

WADC TECHNICAL REPORT 57-266

PART I.

ASTIA DOCUMENT No. AD 130807

# **EFFECTS OF NUCLEAR RADIATION ON ORGANIC FLUIDS**

## **PART I. GAMMA RADIATION STABILITY OF CERTAIN MINERAL OILS AND DIESTER FLUIDS**

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## FOREWORD

This report was prepared by the Organic Materials Branch, Materials Laboratory, Directorate of Research, Wright Air Development Center, with 1st Lt. William L. R. Rice acting as Project Engineer. It was written in cooperation with 1st Lt. James H. Way, Fuel and Oil Branch, Power Plant Laboratory, Directorate of Laboratories, Wright Air Development Center. It is the first part of a series of reports concerning the investigation of the effects of nuclear radiation on organic fluids and lubricants. This project was initiated under Project No. 2133, "Radiation Effects," Task No. 73071, "Radiation Effects on Materials."

All irradiations, both at WADC and in other sources, were conducted under the supervision of Lt. Robert H. Johnson, Analysis and Measurements Branch, Materials Laboratory, Directorate of Research, Wright Air Development Center.

This report covers work conducted during the period January 1956 to March 1957.

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## ABSTRACT

A study was made of the stability to gamma radiation of certain refined petroleum oils and diester fluids. Testing of the irradiated fluids by means of lubricant evaluation procedures, such as viscosity, flash point, and oxidation-corrosion resistance, indicated that for the tests conducted, the mineral oils of the type studied should have general resistance to gamma radiation up to a dosage of about  $1 \times 10^8$  roentgens. The diester fluid di-2-ethylhexyl sebacate (Flexol 201) had very poor stability to gamma radiation over the same range. Addition of 0.5% phenothiazine improved the radiation resistance of this base fluid, except for the loss of oxidative stability experienced at dosage levels of  $1 \times 10^7$  roentgens and lower.

Examination was made of the test data for the irradiated fluids to determine if any property changes followed the radiation exposure history. This was to discover if either the mineral oil or diester type fluids could be used as secondary reference standards for radiation calibration of non-uniform geometries exposed to nuclear radiation, such as bearing assemblies or circulating fluid loops. The changes offering the most promise were the increase in neutralization number of the diester fluid and the gas evolution of the mineral oils.

## PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:



R. T. SCHWARTZ  
Chief, Organic Materials Branch  
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## I. INTRODUCTION

A. Background

Although organic compounds have relatively poor stability to nuclear radiation, their use in a radiation environment is not necessarily precluded. Where low radiation levels are encountered, it is preferable to use an organic liquid if it has engineering properties superior to a substitute material of greater radiation resistance. The limit of use is that point at which radiation induced breakdown changes the fluid properties to such an extent that it no longer performs its necessary functions. This breakdown limit is used in defining the term "damage threshold," which is an arbitrary term of changing definition. For instance, if viscosity is the only critical property, then the change in viscosity determines the allowable maximum exposure. For complex organic systems, such as lubricants, definition of radiation limit is not quite so simple. It is generally necessary to perform the maximum number of lubricant tests to determine the limits of usability. Obviously, the true criterion of a lubricant is whether it will give satisfactory performance in the end item application, be it a swivel chair caster or a turbojet engine. This is impractical in many instances, due to the expense of testing or time required. For radiation evaluation, the additional limitation is imposed of lack of irradiation space. Irradiating quantities in excess of a gallon to any appreciable dosage requires sizeable source capacity and often results in delay of other scheduled irradiations. The compromise usually adopted is to conduct laboratory tests on relatively small volumes of irradiated fluid samples. The test program reported herein was conducted on such a basis.

Associated with any test involving nuclear radiation is the problem of determining the amount of radiation energy received by the sample. This is particularly important in dynamic tests in the presence of nuclear radiation. An example of the magnitude of such a determination would be the use of a bearing rig adjacent to a radiation source. The non-uniform geometry of the metal components makes it almost impossible to calculate the dosage a test fluid received, since self shielding varies according to location in the rig and the fluid would not receive the same dose at all locations during the test. A possible solution to this problem is to consider the property changes in the fluid itself and relate such changes to a standard radiation dosage. This

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would entail running the rig under no-load conditions to eliminate the effects of temperature, shear breakdown, and related test effects. When run for the same time period as the standard test, this no-load test would expose the experimental oil to the same radiation dosage. Then by comparing the property changes of the irradiated oil to those of the same oil after irradiation to varying dosages in a standard and well-calibrated source, it would be possible to estimate the dosage received during the test. This sort of "secondary reference standard" is attractive for organic liquids because the circulation of the oil during the test serves to integrate the dosage received and give bulk property changes commensurate with those of an irradiation in a well-calibrated source. An organic would be more attractive than a water base reference fluid because it would not corrode the test apparatus and would be similar in nature to the experimental oils used. To this end, a typical mineral oil and diester base stock were irradiated from  $5 \times 10^6$  roentgens up to  $1 \times 10^8$  roentgens and evaluated to see if any of the typical laboratory tests could be related to the dosage received.

This report is concerned with a preliminary survey of the stability to gamma radiation of certain refined petroleum oils and diester fluids. The petroleum oils are typical aircraft reciprocating engine lubricants as defined by Specification MIL-L-6082. The diester fluids evaluated are typical of the synthetics used in aircraft turbine engine lubricating oils. Data are reported for a blend of di-2-ethylhexyl sebacate (Plexol 201) with 0.5% phenothiazine, to show the effect of an inhibitor on the radiation resistance of a diester fluid.

The secondary objective of this report was to determine the stability of diesters and mineral oils to gamma radiation to see if any property change in either type of fluid could be related to the dosage received so that the fluid could be used to calibrate non-uniform irradiation volumes.

## B. Radiation Environment

The test oils were irradiated in one of two sources. One source is the spent fuel elements canal of the Materials Testing Reactor, Arco, Idaho. This facility consists of MIR fuel elements arranged around a test rack, with canisters placed in the rack for irradiation of the samples. Ambient temperature is about 75°F. Quart samples of the oils were placed in the stainless steel canisters, which have a one-eighth inch wall thickness, and irradiated to the specified dosage. The dose rate



for this facility is a function of the age of the fuel elements in the rack, varying from  $5 \times 10^5$  r/hr to  $3 \times 10^7$  r/hr. The average gamma energy is estimated to be in the range 200 Kev to 2 Mev, the importance of intermediate energy groups varying according to the age of the fuel element. Calibration is made by the ceric-cerous technique, with routine checks taken using ion chambers. Dose values for this facility are reported on the basis of the unperturbed irradiation volume, that is, before the canister is placed in-source. An estimate of the accuracy of the dosimetry indicates a possible error of plus or minus ten per cent. Perturbation error due to the insertion of the metal canister has not been calculated, but has been estimated to be about an additional twenty per cent.

The other source used was the 1500 curie  $\text{Co}^{60}$  pipe at the Wright Air Development Center. Samples of about 400 ml were irradiated in the source in glass ampoules at an ambient temperature of about 75°F. The dose rate was about  $3.8 \times 10^5$  r/hr, with an average gamma energy of 1.25 Mev. Dosimetry was by the ferrous-ferric technique and by means of ion chambers. Recent measurements were made using Sigoloff dosimeters. The estimated accuracy of dosimetry is plus or minus ten per cent. Because of the small sample size and the use of glass containers, the dosage values reported for this source are probably quite close to the dosage received by the sample.

The unit of radiation used in this report is the roentgen, defined as follows:

roentgen: That quantity of X- or gamma radiation which will produce, as a consequence of ionization, one electrostatic unit of electricity, of either sign, in 1 cc of dry air, as measured at 0°C and standard atmospheric pressure. One roentgen is equivalent to the absorption of 87.1 ergs/gm C (based on 34.0 Ev/ion pair), 87.7 ergs per gm of air, 95.8 ergs/gram of tissue, or 96.7 ergs/gm of water.

## II. RESULTS AND DISCUSSION OF RESULTS

### A. Test Procedures

The test results for the irradiated oils are given in Tables I through VI and Figures 1 through 4. All tests were conducted in accordance with the procedures described in Federal Test Method Standard Number 791 (3).



## B. Radiation Stability of Mineral Oils and Diester Fluids

The stability of two grades of mineral oil was investigated after exposure in the MTR gamma canal, the data being shown in Table I. At  $1 \times 10^8$  roentgens an increase occurred in the 100°F viscosity for both the 1065 and 1100 mineral oil. Neutralization number showed an increase for the 1100 oil but not for the 1065 oil. The panel coke value went up somewhat for the lighter 1065 oil but showed a decrease in the case of the 1100 fluid. Other properties, such as pour point, flash point, carbon residue, and copper strip corrosion showed little change.

A more thorough study was made of mineral oil stability by subjecting a 1065 oil to increasing dosage levels in the WADC Co<sup>60</sup> source. The test data are shown in Table II. No property change was observed with increasing radiation dosage. Up to the upper level of  $1 \times 10^8$  roentgens there were so few variations in the fluid properties that it can be concluded that for the tests conducted, the oil was stable to  $1 \times 10^8$  roentgens.

Table III shows the results of the testing of several diesters after irradiation at the MTR gamma canal. The di-2-ethylhexyl adipate, azelate, and sebacate esters all showed the same general change in properties. The viscosity at 210°F is relatively unaffected, but the viscosity determined at lower temperatures, such as -65°F, indicates a more marked susceptibility to change with increasing radiation dosage. This change in the low temperature viscosity is typical, and indicates a loss in low temperature properties as a result of irradiation. The pour point was below -75°F for all control and irradiated fluids. The flash point reduction is typical of data obtained for diester fluids, ranging from 35°F to 75°F, depending on the fluid. Evaporation was essentially unchanged, while neutralization number increased greatly with irradiation. Coke values showed little change for the sebacate, but increased somewhat for the adipate and azelate. Rubber swell showed an increase with dosage. Foaming was acceptable.

Table IV gives the test data for a commercial di-2-ethylhexyl sebacate, Flexol 201, irradiated in the WADC Co<sup>60</sup> source. Viscosity shows little change up to  $1 \times 10^8$  roentgens for the higher temperatures, but does change somewhat for the lower temperature viscosities. As can be seen, specific gravity, density at 130°F, and refractive index show little change up to  $1 \times 10^8$  roentgens. Flash point again decreases, dropping from 415°F to a value of 255°F, a change of 160°F. Fire point did not drop as much, showing a maximum change of 55°F. Autogenous ignition

TABLE I  
EFFECTS OF FUEL ELEMENT GAMMA RADIATION ON REFINED MIL-L-6082 PETROLEUM OILS

Dosage (Roentgens)	Grade 1065				Grade 1100				
	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>
Viscosity (centistokes) 210°F 100°F	12.33 119.8	12.74 123.6	13.50 135.8	20.17 263.7	19.82 259.0	20.97 280.5			
	-5	-5	-5	-5	-5	-5			
Pour Point (°F)	455	450	445	495	490	485			
Flash Point (°F)	0.01	0.01	0.02	0.01	0.45	0.75			
Neutralization Number (mg KOH/gm oil)	128	166	164	112	125	50			
Coking at 600°F (mg) (1)	0.58	0.64	0.65	0.17	0.36	0.40			
Conradson Carbon Residue (%)	1-A	1-A	1-A	1-A	1-A	1-A			
Copper Strip Corrosion (2)									

(1) False bottom, 4 hour test  
 (2) 212°F, 3 hour test (1-A is a slight tarnish)  
 Note: Samples were irradiated at MTR gamma canal.



TABLE II

EFFECTS OF CO<sup>60</sup> GAMMA RADIATION ON A REFINED PETROLEUM OIL (MIL-L-6082A - GRADE 1065)

	0	5.0 x 10 <sup>6</sup>	1.0 x 10 <sup>7</sup>	3.3 x 10 <sup>7</sup>	1.0 x 10 <sup>8</sup>
Dosage (Roentgens)					
Viscosity (centistokes)					
400°F	2.18	2.08	2.04	2.02	2.08
210°F	11.39	11.04	10.95	10.90	11.35
130°F	47.14	46.20	46.03	46.37	49.44
100°F	103.8	101.5	105.1	102.4	109.7
Viscosity at 130°F (cp)	40.50	39.69	39.52	39.86	42.53
Specific Gravity, 68/68°F	0.8823	0.8823	0.8818	0.8829	0.8834
Density at 130°F (gm/cm <sup>3</sup> )	0.860	0.860	0.858	0.861	0.861
Refractive Index at 25°C	1.4825	1.4825	1.4826	1.4828	1.4835
Flash Point (°F)	455	455	450	460	445
Fire Point (°F)	525	525	520	535	525
Autogenous Ignition Temperature (°F)	720	720	705	705	710
Evaporation, % (6½ hours at 400°F)	5.43	5.66	5.43	5.04	5.88
Neutralization Number (mg KOH/gm oil)	0.01	0.01	0.01	0.01	0.01

TABLE II (Continued)  
EFFECTS OF CO<sup>60</sup> GAMMA RADIATION ON A REFINED PETROLEUM OIL (MIL-L-6082A - GRADE 1065)

Dosage (Roentgens)	0	5.0 x 10 <sup>6</sup>	1.0 x 10 <sup>7</sup>	3.3 x 10 <sup>7</sup>	1.0 x 10 <sup>8</sup>
Miniaturized Oxidation-Corrosion Test (1)					
Weight Loss on Metals (mg/cm <sup>2</sup> )					
Magnesium	0.033*	0.067*	0.118*	0.016	0.016
Aluminum	0.016*	0.016*	0.000	0.016	0.016
Copper	1.981	0.948	0.220	1.524	1.710
Silver	0.016	0.016	0.050*	0.016*	0.033
Steel	0.050*	0.050*	0.033*	0.016	0.016
100°F Viscosity (centistokes)					
Before Test	103.8	101.5	105.1	102.4	109.7
After Test	243.0	201.0	174.8	500.6	283.7
% Change	134.1	98.0	66.3	388.9	158.6
Neutralization Number					
Before Test	0.01	0.01	0.01	0.01	0.01
After Test	5.26	3.97	5.13	5.26	4.10
Change	5.25	3.96	5.12	5.25	4.09
Evaporation Loss (%)	0.50	0.47	0.48	0.46	0.50

(1) 72 hours at 347°F

\* Weight gain

Note: Samples were irradiated in WADC Co<sup>60</sup> source.



TABLE III  
EFFECTS OF FUEL ELEMENT GAMMA RADIATION ON DIESTER FLUIDS

	D1-2-Ethylhexyl Adipate			D1-2-Ethylhexyl Azelate			D1-2-Ethylhexyl Sebacate		
	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>
Dosage (Roentgens)									
Viscosity (centistokes)									
210°F	2.75	2.85	3.08	3.39	3.54	3.76	3.39	3.42	3.78
100°F	9.76	10.28	11.49	12.55	13.26	14.73	12.61	13.22	15.25
-65°F	5898	6613	8653	7110	7834	10,571	7964	8824	12,063
Pour Point (°F)	B -75 (2)	B -75	B -75	B -75	B -75	B -75	B -75	B -75	B -75
Flash Point (°F)	375	350	330	405	375	330	410	385	275
Evaporation, % (6½ hours at 400°F)	45.0	33.4	43.1	17.0	22.1	17.4	15.6	19.8	12.2
Neutralization Number (mg KOH/gm oil)	0.64	1.76	4.09	0.01	1.08	3.19	0.34	1.46	4.50
Coking at 600°F (mg) (1)	25	5	36	15	10	34	24	5	24
Rubber Swell, "H" Stock (%)	32.6	34.7	38.8	21.1	22.6	27.9	17.1	18.7	26.1
Foaming (MIL-L-7808 Control)	Pass	Pass	Pass	Pass	Pass	Pass	Pass	Pass	Pass

(1) False bottom, 4 hour test  
(2) B -75 means below -75°F  
Note: Samples were irradiated at MIR gamma canal



TABLE IV  
EFFECTS OF CO<sup>60</sup> GAMMA RADIATION ON DI-2-ETHYLHEXYL SEBACATE (PLEXOL 201)

	0	5.0 x 10 <sup>6</sup>	1.0 x 10 <sup>7</sup>	4.3 x 10 <sup>7</sup>	1.1 x 10 <sup>8</sup>
Dosage (Roentgens)					
Viscosity (centistokes)					
400°F	1.09	1.08	1.09	1.13	1.22
210°F	3.31	3.36	3.41	3.58	3.98
130°F	8.05	8.05	8.14	8.82	10.22
100°F	12.70	12.72	12.99	14.11	16.62
Viscosity at 130°F (cp)	7.18	7.19	7.26	7.88	9.14
Specific Gravity, 68/68°F	0.9148	0.9154	0.9142	0.9166	0.9166
Density at 130°F (gm/cm <sup>3</sup> )	0.891	0.892	0.892	0.893	0.893
Refractive Index at 25°C	1.4478	1.4825	1.4481	1.4489	1.4502
Flash Point (°F)	415	425	435	400	255
Fire Point (°F)	495	505	480	500	440
Autogenous Ignition Temperature (°F)	775	775	775	780	755
Evaporation, % (6½ hours at 400°F)	11.44	11.78	12.25	12.96	11.47
Neutralization Number (mg KOH/gm oil)	0.01	0.32	0.78	3.07	6.92

TABLE IV (Continued)  
EFFECTS OF CO<sup>60</sup> GAMMA RADIATION ON DI-2-ETHYLHEXYL SEBACATE (PLEXOL 201)

Dosage (Roentgens)	0	5.0 x 10 <sup>6</sup>	1.0 x 10 <sup>7</sup>	4.3 x 10 <sup>7</sup>	1.1 x 10 <sup>8</sup>
Miniatized Oxidation-Corrosion Test (1)					
Weight Loss on Metals (mg/cm <sup>2</sup> )					
Magnesium	16.923	28.764	20.988	21.937	30.764
Aluminum	0.016*	0.000	0.016	0.000	0.016
Copper	3.777	1.897	2.507	3.083	1.355
Silver	0.033*	0.067*	0.016*	0.016*	0.033
Steel	0.000	0.033*	0.016	0.033	0.050
100°F Viscosity (centistokes)					
Before Test	12.70	12.72	12.99	14.11	16.62
After Test	(2)	(2)	(2)	Solid	Solid
% Change					
Neutralization Number					
Before Test	0.01	0.32	0.78	3.07	6.92
After Test	11.42	15.65	13.34	13.47	27.71
Change	11.41	15.33	12.56	10.40	20.79
Evaporation Loss (%)	3.28	2.77	3.33	2.26	2.37

(1) 72 hours at 347°F

(2) Oil turned to sludge

\* Weight gain

Note: Samples were irradiated in WADC Co<sup>60</sup> source.



temperature experienced little change. Evaporation was also unaffected. The greatest property change noted was that for the neutralization number, which went from 0.01 mg KOH/gram of oil to a high of 6.92, indicating a great increase in the acidic constituents as a result of irradiation. As could be expected, the uninhibited oil corroded certain metals during the 347°F oxidation-corrosion test, showing the greatest attack on magnesium and copper. The most important consequence of this test is that it showed little corrosion on the aluminum, silver, or steel and a relatively small increase in the very large corrosion on magnesium and copper.

The data of Table V illustrate the effect of phenothiazine on the radiation stability of Flexol 201. Viscosities again show the characteristic increase with dosage. It is interesting to note that in some manner the phenothiazine inhibits the decrease in flash point, the flash remaining constant up to  $3 \times 10^8$  roentgens. Evaporation, coking, neutralization number, and rubber swell all show an increase with radiation dosage. The static corrosion increases with irradiation, particularly for the copper. Except for corrosion on magnesium, the 347°F oxidation-corrosion test shows little effect on metals. The neutralization number change for this test also increased for the larger dosages, but not as much as for the uninhibited oil.

Figure 1 illustrates the change in oxidation stability of an inhibited diester with increasing radiation dosage. The irradiated base stock reported in Table IV was blended with 0.5% phenothiazine after each irradiation and oxidized in the Shell Development Company version of the Dornte apparatus (347°F, copper wire catalyst, circulated oxygen). The induction period, or time to reach the break point of the  $O_2$  absorption curve, was changed by the irradiation. The induction period fell quite rapidly, until at  $2.5 \times 10^7$  roentgens, the oil absorbed oxygen at a constant rate with no induction period break point observable. This loss of oxidative stability as a result of irradiation is a common characteristic of oils exposed to nuclear radiation and is reported by Back (1) and the Shell Development Company (5).

Figure 2, which is taken from the data of Figure 1, gives an interesting relationship. Taking the induction period as the time to absorb 0.5 moles of oxygen per 500 grams of oil, and plotting this value against the radiation dosage, the curve of Figure 2 is obtained. In the range of  $1 \times 10^7$  to  $1.1 \times 10^8$  roentgens, a log-log plot gives a straight line relationship between the oxygen uptake and the dosage.



TABLE V

EFFECTS OF GAMMA RADIATION ON AN INHIBITED DIESTER  
(Di-2-Ethylhexyl Sebacate (Plexol 201) plus 0.5% Phenothiazine)

Dosage (Roentgens)	0	3 x 10 <sup>7</sup>	1 x 10 <sup>8</sup>	3 x 10 <sup>8</sup>
Viscosity (centistokes)				
210°F	3.27	3.43	3.60	4.30
100°F	12.69	13.08	13.98	17.68
-65°F	10,118	9,762	13,931	17,978
Flash Point (°F)	460	460	455	460
Evaporation, % (6½ hrs., 400°F)	7.7	10.8	11.2	15.0
Coking at 600°F (mg)	5 (2)	13 (2)	74 (2)	91 (2)
Neutralization No. (mg KOH/gm)	0.06	0.12	0.24	0.74
Rubber Swell, "H" Stock (%)	16.7	17.5	20.4	29.2
Gear Test (lb./inch)	2180	2210	2090	2250
Foaming (1)	Pass	Pass	Pass	Pass
450°F Corrosion (mg/in <sup>2</sup> )				
Silver	-0.18	-0.23	-0.22	-0.46
Copper	-0.80	-0.83	-0.98	-3.08
347°F Oxidation-Corrosion				
Weight Change (mg/cm <sup>2</sup> )				
Copper	-0.02	0.02	-0.04	0.07
Silver	-0.02	0.01	0.00	0.04
Steel	0.00	0.05	-0.04	-0.11
Aluminum	0.00	0.04	0.00	0.07
Magnesium	-0.16	-0.01	-5.26	-10.21
100°F Viscosity Change (%)	2.60	1.26	13.23	22.06
Neutralization Number Change	0.23	1.86	3.58	3.77

(1) MIL-L-7808 control

(2) False bottom, 4 hour test

Note: Samples irradiated at MTR gamma canal

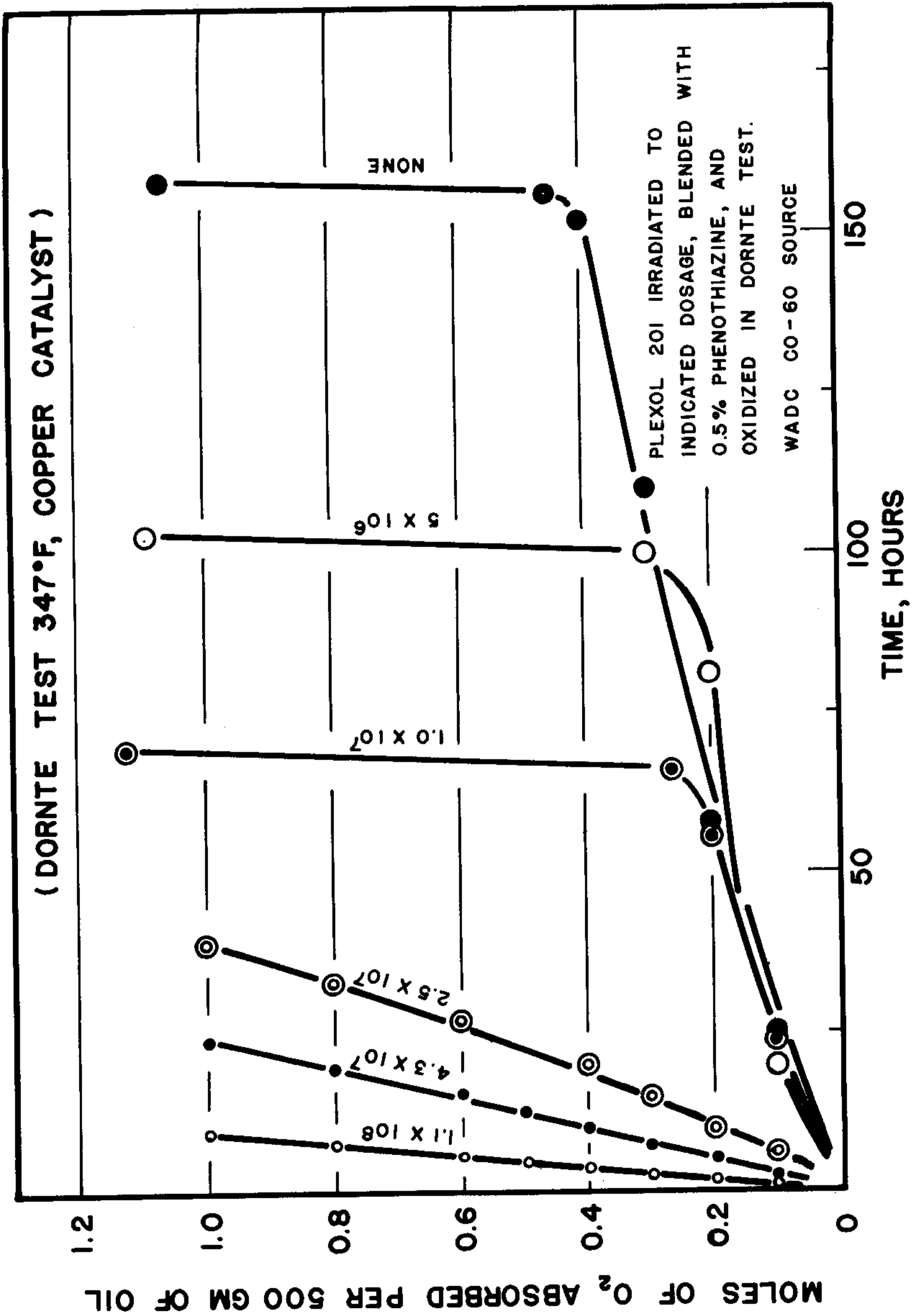


FIGURE 1 : EFFECT OF GAMMA RADIATION ON INHIBITOR SUSCEPTIBILITY OF PLEXOL 201



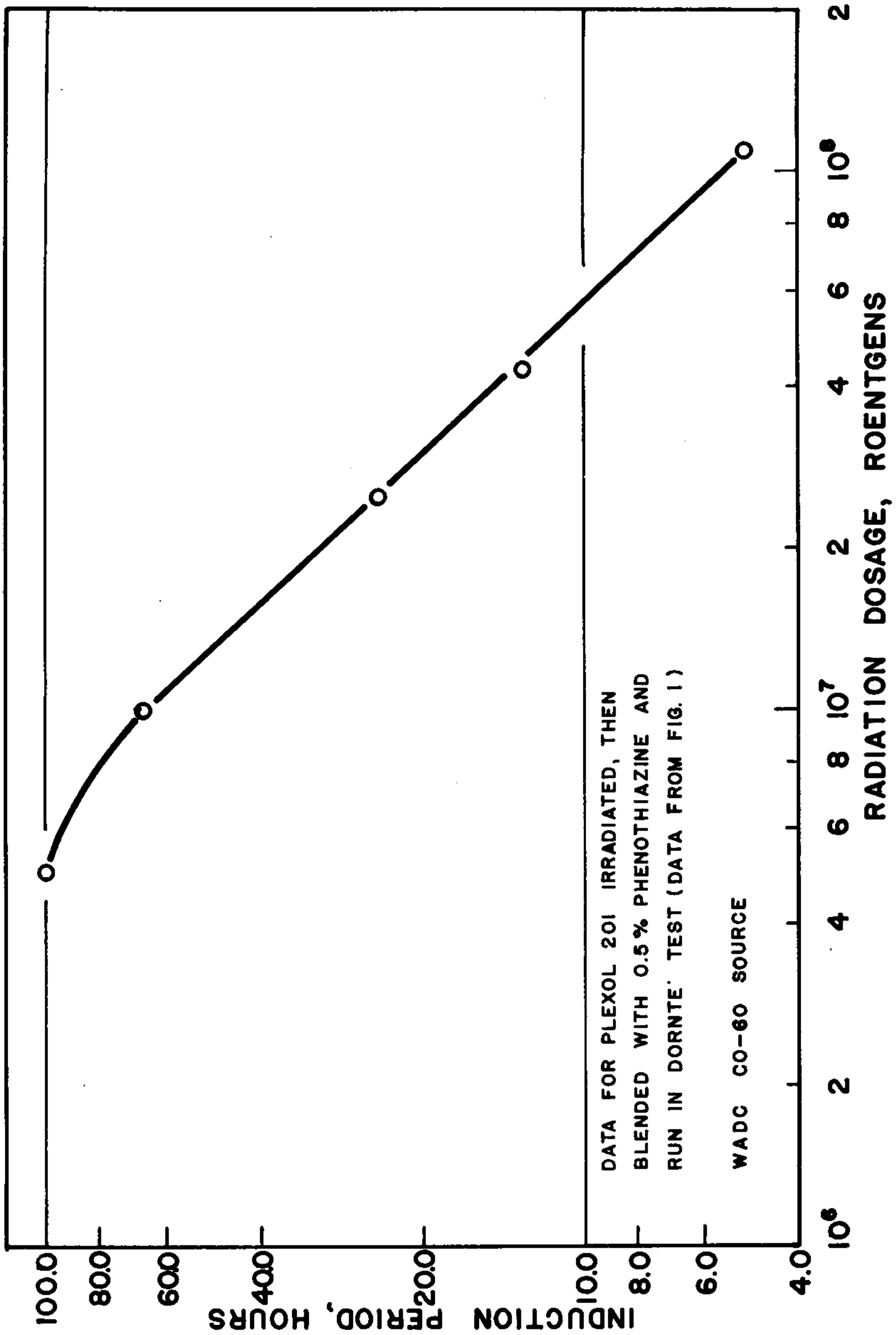


FIGURE 2 : DIESTER INDUCTION PERIOD VS GAMMA DOSAGE

### C. Mineral Oils or Diesters as Secondary Dosimetry Standards

The radiation stability of the mineral oils and diesters studied is such that in the range of  $1 \times 10^7$  to  $1 \times 10^8$  roentgens, only a few tests can be used to trace the radiation exposure. Infrared analysis, which was conducted on all of the irradiated mineral oils and diesters, showed no change in the infrared spectra up to the maximum dosage. For commercial diesters of the Flexol 201 variety, all changes in the range from 5 to 12 microns are masked by the base stock, so the history of the radiation induced changes cannot be traced. The mineral oil reported in Table II did not show any change in infrared spectra with increasing radiation dosage, which is consistent with the lack of change in the general physical and chemical properties of the fluid. Infrared analysis does not follow the dosage for either the diester or mineral oil type fluid up to  $1 \times 10^8$  roentgens and cannot be used to indicate exposure to gamma radiation for these fluids.

The induction period of Figure 2 has a possibility of use for testing irradiated diester formulations, since it gives a straight line relationship over an order of magnitude change in radiation dosage. Since this is not a standard test, it would not be easy to establish the induction period change as the basis for evaluating a secondary reference fluid of the diester type. A simpler technique would be preferable.

The change in neutralization number of the diester Flexol 201 appears to have promise as a method of determining the exposure to radiation. Figure 3 shows the data on neutralization number change for the irradiated diesters. Although the data are preliminary, it would appear that from  $1 \times 10^7$  to  $1 \times 10^8$  roentgens this test would be feasible for determining the dosage received.

Of the property changes investigated, the one that appears most promising over a wide range of exposures is the gas evolution of the exposed material. Table VI and Figure 4 illustrate some data on gas evolution of irradiated mineral oils. As can be seen, a loglog plot of gas evolution against radiation dosage shows the gassing to be a power function of the dosage. This indicates the possibility of a technique usable over a wide range, i.e., from  $1 \times 10^7$  through  $1 \times 10^9$  roentgens. More accurate determinations at the WADC Co<sup>60</sup> source on a wide range of fluids show the same sort of straight line relationship. All of the fluids irradiated, mineral oils, silicones, and disiloxanes, gave a straight line relationship on loglog paper for gassing with increasing dosage.



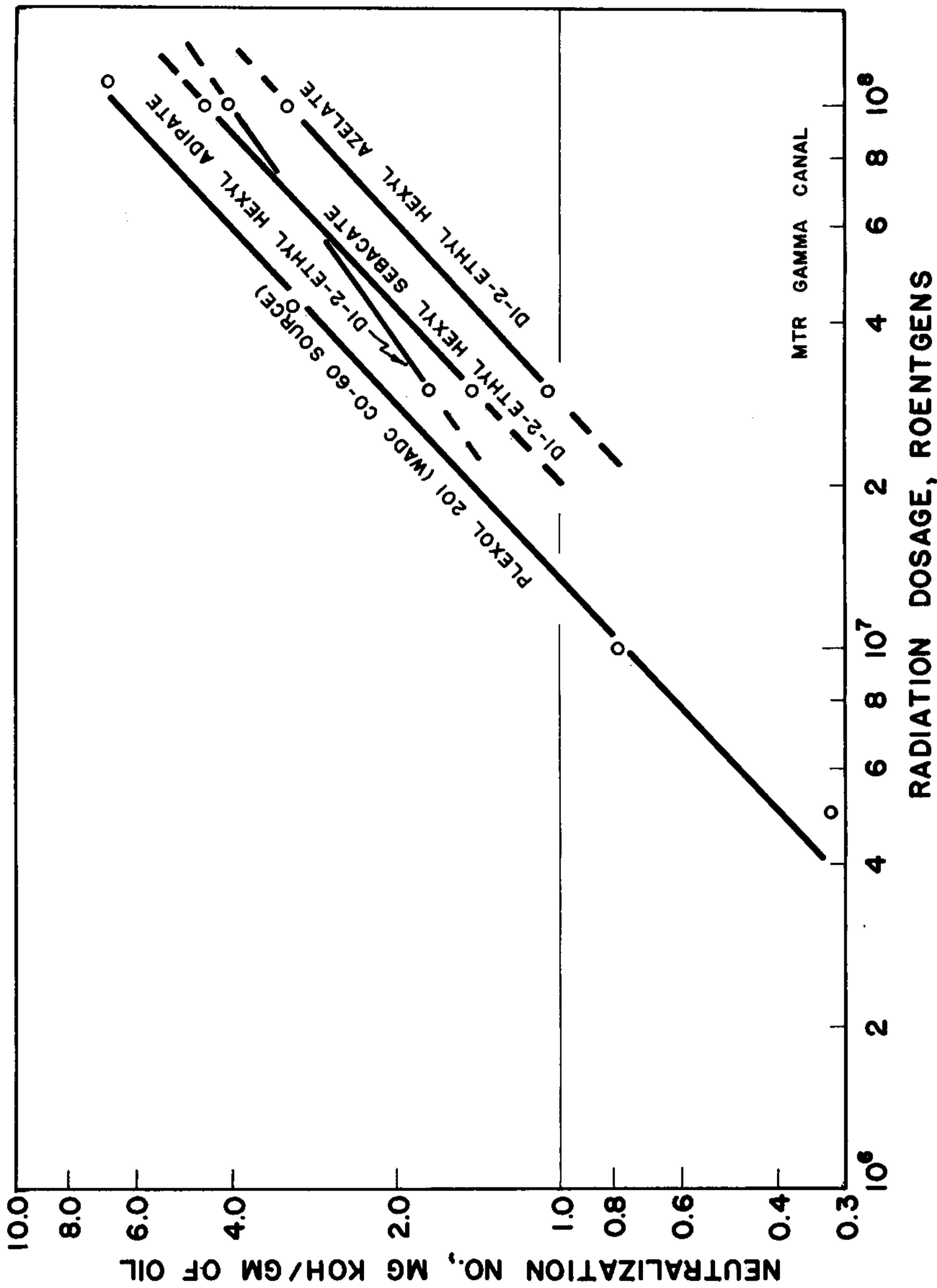


FIGURE 3: EFFECT OF DOSAGE ON DIESTER NEUTRALIZATION NUMBERS

TABLE VI  
GAS EVOLUTION OF IRRADIATED PETROLEUM OILS

MIL-0-6081		MIL-L-6082					
Grade 1005	Grade 1010	Grade 1065	Grade 1100				
Dose	ML Gas/ML Liq.	Dose	ML Gas/ML Liq.				
Dose	ML Gas/ML Liq.	Dose	ML Gas/ML Liq.				
1.0 x 10 <sup>7</sup>	0.09	1.0 x 10 <sup>7</sup>	0.19	1.1 x 10 <sup>7</sup>	0.23	1.1 x 10 <sup>7</sup>	0.2
1.0 x 10 <sup>7</sup>	0.07	1.0 x 10 <sup>7</sup>	0.22	9.9 x 10 <sup>6</sup>	0.19	3.1 x 10 <sup>7</sup>	0.0
3.0 x 10 <sup>7</sup>	0.25	3.0 x 10 <sup>7</sup>	0.66	3.0 x 10 <sup>7</sup>	0.74	1.0 x 10 <sup>8</sup>	1.9
3.0 x 10 <sup>7</sup>	0.26	3.0 x 10 <sup>7</sup>	0.68	3.0 x 10 <sup>7</sup>	0.84	3.0 x 10 <sup>8</sup>	6.5
1.0 x 10 <sup>8</sup>	0.97	9.9 x 10 <sup>7</sup>	1.87	1.0 x 10 <sup>8</sup>	4.17	1.0 x 10 <sup>9</sup>	20.0
1.1 x 10 <sup>8</sup>	0.98	1.0 x 10 <sup>8</sup>	2.03	1.0 x 10 <sup>8</sup>	2.22	3.0 x 10 <sup>9</sup>	53.4
3.0 x 10 <sup>8</sup>	2.17	3.0 x 10 <sup>8</sup>	5.76	3.0 x 10 <sup>8</sup>	6.70		
3.0 x 10 <sup>8</sup>	1.90	3.0 x 10 <sup>8</sup>	5.84	3.4 x 10 <sup>8</sup>	9.43		
9.5 x 10 <sup>8</sup>	9.65	1.0 x 10 <sup>9</sup>	20.6	1.0 x 10 <sup>9</sup>	21.7		
1.0 x 10 <sup>9</sup>	9.75	1.0 x 10 <sup>9</sup>	19.7	1.0 x 10 <sup>9</sup>	26.4		

Note: Samples irradiated in MTR gamma canal.



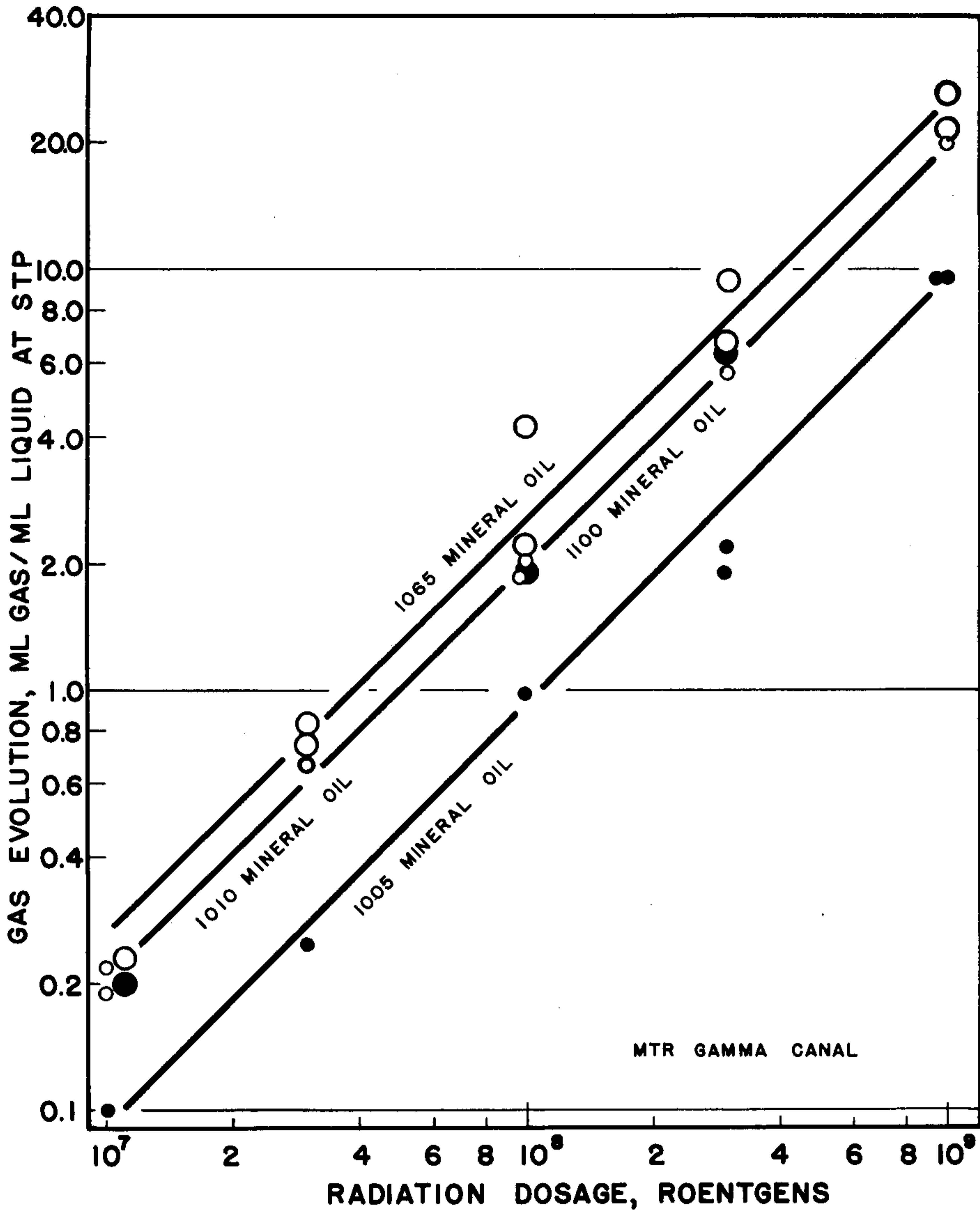


FIGURE 4: EFFECT OF GAMMA RADIATION ON GASSING OF MINERAL OILS

## III. CONCLUSIONS

A. Radiation Stability of Mineral Oils and Diester Fluids

For the fluids tested and reported herein, it is concluded that the mineral oils meeting Specification MIL-L-6082 should possess general stability to gamma radiation up to the level of  $1 \times 10^8$  roentgens, with relatively minor changes in a few properties such as viscosity.

The uninhibited diester fluids show much greater susceptibility to gamma radiation in the same range. Viscosity shows an increase, especially at low temperatures, flash point goes down radically, and neutralization number shows a large increase. Also, addition of an inhibitor (phenothiazine) to the irradiated di-2-ethylhexyl sebacate (Plexol 201) does not prevent loss of oxidative stability.

Over the same radiation range (up to  $1 \times 10^8$  roentgens), Plexol 201 inhibited with 0.5% phenothiazine prior to irradiation shows a slight improvement over the base stock. The viscosity does not increase to as great an extent with irradiation, the flash point remains stable, and the neutralization number does not increase as much. Since the base stock and the inhibited formulation were irradiated in different sources, the data can not be compared too closely, but it is evident that a general increase in capabilities of the ester is observed if it is inhibited. However, it must be emphasized that extensive tests indicate that the decrease in oxidative resistance of irradiated aliphatic hydrocarbons and diester fluids as measured by the Dornte test is not hindered by the addition of inhibitor prior to or following irradiation (5).

It can be concluded that on the basis of this and other work (6), diesters do not have as great a resistance to property changes as do mineral oils following exposure to increasing dosages of gamma radiation.

B. Dosimetry by Means of Diester or Mineral Oil Property Changes

Of the tests performed, only two appear to have promise for following the radiation exposure history. One is the change in neutralization number of the diesters with increasing gamma dosage and the other is the gas evolution of a fluid with increasing dosage, both properties being power functions of the radiation



exposure. Since both tests are relatively easy to perform, there would not be any great difficulty in establishing standards for measuring the dosage. The gas evolution would be particularly convenient, for it would only necessitate a calibrated gauge for attachment to the system, the pressure reading being directly related to the radiation exposure.

#### IV. FUTURE PLANS

Evaluation of the stability of diesters and mineral oils to higher radiation levels has already been undertaken. Testing is now in progress on mineral oils irradiated to  $1 \times 10^9$  roentgens by WADC and the data will be reported when completed. Diesters and mineral oils are being extensively studied under contract by the Esso Research and Engineering Company (6) which is continuing the work along this line. Information on the effects of additives in base stocks has been reported in references (4) and (5) and will be presented in future reports.

Measurement of fluid property changes to reflect radiation exposure is being undertaken by Air Force contractors and will be discussed in future contract reports.

## BIBLIOGRAPHY

- (1) Bach, Radiolytic Oxidation of Organic Compounds, Proceedings of the International Conference on Peacetime Uses of Atomic Energy, Vol. 7, "Nuclear Chemistry and Effects of Radiation," United Nations, New York (1956).
- (2) Collins, C. G. and Calkins, V. P., Radiation Damage to Elastomers, Organic Liquids, and Plastics, Apex 261, General Electric, Atomic Products Division, Aircraft Nuclear Propulsion Department, Cincinnati 15, Ohio (September 1956).
- (3) Federal Test Method Standard Number 791, Lubricants, Liquid Fuels, and Related Products: Methods of Testing, General Services Administration, Washington 25, D. C. (15 December 1955).
- (4) Wright Air Development Center Technical Report 56-646, Effects of Radiation on Aircraft Fuels and Lubricants, Summary Report, September 1, 1955 through December 31, 1956, California Research Corporation report on Air Force Contract AF 33(616)-3184.
- (5) Wright Air Development Center Technical Report 57-177, Engine Oil Development for Wright Air Development Center, Summary Report, September 15, 1955 through November 1, 1956, Shell Development Company report on Air Force Contract AF 33(616)-3182.
- (6) Wright Air Development Center Technical Report 57-255, Nuclear Radiation Resistant Turbine Engine Lubricants, Summary Report, Report by A. H. Matuszak, February 1, 1956 to February 1, 1957, Esso Research and Engineering Company report on Air Force Contract AF 33(616)-3181.



