation of metals under the action of pulses, and an important question is that of the thermodynamic equilibrium of the evaporation process.[1] Near the liquid–vapor equilibrium line and in the transcritical region, a rigorous theoretical calculation of the characteristics of metals is impossible because of the presence of a strong interparticle interaction of complex structure, while static experiments are limited by the simultaneous action of the high temperatures and pressures that are typical of metals. It is therefore of great importance to investigate the properties of metals near the evaporation line by dynamic methods.

Dynamic investigation methods with the aid of strong shock waves make it possible to obtain, in a wide range of parameters, the caloric characteristics of metals—the pressure P, the specific volume V, and the internal energy E.[2,3] This information was obtained for many metals in experiments on shock compression of solid and porous samples[3–6] and by recording the release isentropes.[7–9] By using these data we can, without resorting to models, construct a phenomenological equation of state in the form of the function $E(P, V)$. In this case the equation of state, however, does not contain such important material characteristics as the temperature and the entropy. Zel'dovich[10] was the first to call attention to the possibility of determining, in view of the adiabaticity of the release process, the entropy of shock compression from the final parameters of the expanding substance. If a metal expands in the state of an ideal gas or plasma, its entropy can be calculated from the pressure and density (or temperature) measured in the course of the dilatation.

The program proposed in[10] was not realized because of the great difficulty of obtaining, in shock compression, the high-energy states needed to reach the ideality region. Another difficulty lies in the exact registration of high expansion velocities on the order of dozens of km/sec. In the case of lower-intensity shock waves, the entropy of copper was determined[11,12] from the residual temperature of the solid and liquid metal. In[12], the entropy of shock compression of sodium, strontium, barium, and uranium was determined from spectroscopic measurements of the fraction of the evaporated metal after expansion in vacuum, and the object of the investigation was the states within the two-phase region.

We have developed a new variant of obtaining entropy information, based on an investigation of the evaporation process when metal samples, previously compressed by shock waves of various intensities, expand in air. The release pressure at which the evaporation started, yielded the point of intersection of one of the isentropes with the phase boundary, whose parameters, including the entropy, were assumed known. The start of the evaporation upon release can be observed if the relaxation time of the metastable states that are produced below the saturation curve is much shorter than the characteristic time of the evaporation process. Otherwise the metal will expand along the adiabats of the heated liquid or supercooled vapor. It is possible, however (see[14]) that owing to the presence of a large number of charged particles in the metal on the saturation line, the relaxation is of the order of 10^{-9} sec. At such times, the difference between the temperatures of the metastable and equilibrium states, i.e., the superheat of the liquid metal, does not exceed several degrees.

The question of evaporation of shock-compressed metals in release waves was investigated in experiment us-

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ing lead as an example. The investigation yielded, for a wide range of shock pressures, the rate of isentropic expansion and displayed the effects of metal evaporation in the course of the dilatation. The experimental data yielded new thermodynamic information on the properties of the liquid phase of lead at high pressures and temperatures.

2. PROCEDURE

The evaporation effects were ascertained by measuring the rate of isentropic expansion, in air with initial pressure $P_{in}=1$ bar, of lead samples compressed by shock waves of various intensities. The experiment yielded the dependence of the rate of expansion W of the lead into the air on the mass velocity U in the shock wave.

A schematic $P-V$ diagram of the experiment (Fig. 1) shows that the final states of some of the release isentropes lie inside the two-phase region. When they cross the phase boundary, the isentropes experience a kink[15]:

$$\left(\frac{\partial V}{\partial P}\right)_s^{II}-\left(\frac{\partial V}{\partial P}\right)_s^{I}=-\frac{T}{C_p}\left(\frac{dS}{dP}\right)^2. \quad (1)$$

The superscripts I and II pertain here respectively to the pure phases and to their mixture, while the derivative in the right-hand side is taken along the equilibrium line. The change of the rate of release is connected with the isentrope slope $dW/dP=(-(\partial V/\partial P)_S)^{1/2}$; it follows from (1) that the entry of the isentrope into the two-phase region from the liquid phase (evaporation) or from the gas phase (condensation) causes an additional increase of the rate W. The dependence of the expansion rate on the parameters of the initial state in the compression wave is monotonic if the metal expands along the metastable adiabats (dashed in Fig. 1) and has two kinks in the case of equilibrium expansion. The more noticeable of the two kinks corresponds to the start of the evaporation, because of the substantial difference between the compressibilities in the liquid and the paraphase states. The presence or absence of a kink on the experimental $W(U)$ curve makes it thus possible to confirm that the metal is evaporating.

Let us estimate the magnitude of the expected change in the slope of the experimental curve at the start of the evaporation (isentrope ab in Fig. 1). Using the inequalities $(\partial V/\partial P)_S^I \ll (\partial V/\partial P)_S^{II}$ and $\delta P \ll dP$ (the latter follows from $\rho_{air} \ll \rho_{Pb}$) we can determine (see Fig. 1a) the slope discontinuity

$$\Delta\left(\frac{dW}{dU}\right)_b \approx \left(\frac{dW}{dP}\right)_{II}\left(\frac{dP}{dU}\right)_b=\left(\frac{dW}{dP}\right)_{II}\left(\frac{dP}{dS}\right)_b\left(\frac{dS}{dU}\right)_a. \quad (2)$$

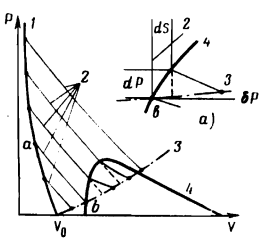

FIG. 1. $P-V$ diagram of the experiment: 1—shock adiabat, 2—release isentropes (the metastable isentropes are shown dashed), 3—final-state curve in the case of release in air, 4—boundary of liquid–vapor two-phase region.

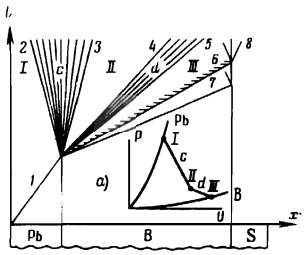

FIG. 2. $x-t$ and $P-U$ diagrams (a) of release wave with evaporation taken into account. Constant flow regions: I—initial state, II—phase boundary, III—final state; c, d—centered release waves. 1—D_{Pb}, 2—$U_I-C_I^L$, 3—$U_{II}-C_{II}^L$. 4—$U_{II}-C_{II}^{L+G}$, 5—$U_{III}-C_{III}^{L+G}$, 6—U_{III}, 7—D_A, 8—D_S; D and U are the wave and mass velocities, C is the speed of sound, A—air, S—screen.

Substituting in (2) the condition that the entropy increase on the shock adiabat

$$dS/dU=U^2D_u'/TD$$

as a function of its kinematic parameters (the mass and wave velocities U and D) we obtain ultimately, taking (1) into account,

$$\Delta\left(\frac{dW}{dU}\right)_b \approx \left(\frac{T}{C_p}\right)_b^{1/2}\left(\frac{U^2D_u'}{TD}\right)_a. \quad (3)$$

The increase of the slope dW/dU when the isentrope enters the two-phase region (not too close to the critical point; $T \leq 0.7\,T_c$), turns out according to the estimate (3) to be close to unity for various metals. This means that the kink can be registered experimentally.

Evaporation of the expanding metal at sufficiently large shock-compression entropies leads to a release wave of unique structure, having three regions of constant flow (Fig. 2). The intermediate region is due to the presence of two sound velocities on the phase boundary in state II (kink on the release curve in the coordinates P and U, Fig. 2a), and corresponding two perturbation-propagation velocities—in the liquid $U_{II}-C_{II}^L$ and in the vapor-liquid mixture $U_{II}-C_{II}^{L+G}$. The vapor content increases from zero to the maximum value on going from state II to state III. According to calculations, the thickness of the state-III layer with partially evaporated metals is several tenths of a millimeter under the experimental conditions, and is sufficient for reliable registration of two-phase states.

The expansion rate was determined from the time of passage of the free lead surface through the air gap (Fig. 2) between the sample and the transparent organic-glass screen. The glow time in the gap was registered with an SFR-3M image-converter streak camera. The glow started when the shock wave emerged to the surface of the sample and was "cut off" when the metal struck the screen. In a number of experiments the organic glass was replaced by a copper plate in which were inserted electric contacts that applied a voltage on the oscilloscope plates when they were closed. This determined not only the time of passage of the lead through the gap but also the parameters of the shock wave pro-

TABLE I. Experimental results on isentropic expansion of shock-compressed lead in air ($P_{in.air}$ = 1 bar).

Run No.	Initial state on the shock adiabat		Final state in release wave			
	U, km/sec	P, Mbar	W, km/sec	P, bar	Δ, km/sec	$\delta \cdot 10^2$
1	1.03	0.42	2.22	75	0.16±0.03	8.0
2	1.33	0.60	2.86	118	0.20±0.04	7.5
3	1.81	0.99	3.84	212	0.22±0.05	6.1
4	2.19	1.31	4.78	325	0.40±0.06	9.1
5	2.45	1.56	5.56	480	0.66±0.07	13.4
6	2.76	1.90	6.39	570	0.87±0.08	15.8
7	3.00	2.16	6.97	680	0.97±0.08	16.1
8	3.94	3.40	9.60	1280	1.72±0.11	21.8

duced in the copper screen when the expanding lead was stopped.

The experiments were performed with explosive propellant systems with known mechanical characteristics, which determined the parameters of the initial states of the lead (prior to the expansion) accurate to 1% (relative to U). The velocity W of the expansion front was recorded with the same accuracy.

3. RESULTS OF EXPERIMENT. EVAPORATION EFFECTS

The experiments on the expansion of the lead were performed in a wide range of shock compression, from 0.4 to 3.4 Mbar. The results are shown in Table I, where each experimental figure represents an average of 3–5 experiments. The table lists also the initial-state pressure P_1 on the Hugoniot adiabat (corresponding to[16]) and the release pressures P_3 calculated from the shock adiabat of air.[17] It is known that at low initial pressures the expansion velocity is approximately double the mass velocity in the compression wave. The last column of the table gives therefore the absolute and relative differences, $\Delta = W - 2U$ and $\delta = \Delta/2U$, respectively, which reveal most clearly the influence of the irreversible heating in the shock-wave front on the release process. The errors listed in the table correspond to the aforementioned accuracy of U and W. The possible error of δ is the same for all the runs and is equal to 1.4×10^{-2}.

The results are illustrated in Fig. 3, which shows also data obtained by others.[18,19] The experimental expansion curves $\Delta(U)$ and $\delta(U)$ have clearly noticeable kinks. They correspond on the Hugoniot adiabat of lead to the state $U_a = 2.20 \pm 0.05$ km/sec and $P_a = 1.32 \pm 0.04$ Mbar. At higher shock-compression pressures the lead is partially or fully evaporated as it expands in the air, and at lower pressures it remains in the condensed state. The evaporation isentrope crosses the equilibrium line at $W_b = 4.80 \pm 0.10$ km/sec and $P_b = 330 \pm 10$ bar. The inflection of the $\delta(U)$ curve at 1 km/sec, as shown by calculations with a model equation of state, is due to the melting of the lead.

Two additional experimental runs were performed shock-compressed at 1.90 Mbar. In the first we measure the rate of expansion at an initial air pressure $P_{in} = 0.1$ bar. Under these conditions we registered an expansion velocity $W = 6.74$ km/sec and a pressure $P_3 = 65$ bar. Comparison with the data of Table I shows that lowering the final pressure from 600 to 65 bar has increased W by 350 m/sec. If the expansion were to follow the adiabat of a superheated liquid, the increase of the velocity, at the same pressure drop, would not exceed several dozen meters.

In the second run, at the same initial-state parameters but with the expansion in air at atmospheric pressure, we measured the shock-wave velocity in the copper screen after it was struck by the expanding lead. The lead velocity (see Table I) was 6.39 km/sec. The parameters of the shock wave produced in the copper were $D = 5.94$ km/sec, $U = 1.37$ km/sec, and $P = 0.73$ Mbar. According to estimates, the impact of the superheated liquid lead should have produced in the copper screen a much higher pressure, ~1.5 Mbar.

4. DISCUSSION OF RESULTS

Our experiments show that the decay of the shock on the metal–air interface, at a definite shock-wave amplitude, is accompanied by evaporation with a relaxation time $\tau \ll 10^{-6}$ sec, which the characteristic time of motion of the lead through the gap. This circumstance supports the conclusions of[14] that the evaporation of a metal in the front of a release wave at a temperature of several thousand degrees is in thermodynamic equilibrium.

Measurements of the rate of expansion of lead have revealed on one of the isentropes states that belong to the equilibrium line ($P_b = 330$ bar) and to the Hugoniot adiabat ($P_a = 1.32$ Mbar). The thermodynamic interpretation of this result presupposes that the parameters of the saturation line are known. For lead and a number of other metals these parameters were calculated in[20], according to which the experimentally singled out isentrope of the start of the evaporation has at $P_b = 330$ bar the values $T_b = 4000 \pm 100$ K, $S_b - S_0 = 0.44 \pm 0.01$ J/g-deg (S_0 is the entropy of lead under normal conditions).

An entropy of 0.44 J/g-deg is produced on account of the irreversible heating of the lead in the front of a shock wave of amplitude 1.32 Mbar.

The thermal characteristics of shock-compressed

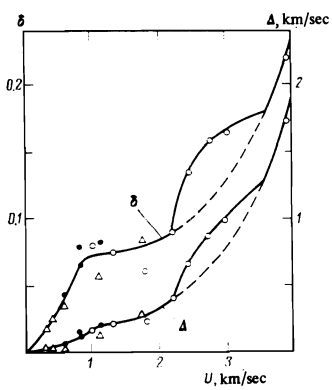

FIG. 3. Deviation from the doubling law in isentropic expansion of lead in air. Equilibrium and metastable (dashed) expansion at $P_{in.air} = 1$ bar ($\Delta = W - 2U$, $\delta = \Delta/2U$); ○—our results, ●—[18], △—[19].

TABLE II. Thermodynamic parameters on the shock adiabat of lead.

P, Mbar	V, cm³/g	Calc. according to [21]				Our calculations			
		T, K	$S-S_0$, J/g-deg	C_V, J/g-deg	γ	T, K	$S-S_0$, J/g-deg	C_V, J/g-deg	γ
0.6	0.0595	3060	0.226	0.133	1.51	2300	0.249	0.147	1.02
0.8	0.0566	4770	0.278	0.144	1.36	3670	0.312	0.160	0.92
1.0	0.0543	6550	0.317	0.155	1.24	5120	0.361	0.172	0.85
1.5	0.0502	11 000	0.389	0.183	1.06	8940	0.453	0.201	0.75
2.0	0.0474	15 400	0.443	0.211	0.94	12 800	0.520	0.226	0.71
2.5	0.0452	19 600	0.487	0.237	0.87	16 700	0.574	0.250	0.68
3.0	0.0434	23 600	0.525	0.261	0.81	20 500	0.621	0.270	0.66
4.0	0.0406	31 100	0.589	0.306	0.73	27 900	0.698	0.307	0.65
5.0	0.0387	38 100	0.643	0.347	0.67	35 000	0.762	0.338	0.64

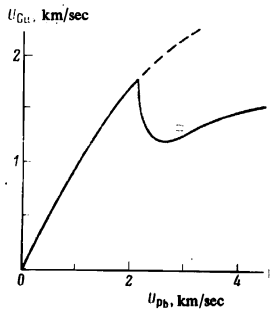

FIG. 4. Mass velocity in copper screen when lead is stopped. Solid line—calculation with allowance for evaporation, dashed—stopping of metastable liquid, □—our experiment.

metals were calculated earlier[3,6] from semi-empirical equations of state containing many free parameters. The most consistent of the proposed ones is the equation obtained in[21] and takes into account the contribution of the electrons, the melting, and the anharmonicity of the oscillations of the liquid-metal atoms. The parameters of the shock adiabat of lead as calculated in[21] are given in the left-hand side of Table II. A pressure of 1.32 Mbar corresponds in this case to an entropy $S_b - S_0 = 0.37$ J/g-deg, which is much less than the entropy obtained in the interpretation of the experiments in accordance with[20].

Using the new thermal information, we have obtained for the equation of state of compressed lead a variant in which the effect of the temperature on the specific heat of the liquid metal is much less than in[21]. The Grüneisen coefficient $\gamma = V(\partial P/\partial E)_V$ of the liquid state has likewise a different temperature dependence. The shock-adiabat parameters calculated from the new equation of state are given in the right-hand side of Table II. They differ from those of[21] in the larger values of the entropy and specific heat, and in the smaller values of the temperature and the Grüneisen coefficient on the shock adiabat in the band $P = 0.5$–5.0 Mbar. With increasing pressure, owing to the increasing contribution of the electrons to the equation of state, the temperature and specific-heat differences decrease. The distribution of the entropy on the shock adiabat shows that the critical entropy ($S_b - S_0 = 0.58$ J/g-deg[20]) is reached at $P_I = 2.6$ Mbar. The expansion after a shock compression of 3.4 Mbar reduces the lead to the state of weakly-ionized nonideal gas.

The derivation of a thermodynamically complete equation of state of lead in a wide region of the phase diagram, including liquid, plasma, and other states, is the subject of a separate investigation. We have presented here preliminary results of a calculation of the rate of expansion along metastable release isentropes (dashed lines in Fig. 3) and the parameters of the shock waves in a copper screen that stopped the expanding lead (Fig. 4). The abrupt decrease of the mass velocity is due to formation, at 2.2 km/sec, of a low-density layer of partially evaporated lead.

The first results of this investigation were obtained by the untimely deceased M. I. Brazhnik, who did much to organize and develop dynamic methods in the Soviet Union. The authors express deep gratitude to V. E. Fortov and I. I. Sharipdzhanov for useful discussions throughout the course of the work.

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