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RESEARCH ON THE RADIATION STABILITY
OF ORGANIC FLUIDS

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RESEARCH ON THE RADIATION STABILITY OF ORGANIC FLUIDS

A B S T R A C T

Additional knowledge has been gained pertaining to effects of irradiation variables, chemical components, etc., on the stability of organics. This basic program has been supplemented with an applied program wherein research was undertaken of particular interest in reactor moderator-coolant and lubricant applications. Irradiations of the capsule type in the absence of air were conducted in the MTR and BNL and X-10 reactors under a variety of conditions in investigations of effects of temperature, flux, dosage, and organic chemical structure. The MTR gamma facility and the California Research Co⁶⁰ source were also used in work with gammas.

With highly aromatic organics at BNL, damage measured by viscosity change was found independent of irradiation temperature up to 371°C (700°F) and $N^* \sim 5$. Coke formation was minor under these conditions, although it was universal at 426°C (800°F) and $N \sim 3$. Damage generally depended on temperature up to 371°C at $N \sim 8$. Up to $N \sim 5$, a log function of viscosity change was found to vary linearly with radiation dosage.

Many fused ring and nonfused ring aromatics were stable at 371°C and $N \sim 3$. Viscosity changes were slight, and gassing amounted to about 2 to 5 ml gas/ml fluid. Simple molecular distillations showed materials, so exposed, e.g., biphenyl, naphthalene, and 1,3-diphenylbenzene, to be 93%-97% distillable, of which 72%-81% was pure starting material. At $N \sim 8$ and 371°C, gas evolution increased by a factor of ~ 10 . Biphenyl was the only organic to remain fluid under these conditions. However, it exhibited a large increase in viscosity. It is apparent from this that a continuous purification system, probably a distillation technique, is a necessary prerequisite to the successful use of an organic as a moderator-coolant.

The stabilities of solutions of highly viscous polymers in low viscosity alkylbenzenes were studied in the presence of air with gamma radiation. Styrene-alkene copolymer, polyalkene, and polyester solutions markedly decreased in viscosity on irradiation; whereas, polystyrene solutions merely increased slowly. This discovery makes possible the formulation of "constant viscosity" fluids for various dosages and is the basis for a series of new lubricants, covering a wide viscosity range, designed for maximum resistance to viscosity change on irradiation.

Comparative stabilities of representative types of organics with combined and gamma radiation have allowed the tentative determination, based on viscosity change, that one fast neutron (>1 mev) causes ~ 16 times the damage of one gamma (1.6 mev). Thermal neutrons cause insignificant damage. As a result of this study, it is suggested that the radiation dosage to cause damage be expressed in terms of "equivalent fast neutrons," a number obtained by adding to the number of fast neutrons the damage equivalent number each of gamma rays and epithermal neutrons. This should aid in the interpretation of existing data for various applications.

$$*N = \text{Reactor dosage} \times 10^{-18}, \text{ thermal neutrons/cm}^2$$

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1. INTRODUCTION

1.1 Previous Research and Publications

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Frequent inquiries are received regarding the scope and results of previous research and the availability of past reports. For this reason, Table 1.0 notes reports and publications of the progress of California Research studies on the radiation stability of organic fluids. Technical progress reports are now being issued about twice yearly. The four most recent reports are available through the AEC Technical Information Service at Oak Ridge, Tennessee. Future publications of this sort will also be available from this source.

Since January 1953, monthly Progress Letters have been written. These consist of one page and cover general objectives and over-all results of the research effort. They are distributed through the AEC San Francisco Operations Office to individuals known to be interested in the developments in this field of research. Personnel desiring copies of future Progress Letters should direct inquiries to Dr. I. F. Zartman, AEC Division of Engineering, Washington 25, D. C.

California Research personnel participate in conferences and symposia at such times as the subject matter relates to the uses and applications of organic fluids in programs of AEC interest.

1.2 Present Research and Organization

This report covers a nine-month period from the beginning of calendar 1953. During this period, there were two objectives of the work reported:

- a. to conduct basic research on the radiation stability of organic fluids,
- b. to relate results to AEC applications.

To further these basic objectives, the research effort was directed along several avenues, as follows:

The Basic Program is concerned with a study of the effects of irradiation variables, chemical structure, etc. on the radiation stability of organic fluids. Reactor dosages up to approximately 8×10^{18} thermal neutrons/cm²,¹ temperatures to 426°C (800°F), and fluxes to about 10^{14} thermal neutrons/cm²/sec. were used. Relative stabilities of various chemical structures and the inhibition effects of chemical additives were also studied. During this report period, approximately 250 fluid samples were exposed to combined radiations in various reactors, including ORNL X-10, MTR, and BNL. In addition, about 210 samples of organic fluids were exposed to pure gamma radiation at dosages

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up to 10^{10} R. Exposures were made in the MTR gamma facility where gamma fluxes are about 10^7 R per hour and in the California Research Co^{co} source where a flux of about 2×10^5 R per hour prevails. Initial attempts to assess the relative thickening effects of neutrons and gamma photons on organic fluids have also been made.

The Applied Program is concerned with the application of the basic knowledge to specific problems within the AEC reactor development program. The possible use of organic fluids as moderator-coolants has received the major emphasis. Thermal stability studies have been conducted, chiefly on polynuclear aromatic materials, at 493°C (920°F). Certain of these have subsequently been exposed to combined reactor flux up to 426°C (800°F). Work on lubricants has also been continued on the basis of specific requests received for radiation stable materials of this sort. Several oils and greases have been formulated corresponding, respectively, to Society of Automotive Engineers (SAE) and to the National Lubricating Grease Institute (NLGI) grades. The stability of these formulations to gamma rays is being studied in both the California Research Co^{co} and in the MTR gamma facility. Due to increasing interest in lubricants, a separate report, California Research-AEC Report No. 7, devoted entirely to this subject, will issue shortly.

Research on organic shield materials has received little emphasis during this report period. The survey conducted on this subject prior to May 1951 seems still pertinent to aspects of this problem raised more recently. A few alcohols and amines, high hydrogen content organics, have been exposed both to combined radiations and to gamma radiations in new work.

A Consulting Program exists whereby California Research personnel endeavor to assist organizations in reactor development with problems involving organics. In this way, the accumulated fund of knowledge on this subject yields the greatest benefits. Also, intimate contact with new problems allows the course of new research to be charted so as to provide information of most interest. Cooperative work has been instituted in two instances in which existing parallel programs of research could be mutually complemented. This work has expedited an exchange of information and samples to a degree not otherwise possible.

A problem related to consulting work is that of supplying samples of promising organics for test work by interested personnel. In this supply program, small samples have been provided gratis. Larger quantities (one gallon or more) have been paid for by the requestor on a cost basis, which includes charges for materials, preparation time, and control tests, if required.

Along with the discussions of the foregoing items, a section in Report No. 6 is devoted to future work. Included here are details and justification of the proposed new effort.

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2. BASIC PROGRAM

2.1 Research with Combined Radiation

2.11 Experimental Approach

The principal projected uses of organic fluids in power reactors include such applications as moderator-coolants, shielding materials, lubricants, and power transmission fluids. These applications will involve exposure either to combined radiations or to gamma flux. For this reason, samples investigated in the present work were irradiated in reactors or in a gamma source. The latter work will be reported under a subsequent heading. In the former case, the Brookhaven and the Oak Ridge graphite reactors and the Materials Testing Reactor were used. All experiments reported involved capsule-type irradiations wherein before and after observations were made.

The exposures conducted are expressed in terms of thermal neutron dosage, primarily because these data are the most readily available; and, in a given reactor situation, the fast and gamma flux is proportional to the thermal neutron flux. Recognizing that a knowledge of the effects of the components of combined flux is extremely important, considerable monitoring work was undertaken in order to assess quantitatively the relative levels of fast flux, thermal flux, and gamma flux. The objective is to determine the damaging ability of these various components in terms of a particular criterion. Until a fairly complete definition of the relative effects of these components is determined, the thermal neutron dosage terminology will continue to be used. The dosage term presently used is defined as N , which equals dosage $\times 10^{-18}$, expressed as thermal neutrons/cm². Another method of expression would be in terms of energy absorbed. In order to use this basis, the energy spectra of the gamma and neutron fluxes must be known with a reasonable degree of accuracy. A first step in evaluating the relative damage of the components of combined flux has been taken, and subsequent sections will illustrate relative indexes of damage in terms of incident flux. Section 2.4 presents a discussion of the validity and limitations of such an approach to the use of energy absorbed as a measure of dosage received.

The materials irradiated in the present work were chosen mainly from chemical structures known or expected to be relatively radiation stable on the basis of previous studies. Other materials, on which knowledge was insufficient to permit estimation of radiation stability, were also irradiated. Many materials were chosen on the basis of possible end uses. Here, particular attention was paid to what might be termed "fatigue" testing; i.e., an attempt to determine limiting temperatures and limiting dosages for the most stable structures. The end point in mind was the extension of the basic program to cover moderator-coolant situations.

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2.12 Equipment

Various types of capsules were used to contain the samples of organic fluids exposed to combined radiations. Several of these capsules are illustrated in Figure 2.0. The numbers below the figure refer to internal volume in ml of each container. In exposures below 140°C (285°F), either a semihard 2S aluminum or a 58135-type magnesium was used, with the latter preferred due to somewhat better tensile strength. For temperatures above 140°C, containers of Types C-12 and C-22, shown in Figure 2.0, were fabricated from 410 stainless steel. This material was necessary to attain satisfactory tensile strengths at higher temperatures up to 800°F (426°C).

All vessels were designed to withstand the estimated maximum decomposition pressures of the fluids. For the 410 SS capsules, typical tests show that they will hold up to 7500 psi at 700°F (371°C). Magnesium-type vessels have, in general, been designed to hold 5000 psi of hydraulic pressure at low temperatures.

As illustrated by container C-17G in Figure 2.0, all vessels feature a bursting disc. This disc serves two purposes. First, its thickness is selected so that it will rupture about 2000 psi below the maximum bursting pressure of the vessel. Thus, should excessive pressures be generated, the safety disc will rupture rather than the entire vessel. Second, the disc is also designed for penetration by a hollow needle. This needle first seals against a small annulus of soft material, such as aluminum, and then punctures the disc. The gases contained in the vessel then enter a manifold system containing pressure gages so that the pressure of evolved gas can be measured accurately. Outlets are attached to this system so that samples of the gas may be collected for subsequent analysis. Usually, five ml of a fluid is introduced into the capsules; although as large a quantity as 15 ml has been exposed in some of the larger vessels.

The mechanics of actual exposure vary somewhat with the type of facility used. For example, in ORNL Hole 19, capsules of the C-31 type were placed by hydraulic pressure directly and individually in the water-cooled hole. Similarly, vessels of the C-17 type were exposed in the VH-3 hydraulic facility at the MTR. C-18.5 containers were exposed 16 at a time in an outer aluminum basket in the MTR VG-51 facility. Vessels of the C-12 and C-22 type, of 410 stainless steel, were contained in outer ovens when exposed at elevated temperatures in the Brookhaven Reactor. These ovens, which have been described in detail in previous reports, are equipped with heaters which can be controlled from the reactor face. Usually, about 18 capsules can be fitted into such an oven; and as many as four ovens have been exposed concurrently. Magnesium vessels of the C-12.2 type have been exposed similarly in the Brookhaven Reactor but at lower temperatures.

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2.13 Monitoring

It is apparent that for the quantitative assessment of the comparative degradation of organic fluids, which might be ascribed, for example, to slow or fast neutrons, intensities must be known for the individual components of combined flux. Although the neutron and gamma energy spectra in Hole 19 of the ORNL graphite reactor are known experimentally (Sisman, Bopp - ORNL-525), those at Brookhaven and at the MTR have been based on theoretical calculations. To extend this information, monitoring work on the various components was undertaken.

To determine the comparative prevalence of fast neutrons, neptunium-237 (Np^{237}) monitors were exposed in various reactors. These monitors, which consist of Np^{237} plated on platinum foils, were wrapped in aluminum to prevent loss of fission products and surrounded by cadmium-silver shields to absorb thermal and epithermal neutrons. In this method, developed by Dr. R. Fiore of Argonne, strontium-89 (Sr^{89}) is separated from a solution of the irradiated neptunium, platinum, and aluminum. Counting of this material yields data on fast neutron (above about 1 mev) dosage. An accuracy of about $\pm 25\%$ is ascribed to dosages determined in this manner. The principal source of error is the fission yield of Sr^{89} .

Foils of cobalt-aluminum (Co-Al) were used in thermal flux evaluations. The alloy employed contains $0.620\% \pm 0.006\%$ cobalt, as determined by activation analysis in comparison with a cobalt standard. Such activation was measured by gamma ray counting in the present work. An accuracy of about $\pm 10\%$ is ascribed to dosages determined by this means.

In an attempt to determine the gamma component of combined flux, a small amount of work was undertaken with a calorimetric device. This was first developed and used by Mr. C. Hogg at the MTR and is described in IDO-16093-PPCo. A schematic sketch of this device is illustrated in Figure 2.1. It consists primarily of a center rod of comparatively high molecular weight material, such as lead, encased in an outer shell of aluminum. Thermocouples are imbedded at two points in the lead rod. Insulation is placed around the rod inside the outer shell. In this arrangement, heat induced in the lead rod by gamma activity can only be dissipated toward the uninsulated end. This creates a temperature differential between the two thermocouples, the magnitude of which is a function of the gamma energy and flux. The heat losses to the insulation are comparatively small, being in the range of 5%. Theoretically, heating due to fast neutrons is less than 1/1000 the gamma heating for a high density material such as lead. Thus far, this device (see Figure 2.2) has been used only in the Brookhaven Reactor.

In Table 2.0 are presented summary data of values for thermal flux, fast flux, and gamma flux in the reactor positions in which samples were exposed. Epithermal figures are not included

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because they are available only for ORNL (60% of thermal). The fluxes have not all been monitored during the irradiations so might be considered, at this point, as best values.

TABLE 2.0

FLUX COMPONENTS IN REACTOR POSITIONS USED

Position, (Q)	Thermal Neutrons, $n/cm^2/sec. \times 10^{-12}$	Fast Neutrons, Per Cent of Thermal	Gamma, Per Cent of Thermal
ORNL Hole 19	1*	4*	50
BNL E-25	3.2*	23*	30*
MTR VH-3	137*	7	90
MTR VG-51	1.2*	0.4*	56*

*Monitored values; others calculated.

The values for ORNL Hole 19 are taken from ORNL-928 (Sisman and Bopp). California Research has monitored the Brookhaven facility and the two MTR facilities. Of the gamma values, that for the BNL positions was monitored by California Research and that for the MTR VG-51 was monitored by Mr. Hogg. Similar gamma calorimeters were used. The other two values represent the best available calculated information. Of paramount note is the different ratios of the three components of combined flux for different reactors. This points to the limitation of expressing dosages in terms of the thermal neutron component alone. To be valid, the use of the thermal neutrons as a dosage index would require that there be no differences in damaging effects by the various components and/or that their flux ratios be the same in different reactors. The latter premise is obviously incorrect, as is the former, which point will be shown in subsequent sections.

Table 2.1 presents the results from Co-Al and Np^{237} monitors of reactor neutron flux obtained since October 1952. Values for the thermal neutron dosage are shown for both the foil evaluation by gamma count and also the comparable Operations value, where available. These latter figures from Brookhaven present an interesting comparison with the Co-Al monitor data. The Operations values of thermal neutron dosage are also shown for the positions occupied by the individual series of samples because placement varied somewhat from that of the actual monitor.

The first group of exposures comprises Series 10 through Series 16, all irradiated in the Brookhaven Reactor, Hole E-25. For these, agreement is good between dosage values obtained by Co-Al monitoring and values obtained from Operations. The ratio

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ranges from 0.92 to 1.10. The Np^{237} monitor of this group shows, for these exposure conditions, a fast neutron flux of about 23% of the thermal neutron flux.

Figure 2.3 portrays this information graphically. This is a position plot of the ovens (containing samples) and the Co-Al monitors of Series 10 through 16. Both are shown in terms of distance from the reactor centerline in relation to the appropriate dosage curve obtained by calculated values from BNL Operations. Figure 2.3 again indicates good agreement between these sets of values.

The next values to consider in Table 2.1 are those that were obtained from the exposure of Series 18 in the MTR VG-51 facility. Here previous monitoring data were not available; hence, the dosage received by the fluid samples was taken as that indicated by the Co-Al foil monitors. A Np^{237} monitor was also exposed in this group and yielded an approximate value of 0.4% for the fast neutron flux as a per cent of thermal flux.

Results of the monitors exposed with Series 19 in the MTR VH-3 hydraulic rabbit are next considered in Table 2.1. Again, because of the absence of previous monitoring data, dosages received are taken as those indicated by the foils. The Np^{237} monitor of this group has not been processed as yet. Hence, a calculated ratio of fast flux as 7% of thermal flux has been used throughout.

The next group of exposures indicated in Table 2.1 relates to Series 20 through Series 23, exposed in the Brookhaven Reactor. For these six-week exposures, the ratio of neutron dosage determined by foil monitoring and as determined from Operations is in rather poor agreement. Figure 2.4 shows this information graphically. Figure 2.4 is again a position plot of both foil monitors and samples in terms of dosage received at various positions from the Brookhaven Reactor centerline. The dosages received by the four Co-Al monitors fall considerably below the values shown on the calculated curve. Deviations from the calculated curve are shown in Figure 2.5. The nature of this deviation suggests that a neutron "sink," such as caused by the presence of a high cross-section material, existed at this position during these exposures. For this reason, the dosages ascribed to Series 20 through Series 23 are taken from the monitor values rather than from the Operations traverse.

Additional foils were exposed with the group of samples compromising Series 24, 25, and 26, as noted in Table 2.1. These values are also shown in Figure 2.4, and agreement between monitoring and Operations data is good. The Np^{237} monitor irradiated in this group yields a ratio of fast flux to thermal flux of 18% for the Brookhaven Hole E-25. This is enough at variance with the previous 23% figure that a third Np monitor is currently being irradiated in the same position.

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The last group of monitors noted in Table 2.1 was exposed in the MTR hydraulic rabbit facility in conjunction with Series 28 and 29. In this group of exposures, the monitors were exposed in pairs so that the accuracy of the processing technique could be checked. During processing, the identity of the monitors was coded so as to preclude preconceptions of the correct values to be obtained. Thermal neutron fluxes calculated from these dosage values are shown in Table 2.1A.

TABLE 2.1A

AGREEMENT OF Co-Al MONITORS

Co-Al Monitor	nvt	nv
A	9.48×10^{18}	1.350×10^{14}
H	9.36	1.335
B	4.57	1.338
G	4.62	1.351
C	1.816	1.346
F	1.848	1.368
Average		1.348×10^{14}
Average Deviation		0.6%

These flux values are in remarkably good agreement, particularly so because the three sets of duplicate monitors were exposed for different durations on different dates. This indicates that reactor power level was maintained quite constant during this period and also that the gamma counting technique used is quite reliable.

2.14 Indexes of Damage

Viscosity change continues to be the primary criterion of deterioration in the present work. This was originally chosen because it is perhaps the single most significant property of a fluid and is clearly defined in all military, commercial, and industrial specifications for organic fluids, whatever be their use. Although viscosity change is a gross index of damage in that it gives no indication of what has changed or to what extent, it is a significant index for any engineering utilization anticipated or required of a fluid.

As a criterion of viscosity change, η_1 (or η_{10}) is used and is defined as "the viscosity of an irradiated fluid at the temperature, T_1 (or T_{10}), at which the viscosity of the starting material was one centistoke (cs) (or 10 cs)." This index is an

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attempt to establish a common comparative basis for fluids of widely differing viscosities. By this means, the initial viscosity of each fluid begins at one cs; and viscosity change can be measured in terms of deviation from this common point.

A second index of damage used is that of gas evolution and is expressed as "ml of gas generated per ml of fluid charged." This value is determined by measurements of the pressure of the gas generated, as described previously in Section 2.12. Although the significance of gas generation is perhaps not as great as that attached to viscosity change, it is of importance to those concerned with systems in which gas handling is a problem.

The third index of damage used is the change in purity of the fluid on irradiation. This is determined by infrared and mass spectroscopy on original and irradiated materials. This value is helpful in situations in which high polymers, lacquers, or gum-like materials must be replaced via a purification system. However, purity values have no simple relation to viscosity change or gas evolution, and it must be borne in mind that mere change in purity does not necessarily reflect decreased usability.

2.15 Results of Irradiations

2.151 General

During this report period, approximately 16 series of samples of organic fluids were exposed to combined reactor flux under a variety of conditions. A summary of these exposures is presented in Table 2.2. This table summarizes, by series, the general information as to the purpose of the exposures and the irradiation conditions which prevailed. In like fashion, data on the 210 samples exposed to gamma radiation are summarized in Table 2.2 by series. Complete information on individual samples is given in Table 2.3 (four pages). The discussion to follow deals with the purpose of each series of samples exposed.

Series 17, 18, and 19 were exposed in ORNL Hole 19, MTR VG-51, and MTR VH-3, respectively, for a dosage of $N^1 = 2.4$, 2.0 , and 0.74 , respectively. Magnesium containers of Types C-31, C-18.5, and C-17 (Figure 2.0) were used with an initial one atmosphere of helium. Other exposure data are tabulated on Page 1 of Table 2.3.

Each series comprises identical samples of 15 typical organic fluids, i.e., ethers, esters, and alkylaromatics, some with and some without chemical additives. The purpose of these exposures was to determine the relative effect of the fast neutron, thermal neutron, and gamma photon components of combined flux (Series G-2, consisting of the same samples of fluids, was exposed to pure gamma flux to the approximate dosage of the gamma portion of the ORNL exposure). Results will be discussed in Section 2.3.

¹ Dosage $\times 10^{-18}$, expressed as thermal neutrons/cm²

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Series 20, 21, 22, and 23 were exposed in the Brookhaven Reactor in 410 stainless steel containers (C-12) sealed under helium. The purpose of these series was to note the effects of prolonged dosage at elevated temperatures. It had been intended to expose two series at 315°C (600°F) and two at 371°C (700°F), one pair for half the final dosage. However, malfunctions of heating elements precluded this exposure; and conditions which actually prevailed are noted, as follows:

Series 20	170°C (338°F)	N = 7.9
21	315	7.5
22	170	7.8
23	371	7.6

Samples of the same materials were exposed in all four series. These materials were all aromatics, some polynuclear aromatics which would be considered for use as moderator-coolants; others alkylaromatics with and without additives. Data are summarized in Table 2.3, and results are discussed in Section 2.152.

Series 24 was irradiated in the Brookhaven Reactor in 410 stainless steel vessels of Type C-12, sealed under helium. Materials used in Series 20 through 23 were also included herein, with the exception that an alkylaromatic with various additives was eliminated, and three silanes were included. The purpose of this series was to extend the study of temperature effects to 426°C (800°F). Thermal nvt was approximately 2.67×10^{18} . Data are summarized in Table 2.3, and results are discussed in Section 2.152.

Series 25, consisting of 18 samples, was exposed in the Brookhaven Reactor to a dosage of $N \sim 2.29$ at 170°C. Magnesium capsules of the C-12.2 type, sealed under helium, were used. The results are tabulated on page 3 of Table 2.3. The purpose was to note the radiation stability of representative high hydrogen content amines and alcohols. This is discussed under Section 3.3 on shielding materials. The second purpose was to observe the behavior of certain polymer thickeners and additives in an alkylbenzene. These results are discussed in Section 2.153.

Series 26 was exposed in the Brookhaven Reactor at 371°C (700°F) for a dosage of $N \sim 2.78$. Capsules of 410 stainless steel of the C-22 type were used, sealed under helium (see Table 2.3, page 4). The materials studied were all polynuclear aromatics. The purpose of this set of exposures was to provide larger samples (approximately 30 ml each) for subsequent fractionation and analysis, such as by distillation and infrared spectrometry, respectively. Analyses of this series are still in progress. A simple one-stage molecular distillation has been conducted on all the samples where sufficient material was recovered from the irradiation. At the present time, the data are perhaps more pertinent to the applied program and, hence, are discussed in Section 3.12 on Radiation Stability of Moderator-Coolants. It is hoped that detailed analyses, to be available and reported in the future, will contribute to the basic understanding of the relative stability and the mechanism of deterioration upon irradiation of organic fluids.

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Series 27 samples (Table 2.3, page 4) were exposed in the MTR hydraulic rabbit (VH-3) in magnesium containers of the C-17 type, sealed under helium. Exposure temperature was about 30°C, and thermal nvt's progressed to about 4.1×10^{18} . The purpose of this series was to determine the effect of increasing dosage on the thickening of n-butylbenzene. It had been intended to extend these exposures to an nvt of 10^{19} . However, difficulty in obtaining stated high fluxes reduced the actual exposures by a factor of two. Results of these exposures are discussed in Section 2.154 under the Effect of Flux Level.

Series 28 is an extension of Series 19 relating to the effect of flux components. These exposures were conducted in the MTR hydraulic rabbit to a thermal nvt of about 2×10^{18} for subsequent comparison with Series 17 and 18. Exposure has been completed on these samples, and evaluations are in progress.

Series 29 is an extension of Series 27 exposed in the MTR hydraulic rabbit. Samples of n-butylbenzene were irradiated progressively to a thermal nvt of about 10^{19} . Exposures have been completed on these samples, and evaluations are in progress.

Series 30, 31, and 32 were exposed in the Brookhaven Reactor at temperatures of 385°C (725°F), 399°C (750°F), and 413°C (775°F), respectively. Capsules of 410 stainless steel of the C-22 type, sealed under helium, were used. The purpose of this set of exposures was to determine the highest practical temperature that organics might withstand for a dosage of $N \approx 2.8$. If, as discussed in Section 2.153, 426°C (800°F) is too severe, and 371°C (700°F) is still satisfactory, it is expected that these intermediate temperatures will further define the limit of usefulness of organics. Materials contained in these series are those which were promising in the exposures of Series 20 through 26. In addition, several silanes, furnished by personnel of the National Bureau of Standards, were included. The identity of these samples is noted in Table 2.4. Exposures were completed on October 7, 1953, and evaluations are in progress.

2.152 Effect of Temperature and Dosage

Significant data from Series 20, 21, 22, and 23, devoted to this subject, are noted in Table 2.3A (page 12). This information is summarized from pages 2 and 3 of Table 2.3. At the pile ambient temperature of approximately 170°C, the indexes of damage for Series 20 and Series 22 agree within reasonable limits. For Series 21 at 315°C, the η_1 's are generally higher. Of the ten materials noted in Table 2.3A, five have higher η_1 's at 315°C, two have lower η_1 's, and three are indeterminate, thus showing a general tendency toward greater viscosity increase with increasing temperature of irradiation. It is interesting to note that the didodecyl selenide inhibitor appears to have no stabilizing effect in the biphenyl, either at 170°C or at 315°C with the high dosage condition which prevailed. The additives used in amylbiphenyl (noted in Table 2.3) were also of dubious value under these conditions.

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TABLE 2.3A

THE EFFECT OF TEMPERATURE ON RADIATION DAMAGE

Series	S-20	S-22	S-21	S-23
Temperature, °C	170	170	315	371
Thermal n/cm ² x 10 ⁻¹⁸	7.9	7.8	7.5	7.6
<u>Material</u>	<u>Index of Damage, η_1</u>			
Biphenyl	7.3	11	27	16
Biphenyl + 5% O-1*	7.5	11	36	B**
Phenanthrene	5.7	†	8.5	†
3-Methylbiphenyl	Lost	18	37	B
Amylbiphenyl	16.0	B	B	B
Dowtherm "A"	24.0	31	Solid	B
Naphthalene	10	13	Lost	B
1,2-Diphenylbenzene	6	8	3.5	Coke
1-Phenylnaphthalene	8	12	8	Black Tar
Phenylcyclohexane	150	B	Lost	B

*O-1 = Didodecyl Selenide

**B = Safety disc ruptured

†Not included

Series 23 at 371°C was definitely more severe than the others. Of the 18 materials irradiated, only biphenyl displayed any degree of fluidity at 250°F (121°C). The η_1 for biphenyl was actually lower than for the 315°C exposure. Thus, these exposures also show that increasing temperature accelerates thickening generally. However, the dosage level is of significance in the latter effect because, as will be shown in Section 3.122, temperatures up to 371°C have no accelerating effect on the thickening of these organics at lower dosages of $N \approx 3$. At the higher dosages, the fluids are very close to the coking stage and are apparently more susceptible to the relatively small thermal effects. At the lower dosages, such effects are evidently small compared to those from radiation.

Certain of the materials used in Series 20 through 23, namely, chrysene and pyrene, have melting points too high for convenient viscosity determinations. The melting points on both of these decreased on irradiation. For chrysene, the decrease was about 50°C for all the exposures; while for pyrene, the decrease was about 30°C. The lowered melting points reflect mixtures of irradiation products. Chrysene showed particular promise in Series 23 where it was not coked as was pyrene and where only one other organic, biphenyl, was of interest.

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2.153 Effect of Chemical Components

The relative severity of the three exposures in Series 17, 18, and 19, in order of decreasing damage in terms of η_1 , is 17, 19, and 18. The degree of thickening experienced in Series 17 is termed moderate. At this dosage, uninhibited ethers, esters, and long chain alkylaromatics are either past their limit of usefulness or approaching marginal conditions. Chemical additives improve the radiation stability of these materials to the point where many of them can still be used. Biphenyl, the most stable organic irradiated in these series, suffered negligible damage in these instances.

The behavior of the poly(propene oxide)-C (capped with a phenyl group on one end and an ester group on the other end of the chain) is rather unusual. Without additives, this fluid thickened to a near solid. With additives, the material decreased in viscosity in all three series. Chemical additives were also effective in reducing thickening of octadecylbenzene and diethyl phthalate. The sebacate would have become solid at this dosage if exposed uninhibited. Chemical additives prolonged the usefulness and enhanced radiation stability of this ester in these exposures.

Of particular note is the behavior of the inhibited cetane. This straight chain hydrocarbon (hexadecane) is, next to the silicones and aliphatic esters, the least radiation stable of the chemical structures. However, in Series 17, 18, and 19, its comparative damage is amongst the least of all fluids exposed. This is attributed to the excellent inhibitor response of this fluid to the particular additives selected. It is a rather striking example of the protection afforded less stable materials by selected chemical additives.

The behavior of polymer thickeners studied in Series 25 is important in the lubricant development program to be reported at a later date (Report No. 7). Some of these materials depolymerize upon irradiation, while others tend to increase in polymer length and viscosity. It will later be shown that the behavior of poly(lauryl methacrylate) and polystyrene differ in this manner. The latter material contained in an alkylbenzene underwent a gradual thickening upon irradiation; whereas, the methacrylate depolymerized initially and subsequently thickened. The addition of 2% iodonaphthalene to the polystyrene-containing fluid resulted in a considerably lower η_1 . This indicates that the iodine-containing material, in some manner, promoted depolymerization of the polystyrene.

Of the additives tried in the alkylbenzene, neither the selenide-phenothiazine combination nor the dithiophosphate was as effective as the selenide by itself. The new material, dibenzothiophene, with an η_{10} of 25, warrants further investigation; the triphenylmethylsilane, with an η_{10} of 49, is less interesting.

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Although it was not the primary purpose of Series 26 to intercompare fluids, it should be noted that biphenyl, methylbiphenyl, naphthalene, phenylnaphthalene, and 1,2-diphenylbenzene evidenced comparatively small viscosity increases. The triphenylmethylsilane evidenced a considerably lower η_{10} in Series 26 than under the milder conditions of Series 25. This suggests that the Series 26 exposure promoted liquid breakdown products of this material. Acenaphthene underwent a considerable viscosity increase and is not considered particularly promising.

2.154 Effect of Flux Level and Dosage

It is important to know how viscosity change of an organic fluid varies with radiation dosage. Likewise, the effect on viscosity change of the rate at which a given amount of radiation is applied (flux level) is important. Both of these variables were investigated in experiments in which samples of n-butylbenzene were exposed under similar conditions in the Brookhaven Reactor and in the MTR VH-3 facility. Samples were removed periodically in these experiments and viscosities measured to ascertain the increase in viscosity with increasing dosage. The results of these two series of experiments are illustrated in Figure 2.6.

Both exposures were conducted under helium. The Brookhaven work was in aluminum, and pressure generated was bled off; whereas, magnesium vessels were used in the MTR work, and pressure was allowed to build up. The Brookhaven exposures were at about 140°C, and the MTR exposures were at about 30°C. Past work has shown this temperature differential to have a negligible effect on viscosity change. Also, the container material and pressure differences should be inconsequential. The overriding differences in the two sets of data are twofold: The flux level which, in terms of thermal neutron flux, was higher by a factor of 55 at the MTR; and the ratio of fast flux to thermal flux, which was 22% at Brookhaven and about 7% at the MTR.

Four sets of curves are shown in Figure 2.6 to compare the thickening of n-butylbenzene exposed in the two facilities. These are plots of the usual function of viscosity in terms of $\log \eta_1$ against dosage and are actually the same data presented in different ways. The first set of curves portrays the variation of η_1 as a function of thermal neutron dosage. The two curves are actually fairly close; and, at a dosage of 2.8, the deviation is about 22%. A higher slope results for the data from Brookhaven, indicating one of two possibilities: (1) that for a given dosage, expressed as thermal neutrons, the higher proportion of fast and possibly epithermal neutrons at Brookhaven results in greater thickening; or (2) that the flux level at the MTR represents a less stringent exposure.

The second set of curves shows the same indexes of damage plotted in terms of fast neutron dosage. In this case, the slope of the MTR data is considerably higher than that of

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the Brookhaven results. This would appear to indicate that the higher flux level prevailing at the MTR results in greater thickening to this organic fluid. These curves are misleading inasmuch as total damage is plotted against partial dosage; that is, only the fast neutron dosage is considered, and thermal and epithermal neutrons and gammas are ignored. The third set of curves shows the same indexes of damage plotted in terms of gamma dosage. It should be noted that the values of gamma flux used were 30% of thermal neutrons for Brookhaven and 90% for the MTR position. This representation is not valid by the same consideration noted above, namely, that total index of damage is plotted against only a portion of total dosage.

The fourth set of curves is an attempt to reconcile the previous two representations by plotting $\log \eta_1$ against an equivalent fast neutron dosage. This equivalency is based on the relationship that a 1.6 mev gamma has approximately 1/16 the damaging effect (as indicated by fluid thickening) as does a fast neutron whose energy is greater than ~ 1 mev. The development of this relationship is presented in Section 2.3 of this report. The various assumptions and calculations pertaining to its development are treated in full at that point. Thermal neutrons are neglected because it will be shown that their effect is negligible. As portrayed in the last curves, the exposures in the MTR reflect a higher slope than those of the Brookhaven radiations, indicating a greater rate of thickening or damage in the former. At the equivalent dosage of 0.5, this separation of the two curves is approximately 50%. This difference is large enough to be significant and may be due to one or more of three factors: (1) that the higher flux level prevailing in the MTR exposures increases the rate of thickening of organic fluids, (2) the ratio of epithermal to fast neutron flux at the MTR is much greater than that at Brookhaven, or (3) that the assumptions and experimental uncertainties extant in developing the dosage equivalency are large enough to encompass this degree of error.

No measurements of epithermal flux are available for the MTR and Brookhaven facilities used in this work. Monitoring operations are under way to gather such data. These should allow a fuller treatment of the effect of flux level and radiation dosage on viscosity change.

2.16 Work in Progress

Exposures have been completed on Series 28, relating to the effect of flux level in the MTR, and on Series 29, also an MTR exposure, relating to the effect of dosage. These samples have been received at Richmond but have not as yet been evaluated. Series 30, 31, and 32, exposed in the Brookhaven Reactor at 385°C, 399°C, and 413°C, respectively, were removed from the reactor on October 7. Evaluations on these samples are also in progress.

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2.2 Research with Gamma Rays

2.21 Experimental Approach

This study of effects on organics of pure gamma rays was undertaken for two primary reasons. With respect to the basic program, it is important to separate the effects of the components of combined radiations. Of these, gamma rays are most readily obtained in a pure state. From a practical viewpoint, many uses of organic fluids involve exposure to gamma irradiation only. Some such uses are (1) radiation shielding, (2) secondary heat transfer fluids, and (3) lubricants.

In keeping with the stated purposes of the work, materials irradiated fall into three groups: (1) those being studied in other work with combined radiations, (2) those of particular interest as shielding materials (which require high N_H), and (3) alkylaromatic solutions of polymers. The last group is being studied with a view toward eventual application in lubricating oil and grease formulations. The criteria of stability in this work are the same as previously discussed for combined radiations (Section 2.14).

The gamma irradiations have been conducted at two locations. For low dosage studies, the California Research Co^{co} source has been used. A recent revision of the values supplied by BNL places this source at nominally 600 curies and about 1.7×10^5 R per hour. High dosage level irradiations have been carried out in the MTR gamma facility, which runs up to $\sim 3 \times 10^7$ R per hour. The advantage of the one hundredfold increase in flux available at the MTR is obvious, but the use of both sources has enabled a more complete picture of dosage effects to be ascertained in certain cases of special interest. An example of the latter is in the study of polymer thickeners in alkylaromatic solutions.

2.22 Equipment

The capsules used for irradiations have been adequately described in Section 2.12. Magnesium capsules were used at the MTR, either sealed in helium or open to the air, as required. In the California Research Co^{co} source, open 1020 steel cells were used for Series G-1 and 4. All later series have been run in pyrex cells having an individual capacity of 10 cc. Sixteen of these cells were irradiated at one time. The MTR gamma facility, which consists of discharged fuel elements surrounding an irradiation zone, is situated under 20 feet of water and, thus, requires water-tight casings. Casings of 1020 steel were used for Series G-2 and G-3. However, appreciable corrosion occurred; and, for subsequent series, aluminum casings were used. The latter are fitted with a solid aluminum extension which aligns the samples automatically in the centerline of the fuel assembly and also serves to sink the casing.

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2.23 Dosimetry

For the California Research Co⁶⁰ (Brookhaven tube type) source, the flux data have been used as supplied by BNL (almost uniformly 1.69×10^5 R per hour along the axis of the cylinder). For the irradiations at the MTR, all dosages were calculated from chemical dosimetry (ceric-cerous) measurements performed by the MTR personnel. The method is adequately described in the literature.¹

Two problems arise with respect to the calculation of dosages in the MTR gamma facility. The active length of the fuel elements is about 24 inches. The flux varies with the vertical traverse of the fuel element by as much as fourfold from the center to the ends. In Series G-3 and subsequent series, only 12 inches of the total active space has been utilized, six inches on either side of the center. By this means, the maximum difference in flux between the center and extreme ends of the sample casing is reduced to about 10%.

The second problem in the dosage calculation is that of accounting for the decay of the activity of the fuel elements with time². The total dosage must be obtained by integrating the activity over a period of time. This calculation is relatively simple for fuel elements of known and identical age. The present practice of mixing elements of differing ages at the MTR gamma facility reduces the mathematics to a form which cannot be readily solved. This problem can be surmounted only by taking more frequent dosimetry measurements which will allow a graphical integration. The over-all accuracy in the gamma dosages thus obtained is probably about $\pm 20\%$.

2.24 Results of Irradiation

2.241 General

During this report period, ten series of organic samples were exposed to gamma radiation. These exposures have been designated "G" series, each having a particular purpose. This purpose, together with pertinent results of each series of samples, is summarized in the following paragraphs. Table 2.2 gives pertinent exposure aspects for each gamma series.

Series G-1 comprised 16 samples of conventional and special lubricants. These were exposed in the California Research Co⁶⁰ gamma source for a dosage of 5.1×10^6 R. The samples included seven greases and nine oils. Evaluations are complete on these lubricants, and their presentation will be made in CRC-AEC Report No. 7 which is to be devoted to lubricants.

¹ J. Weiss, "Nucleonics," 10, 28-31, 1951

² T. R. Wilson, Jr., IDO-16076-PPCo.

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Series G-2 (CRC-501-1) consisted of 15 representative samples of different types of organic fluids which were identical to those in Series ("S") 17, 18, and 19. The purpose of this exposure was to explore the effects of the components of combined flux by establishing the damage due to pure gamma rays. The application of this is described in Section 2.3. The Series G-2 samples were irradiated in the MTR gamma facility for approximate dosages of 10^9 R. In Table 2.5 are shown the results of these exposures in terms of gas formed and viscosity increase. On an over-all basis, the damage suffered by these fluids was minor; i.e., viscosity increases were, in general, less than 25%, and gassing was moderate. An exception was the poly(propene oxide)-C, which thickened excessively when uninhibited. By contrast, the addition of additives caused this fluid actually to decrease in viscosity. The esters were the next least resistant as a class, evidencing the greatest thickening and gas evolution. Additives appreciably increased the stability of the esters. Biphenyl displayed no appreciable damage. Of particular interest is inhibited cetane (hexadecane). Although gassing was quite high, viscosity increase was very low. It would be expected that this material, uninhibited, would become very viscous.

Series G-3 (CRC-501-2) included the same materials as Series G-2 and was irradiated in the MTR facility to a dosage of from 4 to 10×10^9 R. This exposure can be termed moderately severe; i.e., uninhibited esters and mineral oils were no longer suitable for use. The isobutyl and n-butyl hexanoates and the diethyl phthalate became solids as did an uninhibited octadecylbenzene and a poly(propene oxide). Inhibitors reduced both thickening and gassing in the latter two materials. More resistant structures, such as an s-butylbenzene, displayed moderate thickening and moderate gassing; while biphenyl displayed negligible gas formation and a slight increase in 210°F viscosity.

This gamma dosage is marginal for the less resistant structures, such as esters and mineral oils, even with the incorporation of chemical additives. Aromatic materials, specially formulated with selected additives, would be recommended for use at this dosage. The samples presented in Table 2.5 constitute but half of those exposed in Series G-3. The other 18 samples are still being evaluated. They comprise a range of fluids including several special lubricants and certain potential shielding materials.

Series G-4 samples were irradiated in the California Research Co⁵⁰ gamma source progressively to maximum dosages of 4×10^9 R. Exposures were made in 1020 steel containers open to air. This series involved a study of the stability of polymeric thickeners in low viscosity alkylaromatic fluids. The base material chosen in this work, of interest in lubricant development, was a mixed alkylbenzene (MW ~ 250), inhibited with a dialkyl selenide and quinizarin for optimum radiation resistance. Polymer thickening agents used included a polyester, a polybutene, a poly(alpha-methylstyrene), and a butene-styrene copolymer.

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Samples of thickened oils were exposed for varying lengths of time so that the gradual change in viscosity as a function of gamma dosage could be determined. The data from this series are included in Table 2.6 and are discussed in Section 2.243.

Series G-5 and G-6 (CRC-501-3 and 501-4) are an extension of the work begun in Series G-4. Samples of the same formulations were exposed in the MTR gamma facility for dosages of approximately 10^9 and 10^{10} R, respectively. These exposures were made in magnesium containers sealed under helium. Although not strictly comparable to Series G-4, due to the container difference, these exposures represent dosages higher by a factor of 2-1/2 and 25, respectively, compared with Series G-4. Exposures have been completed on both of these series, but evaluations have not been completed. The identities of the samples are presented in Table 2.7.

Series G-7 (CRC-501-5) includes 15 samples having the same identities as samples of Series G-2 and G-3. The purpose of this exposure was to obtain an intermediate dosage (5×10^9 R) between that of the two previous series. As already noted, the dosage of $\sim 10^9$ R in Series G-2 was comparatively mild; whereas, the dosage of 10^{10} R in Series G-3 was moderately severe. The exposure has been completed on this series, and evaluations are in progress. The identities of the samples are tabulated in Table 2.7.

Series G-8 and G-9 (CRC-501-6 and 501-7) each consist of 16 samples of special radiation resistant lubricants, six of which are greases and ten of which are fluids. The fluid formulations are based on alkylaromatic base stocks, thickened to the desired viscosity with polymers studied in Series G-4. The greases were formulated from similar fluids and were thickened to the grease consistency by the use of silica. The purpose of these two series was to observe the radiation resistance of these specially formulated lubricants. Dosages for both series are $\sim 5 \times 10^9$ R. These irradiations are identical, except that Series G-9 was open to the air; whereas, Series G-8 samples were sealed under helium. This arrangement will provide an observation as to how air affects these new materials. Exposures on these samples have been completed, evaluations are in progress, and results will be reported in CRC-AEC Report No. 7.

Series G-10, consisting of 24 samples, was exposed in the California Research Co.[®] gamma source in pyrex containers open to the air. An additional thickener, that is, a poly-(alkene oxide), was studied in the same fashion as the samples of Series G-4. A special fluid, comprising 50% poly(propene oxide)-C and 50% mixed alkylbenzenes (MW ~ 350) with the usual inhibitors, was also exposed in the same manner. The purpose of this particular fluid combination was to observe the results of blending two fluids, one of which tends to decrease and the other of which tends to increase in viscosity upon irradiation. Exposures were made progressively to about 2×10^9 R. The results on these two fluids are contained in Table 2.6.

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Several of the radiation resistant lubricants noted in Series G-8 and G-9 were also included in Series G-10. The purpose was to obtain an additional point at a lower dosage ($\sim 2 \times 10^8$ R) than that which would be obtained in the MTR exposures. These results have not yet been evaluated and also will be presented in the forthcoming Report No. 7.

2.242 The Effect of Dosage

An approximation of the effect of gamma dosage on the thickening of an organic fluid is portrayed in Figure 2.7. The data for the three fluids shown are taken from Table 2.5 which presents the complete results for Series G-2 and G-3. The curves of Figure 2.7 are considered typical of the over-all results. The origin in every case is connected to the outer points. Straight line plots are, thus, assumed because only two points are available. However, from past work, the assumption appears to be a reasonable one; large deviations from linearity would not be expected.

As expected, the ester shows the highest slope, hence, the least radiation resistance. The ether is intermediate; and the alkylaromatic has a markedly lower slope, hence, greatest radiation stability. The pattern shown in Figure 2.7 closely follows that previously established for exposures to combined radiations. It is evident from Figure 2.7 that a gamma dosage of approximately 10^{10} R (10^{12} gammas/cm²) represents a moderately severe exposure.

2.243 Study of Thickeners

In Figure 2.8 are plotted the results of the G-4 and G-10 exposures in the Co⁶⁰ gamma source. This figure shows the relationship of viscosity change to gamma dosage for the several fluids containing polymer thickeners. It will be noted that behavior varied considerably. The samples containing poly(lauryl methacrylate) and the poly(alkene oxide) evidenced an almost immediate decrease in viscosity. This is attributed to a breakdown in the high molecular weight polymer. In the case of the poly(lauryl methacrylate), a minimum point was reached at a dosage of $\sim 3.1 \times 10^8$ R. At this dosage, it appears that depolymerization has progressed to a saturation point; and from then on, over-all thickening of the fluid mixture will occur.

Samples containing the polybutene and the styrene-butene copolymer displayed a marked viscosity decrease also, although this is less pronounced than for the two materials already noted. The poly(alpha-methylstyrene) sample displayed a slight viscosity increase as almost a straight line function. The possibilities are at once apparent that by combining this polystyrene and, for example, the polybutene, a fluid having essentially constant viscosity with increasing dosage would result. Combinations of this sort have been exposed in other series, although results are not available as yet.

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The formulation denoted as Fluid "X" displayed this tendency of constant viscosity. This fluid comprises a poly-(propene oxide) and an alkylbenzene which show divergent trends of thickening with dosage. For a dosage of $\sim 1.6 \times 10^8$ R, viscosity increase was essentially nil for this fluid. At this juncture, the curve appears to drop sharply, although additional points are needed for further definition.

The use of polymer thickeners is a promising means of formulating higher viscosity lubricants and other materials from comparatively low viscosity alkylaromatic base stocks. The results shown in Figure 2.8 indicate that such an approach is feasible from a standpoint of radiation resistance. It is evident, of course, that the thickener itself must possess a certain degree of radiation stability so that the higher viscosity is not immediately lost due to depolymerization. Of the materials thus far tried, the poly(alpha-methylstyrene) is the most promising.

2.25 Work in Progress

Although exposures have been completed on all of the samples contained in Series G-1 through G-10, evaluations are in various degrees of completion. Specifically, Series G-3 is half completed; Series G-5, G-6, G-7, G-8, and G-9 are still in the process of evaluation. Due to the present state of completion, several of the objectives of the gamma work have only partially been attained. A reassessment of these objectives must await the complete evaluation of the exposure presently under way.

2.3 Components of Combined Radiations

2.31 Qualitative Evaluation

In the assessment of damage to organics by components of combined radiations, it is not practical or possible to expose appreciable quantities of organic fluids to significantly high pure thermal, epithermal, or fast neutron fluxes. Thus, other means had to be used. The approach was to expose series of samples to four different conditions in which the ratios of the individual components of radiation varied. In this manner, the relative effect of each component was estimated. The four sets of exposures involved follow:

- a. Series G-2 exposed to pure gamma flux from the MTR gamma facility.
- b. Series 17 exposed to combined radiations from the ORNL reactor for a dosage of $N \approx 2.4$.
- c. Series 18 exposed to the same approximate thermal neutron dosage in the MTR VG-51 facility in which the fast neutron flux is comparatively low.
- d. Series 19 exposed in the MTR VH-3 facility where a higher fast to thermal neutron ratio exists than in (b). This exposure was to have been to a

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thermal neutron dosage equivalent to that in (b) and (c), but less than half the intended dosage was actually received. This experiment is being extended to the desired dosage.

TABLE 2.8

COMPONENTS OF COMBINED RADIATIONS
AT VARIOUS FACILITIES

	Series 17 ORNL Hole 19	Series 19 MTR VH-3	Series G-2 MTR Gamma Facility	Series 18 MTR VG-51
Thermal Neutrons/ $\text{cm}^2 \times 10^{18}$	2.4	0.74	0	2.0
Fast Neutrons/ $\text{cm}^2 \times 10^{18}$	0.096	0.052	0	0.008
Gamma: Gamma/ $\text{cm}^2 \times 10^{18}$	1.2	0.66	1.26	0.96
R $\times 10^8$	9.5	5.3	10	7.6
Relative Viscosity Change	2.0	1.5	1.4	1.28

From the prevailing flux conditions, noted previously in Table 2.0, the "best values" for the components of combined dosage have been calculated and are indicated in Table 2.8 for Series 17, 18, and 19. Using viscosity change as a criterion, the severity of the exposure decreases from left to right in Table 2.8. This indicates that the ORNL exposure was the most severe, followed by the MTR VH-3, the MTR gamma, and the MTR VG-51, in that order. The damage undergone by the organic fluids in the ORNL irradiation would be considered moderate; i.e., an uninhibited poly(propene oxide)-C turned solid. More resistant materials, such as biphenyl, were relatively unaffected. Under these conditions, paraffinic materials, such as conventional lubricating oils, would be at or past their limit of usefulness in the absence of suitable additives. The damage suffered by Series 18, the least affected of the four, is considered negligible; i.e., viscosity increases range up to about 25% maximum.

The thermal neutron and gamma dosages are about 25% higher for the ORNL Series 17 than for the MTR Series 18. However, the main difference in exposure is in the fast neutron dosage, which is higher by a factor of 12 for the Series 17 samples. Values of the index of damage, η_1 , for these samples are approximately 70% greater than those for the Series 18 samples. This indicates that the fast neutrons contributed most to the fluid deterioration because the other components are present in preponderance.

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In comparing the MTR gamma Series G-2 with the MTR Reactor Series 18, a slightly higher (of the order of 15%) deterioration was noted for the gamma samples. For these samples, the pure gamma dosage is approximately 25% higher than the gamma component of the pile exposure. The dosage of 2×10^{18} thermal neutrons/cm² (and about 10^{16} fast neutrons/cm²) in the Series 18 exposure then appears to have had negligible thickening effect. This establishes gamma photons as important contributors to thickening of organic fluids and the thermal neutrons as negligible. It is noteworthy that it also appears that a fast neutron dosage of 10^{16} is a negligible radiation dosage. This roughly establishes a tolerance level below which conventional materials can be used in ionizing atmospheres. It must be remembered that these conclusions are drawn from exposures in magnesium containers in the absence of air.

2.32 Quantitative Evaluation

Thus far, the basis has been established for assuming that thermal neutrons exert a negligible thickening effect on organic fluids and that fast neutrons and gamma photons are the principal damaging agents. In attempting to separate the effects of the latter two components, the thickening due to pure gamma exposure in Series G-2 should first be considered. If a straight line relationship is assumed between the logarithm of viscosity index of damage, η_1 , and gamma dosage, a proportionate damage due to the gamma portion of the combined flux in Series 17, 18, and 19 can be interpolated. This interpolation is reasonable, as previously demonstrated in Figure 2.7. If the gamma portion of η_1 is subtracted from the total increment of damage, that which remains must be an assessment of the thickening due to the action of fast neutrons (including epithermal). This procedure has been used, and the data are portrayed in Table 2.9. These result finally in the determination of the ratio of damage by fast neutrons to that by gammas.

The average values of these ratios in Table 2.9 are as follows: 2.81 for Series 17, 2.53 for Series 19, and 0.26 for Series 18. These figures then must be adjusted for the relative flux of fast neutrons and gammas in each case. The ratios used are as follows: For Series 17, fast neutrons and gamma rays as 4% and 50% thermal flux, respectively; for Series 19, 7% and 90%, respectively; for Series 18, 0.4% and 56%, respectively. Multiplying by the proper ratio of gammas to fast neutrons yields the following figures: 34 for Series 17, 33 for Series 19, and 31 for Series 18. This ratio relates the apparent damaging effect of one fast neutron to that of one gamma photon. It is fortuitous that the three values agree so closely (especially for Series 18 where the individual values vary markedly).

In arriving at these ratios, it must be pointed out that the portion of damage ascribed to fast neutrons was actually caused by all neutrons above thermal energy. However, only fast neutrons (as determined by Np²³⁷ monitors) were taken into

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account in the dosage determinations. This leaves unaccounted for the epithermal neutrons, that is, those whose energies are below 1 mev but greater than thermal. In Section 2.43, it is noted that (in Hole 12, ORNL) epithermal neutrons below 1 mev are roughly equivalent to the aggregate of fast neutrons above 1 mev in terms of damaging effect on organic fluids. While this relationship was determined for a specific position in a specific reactor, it is assumed to apply to the data at hand for lack of further information. With such an assumption, the previous ratio should be reduced by a factor of about two. This results in 16 as the ratio of damage of one fast neutron to one gamma photon. It also should be noted that this is an average value and, as such, may not represent any particular fluid. In fact, the scatter of the individual ratios indicates that the relationship varies as a function of structure.

This ratio is in terms of the incident flux, that is, the impinging particles or rays. It is believed that a ratio in these terms has significance from both a practical and theoretical standpoint. Practical inquiries, for example, as received by California Research concerning the use of organic fluids, usually specify the radiation condition only in terms of flux. From a theoretical viewpoint, the fluid suffers damage by virtue of energy absorption. The difficulty of expressing the results of radiation damage studies in terms of energy absorption, and the relative merit of expressing such data in terms of incident flux, will be discussed in the Section 2.4, to follow.

2.4 The Relation of Incident Flux and Energy Absorption to Radiation Damage

2.41 Introduction

In the study of radiation damage in reactors, the most commonly employed measure of the amount of radiation received is simply the total incident flux. In fact, most often only the thermal flux is given; even though, in the majority of instances, (e.g., in the study of organic fluids) the damage is attributed to the action of only gamma rays and the neutrons above thermal energy. This can be done because, in a fixed position in a given reactor, the gamma and fast neutron fluxes are proportional to the thermal flux. In the present discussion, only the fast (and resonance or epithermal) neutron and gamma ray components of the reactor flux and their effects upon organic fluids shall be considered.

The question arises as to whether the results of radiation damage are better expressed in terms of actual energy absorbed rather than incident flux. This problem is of interest in two situations: first, in the translation of radiation damage studies from one reactor medium to another; and, second, in the separation of the individual effects of radiation components. The choice of a representation of dosage involves the considerations

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that incident flux is the easiest to determine, while energy absorbed reduces all radiation components to a common base. Some evidence is available¹ to indicate that for equal amounts of energy absorbed, damage is the same for any type of radiation. The value of a relationship such as this would be undisputed; however, there will be developed in the following discussion a basis for the preferential use of incident flux in studies, such as the present one, on the radiation damage to organic fluids by combined reactor flux. First to be considered are some of the theoretical and experimental requirements of the procedure for determining the energy absorption by materials exposed to reactor radiations.

2.42 Reactor Flux Energy Dissipation

The problem of determining the absorption of energy from neutrons can be illustrated by the method given by Richardson². The total energy transfer on neutron scattering is given by the relationship:

$$Q_n = N_s K \int_0^{\infty} (nv)_u \sigma_s dE \quad (1)$$

where N_s = number of scattering atoms

K = average fraction of kinetic energy lost per collision; ($K = 2A/(A + 1)^2$, where A = atomic weight of the scattering atom)

$(nv)_u$ = neutron flux with energy between E and $E + dE$

σ_s = scattering cross-section per atom.

Q_n is then evaluated as follows:

- (a) values of σ_s as a function of energy, E , are obtained; these are readily available from various sources³;
- (b) a neutron energy spectrum, i.e., values of $(nv)_u$ as a function of (E) must be assumed for the particular position in the pile;
- (c) values of (E) versus the product $KN_s(nv)_u\sigma_s$ are plotted, and the area under the curve then gives Q_n .

¹ Sisman, O., Symposium, "Use of Organic Materials in Nuclear Reactors," North American Aviation, Aug. 1953.

² Richardson, D. M., ORNL-129, "Calorimetric Measurement of Radiation Energy Dissipated by Various Materials Placed in the Oak Ridge Pile," Oct. 1, 1948.

³ "The Science and Engineering of Nuclear Power," Vol. I, Appendix C; or, "Neutron Cross-Sections," AECU-2040, May 15, 1952.

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The point to be noted here is the requirement (b) for the neutron flux versus energy spectrum. To reduce neutron radiation damage data to an energy absorbed basis and to apply this information practically would require that flux versus energy spectra, as a function of spatial distribution, be known for all types of reactors of interest. Such information is not readily available. The calculation of neutron flux as a function of energy is very complex, as given, for example, by Goldberger et al.¹ for a Hanford-type graphite reactor. It should be noted that the latter calculations were made with the assumption that the pile consisted of an infinite square array of line sources, all of equal strength. These conditions are well approximated only near the center of the pile.

Furthermore, at best, only a relative flux versus energy relationship can be calculated. Also, the actual heating effect in a pile is the result of the absorption of both neutrons and gamma rays. As a consequence, in order to determine the true Q_n , calorimetric heating measurements on at least two different substances must be performed at the point in the pile under consideration. Then, the factors of the ratio of the true neutron flux to the assumed flux and the gamma heating contribution can be solved, as done by Richardson².

The calculation of the contribution of gamma rays to energy absorption in the pile should be made in a manner exactly analogous to that for neutrons already described (Equation 1). Calculated flux and energy spectra for pile gamma rays appear to be less readily available than those for neutrons. Certain simplifications in the general behavior of gamma rays, however, lend themselves to the problem; namely, that, for most elements, the ratio of the true energy absorption coefficient is roughly independent of energy in the region around 1 mev. For this reason, the gamma portion of the calorimetric measurements discussed can be expressed simply as the ratio of the heating effects to some "standard." The details of this treatment are not pertinent here. However, for the same reason, it is possible to treat pile gamma ray effects, in general, by merely assuming a reasonable, average value of the energy. Hence, gamma ray energy absorption in a reactor can be calculated satisfactorily from the simple relationship:

$$Q\gamma = E_{av.} \times f_E \times \mu_T \quad (2)$$

where, $Q\gamma$ = energy transfer on gamma scattering [analogous to Q_n , determined by Equation (1)]

$E_{av.}$ = average gamma energy

f_E = apparent total gamma flux (a function of the average energy, $E_{av.}$)

μ_T = true energy absorption coefficient.

¹ Goldberger, N., Stephenson, J., and Weinberg, A. M., CP-2439, "Neutron Flux in W Lattice," Nov. 30, 1944.

² Richardson, D. M., ORNL-129

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It should be recognized, however, that this treatment is an approximation; and the choice of average energy is, to some degree, arbitrary.

It is apparent that the specific treatment of radiation damage, from the viewpoint of energy absorbed, in different nuclear reactors would be a formidable task. The problem would be appreciably simplified only for conditions wherein it could be assumed that the neutron energy spectra are similar. However, in such cases, it would be just as significant and simpler to make comparisons on the basis of incident flux.

2.43 Reactor Incident Flux

The requirements for an adequate basis of representation of the results of radiation damage in terms of incident flux are quite simple to satisfy. The neutron flux in definite ranges of pile energies (i.e., thermal, resonance, and fast) can be readily determined by well known methods of metal foil activation. In the present studies of radiation damage, the neutron flux determination has been restricted to (1) thermal neutron flux, as determined by cobalt activation, and (2) fast flux (greater than 1 mev), as determined by neptunium fission. The individual methods have been described in Section 2.13.

An important omission, however, is the value of the resonance neutron flux (i.e., with energy between thermal and 1 mev). While the thermal neutrons contribute little or nothing to the radiation damage of organic fluids, pile resonance flux should approach the damaging effects due to fast flux. This conclusion is supported by considerations of energy absorption. Richardson¹ gives a plot of values for E versus $KN(nv)_{0.68}$ for various elements from which relative Q_n values were computed as previously discussed (see Equation (1), Section 2.42) for a typical position (Hole 12) in a graphite reactor of the Oak Ridge type. A rough estimate can be made of the relative contributions of the 0 to 1 mev (resonance) neutrons and the 1 to ∞ mev (fast) neutrons to the total Q_n by determination (graphic) of the appropriate areas under these curves. Thus, it is found that the ratio of energy absorbed (or, presumably, also of radiation damage) for the total resonance to the total fast flux is about 1.3 and 1.0 for carbon and hydrogen, respectively. These figures apply, of course, to a particular situation in a graphite reactor where the ratio of fast to resonance flux is about 15. It is apparent then that a complete accounting of the incident neutron flux should include the resonance flux, which can be determined by foil activation (e.g., cadmium ratio measurements on indium foils, as described by Hughes²).

The only method which appears to be available for the determination of pile gamma flux is that which uses gamma heating effects, that is, by use of the relationship of Equation (2) discussed in Section 2.42. Hence, there is little to choose

¹ Richardson, D. M., ORNL-129

² Hughes, D. J., "Pile Neutron Research" (1953) pp 129-134

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between expressing gamma flux in terms of energy absorbed or incident flux. However, the latter is actually the simpler of the two, especially because the determination can be made independent of any neutron heating effects by measuring the gamma heating in a heavy element with low thermal neutron cross-section, such as lead. The very simple calorimetric principle described by Hogg¹ has been used for this purpose and is discussed in Section 2.13.

2.44 Relative Effects of Flux Components

Finally, it is of interest to show that the experimental results obtained in Section 2.32 on the relative radiation damage of fast neutrons and gamma rays, based on incident flux, is consistent with the available information on energy absorption. Richardson² has tabulated, for various elements, values of the energy dissipation of the neutron (total fast plus resonance) and the gamma flux in Hole 12 in the Oak Ridge graphite reactor.

TABLE 2.10

RELATIVE ENERGY ABSORPTION PER
ATOM IN HOLE 12, ORNL*

Element	Gamma	n (total)	n (fast)	n (resonance)
Hydrogen	1.000	9.12	4.0	5.1
Carbon	5.97	1.36	0.69	0.67
Oxygen	7.98	1.18	0.52	0.66

*Relative flux: $\phi = 1.00$, n (fast) = 0.083,
 n (resonance) = 1.27

In Table 2.10, these data have been converted to a relative basis of energy absorption per atom, arbitrarily setting the gamma energy absorption for hydrogen equal to unity. In addition, by using the ratio of the resonance to the fast neutron energy dissipation as determined in Section 2.43, the total neutron effect has been divided up into the resonance (0 to 1 mev) and fast (above 1 mev) components. The values for the relative flux were obtained from the data of Richardson² and Bopp and Sisman³. Now, if radiation damage is a function only of energy absorbed, these data will allow the calculation of the relative damage to any organic fluid caused by the reactor flux components. It is apparent that this will be a function of composition. The apparent ratio of damage

¹ Hogg, C. H., "Gamma Heating Measurements in the MTR," IDO-16093-PPCo.

² Richardson, D. M., ORNL-129

³ Bopp, C. D., and Sisman, O., "The Neutron Flux Spectrum and Fast and Epithermal Flux in Hole No. 19 of the ORNL Reactor," ORNL 525, July 28, 1950

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to correspond with the value of 16 calculated in Table 2.9 from the radiation thickening of organic fluids will be given by the relation:

$$\text{Damage for, } \frac{1 \text{ fast neutron}}{1 \text{ gamma}} = \left(\frac{1}{0.083} \right) \times \frac{4.0 a + 0.69 b + 0.52 c}{1.00 a + 5.97 b + 7.98 c}$$

where a/b/c is the atomic ratio of C/H/O in the compound. Typical values are given in Table 2.11.

TABLE 2.11

RATIO OF n/γ DAMAGE CALCULATED FROM ENERGY ABSORPTION

Compound	C/H/O	"Damage," n (fast)/γ
Decane	0.455/1/0	14.0
Benzene	1/1/0	8.1
Naphthalene	1.25/1/0	6.9
Dibutyl Phthalate	0.728/1/0.182	8.2

Because the average deviation from the mean for the best two series of experiments in Table 2.9, S-17 and S-19, corresponds to about + 9 out of 16, the agreement of the calculated values of Table 2.11 with the value of 16 from thickening data is seen to be entirely reasonable. This is especially so in view of the questionable validity of the two basic assumptions, namely, (1) that η_1 is a linear function of the degree of structural damage and (2) that energy absorption alone determines the extent of damage in organic fluids. With respect to (2), it is pertinent to note that organic structural differences are not taken into account in the calculation of energy absorption. For example, resonance, which certainly has a major effect on radiation stability, is not considered. Energy absorption should thus be inadequate as a basis for radiation damage comparisons of organic compounds of basically different structure.

In conclusion, it appears that, in a broad study of the radiation stability of organic fluids, representation of the results on the basis of incident flux rather than on an energy absorption basis is certainly simpler and more practical and may actually prove to be the more realistic method of comparison of relative stability of fluids and relative damaging power of reactor flux components. Further, it appears feasible to express radiation damage, e.g., viscosity change, as a function of "equivalent fast neutron dosage." This dosage function should be obtained by adding to the fast neutron dosage, much as in the last curve of Figure 2.6, the damage equivalent each of epithermal neutron and gamma dosages.

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3. APPLIED PROGRAM

3.1 Research on Moderator-Coolants

3.11 Thermal Stability

3.111 Introduction

The proposed use of organic compounds as coolants and moderators in nuclear reactors requires that these substances, in the presence of combined radiations, be able to withstand as high temperatures as possible, preferably the order of 538°C (1000°F). Studies of the thermal stability of organic compounds were undertaken in the moderator-coolant development program to establish the maximum stability temperature for organics and to screen promising chemical structures for further study in the presence of radiation. The present work was concerned almost entirely with aromatic hydrocarbons or simple modifications of the same. Some data are available in the literature on the thermal cracking of aromatic hydrocarbons¹; however, the conditions given are too varied to allow adequate intercomparison of stabilities or application to proposed environments.

From purely thermodynamic considerations, all organic compounds are unstable with respect to decomposition into the elements at elevated temperatures. On this basis also, increased instability with increasing molecular weight is predicted. Probably more significant is the fact that the latter is to be expected on statistical grounds². It should be noted, however, that the end product "coke" obtained in the pyrolyses of aromatic hydrocarbons is not carbon but is a highly condensed aromatic³. This was borne out in the present work wherein, even under the most drastic exposures, less than one mole of hydrogen per mole of original compound was evolved. (The theoretical moles of hydrogen per mole of organic is between about four and seven for most of the compounds studied.) These observations suggest that the problem of stability at elevated temperatures is basically a kinetic one. There would appear to be some hope, then, for finding some "magic" structure of maximum stability. However, the available information would lead to the conclusion that the simplest structures probably would be the most stable. Accordingly, the present work was started with the study of the effect of temperature using biphenyl and naphthalene.

3.112 Experimental Method

The 26 compounds studied (three of which were nonaromatics) are listed in Table 3.0 with the sources of supply and the melting points. The best commercial grades were obtained, where available, and used without further purification. The synthesized compounds were purified by crystallization or distillation.

¹ Sachanen, A. N., "Conversion of Petroleum," pp 87-110, Rheinhold (1948)

² Obreimov, I. F., Zhur, Eksptl. Teoret. Fiz. 19, 396-406 (1949); c.f., Chem. Abstr. 46, 9906 f (1952)

³ Hadzi, D., Fuel, 32, No. 1, 112-113 (1953)

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The pyrolyses were carried out in welded stainless steel (Type 304) vessels. A 20-gram charge of organic compound was usual. The loaded bombs were flushed several times with nitrogen to remove air and then were adjusted to an initial pressure of exactly 300 psig of nitrogen. No difficulty was observed with sublimation, even with naphthalene. Automatic control maintained furnace temperatures to within $\pm 2^\circ\text{F}$. A system of continuous pressure recording was used.

The main criterion of stability was gas evolution. This was measured in two ways. First, values were obtained from the final pressure readings after cooling the bombs to room temperature. More reliable values were obtained by mass spectrographic analysis of the gas phase and evaluation based on the initial nitrogen charge. Both values are given in the tables, and agreement is generally satisfactory. As a more readily apparent index of damage, the mass spectrographic gas determination is also given as moles of gas (total hydrogen and C_1 to C_5 hydrocarbons only) per mole of initial charge. Obviously, values of this latter index which are of the order of unity would indicate extensive damage. However, this is not completely unambiguous. Hydrogen and methane were found to be the chief constituents of the gas phase, in general, with decreasing amounts of higher paraffin hydrocarbons and usually small amounts of benzene. Unsaturated paraffins were not detected in any appreciable quantity. The gaseous hydrocarbon-to-hydrogen ratio increased as coking increased, but the individual ratios varied considerably depending upon the nature of the original aromatic compound.

For the purposes of this study, coke is defined as the relatively infusible and/or nonvolatile fraction of the pyrolysis residue. This was estimated in many cases from the simple manual recovery of the sample. The per cent liquid, quoted in the tables, represents liquid saturated with respect to the solid phase at room temperature. In cases of special interest, a simple one-stage high vacuum "molecular" distillation was performed to separate coke from the volatile solid fraction. Treatment in the latter separation was for one hour at 315°C (600°F) at about one-micron pressure. The coke values are not as suitable as the gas evolution data for the evaluation of relative thermal stabilities; however, the former are of perhaps more practical interest. In any case, all the factors must be considered in evaluating a given compound for possible use as a heat transfer medium.

3.113 Experimental Results

The Effect of Temperature - The study of the effect of temperature was made not particularly from the point of view of practical application but in order to find the highest temperature which would give reasonably rapid screening results. Because it was hoped to establish relative stabilities, appreciable decomposition was necessary. It has been previously shown¹ that 1000°F (538°C) is too severe for this purpose.

¹ Bolt, R. O., and Carroll, J. G., Cal. Res-AEC Report No. 4, Jan. 9, 1953, pp 16-18

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For reasons previously noted, biphenyl and naphthalene were chosen for this preliminary study. The data, including some for o- and p-terphenyls, are shown in Table 3.1. On the basis of these results, 920°F (493°C) was chosen as a suitable temperature for further study. Naphthalene and biphenyl were of comparable stability with respect to gas evolution at this temperature. A marked difference in the effect on gas evolution with increasing temperature was readily apparent, however. The evolution from naphthalene increased about five times faster than that from biphenyl in going from 920°F to 950°F (510°C) or 980°F (526°C). In the homologous compounds, biphenyl, o-terphenyl, and p-terphenyl, the increase in gas evolution in going from 920°F to 950°F appeared to be constant, 5.0, 4.6, and 4.7, respectively. No simple relationship (for example, of the Arrhenius type) is apparent between temperature and gas evolution. This is not unexpected in view of the complexity of the reactions occurring during pyrolysis.

The effect of added quinoline on biphenyl and naphthalene at 980°F is included in the data of Table 3.1. An appreciable effect was noted on the naphthalene only; more will be said of additives in a later section. All of the remaining data to be presented in the sections to follow were obtained at 920°F.

Polyphenyl vs. Condensed Ring Aromatics - A comparison of the effect on stability of some of the simplest structural variations of the homologs of biphenyl and naphthalene is given in Table 3.2. Excluding naphthalene, the polyphenyls as a class were more stable than the condensed ring aromatics. In fact, there would appear to be little reason for pursuing any further the study of the latter class of compounds. The data on the polyphenyls illustrate the not unexpected effect of decreasing stability with increasing molecular weight.

Infrared spectroanalysis of the volatile fractions recovered from biphenyl and naphthalene after 24-hours' exposure at 920°F gave purities of 58% and 95%, respectively. Thus, naphthalene appears appreciably more stable than indicated by gas evolution or coke formation alone. (The effect of longer exposure will be discussed later in the section on additives.) A further difference noted between all of the members of the two classes of aromatics examined is in the character of the "coke" formed during pyrolysis. The condensed ring aromatics produced a true, hard, infusible coke which formed as a separate phase. The polyphenyls, on the other hand, formed "resins" rather than coke which, although relatively nonvolatile, were still soluble in the melt. This difference could be of considerable practical importance and favors further the polyphenyls as a class.

Typical pressure versus time data are illustrated by the curves of Figure 3.0. It is apparent that no singular interpretation of the curves can be made. Relatively rapid attainment of a pressure plateau can be the result of very rapid and extensive

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initial decomposition or of good stability as in the cases of anthracene and naphthalene, respectively. The slow, continuous rates of rise in pressure in the cases of p-terphenyl and p-quatrphenyl are practically identical; although the total gas evolution of the latter was over three times greater than the former. Phenanthrene shows a type of complex pressure versus time behavior which will become even more apparent in studies at longer exposures to be described. In keeping with the general complexity of the pressure behavior, the individuality of the ratio of hydrocarbons (methane to pentane) to hydrogen in the evolved gas is apparent in the values listed in Table 3.2.

The Effect of Substituents on Naphthalene - The effects of simple substitution on naphthalene are shown in Table 3.3. The alkyl side chains in the methyl naphthalenes and in acenaphthene increased the gas evolution very markedly. The fluoronaphthalenes exhibited good stability in regard to gassing. However, 1-fluoronaphthalene, the best in this regard, was converted from the original liquid at ordinary temperatures to about 65% solid product after thermal exposure.

The Effect of Structure Modification - Examined under this section are a number of compounds typical of a variety of structures which do not fit into previously discussed classifications and which might be expected to have good thermal stability. Included in Table 3.4 are data for cetane and decalin. The latter were run only to confirm the expected, extremely poor stability of nonaromatic hydrocarbons under the present conditions. None of the compounds studied show any particular promise at 920°F. These include aromatic derivatives of methane and ethylene, aromatic silanes, an aromatic phosphate, and a halogenated benzene. In the latter case, the use of a Vycor liner reduced the gas evolution; but the decomposition was still very extensive. A perfluorinated hydrocarbon of average composition corresponding to $C_{21}F_{44}$ was still liquid, with little or no coke formed after 24-hour exposure at 920°F; however, extensive cracking to lighter fluorohydrocarbons had occurred.

The Effect of Additives - The kinetic nature of the problem of thermal stability has been already noted to be implied from the basic thermodynamic instability of organic compounds. Also, it is generally accepted¹ that a free radical mechanism predominates in the thermal decomposition of hydrocarbons. A fruitful approach to the problem of increasing the thermal stability of organic compounds would then appear to be that of attempting to find inhibiting additives. The thermal stability of the additive itself, albeit in dilute solution in the organic fluid, is a complication to be considered.

Naphthalene and biphenyl were chosen for these studies, the results of which are shown in Table 3.5. To increase the thermal damage at 920°F, in order that the effect of additives

¹ Rice, F. O., A.C.S. Petrol. Div. Sympos. on Catalysis in Hydrocarbon Chemistry, No. 24, 5-7 (Sept. 1952)

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could be more readily assessed, the exposure time was increased to 72 hours. The unexpected result found was that the gas evolution increased about 7- and 38-fold for biphenyl and naphthalene, respectively, for the threefold increase in exposure time. This is shown in the pressure versus time data illustrated in Figure 3.1. It is apparent that the observed effects are the result of "inhibition" periods of about 20 and 34 hours for biphenyl and naphthalene, respectively. For the latter compound, this period was reduced to approximately 20 and 12 hours at temperatures of 950°F and 980°F, respectively. Biphenyl appears to show a similar effect. In an attempt to determine the cause of this inhibition period, runs were made with both biphenyl and naphthalene in Vycor (96% silica glass) liners; and a run was made with specially purified naphthalene. The latter was acid and alkali washed and then recrystallized several times from alcohol. None of these variations had any significant effect on the stability or pressure-time behavior. The effect of the Vycor in increasing markedly the hydrocarbon-to-hydrogen ratio in the gas from both naphthalene and biphenyl is notable.

None of the additives tried had any beneficial effects on the thermal stability. Where the total gas evolved was perceptibly decreased in two instances (for naphthalene), the inhibition period was shortened, however. It is interesting to note that an increase in the gaseous hydrocarbon-to-hydrogen ratio appears to go along with the decreased inhibition periods in every case. In general, an increase in the above ratio is associated with increased decomposition whether this be due to higher temperature, longer exposure, or catalytic effects. The lack of inhibiting effect of nitric oxide is surprising in view of its known behavior in this regard with paraffins^{1,2}. Acridine and similar compounds are known to function as inhibitors of catalytic cracking³. These were not effective in the present study of thermal cracking. Naphthacene was tried because of its known behavior as an inhibitor of liquid phase free radical reactions⁴ (at ordinary temperatures) but showed catalytic activity in the present study. Iodine is a known catalyst for paraffin cracking⁵ and was found to function extremely efficiently in this regard for biphenyl in the present study. The pressure-time data are shown for naphthacene and iodine on Figure 3.1. The problem of finding an inhibiting additive does not appear hopeful at present in view of the results discussed and in view of the similar experiments of other workers in this field¹.

¹ Rice, F. O., A.C.S. Petrol. Div. Sympos. on Catalysis in Hydrocarbon Chemistry, No. 24, 5-7 (Sept. 1952)

² Sachanen, A. N., "Conversion of Petroleum, p 24, Rheinhold (1948)

³ Vogl, H. H., et al, 3rd World Petrol. Congr. (1951) Sect. IV, Subsect. II, Preprint 5

⁴ Kooyman, E. C., and Farenhorst, E., Trans. Farad. Soc. 49 58-67 (1953)

⁵ Same as ² above, except p 23

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The data of Table 3.5 include a run made with the eutectic mixture of naphthalene and biphenyl (44 and 56 weight per cent, respectively) which melts at 103°F (39.4°C). The observed gas evolution (0.58) is appreciably greater than that which would be calculated (0.41) from the composition. The pressure-time behavior of this mixture is included on Figure 3.1.

3.114 Conclusions

The results of the present study at 920°F (493°C) demonstrate the outstanding thermal stability of the simplest polycyclic aromatic structures, namely, biphenyl and naphthalene. Maximum exposures, resulting in little or no damage, at this temperature were about 30 and 20 hours for naphthalene and biphenyl, respectively. However, for longer exposures, where appreciable damage occurred, biphenyl was better than naphthalene. To a considerable degree, the latter effect was due to a basic difference in the condensation products formed from polyphenyls as compared to condensed ring aromatics in general. The former produced fusible resins soluble in the melt, rather than true infusible cokes. As the number of aromatic rings was increased, polyphenyls were found to be more stable than condensed ring aromatics in every respect. It appears unlikely that any polynuclear aromatic of the condensed ring type would show any promise as a thermally stable heat transfer fluid at 920°F.

None of the structure modifications of aromatic compounds studied showed any promise with regard to thermal stability. These studies included representative examples of aromatic methane derivatives and silanes which are considered to have unusually high thermal stability for organic compounds. The majority of the compounds studied showed such extensive damage in 24-hour exposures at 920°F that it did not appear feasible to attempt to establish relative stabilities at this temperature.

The search for inhibiting additives to prolong thermal stability was unsuccessful. However, the pressure versus time data for naphthalene and biphenyl indicate that the apparent stability of these compounds was due to an inhibiting factor of, as yet, unknown origin. It, thus, appears to be of interest to pursue the study of additives further.

3.115 Future Work

The continued pursuit of a unique organic structure of optimum thermal stability does not appear to be a fruitful avenue for further work. However, properties other than thermal stability may be of importance (e.g., melting point, boiling point, viscosity, etc.) from an engineering standpoint. Hence, some further work with modifications of the simplest organic structures, especially with biphenyl, may be of interest; methyl-, fluoro-, and tri-fluoromethyl-substituted biphenyls, for example. Diphenylene, though not readily available, would be of some interest to complete the picture on the basic aromatic structures.

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Additional additive studies are perhaps the most likely course which would lead to increased thermal stability. In addition to the usual liquid phase additives, it appears of interest to examine the effect of variation of the initial gas phase (e.g., ethylene, hydrogen, and methane) over the organic fluid. In the same category, a significant study would be the determination of the effect of some simple purification cycle on the stability (i.e., on the inhibition period) of naphthalene (or biphenyl) which has been subjected to thermal exposure.

Particular physical specifications for an organic heat transfer fluid will very probably have to be met by employing mixtures. Possible eutectic mixtures, especially of biphenyl with other polyphenyls, should be studied with this view.

The temperature of 920°F has proved to be still too high to obtain relative stabilities for the majority of organic compounds. For this reason, and also because actual working temperatures being contemplated for organic fluids in nuclear reactors are as low as 750°F (399°C), thermal stability studies at temperatures below 920°F should be undertaken. Much longer exposures would be necessary at the lower temperatures, and the experiments should probably be of the capsule type as employed for irradiation studies.

3.12 Radiation Stability

3.121 Experimental Approach

In addition to the thermal stability experiments already described for the development of possible moderator-coolants, radiation stability tests at high temperatures were conducted. These were a necessary next step in the screening process. The maximum temperature to which it was possible to use these organics in a radiation field may thus be established. Temperatures up to 426°C (800°F) were attained, with dosages, expressed as thermal neutrons, approaching $10^{18}/\text{cm}^2$. The samples were contained in Series 20, 21, 22, 23, and 26, the particular exposures of which have been noted previously in Tables 2.2 and 2.3. Discussion of the results is placed in this section because these experiments pertain almost entirely to the moderator-coolant work.

Evaluation criteria included viscosity change, gas evolution, and analysis by infrared spectrophotometry. Molecular distillation was also carried out on certain of the irradiated materials. Irradiations were conducted in the Brookhaven Reactor in capsules of 410 stainless steel of the C-12 and C-22 type described previously. The techniques of sample evaluation have also been described adequately in previous sections.

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3.122 Experimental Results

Viscosity Change - Figures 3.2 through 3.6 represent typical plots of the index of viscosity change, η_1 , versus dosage for a number of organics irradiated at the different temperatures shown. The curve for n-butylbenzene, previously shown in Figure 2.6, is repeated in Figure 3.2 to demonstrate the adherence to a straight line function for this $\log \eta_1$ -dosage relationship to a dosage of $N \sim 3$. These data, as noted, were all obtained at 140°C . The amylobiphenyl curve reflects the straight line relationship to an N of ~ 4 , irrespective of temperatures which range from 140°C (284°F) to 371°C (700°F). This is very important and points out that, in the "linear" region of this curve, the effect of temperature is dominated by the more important effect of ionizing radiations. It is to be noted that, in this region, the slope of the amylobiphenyl is less than that of the n-butylbenzene, demonstrating greater "stability" of the amylobiphenyl for the lower dosages. The amylobiphenyl curve is seen to deviate upward in the regions of higher dosage to an N of 8. However, only one point is available on the curve to illustrate this behavior.

Figure 3.3 is a similar plot for biphenyl and 3-methylbiphenyl. The initial portion of the curve again reflects the straight line relationship to an N of ~ 4 to 5, once more irrespective of temperatures ranging from 140°C to 371°C . The 426°C (800°F) exposures for both fluids resulted in no fluid portion remaining for evaluation. It is interesting that the points for both materials are so closely grouped that one curve suffices to connect both sets of data. The slope of this curve is approximately the same as that for the amylobiphenyl; and, again, the upward deviation from a straight line beyond an N of ~ 5 is shown. The grouping of the points in the region of $N = 8$ more firmly establishes this tendency for the curve to deflect upward. For these materials, the increase of temperature in the higher regions of dosage, with the exception of the 371°C exposure of biphenyl, appears to have an accelerating effect on the thickening.

Figure 3.4 is a similar plot for Dowtherm "A" ($\sim 75\%$ diphenyl oxide and 25% biphenyl). This curve further illustrates the points already mentioned. The slope is somewhat higher than that of biphenyl, showing that the diphenyl oxide is somewhat less stable than biphenyl itself. This is also indicated by the fact that the 315°C (600°F) exposure at about an N of 8 resulted in the material becoming coked, a situation not experienced with biphenyl alone.

Figure 3.5 is a similar plot for naphthalene and 1-phenylnaphthalene. Observations previously made are further verified in these two curves. For naphthalene, the 315°C sample, at an N of 8, was lost, while the 371°C sample was also lost by virtue of a ruptured bursting disc. Comparatively little difference in slope is indicated between the two materials.

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Figure 3.6 is an illustration of the same relationship for 1,2-diphenylbenzene (o-terphenyl). The curve again depicts the straight line portion, irrespective of temperatures to 371°C to an N of ~5. From there on, the deviation from linearity is again observed. In the 371°C exposure, at an N of ~8, the terphenyl was reduced to a coke.

Table 3.6 is a summary of the radiation stabilities of the various materials discussed in Figures 3.2 through 3.6.

TABLE 3.6

STABILITIES OF AROMATICS IRRADIATED AT 140°C-371°C

Compound Identity	Relative Index at 4 x 10 ¹⁸ n/cm ² Slope* Substance/Slope 1,2-Diphenylbenzene
1,2-Diphenylbenzene	1.0
s-Amylbiphenyl	1.1
Biphenyl	1.3
3-Methylbiphenyl	1.3
1-Phenylnaphthalene	1.4
Naphthalene	1.6
Diphenyl Oxide (75%) - Biphenyl	1.8
n-Butylbenzene	2.8

* Log η_1/nvt

Comparison is made of the actual slope ($\log \eta_1/nvt$) of the curves for the various materials at N = 4, as compared to that for 1,2-diphenylbenzene which was arbitrarily assigned a value of unity. Because the slope is an index of radiation stability in that it represents the rate of thickening with irradiation, the diphenylbenzene emerges as the most stable by the viscosity change criterion up to about 371°C and N = 4. At dosages higher than N ~4, and particularly at N = 8 where the temperature effect becomes increasingly important, the very marked increase in slope of the curves (depicted in Figures 3.2 through 3.6) renders the determination of relative stabilities difficult.

Gas Evolution - The present data on gas evolution must be considered as semiquantitative because of difficulties experienced in measurement. These difficulties resulted from solubility of the evolved gases in the liquid organic. This caused foaming and contamination of the penetrometer gage

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bourdon tubes with liquid which proved difficult to remove. Nevertheless, the data give some indication of the gassing tendencies of the organics at high temperatures in the presence of radiation.

The information in Table 3.7 illustrates gassing of certain of the fluids exposed at 315°C (Series 16), 371°C (Series 15), and 426°C (Series 24).

TABLE 3.7

GAS EVOLUTION IN REACTOR IRRADIATIONS

Exposure Temperature* Compound	Ml Gas/Ml Material Charged		
	315°C	371°C	426°C
Biphenyl	3	2	42
3-Methylbiphenyl	8	9	35
s-Amylbiphenyl	23	52	Blown
Naphthalene	4	5	46
1,2-Diphenylbenzene	5	4	22
1-Phenylnaphthalene	7	5	Blown

*BNL Exposures, nvt = $\sim 3 \times 10^{18}$ n/cm²

Dosages for these exposures were approximately equivalent. The principal difference in gas generated appears related to the presence or absence of aliphatic side chains. Typically, biphenyl resulted in low gas formation, with increased gassing for 3-methylbiphenyl and a further increase for amylbiphenyl. The other polynuclear aromatics without side chains showed gassing equivalent to biphenyl. Although there was little difference between 315°C and 371°C exposures (except for the amylbiphenyl), 426°C caused markedly increased gas evolution. This increase corresponds to a greatly increased coking.

Table 3.8 contains similar gassing data for exposures of approximately $N = 8$ at temperatures of 170°C (Series 20), 315°C (Series 21), and 371°C (Series 23).

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TABLE 3.8

GAS EVOLUTION IN REACTOR IRRADIATIONS

Exposure Temperature*	Ml Gas/Ml Material Charged		
	170°C	315°C	371°C
Compound			
Biphenyl	7	19	17
3-Methylbiphenyl	19	61	Blown
s-Amylbiphenyl	168	Blown	Blown
Naphthalene	7	Blown	Blown
1,2-Diphenylbenzene	7	11	58
1-Phenylnaphthalene	11	33	55

*BNL Exposures, nvt = $\sim 8 \times 10^{18}$ n/cm²

These data, from Table 2.3, page 2, are in accord with those of Table 3.7. A comparison of the two tables indicates that the increased dosage has a very large effect on the amount of gas generated in these materials upon irradiation. In comparing values for the different dosages at 315°C and at 371°C, it is readily seen that the higher dosage results in about ten times as much gas formed and actually represents a more severe condition, from the standpoint of gassing, than the temperature increase from 371°C to 426°C.

Analytical Data - In order to ascertain purity changes on irradiations at elevated temperatures, infrared spectrographic analyses were carried out on the samples of Series 26 (N = 2.8; temperature, 371°C). These results are shown in Table 3.9.

TABLE 3.9

ANALYSIS OF IRRADIATED AROMATICS*

Compound	% Purity of Crude by I.R. Anal.	% Distillate	% Purity of Distillate by I.R. Anal.	% Purity of Crude Based on Distillate**
Biphenyl	79	97	81	79
3-Methylbiphenyl	-	92	83	76
Naphthalene	76	95	81	77
1,3-Diphenylbenzene	76	93	72	67
1,2-Diphenylbenzene	72	69	85	59
1-Phenylnaphthalene	~ 43	69	~ 35	~ 24

* From BNL exposure at 371°C and nvt = 2.8×10^{18} n/cm²

** (% Purity of Distillate) x (% Distillate)

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The values in the first column of Table 3.9 give the apparent per cents of unchanged material in the whole (crude) irradiated samples. The infrared spectra are, however, difficult to interpret because of interferences by the irradiation products. To alleviate this difficulty and to gain information on purification methods, the crude materials were distilled and the distillate analyzed by the infrared method. These data occupy the remaining columns of Table 3.9.

A simple molecular distillation procedure was conducted to a still temperature of 315°C (600°F) at about one micron pressure. The residues in all cases were dark, resinous solids. It is felt that the infrared analyses of the distillates should be more reliable than those determined directly on the crude material. Thus, the best values of purity of the crude, listed in the last column of Table 3.9, are based on the per cent distillate and the purity of the distillate.

As is to be expected, a simple, one-stage distillation of the crude irradiated samples effects only a partial purification; however, such an overhead should be suitable for recycling in a reactor moderator-coolant. Future studies are planned to extend this separation technique by employing fractional distillation, silica gel adsorption (an example of which is shown in Figure 3.7), and similar methods.

General Comparisons - Table 3.10 summarizes the radiation stability of the six materials of Series 26 by the three indexes of damage: viscosity change, gassing, and purity change. The index of viscosity damage, read from the curves of Figures 3.2 to 3.6 at $N = 2.8$, is also included. Actually, the organics are rather closely grouped by all three indexes. This is not surprising because of the similarities in structure of the materials.

TABLE 3.10

STABILITIES OF IRRADIATED AROMATICS*

Compound	η_1 at $N = 2.8$ from Figs. 3.2 to 3.6	η_1 from Series 26	ML Gas/ ML Liq.	Best Purity Value
Biphenyl	1.6	1.4	2	79
3-Methylbiphenyl	1.6	1.4	6	76
Naphthalene	1.8	1.8	5	77
1-Phenylnaphthalene	1.7	2.4†**	~8	~24
1,2-Diphenylbenzene	1.4	1.5†	5	59
1,3-Diphenylbenzene	-	1.5†	4	67

*From BNL exposure at 371°C and 2.8×10^{18} nvt

**Value apparently unduly high

†Estimated

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It is apparent that the viscosity stability criterion, η_1 , is the least sensitive; the spread between the actual η_1 measurement of Series 26 and the "average" taken from the curves of Figures 3.2 through 3.6 should be noted particularly. Lack of correlation between the various indexes is due largely to the fact that viscosity is a complex function of composition and varies in an individual manner for each compound. An interesting result of this is that a loss of purity need not correspond to a change in viscosity. This is illustrated in the cases of the diphenylbenzenes.

3.123 Conclusions

From the radiation stability data cited from Series 20 through 24 and Series 26, several tentative conclusions can be made. For dosages up to an N of ~ 5 , radiation damage, as measured by viscosity change, appears to be independent of temperatures to 371°C. At temperatures above this value, specifically at 426°C, all the materials mentioned in this section were no longer fluids at dosages of about N = 3. This indicates that, at some temperature between 371°C and 426°C, points of discontinuity in the curves occur; and temperature assumes an important role in the degradation. In this mild dosage region, a log function of viscosity increase varies linearly with radiation dosage. At the higher dosages, in the region of an N of ~ 8 , the effect of temperature is not conclusive; although, in most instances, higher temperatures appear to have imparted greater damage. In this dosage region, the plot of the log function of viscosity increase versus dosage is not a straight line and, instead, deviates markedly in the direction of greater damage. At the mild dosages, gas evolution appears to be independent of temperature up to 371°C for aromatics and alkyl-aromatics. The amount of gas generated was greatly accelerated by exposures at 426°C or by employing dosages to an N of ~ 8 .

Biphenyl and the terphenyls appear to be affected to about the same degree by irradiation at 371°C and an N of ~ 2.8 . It is reemphasized that agreement between the three indexes of damage is not to be expected because each one is dependent to some degree upon a different aspect of the degradation of the original compounds.

3.124 Application of Organics as Moderator-Coolants

From the standpoint of this use in a reactor, it is evident that several aspects of the changes which occur in organic fluids upon irradiation must be considered. The information cited on viscosity and purity changes and gas evolution gives a quantitative picture of the extent of these occurrences as a function of dosage and temperature for typical materials believed to be among the most radiation stable. In reactor design, viscosity change is of most importance because the fluid must be circulated and the handling equipment must be chosen with the property of viscosity change in mind.

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Gas evolved is also important in that this must be contained or vented.

The formation of breakdown products upon irradiation is not necessarily of concern, except as their radiation stability departs from that of the original material, as they affect viscosity, lead to deposit formation, or promote excessive gassing. Deposit formation is particularly important in that surfaces in the circulating system would become contaminated. Such contamination might have a major effect on heat transfer and thermal conductivity and perhaps hamper circulation. It is evident that provisions for removal of this type of material must be made. It is believed that practical techniques for such removal can be adapted either from distillation, absorption, or other methods. In the regions of temperature and dosage discussed, it is evident that some such purifying system would be essential.

3.2 Research on Lubricants

During calendar 1953, there has been a growing interest in radiation resistant lubricants for use by AEC contractors. Section 6 presents the program of supply of such materials resulting from inquiries from various personnel.

Up to the present, California Research has formulated six radiation resistant oils based on physical properties adapted from the SAE gear and motor oil designations. Table 3.11 tabulates the viscosity specifications for these materials. Five greases have also been formulated to conform to the National Lubricating Grease Institute grades 0 through 5. Their designations are shown in Table 3.12.

TABLE 3.12

RADIATION RESISTANT GREASES

Notebook No. 1961-46-	1	2	3	4	5
NLGI Grade	0	1	2	3	4
ASTM Worked Penetration:					
Range	355/385	310/340	265/295	220/250	175/205
ASTM Worked Penetration	366	314	296	233	177
ASTM Drop Point, °F	← 500+ →				

Samples of these formulations of oils and greases have been exposed to gamma irradiation in Series G-8 and G-9. However, evaluations have not been completed as yet.

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Because of the renewed interest in radiation resistant lubricants, this subject will be covered in an individual report, CRC-AEC No. 7, to be issued later this fiscal year. In this report, the status and results of irradiations on the particular lubricants mentioned, as well as irradiation data obtained on other lubricants, will be presented. Portions of Series G-1, G-3, G-4, G-5, G-6, and G-10, which pertain to studies of either conventional or special lubricants, will be involved.

3.3 Research on Fluid Hydrogenous Shields

During the past two years, little work has been conducted on the development of fluid shielding materials. Research performed in 1950 is still pertinent to this subject.

Past experience has shown that radiation stability generally varies inversely with hydrogen content. Ideally speaking, liquid methane would present the maximum obtainable N_H for organic fluids; while the minimum N_H would occur for polynuclear aromatics. The latter materials should evidence by far the greater stability toward ionizing radiations. The problem is to find some intermediate fluid possessing both good radiation stability and high N_H . Aliphatics possessing functional groups, such as amines or alcohols, certainly have good hydrogen contents; but their stability is questionable.

During the present report period, a few of these were exposed to combined flux and to gamma flux. Exposures were made to expand the general knowledge in this field and to answer inquiries as to the radiation stability of these materials. Results of the exposure to the combined flux of the Brookhaven Reactor are shown in Table 2.3B.

TABLE 2.3B

RADIATION STABILITY OF FLUID HYDROGENOUS SHIELDS -
FROM TABLE 2.3, PAGE 3, SERIES 25 - BNL - 170°C - N=2.29

Fluid	Atoms of H_2/cc , $N_H \times 10^{-22}$	Gas Formed, ml/ml Fluid	Index of Damage
n-Dodecylamine	7.1 at 20°C	-	Rubbery at 100°C
n-Dodecylamine, 5% 0-1*	-	-	Solid at 100°C
n-Hexylamine	6.9 at 20°C	-	$\eta_1 = 30$
Tributylamine	6.9 at 20°C	27	$\eta_1 = 4.6$
2-Phenylethylamine	4.8 at 24°C	10	$\eta_1 = 50$
Dodecanol	7.0 at 24°C	1	$\eta_1 = 4$
Dodecanol, 5% 0-1*	-	-	Solid at 100°C
Water	6.7 at 4°C	-	-

*0-1 = Didodecyl
selenide

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None of the materials in Table 2.3B were found particularly radiation stable. With the amines, the superiority of the tertiary structure is evident and merits further investigation. The use of the selenide additive with dodecylamine may even have been detrimental, as the results indicate. The same situation exists with the alcohol. Actually, the dodecanol without additive shows some promise as a shield material under mild exposure conditions, although the viscosity damage suffered is rather high. With four exceptions, the η_1 's of the 45 samples of Series 17, 18, and 19, which were exposed to comparable dosages and lower temperatures, were lower than the value for dodecanol. Samples of the materials of Table 2.3B have been exposed in the MTR gamma facility for an approximate dosage of 10^{10} R. The exposure has been completed, and evaluation is in process on these samples.

4. CONSULTING PROGRAM

4.2 MTR Loop

By letter of July 3, Dr. I. F. Zartman advised California Research that the design and construction of a loop for exposing organic materials in reactors had been assigned to the Phillips Petroleum Company. California Research Corporation was asked to act in an advisory capacity in the construction of this equipment. A meeting was held on July 9 in response to an invitation of Mr. J. R. Huffman of Phillips on June 30. California Research personnel met with representatives of North American Aviation and Phillips; and discussions included preliminary design concepts, organic materials of interest to both North American and to California Research, and also the type of data of interest

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to all concerned. It was the consensus that the first loop be operated in the gamma facility at the MTR and that a second loop be constructed to operate in a reactor, if funds permitted. It is visualized that this loop will be a facility of the Materials Testing Reactor, available to anyone having need for it.

4.3 Argonne National Laboratory

For some time, Argonne National Laboratory has been concerned with a lubrication problem involving the use of a radiation resistant grease lubricant. This problem exists in the Experimental Breeder Reactor being operated in Idaho and comprises the lubrication of certain components of control rods. These rods are actuated by electric motors and must be free to move within the limits of the power output of the driving motor. Neutron fluxes of about 10^{12} neutrons/cm² prevail in an ambient temperature ranging from 30°C to 100°C. The exposure of the lubricant constitutes a rather severe condition in that a thin film is exposed to oxidative conditions in the radiation zone. At present, a Dow Corning silicone or a halocarbon-type grease is being used, with less than satisfactory operation because this grease must be changed at frequent intervals. It is well known, of course, that silicones are among the least radiation resistant of organic types.

On previous occasions, experimental greases formulated by California Research have been provided Argonne for this application. These greases were exposed in Argonne's CP-3 reactor and were considered unsatisfactory on the basis of gas formed. On June 11, three samples of experimental greases were provided Dr. W. K. Anderson of Argonne for his scrutiny. These three greases are designated 1961-43-2, -3, and -4, respectively, in Table 4.0. Of the three, the 1961-2 was satisfactory from a radiation stability standpoint but unsatisfactory in a thermal stability test for one month at 100°C. The 1961-3 and 1961-4 versions were reported to have excellent thermal properties, that is, resistance to "caking" in the thin film exposures at 100°C. However, their radiation stability, as evinced by gassing, was considered unsatisfactory. It was requested that materials similar in thermal properties but having increased radiation stability be supplied. Two such materials were provided Dr. Anderson in Los Angeles on August 6, 1953. The composition of these two greases is noted in the attached Table 4.0 as Nos. 1961-48-3 and -4.

4.4 Knolls Atomic Power Laboratory

During this report period, personnel of Knolls Atomic Power Laboratory have presented problems involving need for radiation resistant lubricants. Lubrication of remote handling machinery for processing SIR fuel elements is required in a gamma atmosphere for $\sim 10^9$ - 10^{10} R. Close liaison through correspondence and incidental visits has been maintained with personnel under Mr. R. Boice of the Remote Fuel Handling Subunit in this regard. Samples of lubricants have been supplied as requested and as noted subsequently in Table 6.0.

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5. COOPERATIVE PROGRAM

5.1 North American Aviation

A cooperative program with North American Aviation has as its immediate purpose the determination of the equivalence of electron and reactor flux with respect to radiation damage to organic compounds, as measured by viscosity and purity changes. For this study, n-butylbenzene (research grade) was used. The reactor exposures were made at the MTR (VH-3) and have been discussed previously as Series 27. These involved irradiation in magnesium capsules under a helium atmosphere at $\sim 30^{\circ}\text{C}$. The electron irradiations were performed with the North American Aviation Statitron (electrostatic generator). Here, samples were irradiated in an aluminum cell (stirred) under helium at $\sim 33^{\circ}\text{C}$. There is still some question regarding the actual dosage delivered in the electron irradiations, apparently due to uncertainties in voltage measurement. The results from the Statitron and reactor work are tabulated in Table 5.0. The index of viscosity damage, η_1 , and the infrared spectra were determined for all irradiated samples. In addition, for the electron bombarded samples, mass spectrographic analyses were made.

In general, the infrared analyses were not completely satisfactory. The character of the absorption peaks in the region of 13.4 and 14.3 microns shows little apparent interference from components other than the parent substance. Yet, all indications are that the purities calculated therefrom are much too high. This may indicate the formation of polyalkylphenyls which absorb strongly in the same spectral region as the original n-butylbenzene. In the case of the 100 Mah electron-irradiated sample, good apparent agreement was observed between the purity based on mass spectrographic analysis compared with that calculated from the infrared absorption in the region of 9.4 and 10.7 microns. For this reason, the infrared analyses given in Table 5.0 are based on the absorption in this region of 9.4 and 10.7 microns.

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Figure 5.0 shows for the reactor work the relationships for both per cent purity (from the infrared determinations) and η_h versus reactor dosage. The η_h and purity data for the 100 μ ah electron irradiation applied to these curves show an equivalent reactor dosage of $\sim 2.75 \times 10^{18}$ thermal nvt from the η_h and $\sim 2.20 \times 10^{18}$ thermal nvt from the purity. The former is considered the more accurate. The difference between these two dosages represents an uncertainty on the purity curve of $\sim 4\%$, well within the experimental error.

The electron and the reactor irradiations can be arbitrarily put on an equal dosage basis by consideration of the total energy absorption in each case. Using the calorimetric data of Richardson (ORNL-129), the 100 μ ah electron irradiation is found equivalent to $N = 9.3$ at position H-12 in the ORNL graphite reactor. Complete data for conversion to the MTR equivalence are not available. However, using the fast neutron and gamma fluxes and apparent damage ratio at the MTR and ORNL (see Tables 2.0 and 2.9), it can be estimated that the 100 μ ah electron irradiation is roughly equivalent, on an energy absorption basis, to $N = 5$ at the VH-3 facility in the MTR. The apparent energy equivalence found from the viscosity data is only 56% of this value ($2.75/5$) and, from the purity data, only 44% ($2.20/5$). It is, thus, apparent that either (1) electrons actually cause less damage than combined radiation from an energy absorption standpoint, (2) the calculated equivalence between electrons and combined reactor radiation is in error, and/or (3) the apparent dosage given the electron irradiated sample is incorrect. Although there is considerable uncertainty in (2), recent information from North American personnel indicates that (3) is likely true. Research is now proceeding to correct this difficulty.

5.2 National Bureau of Standards

Contact was established with Dr. C. W. Beckett, Chief, Thermodynamics Section of the National Bureau of Standards, in latter 1952. Dr. Beckett, together with other groups coordinated with his, was concerned with the synthesis of organo-silicon and fluorine-containing materials for possible use as reactor moderator-coolants. The Bureau was concentrating on synthesis work, and it was agreed that California Research would conduct, along with the regular program, a number of irradiation screening tests on the new products.

In response to an invitation by California Research, Dr. Beckett provided several samples of organo-silicon compounds, two of which were exposed in Series 24 at 426°C , in addition to samples exposed in Series 25 at 170°C . Two of these materials, the tetrabenzylsilane and the dibenzothiophene, merited further study and have been included in subsequent exposures. At California Research request, additional samples were supplied by Dr. Beckett in September 1953 and were irradiated in

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Series 30, 31, and 32. A description of all the samples received from Dr. Beckett is included in Table 5.1, along with the disposition of these materials.

6. SUPPLY PROGRAM

6.1 Samples Supplied

California Research Corporation has supplied samples of radiation stable organic fluids upon request, when possible. Usually, samples of gram or ounce quantity for research purposes have been supplied gratis. When the sample reaches gallon or pound size, normally a special preparation is required. Such samples are then supplied on a cost basis, which includes actual charges made for materials; transportation, if prepaid; and man-hours directly needed for the preparation work and the performance of simple control tests. A listing of samples furnished on this basis is attached as Table 6.0.

7. FUTURE PROGRAM

7.1 Basic Program

Reactor Radiations

Effect of Dosage - The hydraulic rabbit facility (VH-3) at the Materials Testing Reactor is to be utilized in this work. Experiments are already under way to determine the effects of combined radiation to a dosage of $\sim 10^{19}$ n/cm², expressed as thermal neutrons, on n-butylbenzene. Similar studies will be extended to other materials of interest as potential moderator-coolants, shield materials, lubricants, etc. It is anticipated that this work will include, in addition to butylbenzene, a straight chain hydrocarbon, such as cetane; biphenyl, or similar compounds; an aliphatic monoester; an aliphatic diester; an aliphatic ether; and an aliphatic polyether. One or more of these compounds also is to be compounded with additives to ascertain how these affect the rate of deterioration.

Information derived from these experiments will be invaluable in predicting the performance of various organics under dosage conditions ranging from mild to severe. It is anticipated that the principal criterion of deterioration will be viscosity change, although accurate data on gas evolution are to be obtained in conjunction with analytical work to be described.

Effect of Flux Magnitude - Fifteen materials of basic structure have already been irradiated at ORNL and in the MTR. In the case of the latter irradiations, about a fiftyfold increase in fast flux was used. Comparisons of the results of these irradiations are expected to give information

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pertinent to the effect of flux magnitude. During the coming year, it is anticipated that a few additional experiments will be conducted on this subject, as required, to amplify adequately the definition of the flux magnitude variable.

Effect of Temperature - Considerable work on this subject is to be conducted under the applied program pertinent to moderator-coolants. This work will embrace organics expected to have maximum thermal and radiation stability. This is only one class of material which eventually will find application as a result of the present research. The effect of temperature is important to other classes of compounds as well. It is anticipated that samples from these other classes will be irradiated under different temperature conditions to ascertain how changes in temperature affect the radiation stability.

Effect of Dosage - A question has been raised repeatedly as to the effect of pressure in the irradiation capsule on the deterioration of organics. Although the nature of the reactions taking place with organics as a result of irradiation is such that it seems unlikely that a back-reaction could be made to take place, it does seem possible that, in certain instances, e.g., where formation of unsaturated materials is high, hydrogen pressure might retard decomposition. In order to check this possibility, it is anticipated that several samples will be irradiated with and without hydrogenation catalysts. If hydrogen pressure is beneficial, the hydrogen gas normally evolved may be less in a capsule with an effective hydrogenation catalyst than in one without such a catalyst. Another approach would be to undertake irradiations in open and in closed containers, the former fitted with check valves to permit the escape of evolved gas but prevent the introduction of air. Intercomparison of these results with those of past work may give important information on the effect of pressure.

Effect of Chemical Components

Catalysts - For almost any application of organics in the radiation field, metals of various identities will be used. Therefore, the effect of these on deterioration is important. It is planned to irradiate certain selected organics in stainless steel containers in which strips of various metals are placed. Comparison of the results from fluids so irradiated with those without metallic strips is expected to give adequate information on this subject.

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General Effects - New organics are in the offing and show some promise for various applications. Thus, silanes, polyfluoroaromatic compounds, and tertiary amines merit further investigation from preliminary results. Continued screening tests are, therefore, planned to gain needed information on special and new materials. Additional work is also being undertaken with various additives and additive combinations as these appear promising for alleviating the effects of radiation. In this regard, nitric acid, among others, should be investigated as a possible inhibitor of damage.

Analytical Work - The analysis of irradiated organics will be continued and expanded. Such data will be primarily aimed at determining the true extent of damage. However, where possible, the nature of the breakdown products will be determined. Such information may be useful in predicting more stable structures. The application of suitable fractionation methods will be made, which will not only simplify the analyses but will aid in the design of practical purification schemes. This latter, of course, is of primary importance for moderator-coolant applications.

These analyses will be especially pertinent to many of the fluids to be studied in the future work described in the sections pertaining to the effect of dosage, temperature, and flux components. Correlation of the three damage indexes, per cent purity, viscosity, and gas evolution, should also result from these studies.

Monitoring - A most important part of the research work in reactors is the determination of individual fluxes of the components of combined radiations in the various reactor facilities used. A considerable program of monitoring is, therefore, planned for the future. At present, little information is available on the magnitude of the epithermal (or, resonance) flux in the Brookhaven Reactor or the MTR. It is anticipated that existing data on gamma flux and fast and slow neutron fluxes will be supplemented by the data on the epithermal flux in these facilities. Checks on the fast flux at ORNL, Brookhaven, and at the MTR are also planned, along with measurements of gamma flux at Brookhaven. It is also hoped that a means may be derived for determining the gamma flux at Hole VH-3 of the MTR to complete the measurements of flux components there.

These monitoring operations will play a principal part in the effort planned to determine the comparative effects of gamma rays and neutrons. The cooperative program with North American Aviation is already set up to investigate the relative effects of electrons (from the North American Aviation Statitron) and gamma and combined radiations from reactors. Although this work is barely under way, it is probable that answers on this problem will be obtained in the near future. The irradiations

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of n-butylbenzene were first undertaken because considerable information on this material from both gamma and combined radiation facilities is available. Similar work on 3-methylbiphenyl is anticipated.

Gamma Rays - A major objective of further gamma irradiations will be to establish the equivalence between gamma rays and neutrons as to their relative damaging effects on organic fluids. A continuation of the present work, whereby basic types of organics have been exposed for successively higher dosages, is desirable. Analytical data will supplement viscosity and gassing data in this effort. More complete data on divergent types of organic fluids are also necessary in order to determine any possible effects of chemical structure on the comparative damaging effects of gamma rays and neutrons. This relative effect should vary considerably between different types of organic fluids.

Dosimetry methods for determining gamma flux, especially under combined radiation conditions, require further refinement. Further experiments will be made with gamma calorimeters in this work. If feasible, small calorimeters should be constructed and exposed concurrently with fluid samples in both reactor and gamma facilities. The feasibility of constructing a small calorimeter for exposure in the MTR VH-3 hydraulic tube facility will be investigated.

7.2 Applied Program

Moderator-Coolants

Effect of Dosage - Data have been obtained as to dosage effects at an $N \sim 3$ and also $N \sim 8$. Information is required at intermediate dosages. For this work, only stable materials, such as those suited for moderator-coolant use, will be chosen; and exposures will be at 315°C (600°F) and 371°C (700°F), the temperatures used in previous irradiations.

Effect of Temperature - Experiments conducted at 426°C (800°F) indicated that this temperature is too severe for the use of organics in radiation zones. Similar experiments at 371°C (700°F) show this temperature to be well within the range of usability. It is, thus, desirable to establish the temperature ceiling between 371°C and 426°C for use with organics. Present tests are being conducted at temperatures of 384°C (727°F), 399°C (750°F), and 414°C (775°F) for thermal nvt's of approximately 2.5×10^{18} . Repetition of exposures at 800°F in capsules having larger free space to minimize rupture of bursting discs and subsequent loss of sample may also be desirable. Such exposures would be performed either at Brookhaven or at the MTR in equipment similar to that previously used. If many experiments are warranted at higher temperatures, it may be well to consider a permanent heated section to be established in one of the MTR facilities.

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Loop Experiments - It is expected that the circulating loop will be available at the MTR for use during calendar 1954. California Research Corporation will be interested in performing experiments on selected moderator-coolant type materials, probably at a temperature of $\sim 371^{\circ}\text{C}$ (700°F). This type of an experiment is the logical sequel to capsule exposures and will provide information not otherwise available regarding engineering data, such as heat transfer, pumpability, coking, corrosivity, etc. The usual measurements of viscosity, gassing, and chemical analysis would be performed on irradiated samples taken during exposure.

An important item in this loop work is the establishment of required purification procedures, such as distillation or absorption techniques, for the organic. This is the initial step toward determining the conditions necessary for continuous operation. In support of this, it is planned to irradiate a large sample of organic in the MTR gamma facility for use in a further investigation of purification methods.

Thermal Stability Tests - Extensive thermal stability tests are not planned for the future because the effects of temperatures of the order of 371°C (700°F) appear to be secondary to the effects of radiation in proposed applications. Instead, radiation stability tests at high temperatures will be emphasized. However, a few thermal stability tests are contemplated to assess the damage to organics at 371°C to 425°C under conditions and by criteria analogous to the capsule-type irradiation studies. Comparison of these results with those of irradiation tests will permit a more complete answer relative to the effects of radiation alone at these temperatures.

Lubricants

The study of radiation stability of the special lubricants designed for maximum radiation resistance will be continued. Initial gamma exposures have been completed, though these are not yet evaluated. Such evaluations will serve to indicate the direction reformulation and retesting should take. No appreciable functional test work in bearings, gears, etc., is contemplated on the present project this year.

R. O. Bolt

R. O. BOLT

J. G. Carroll

J. G. CARROLL

B. J. Fontana

B. J. FONTANA

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The authors wish to extend their thanks to Dr. Rocco Fiore of Argonne National Laboratory, who supplied the cobalt and neptunium monitors and initially did the chemical processing thereof. Dr. Fiore also instructed California Research personnel in the methods used and has continued to provide monitors and information on the validity of the techniques.

Personnel of AEC reactor sites have contributed, in a large degree, to the progress of the work performed. Messrs. W. B. Lewis and C. E. Stoops assisted in the irradiations performed at the MTR. The exposures in the Oak Ridge Reactor were performed through the courtesy of Messrs. O. Sisman and C. D. Bopp of Dr. D. S. Billington's Solid State Division. Personnel of Brookhaven National Laboratory have contributed to the success of the experiments performed at that site. Particular credit is due to Dr. Marvin Fox, Chairman of the Reactor Science and Engineering Department, and to Messrs. R. W. Powell, J. J. Floyd, and F. P. Reeves of Reactor Operations.

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TABLE 1.0

PUBLICATIONS

Type	Date	Description	Distribution (or Sponsoring) Agency
I. <u>Technical Reports</u>			
1. CRC-AEC No. 6	Oct. 30, 1953	<u>Progress Report</u> for period Jan. 1, 1953, to Sept. 30, 1953	U.S. Atomic Energy Commission Technical Information Service Oak Ridge, Tennessee
2. CRC-AEC No. 5	Feb. 28, 1953	<u>Report</u> on Certain Shield Materials for Knolls Atomic Power Laboratory	
3. CRC-AEC No. 4 (TID-5132)	Jan. 9, 1953	<u>Progress Report</u> for period June 1, 1952, to December 31, 1952	
4. CRC-AEC No. 3 (TID-5094)	July 1, 1952	<u>Summary Report</u> for period April 30, 1951, to May 30, 1952	
5. CRC-AEC No. 2	Feb. 21, 1952	<u>Progress Report</u> for period Oct. 1, 1951, to Feb. 1, 1952	
6. CRC-AEC No. 1	Oct. 10, 1951	<u>Progress Report</u> for period May 1, 1951, to Oct. 1, 1951	
7. NEPA-1844	Apr. 30, 1951	<u>Final Report</u> for period Jan. 26, 1948, to April 30, 1951, "The Effects of Fission Radiations on Lubricants and Lubrication"	
II. <u>Progress Letters</u>			
1. Commencing with January 1953, a monthly Progress Letter covering general objectives and results has been distributed to those personnel and agencies known to be interested in the course of this research.			San Francisco Operations Office U.S. Atomic Energy Commission
III. <u>Prepared Discussions</u>			
1. B. J. Fontana	Aug. 5, 1953	"Thermal Stability of Aromatic Compounds at 493°C (920°F)"	(North American Aviation) (Conference on "Use of Organic Materials in Nuclear Reactors")
2. J. G. Carroll	Aug. 5, 1953	"Certain Effects of Radiation Dosage and Type on the Stability of Organic Fluids"	
3. R. O. Bolt	Aug. 5, 1953	"Some Effects of Temperature on the Radiation Stability of Aromatic Hydrocarbons"	
4. R. O. Bolt	June 2, 1953	Review of Progress	
5. R. O. Bolt	March 24, 1953	"Some Aspects of Radiation Damage to Organics"	
IV. <u>Reactor Science and Technology</u>			
	Dec. 1951 April 1952	Three survey articles based on NEPA-1844	U.S. Atomic Energy Commission Technical Information Service Oak Ridge, Tennessee

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TABLE 2.1
FOIL MONITORS OF REACTOR NEUTRON FLUX

Series	Reactor	Space	Foil			Distance from Reactor $\bar{G}$, in.	Exposure				Thermal Neutron Dosage, $n/cm^2 \times 10^{18}$		Ratio: Foil OFS	Fast Neutron Dosage, $n/cm^2 \times 10^{18}$
			Type	No.	Wt., mg		Date In	Date Out	Hours	MWH	Foil	Operations		
S-11	BNL	E-25	Co-Al ⁽¹⁾	2	7.00	1 (E)	9-23-52	10-1-52		3,582	-	-	-	
S-10	BNL	W-25	-			9 (W)					-	1.72	-	
S-12	BNL	E-25	-			29 (W)					-	1.63	-	
	BNL	E-25	-			33 (E)					-	1.58	-	
	BNL	W-25	Co-Al	3	8.35	37.5 (W)					-	-	-	
	BNL	E-25	Co-Al	1	7.30	41.5 (E)					1.61	1.51	1.07	
	BNL	E-25	Np ⁽²⁾	2	136.4	20 (E)	9-23-52	10-28-52		16,540	-	7.82	-	1.8 ± 0.35 ⁽³⁾
S-14	BNL	E-25	-			10 (E)	9-23-52	10-28-52		16,540	-	8.00	-	
	BNL	W-25	Co-Al	4	6.60	1 (W)	10-1-52	10-14-52		6,492	2.89	3.14	0.92	
S-16	BNL	W-25	-			8 (W)				6,492	-	3.12	-	
	BNL	W-25	Co-Al	7	8.32	22 (W)				6,492	2.86	3.10	0.92	
S-15	BNL	W-25	-			29 (W)				6,492	-	2.93	-	
	BNL	W-25	Co-Al	5	6.78	37 (W)				6,492	2.65	2.83	0.94	
S-13	BNL	E-25	-			41 (E)		10-21-52		9,738	-	4.14	-	
	BNL	E-25	Co-Al	6	6.15	49.5 (E)		10-21-52		9,738	4.27	3.86	1.10	
S-17	ORNL	19	-	-	-	-					-	2.4 ± 10%	-	
S-18	MTR	VG-51	-	-	-	-	12-11-52	1-1-53		23,896	-	-	-	0.008 ⁽⁴⁾
	MTR	VG-51	Np		474.54	+16*					-	-	-	
	MTR	VG-51	Co-Al	-	7.05	-17*					2.19	-	-	
	MTR	VG-51	Co-Al	-	7.10	+11*					1.98	-	-	
S-19	MTR	VH-3					1.5-Hr. Exposures Between 3-20-53 and 3-22-53							
	MTR	VH-3	Np		61.74		2-18-53	2-19-53	28.6		-	-	-	(5)
	MTR	VH-3	Co-Al	FC-1	6.95	**	3-20-53	3-20-53	1.5		0.74	-	-	
	MTR	VH-3	Co-Al	FC-2	7.11	**	3-21-53	3-21-53	1.5		0.76	-	-	
	MTR	VH-3	Co-Al	FC-4	8.54	**	3-22-53	3-22-53	3.75		2.10	-	-	
S-22	BNL	E-25	Co-Al	D	8.25	1	12-2-52	1-14-53	825	18,982	7.7	9.22	0.84	
S-23	BNL	E-25	-			8 (E)					-	7.8	-	
	BNL	W-25	-			8 (W)					-	7.6	-	
	BNL	W-25	Co-Al	B	6.95	14 (W)					7.6	9.10	0.84	
S-20	BNL	E-25	-			24 (E)					-	7.9	-	
S-21	BNL	W-25	-			24 (W)					-	7.5	-	
	BNL	E-25	Co-Al	A	8.10	33 (E)					7.4	8.45	0.86	
	BNL	W-25	Co-Al	C	8.35	33 (W)					8.0	8.45	0.95	
S-26	BNL	E-25	Co-Al	F	7.20	2 (E)	3-10-53	3-21-53	229	5,747	2.62	2.80	0.94	
S-24	BNL	E-25	-			9 (E)					-	2.78	-	
	BNL	E-25	-			25 (E)					-	2.67	-	
	BNL	E-25	Np		81.84	36 (E)					-	2.52	-	0.45 ⁽³⁾
S-25	BNL	E-25	-			50 (E)					-	2.29	-	
			E		7.20	77 (E)					1.68	1.62	1.04	
S-27	MTR	VH-3					In series with S-19					-	-	
S-28	MTR	VH-3				**	3.75 to 19.5 Hr. Exposures					-	-	
S-29	MTR	VH-3				**	Between 7-2-53 and 7-9-53					-	-	
	MTR	VH-3	Co-Al	E (C)	7.05	**	7-2-53	7-2-53	3.75		1.82	-	-	
	MTR	VH-3	Co-Al	F	8.35	**					1.85	-	-	
	MTR	VH-3	Co-Al	F (B)	8.60	**	7-5-53	7-5-53	9.5		4.57	-	-	
	MTR	VH-3	Co-Al	G	6.90	**					4.62	-	-	
	MTR	VH-3	Co-Al	G (A)	8.55	**	7-9-53	7-9-53	19.5		9.48	-	-	
	MTR	VH-3	Co-Al	H	6.95	**					9.36	-	-	

(1) 0.62% Cobalt

(2) Neptunium²³⁷

(3) Fast Flux as Per Cent of Thermal Flux in BNL Hole 25

$$a. \frac{1.8 \times 10^{18}}{7.82 \times 10^{18}} \times 100 = 23\%$$

$$b. \frac{0.45 \times 10^{18}}{2.52 \times 10^{18}} \times 100 = 18\%$$

* From $\bar{G}$ of lattice in VG-51 [(-) = below $\bar{G}$; (+) = above]

** Exposures at "stop" in VH-3 facility

(4) Fast Flux as Per Cent of Thermal Flux in MTR VG-51

$$\frac{0.008 \times 10^{18}}{2 \times 10^{18}} \times 100 = 0.4\%$$

(5) Foil being processed

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TABLE 2.2
RESUME OF EXPOSURES

Series	No. of Samples	Container			Atm.	Type	Space	Approx. Temp., °C	E X P O S U R E						Gamma Dosage		Purpose	
		Material	Capacity, ml	Fluid					Neutron Flux, n/cm ² /sec. x 10 ¹²		Gamma Flux, R/Hr. γ/cm ² /sec. x 10 ⁸		Neutron Dosage, n/cm ² x 10 ¹⁸		Gamma Dosage, R x 10 ⁸ γ/cm ² x 10 ¹⁸			
									Thermal	Fast	x 10 ⁸	x 10 ¹²	Thermal	Fast	x 10 ⁸	x 10 ¹⁸		
S-17	15	Mg	31	10	He	ORNL	H-19	30	1.0	0.04	1.4	0.5	2.4	0.096	9.5	1.2	Effect of Flux	** (1)
S-18	15	Mg	18.5	6	He	MTR	VG-51	100	1.2	0.005	1.9	0.67	2	0.008	7.6	0.96	Effect of Flux	
S-19	15	Mg	17	10	He	MTR	VH-3	30	137	9.6	372	123	0.74	0.052	5.3	0.66	Effect of Flux Level	
S-20	18	410 SS	12	5	He	BNL	E-25	170	2.98	0.65	2.57	0.90	7.9	1.74	18.8	2.37	Effect of Dosage	
S-21	18	410 SS	12	5	He	BNL	W-25	315	2.98	0.65	2.57	0.90	7.5	1.65	17.8	2.25	Effect of Dosage	
S-22	18	410 SS	12	5	He	BNL	E-25	170	3.11	0.68	2.65	0.93	7.8	1.72	18.5	2.34	Effect of Dosage and Temp.	
S-23	18	410 SS	12	5	He	BNL	W-25	371	3.11	0.68	2.65	0.93	7.6	1.67	18.0	2.28	Effect of Dosage and Temp.	
S-24	18	410 SS	12	5	He	BNL	E-25	426	3.23	0.71	2.77	0.97	2.67	0.59	6.3	0.80	Effect of Temperature	
S-25	16	Mg	12.2	5	He	BNL	E-25	170	2.76	0.61	2.37	0.83	2.29	0.50	5.5	0.69	Shield Materials and Additives	
S-26	19	410 SS	22	15	He	BNL	E-25	371	3.38	0.74	2.89	1.01	2.78	0.61	6.4	0.81	Larger Sample for Analysis	
S-27	10	Mg	17	10	He	MTR	VH-3	30	137	9.6	372	123	4.1 [†]	0.29 [†]	29.3 [†]	3.69 [†]	Effect of Dosage	(2)
S-28	15	Mg	17	10	He	MTR	VH-3	30	137	9.6	372	123					Repeat of S-19	
S-29	10	Mg	17	10	He	MTR	VH-3	30	137	9.6	372	123					Repeat of S-27	(4)
S-30	18	410 SS	22	10	He	BNL	E-25	385					~2.5				Effect of Temperature	
S-31	18	410 SS	22	10	He	BNL	E-25	399					~2.5					
S-32	18	410 SS	22	10	He	BNL	E-25	412					~2.5					
G-1	16	1020 SS	12	10	Air	Gamma	CRC	25	-	-	0.17	0.06	-	-	5.8	0.73	Conventional Lubricants ⁽⁵⁾	(1) (2,5) (2,5)
G-2	15	Mg	17	10	He	Gamma	MTR	25	-	-	~10	~3.5	-	-	10	1.26	Effect of Flux Components ⁽¹⁾	
G-3	36	Mg	17	10	He	Gamma	MTR	25	-	-	~10	~3.5	-	-	100	12.6	Effect of Gamma Dosage ^(3,1)	
G-4	42	1020 SS	12	10	Air	Gamma	CRC	25	-	-	0.17 [†]	0.06 [†]	-	-	2.25 [†]	2.84 [†]	Study of Thickeners	
G-5	16	Mg	17	10	He	Gamma	MTR	25	-	-	~10	~3.5	-	-	10	1.26		
G-6	16	Mg	17	10	He	Gamma	MTR	25	-	-	~10	~3.5	-	-	100	12.6		
G-7	16	Mg	17	10	He	Gamma	MTR	25	-	-	~10	~3.5	-	-	50	6.3	In Series with G-2, G-3 ⁽²⁾	
G-8	16	Mg	17	10	He	Gamma	MTR	25	-	-	~10	~3.5	-	-	50	6.3	Special Lubricants and Greases ^(2,5)	
G-9	16	Mg	17	10	Air	Gamma	MTR	25	-	-	~10	~3.5	-	-	50	6.3		
G-10	24	Glass	12	10	Air	Gamma	CRC	25	-	-	0.17 [†]	0.06 [†]	-	-	2.25 [†]	2.84 [†]	Study of Thickeners and Lubricants ^(1,5)	

[†] Maximum values

* Approximate level of gamma dosage due to exposure variables

⁽¹⁾ Completed. Information contained in CRC-AEC No. 6, September 1953.

⁽²⁾ Exposure completed. Not evaluated.

⁽³⁾ Exposure completed. Partially evaluated.

⁽⁴⁾ In Reactor 9/25/53. ⁽⁵⁾ To be presented in CRC-AEC No. 7

** S-19 and S-27 received insufficient dosage for purpose intended.

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TABLE 2.3
RESULTS OF IRRADIATIONS OF ORGANIC FLUIDS IN NUCLEAR REACTORS

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Fluid	Container No.	Viscosity, Centistokes				Gas Formed		Index of Damage ++			
		at 100°F		at 210°F				T _i	η _i	T ₁₀	η ₁₀
		Orig.	Irr.	Orig.	Irr.	psi	ml/ml Oil				
Series 17 - Irradiated under helium in closed 410 SS vessels in Hole 19 of ORNL at ~30°C for dosage expressed as thermal neutrons/cm ² of ~2.4 x 10 ¹⁸ .											
Poly(propene oxide)-C + A*	LM-1	45.5	34.4	8.70	7.10	165	8	-	-	197	8.1
Octadecylbenzene + A	LM-2	10.9	17.1	2.72	3.24	117	6	-	-	102	16
Di(2-ethylhexyl) Sebacate + A	LM-3	12.7	24.4	3.40	5.59	175	8	-	-	115	19
Cetane + A	LM-4	3.16	4.37	1.28	1.63	240	12	258	1.25	18	18
Diethyl o-Phthalate + A	LM-5	6.75	12.5	1.80	2.70	66	3	302	1.37	80	19
Biphenyl	LM-6	-	-	1.00	1.13	n11	n11	210	1.13	-	-
s-Butylbenzene	LM-7	0.89	1.34	0.50	0.68	80	4	84	1.53	-	-
Diisooamyl Ether	LM-8	1.02	3.32	0.54	1.20	405	19	103	3.15	-	-
Isobutyl Hexanoate	LM-9	1.30	5.45	0.66	1.69	385	18	136	3.35	-	-
n-Butyl Benzoate	LM-10	2.16	4.48	0.92	1.52	-	-	196	1.67	-	-
n-Butyl Phenyl Ether	LM-11	1.58	2.72	0.66	1.03	80	4	150	1.62	-	-
Poly(propene oxide)-C	LM-12	57.6	Sol1d	10.1	Sol1d	215	10	-	Sol1d	211	Sol1d
Octadecylbenzene	LM-13	11.2	505	2.77	14.1	-	-	-	-	103	420
Diethyl o-Phthalate	LM-14	6.36	18.2	1.64	3.40	83	4	305	1.63	78	33
n-Butyl Hexanoate	LM-15	1.32	6.69	0.67	2.26	325	16	138	4.20	-	-
Series 18 - Irradiated under helium in closed magnesium vessels in VG-51 of the MTR (CRC-2) at ~26°C for dosage expressed as thermal neutrons/cm ² of ~2 x 10 ¹⁸ .											
Poly(propene oxide)-C + A	1	45.5	39.5	8.70	7.50	50	7	-	-	197	8.6
Octadecylbenzene + A	2	10.9	12.4	2.72	2.98	25	4	-	-	102	12
Di(2-ethylhexyl) Sebacate + A	17	12.7	16.2	3.40	4.03	70	10	-	-	115	13
Cetane + A	4	3.16	3.43	1.28	1.37	90	13	258	1.06	18	13
Diethyl o-Phthalate + A	5	6.75	8.75	1.80	2.15	38	5	302	1.17	80	13
Biphenyl	20	-	-	1.00	1.01	n11	n11	210	1.01	-	-
s-Butylbenzene	7	0.89	0.98	0.50	0.53	20	3	84	1.10	-	-
Diisooamyl Ether	8	1.02	1.44	0.54	0.71	135	19	103	1.39	-	-
Isobutyl Hexanoate	9	1.30	2.13	0.66	0.97	110	16	136	1.57	-	-
n-Butyl Benzoate	10	2.16	2.66	0.92	1.05	-	-	196	1.14	-	-
n-Butyl Phenyl Ether	11	1.58	1.67	0.66	0.74	19	3	150	1.09	-	-
Poly(propene oxide)-C	12	57.6	271	10.1	31.8	90	13	-	-	211	31
Octadecylbenzene	13	11.2	15.3	2.77	3.44	35	5	-	-	103	14
Diethyl o-Phthalate	14	6.36	9.34	1.64	2.24	25	4	305	1.19	78	15
n-Butyl Hexanoate	15	1.32	2.03	0.67	0.92	105	15	138	1.47	-	-
Series 19 - Irradiated under helium in closed magnesium vessels in VH-3 of the MTR (CRC-1) at ~30°C for dosage expressed as thermal neutrons/cm ² of ~0.74 x 10 ¹⁸ .											
Poly(propene oxide)-C + A	1	45.5	38.6	8.70	7.37	295	14	-	-	197	8.5
Octadecylbenzene + A	2	10.9	14.7	2.72	3.33	225	11	-	-	102	14
Di(2-ethylhexyl) Sebacate + A	3	12.7	14.5	3.40	3.70	385	18	-	-	115	11
Cetane + A	4	3.16	3.98	1.28	1.55	-	-	258	1.18	18	16
Diethyl o-Phthalate + A	5	6.75	10.1	1.80	2.40	138	7	302	1.27	80	15
Biphenyl	6	-	-	1.00	1.04	-	-	210	1.04	-	-
s-Butylbenzene	7	0.89	1.07	0.50	0.58	103	5	84	1.20	-	-
Diisooamyl Ether	8	1.02	1.67	0.54	0.83	-	-	103	1.63	-	-
Isobutyl Hexanoate	9	1.30	3.55	0.66	1.72	510	24	136	2.68	-	-
n-Butyl Benzoate	10	2.16	3.26	0.92	1.21	115	6	196	1.33	-	-
n-Butyl Phenyl Ether	11	1.58	1.89	0.66	0.85	87	4	150	1.28	-	-
Poly(propene oxide)-C	12	57.6	3150	10.1	234	345	17	-	-	211	225
Octadecylbenzene	13	11.2	27.3	2.77	5.27	255	12	-	-	103	25
Diethyl o-Phthalate	14	6.36	13.0	1.64	2.73	110	5	305	1.37	78	22
n-Butyl Hexanoate	15	1.32	2.69	0.67	1.17	585	28	138	1.89	-	-

* A = 5 Wt. per cent didodecyl selenide, 5% idonaphthalene, and 0.1% quinizarin.

++ η = Viscosity of irradiated material at temperature, T, at which original viscosity is one (η_i) or ten (η₁₀) centistokes.

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TABLE 2.3
RESULTS OF IRRADIATIONS OF ORGANIC FLUIDS IN NUCLEAR REACTORS

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Fluid	Container No.	Viscosity, Centistokes				Gas Formed		Index of Damage ⁺⁺			
		at 100°F		at 210°F				T ₁	η ₁	T ₁₀	η ₁₀
		Orig.	Irr.	Orig.	Irr.	psi	ml/ml Oil				
<u>Series 20 - Irradiated under helium in 410 SS closed vessels in Hole E-25 of BNL at ~170°C for dosage expressed as thermal neutrons/cm² of ~7.2 x 10¹⁶.</u>											
Biphenyl	1	Solid	-	1.00	7.28	78	7	210	7.3	-	-
Phenanthrene	2	1.38*	-	0.96**	25.2	47	4	310	5.7	-	-
3-Methylbiphenyl	3	3.22	-	1.11	-	8	1	-	-	46	Empty
Amylbiphenyl	4	10.8	-	2.21	198.3	635	61	325	16	103	3 x 10*
Dowtherm "A"	5	2.65	13.9*	1.05	28.8	150	14	218	24	22	4 x 10*
Naphthalene	6	0.66*	-	0.80	6.33	63	6	170	10	Partially Lost	
1-Fluoronaphthalene	7	1.55	-	0.71	35.1	27	3	153	140	Partially Lost	
1-Phenylnaphthalene	8	17.1	27.6*	2.51	70.3	109	10	322	8	120	3 x 10*
1,2-Diphenylbenzene	9	2.64*	-	4.42	130.0	78	7	372	6	166	900
Chrysene	10	Solid	-	Solid	-	10	1	m.p. = orig. 252-254, irr. 238-240°C			
Amylbiphenyl + 5% O-1*, 0.1% D-2*	11	9.55	-	2.10	116	605	58	-	-	98	~2 x 10*
Amylbiphenyl + 5% R-6*	12	9.29	-	2.01	-	8	1	Capsule Empty			
Amylbiphenyl + 5% O-1, 5% R-6, 0.1% D-2	13	-	-	-	-	-	-	Disc Blown			
Phenylcyclohexane	14	2.09	-	0.92	124	535	51	195	150	Partially Lost	
Biphenyl + 5% O-1	15	0.82*	5.16*	1.04	8.40	117	11	218	7.5	-	-
Amylbiphenyl + 5% O-1	16	9.51	-	2.10	308	678	64	-	-	98	~1 x 10*
Octadecylbenzene + 5% O-1, 5% R-6, 0.1% D-2	17	10.9	-	2.72	-	-	-	Disc Blown			
Pyrene	18	Solid	-	Solid	-	51	5	m.p. = orig. 142-147, irr. 113-117°C			
<u>Series 21 - Irradiated under helium in closed 410 SS vessels in E-25 of the Brookhaven Reactor at ~315°C for dosage expressed as thermal neutrons/cm² of ~7.5 x 10¹⁶.</u>											
Biphenyl	37	Solid	-	1.00	27.1	203	19	210	27	Partially Lost	
Phenanthrene	38	1.38*	23.3*	0.96**	58.0	-	-	310	8.5	-	-
3-Methylbiphenyl	39	3.22	27.2*	1.11	45.6	635	61	225	37	-	-
Amylbiphenyl	40	10.8	-	2.21	-	-	-	Disc Blown			
Dowtherm "A"	41	2.6	-	1.05	-	425	41	Solid			
Naphthalene	42	Solid	-	0.80	-	-	-	Sample Lost			
1-Fluoronaphthalene	43	1.55	-	0.71	-	-	-	Disc Blown			
1-Phenylnaphthalene	44	17.1	-	2.51	44.7	318	33	322	8	120	3000
1,2-Diphenylbenzene	45	2.64*	17.3*	4.42	42.4	117	11	372	3.5	166	180
Chrysene	46	-	-	-	-	285	27	m.p. = orig. 252-254, irr. 190-195°C			
Amylbiphenyl + 5% O-1, 0.1% D-2	47	9.55	-	2.10	-	613	59	Black Tar			
Amylbiphenyl + 5% R-6	48	9.29	-	2.01	-	-	-	Disc Blown			
Amylbiphenyl + 5% O-1, 5% R-6, 0.1% D-2	49	-	-	-	-	-	-	Disc Blown			
Phenylcyclohexane	50	2.09	-	0.92	-	1235	116	Partially Lost			
Biphenyl + 5% O-1	51	0.82*	-	1.04	44.0	312	30	218	36	-	-
Amylbiphenyl + 5% O-1	52	9.51	-	2.10	-	855	83	Black Tar			
Octadecylbenzene + 5% O-1, 5% R-6, 0.1% D-2	53	10.9	-	2.72	-	-	-	Disc Blown			
Pyrene	54	-	-	-	-	125	12	m.p. = orig. 142-147, irr. 119-121°C			
<u>Series 22 - Irradiated under helium in closed 410 SS vessels in E-25 of the Brookhaven Reactor at ~170°C for dosage expressed as thermal neutrons/cm² of ~7.8 x 10¹⁶.</u>											
Biphenyl	19	Solid	-	1.00	11.3	59	6	210	11	Partially Lost	
4,4'-Difluorobiphenyl	20	Solid	-	1.07	-	47	5	-	-	Infusible	
3-Methylbiphenyl	21	3.22	13.5*	1.11	21.9	195	19	225	18	-	-
Amylbiphenyl	22	10.8	-	2.21	-	-	-	Disc Blown			
Dowtherm "A"	23	2.65	19.7*	1.05	35.5	149	14	218	31	-	-
Naphthalene	24	0.66*	4.80*	0.80	7.35	81	8	170	13	-	-
1-Fluoronaphthalene	25	1.55	-	0.71	-	41	4	Black Tar			
1-Phenylnaphthalene	26	17.1	-	2.51	122	117	11	322	12	120	9000
1,2-Diphenylbenzene	27	2.64*	56.9*	4.42	202.2	61	6	372	8	166	1700
Chrysene	28	-	-	-	-	24	2	m.p. = orig. 252-254°C, irr. 190-195°C			
Amylbiphenyl + 5% O-1, 0.1% D-2	29	9.55	-	2.10	-	-	-	Disc Blown			
Amylbiphenyl + 5% R-6	30	9.29	-	2.01	-	-	-	Disc Blown			
Amylbiphenyl + 5% O-1, 5% R-6, 0.1% D-2	31	-	-	-	-	-	-	Disc Blown			
Phenylcyclohexane	32	2.09	-	0.92	-	-	-	Disc Blown			
Biphenyl + 5% O-1	33	0.82*	7.33*	1.04	12.8	161	15	218	11	-	-
Amylbiphenyl + 5% O-1	34	9.51	-	2.10	-	885	94	Solid			
Octadecylbenzene + 5% O-1, 5% R-6, 0.1% D-2	35	10.9	-	2.72	-	-	-	Disc Blown			
Pyrene	36	Solid	-	Solid	-	61	6	m.p. = orig. 142-147, irr. 105-110°C			

⁺⁺ η = Viscosity of irradiated material at temperature, T, at which original viscosity is one (η₁) or ten (η₁₀) centistokes.

* Viscosity at 250°F

** Viscosity at 318°F

* Additive Code: (Per cent by weight)

O-1 = Didodecyl Selenide
O-2 = Phenyl Dibutyl Dithiophosphate
O-15 = Phenothiazine
D-2 = Quinizarin
R-6 = Iodonaphthalene

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TABLE 2.3

RESULTS OF IRRADIATIONS OF ORGANIC FLUIDS IN NUCLEAR REACTORS

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Fluid	Container No.	Viscosity, Centistokes				Gas Formed		Index of Damage ⁺⁺			
		at 100°F		at 210°F		psi	ml/ml Oil	T.	η ₁	T ₁₀	η ₁₀
		Orig.	Irr.	Orig.	Irr.						
Series 23 - Irradiated under helium in closed 410 SS vessels in E-25 of the Brookhaven Reactor at ~371°C for dosage expressed as thermal neutrons/cm ² of ~7.6 x 10 ¹⁸ .											
Biphenyl	55	Solid	-	1.00	16.1	195	19	210	16	-	-
4,4'-Difluorobiphenyl	56	-	-	-	-	-	-		Leak		
3-Methylbiphenyl	57	3.22	-	1.11	-	-	-		Disc Blown		
Amylbiphenyl	58	10.8	-	2.21	-	-	-		Disc Blown		
Dowtherm "A"	59	2.65	-	1.05	-	-	-		Disc Blown		
Naphthalene	60	Solid	-	0.80	-	-	-		Disc Blown		
1-Fluoronaphthalene	61	1.55	-	0.71	-	-	-		Disc Blown		
1-Phenyl naphthalene	62	17.1	-	2.51	-	580	55		Black, high m.p. solid		
1,2-Diphenylbenzene	63	-	-	4.42	-	610	58		Coke		
Chrysene	64	Solid	-	Solid	-	205	20		m.p. = orig. 252-254, irr. 195-198°C		
Amylbiphenyl + 5% 0-1*, 0.1% D-2	65	9.55	-	2.10	-	-	-		Disc Blown		
Amylbiphenyl + 5% R-6	66	9.29	-	2.01	-	360	34		Coke		
Amylbiphenyl + 5% 0-1, 5% R-6, 0.1% D-2	67	-	-	-	-	-	-		Disc Blown		
Phenylcyclohexane	68	2.09	-	0.92	-	-	-		Disc Blown		
Biphenyl + 5% 0-1	69	0.82*	-	1.04	-	-	-		Disc Blown		
Amylbiphenyl + 5% 0-1	70	9.51	-	2.12	-	-	-		Disc Blown		
Octadecylbenzene + 5% 0-1, 5% R-6, 0.1% D-2	71	10.9	-	2.72	-	-	-		Disc Blown		
Pyrene	72	Solid	-	Solid	-	113	11		Coke		
Series 24 - Irradiated under helium in closed 410 SS vessels in E-25 of the Brookhaven Reactor at ~426°C for dosage expressed as thermal neutrons/cm ² of ~2.67 x 10 ¹⁸ .											
3-Methylbiphenyl	61	3.29	-	1.18	-	355	35		Coke		
Amylbiphenyl	62	9.78	-	2.08	-	-	-		Disc Blown		
Biphenyl	63	Solid	-	1.00	-	445	42		Heavy Asphalt		
1-Phenyl naphthalene	64	17.1	-	2.51	-	-	-		Disc Blown		
Octadecylbenzene + 5% 0-1, 2% R-6, 0.1% D-2	65	11.6	-	2.78	-	-	-		Disc Blown		
Naphthalene	66	Solid	-	0.80	-	485	46		Coke		
Dowtherm "A"	67	2.64	-	1.01	-	n11	n11		Coke		
1,3-Diphenylbenzene	68	Solid	-	3.71	-	405	39		Coke		
Phenylcyclohexane	69	2.09	-	0.92	-	-	-		Disc Blown		
1-Fluoronaphthalene	55	1.55	-	1.81	-	-	-		Disc Blown		
1-Methylnaphthalene	56	2.09	-	0.88	-	412	39		Coke		
2-Methylnaphthalene	57	-	-	-	-	-	-		Disc Blown		
1,2-Diphenylbenzene	58	Solid	-	4.42	-	235	22		Low m.p., High Vis. at 100°C		
Acenaphthene	59	-	-	1.58	-	605	58		Coke		
3-Methylbiphenyl + 5% 0-1	60	3.37	-	1.34	-	685	65		High m.p. solid		
Tetraphenylsilane	C-9	-	-	Solid	-	72	7		Rubbery Solid		
Tetrabenzylsilane	C-19	-	-	Solid	28.0	408	39		Black, Semisolid		
Triphenylmethylsilane	77	-	-	3.60	-	282	27		Infusible Plastic		
Series 25 - Irradiated under helium in closed magnesium vessels in E-25 of the Brookhaven Reactor at ~170°C for dosage expressed as thermal neutrons/cm ² of ~2.29 x 10 ¹⁸ .											
n-Dodecylamine	21M	3.28	-	1.25	-	n11	n11		Rubbery at 100°C		
Di-n-hexylamine	33M	2.01	-	0.89	-	n11	n11		Sample Lost		
n-Hexylamine	37M	0.93	22.0	0.48	4.77	-	-	92	30	-	-
2-Phenylethylamine	1M	1.92	-	0.80	23.8	100	10	176	50	-	-
Tributylamine	12M	1.35	7.08	0.65	2.14	275	27	137	4.6	-	-
n-Dodecylamine + 5% 0-1	2M	2.88	-	1.19	-	n11	-		Solid at 100°C		
Dodecanol	7M	12.31	169	2.40	15.2	8	1	333	4	110	123
Dodecanol + 5% 0-1	4M	12.6	-	2.27	-	n11	n11		Solid at 100°C		
Mixed Alkylbenzenes (MW ~250) + 5% 0-1, 2% S-16, 0.1% D-2	6M	14.0	19.8	2.67	3.65	185	18	-	-	116	14
Mixed Alkylbenzenes (MW ~250) + 5% 0-1, 4% S-14, 0.1% D-2	2M	16.5	14.9	4.17	2.97	19	2	-	-	132	9.4
Mixed Alkylbenzenes (MW ~250) + 5% 0-1, 2% S-16, 2% R-6, 0.1% D-2	46M	13.7	11.5	2.77	2.69	315	31	-	-	116	8.6
Mixed Alkylbenzenes (MW ~250) + 5% 0-1, 0.1% D-2	30M	7.55	11.9	1.98	2.80	12	1	-	-	85	16
Mixed Alkylbenzenes (MW ~250) + 2% 0-1, 1% 0-15, 0.1% D-2	32M	7.52	23.4	1.93	4.28	370	36	-	-	86	33
Mixed Alkylbenzenes (MW ~250) + 5% 0-2, 0.1% D-2	45M	8.41	16.0	2.06	3.27	4	4	-	-	92	20
Dibenzothiophene	C-20**	Solid	Solid	1.91	2.94	n11	n11	~310	~1.37	~85	~25
Triphenylmethylsilane	52**	2.30*	330	3.60	11.4	23	2	-	-	149	49

*Additive Code: (Per cent by weight)

0-1 = Didodecyl Selenide
 0-2 = Phenyl Dibutyl Dithiophosphate
 0-15 = Phenothiazine
 D-2 = Quinizarin
 R-6 = Iodonaphthalene
 S-14 = Poly(lauryl methacrylate)
 S-16 = Poly(alpha-methyl styrene)

⁺⁺ η = Viscosity of irradiated material at temperature, T, at which original viscosity is one (η_1) or ten (η_{10}) centistokes.

** 410 Stainless vessel

† Viscosity at 250°F

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TABLE 2.4

IRRADIATIONS IN PROGRESS*

Series 30, 31, and 32 - Exposed in 410 SS
Vessels in Brookhaven Reactor for an
Approximate Dosage of 2.5×10^{18} n/cm².

Identity	Melting Point, °C	Container No.		
		S-30	S-31	S-32
		371°C (725°F)	399°C (750°F)	426°C (775°F)
Biphenyl	71	1	19	37
Naphthalene	79	2	20	38
1,3-Diphenylbenzene	87	3	21	39
1,4-Diphenylbenzene	213	4	22	40
Biphenyl-Naphthalene (56-44 wt. per cent)	39	5	23	41
Biphenyl-1,3-Diphenylbenzene (50-50 wt. per cent)	44	6	24	42
1,3,5-Triphenylbenzene	176	7	25	43
Phenanthrene	99	8	26	44
Biphenyl-1,3-Diphenylbenzene** + 2% Naphthalene	-	9	-	-
Diphenylacetylene	60	10	27	45
Fluorolube	(Liquid)	-	28	46
Dowtherm "A"	(Liquid)	11	29	47
Triphenyl Phosphate	50	12	30	48
Triphenylbenzylmethane	143	13	31	49
Triphenylbenzylsilane	100	14	32	50
Tetrabenzylsilane	128	15	33	51
Diphenyldibenzylsilane	60	16	34	52
Polybenzyl	~85	-	35	-
Biphenyl-1,3-Diphenylbenzene** + 5% Dibenzyl Selenide	-	17	36	53
Biphenyl-1,3-Diphenylbenzene** + 5% Thianthrene	-	18	-	54

*Exposures completed October 7, 1953;
evaluations in progress.

**50-50 Wt. per cent.

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TABLE 2.5

RESULTS OF GAMMA IRRADIATIONS OF ORGANIC FLUIDS

Fluid	Container No.	Dosage: **R x 10 ⁸	Viscosity, Centistokes				Gas Formed		Index of Damage ⁺⁺			
			at 100°F		at 210°F							
			Orig.	Irr.	Orig.	Irr.	psi	ml/ml Oil	T ₁	η ₁	T ₁₀	η ₁₀
Series G-2 (CRC-501-1) - Irradiated under helium in closed magnesium vessels in the MTR gamma facility at ~25°C for dosages as indicated.												
Poly(propene oxide)-C + A*	31	8.4	45.5	38.3	8.70	7.20	185	9	-	-	197	8.4
Octadecylbenzene + A	32	10.5	10.9	13.5	2.72	3.14	135	7	-	-	105	13
Di(2-ethylhexyl) Sebacate + A	33	10.4	12.7	14.4	3.40	3.76	235	11	-	-	115	11
Cetane + A	34	10.5	3.16	3.61	1.28	1.42	375	18	257	1.09	25	12
Diethyl o-Phthalate + A	35	10.5	6.75	9.16	1.80	2.22	115	6	302	1.18	80	14
Biphenyl	36	8.4	-	-	1.00	1.01	n11	n11	270	1.01	-	-
s-Butylbenzene	37	10.4	0.89	1.00	0.50	0.54	42	2	83	1.13	-	-
Diisoamyl Ether	38	8.4	1.02	1.55	0.54	0.75	365	17	103	1.52	-	-
Isobutyl Hexanoate	39	10.5	1.30	5.50	0.66	0.76	325	16	137	2.15	-	-
n-Butyl Benzoate	40	8.4	2.16	2.87	0.92	1.13	47	2	197	1.23	6	16
n-Butyl Phenyl Ether	41	10.4	1.58	1.77	0.66	0.76	50	2	150	1.13	-2	10
Poly(propene oxide)-C	42	9.5	57.6	905	10.1	91.5	210	10	-	-	210	91
Octadecylbenzene	43	9.5	11.2	17.9	2.77	3.94	119	6	-	-	107	16
Diethyl o-Phthalate	44	9.5	6.36	11.7	1.64	2.59	63	3	283	1.50	78	19
n-Butyl Hexanoate	45	10.4	1.32	2.89	0.67	1.20	385	18	139	1.97	-40	55
Series G-3 (CRC-501-2) - Irradiated under helium in closed magnesium vessels in the MTR gamma facility at ~25°C for dosages as indicated.												
Poly(propene oxide)-C + A	46	40	45.5	20.2	8.70	4.79	435	21	-	-	197	5.4
Octadecylbenzene + A	47	40	10.9	21.7	2.72	4.36	365	17	-	-	105	19
Di(2-ethylhexyl) Sebacate + A	48	40	12.7	47.8	3.40	9.17	-	-	-	-	115	36
Cetane + A	49	40	3.16	5.41	1.28	1.95	835	40	257	1.48	25	20
Diethyl o-Phthalate + A	50	71	6.75	468	1.80	19.2	405	19	302	5.3	80	1250
Biphenyl	51	71	-	-	1.00	1.13	n11	n11	210	1.13	-	-
s-Butylbenzene	52	71	0.89	2.03	0.50	0.93	275	13	83	2.42	-	-
Diisoamyl Ether	53	71	1.02	14.1	0.54	4.57	1700	81	103	13.6	-	-
Isobutyl Hexanoate	54	88	1.30	-	0.66	-	535	26	Solid			
n-Butyl Benzoate	55	88	2.16	1866	0.92	67.2	365	17	197	90	-	-
n-Butyl Phenyl Ether	56	88	1.58	17.0	0.66	3.03	305	15	150	6.5	-	-
Poly(propene oxide)-C	57	88	57.6	-	10.1	-	1950	93	Solid			
Octadecylbenzene	58	96	11.2	-	2.77	-	1400	67	Solid			
Diethyl o-Phthalate	59	96	6.36	-	1.64	-	515	25	Solid			
n-Butyl Hexanoate	60	96	1.32	-	0.67	-	-	-	Viscous Liquid Lost			

⁺⁺ η = Viscosity of irradiated material at temperature, T, at which original viscosity is one (η_1) or ten (η_{10}) centistokes.

** One R = 1.26×10^8 gamma/cm²

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TABLE 2.6
IRRADIATION OF ORGANIC FLUIDS TO CO⁶⁰ GAMMA RAYS⁽¹⁾ AT 25°C

Thickener(2)	Container No.	Exposure		Viscosity, Centistokes		Viscosity Index	Index of Damage	
		Days	R x 10 ⁶	at 100°F	at 210°F		T ₁₀	Y ₁₀
<u>SERIES G-4 - Open to air - 1020 steel containers</u>								
<u>30% S-16</u>	-	0	0	19.6	2.98	-95	129	10.0
	3-A	9.19	0.37	19.8	3.17	-50		10.3
	3-A	21.25	0.86	20.0	3.19	-45		10.4
	3-A	21.80	0.88	20.1	3.20	-45		10.5
	3-B	22.33	0.90	20.1	3.19	-45		10.5
	3-B	30.99	1.26	20.3	3.21	-45		10.6
	3-C	52.24	2.12	20.8	-	-		-
	1-A	76.46	3.10	20.9	3.27	-45		11
	1-B	89.07	3.61	21.0	3.28	-45		11
	1-C	98.26	3.98	21.3	3.30	-45		11.2
<u>4% S-14</u>	-	0	0	17.3	4.35	180	135	10.0
	1-A	9.19	0.37	12.2	3.06	120		7.0
	1-A, 1-B	21.25	0.86	11.1	2.74	93		6.3
	1-A	21.80	0.88	10.6	2.65	90		6.0
	1-B	30.99	1.26	10.1	2.51	76		5.8
	1-C, 1-D	52.24	2.12	9.98	2.46	65		5.7
	2-A	76.46	3.10	9.81	2.39	56		5.5
	2-B	89.07	3.61	9.81	2.38	55		5.5
	2-C	98.26	3.98	9.94	2.39	52		5.6
	<u>8% S-15</u>	-	0	0	16.5	3.86	147	130
2-A		9.19	0.37	15.5	3.42	109		9.1
2-A, 2-B		21.25	0.86	14.4	3.20	95		8.4
2-A		21.80	0.88	14.6	3.25	97		8.6
2-B		30.99	1.26	14.0	3.11	88		8.2
2-C, 2-D		52.24	2.12	13.4	2.96	70		7.8
3-A		76.46	3.10	13.2	2.90	65		7.7
3-B		89.07	3.61	13.0	2.88	67		7.5
3-C		98.26	3.98	13.0	2.86	62		7.5
<u>8% S-17</u>		-	0	0	18.6	4.27	160	137
	4-A	9.19	0.37	17.4	4.25	175		9.7
	4-A, 4-B	21.25	0.86	15.9	3.84	155		8.8
	4-A	21.80	0.88	16.2	3.94	165		9.0
	4-B	30.99	1.26	15.3	3.69	150		8.5
	4-C	52.24	2.12	14.5	-	-		-
	4-A	76.46	3.10	13.4	3.17	112		7.3
	4-B	89.07	3.61	13.2	3.13	110		7.2
	4-C	98.26	3.98	13.0	3.07	105		7.1
	<u>Base Oil</u>	-	0	0	7.55	1.97	30	86
1-D		67.27	2.73	7.89	2.02	30		10.5
<u>SERIES G-10 - Open to air - Pyrex containers</u>								
<u>4% S-18</u>	-	0	0	14.3	3.72	170	130	10.0
	15	1	0.04	14.1	3.67	170		9.0
	13	5	0.20	13.3	3.47	160		8.4
	15	10	0.41	12.2	3.17	140		7.7
	15	20	0.82	11.2	2.87	115		7.0
	16	40	1.63	10.0	2.65	110		6.4
	10	47	1.90	9.8	2.60	105		6.2
	<u>Fluid "X" (a)</u>	-	0	0	34.0	6.02	130	170
12		1	0.04	34.1	6.03	130		10
15		5	0.20	34.0	5.97	128		10
13		10	0.41	34.0	5.94	128		9.9
12		20	0.82	34.2	5.87	123		9.8
14		40	1.63	35.2	5.90	120		9.7
9		47	1.90	33.2	5.57	115		9.2

⁽¹⁾ 1.69 x 10⁶ R/Hr. (4.06 R/Day) - 600-Curie Source

⁽²⁾ Base Oil: Mixed Alkylbenzenes (MW ~250); 5% Didodecyl Selenide
0.1% Quinizarin

⁽³⁾ 50% Poly(propene oxide)-C; 45% Mixed Alkylbenzenes (MW ~350), 5% Didodecyl Selenide,
0.05% Quinizarin

Additive Code:

S-14 Poly(lauryl methacrylate)
S-15 Polybutene
S-16 Poly(alpha-methyl styrene)
S-17 Styrene-Butene Copolymer
S-18 Poly(alkane oxide)

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TABLE 2.7

GAMMA IRRADIATIONS IN PROGRESS -
THE EFFECT OF THICKENERS AND OF CHEMICAL COMPOSITION

Base Fluid	INHIBITORS*								Container No.		
	Oxidation		Radical Reaction		Metal Deactivator		Special				
	%	No.	%	No.	%	No.	%	No.	G-5	G-6	
<u>Series G-5 (CRC-501-3) for ~10⁸ R and Series G-6 (CRC-501-4) for ~10¹⁰ R exposure in MTR facility at 25°C in magnesium containers sealed under helium.</u>											
Mixed Alkylbenzenes (MW ~250)	5	0-1	-	-	0.1	D-2	30	S-16	1	17	
↓	5	0-1	-	-	0.1	D-2	-	-	2	18	
	5	0-1	-	-	0.1	D-2	4	S-14	3	19	
	5	0-1	-	-	0.1	D-2	8	S-15	4	20	
	5	0-1	-	-	0.1	D-2	8	S-17	5	21	
	5	0-1	-	-	0.1	D-2	4	S-18	6	22	
	5	0-1	-	-	0.1	D-2	{30 2.5	S-16 S-17	7	23	
	-	-	-	-	-	-	-	-	8	24	
	5	0-1	-	-	0.1	D-2	50	S-16	9	25	
	5	0-1	-	-	0.1	D-2	20	S-16	10	26	
	5	0-1	-	-	0.1	D-2	10	S-16	11	27	
	5	0-1	-	-	0.1	D-2	30	S-17	12	28	
	5	0-1	-	-	0.1	D-2	20	S-17	13	29	
	Mixed Alkylbenzenes (MW ~350)	5	0-1	-	-	0.1	D-2	-	-	14	30
	Mixed Alkylbenzenes (MW ~350)	5	0-1	-	-	0.1	D-2	20	S-16	15	31
	Amylbiphenyl	5	0-1	-	-	0.1	D-2	35	S-16	16	32
<u>Series G-7 (CRC-501-5) for ~5 x 10⁸ R exposure in MTR facility at 25°C in magnesium containers sealed under helium.</u>											
									G-7		
Poly(propene oxide)-C	5	0-1	5	R-6	0.1	D-2	-	-		33	
Octadecylbenzene	5	0-1	5	R-6	0.1	D-2	-	-		34	
Di(2-ethylhexyl) Sebacate	5	0-1	5	R-6	0.1	D-2	-	-		35	
Cetane	5	0-1	5	R-6	0.1	D-2	-	-		36	
Diethyl o-Phthalate	5	0-1	5	R-6	0.1	D-2	-	-		37	
Biphenyl	-	-	-	-	-	-	-	-		38	
s-Butylbenzene	-	-	-	-	-	-	-	-		39	
Diisooamyl Ether	-	-	-	-	-	-	-	-		40	
Isobutyl Hexanoate	-	-	-	-	-	-	-	-		41	
n-Butyl Benzoate	-	-	-	-	-	-	-	-		42	
n-Butyl Phenyl Ether	-	-	-	-	-	-	-	-		43	
Poly(propene oxide)-C (SL-464)	-	-	-	-	-	-	-	-		44	
Octadecylbenzene	-	-	-	-	-	-	-	-		45	
Diethyl o-Phthalate	-	-	-	-	-	-	-	-		46	
n-Butyl Hexanoate	-	-	-	-	-	-	-	-		47	
Amylbiphenyl	5	0-1	-	-	0.1	D-2	35	S-16		48	

*Additive Code:

0-1 Didodecyl Selenide
D-2 Quinizarin

S-14 Poly(lauryl methacrylate)
S-15 Polybutene

S-16 Poly(alpha-methyl styrene)
S-17 Styrene-Butene Copolymer
S-18 Poly(alkene oxide)

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TABLE 2.9
EFFECT OF FLUX COMPONENTS ON THE THICKENING OF ORGANIC FLUIDS

Fluid	Index of Damage						Index of Gamma Damage ⁽¹⁾									
	S-17		S-19		S-G-2		S-18		S-17		S-19		S-G-2		S-18	
	η_1	η_{10}	η_1	η_{10}	η_1	η_{10}	η_1	η_{10}	η_1	η_{10}	η_1	η_{10}	η_1	η_{10}	η_1	η_{10}
Poly(propene oxide)-C; A *	-	8.1	-	8.5	-	8.4	-	8.6	-	8.3		9	-	8.4	-	8.5
Octadecylbenzene; A	-	16	-	14	-	13	-	12	-	12.6		11.4	-	13	-	12
Di(2-ethylhexyl) Sebacate; A	-	19	-	11	-	11	-	13	-	11.1		10.6	-	11	-	10.9
Cetane; A	1.25	18	1.18	16	1.09	12	1.06	13	1.08	11.9	1.04	11.0	1.09	12	1.06	11.5
Diethyl o-Phthalate; A	1.37	19	1.27	15	1.18	14	1.17	13	1.16	13.5	1.08	11.7	1.18	14	1.13	12.6
Biphenyl	1.13	-	1.04	-	1.01	-	1.01	-	1.01	-	1.01	-	1.01	-	1.01	-
s-Butylbenzene	1.53	-	1.20	-	1.13	-	1.10	-	1.11	-	1.06	-	1.13	-	1.08	-
Diisocamyl Ether	3.15	-	1.63	-	1.52	-	1.39	-	1.60	-	1.30	-	1.52	-	1.45	-
Isobutyl Hexanoate	3.35	-	2.68	-	2.15	-	1.57	-	2.0	-	1.47	-	2.15	-	1.74	-
n-Butyl Benzoate	1.67	-	1.33	-	1.23	-	1.14	-	1.31	-	1.13	-	1.23	-	1.20	-
n-Butyl Phenyl Ether	1.62	-	1.28	-	1.13	-	1.09	-	1.11	-	1.06	-	1.13	-	1.08	-
Poly(propene oxide)-C	Solid	-	-	-	91	-	31	-	91.5	-	34.4	-	91	-	59	-
Octadecylbenzene	-	420	-	225	-	16	-	14	-	15.7	-	12.8	-	16	-	14.3
Diethyl o-Phthalate	1.63	33	1.37	22	1.50	19	1.19	15	1.50	18.9	1.25	14.2	1.50	19	1.37	16.5
n-Butyl Hexanoate	4.20	-	1.89	-	1.97	-	1.47	-	1.85	-	1.41	24	1.97	55	1.63	35
Neutron Portion of Total Damage ⁽²⁾									Ratio: ⁽³⁾ Damage by Neutrons Damage by Gammas							
S-17 S-19 S-18									S-17 S-19 S-18							
$\eta_1 - \eta_{10}$ $\eta_{10} - \eta_{10}$ $\eta_1 - \eta_{10}$ $\eta_{10} - \eta_{10}$ $\eta_1 - \eta_{10}$ $\eta_{10} - \eta_{10}$ $\eta_1 - \eta_{10}$									⁽⁴⁾ $(\eta_1)_x$ ⁽⁴⁾ $(\eta_{10})_x$ $(\eta_1)_x$ $(\eta_{10})_x$ $(\eta_1)_x$ $(\eta_{10})_x$							
Poly(propene oxide)-C; A	-	-0.2	-	-0.5	-	0.1	-	-	-	-	-	-	-	-	-	-
Octadecylbenzene; A	-	3.4	-	2.6	-	0	-	0	-	1.3	-	1.9	-	-	-	-
Di(2-ethylhexyl) Sebacate; A	-	7.9	-	0.4	-	2.1	-	2.1	-	7.2	-	0.7	-	2.3	-	-
Cetane; A	0.17	6.1	0.14	5.0	0	1.5	2.1	3.5	3.5	5.0	-	1.0	-	-	-	-
Diethyl o-Phthalate; A	0.21	5.5	0.19	3.3	0.04	0.4	-	1.6	2.3	0.5	0.30	0.15	-	-	-	-
Biphenyl	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
s-Butylbenzene	0.42	-	0.14	-	0.02	-	3.8	-	2.3	-	0.25	-	-	-	-	-
Diisocamyl Ether	1.55	-	0.33	-	-0.06	-	2.6	-	1.1	-	-0.13	-	-	-	-	-
Isobutyl Hexanoate	1.35	-	1.21	-	-0.17	-	1.4	-	0.5	-	-0.23	-	-	-	-	-
n-Butyl Benzoate	0.36	-	0.20	-	0.06	-	1.2	-	1.5	-	0.30	-	-	-	-	-
n-Butyl Phenyl Ether	0.51	-	0.22	-	0.01	-	4.6	-	3.7	-	0.12	-	-	-	-	-
Poly(propene oxide)-C	Solid	-	-	-	-28	-	-	-	-	-	7.5	-	-0.47	-	-	-
Octadecylbenzene	-	404	-	12.2	-	-0.3	-	-	-	-	4.4	-	-0.07	-	-	-
Diethyl o-Phthalate	0.13	14.1	0.12	8.0	-0.18	-1.5	-	1.6	-	1.9	0.50	-0.23	-	-	-	-
n-Butyl Hexanoate	2.35	-	0.48	-	-0.16	-	2.8	-	1.2	-	-0.25	-	-	-	-	-
Average									2.8±1.3 2.5±1.5 0.3±0.5							
Ratio: $\frac{\text{Gamma Dosage}}{\text{Fast Neutron Dosage}}$									$\frac{12 \times 10^{17}}{96 \times 10^{15}}$ $\frac{6.6 \times 10^{17}}{52 \times 10^{15}}$ $\frac{2.6 \times 10^{17}}{8 \times 10^{15}}$ ↓ ↓ ↓ 12.5 12.7 120							
Ratio of Damage: $\frac{1 \text{ Fast Neutron}^{(4)} > 1 \text{ meV}}{1 \text{ Gamma Photon of } 1.6 \text{ meV}}$									34 31 (31)							
⁽⁴⁾ Damage includes thickening due to neutrons of all energies. Fast neutron flux designated as that greater than 1 meV as determined by ²³⁷ Np. Thermal neutrons exert negligible effect on thickening. Epithermal neutrons below 1 meV are about equivalent to fast neutrons above 1 meV as to thickening effect on organic fluids. See text. Therefore, Ratio of Damage: $\frac{1 \text{ Fast Neutron} > 1 \text{ meV}}{1 \text{ Gamma Photon of } 1.6 \text{ meV}}$																

*A = Containing 5% didodecyl selenide, 5% iodonaphthalene, and 0.1% quinizarin.

(1) = The η_1 's and η_{10} 's were obtained by interpolating at the respective gamma portions of combined dosage for Series 17, 18, and 19 from straight line plots of $\log \eta_1$ vs. gamma dosage for Series G-2.

(2) = By difference.

(3) = Unweighted for flux levels.

(4) = $(\eta_1)_x = \frac{\eta_1 - \eta_{10}}{\eta_{10} - 1}$; $(\eta_{10})_x = \frac{\eta_{10} - \eta_{10}}{\eta_{10} - 1}$

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TABLE 3.0

MATERIALS USED IN THERMAL STABILITY STUDIES

Compound	Source*	Melting Point, °C**
Naphthalene	B. (C.P. anal.)	79
Anthracene	E. No. 480	215-217
Phenanthrene	M. No. 3468	97-99
Chrysene	E. No. 4217	(253-254)
Pyrene	M. No. 1262	148-149
Biphenyl	E. No. 721	69.5-70.5
o-Terphenyl	E. No. 2301	56-57
m-Terphenyl	M. No. 7076	86-87
p-Terphenyl	E. No. 2103	212-213
p-Quatraphenyl	(Synthesized by N.A.A.)	308-312
1,3,5-Triphenylbenzene	(Synthesized)	174.0-176.3
1-Methylnaphthalene	Reference Fuel Grade (99%)	(-31)
2-Methylnaphthalene	E. No. 2416	34-35
Acenaphthene	E. No. 597	93.5-94.5
1-Fluoronaphthalene	E. No. 3304	(-8)
2-Fluoronaphthalene	E. No. 6168	59-61
Cetane	Reference Fuel Grade	(19)
Decalin	E. No. P-1905 (Redistilled)	(ca -40)
Stilbene	E. No. X-1817	(124)
Triphenylmethane	E. No. 833	93-94
Triphenylbenzylmethane	(Synthesized)	142.8-143.1
Triphenylmethylsilane	(Synthesized)	65.5-67
Triphenylbenzylsilane	(Synthesized)	99.3-99.8
Triphenylphosphate	Mo. -	(49-50)
1,2,4-Trichlorobenzene	E. No. 1641	(17)
Perfluoroheneicosane	-	(Liquid)

* B = Baker Chemical Co.
 E = Eastman Organic Chemicals
 M = Matheson Co.
 Mo. = Monsanto Chemical Co.

** Melting points as given by commercial supplier or determined for synthesized compounds; values in parentheses are literature values.

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TABLE 3.1

EFFECT OF TEMPERATURE ON THERMAL STABILITY
 24-HOUR EXPOSURE - 304 SS VESSEL - 300 PSIG INITIAL NITROGEN PRESSURE

Compound	Run No.	Temp., °F	Gas Evolution*		Moles Gas Per Mole	% Distribution of Products**		
			cc/Gram (Press.)	(M.S.)		Coke	Volatile Solid	Liquid
			(+10)					
Naphthalene	6	980	75	150	0.85 (I)	~57 (r)	~33	-
Naphthalene***	6	980	94	96	0.55 (I)	~60 (r)	~34	-
Naphthalene	8	950	48	49	0.28 (I)	? (r)	<97	-
Naphthalene	3	920	25	2	0.014 (E)	~0.1(r)	>99	-
Naphthalene	12	920	9	3	0.018 (E)	~0.1(a)	>99	-
Biphenyl	6	980	30	28	0.18 (I)	<77 (r)	?	21
Biphenyl***	6	980	44	26	0.17 (I)	<75 (r)	?	23
Biphenyl	8	950	25	16	0.10 (I)	? (r)	<55	44
Biphenyl	3	920	13	4	0.020 (E)	1.4(a)	98	-
p-Terphenyl	8	950	11	27	0.27 (I)	<98 (r)	?	-
p-Terphenyl	10	920	2	7	0.057 (I)	56 (a)	32	11
o-Terphenyl	8	950	48	37	0.37 (I)	<92 (r)	?	6
o-Terphenyl	10	920	11	9	0.081 (I)	39 (a)	35	25

* E = "Equilibrium" value

I = Still increasing

** Coke and volatile solid determined by

(r) = manual separation, (a) = Vacuum distillation

Difference from 100% = Gas

*** 0.55 Wt. Per Cent quinoline added

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TABLE 3.2

THERMAL STABILITY OF POLYPHENYL VS. CONDENSED RING AROMATICS
 TEMPERATURE = 920°F - 24-HOUR EXPOSURE -
 304 SS VESSEL - 300 PSIG INITIAL NITROGEN PRESSURE

Compound	Run No.	Gas Evolution*				% Distribution of Products**			I.R Purity of Volatile Solid, %
		cc/Gram		Moles Gas Per Mole	Hydrocarbon-to-Hydrogen Ratio	Coke	Volatile Solid	Liquid	
		(Press.)	(M.S.)						
		(±10)							
Biphenyl	3	13	4	0.020 (E)	0.25	1.4	98	-	58
m-Terphenyl	10	0	5	0.037 (I)	0.61	35	53	12	36
p-Terphenyl	10	2	7	0.057 (I)	0.68	56	32	11	
o-Terphenyl	10	11	9	0.081 (I)	2.0	39	35	25	
p-Quatraphenyl	21	13	18	0.19 (I)	0.80	76	22	-	
1,3,5-Triphenyl-benzene	26	14	11	0.13 (I)	0.58	73	26	-	95
Naphthalene	12	9	3	0.018 (E)	0.49	0.1	>99	-	
Anthracene	14	116	121	0.96 (E)	8.8	79	12	-	
Phenanthrene	14	103	101	0.81 (I)	2.9	78	14	-	
Chrysene	14	93	93	0.96 (I)	2.6	90	4	-	
Pyrene	14	116	111	0.99 (I)	0.65	87	8	-	

* E = Equilibrium value
 I = Still increasing

** Coke and volatile solid determined by vacuum distillation in every case.

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TABLE 3.3

EFFECT OF SUBSTITUENTS ON THE THERMAL STABILITY OF NAPHTHALENE
 TEMPERATURE = 920°F - 24-HOUR EXPOSURE -
 304 SS VESSELS - 300 PSIG INITIAL NITROGEN PRESSURE

Compound	Run No.	Gas Evolution*			% Distribution of Products**		
		cc/Gram		Moles Gas Per Mole	Coke	Volatile Solid	Liquid
		(Press.)	(M.S.)				
		(±10)					
Naphthalene	12	9	3	0.018	0.1	>99	- (a)
1-Methylnaphthalene	12	166	170	1.08	~61	~26	- (r)
2-Methylnaphthalene	12	94	209	1.32	~59	~26	- (r)
Acenaphthene	10	155	155	1.07	?	<87	- (r)
1-Fluoronaphthalene	12	11	3	0.017	?	<65	35 (r)
2-Fluoronaphthalene	26	13	6	0.041	7	93	- (a)

* Equilibrium values in every case

** Coke and volatile solid determined by:

- (a) Vacuum distillation
- (r) Manual separation

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TABLE 3.4

EFFECT OF STRUCTURE MODIFICATIONS ON THERMAL STABILITY
 TEMPERATURE = 920°F, 24-HOUR EXPOSURE***

Compound	Run No.	Gas Evolution*			% Distribution of Products		
		cc/Gram		Moles Gas Per Mole	Coke**	Volatile Solid	Liquid
		(Press.)	(M.S.)				
Stilbene	21	(+10) 88	82	0.66 (I)	~64	-	~31
Triphenylmethane	21	62	59	0.65 (I)	~70	-	~26
Triphenylbenzylmethane	26	90	83	1.20 (I)	~71	?	23
Triphenylmethylsilane	19	57	50	0.61 (I)	~98	-	-
Triphenylbenzylsilane	26	59	55	0.82 (I)	~89	-	8
Triphenylphosphate	21	34	40	0.52 (I)	~74	-	~22
1,2,4-Trichlorobenzene ^a	19	123	117	0.93 ^c (I)	~81	-	-
1,2,4-Trichlorobenzene ^b	19	64	-	~0.52 ^c (I)	~90	-	-
Perfluoroheneicosane	26	6	16	~0.76 ^d (E)	-	-	~80
Cetane ^a	3	244	136	1.37 (I)	~86	-	-
Decalin ^a	3	252	233	1.43 (I)	~45	-	33

* E = Equilibrium value
 I = Still increasing

** Coke and volatile solid determined by manual recovery in every case.

*** 304 SS Vessels - 300 psig initial nitrogen pressure

^a Seven-hour exposures at 915°F

^b Vycor liner

^c HCl only in gas phase

^d Light fluorocarbons only in gas phase

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TABLE 3.5

EFFECT OF ADDITIVES ON THE THERMAL STABILITY
OF NAPHTHALENE AND BIPHENYL
TEMPERATURE = 920°F EXPOSURE = 72 HOURS
304 SS VESSELS - 300 PSIG INITIAL NITROGEN PRESSURE

Additive		Gas Evolution					% Distribution of Products†			Series No.	
Wt. %	Compound	cc/Gram		Moles Gas Per Mole	Inhibition Period, Hours	Hydrocarbon-to-Hydrogen Ratio	Coke	Volatile Solid	Liquid		
		(Press.)	(M.S.)								
		(±10)									
		NAPHTHALENE									
	None	109	121	0.69 (E)	34	3.3	63	25	4 (a)	16	
	None*	132	124	0.71 (E)	29	5.5	<79	>12	- (r)	16	
	None**	106	111	0.64 (E)	30	3.9	<72	>22	- (r)	23	
1.5	Nitric Oxide	101	103	0.58 (E)	26	5.3	~72	~21	- (r)	28	
2.4	Thianthrene	115	132	0.74 (E)	21	5.2	<73	>17	- (r)	23	
2.4	Acridine	116	127	0.72 (E)	~11	4.9	~74	~17	- (r)	28	
1.5	Naphthacene	101	105	0.60 (E)	~6	4.4	~76	~17	- (r)	28	
		BIPHENYL									
	None	23	21	0.13 (I)	20	0.47	41	20	38 (a)	16	
	None*	32	28	0.19 (I)	15	1.0	<64	~26	8 (r)	16	
1.5	Nitric Oxide	27	23	0.15 (I)	20	0.90	<81	?	17 (r)	28	
2.4	Thianthrene	22	21	0.14 (I)	21	0.66	<61	?	38 (r)	23	
2.4	Triphenylmethane	-	~113	~0.7 -	-	~0.9	~77	?	14 (r)	23	
2.4	Iodine	80	164	1.09 (E)	0	6.8	~69	?	17 (r)	23	
		NAPHTHALENE-BIPHENYL***									
	None	89	94	0.58 (E)	19	3.8	~70	?	23 (r)	28	

* Vycor Liner

** Specially purified naphthalene

*** 44-56 Wt. Per Cent, resp.

[†] Coke and volatile solid determined by:

(a) = Vacuum distillation

(r) = Manual recovery

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TABLE 3.11

RADIATION RESISTANT LUBRICANTS

SAE No.	Equivalent Viscosity Range in SSU at 130°F for 50 V.I. Oil	CAL RESEARCH EQUIVALENT LUBRICANT					
		No.	Viscosity in SSU at 130°F		Viscosity in SSU at		Viscosity Index
			Range	Actual	100°F	210°F	
5W	55-65	← RRL-1	55-75	55	78.0	37.5	112
10W	67-76						
*10	90-120	← RRL-2	125-175	148	291	56.2	124
20W	100-120						
20	110-220	← RRL-3	255-315	298	767	71.8	79
30	220-340						
40	340-485	← RRL-4	500-600	529	1394	111	101
50	485-760						
*60	720-950	← RRL-5	1000-1100	1065	3257	175	95
*70	950-1250						
80	200-300	← RRL-6	2000-2200	2180	6356	358	112
90	300-530						
140	900-1950	← RRL-6	2000-2200	2180	6356	358	112
250	>1950						

* This designation discontinued.

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TABLE 4.0

GREASE SAMPLES TO ARGONNE NATIONAL LABORATORY

Cal Research Designation	1961-43-2	1961-43-3	1961-43-4	1961-48-3	1961-48-4
Oil Viscosity at 210°F, SSU	72	210	527	127	150
ASTM Worked Penetration	275	273	256	286	290
Oil Type and Per Cent by Weight					
Alkylaromatic	71.8	-	-	17.4	-
Petroleum Bright Stock	-	71.8	-	-	43.4
Petroleum Bright Stock	-	-	71.8	-	-
Poly(alpha-methylstyrene)	-	-	-	54.3	39.2
Styrene-Alkene Copolymer	-	-	-	10.9	-
Additives, Per Cent by Weight					
Du Pont Fumed Silica	13	13	13	13	13
Graphite	10	10	10	-	-
Didodecyl Selenide	5	5	5	4.35	4.35
Quinizarin	0.2	0.2	0.2	0.05	0.05

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TABLE 5.0

COMPARISON OF ELECTRON AND REACTOR RADIATION DAMAGE TO N-BUTYLBENZENE

Facility	Dosage ⁽¹⁾	Viscosity, Centistokes		Index of ⁽²⁾ Damage, η_1	Per Cent Purity	
		100°F	210°F		I.R. ⁽³⁾	M.S.
-	0	0.95	0.54	(1.00)	(100)	(100)
MTR, VH-3	N = 0.37	1.07	0.58	1.13	90	-
MTR, VH-3	N = 1.11	1.31	0.68	1.38	79	-
MTR, VH-3	N = 2.06	1.69	0.81	1.82	-	-
MTR, VH-3	N = 4.12	3.61	1.44	4.00	60	-
NAA, Statitron	5.56 μ ah	0.99	0.55	1.04	96	~100
NAA, Statitron	13.5 μ ah	1.02	0.56	1.08	93	~100
NAA, Statitron	100.0 μ ah	2.16	0.97	2.35	70	70

(1) N = Combined Radiation Dosage x 10^{-18} ,
thermal neutrons/cm²
 μ ah = Microampere-hours at nominal voltage of 1 mev

(2) T₁ = 92°F

(3) Based on average of values obtained from
peaks at about 9.4 and 10.7 microns.

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TABLE 5.1

SAMPLES OF ORGANICS RECEIVED FROM
NATIONAL BUREAU OF STANDARDS

Identity	Melting Point, °C	Disposition
<u>Group I - Received March 1953</u>		
Tetraphenylsilane	235	Exposed in Series 24 at 426°C - See Table 2.3 Under consideration for future test work.
p-Tolyltriphenylsilane	138	
Tetra-p-tolylsilane	228	
Tri-p-tolylphenylsilane	-	
Tetrabenzylsilane	128	Exposed in Series 24 at 426°C - See Table 2.3 - Merits further work.
Dibenzothiophene	98	Exposed in Series 25 at 170°C - See Table 2.3 - Merits further work.
<u>Group II - Received Sept. 1953</u>		
Tetrabenzylsilane	128	Being irradiated in Series 30, 31, and 32
Dibenzylidiphenylsilane	60	Being irradiated in Series 30, 31, and 32
Polybenzyl	~85	Being irradiated in Series 31
Fluoranthrene	-	Under consideration for further test work.
Perylene	275	
Triphenylene	-	
2-Methyl-3-hexyne	40	

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TABLE 6.0

SAMPLES FURNISHED

Quantity	Identity	Date	To	Attention
20 Gal.	Mixed Alkylbenzenes (MW ~350)	12-26-52	GE	F. Lutter
5 Gal.	Special Oil - "SAE 30" (1961-34-2)	1-2-53	GE-Schenectady	H. E. Wicks
5 Gal.	Special Oil - "SAE 20" (1961-34-1)	1-2-53	↓	↓
10 Lb.	Special Grease (1961-34-3)	1-2-53		
25 Gal.	Special Hydraulic Oil (1961-37-1)	1-23-53		
5 Gal.	Special Oil - "SAE 40" (1961-36-2)	1-27-53		
300 Ml.	n-Butylbenzene (1961-38-1)	2-12-53	North Amer. Av.	H. W. Pearlman
25 Ml.	3-Methylbiphenyl (1961-38-2)	2-12-53	North Amer. Av.	H. W. Pearlman
10 Lb.	Special Grease (1961-40-1)	2-27-53	GE-Schenectady	H. E. Wicks
10 Lb.	Special Grease (1961-34-3)	2-27-53	GE-Schenectady	H. E. Wicks
1 Gal.	Aromatic Oil (1961-42-1)	4-13-53	Brookhaven	R. W. Powell
1 Gal.	Special Oil (1961-42-2)	4-13-53	Brookhaven	R. W. Powell
3 "Gal."	Special Grease (1961-44-1)	6-5-53	GE-Schenectady	H. E. Wicks
2 Oz.	Special Greases (3) (1961-43-2,3,4)	6-11-53	Argonne	W. K. Anderson
1 Qt.	Special Oils (4) (1961-47-1,2,3,4)	7-23-53	KAPL	J. Barnard
2 Oz.	Special Greases (2) (1961-48-3,4)	8-6-53	Argonne	W. K. Anderson
10 Gal.	Special Oil, RRL-3 (1961-49-1)	8-17-53	GE-Schenectady	H. E. Wicks
20 Gal.	Mixed Alkylbenzenes (MW ~250) (1961-49-2)	8-20-53	GE-Oak Ridge	L. E. Stanford
10 Gal.	Special Oil, RRL-4 (1961-49-3)	9-8-53	GE-Schenectady	H. E. Wicks

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FIGURE 2.0

SAMPLE CONTAINERS USED IN IRRADIATION

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CAPSULE - VOL., ML.

C-31

C-12.2 C-17

C-17G

C-22

C-12

C-18.5

PR 538675

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FIGURE 2.1

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FIGURE 2.2
FIRST GAMMA CALORIMETER USED IN E-25 OF BNL REACTOR

PR 538676

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FIGURE 2.3
Co-Al MONITORS IN THE BROOKHAVEN REACTOR
HOLE 25
SERIES 10, 11, 12, 13, 15, 16

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FIGURE 2.4
Co-Al MONITORS IN THE BROOKHAVEN REACTOR
HOLE 25
SERIES 20, 21, 22, 23, 24, 25, 26

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352 055

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FIGURE 2.5
DEVIATION OF THERMAL NEUTRON DOSAGE FROM CALCULATIONS
BROOKHAVEN REACTOR
SERIES 20, 21, 22, 23

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FIGURE 2.6
THE THICKENING OF n-BUTYLBENZENE
IN TERMS OF RADIATION COMPONENTS

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FIGURE 2.7
VISCOSITY CHANGE WITH GAMMA DOSAGE
UNDER ONE ATMOSPHERE OF HELIUM

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FIGURE 2.8
STABILITY OF POLYMER THICKENERS TO GAMMA PHOTONS *

FLUX $\sim 1.69 \times 10^5$ R/HR - 25°C.

BASE OIL: MIXED ALKYL BENZENE (MW ~ 250) WITH 5% DIDODECYL SELENIDE AND 0.05% QUINIZARIN

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FIGURE 3.0
GAS EVOLUTION AT 493°C
POLYPHENYL VS. CONDENSED RING AROMATICS

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FIGURE 3.1
GAS EVOLUTION AT 493°C.
EFFECT OF ADDITIVES

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FIGURE 3.2
EFFECTS OF DOSAGE AND TEMPERATURE
ON DAMAGE TO ORGANICS

MAGNESIUM CONTAINERS, HELIUM ATMOSPHERE
IRRADIATION TEMP., °C., AS SHOWN
BNL REACTOR

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FIGURE 3.3
EFFECTS OF DOSAGE AND TEMPERATURE
ON DAMAGE TO ORGANICS

MAGNESIUM CONTAINERS, HELIUM ATMOSPHERE
IRRADIATION TEMP., °C., AS SHOWN
BNL REACTOR

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FIGURE 3.4
EFFECTS OF DOSAGE AND TEMPERATURE
ON DAMAGE TO ORGANICS

MAGNESIUM CONTAINERS, HELIUM ATMOSPHERE
IRRADIATION TEMP., °C., AS SHOWN
BNL REACTOR

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FIGURE 3.5
EFFECTS OF DOSAGE AND TEMPERATURE
ON DAMAGE TO ORGANICS

MAGNESIUM CONTAINERS, HELIUM ATMOSPHERE
IRRADIATION TEMP., °C., AS SHOWN
BNL REACTOR

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FIGURE 3.6
EFFECTS OF DOSAGE AND TEMPERATURE
ON DAMAGE TO ORGANICS

MAGNESIUM CONTAINERS, HELIUM ATMOSPHERE
IRRADIATION TEMP., °C., AS SHOWN
BNL REACTOR

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FIGURE 5.0
PURITY AND VISCOSITY CHANGES IN
REACTOR IRRADIATIONS OF n-BUTYLBENZENE

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