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***Evaluation of the Effects of Radiation-Induced
Conductivity on Charge Separation in Nuclear
Weapons During Radiographic Inspection***

Analysis and Experiments

***Eugene H. Farnum
Michael D. Holder***

November 4, 1997

I Summary

Radiography is routinely used for non-destructive inspection of nuclear weapons at Pantex. For example, X radiography can be used to observe the positions of valves, to verify that a stronglink is in the safe position, or to inspect internal mechanical assembly details. Because of the presence of heavy metals in warheads, such operations are carried out with high-energy X-rays produced by linear accelerators (Linacs), and substantial doses can be accumulated, especially if images from more than one direction are required.

In December 1996, the basis for safety assurance of Linac operations at Pantex was called into question. Questions concerned the level of electrical charge separation in the high explosive (HE) dielectric and possible consequences of high electrical fields.

Linac operations, which affect other critical missions at Pantex, were suspended and the Weapons Labs were asked to perform a critical analysis to determine what controls were required to assure safety. The postulated mechanism by which fields could build-up involved creation of charge (primarily Compton Electrons) by incident X-ray photons, and their accumulation in the high explosive. Building on a model originally developed by Mike George¹ at Los Alamos for somewhat different conditions, Livermore developed a model which predicted the voltages which would occur in the vicinity of the detonator cables, and showed how these voltages depend on bulk resistivity of the HE and on the dose and dose rate. We proposed that the effects of radiation induced conductivity (RIC) would dominate, and showed that at steady state, neither dose nor dose rate would affect the voltage. We also proposed a series of experiments on HE assemblies to measure the RIC and to confirm the level of voltages attained.

The experiments were conducted in March and April at the Los Alamos high-energy Microtron X-ray machine. Other experiments were conducted at Livermore. These efforts were successful and showed that voltages were insignificant, and did not depend on dose or dose rate. Operations were resumed at Pantex on April 19, 1997 after joint presentations of these models and experimental results to the Department of Energy's Deputy Assistant Secretary for Military Application and Stockpile Management (DASMASM), and to members of the Defense Nuclear Facilities Safety Board (DNFSB.)

II The Compton Effect

Absorption of X rays in materials is the result of the photoelectric effect, electron-positron pair production, and the Compton Effect (figure 1). Absorption of photons can stimulate nuclear effects, photoneutrons, photoprotons, and photofissions, however the cross-sections are extremely small. At the photon

Figure 1. Absorption in Iron [D. W. Kerst, *American Scientist*, 35, (1947)]

energies available from the Pantex Linac, largely between 1 and 4 MeV, the Compton effect dominates.

At the 1922 meeting of the American Physical Society Arthur H. Compton asserted that the scattering of X rays by matter could be described as quanta of radiation bouncing off electrons. Using this approach Compton was able to describe the increase in wavelength of scattered X rays. Compton came to this result despite his best efforts to describe the dispersion of X rays using classical wave theory. He, like many of his peers, was reluctant to apply Einstein's hypothesis of light quanta.

Compton also described the behavior of the electrons struck by photons which recoil with an energy that is a function of the incident and scattered radiation angles. Recoil electrons may be boosted into a higher energy level within the atom, perhaps into the conduction band of the bulk material, or liberated from the atom entirely. It is those liberated electrons that may separate onto metal conductors in a weapon assembly or into insulators like HE, and given sufficient quantity, could provide stored electrical energy sufficient to initiate the main weapon detonators.

III Conductivity

If all of the materials in weapons were conductors, there would be no charge separation, and no safety question. All Compton electrons produced during radiography would be quickly and safely returned to their place of origin. But within weapon assemblies there are insulating materials, most notably the high explosives (HE) which are excellent insulators. These insulators can trap electrons and slow their return to their place of origin.

Charge separation in the HE presents a unique situation in weapon assemblies. Flat detonator cables are routed along the surface of the HE under case metal material. In this configuration a capacitor is formed between the flat cable and the metal case. Given an electrical field in the HE, a charge develops in the cable-case capacitor. The charge in the cable-case capacitor could, given a large X-ray dose and no conduction, reach a level sufficient to provide the energy required to fire detonatorsⁱⁱ.

The conductivity of the HE and the Compton current define a maximum steady-state field achievable for a given X-ray flux. Figure 2 shows a circuit model for this effect. The maximum voltage in the capacitance of the HE is the product of the HE resistance and the Compton current.

PBX-9501 explosive exhibits a normal conductivityⁱⁱⁱ of $5 \times 10^{-13} \Omega^{-1}\text{cm}^{-1}$, and PBX 9502 is $2 \times 10^{-15} \Omega^{-1}\text{cm}^{-1}$.

Figure 2. The steady-state electric field developed in the explosive dielectric is proportional to the Compton electron flux, and inversely proportional to the normal conductivity of the explosive.

The Linatron 3000 X-ray source used at Pantex has a coarse and fine pulse structure (figure 3). The fine structure has 83 ps pulses separated by 250 ps. These are combined into 4 μ s coarse pulses that are separated by 3 ms. The overall duty factor, the fraction of time the beam is actually on, is about 1/3000. Therefore the peak flux is 3000 times greater than the average dose rate, or about 5×10^3 rad/s during normal operations.

Calculating the maximum steady-state field using the peak flux and the normal conductivity predicts an electric field of 1.5×10^5 V/m in PBX 9502 at 3 meters from the Linac. Applying this value to the cable-case capacitor configuration results in 23 μ J of stored energyⁱⁱ, an amount insufficient to fire detonators by orders of magnitude.

This mechanism does not provide assurance should something happen to dramatically reduce the distance between the Linac and the weapon. The peak flux in a pulse can be as high as 2×10^5 rad/s at one meter from the source, and higher still at the minimum distance (40 cm) limited by the Linac housing. The flux is proportional to the inverse square of the distance, so the safety margin could be dramatically reduced.

IV Radiation Induced Conductivity (RIC)

RIC in insulators is a well-known phenomenon^{iv} that decreases the resistivity of the insulator while it is exposed to ionizing radiation. The RIC effect occurs when electrons are excited into the insulator's conduction band by the ionization process. The lifetime of the excited electron depends on the available extrinsic and intrinsic traps and is very short (decay time of RIC has not been measured directly but is considered to be < 100 ps in most insulators^{iv}). Thus we assume for this analysis that the radiation-induced conductivity vanishes immediately when the irradiation is stopped. The RIC effect is expressed by the formula^{iv}:

$$\sigma = \sigma_0 + k \phi^\delta \quad (1)$$

where σ is the electrical conductivity, σ_0 the conductivity in the absence of radiation, ϕ is the radiation ionizing flux and k and δ are constants. In nearly all cases, $\delta = 1.0$ and, in cases where RIC dominates, $\sigma \gg \sigma_0$. Thus $\sigma \cong k \phi$.

The value of the RIC efficiency constant, k depends on the material, its purity, its degree of crystallinity and extrinsic defects caused by irradiation. For crystalline ceramics it is remarkably similar^{iv}, being about 8×10^{-15} s / [Ω cm rad] for most ceramics and varying from 5×10^{-15} s / [Ω cm rad] up to 5×10^{-13} s / [Ω cm rad] in the most pure and undamaged materials^{iv}. In polymers, however, the

Figure 3. LINAC Fine Pulse Structure

measured values of k vary by more than four orders of magnitude, generally increasing with increasing crystallinity^{v,vi}. Most polymers measured show lower RIC constants than ceramics, their k values varying between 10^{-15} s / [Ω cm rad] and 10^{-19} s / [Ω cm rad] with polytetrafluoroethylene and polyimide being among the highest. The value of k has not been measured for explosives. However, since explosives are mostly crystalline we estimated that PBX 9502 will have a k of 5×10^{-15} s / [Ω cm rad], at the lower end of the values measured for crystalline ceramics and the upper end of those measured for polymers. Using this value of k as an example, irradiating PBX 9502 with a peak flux of only 0.4 rad/s will increase its conductivity ($\sigma_0 = 2 \times 10^{-15} \Omega^{-1} \text{ cm}^{-1}$) by a factor of 2.

V Analysis

The important contribution of RIC is that both the generation rate of Compton electrons and the conductivity of the explosive are proportional to the X-ray flux. At some level of electric field in the explosive the return current due to RIC balances the forward Compton electron current. In this limiting equilibrium condition the charge added per pulse will be balanced by current during the pulse due to RIC. Figure 4 shows a circuit model including the RIC effect. The common switch elements show how the RIC effect and Compton current are coupled and occur at the same time during X-ray pulses. We wish to determine the electric field in the explosive at which the Compton current and the return leakage current are equal, a condition which limits the stored energy.

The following analysis assumes, for simplicity, that all the Compton electrons deposited in a given thickness of explosive collect on an imaginary electrode imbedded in the explosive. The model assumes that RIC is large so $\sigma \gg \sigma_0$, and that when the X-ray pulse is off negligible current flows. Therefore, we consider only RIC that happens during the pulse and assume that the charge is fully separated to the imaginary electrode. The pulse is assumed to be a square shape in time. All units are evaluated per unit area. The following nomenclature will be used.

- Q = charge transferred into explosive per X-ray pulse
- i_r = return current caused by voltage V across explosive
- i_x = Compton electron current caused by the X-ray flux ϕ
- ϕ = X-ray flux (rad/cm² s)
- t = width of a single pulse in time
- Φ = X-ray fluence in one pulse = ϕt (rad/cm²)
- V = voltage developed across electrodes at equilibrium (steady state) during irradiation
- d = electrode separation
- A = electrode area in beam

Figure 4. Equivalent circuit with radiation-induced conductivity contribution. The switch elements represent the coupling between Compton electron currents and RIC which occur simultaneously during the x-ray pulse.

- R = resistance of a unit area of the explosive during irradiation
 σ = RIC during irradiation
 B = charge separation efficiency (coulombs / rad)
 k = RIC generation efficiency (s / Ω cm rad)

The resistance of the explosive due to RIC in the irradiated region is given by:

$$R = \frac{d}{A\sigma} = \frac{d}{\sigma} \text{ per unit area,} \quad (2)$$

and from (1) above, with $\delta = 1$ and $\sigma_0 \cong 0$, (2) becomes:

$$R = \frac{d}{k\phi} \text{ per unit area.} \quad (3)$$

Then the return current for a unit area is :

$$i_r = \frac{V}{R} = \frac{Vk\phi}{d}. \quad (4)$$

The Compton charge transferred per unit area during a single pulse, Q , is proportional to the fluence in the pulse, so:

$$Q = B\Phi = B\phi t. \quad (5)$$

This represents a current, i_x , where $Q = i_x t$. (6)

Therefore, from (5) and (6), $i_x = B\phi$. (7)

Equilibrium is achieved when the charging current from the Compton electrons is equal to the return current from the voltage induced by the Compton charging. Thus during the pulse, $i_x = i_r$ or from (4) and (7):

$$\frac{Vk\phi}{d} = B\phi. \quad (8)$$

and solving for V (9)

$$V = \frac{dB}{k}.$$

The electric field, E , is then: (10)

$$E = \frac{B}{k}.$$

Thus, when the RIC effect dominates the conductivity of the explosive, the equilibrium voltage during X-ray irradiation is independent of flux and depends only on the RIC generation efficiency, the Compton charge generation efficiency and, in this simplified model, the charge separation.

VI Measurements of RIC in PBX 9502 & PBX 9501

RIC generation efficiencies for ceramics and polymers have been reported, but for explosives have not. RIC in cylindrical samples of explosives PBX 9502 and PBX 9501 were measured while irradiated by the 10 MeV Microtron X-ray source at Los Alamos. The X-ray dose rate was 18.7 rad/s, at 60 Hz pulse rate, with a duty factor of 6×10^{-5} . The peak flux was 3.1×10^5 rad/s. The X-ray beam was collimated to a 34.5 mm diameter spot approximately in the center of the 80 mm diameter explosive disk.

The explosive samples (figure 5) were glued with a fine layer of Sylgard® 184 encapsulant between two stainless steel plates, which act as the electrodes. The explosive-metal-plate sandwiches were mounted in an aluminum housing and insulated from the housing by nylon® mounting hardware to provide isolation and shielding. The sample assemblies were illuminated with X-rays from the Microtron while conductivity measurements were made.

Given an ideal sample and electrode configuration the conductivity could be measured by impressing a known voltage across the sample, and then measuring the resulting current. Conductivity could then be calculated from the voltage, current, and the geometry of the sample assembly. This approach is inaccurate given the low conductivity of the explosive material, surface leakage, and the difficulty of connecting electrodes to it. This inaccuracy can be reduced by measuring the current as a function of voltage over a wide range.

A Keithley 6517 electrometer, driven by a computer data acquisition system, was used to measure the conductivity of the sample (figure 6). The conductivity was determined by impressing a known voltage across the sample while measuring the resulting current at up to 100 different impressed voltages between -600 and +600 volts D.C. Given ideal circumstances the resulting data would form a straight line through zero. The inverse slope of the line would then be equal to the resistance of the sample. In practical measurements the electrode contact resistance, polarization, and other effects produce a nonlinearity around zero volts. The data is analyzed by fitting the slope of the data, by least-squares regression, on either side of zero volts where it is linear. The slope of the data above zero volts and the slope below zero are then averaged to determine the resistance of the sample. 3 beam-off measurements, 2 before irradiation and 1 after, were analyzed for each sample. The average value of the 3 slopes was used to calculate σ and σ_0 . For PBX 9502 (figure 7), 3 measurements taken while the sample was irradiated were analyzed and

Figure 5. Explosive Assembly for RIC Measurement. Dimensions in mm.

Figure 6. Conductivity Test Layout

**Fig. 7. Conductivity of PBX 9502 in 10 MeV Beam
2.8 cm Diameter Beam, 0.3 cm Thick Sample, 1120 rad/min.**

averaged to calculate σ . For PBX 9501 (figure 8), 5 measurements taken while the sample was irradiated and were analyzed and averaged to calculate σ and σ_0 . In PBX 9501 the difference between the irradiated and unirradiated slopes appears small due to the higher normal conductivity, the low duty cycle of the X-ray source, and the small area of the sample being illuminated.

For PBX 9502 The average of three measured slopes was $3.1 \times 10^{-12} \Omega^{-1}$, or $3.2 \times 10^{11} \Omega$, while the sample was irradiated. The standard deviation(s) of the slopes was $1.3 \times 10^{-12} \Omega^{-1}$. The average of three measured slopes was $2.4 \times 10^{-14} \Omega^{-1}$ ($s=4.7 \times 10^{-16}$) with the beam off.

Since $\sigma_0 \ll \sigma$ for PBX 9502, we can ignore the beam-off slope and calculate RIC from the resistance, R , the irradiated area A_{irr} , and the thickness of the sample, d .

$$\text{Conductivity, } \sigma = \frac{d}{R \cdot A} = \frac{0.3 \text{ cm}}{3.2 \times 10^{11} \Omega \cdot 9.3 \text{ cm}^2} = 1.0 \times 10^{-13} \Omega^{-1} \text{ cm}^{-1}$$

Scaling for the X-ray pulse duty cycle gives an instantaneous RIC of $1.7 \times 10^{-9} \Omega^{-1} \text{ cm}^{-1}$ at the peak flux of $3.1 \times 10^5 \text{ rad/s}$. The conductivity of the sample measured without X-ray irradiation was $1.5 \times 10^{-16} \Omega^{-1} \text{ cm}^{-1}$. Thus during irradiation, the conductivity in PBX 9502 increases by 7 orders of magnitude.

For PBX 9501 The average of three measured slopes was $7.8 \times 10^{-10} \Omega^{-1}$ ($s=9.8 \times 10^{-11}$), or $1.2 \times 10^9 \Omega$, while the sample was irradiated. The average of three measured slopes was $6.5 \times 10^{-10} \Omega^{-1}$ ($s=4.7 \times 10^{-11}$) with the beam off.

In this case, we cannot ignore the effect of the beam-off conductivity in calculating RIC. The beam-on slope, R_{irr}^{-1} becomes

$$R_{irr}^{-1} = \frac{\sigma A}{d} + \frac{\sigma_{RIC} A_{irr}}{d} \left(\frac{t_{on}}{t_{off}} \right)$$

Where $\frac{t_{on}}{t_{off}}$ is just the Microtron duty factor, 6×10^{-5} , and $\frac{\sigma_0 A}{d} = R_{off}^{-1}$.

$$\text{Thus, } \sigma_{RIC} = \left(R_{irr}^{-1} - R_{off}^{-1} \right) \left(\frac{d}{A} \right) \left(\frac{t_{off}}{t_{on}} \right). \quad (11)$$

For PBX 9501, $\sigma_{RIC} = 7 \times 10^{-8}$ ($s=6 \times 10^{-8}$) $\Omega^{-1} \text{ cm}^{-1}$. It should be noted that for PBX 9501, the difference between the irradiation on and off slopes is small. Thus the error in calculating the σ_{RIC} is large, with an estimated standard error of 80%.

Fig. 8. Conductivity of PBX 9501 in 10 MeV Beam
2.8 cm Diameter Beam, 0.3 cm Thick Sample, 1120 rad/min.

The conductivity of the sample measured without X-ray irradiation was $3.9 \times 10^{-12} \Omega^{-1} \text{ cm}^{-1}$. Thus, during irradiation the conductivity in PBX 9501 increases by approximately 5 orders of magnitude.

The RIC coefficient, k , can be calculated from the average slopes of the irradiated and unirradiated conductivity measurements, in a similar manner:

$$\text{RIC Coefficient, } k = \frac{\sigma_{RIC}}{\phi} = \frac{(R_{irr}^{-1} - R_{off}^{-1}) \left(\frac{d}{A} \right) \left(\frac{t_{off}}{t_{on}} \right)}{\bar{\phi} \left(\frac{t_{off}}{t_{on}} \right)} = \frac{(R_{irr}^{-1} - R_{off}^{-1}) d}{\bar{\phi} A_{irr}}, \quad (12)$$

When $\bar{\phi}$ is the average flux.

$$\text{For PBX 9502, } k = \frac{(3.1 \times 10^{-12} - 2.4 \times 10^{-14}) \cdot 0.3 \text{ cm}}{18.7 \frac{\text{Rad}}{\text{s}} \cdot 9.3 \text{ cm}^2} = 5.3 \times 10^{-15} \text{ s} / [\Omega \text{ cm rad}]$$

(s = 2.2×10^{-15})

$$\text{For PBX 9501, } k = \frac{(7.8 \times 10^{-10} - 6.5 \times 10^{-10}) \cdot 0.3 \text{ cm}}{18.7 \frac{\text{Rad}}{\text{s}} \cdot 9.3 \text{ cm}^2} = 2.2 \times 10^{-13} \text{ s} / [\Omega \text{ cm rad}]$$

(s = 1.9×10^{-13})

VII Expected E-Fields in Explosives During Radiography

The equilibrium voltage expected between the metal plates in the PBX 9502 explosive sample assembly can be calculated. We use k in PBX 9502 = $5.3 \times 10^{-15} \text{ s} / [\Omega \text{ cm rad}]$ as measured. To obtain the Compton charge generation efficiency, B , let us take the deposition charge density in the outer zones of the explosive $8.1 \times 10^{-13} \text{ coulombs/g/rad}^{\text{vii}}$ which in a unit area of explosive 0.3-cm thick, with a density of $1.88 \text{ g} / \text{cm}^3$ deposits a charge of $2.4 \times 10^{-13} \text{ coulombs} / \text{rad}$. Thus $B = 2.4 \times 10^{-13} \text{ C/rad}$ and the voltage necessary to achieve equilibrium during the pulse is given by equation 9:

$$V = \frac{(0.3 \text{ cm})(2.4 \times 10^{-13} \text{ C / Rad})}{5.3 \times 10^{-15} \text{ s} / [\Omega \cdot \text{cm} \cdot \text{Rad}]} = 14 \frac{\text{C}\Omega}{\text{s}} = 14 \text{ V}$$

and the resulting electric field is $E = \frac{B}{k} = 45 \text{ V/cm}$.

VIII Voltage Measurements

Samples of explosives PBX 9502 and PBX 9501, with potted detonator cables to simulate the weapon configuration (figure 9), were irradiated with the 10 MeV Microtron X-ray source. Voltages were measured between the outer metal plate(closest to the X-ray source) and the outer detonator cable conductor, inner detonator conductor, and inner metal plate(farthest from the source).

The same Keithley 6517 electrometer, with slightly different schematic configuration (figure 10), was used to measure the voltage on the various conductors in the assembly with respect to time. All voltages were measured with respect to the outer metal plate.

Figure 11 shows the voltage that appears on the outer detonator cable conductor in an assembly with PBX 9502 while illuminated with X rays. During the measurement the beam was switched off for 4 minutes, and then on again. The charge did not dissipate significantly while the beam was off. This is consistent with the low conductivity of the Kapton® insulator in the cable and of PBX 9502. Figures 12 and 13 show the measured voltages on the inner detonator conductor and inner plate.

Figures 14 and 15 show the measured voltages on the outer detonator conductor and inner detonator conductor with PBX 9501.

Figure 16 shows the measured voltage between the outer metal plate and the inner metal plate during two long X-ray exposures of the PBX 9501 sample assembly. During each run the beam was switched off for a period, and then on again. The voltage across the metal plates dissipated significantly while the beam was off. This is consistent with the higher normal conductivity of the PBX 9501.

IX Discussion

Both PBX 9501 and PBX 9502 have RIC coefficients in the predicted range, increasing conductivity during irradiation by 7 orders of magnitude for PBX 9502 and 5 orders of magnitude for PBX 9501. Given these k values RIC dominates over normal conductivity during radiography operations.

Voltages measured on the various conductors in HE/detonator cable assemblies were universally small. In each case the voltage achieved after X-ray exposure for up to 15 minutes was less than 1 volt. This value is substantially lower than predicted using the simplified RIC circuit model for the sample geometry, and is 2 orders of magnitude smaller than the level predicted without RIC.

Figure 9. Explosive-Metal Assembly for Voltage Measurement. Dimensions in mm.

Figure 10. Voltage test layout

Figure 11. Voltage developed between the outer case and the outermost detonator cable vs time. The dose rate was 22.8 rad/s at the sample.

Figure 12. Voltages developed between the outer-plate and the innermost detonator cable conductor vs. time. The dose rate was 22.8 rad/s at the sample. Two measurements with the test device in the beam and one run with it out of but next to the beam are shown.

Figure 13. Voltages developed between the end-plate facing the beam and the innermost end-plate vs. time. The dose rate was 22.8 rad/s at the sample. Two measurements with the test device in the beam and one run with it out of but next to the beam are shown.

Figure 14. Voltages developed between the end-plate facing the beam and the outermost detonator cable conductor vs. time. Measurements with the test device in the beam and with it out of but next to the beam are shown. X-ray flux was 18.7 rad/s at the sample.

Figure 15. Voltages developed between the end-plate facing the beam and the innermost detonator cable conductor vs. time. Measurements with the test device in the beam and with it out of but next to the beam are shown. X-ray flux was 18.7 rad/s at the sample.

Figure 16. Voltages developed between the end-plate facing the beam and the innermost end-plate vs. time. Measurements with the test device in the beam and with it out of but next to the beam are shown. The dose rate was 18.7 rad/s at the sample. The voltage decay with the beam turned off indicates the leakage resistance of the HE.

Measured voltages reached an equilibrium condition quickly for high fluxes, and more slowly for lower fluxes as in the out-of-beam cases. This phenomenon reflects the change of RC time constant in the explosive due to the RIC reduction in resistance.

X Conclusions

The following conclusions may be drawn from these analyses and experiments:

- The RIC effect limits the maximum charge accumulation in X-irradiated weapons for fluences greater than a few rads.
- The accumulated equilibrium charge and voltage are independent of the dose rate (flux).
- The accumulated charge and voltage are independent of the pulse structure of the X-ray machine.
- Since the charges are dissipated during the X-ray pulse, the total dose (fluence) does not affect charge accumulation and waiting periods are unnecessary.
- During radiography operations the charges separated in the HE and in the detonator cable-case capacitor are insufficient to fire detonators by many orders of magnitude.

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