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PRELIMINARY STUDIES ON PULSED ELECTRIC FIELD BREAKDOWN OF LEAD AZIDE

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Preliminary results from a study of the electric field induced breakdown of lead azide are reported. Specifically, ~~we discuss~~ the effects of contacts on the initiation of explosives by electric fields, and present preliminary measurements of the transient current in lead azide during the application of a pulsed voltage pulse.* A circuit which eliminates the capacitive transient due to the voltage pulse is described.

* are discussed.

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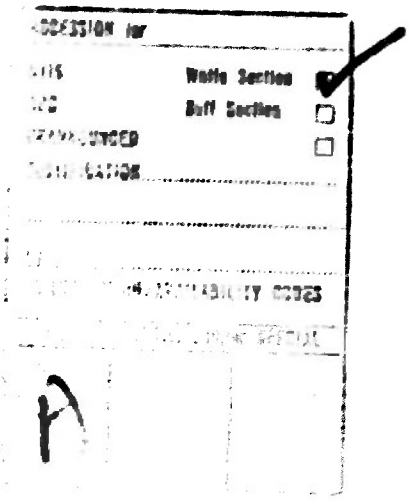
FOREWORD

This program was sponsored by the Nuclear Weapons Effects Program Office (NWEPO), Harry Diamond Laboratory, Adelphi, Maryland under DA Project No. 1W162118AD51 on Nuclear Weapons Effects for Army Projects - Electrical Properties of Lead Azide. The technical monitor for this work was Mr. James. H. Gwaltney, NWEPO.



INTRODUCTION

Lead azide has been observed to initiate when subjected to pulsed radiation during underground nuclear tests (Ref 1-3). Initiation occurred at radiation dose levels well below the thermal initiation level. It has been proposed that the initiation is caused by dielectric breakdown in an intense electric field produced in the material by the radiation pulse (Ref 2-4). Possible mechanisms for generation of this electric field are discussed elsewhere (Ref 4-8). In order to understand the initiation of lead azide by radiation-induced electric fields, it is desirable to know the behavior of the material during the application of a high voltage pulse with risetime and duration approximating that of the induced field. We describe here the development of a technique by which a well characterized voltage pulse can be applied to a pressed pellet explosive sample and the associated conduction current simultaneously monitored. In addition, we have shown that contact and surface effects dominate the dc-electric field initiation properties of lead azide; and the importance of considering this when relating pulsed field initiation results to observations from underground nuclear tests is discussed.

BACKGROUND

A. Contact Effects

Several recent investigations of the effect of electric fields on explosive azides, both in pressed pellet (Ref 9,10) and single crystal (Ref 11,12) form, have attempted to establish a threshold field for initiation or breakdown. Leopold (Ref 9) found a breakdown field of $\sim 3 \times 10^4$ volt/cm when lead azide pellets were in contact with the electrodes. However, if either one or both of the electrodes was "stood off" by a thin mylar sheet, no breakdown was observed. For example, with steady state applied voltages up to 4800 volts across a mylar/lead azide/mylar sandwich, corresponding to an average electric field of 7.4×10^4 volts/cm in the lead azide, initiation did not occur. It appears that the presence of electrical contacts on the surface of the lead azide plays an important role in electric field-assisted initiation. This conclusion is further strengthened by recent Russian work that reported experiments on pressed pellets of copper and thallium azide in which they varied the contact metals and found a dependence of the breakdown field on the metal work function (Ref 10). They attributed this to current injection presumably from ohmic contacts. Subsequently, Mark and Gora (Ref 13) performed an analysis of the Russian work, based on classical Schottky barrier theory, showing that the results are well described in terms of a barrier formed at the metal/lead azide interface whose magnitude depends on material properties, the metal work function and the applied voltage. In their model, initiation is associated with a critical field at the interface, in the region of the Schottky barrier between the metal electrode and the azide sample. Their discussion explicitly takes into account the

particulate nature of the pressed pellets.

In any case, Leopold's results on pressed pellets suggest that the presence of a conducting contact is necessary for initiation to occur at all, at least for average electric fields $\bar{E}$ in the range of those causing initiation in the presence of contacts ($3\text{-}4 \times 10^4$ volts/cm). An average electric field $\bar{E}$ leading to initiation in single crystal samples of lead azide, using a Au/lead azide/Au sandwich geometry, has been reported (Ref 12) to be 3.5×10^4 volts/cm. We will present below experiments performed with dc voltages which extend Leopold's pressed pellet results to higher $\bar{E}$ values; and briefly discuss alternative interfacial mechanisms which may be responsible for initiation.

B. Pulsed Electric Field Measurements

The application of pulsed electric fields to lead azide does not exactly simulate the conditions experienced by a sample exposed to a radiation-induced electric field. It in effect separates out one of the phenomena occurring in the radiation environment, others being the increase in conductivity due to the "injection" of charge carriers by photo-compton processes and the production of charge carriers by excitation processes in the bulk.

In this work, we have developed a technique by which a well characterized voltage pulse is applied to an explosive sample and the associated conduction current simultaneously monitored. This measurement can in principle provide information about changes in conductivity occurring in response to an applied electric field in the absence of other radiation effects.

EXPERIMENTAL

A. Contact Effects

Samples were mounted between mylar insulators in the capacitive geometry shown schematically in Fig. 1. The experiment was conducted in a vacuum chamber at 2×10^{-5} torr. (The high voltage experiments reported by Leopold (Ref 9) were performed with the samples immersed in oil). Care was taken to avoid discharges from the high voltage points of the sample holder to ground. The voltage was increased in steps of 500 volts to a maximum of 5 kV, and was held constant for 60 sec. at each step. Values for $\bar{E}$ (in the sample) were calculated from the applied voltage V_a, the thickness L appropriate to a given sample, the mylar insulator thickness $\ell/2$, and the dielectric constant ratio κ [dielectric constant (lead azide)/dielectric constant (mylar)], by using the relation appropriate to perfect dielectrics:

$$\bar{E} = \frac{V_a}{\kappa \ell + L}$$

No initiation occurred in the pressed pellet samples (pressed to ~ 3.5 gm/cm^3 density, 1-3 x 10^{-2} cm thick), using the insulated electrodes, up to the highest $\bar{E}$-value applied, 1.4 x 10^5 V/cm. This value was determined by the upper limit (5kV) of the power supply and sample geometry. Our highest $\bar{E}$ is a factor of four greater than the $\bar{E}$-values that lead to initiation in contacted samples (Ref 10-12), and a factor of two greater than the highest $\bar{E}$-value reported by Leopold (Ref 9). The same result (no initiation) held for single crystal samples (grown by the method of Garrett (Ref 14) and cut and polished to provide parallel flat surfaces), with the highest $\bar{E}$-value attained being 1.02 x 10^5 V/cm. The maximum $\bar{E}$-values were maintained on the sample for about a half-hour, and some samples were subjected to combinations of 400.0 nm irradiation (strongly absorbed by $Pb(N_3)_2$) and strong field, for both polarities, again with no effect. It has been suggested (Ref 4) that the application of such radiation may increase the local field at the irradiated surface due to changes in conductivity, and it has been observed (Ref 4) that the threshold $\bar{E}$-values for contacted single crystal samples decrease by a factor of two with concurrent irradiation.

B. Pulsed Electric Fields

When a voltage pulse is applied to an insulating specimen such as lead azide, it is accompanied by a substantial capacitive current pulse. This displacement current can effectively interfere with the measurement of conduction current in the material. To overcome this problem, we have designed a balanced circuit as shown in Fig. 2. On closing the mercury wetted contact relay S, the voltage +V/2 appears at P_2 and -V/2 appears at P_1. The point P_2 is connected to the sample contact on the high voltage side and to the y-axis of one beam of a dual beam oscilloscope S_1. P_1 is connected to a variable capacitor C_v. The internal resistance of the battery is shunted by the 12 μf capacitance in parallel so that the full voltage appears at P_1 and P_2 in approximately 10^{-7} seconds. The current through the sample is obtained by measuring the voltage developed across the load resistance R_L with the second beam of the oscilloscope connected at S_2. Sensitivity is varied by changing R_L. C_L represents the unavoidable stray capacitance of the measuring circuit (resistors, cables, etc.). The displacement current transient due to the sample capacitance C_s is compensated by the equal and opposite transient through C_v which is adjusted to equal C_s. For a sample of thickness d, the average applied field was $V/(2d)$ where V_{max} was 2000 volts.

The entire circuit was contained in a metal box and the evacuable sample chamber was mounted on top of the box to make leads as short as possible and reduce stray capacitance. This arrangement

is shown in Fig. 3.

Samples were mounted on teflon discs, electroded with silver paint and potted in RTV. Pellets of both PVA and RD 1333 lead azide were tested. PVA lead azide samples pressed to 54,000 psi were exposed to maximum average fields of 4.9×10^4 volts/cm. Four of ten pellets tested initiated in forepump pressure vacuum ($\sim 10^{-2}$ torr). None could be initiated at ambient conditions. The time between application of the voltage and detonation was observed to vary, sometimes being greater than the duration of the oscilloscope trace. Current and voltage traces of an event which began after 22 μs are shown in Fig. 4. Initially the current through the sample was less than 10^{-4} amp. At approximately 22 μs, the current increased rapidly with a corresponding drop in voltage. 16 μs later, there was a sharp drop in the voltage which is evidence for the actual detonation of the pellet.

Several pellets of RD 1333 with densities between 2.62 and 3.26 were exposed to pulsed fields as high as 6.5×10^4 volts/cm at both vacuum and ambient conditions. None of these samples could be initiated even with repeated application of the pulsed field.

As an alternative approach, designed to obtain voltage transients more similar to those produced by the radiation-induced field, a high voltage pulse generator was acquired. The Pacific Atlantic Trading Co. Model PT55 was chosen for the experiments since it is capable of producing 55 kV pulses with 2 ns risetimes and 75 ns pulse width.

A trigger pulse of at least +250V with risetime of 10 ns was required. This was accomplished with a circuit designed around a Potter-Brumfield mercury-wetted-contact relay JML-1240-81. A schematic is shown in Fig. 5.

Lead azide pellets were mounted in specially designed fixtures obtained by one of us (LA) from Sandia* and shown schematically in Fig. 6. The electrode geometry of these fixtures assures a highly uniform electric field in the region where the sample is held. In addition, the electrodes provide a reliable witness to the occurrence of a detonation. A chamber which can be evacuated or filled with various ambient gases was designed to incorporate these fixtures and can be seen in Fig. 7 along with the associated electronics.

Several lead azide pellets tested with the electrodes in direct contact and at ambient conditions were initiated with the 55 kV pulse. We have also made preliminary tests with this system using mylar standoffs between the electrodes and the pellet. With the mylar standoffs and at atmospheric pressure, the lead azide always initiated.

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* See Acknowledgment

However, in one experiment where the edge of the pellet was potted in RTV so that all air gaps between the mylar standoffs were eliminated, initiation did not occur.

DISCUSSION

A. Contact Effects

The results of our experiments on contacted and non-contacted samples in dc fields lead us to conclude that electrode interface effects dominate the field initiation of $Pb(N_3)_2$ when it is directly in contact with the electrode material. There are conflicting possibilities for the role of contacts in the mechanism leading to initiation. The electronic current injection mechanism (Ref 10) offered to account for field initiation in $Cu(N_3)_2$ and TlN_3 explicitly requires ohmic contacts. Hot electronic carrier generation would then be unlikely because of the characteristic field distributions associated with single injection (Ref 15) so that initiation would most probably be a result of either ohmic heating, or of a high density of (reactive) excited electronic states in the form of injected carriers (Ref 16,17). For a model based on electronic carrier emission from a barrier contact (Ref 3), there are three possible mechanisms which rely on high local internal fields to generate hot electrons which lose energy by impact ionization. The first, due to O'Dwyer (Ref 18), assumes that the impact ionization causes avalanche multiplication of electronic carriers which eventually break down at the exit electrode where most of the energy is dissipated by impact - generated carriers. The second mechanism, suggested by DiStefano and Shatzkes (Ref 19), suggests that impact ionization generates a space charge in the volume of the specimen that concentrates the internal field at the emitting electrode and gates an increasing flow of emitted current with eventually catastrophic power dissipation at the entrance electrode. Finally, as there is strong evidence for ionic conductivity dominating the low - field equilibrium current (Ref 20), ionic conductivity may also be playing a role in electrode - dominated initiation. For example, electronic carriers emitted from the electrodes might attach themselves to ions (lattice ions or imperfections such as impurities and vacancies), and high - field anomalies in the ion current then might be responsible for the ultimate initiation. More experimentation is required to determine the exact nature of the interface - dominated initiation process.

These results are particularly important in the analysis of experiments designed to simulate the response of lead azide to a radiation-induced electric field. It has been proposed that the induced field arises in the sample bulk due to the separation of charge produced by the radiation (Ref 2,3,7), a situation presumably different than that of a high field at the sample-electrode interface. In the electroded experiments the contact provides the charge carriers which are emitted into the bulk through the high field region

formed at the interface. One would expect quite different field distributions in the bulk during radiation experiments as a consequence of the photo-carriers generated by the radiation. Therefore, care must be exercised in correlating results from the two types of experiments.

B. Transient Measurements

The circuit designed to compensate for the displacement current arising from the polarization of the sample is quite suitable for the purpose of measuring current through the sample. We suspect, however, that no current is injected from the contacts and that currents originate only by emission from the Schottky barrier when sufficiently high voltages are attained. The current then increases rapidly by one of the avalanching processes discussed above. This is supported by the data shown in Fig. 4. After application of the voltage the measurable current is less than 10^{-4} amp. It is interesting to note that the pellet does not initiate as the voltage is applied but there is a measurable delay time. Also, the actual detonation, indicated by the sharp drop in voltage, follows a short period during which a relatively high current flows through the pellet.

Although the above result demonstrated that the experimental technique is quite applicable to the study of electric field initiation of explosives, it became clear that pulses of shorter duration would have to be higher in magnitude in order to achieve initiation. The original circuit was limited to rise times of $\sim 10^{-7}$ sec and maximum voltage of 2000 volts.

Our preliminary experiments with the shorter rise time high voltage transient generator indicate that with metal contacts lead azide pellets are readily initiated. The next step in the program should be to incorporate the high voltage generator into a balanced circuit similar to the one described above with appropriate instrumentation to record voltage and current response in nanosecond time frames. In addition, the experiments should be performed with both electroded and non-electroded sample geometries. It appears that very high fields can be applied, at least for short times, without initiation if the sample is not contacted. The effect of simultaneous irradiation under this condition would be interesting.

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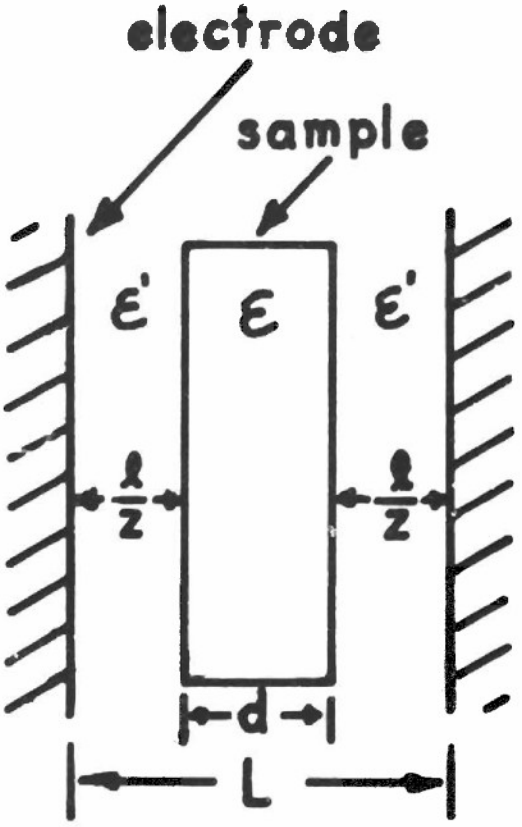


Fig 1 Capacitive Sample Geometry

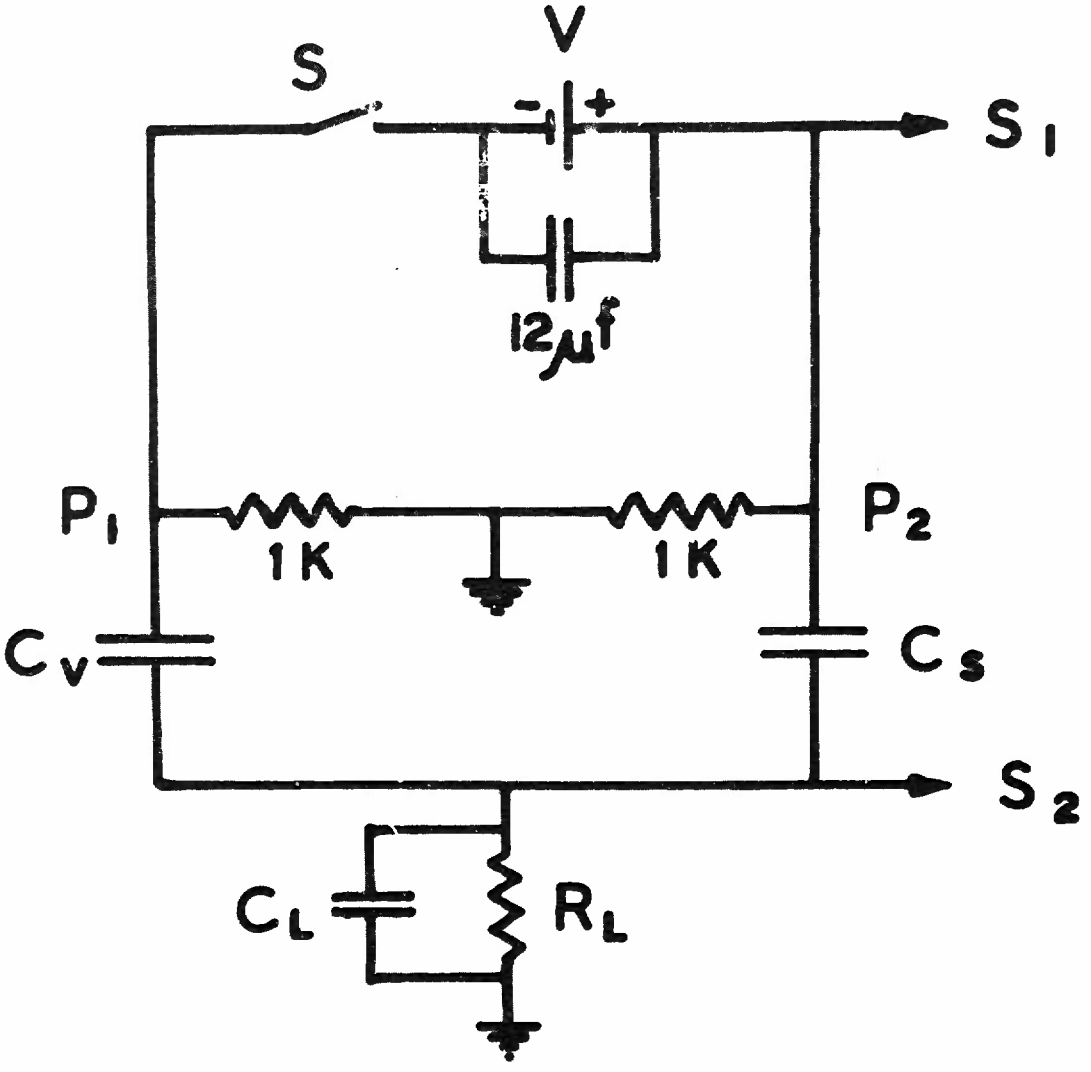
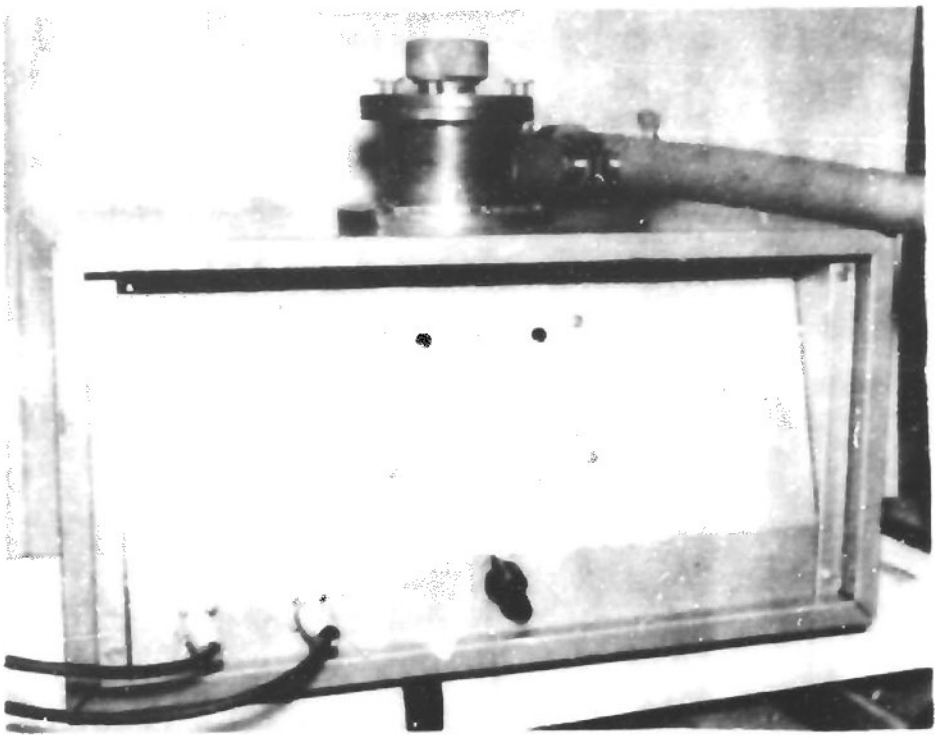


Fig 2 Schematic of the Balanced Circuit



(a)

(b)

Fig 3 Circuit Housing and Sample Chamber

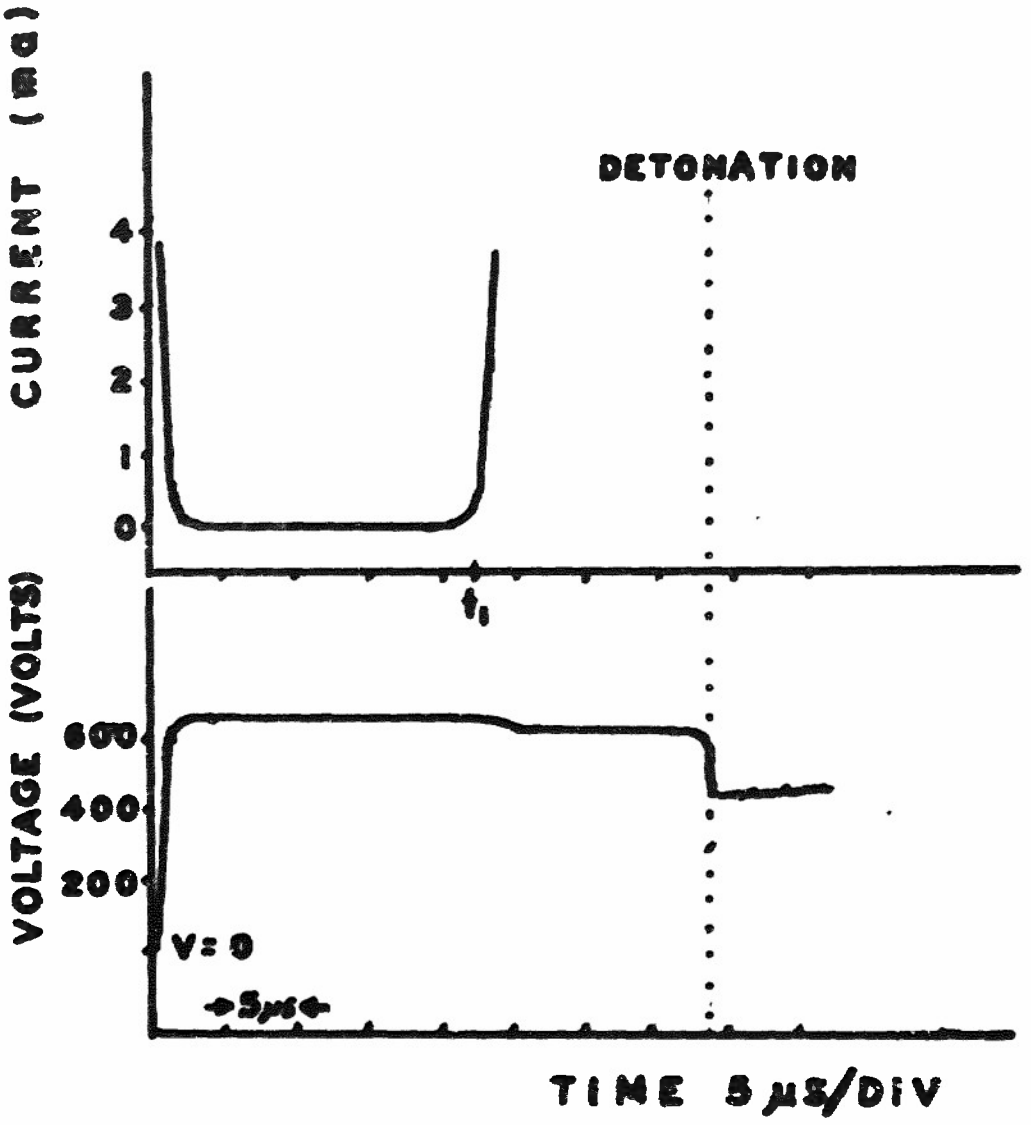


Fig 4 Oscillographs of Current and Voltage vs Time

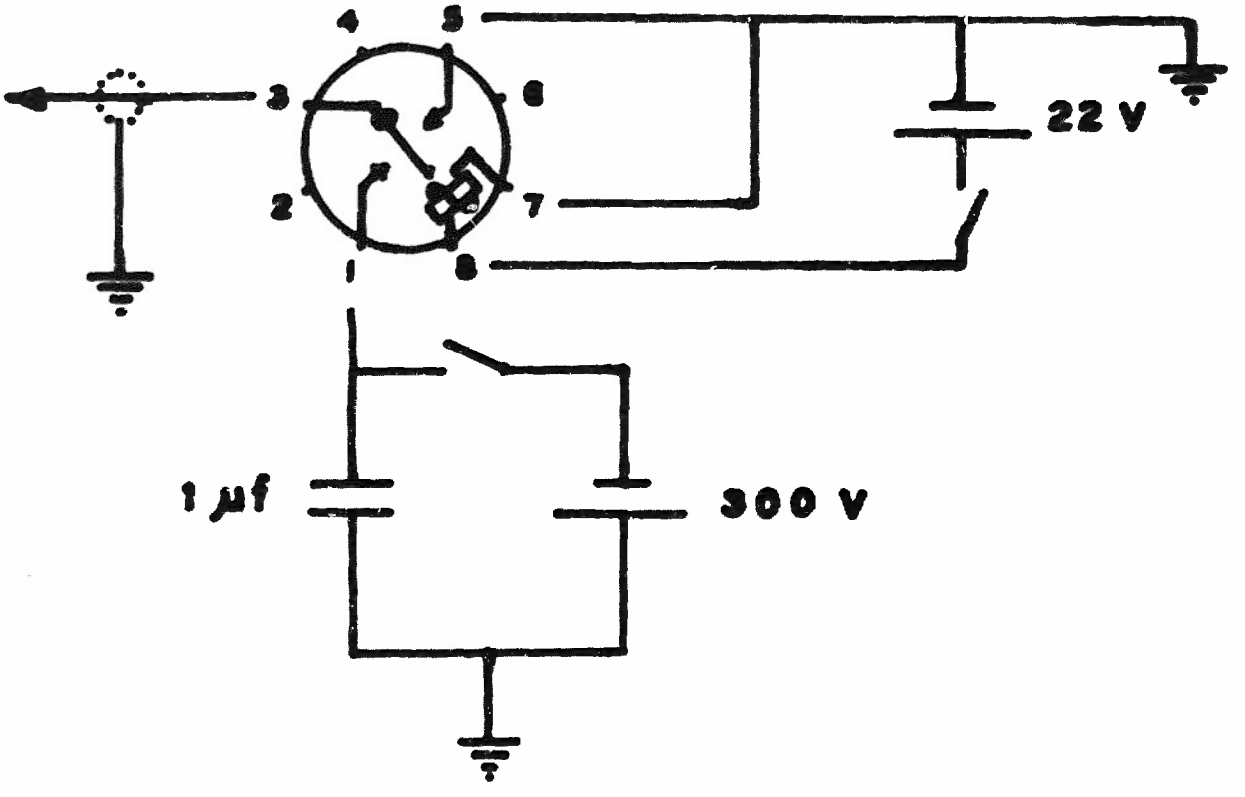


Fig 5 Schematic of the Triggering Circuit

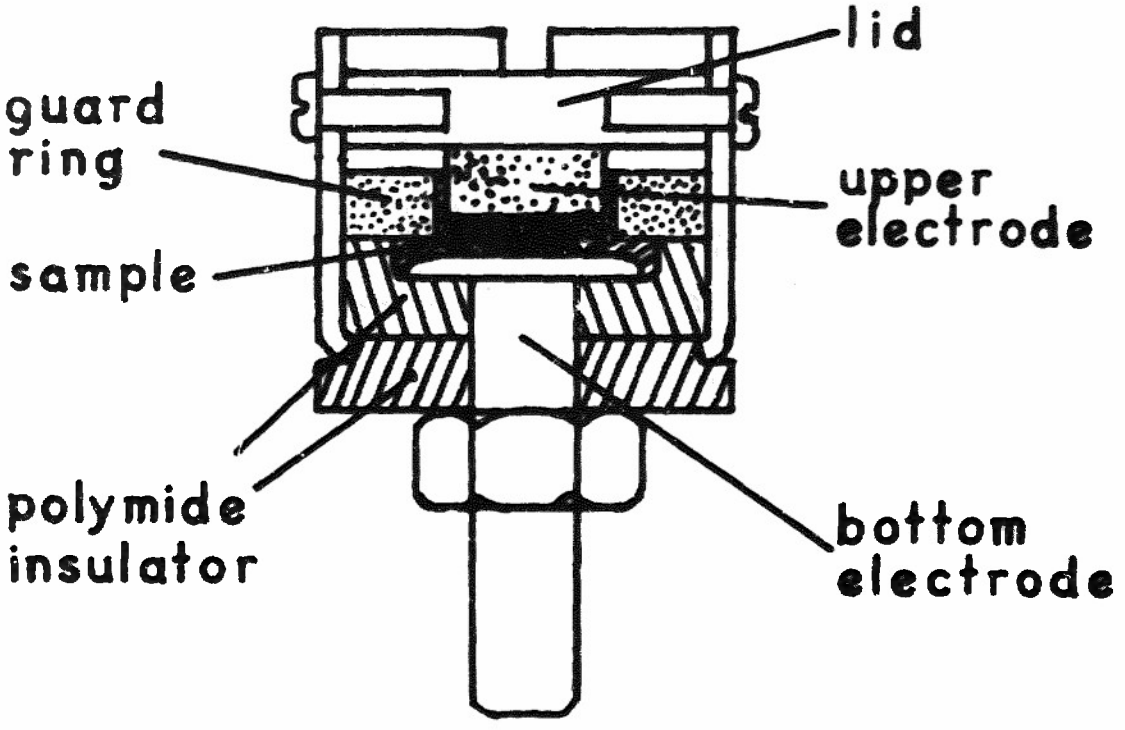


Fig 6 Schematic of the High Voltage Test Fixtures

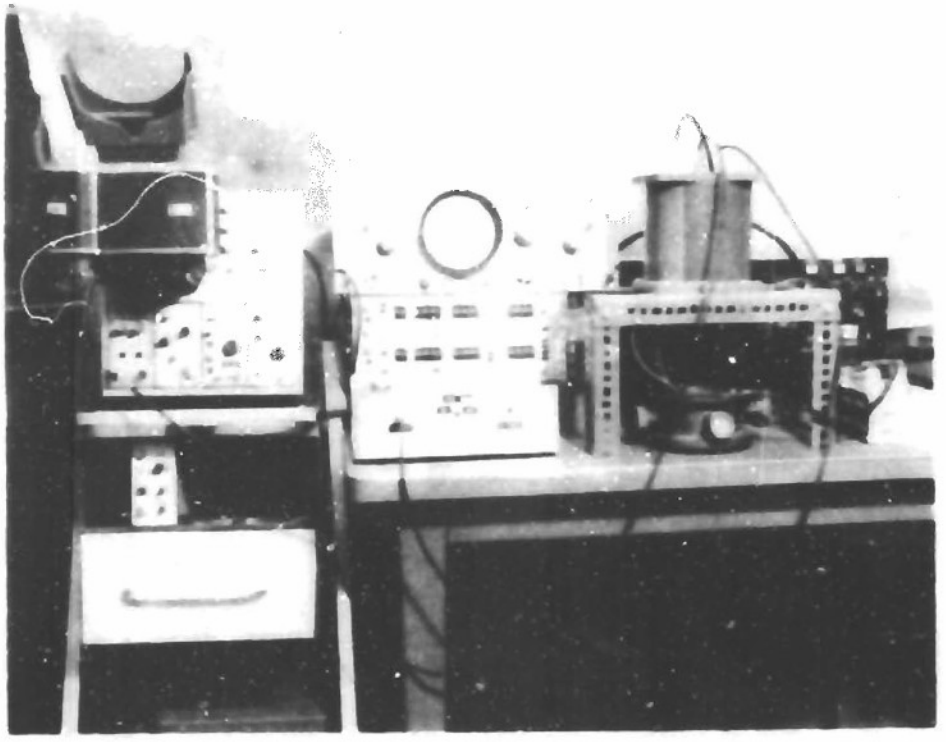


Fig 7 Equipment for the Nanosecond Pulse Experiment

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