OSCILLOGRAPHIC STUDY OF ELECTRICAL EXPLOSION IN COPPER AND GOLD

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An oscilloscope is used to measure electrical resistance as a function of enthalpy in copper and gold during the process of electrical explosion of wire samples. The enthalpy at the initial point of the explosion process was greater in value than at the melting point of the metal. The electrical explosion phenomenon is considered from the viewpoint of the kinetics of the liquid-metal–vapor transition under impulse-heating conditions. The "degeneration" of the initial point of the electrical explosion for liquid-phase heating over periods less than 10^{-7} sec is explained by disruption of the stability of homogeneous-vapor-nucleus formation.

Electrical explosion of a conductor occurs upon heating by a powerful current pulse. There is no unified opinion as to the nature of this phenomenon. It was proposed in [1] that the process might be explained by peculiarities of the kinetics of the liquid-metal–vapor transition. It is thus of interest to measure the thermophysical properties of the liquid metal in the state preceding the electrical explosion. Use of a pulse-recording oscilloscope makes possible such measurements of electrical resistance and enthalpy.

This present study investigated electrical explosion of Cu (99.99%) and Au (99.99%) wires, using the technique described in [2]. A capacitor of 100 μf charged to 3 kV was discharged through a wire specimen 0.3 mm in diameter and 50 mm long, located in air. The discharge process was recorded by a two-beam oscilloscope, producing two synchronous oscillograms of current I through and voltage V across the specimen (Fig. 1). Using a Minsk-22 electronic computer, the oscillograms were processed to determine the dependence of relative metal resistance R/R_0 on molar enthalpy $H = H_T - H_0$ (Fig. 2 for Cu, Fig. 3 for Au) (R_0 and H_0 referred to a temperature $T = 298°K$). The calculations were performed with the formulas

$$R / R_0 = (U - LdI / dt) / R_0 I \tag{1}$$

$$H = \int_0^t IUdt - {}^1/_2 LI^2 \tag{2}$$

where L is the inductance of the specimen ($L < 2.5 \cdot 10^{-7}$ H), t is time. On the voltage oscillograms and R/R_0 versus H curves points corresponding to the commencement (index 1) and completion (index 2) of the metal process were determined. The commencement of the electrical explosion (index 3) was marked by a sharp rise in voltage across the specimen; after this point the oscilloscope beam recording the voltage moved so rapidly that it left practically no trace upon the film. Mathematical processing of the oscillograms was performed only up to this point. The heating time of the liquid phase τ (from point 2 to point 3) for Cu was 1.6 μsec, for Au, 7.7 μsec, which corresponds to heating rates of $25 \cdot 10^8$ deg/sec for Cu and $4.4 \cdot 10^8$ deg/sec for Au. Mean-square random error in determination of R_2/R_0 was 2.4%; for H_2, 2.5%; for R_3/R_0, 2.8%, for H_3, 3%. The maximum possible systematic error did not exceed 10% for R/R_0 and 15% for H; these errors were generated basically by nonlinearity in oscilloscope deviation and distortions introduced by the voltage divider.

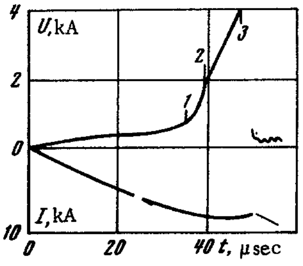

Fig. 2

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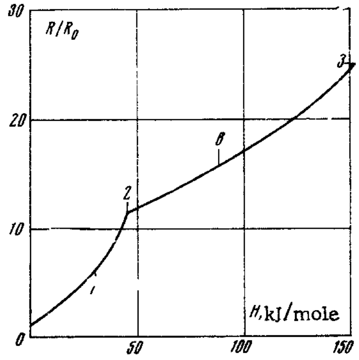

Fig. 2

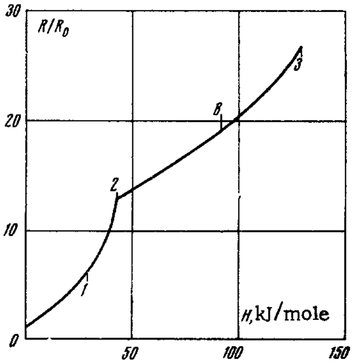

Fig. 3

TABLE 1

	R_1/R_0	R_2/R_0	R_3/R_0	H, kJ/mole		
				H_1	H_2	H_3
Cu	5.70	11.3	25	30	45	152
Au	6.00	13.1	27	29.6	43.3	129

Values of R/R_0 and H for the characteristic points, taken from the data of four experiments for each metal, are presented in Table 1. (Initial resistivity at 298°K for Cu was 1.77 $\mu\Omega \cdot$cm, for Au, 2.39 $\mu\Omega \cdot$cm.) The values of R_1/R_0, H_1; R_2/R_0, H_2 agree satisfactorily with corresponding data for resistance [3] and enthalpy [4] of the solid and liquid phases at the melting point. Comparison of the enthalpy at the initial point of the electrical explosion H_3 with its estimated value H_b at the boiling point T_b under 1 atm pressure [4] shows that for both metals H_3 greatly exceeds H_b. In Figs. 2 and 3 the index b denotes the normal metal boiling point.

In analyzing this experimental fact it must be kept in view that in heating a wire with a powerful current pulse there arises an internal pressure produced by magnetic compression of parallel currents, so that pressure tends to zero at the wire surface. The pressure produced by inertia of the wire material during rapid thermal expansion in the radial direction behaves analogously. Based on these facts, it can be assumed that during the process of pulse heating in open air the surface layer of the metal is under a pressure close to atmospheric, and so the increase in H_3 as compared to H_b observed in the experiments cannot be explained by an increase in liquid-metal boiling temperature under the action of pressure. The difference can be explained by peculiarities of the kinetics of the liquid–vapor phase transition under impulse-heating conditions. We will consider the peculiarities encountered under the conditions of the present experiment.

The smooth course of the oscillogram up to the initial point of electrical explosion (Fig. 1) indicates that after melting the liquid specimen preserved its form, not disintegrating under the action of gravity and magnetic forces. Upon reaching a temperature T_b in the surface layer of the liquid metal there begins heterogeneous formation and growth of vapor nuclei, which develop on existing centers (for example, gas inclusions). However, at a high heating rate, where $\tau = \sim 1$ μsec, the effect of these nuclei on liquid–vapor transition will be insignificant, since they are few in number because of the limited number of existing centers, and the mean velocity of interphase boundary displacement for Cu and Au according to the Hertz–Knudsen equation [5] is ~1 m/sec. For the same reason evaporation through the specimen surface may be neglected. Under conditions of impulse heating at rates of $\sim 10^9$–10^{10} deg/sec superheating of the liquid metal above T_b must occur.

In the superheated liquid homogeneous formation of vapor nuclei is possible, due to fluctuations. The formation rate of such nuclei J (number of critical nuclei arising in unit volume per unit time) under stable conditions at a temperature T is determined by the expression [5]

$$J = B \exp\,[-W_c / kT] \tag{3}$$

where B is a function which in comparison to the exponential term is weakly dependent on T; k is Boltzmann's constant, and W_c is the change in free energy upon formation of a critical nucleus. Upon increase in the degree of superheating $\gamma = (T - T_b)/T_b$, W_c decreases. In the first approximation for a spherical nucleus W_c may be connected with γ by an expression

$$W_c = 16\,\pi\sigma^3 / 3\rho^2\lambda^2\gamma^2 \tag{4}$$

where σ is the surface tension coefficient, ρ is vapor density, and λ is the specific heat of vapor formation.

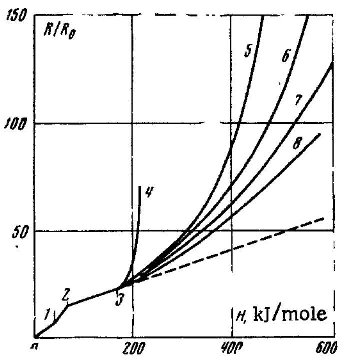

Fig. 4

For an unstable process, after momentary attainment of a temperature T the homogeneous nucleus formation rate J(t) is related to J [see Eq. (3)] as

$$J(t) = J \exp[-\theta / t] \quad (5)$$

where θ is the "relaxation" time for establishment of the stable regime. According to the theoretical evaluations of [5], $\theta = 10^{-7}$-10^{-8} sec.

From Eqs. (3) and (4) it follows that J is a rapidly increasing function of γ, and to attain values $J = 1\ \mathrm{cm}^{-3} \cdot \mathrm{sec}^{-1}$ significant superheating of the metal is necessary. Thus, for example, calculations show that for liquid cesium at p = 1 atm in 1 cm^3 during 1 sec there develops one critical nucleus for a superheating $\gamma = 0.80$; with further increase in superheating to $\gamma = 0.89$, J increases to a value $J = 4 \cdot 10^{23}\ \mathrm{cm}^{-3} \cdot \mathrm{sec}^{-1}$. The latter γ value corresponds to the limiting case of superheating of liquid cesium γ_S, determined by the spinode, calculated with a formula from [6]. This curve is the limit of thermodynamic stability of the phase and satisfies the condition $(\partial p / \partial V)_T = 0$. Experiments [7] have shown that explosive boiling of a number of organic substances upon heating by a current pulse 10^{-5}-10^{-6} sec in duration occurs in the vicinity of spinode. There is thus basis to assume that the initial point of electrical explosion at which the wire resistance increases sharply corresponds to explosive boiling of the superheated metal, which is in a state close to the limit of stability determined by the spinode equation. This is a valid assumption if the duration of liquid-phase heating τ is so short that the effect of heterogeneous nuclei on the metal evaporation process is insignificant, while at the same time τ must be much smaller than θ [see Eq. (5)], since in the opposite case the stability of the homogeneous-nucleus-formation process will be disrupted, and explosive boiling of the liquid metal will occur at higher temperatures $T > T_S$ (T_S is the spinode temperature) in the region of unstable liquid-phase states. The effect of magnetohydrodynamic instability of the liquid cylinder on the heating process must be excluded. In our experiments on copper, deviations from these conditions were evidently insignificant. Comparison of the present data with that of [8] will support such a position.

Figure 4 shows R/R_0 as a function of H, the curves being constructed from the data of [8]. For these curves the points 1, 2, and 3, have the same meaning as in Figs. 2 and 3, while τ values are: 4) 0.10; 5) 0.07; 6) 0.05; 7) 0.02; 8) 0.01 μsec; the dashed line is a continuation of the initial linear portion of the curve R/R_0 versus H for the liquid state, where curves 4-8 coincide. At $\tau = 0.1$ μsec the commencement of electrical explosion is denoted by the sharp inflection in curve 4. For this point [8] determined the values $R_3/R_0 = 23$, $H_3 = 2.5$ kJ/g = 159 kJ/mole, which agree well with the data obtained herein. Thus, for change in τ from 0.1 to 1.6 μsec, H_3 for Cu undergoes practically no change, which is explained by the strong dependence of J on γ [Eqs. (3), (4)] With increase in the heating rate (decrease in τ) in [8] a gradual degeneration of the electrical explosion commencement point (Fig. 4) was noted, reflected by the fact that after the point $R_3/R_0 = 23$, $H_3 = 159$ kJ/mole the curve of R/R_0 versus H inflects upward less sharply, with the curvature decreasing with increase in heating rate. Thus, at heating rates greater than 10^{11} deg/sec the energy introduced into the copper exceeds the enthalpy value at the initial explosion point and does not uniquely define the electrical resistance. This anomalous phenomenon was noted in [9], but no detailed explanation was offered. In light of Eq. (5), it could be proposed that in this case there occurs a transition from stable to unstable homogeneous-nucleus generation, which leads to reduction in J at a given temperature. For $\tau < \theta$ the liquid phase under pulse-heating conditions evidently may be preserved for $T > T_S$, but these states (the branches of curves 5-8 to the right of point 3 in Fig. 4) should be considered as absolutely unstable [the condition $(\partial p / \partial V)_T > 0$ must be fulfilled].

High-speed x-ray techniques indicated the presence of a "layered structure" in the liquid cylinder in this region [10], and so it can be assumed that the unstable states can be achieved only in individual zones, not over the entire specimen volume. The agreement of R_3/R_0 and H_3 values of the present study and [8] indicates good reproducibility of the initial point of the electrical explosion, and also indicates that at sufficiently high heating rates a "layered structure" does not develop to the left of this point [10]. This, in turn, demonstrates the feasility of employing electrical explosion of wires in studies of metal properties near the limits of thermodynamic stability of the liquid phase.

If it is assumed that the values obtained for H_3 for Cu and Au are close to the value H_S for the limit of a stable state at p = 1 atm, they may be used to evaluate the critical temperature of these metals. As calculation by a formula from [7] indicates, the spinode equation for cesium in coordinates p−T, to an accuracy sufficient for interpretation of the results obtained, may be represented in the form

$$p_s / p_c = 10T_s / T_c - 9 \qquad (6)$$

where the index s refers to the spinode and c to critical parameters. This equation is satisfied by experimental data on the spinode for a number of organic liquids [6]; therefore, it may be expected that spinodes in other liquids will satisfy the law for corresponding states. Since under our experimental conditions $p_s = 1$ atm$\ll p_c$, from Eq. (6) we obtain

$$T_c = {}^{10}/_9 T_c = {}^{10}/_9 T_3 = {}^{10}/_9 [T_2 + (H_3 - H_2) / c_2] \qquad (7)$$

where T_2 is the melting temperature of the metal, T_3 is the temperature at the commencement of electrical explosion, c_2 is the mean value of liquid-metal heat capacity over the range T_2 to T_3, which, considering the data of [4], may be equated to the heat capacity at T_2 (for Cu, 31.4 J/mole · deg; for Au, 29.3 J/mole · deg). According to the data obtained for H_2 and H_3, using Eq. (7) for Cu and Au, we have T_c = 5390 and 4820°K.

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