AD NUMBER

AD355669

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Oct 1976, Group 4, DoDD 5200.10, 26 July 1962; AFATL ltr 13 Dec 1977

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ATL-TDR-64-52

(Unclassified Title)
RADIATION EFFECTS ON EXPLOSIVES

(Fourth Quarterly Technical Report)
Ending August 1964

Technical Documentary Report No. ATL-TDR-64-52
October 1964
Project 2511
Task 251102

Directorate of Armament Development
Det 4, Research and Technology Division
Air Force Systems Command
Eglin Air Force Base, Florida

(Prepared under Contract No. AF 08(635)-3192 by TEMPO, General Electric Company, Santa Barbara, California, John A. Berberet, author)

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ABSTRACT

The dimensional changes and loss of weight resulting when selected conventional and fluoro-explosives are exposed to doses of Co^{60} gamma irradiation up to 2×10^9 rad have been observed. The two heterocyclic compounds which were used in these studies, RDX and HMX, behaved similarly and exhibited the greatest dimensional and weight loss changes of all the materials studied. The conventional aromatic compounds, TNT, Tetryl, TNB and DATB, exhibited similar behavior, which was significantly different from that of the two heterocyclic compounds. The radiation stability of these compounds was also significantly greater than that for the heterocyclic compounds.

As a group, the deformation of the fluoro-derivatives followed no similar pattern. Their weight-loss stability was significantly better than that of the heterocyclic compounds, but was exceeded by the aromatic compounds.

Conventional theories of the effect of gamma radiation on the dimensional stability of materials of this type are inadequate to explain the radiation-induced dimensional changes which were observed.

PUBLICATION REVIEW

THIS TECHNICAL DOCUMENTARY REPORT HAS BEEN REVIEWED AND IS APPROVED.

for DAVID K. DEAN
COLONEL, USAF
CHIEF, WEAPONS DIVISION

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INTRODUCTION

When quasi-stable materials in the solid state are exposed to nuclear radiations, macroscopic changes such as dimensional and mass changes (accompanied by the release of gases and vapors), take place. Some materials change color, exhibit microscopic and macroscopic cracking, and change in surface texture. The basic causes of such radiation effects are poorly understood. Likewise, the dependence of these and other radiation-induced phenomena on total dose, dose rates, and environmental conditions, e.g., gaseous environment and temperature, are not known.

When nuclear radiation interacts with these materials, the energy of the radiation deposited in the materials is expended initially either in the form of electron perturbation phenomena, or in the form of atomic displacements. Although of the order of 99 percent of the energy is expended in the form of electron perturbation, there are few bases for associating any of these radiation effects with either of the basic phenomena.

One of the main objectives of the current study is to learn more about the relationship of the observed radiation effects to the basic primary effects. The approach to this problem is simply one of using radiation for which there is a large variation in calculated β-values*, the number of atomic displacements produced per 100 ev of energy absorbed in the material. With all other conditions being the same, it is plausible that those phenomena having their origin in electron perturbations would be little affected, whereas those having a dependence upon displacement phenomena would be grossly affected.

It must be acknowledged that such an approach in a sense might be naive in that some effects may not bear a simple relationship to the two primary phenomena, or they may be complex functions of both of these

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* This is a newly coined term and is chosen to be analogous with the commonly used G-value, a term denoting the number of molecular phenomena of a specified type produced per 100 ev of absorbed energy.

radiation phenomena plus other factors, such as the environmental conditions. Nevertheless, the extremely small amount of work that has been done on basic radiation phenomena in materials of this type, plus the experimental complexities that are encountered in working with them, lend support to the rationality of such an approach at this time.

Three electron radiation sources have been selected for these studies. The first, consisting primarily of Compton electrons (of average energy of the order of 0.5 Mev) from an intense Co^{60} radiation source; the second, 3 Mev electrons from a Van de Graaff accelerator, and lastly, 40 Mev electrons from a Linac accelerator. The displacement cross-section for this range of electron energies varies over several orders of magnitude, and hence there is a good possibility of establishing a correlation between some of the radiation effects and basic phenomena through the use of such sources.

This report covers a portion of the observations made of the effects of radiation on selected material sources, a Co^{60} source having a maximum intensity of about 10^7 rads/hour. The materials studied included six conventional explosives and the fluoroderivatives of four conventional explosive materials.

SECTION I

OBJECTIVES

(U) The immediate objectives of the subject study were to determine the effect of Co^{60} radiation upon the following properties of quasi-stable materials for very high doses:*

1) dimensional stability;

2) loss of weight;

3) color;

4) physical texture;

5) electrical resistivity.

(U) Furthermore, it was desired to obtain some understanding of how the changes in these properties depended upon dose rates as well as total dose. This report covers only those observations made on dimensional stability and weight loss.

(C) The materials studied were four aromatic compounds:

1. TNT (trinitrotoluene)

2. TNB (trinitrobenzene)

3. DATB (diaminotrinitrobenzene)

4. Tetryl (trinitrophenylethylnitramine)

---

* Prior to this study, almost no radiation effects studies with these materials had been done with doses greater than 3×10^8 rad. In this study, doses were taken to levels as high as 2×10^9 rad. The maximum dose rate of the Co^{60} source used in this study was also an order of magnitude greater than that used in any previous studies.

two heterocyclic compounds:

1. β HMX (cyclotetramethylene tetranitramine)
2. RDX (cyclotrimethylenetrinitramine)

and four fluoro-derivatives:

1. TNTF (trinitrobenzotrifluoride)
2. MFTNB (monofluorotrinitrobenzene)
3. DFTNB (difluorotrinitrobenzene)
4. TFEt Tetryl (trinitrophenyltrifluoroethylnitramine)

SECTION II
EXPERIMENTAL PROCEDURE

(U) All materials were irradiated in the form of small cylindrical pellets having dimensions of approximately 1/4-inch diameter by 1/2-inch long. The pellets were made from pure powders pressed at room temperatures with a pressure of about 15,000 psi; no binders were used. The conventional materials were formed in a single pressing operation. The fluoro-derivatives had to be pressed in about five successive steps and as a consequence exhibited circumferential striations about 2 or 3 mm apart. Figure 1 shows an array of samples of the ten different materials. These specific samples were used as "standards" of color and physical texture of the various materials.

(U) When irradiated, the samples were first inserted into ampoules, as shown in Figure 2, consisting of pyrex tubing fitted with ceramic Alsimag 222 plugs at each end. The ampoules in turn were inserted into a stainless steel capsule as shown in Figure 3. This capsule, which had walls of 0.3 inch thickness, served primarily as a containment vessel to ensure that no hazards would result in the event that ignition of one of the materials took place during irradiation. The capsule had been tested to contain a detonation produced by a gram of PETN.

(U) Insertion of the sample into the ampoule, and the ampoule into the capsule was done in a helium atmosphere. Thus, although the sample was exposed to a helium atmosphere at the start of the irradiation, the helium was somewhat diluted by gaseous decomposition products during the course of the irradiation. It is unlikely that this dilution had any significant effect on the sample, for if it had, color changes at the surface of the sample would probably have been different from those produced in the interior. No such differences were noted.

(U) An underwater Co^{60} facility at the Vallecitos Laboratories of the General Electric Company near Pleasanton, California, was used as the irradiation source. The source consisted of eight cobalt rods (Co^{60} canned in stainless steel) 6-inches long, which were clustered about a central irradiation tube (an aluminum tube of 1.25 inches

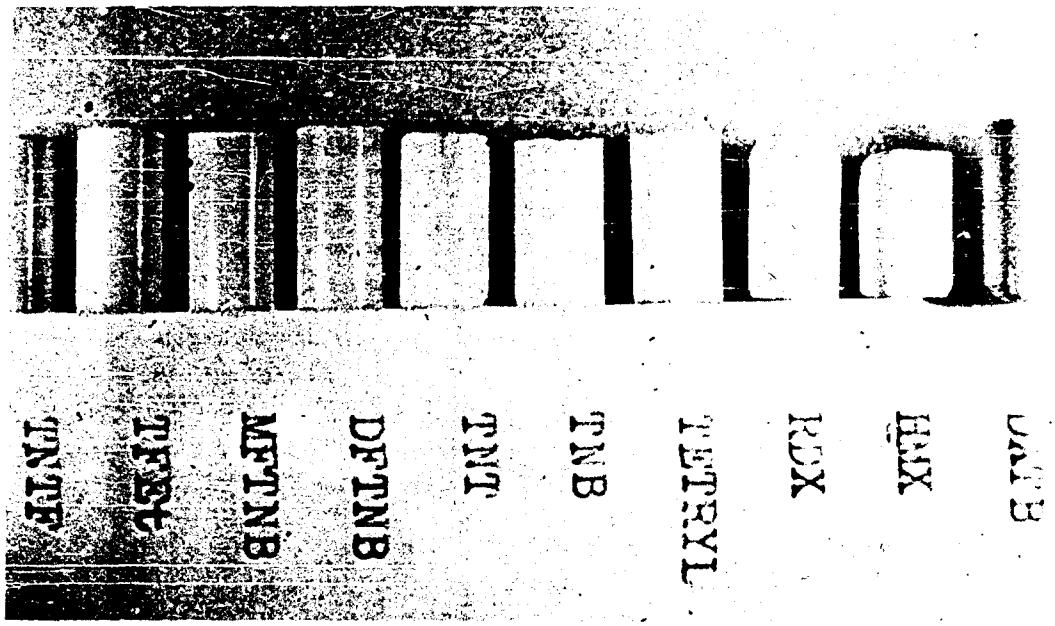

Figure 1. Standard samples of materials exposed to radiation.

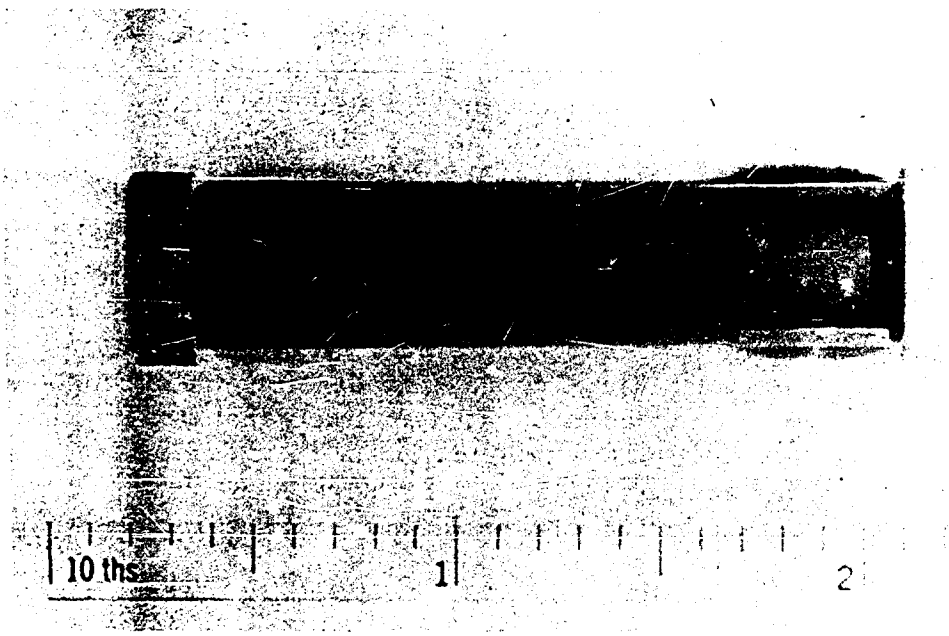

Figure 2. Irradiation ampoule.

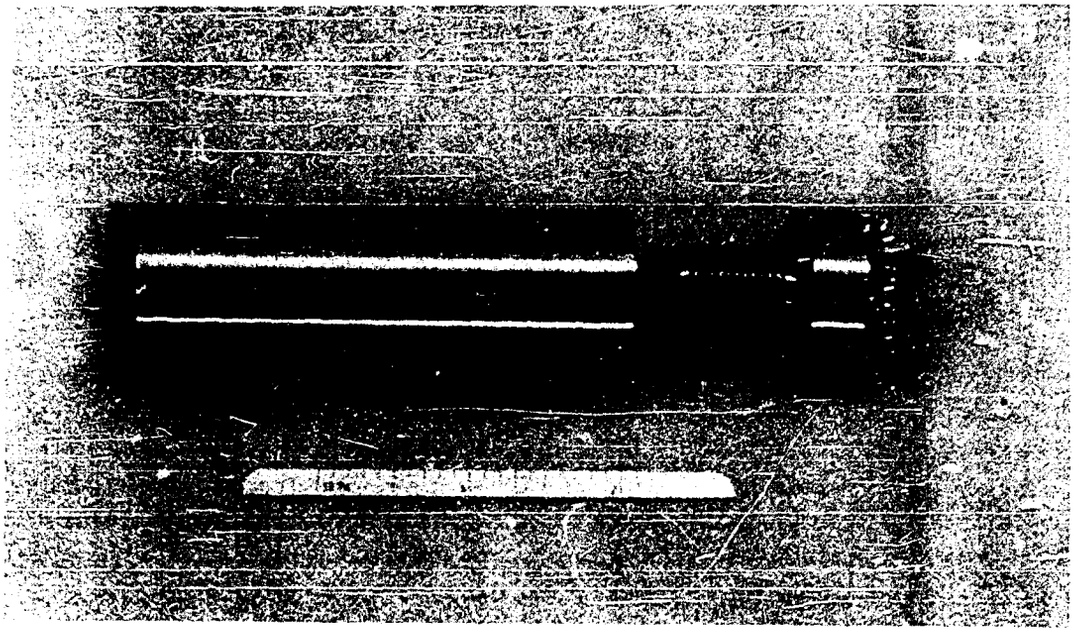

Figure 3. Irradiation capsule.

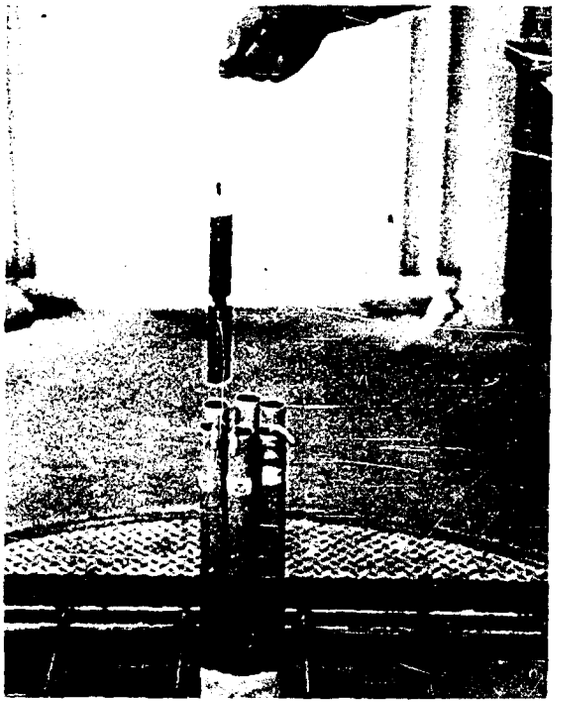

Figure 4.
Samples being charged into irradiation tube.

O.D. and 1/16-inch wall thickness). Six additional aluminum irradiation tubes of the same dimensions were clustered about the cobalt rods and central source tube. This arrangement thus gave seven air-filled irradiation tubes surrounded by water in which two samples could be irradiated in each tube simultaneously. Figure 4 shows the tube arrangement and two samples connected in a tandem manner just prior to being lowered into the irradiation position. (The central tube, not visible in this picture, is surrounded by the six tubes that can be seen.)

(U) Radiation intensities were determined by the ferric-ferrous dosimetry method. The level at the sample position in the central tube was determined to be 9×10^6 rad/hour, and that in the outer six tubes to be 2.3×10^6 rad/hour. The uncertainty in this radiation intensity was estimated to be about ± 20 percent. With the arrangement employed, it was possible to irradiate two samples at the high exposure level, and twelve samples at the low exposure level.

(U) Observations on a specific sample were made periodically during an irradiation. In this procedure, the sample was removed from its irradiation position, weighed, measured, photographed (microscopically as well as macroscopically), encapsulated in a helium atmosphere and returned to its former radiation environment.

(U) For the resistivity studies, the results of which will be reported at a later date, a separate set of samples was used on which only resistivity measurements were made. As with the other samples, these measurements were made periodically throughout an irradiation period so that it would be possible to determine how the resistivity varied as a function of dose.

SECTION III

OBSERVATIONS OF DIMENSIONAL CHANGES

(U) Measurements of the length of samples were made with a microscope having a micrometer attachment; with this system it was possible to make measurements with an uncertainty of less than 2×10^{-4} cm. The largest source of error in this type of measurement was the contamination of the sample surfaces with minute foreign particles.

AXIAL DEFORMATION OF HETEROCYCLIC COMPOUNDS

(U) The axial deformation of two samples of HMX, the dose rate for one being 2.3×10^6 rad/hour, and 9.6×10^5 rad/hour for the second, are shown in Figure 5. The significant aspects of the two curves of Figure 5 are:

1. Both samples first expand, then contract.
2. Although both samples reach their maximum at about the same dose, the maximum for the higher dose rate is about half that for the lower dose rate.

(U) Irradiations of RDX were also made; however, this material lost its physical integrity (crumbled) at such low doses that it was not possible to make any comprehensive studies of the axial deformation. The following two observations with two different samples were made:

Dose Rate (rad/hour)	Dose (rads)	Change in Length (Percent)
2.3×10^6	2×10^7	2.7
2.3×10^6	3.6×10^7	7

(U) When compared with the curve for HMX for the same exposure rate as given in Figure 5, it is seen that the radiation induced axial deformation for these two materials is approximately the same at these exposure levels.

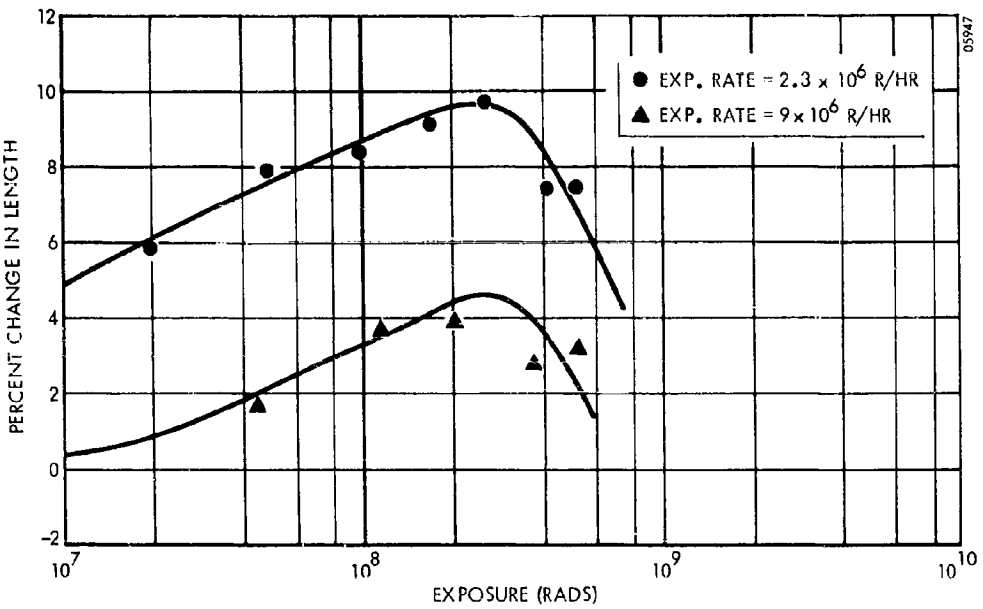

Figure 5. Axial deformation of HMX samples exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)

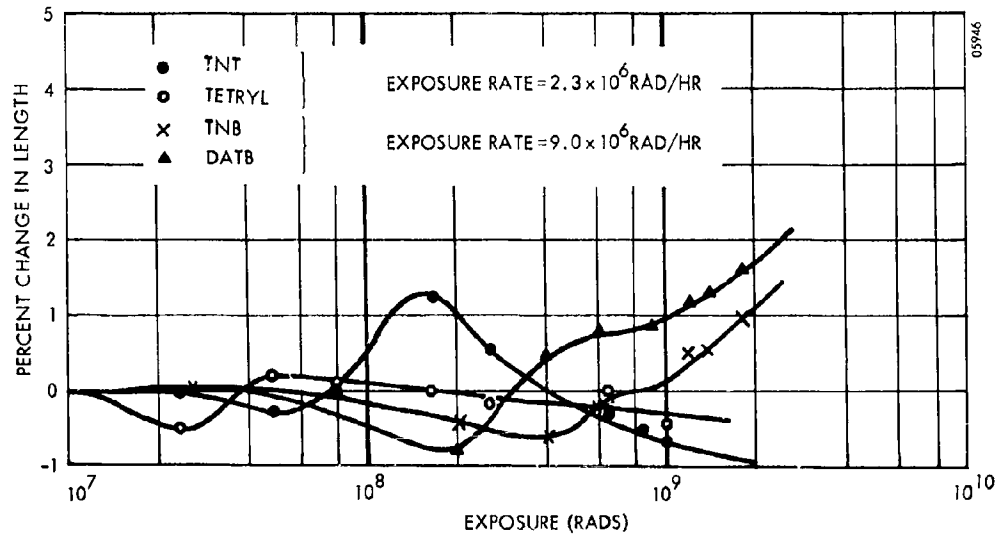

Figure 6. Axial deformation of aromatic compounds exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)

AXIAL DEFORMATION OF AROMATIC COMPOUNDS

(U) The axial deformation of TNT, Tetryl*, TNB, and DATB is shown in Figure 6. The dose rate for TNT and Tetryl was 2.3×10^6 rad/hour, while that for TNB and DATB is 9×10^6 rad/hour.

(U) In contrast with the observations with the heterocyclic compounds, the axial deformation of these materials is first negative, and then becomes positive. A maximum deformation is observed for TNT and Tetryl, while no maximum is observed for TNB and DATB, even for exposures of almost 2×10^9 rads. It is not known whether this absence of a maximum is due to the higher exposure rate, or whether it is characteristic of the materials.

(U) The deformation of these four materials is much less than that of the two heterocyclic compounds. This relative stability is in keeping with the observations of other investigators.

AXIAL DEFORMATION OF THE FLUORODERIVATIVES

(C) The axial deformation of the four fluoroderivatives, TNTF, TFEt Tetryl, MFTNB, and DFTNB, are shown in Figures 7 through 10, inclusive, for two dose rate intensities. The data for TNTF and TFEt Tetryl is similar to that for the heterocyclic compounds, although the magnitude of the dimensional change is much less, especially for TFEt Tetryl. However, in contrast to the observations for HMX, there is no evidence of a strong dependence upon dose rate for any of these four materials.

(C) The completely "negative deformation" or contraction, exhibited by MFTNB, Figure 9, both for exposure rates and for doses up to 10^9 rads, marks the radiation induced behavior of this material as being ostensibly different from that of any of the other materials. Likewise, the behavior pattern of DFTNB shown in Figure 10 is different from the observations made for other materials in that no maxima or minima are observed. Neglecting the zero slope portion of the curve centering about the dose of 2×10^8 rads, it is seen that the sample of DFTNB monotonically expands. (It is believed that this zero slope section is a measurement anomaly, and is probably not characteristic of the true behavior of the material.)

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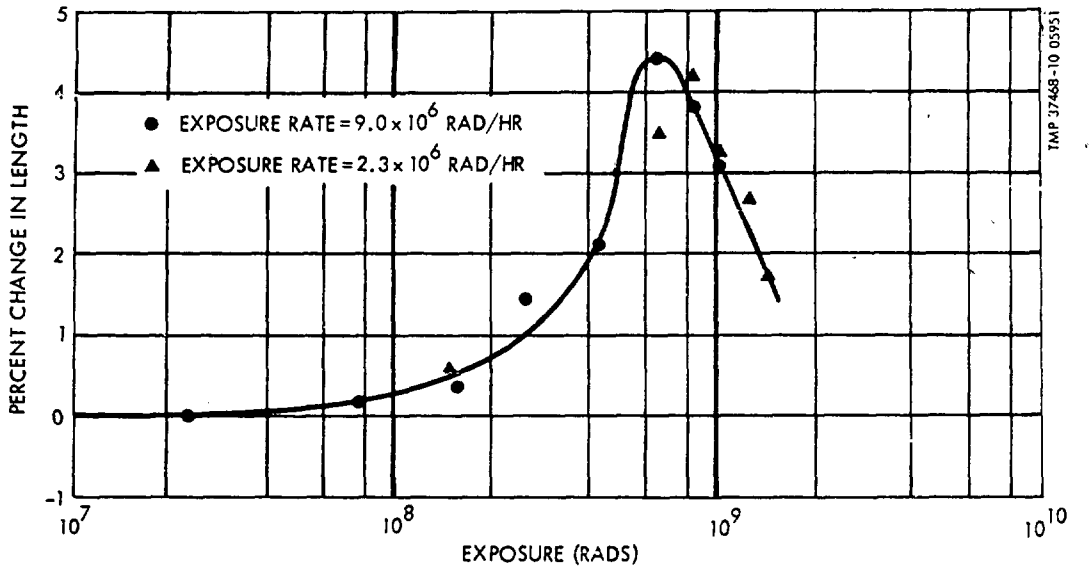
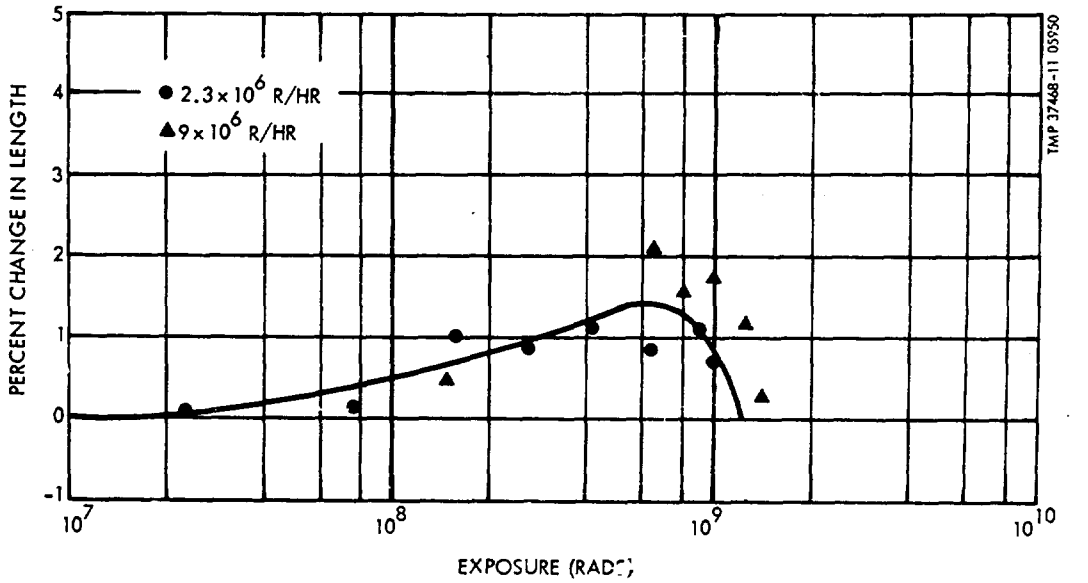
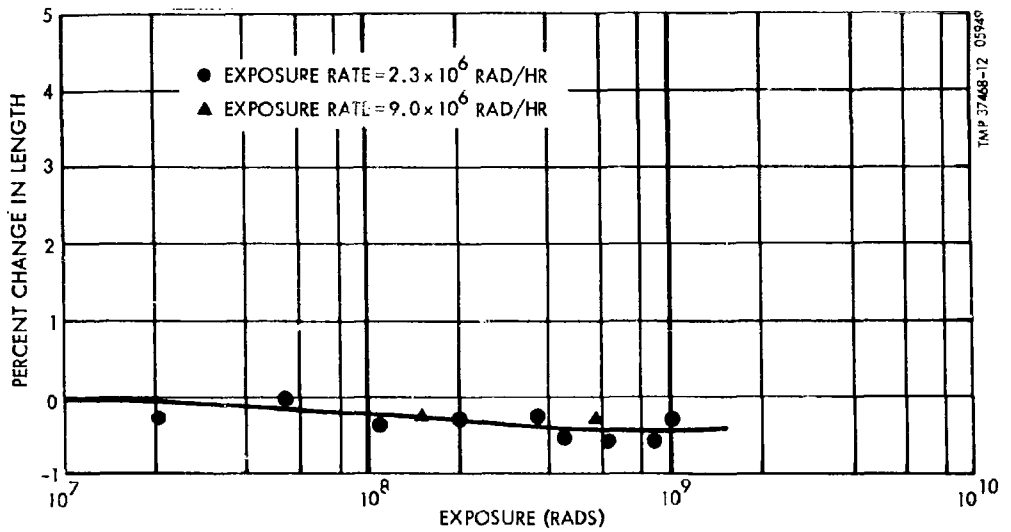
* Although included with the aromatic compounds, Tetryl is actually only "partially" aromatic.



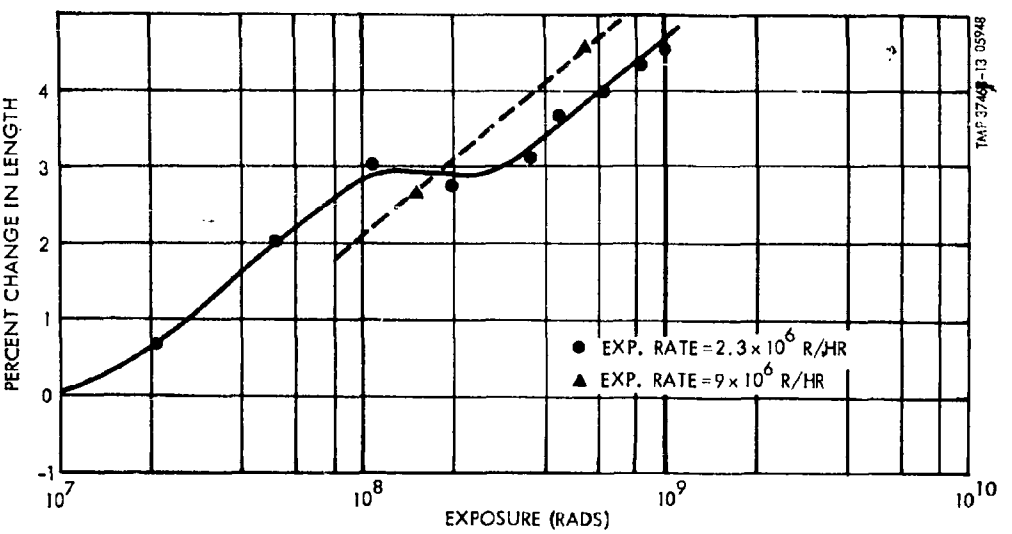
(C) Figure 7. Axial deformation of TNTF samples exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)



(C) Figure 8. Axial deformation of TFEt Tetryl samples exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)



(C) Figure 9. Axial deformation of MFTNB samples exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5 long)



(C) Figure 10. Axial deformation of DFTNB samples exposed to Co^{60} gamma rays. (sample size: ~0.25" diam.)

SECTION IV

RADIATION INDUCED WEIGHT LOSS

(U) The loss of weight as a function of dose for the materials studied, excepting RDX, is shown in Figures 11through 15. When decomposition is induced by radiation, at least a portion, or possibly all, of the decomposition products are gaseous and more or less gradually diffuse from the sample. Non-volatile decomposition products will remain with the host material, where they may remain in an inert state, or may react with the host material, depending upon both the nature of the decomposition products and the host material. Likewise, it has also been shown from other investigations that gaseous radiation-induced decomposition products continue to diffuse from the samples following an irradiation.

(U) Thus, the weight loss data given in Figures 11 through 15 represent only those decomposition products which escaped from the sample from the start to the end of the irradiation period. In no case was a significant dependence of weight loss on dose rate demonstrated. This independence of dose rate suggests that the time for the gas to diffuse from the sample may have been small compared to the total period of irradiation.

(U) The weight loss for a given dose was greatest for HMX, and the least for DATB. As with the dimensional measurements for RDX, it was not possible to make more than one or two measurements of the loss of weight of RDX. In one of these measurements (for an exposure rate of 2.3×10^6 rad/hour), a weight loss of 1.2 percent was observed for an exposure of 4.6×10^7 rads. From Figure 11 it can be seen that this weight loss compares closely with that for HMX.

(C) In comparing the fluoro-derivatives with each other (Figures 14 and 15), it is seen that MFTNB exhibits the least weight loss, and TFEt Tetryl the most. At an exposure of 10^8 rads, the weight loss of MFTNB is very small and compares favorably with DATB.

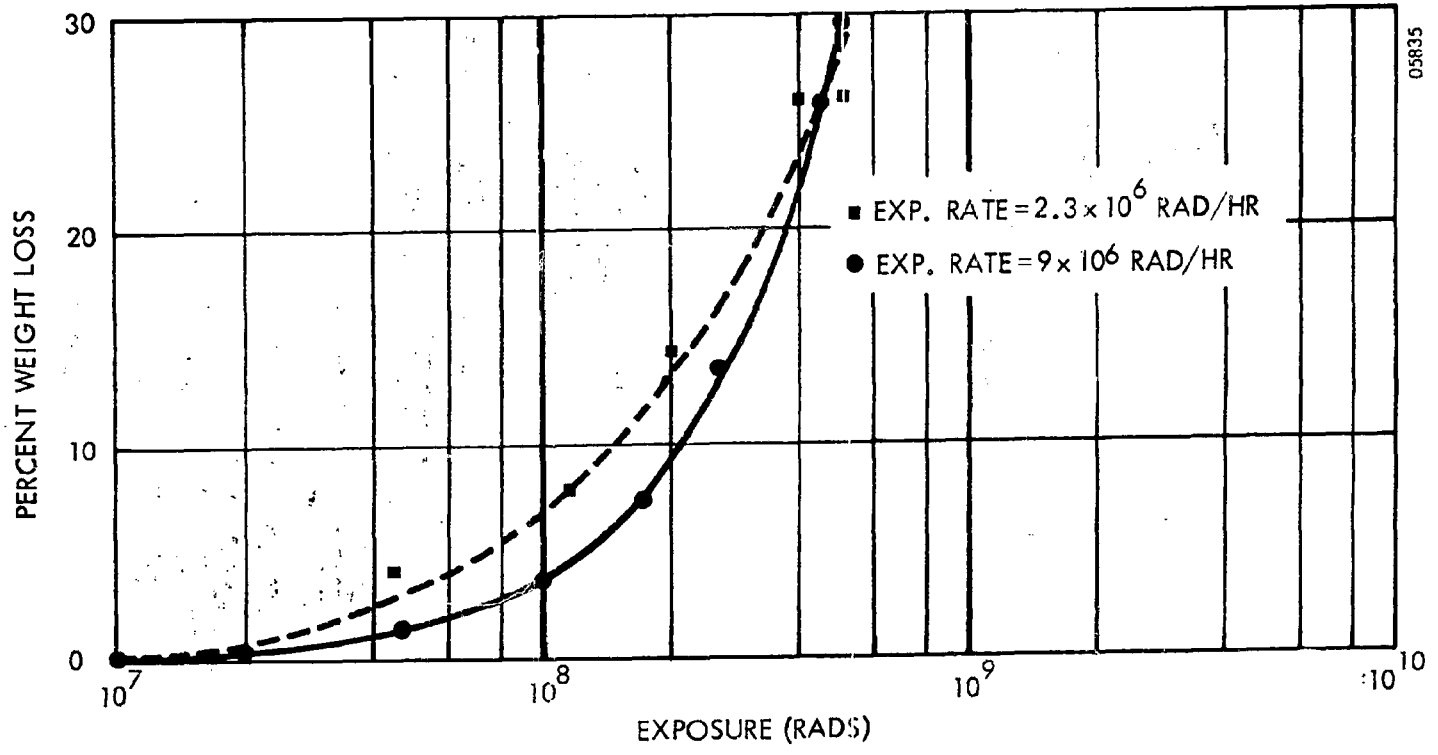
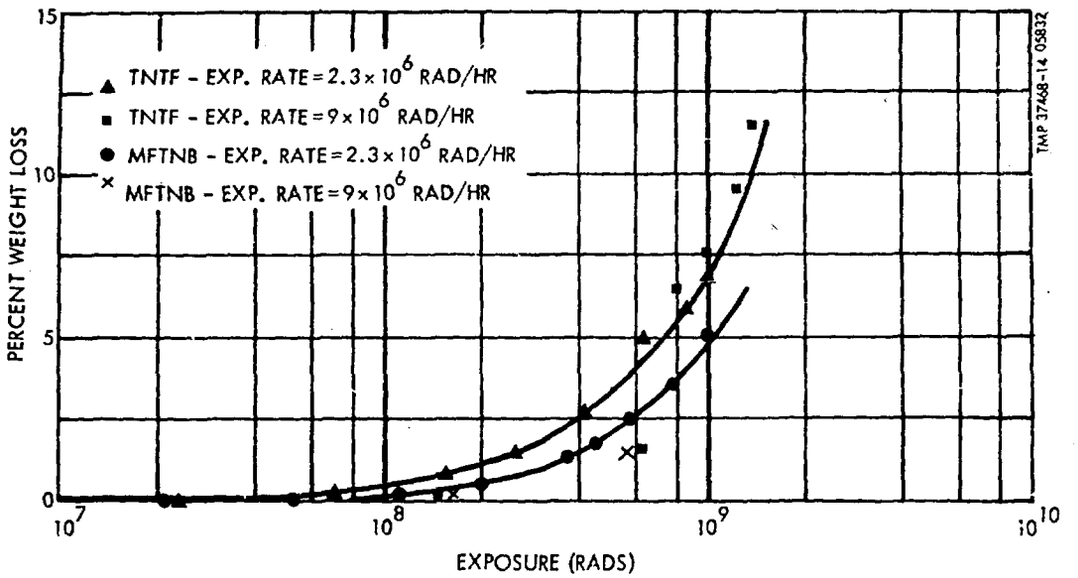
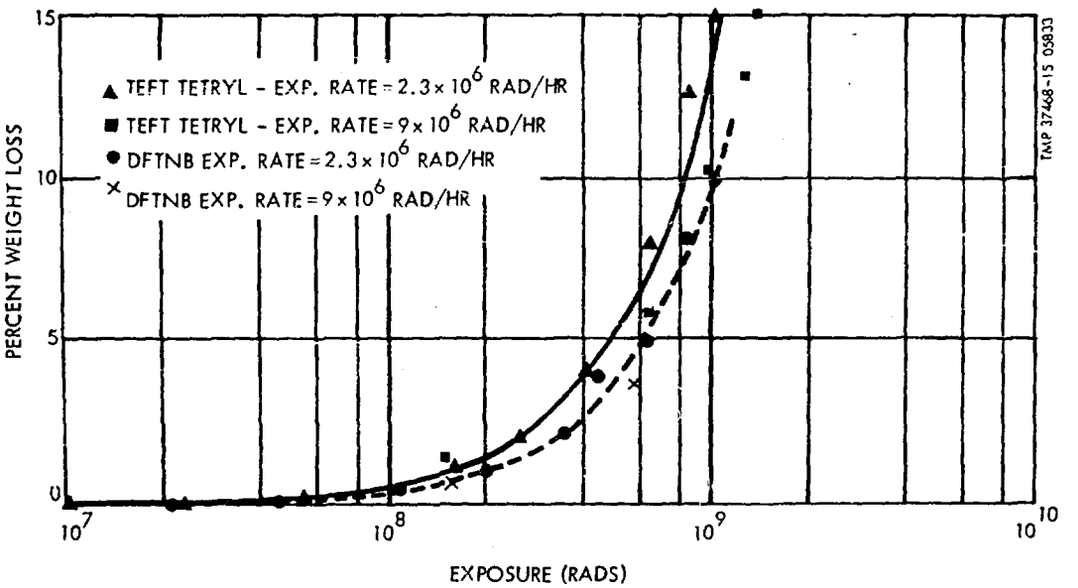


Figure 11. Weight loss of HMX samples exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)



(C) Figure 14. Weight loss to TNTF and MFTNB exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)



(C) Figure 15. Weight loss of TFEt Tetryl and DFTNB exposed to Co^{60} gamma rays. (sample size: ~0.25" diam. x 0.5" long)

SECTION V
CONCLUSIONS

(U) No attempt will be made to state any conclusions concerning the basic mechanisms responsible for the physical deformation until the data from the three series of irradiations has been put into final form and analyzed. However, some conjectures concerning the nature of the deformation process are in order at this time.

(U) The data presented here differ from that presented by other observers in that:

1. In the aromatic compounds a contraction was observed to take place prior to the expansion

2. Many of the materials studied show that a rapid contraction takes place following an expansion.

(U) These phenomena suggest that two basic processes are taking place. One of these processes tends to cause an expansion, and the other, a contraction. It has been hypothesized that the expansion, thought to be a type of plastic deformation, is produced by internal pressures within the sample. These pressures are believed to result from microscopic pockets of gases formed as a consequence of the irradiation induced decomposition. Eventually these pressures become so great that minute fissures to the surface of the sample are produced and the gases escape. Where the number of these fissures or leakage paths become large enough, the internal pressures are diminished and the sample begins to contract.

(C) This theory, however, is inadequate to explain the initial contraction of the aromatic compounds and the continuous contraction of MFTNB.

(U) It is possible that the radiation is having the effect of causing a crystalline phase changes, and, as a consequence, the sample contracts. However, such possibilities are very hypothetical, and it can only be hoped that forthcoming data will give further insight into these processes.

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DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY *(Corporate author)*
TEMPO, General Electric Company
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2a. REPORT SECURITY CLASSIFICATION
Confidential

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4

3. REPORT TITLE
(U) RADIATION EFFECTS ON EXPLOSIVES

4. DESCRIPTIVE NOTES *(Type of report and inclusive dates)*
Fourth Quarterly Technical Report ending August 1964

5. AUTHOR(S) *(Last name, first name, initial)*
Berberet, John A.

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
October 1964	28	

8a. CONTRACT OR GRANT NO.
AF 08(635)-3192

b. PROJECT NO.
2511

Task No.
251102

d.

9a. ORIGINATOR'S REPORT NUMBER(S)
ATL-TDR-64-52

9b. OTHER REPORT NO(S) *(Any other numbers that may be assigned this report)*

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11. SUPPLEMENTARY NOTES

12. SPONSORING MILITARY ACTIVITY
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13. ABSTRACT

(U) The dimensional changes and loss of weight resulting when selected conventional and fluoro-explosives are exposed to doses of Co^{60} gamma irradiation up to 2×10^9 rad have been observed. The two heterocyclic compounds which were used in these studies, RDX and HMX, behaved similarly and exhibited the greatest dimensional and weight loss changes of all the materials studied. The conventional aromatic compounds, TNT, Tetryl, TNB and DATB, exhibited similar behavior, which was significantly different from that of the two heterocyclic compounds. The radiation stability of these compounds was also significantly greater than that for the heterocyclic compounds. As a group, the deformation of the fluoro-derivatives followed no similar pattern. Their weight-loss stability was significantly better than that of the heterocyclic compounds, but was exceeded by the aromatic compounds. Conventional theories of the effect of gamma radiation on the dimensional stability of materials of this type are inadequate to explain the radiation-induced dimensional changes which were observed.

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