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ELECTRICAL BREAKDOWN AND EXCITATION OF DETONATION IN THALLIUM AND COPPER AZIDES

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ABSTRACT:

The mechanism of the electrical breakdown of thallium and copper azides and its connection with the mechanism of exaltation of detonation in solid objects are investigated. The breakdown in TlN_3 is caused by electrons which are injected from cathodes, and in $Cu(N_3)_2$- by holes injected from anodes; both the processes are controlled by nonuniform field distribution in the near contact zones and develop into impact ionization.

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INVESTIGATION OF ELECTRICAL BREAKDOWN AND EXCITATION OF DENOTATION IN THALLIUM AND COPPER AZIDES

Yu. A. Zakharov, Yu. N. Sukhushin

(Presented by the Scientific Seminar of the Department of Radiation Chemistry)

Comparatively, few papers [I-3] , have been devoted to the study of excitation of detonation during the breakdown of explosives although this process is of definite scientific interest [I,4] .

Certain results of the study of electric resistance of the azides of thallium and bivalent copper to voltage impulses with duration of action of nearly 10^{-7} sec are described in this paper for explaining the mechanism of the breakdown and its connection with the mechanism of the excitation of detonation in the said objects. Samples consisted of tablets of 0,2 mm thickness and density similar to a single-crystal. The tablets were fixed on a metallic electrode. The geometry of electrodes "sphere against a plane" - provided a uniform field. Any possible ignition of explosives by corona electrodes was eliminated by placing samples in a protective medium (silicone oil). Voltage and current passing through a sample at the time of breakdown were recorded on an oscillograph.

Problem regarding the source of nonequilibrium charge carriers, which multiply by the mechanism of impact ionization and lead to catastrophic increase in current and destruction of a sample (explosion in our case), is important for explaining the mechanism of the electrical form of breakdown. The nonequilibrium charge carriers can appear as a result of electrical excitation of electrons (foreign electrons situated at the levels of capture), and as a result of injections of electrons from cathode or holes in anode [5,6,7,8,9] . The effect of material and polarity of electrons on the electric strength was investigated by us for studying the source of nonequilibrium carriers during the breakdown of TeN_3 and $Cu(N_3)_2$.

The results are shown in fig. 1-2 in coordinates: cumulative frequency of breakdown-voltage of electric field. Data on 80-120 samples was processed by the method described in paper [IO]. Plane electrodes made of different materials were used. It was detected that the material of anodes exclusively affects the breakdown of $Cu(N_3)_2$ and an order of the influence of anode materials with increasing thermodynamic work function, in which the electric strength increases (Zn-Cu-W) was established [II]. The material of anodes as well as cathodes affects the breakdown of TlN_3. The effect of cathode material is stronger, and the order of metals was established opposite to the order for $Cu(N_3)_2$.

On the basis of the effect of electrode material, it can be concluded that the injection is the source of nonequilibrium carriers during the breakdown of $Cu(N_3)_2$ and TlN_3; The source in the former case is hole injection, and for the latter - holes as well as electrons (double injection [7,9]). The change in the electric strength depending on the work function of the meta. cannot be explained by the difference in the height of potential barrier for injection and extraction of electrons, because in this case different orders for cathodes and anodes should be observed for TlN_3 (by the well-known concepts of the theory of contact phenomena 12). The obtained results can be naturally explained on the basis of the theory of transition processes in the system metal-semiconductor-metal which is being developed by one of the authors. According to this theory, in certain cases (presence of fore.gn levels, small carrier mobility) volume charges present on the contact of the metal-semiconductors play a large role in creating near-breakdown conditions even prior to the application of voltage [I2]. Nonuniform distribution of potential occurs in the external field and a zone of high field voltage is created near the electrodes. This zone transforms with time at a finite rate determined by the mobility of charges. In several cases the initial nonuniform distribution creates such conditions which are sufficient for the injection of carriers and the subsequent impact ionization. Such conditions can also be created during the redistribution of charges. In our case, due to extremely small duration during which voltage is applied and small mobility of carriers, the processes of the transformation of volume charges can be neglected. Then, for the condition....... $\varphi_{n} > \varphi_M$ .... the contact voltages can be expressed as:

$$E_K(t) = \frac{V_{br}(t)}{2\ell_D + (\ell - 2\ell_D)\frac{\Delta\gamma\,\Delta(t)}{(\pm 1)(e^{\pm\Delta\gamma} - 1)}} - \frac{\Delta\varphi_K}{\ell_D\,\Delta(t)} \quad \text{(I)} \qquad (1)$$

$$E_A(t) = \frac{V_{br}(t)}{2\ell_D + (\ell - 2\ell_D)\frac{\Delta\gamma\,\Delta(t)}{(\pm 1)(e^{\pm\Delta\gamma} - 1)}} + \frac{\Delta\varphi_K}{\ell_D\,\Delta(t)} \quad \text{(2)} \qquad (2)$$

The sign "+" corresponds to semiconductors with hole conductivity ($Cu(N_3)_2$), and "-" with electron conductivity (TlN_3). Because $\Delta\gamma = \frac{\Delta\varphi_K}{kT}$ and $\Delta\varphi_K = \varphi_n - \varphi_M$, therefore, the obtained relationship (fig. 1, 2) are qualitatively expressed by formulas (1) and (2) for components (relative value of the electric strength, sign of the effect of electrode polarity, order of metals).

Besides, the results of the investigation of the effect of temperature on electric strengths of TlN_3 and $Cu(N_3)_2$ corresponds to the described concepts - with increase in temperature the electric strength for $Cu(N_3)_2$ increases and decreases for TlN_3.

Special experiments were conducted to study the effect of a hole in the near-breakdown zone with constant current to investigate the sign of carriers at the breakdown of TlN_3 and $Cu(N_3)_2$. Obtained results confirm that the breakdown in TlN_3 is caused by electrons and in $Cu(N_3)_2$ - by holes.

Thus, the breakdown in TlN_3 and $Cu(N_3)_2$ can be explained as follows: the breakdown in TlN_3 is caused by electrons which are injected from cathodes, and in $Cu(N_3)_2$ - by holes injected from anodes; both the processes are controlled by nonuniform field distribution in the near contact zones and develop into impact ionization. This can be described as follows:

Injection of holes: $\partial + {}^{\varepsilon}_{0}N \longrightarrow {}^{\varepsilon}_{-}N$ (3)

Impact ionization by electrons: $e + N_3^- \longrightarrow N_3^0 + 2e$ (4)

Impact ionization of holes: $N_3^0 + N_3^- \longrightarrow 3N_2\uparrow + e$ (5)

The future fate of holes is similar to the processes of thermal decomposition, photolysis, and radiolysis of azides of heavy metals:

$$N_3^0 + N_3^0 \longrightarrow 3N_2\uparrow + h\nu \qquad (6)$$

Three possibilities can be mentioned in connection with the mechanism of the excitation of detonation during the breakdown of TlN_3 and $Cu(N_3)_2$:

1) Excitation due to thermal phenomena in the discharge channel;

2) Excitation due to the action of impact wave in the discharge channel;

3) Excitation as the final stage of the development of breakdown because the mechanism of breakdown, as described by equation (3) - (5), is similar to the mechanism of the excitation of a chemical reaction during detonation in azides of heavy metals as described in 1 .

Special investigations have shown that for copper azides (II) the mechanism III might be applicable, because we did not find such small discharge energy which could accomplish the breakdown of $Cu(N_3)_2$ without exciting detonation up to the very capacities, equal to the capacity of the sample (I-2 nф). Cases of the breakdown of TlN_3 ithout detonation were observed, however, the nature of current oscillogram at the time of breakdown shows that the excitation of detonation occurs as the result of the action of impact wave and not due to the action of heat in the discharge channel (absence of arc stage of discharge).

Thus, the phenomena studied during the breakdown of the azide of thallium and azide of bivalent copper allow to describe the electronic mechanism of the breakdown of these objects, and draw conclusions concerning the mechanism of the excitation of detonation during breakdown.

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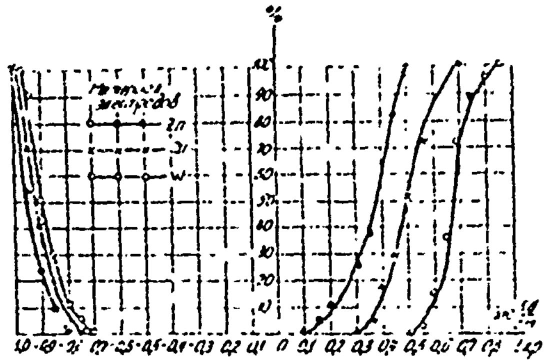

Fig. 1 Effect of material and polarity of electrodes on the breakdown of $Cu(N_3)_2$.

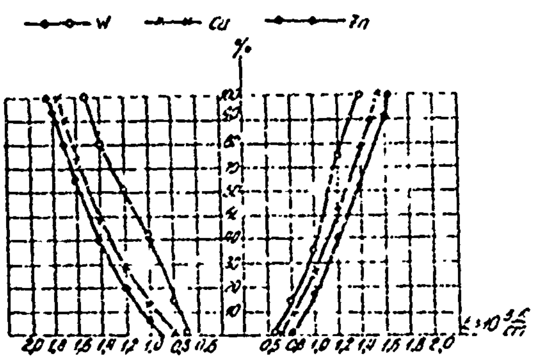

Fig. 2 Effect of material and polarity of electrodes on the breakdown of TlN_3.