APEX
411

IRRADIATION TESTING OF ORGANIC LIQUIDS

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GENERAL ELECTRIC
ATOMIC PRODUCTS DIVISION
AIRCRAFT NUCLEAR PROPULSION DEPARTMENT

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Applied Materials Research Sub-Section

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Abstract

This report summarizes the results of irradiation tests of organic materials conducted in the HB-2 facility of the LITR and in the electron beam of the GE-ANPD 2-Mev Van de Graaff accelerator. Several organic liquids capable of withstanding temperatures up to 650°F have been tested, and descriptions of the tests and typical data are presented.

Introduction

A series of irradiation tests was conducted on organic liquids in the HB-2 experimental facility of the Low Intensity Test Reactor (LITR) and in the beam of the General Electric Aircraft Nuclear Propulsion Department 2-Mev Van de Graaff accelerator. The HB-2 facility in the LITR is a horizontal hole that runs east and west and extends inward from the east side of the reactor to the shell containing the reactor core. Figure 1 is a plan view of the reactor showing the location of the hole with respect to the core and reflector.

Organic liquids of promising radiation stability, as determined by preliminary evaluations conducted by GE-ANPD and outside contractors, were subjected to engineering tests in reactor radiation fields at temperatures up to $650^{\circ}F$. Liquids that have been tested to date include alkylbenzene 350, alkylbenzene 250, Dowtherm-A, Pentalene 195, and number 290.*

This report describes the test facilities and test operation and presents typical test data and their analyses.

The appendix presents a more detailed treatment of the material presented in the main portion of the report.

*Alkylbenzene 350 and alkylbenzene 250 are commercial liquids obtained from the California Research Corporation. They are mixtures of mono- and di-alkylated benzenes. The numbers 350 and 250 designate the average molecular weights of each liquid.

Dowtherm-A is a commercial heat transfer liquid manufactured by the Dow Chemical Company containing a mixture of diphenyl oxide and diphenyl.

The Pentalenes are mixtures of mono-, di-, and poly-amylnaphthalenes. Pentalene 95 is an undistilled mixture, and the Pentalene 195 is a distilled mixture of the amylnaphthalenes obtained from Pentalene 95; both mixtures are commercially available. The numbers 95 and 195 are designations by the Sharples Chemical Company and do not refer to molecular weights of the respective mixtures. The residue of Pentalene 195 after a $625^{\circ}F$ distillation has been designated number 290 by GE-ANPD. The number 290 approximates the molecular weight.

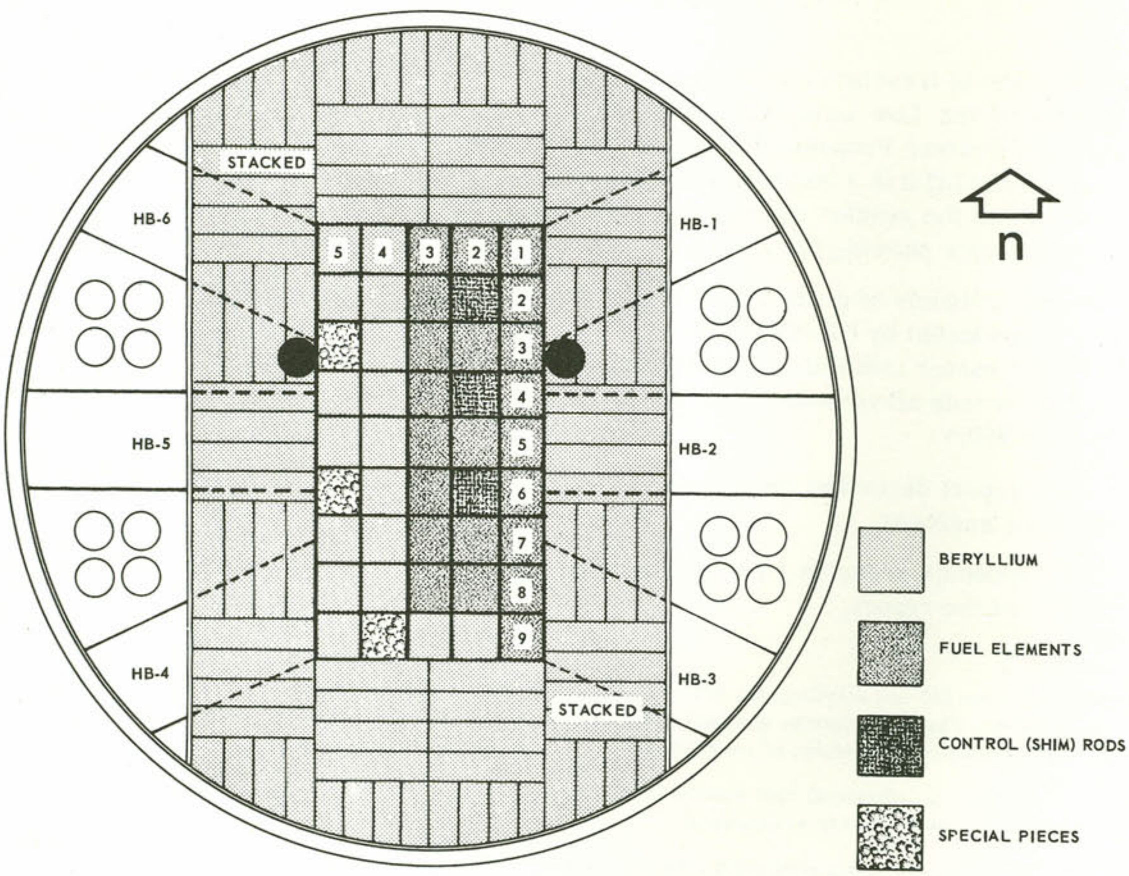

Fig. 1 – Lattice-reflector arrangement of the LITR

Irradiation Facilities and Test Systems

Reactor Irradiation Test System

The experimental loop and accessory equipment used for reactor irradiation testing of the selected organic liquids were designed to be used with the HB-2 facility of the LITR. The system included a canned rotor pump; heat exchangers to heat the test liquid, to cool the liquid for viscosity measurements, and to serve as pump bearing coolant; a can for containing the liquids within the reactor; a filter; a remote-indicating flowmeter; a remote-indicating viscometer; a sump or reservoir; and a wet-test gas meter to measure the off-gas. Figure 2 shows typical equipment used at the LITR. The equipment is inclosed in a gas-tight container filled with carbon dioxide because of the flammable nature of the liquid tested. Figure 3 shows the control panel used with the test equipment. A detailed description of the test system components together with some operating description is given in the appendix.

Van de Graaff Test System

Irradiation of organic liquids in the Van de Graaff accelerator has been found useful in establishing the relative stability of the liquids as measured by gas evolution and viscosity change.* A sketch of a test system devised for use with the GE-ANPD Van de Graaff accelerator is shown in Figure 4. A detailed description of the test system is given in the appendix.

*This work is discussed further in two reports issued by the General Electric Aircraft Nuclear Propulsion Department: V. P. Calkins, "Radiation Damage to Non-Metallic Materials," APEX-167, August 1954; C. G. Collins and V. P. Calkins, "Radiation Damage to Elastomers, Organic Liquids, and Plastics," APEX-261, September 1956.

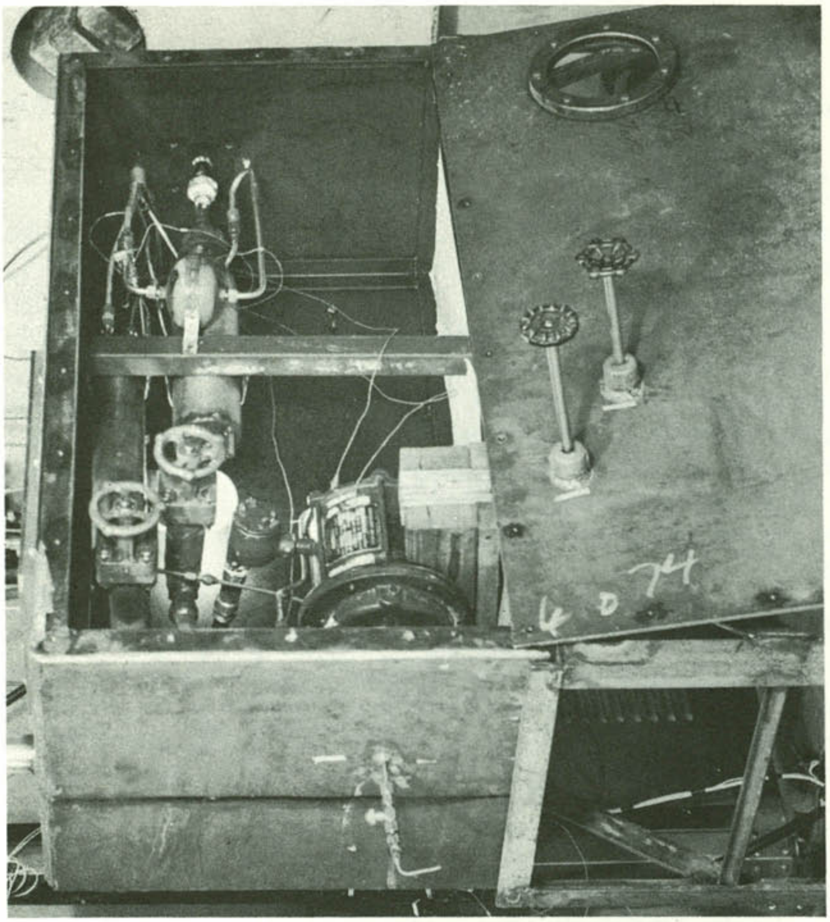

Fig. 2 — A typical hydrocarbon recirculating loop used at the Low Intensity Test Reactor (with side of housing removed)

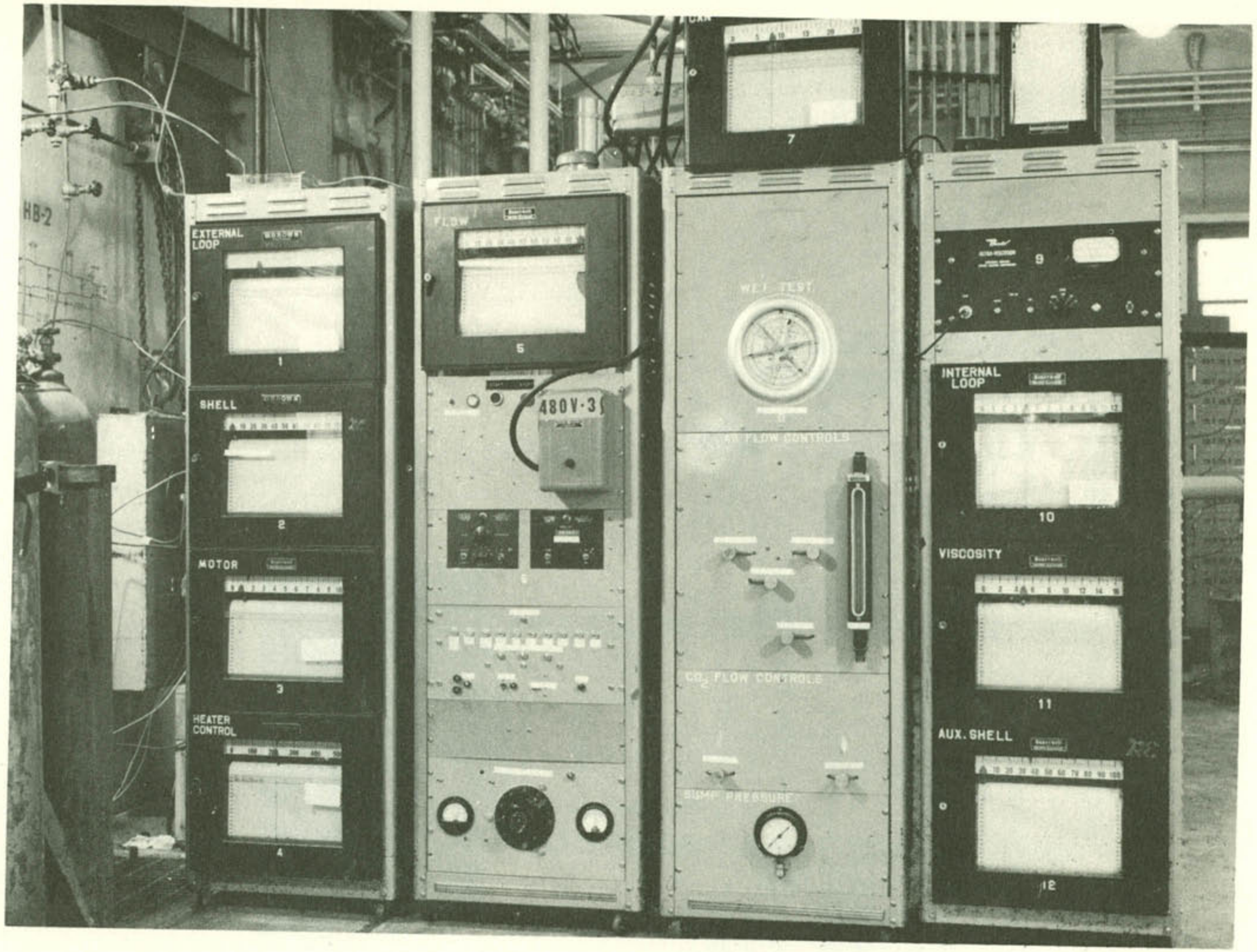

Fig. 3 – Control panel of hydrocarbon recirculating unit at the LITR

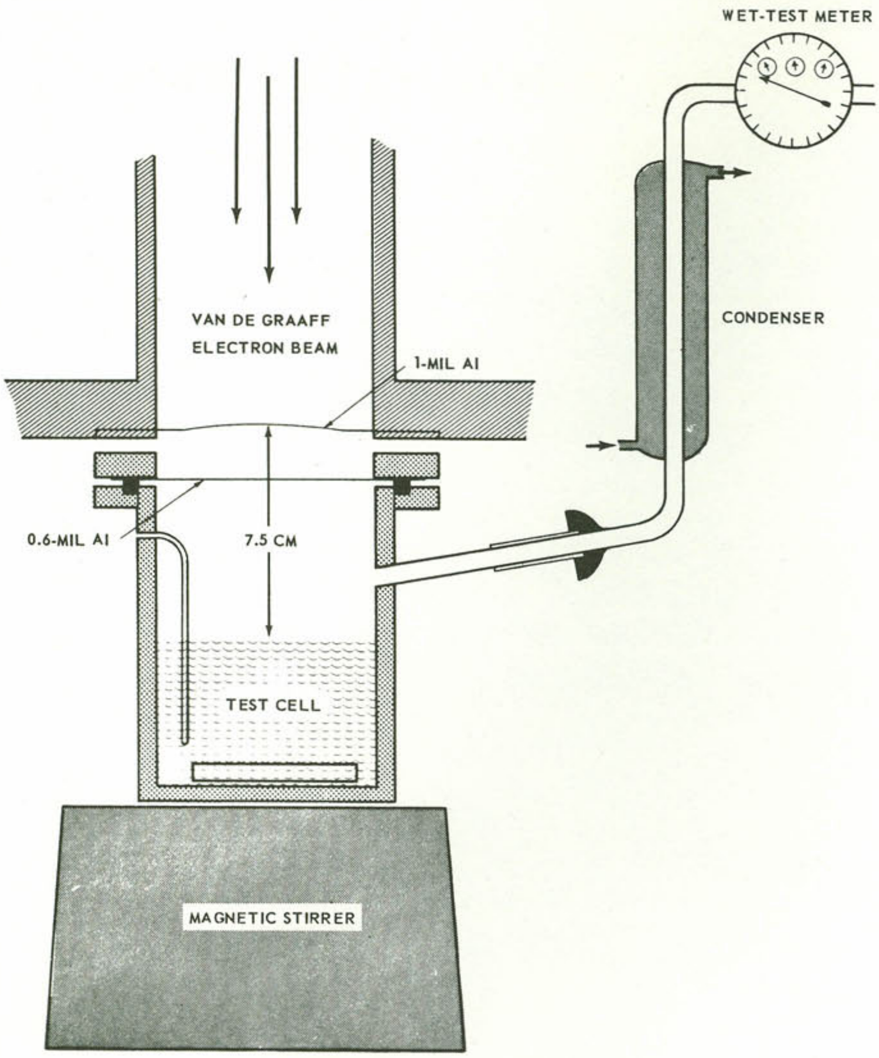

Fig. 4 – A typical test cell for electron irradiation of liquids

Interpretation of Test Data and General Conclusion

A comparison of the data from the 300°F reactor irradiation tests indicated that alkylbenzene 250 was more radiation stable than alkylbenzene 350 on the basis of viscosity change and gas evolution rate as a function of dosage, but was not as stable as Dowtherm-A. These data are presented in Table 1; plots of gas evolution and viscosity versus dosage are shown in Figures 6, 7, and 8. When alkylbenzene 250 was subjected to reactor irradiation tests at 500°F, it was found that the physical property changes with radiation dosage were not markedly different from those measured in the 300°F tests (see Table 3).

In a search for liquids usable at 600°F, Van de Graaff data indicated that Pentalene 95, Pentalene 195, and number 290 showed stability at these temperatures. Of the three liquids tested to date, number 290 appears most promising. A study of hydrogen content and over-all radiation stability is given in Tables 8 and 9 and in Figures 23, 24, 25, and 26.

Operation of Test Systems

Nine reactor irradiation engineering tests on organic liquids were conducted in the HB-2 facility of the LITR. In the initial tests alkybenzene 350, alkylbenzene 250, and Dowtherm-A were tested for radiation stability at an operating temperature of 300°F. Alkylbenzene 250 was subjected to additional testing at temperatures up to 500°F. Gas evolution was measured as a function of irradiation temperature and dosage. A run was also conducted at 450° - 500°F to determine the heat transfer characteristics of alkylbenzene 250 as a function of irradiation dosage.

Approximately 85 irradiations were performed in the Van de Graaff accelerator using a large variety of organic liquids. Twenty-five screening tests were performed using various types of liquids to determine general radiation stability.* Approximately 60 organic liquids, shown by the screening tests to be the most promising radiation-stable types, were subjected to high temperature - high dosage tests.

A detailed description of the irradiation test rigs and a summary of the data obtained from the reactor irradiation and Van de Graaff tests are given in the Appendix.

*Other screening tests were described in a report by V. P. Calkins, "Radiation Damage to Non-Metallic Materials," General Electric Aircraft Nuclear Propulsion Department, APEX-172, August 1954, p. 39.

Appendix
Details of Test Systems, Operation, and Results

Reactor Irradiation Tests

A schematic diagram of a typical hydrocarbon-recirculation loop for reactor irradiation testing is shown in Figure 5. A modified Chemco canned rotor pump was used to avoid the problem of mechanical seals and stuffing boxes. On elevated-temperature tests a small portion of the pump discharge is fed through a cooling coil in which the liquid temperature is reduced to 100° - 150°F and then through the Viscoson, a remote-indicating viscosity sensor, which continuously measures the viscosity and transmits the values to a recorder on the control panel. The cooled liquid flows over the pump bearings and back into the pump inlet line. The main discharge from the pump flows through a heater to the reactor. The heater consists of a noninductively-wound double coil of stainless steel tubing, both ends of which are electrically grounded. By means of a center electrical tap, an electric current is passed through the tubing in two parallel circuits. The heat generated in the wall of the tubing is then transferred with high efficiency to the test liquid, which flows through the tubing.

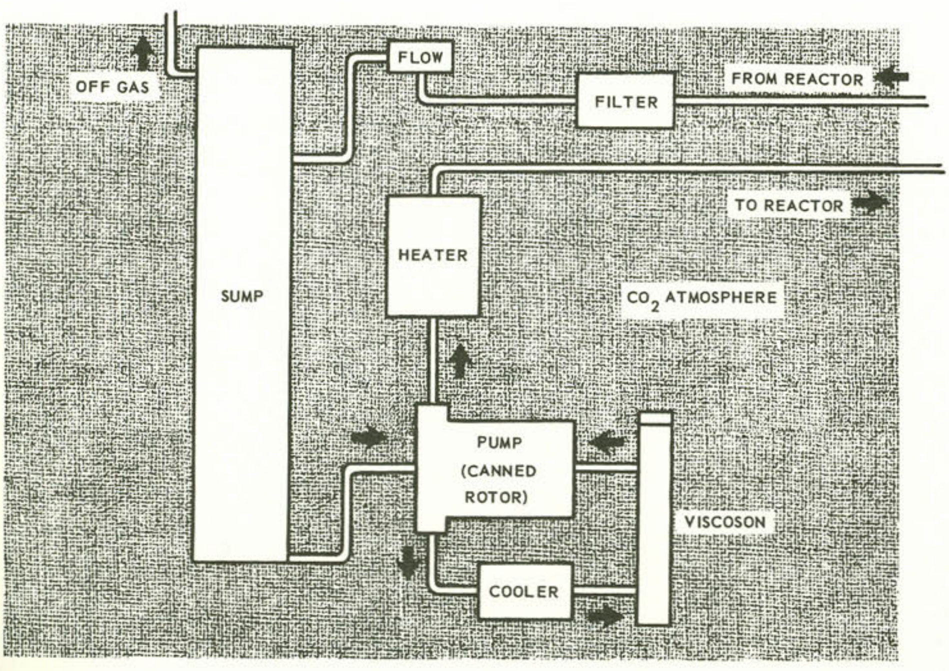

Fig. 5 – Hydrocarbon recirculation loop for reactor irradiation tests

The liquid discharged from the heater flows to a container that is located in the HB-2 test hole of the LITR approximately 13 feet in from the outer shield face of the reactor. This container is held in place by a sealed, water-cooled aluminum probe from which it is thermally insulated by Refrasil insulation. Small-diameter tubing carries the test liquid in and out of the reactor to and from the external pumping system. The pressurized housing containing this external loop is located close against the face of the reactor, as shown in Figure 2. A CO_2 atmosphere is maintained within the box and throughout the reactor probe to minimize fire hazards in the event of a test-loop leak.

The irradiated test liquid coming out of the reactor first passes through a filter to remove any solids that might foul the test loop or flow meter. The filtered liquid then passes through the flow-measuring device and is discharged into the sump, where gaseous decomposition products formed by the reactor irradiation are released by the liquid. The slight pressure (2 inches of water) developed by this action causes the gas to pass through a wet-test meter which is connected to the top of the sump. After being metered, the discharged gas either is collected by water displacement in sample bottles for subsequent analysis or is carried off through the reactor vacuum exhaust system.

The photograph shown in Figure 3 was taken while the alkylbenzene 250 was being tested at 500°F and shows the instrumentation required for the test facility. This equipment provides metering, totalizing, recording, and controlling facilities for the test apparatus. In addition, most of these instruments control units of safety circuits interconnected with the LITR alarm system and the reactor fast-setback or scram controls.

The nine engineering reactor irradiation tests conducted in the HB-2 facility of the LITR using variations of the experimental loop are described in the following sections.

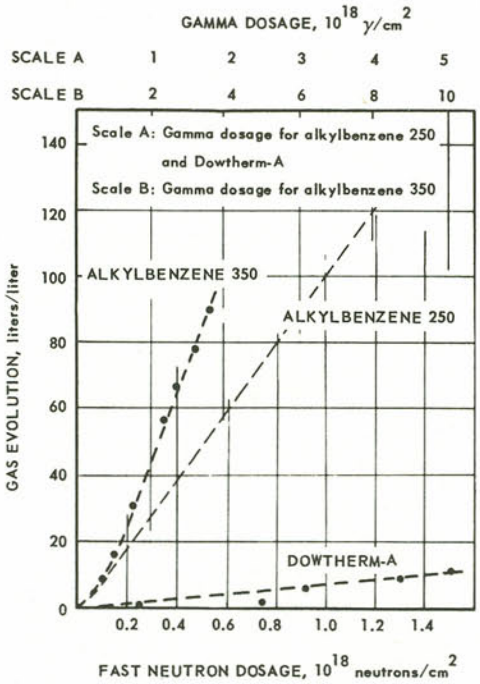

Fig. 6 — Gas evolution from alkylbenzene 350, alkylbenzene 250, and Dowtherm-A as functions of combined reactor radiation (fast neutron plus gamma dosage) at 300°F

Screening Tests at 300°F - In these tests a 700-milliliter aluminum container was used in the reactor irradiation test loop, and alkylbenzene 350, alkylbenzene 250, and Dowtherm-A were irradiated in succession. During the course of the tests the LITR power level was increased from 1900 kilowatts to 3000 kilowatts, and a variety of flux levels was used to give the dosages and data shown in the 300°F irradiation test data summary, Table 1. Figures 6, 7, and 8 compare gas evolution rates and viscosity changes for the three liquids as functions of fast neutron dosage.

Radiation Stability of Alkylbenzene 250 at 450° - 500°F - For this experiment the 700-milliliter stainless steel container was located to support the test liquid in a position 3 inches through 11 inches from the end of the HB-2 hole. A 1/4-inch-thick aluminum plate, which seals the end of this test hole, is positioned against the reactor fuel elements. The average fast-neutron-flux component (0.5 Mev and above) of the reactor radiation for this position with the reactor at the 3000-kilowatt operating level is approximately 6.48×10^{12} n/cm^2-sec, and the thermal-neutron-flux component is approximately 1.7×10^{13} n/cm^2-sec. A flux diagram showing the complete spectrum of the radiation field versus position measured from the 1/4-inch inner plate of the HB-2 facility is shown in Figure 9. The values given are for a reactor power level of 1500 kilowatts. With a flow of 800 cubic centimeters of test liquid per minute through the reactor, a heater input of approximately 2 kilowatts maintained the irradiated liquid at 480° - 500°F. The temperatures of other parts of the test loop under these conditions are shown in Table 2.

The test liquid in the reactor can is heated an additional 35°F during irradiation as a result of approximately 500 watts of nuclear heating.

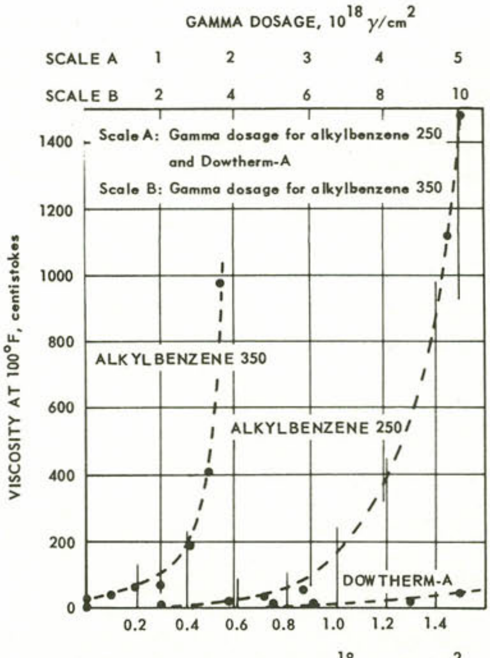

Fig. 7 – Viscosity (at 100°F) of alkylbenzene 350, alkylbenzene 250, and Dowtherm-A as functions of combined reactor radiation (fast neutron plus gamma dosage) at 300°F

TABLE 1

EXPERIMENTAL PHYSICAL PROPERTY CHANGES OF ORGANIC LIQUIDS WITH IRRADIATION

Test	Alkylbenzene (Mol. Wt. 350) Control	Alkylbenzene (Mol. Wt. 350) Irradiated[a]	Alkylbenzene (Mol. Wt. 250) Control	Alkylbenzene (Mol. Wt. 250) Irradiated[b]	DOWTHERM-A Control	DOWTHERM-A Irradiated[b]
Flash point, °F	340	245	280	255	240	190
Fire point, °F	355	290	295	ND[c]	260	225
Density, g/cc						
at: 77°F	0.871	0.914	0.865	0.934	1.058	1.101
100	0.863	0.905	0.860	0.925	1.050	1.092
200	0.827	0.868	0.824	0.888	1.003	1.049
300	0.790	0.835	0.785	0.858	0.954	1.000
400	(0.761)[d]	0.807	0.748	0.814	0.905	0.945
450	(0.748)	(0.795)	(0.737)	(0.808)	0.878	(0.918)
500	(0.738)	(0.786)	(0.726)	(0.797)	0.851	(0.888)
Viscosity,[e] centistokes						
at: 100°F	25.1	974.0	6.56	1658	2.51	45.1
300	1.7	9.3	1.04	13.8	0.63	2.51
450	(0.4)	(< 5)	(< 1)	(< 10)	(0.3)	(< 1)
500	(0.4)	(< 5)	(< 1)	(< 10)	(0.3)	(< 1)
Maximum testing temperature, °F	500	300	500	300	500	300
Hydrogen content hydrogen atoms/cc						
at: 77°F	6.76×10^{22}	6.33×10^{22}	6.30×10^{22}	6.38×10^{22}	3.72×10^{22}	3.78×10^{22}
100	6.7×10^{22}	6.3×10^{22}	6.3×10^{22}	6.3×10^{22}	3.7×10^{22}	3.7×10^{22}
200	6.4×10^{22}	6.0×10^{22}	6.0×10^{22}	6.1×10^{22}	3.5×10^{22}	3.6×10^{22}
300	6.1×10^{22}	5.8×10^{22}	5.7×10^{22}	5.9×10^{22}	3.4×10^{22}	3.4×10^{22}
400	(5.9×10^{22})	5.6×10^{22}	5.4×10^{22}	5.6×10^{22}	3.2×10^{22}	3.2×10^{22}
450	(5.8×10^{22})	(5.5×10^{22})	$(5.4 \times 10^{22}$	(5.5×10^{22})	3.1×10^{22}	$(3.1 \times 10^{22}$
500	(5.7×10^{22})	(5.4×10^{22})	(5.3×10^{22})	(5.4×10^{22})	3.0×10^{22}	(3.0×10^{22})
Specific heat, cal/g-°C						
at: 100°F		(0.47)[f]		(0.47)[f]	0.40[f]	(0.40)[f]
300		(0.57)		(0.57)	0.50	(0.50)
450		(0.65)		(0.65)	0.60	(0.60)
500		(0.67)		(0.67)	0.63	(0.63)
Gas evolution[g]	---	156	---	98	---	7
Maximum radiation testing dosage, fast neutrons/cm^2	---	0.54×10^{18}	---	1.5×10^{18}	---	1.5×10^{18}
Vapor pressure at 450°F, atm	< 1	< 1	< 1	< 1	< 1	< 1
Precipitation or sludging	ND	ND	ND	ND	ND	ND
Film formation	Not tested	Not tested	Not tested	Not tested	Not tested	Not tested
Container materials	Stainless steel or aluminum	SS or Al	SS or Al	SS or Al	SS or Al	SS or Al

[a] Actual Dosage - 0.54×10^{18} fast neutrons/cm^2 plus 3.7×10^{18} gammas/cm^2

[b] Actual Dosage - 1.5×10^{18} fast neutrons/cm^2 plus 4.9×10^{18} gammas/cm^2

[c] ND - None detected

[d] Values given in parentheses are estimated

[e] Viscosity in centistokes x density x 6.72×10^{-4} = Viscosity in lb/(ft-sec)

[f] Based on information furnished by suppliers. Irradiation should not change these properties significantly.

[g] Liters per liter per 10^{18} fast neutrons/cm^2

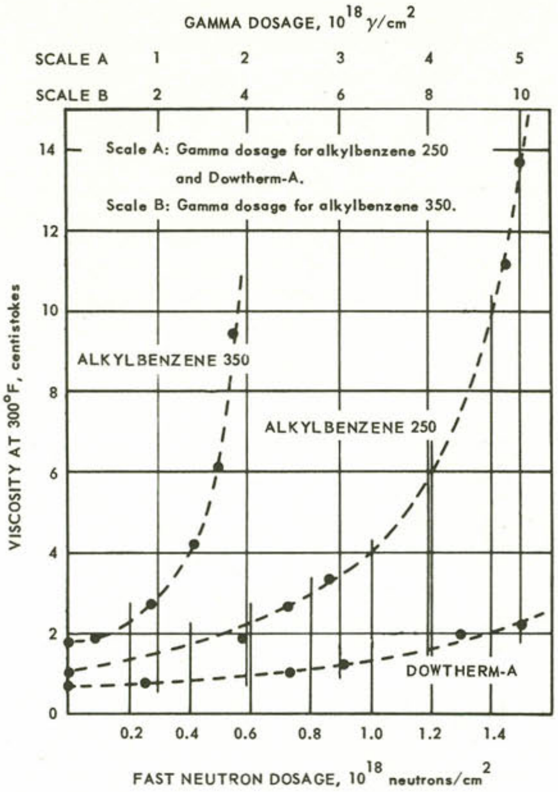

Fig. 8 – Viscosity (at 300°F) of alkylbenzene 350, alkylbenzene 250, and Dowtherm-A as functions of combined reactor radiation (fast neutron plus gamma dosage) at 300°F

During the early part of this test, the gas that evolved as a result of radiation damage to the test liquid in the 700-milliliter container was generated at a rate of 1.5 liters per hour. However, as the experiment continued, the evolution rate steadily decreased and fell to a value of 0.8 liter per hour by the end of the test. The relationship between gas evolution and radiation dosage of the test liquid is shown in Figure 10. Total gas evolution is plotted as a function of the fast neutron ($\geq$ 0.5 Mev) component of the mixed reactor radiation dosage received by the liquid. Similar data from previous experiments conducted at the 300°F irradiation temperature also are plotted in this figure to show the difference in the rates of gas evolution at the two temperatures. Except for the temper-

TABLE 2

TYPICAL TEMPERATURE PROFILE OF REACTOR IRRADIATION TEST LOOP FOR A 490°F LIQUID IRRADIATION TEMPERATURE

Thermocouple Location	Temperature, °F
Sump or heater inlet	290
Heater wall (inlet)	350
Heater wall (outlet)	515
Heater outlet	475
Can inlet	465
Can	490
Discharge to sump	415

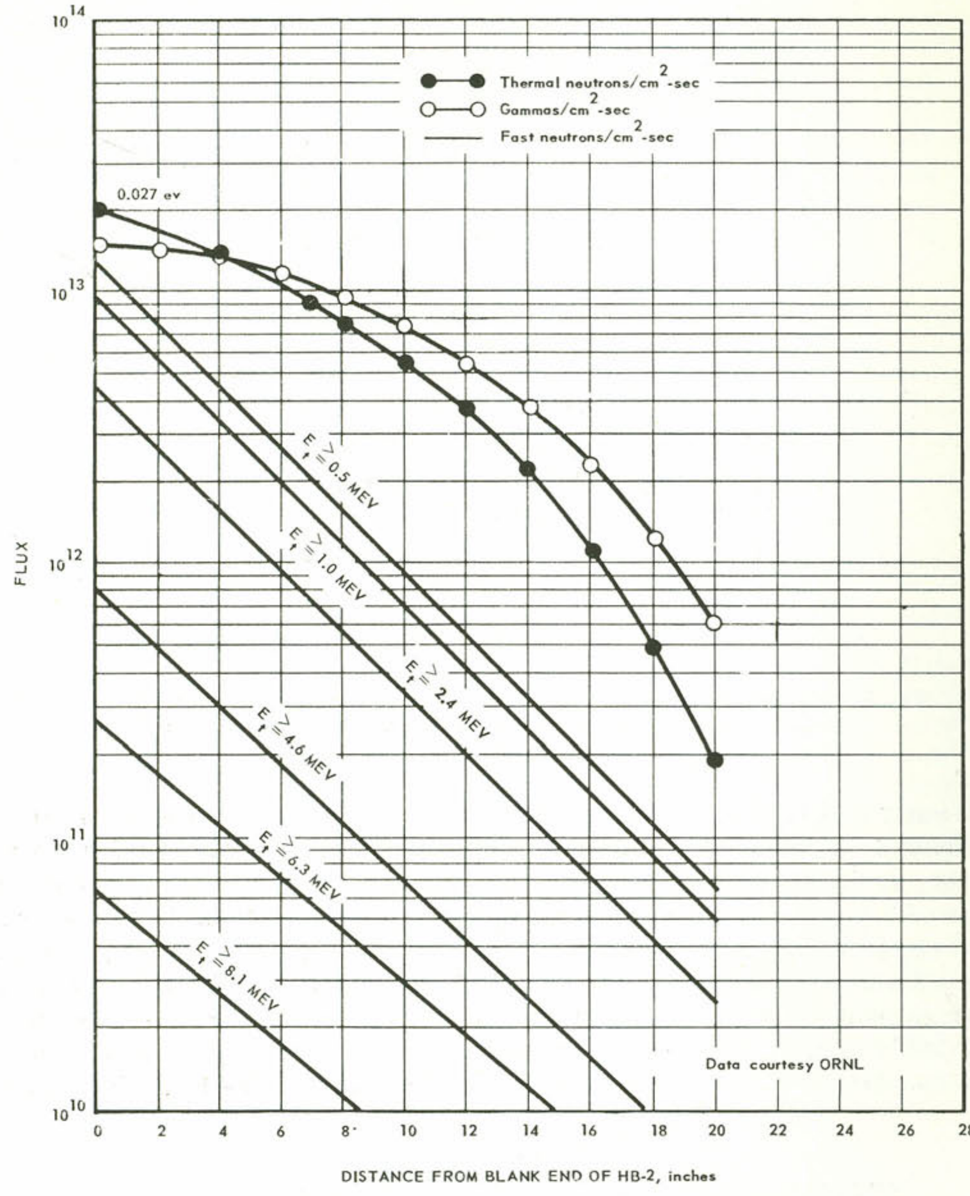

Fig. 9 – Radiation spectrum of the HB-2 hole

ature differences, test conditions were the same. This relationship between gas evolution rate from alkylbenzene 250 and irradiation temperature is further discussed in the following section. A summary of the physical properties of alkylbenzene 250 before and after irradiation is presented in Table 3.

The viscosity of the alkylbenzene 250, originally 7 centistokes at 100°F, increased with irradiation at first very slowly and then very rapidly during the last 10 hours of the test. The data plotted in Figure 11 show the relationship between the viscosity of the alkylbenzene 250 test liquid at various temperatures and the radiation dosage to which the liquid was exposed. Only the fast neutron (≥ 0.5 Mev) component of the reactor radiation is plotted. The viscosity measurements were obtained from specimens removed from the test system periodically during the course of the experiment. Independently of these measurements a continuous record of the viscosity changes produced in the test liquid was also obtained by means of a Viscoson instrument, the probe of which was located in the test-liquid stream after the bypass cooler. The continuous record obtained from this instrumentation showed that the viscosity of the test liquid increased only periodically with radiation and that near steady-state conditions lasted for considerable periods. These steady-state periods were especially noticeable during the last half of the test and are the cause of the break in the 500°F test curve shown in Figure 11. Comparison of the curve for previous 300° tests with the corresponding 500°F test data shows that viscosity changes are less severe at the higher irradiation temperature than at the 300°F temperature. Thermal degradation of large molecules at the higher temperature may account for the difference.

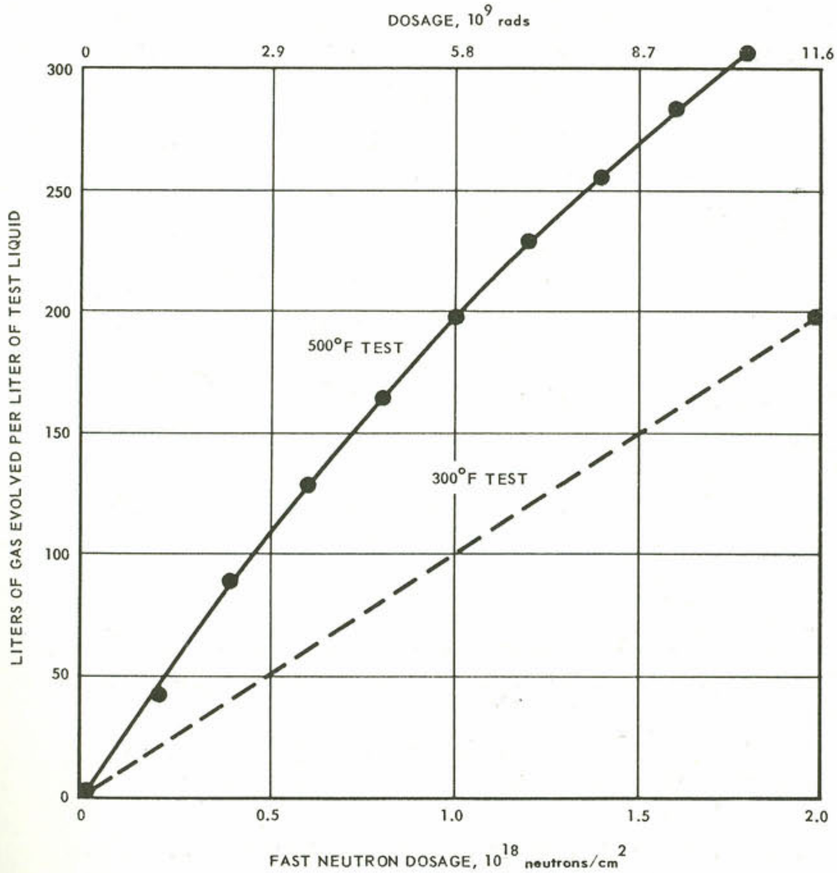

Fig. 10 – Total gas evolution for alkylbenzene 250 versus the fast neutron dosage component of the combined reactor radiation and total energy absorption

TABLE 3

PHYSICAL PROPERTIES OF ALKYLBENZENE 250 BEFORE AND AFTER IRRADIATION AT 300°F AND 500°F

Test	Control	Irradiated[a] at 300°F	Irradiated[a] at 500°F
Flash point, °F	280	255	205
Density, g/cc			
at 100°F	0.860	0.925	0.910
200°F	0.824	0.888	0.884
300°F	0.785	0.858	0.846
400°F	0.748	0.814	0.809
450°F	(0.730)[b]	(0.797)	(0.791)
500°F	(0.713)	(0.779)	(0.773)
Viscosity, centistokes			
at 100°F	7	1658	30
200°F	2	59	7
300°F	1	14	2
400°F	(< 1)	< 10	< 2
500°F	(< 1)	< 10	< 2
Average molecular weight	250	467	320
Hydrogen content, weight percent	12.1	11.4	11.8
Hydrogen content, (H atoms/cc) x 10^{-22}			
at 100°F	6.3	6.3	6.5
200°F	6.0	6.1	6.3
300°F	5.8	5.9	6.0
400°F	5.5	5.6	5.8
500°F	5.2	5.4	5.5
Vapor pressure, atmospheres at 500°F	< 1	< 1	< 1
Gas evolution, liters/liter-5.8×10^9 rads[c]	-	98	198

[a]Mixed reactor radiation of the HB-2 hole of LITR - energy absorbed 8.7×10^9 rads

[b]Values in parentheses are estimated. These values reflect more refined extrapolations of the density data than those presented in Table 1

[c]5.8×10^9 rads is equivalent to a mixed reactor radiation dosage of the HB-2 hole of the LITR having a $\geqq 0.5$-Mev fast neutron component of 1.0×10^{18} n/cm^2

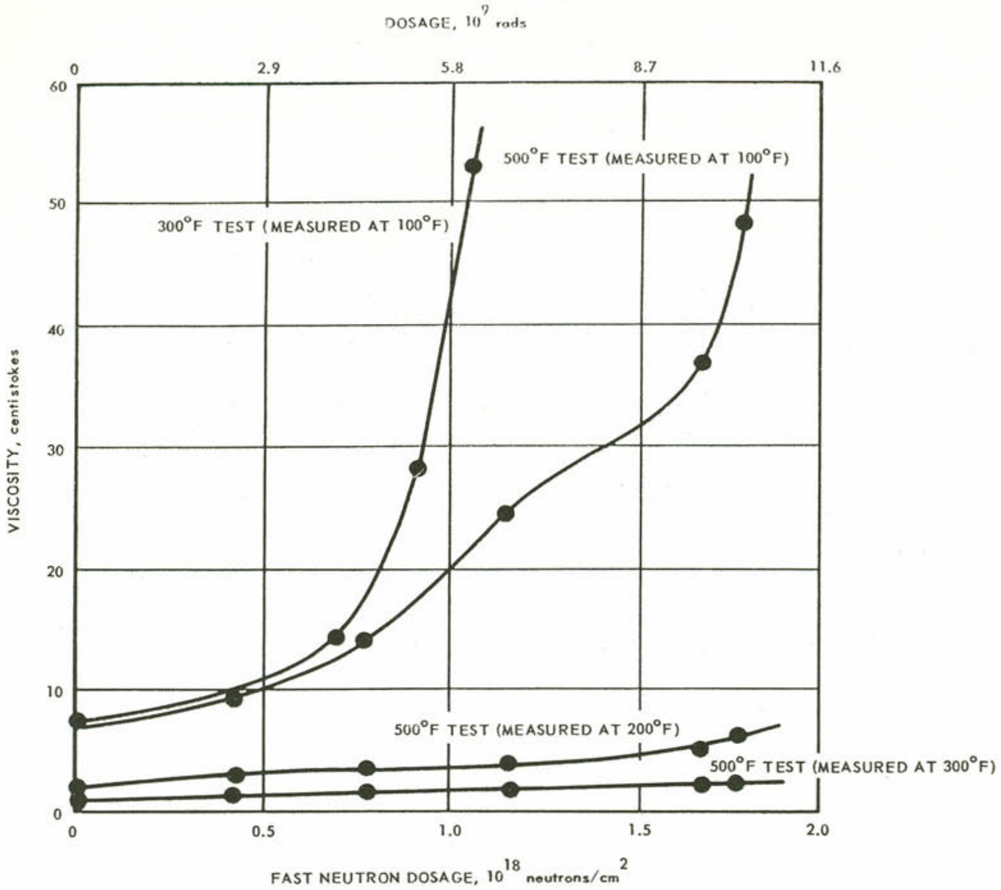

Fig. 11 – Viscosity of alkylbenzene 250 versus the fast neutron dosage component of the combined reactor radiation and total energy absorption

The specimens removed from the test loop during the experiment were subjected to laboratory analysis. The results of these studies are shown in Table 4 in comparison with values obtained from studies of unirradiated alkylbenzene and alkylbenzene irradiated at 300°F.

Effect of Irradiation Temperature on Gas Evolution from Alkylbenzene 250 - Two separate runs were made in this second experiment. In the first run alkylbenzene 250, which previously had been subjected to mixed reactor radiation so that the fast neutron ($\geqq$ 0.5 Mev) dosage component amounted to 0.5×10^{18} n/cm^2, was charged into the test loop and circulated at 300°F, 400°F, and 500°F. For the second run the same experimental conditions were used, but the test liquid was alkylbenzene 250, previously irradiated for 3 times the dosage of the liquid tested in the first run.

The data shown in Table 4 record the gas evolution rates per hour from the 700-milliliter container of alkylbenzene 250 under irradiation at the various temperatures. Very little time was required to obtain equilibrium conditions whenever test temperatures were changed. For this experiment the average fast neutron ($\geqq$ 0.5 Mev) flux component of the reactor radiation was 6.48×10^{18} n/cm^2-sec. The compositions of the gases obtained from the liquids at the various test temperatures are shown in Table 5. Analysis of the results showed excellent agreement between these data and those obtained from previous constant-temperature experiments. Moreover, radiation damage to alkylbenzene 250 as determined by hydrogen evolution appears to be generally independent of temperature. Higher total

TABLE 4

GAS EVOLUTION FROM 700 MILLILITERS OF ALKYLBENZENE 250 AT VARIOUS DOSAGE LEVELS AND TEMPERATURES - CONSTANT FLUX[a]

Temperature, ^oF	Gas Evolution Rate, Liters per hour Dosage[b] 0.5×10^{18} n/cm^2	Dosage[b] 1.5×10^{18} n/cm^2
300	1.04	0.90
400	1.27	1.03
500	1.47	1.18

[a]Average fast neutron flux component of the reactor radiation: 6.48×10^{12} n/cm^2-sec

[b]Fast neutron ($\geqq 0.5$ Mev) dosage (n/cm^2) component of the reactor radiation

TABLE 5

CHEMICAL ANALYSIS OF THE GAS GENERATED BY REACTOR IRRADIATION OF ALKYLBENZENE 250 AT VARIOUS TEMPERATURES

Values Reported as Volume Percent

Temperature, ^oF	Dosage[a] $\times 10^{-18}$	H_2	Combustibles[b]	Illuminants[c]	CO	CO_2	O_2	Inerts[d]
150	0.5	61.4	24.1	11.8	0.9	0.9	0.9	0.0
300	0.5	62.0	20.0	14.0	4.0	0.0	0.0	0.0
300	1.5	66.0	12.0	16.0	6.0	0.0	0.0	0.0
500	0.5	46.2	27.3	18.3	5.8	0.8	1.6	0.0
500	1.0	48.7	22.1	18.3	8.7	2.2	0.0	0.0
500	1.5	52.2	21.7	18.7	6.1	1.3	0.0	0.0

[a]Fast neutron ($\geqq 0.5$ Mev) dosage of test liquids at the time of gas sampling

[b]Combustible saturated hydrocarbons

[c]Illuminants (unsaturated hydrocarbons)

[d]The residue remaining (N_2 or He) after removing all reactive components

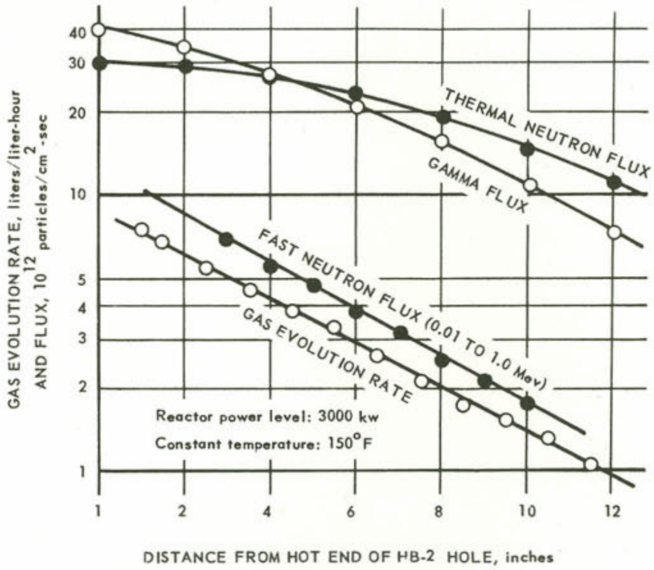

Fig. 12 – Comparison of gas evolution rate from alkylbenzene 250 at various positions within the HB-2 hole of the LITR with the flux pattern of the hole

gas evolution rates at the higher temperatures are attributed to the probability that more of the products of radiation damage are volatile at the high temperatures.

Effect of Flux on Radiation Damage to Alkylbenzene 250 - With minor exceptions the general test facility used for this test was as described for previous experiments. In place of the large container used for previous tests, a small 100-milliliter aluminum container 3-inch ID by 1 inch long, was used for this test. Probe connections were such that it was possible to move this can with the test liquid circulating through it while the reactor was active. Thus, the test liquid could be supported in any position in the range 1/2 inch to 15 inches from the end of the test hole. It was not necessary in this test to use the heater since the test liquid was circulated at ambient temperatures.

During the first run of this experiment, the LITR was held at the 3000-kilowatt operating level while the can containing the circulating alkylbenzene 250 test liquid was held for four hours at each of 15 different positions within the test hole, starting 1/2 inch from the core end of the hole. In the second run this test was repeated, but in the reverse order, starting 15 inches from the core end.

At each location gas evolution data were obtained. It was found that steady-state conditions could be obtained in less than 1 hour after each move. The measured gas evolution rates from the 100-milliliter container ranged from 0.76 liter per hour at an average distance of 1 inch from the core end of the test hole to 0.07 liter per hour at the 15-inch position. The relationships between gas evolution rates at these various positions and the flux levels of the various radiation components at these positions are shown in Figure 12. These data show that changes in gas evolution rates with position closely follow the fast neutron and gamma flux profile in this test hole.

In the third run of this experiment, the power level of the reactor was decreased from 3000 kilowatts to 1500 kilowatts and gas evolution measurements were made at various positions in the HB-2 hole as in previous runs. It was found that gas evolution rates at the lower power level were 50 percent of the rates obtained at the 3000-kilowatt level.

TABLE 6

PROPERTIES OF THE LIQUID CONDENSED FROM THE OFF-GAS LINE IN ALKYLBENZENE 250 REACTOR IRRADIATION TEST EXPERIMENT

Viscosity, centistokes	
at 100°F	1.46
200°F	0.76
300°F	0.51
Density, g/ml	
at 100°F	0.816
200°F	0.775
Flash point	
Small cup, °F	< 100
Average molecular weight	176

Effect of Irradiation on the Heat Transfer Coefficient of Alkylbenzene 250 - A reactor irradiation test was run in the LITR using alkylbenzene 250 to establish further the effects of radiation fields on film formation and heat transfer characteristics. The test equipment was similar to that used in previous tests except that additional instrumentation was used to give more accurate heat transfer data. The alkylbenzene 250 was exposed to a mixed radiation flux (per square centimeter per second) of 1.8×10^{13} thermal neutrons, 9.4×10^{12} fast neutrons, and 2.3×10^{13} gammaphotons; the maximum temperature of the liquid was maintained at 460° - 500°F. The experiment was operated a total of 136 hours with the LITR at the 3000-kilowatt power level. The experiment was terminated at will and not because of increased viscosity. An analysis of the effect of radiation dosage on the heat transfer characteristics is given at the end of this appendix.

As in previous experiments, liquid specimens and gas samples were taken during the course of the test. However, in this experiment a trap was inserted in the off-gas line to collect any volatile liquid that might distill. It was found that four milliliters of the liquid condensed out with every liter of gas evolved. Table 6 shows some of the properties of this low boiling decomposition liquid.

Van de Graaff Testing

The 2-Mev Van de Graaff electron accelerator was used to evaluate radiation damage to several types of organic liquids. Since the Van de Graaff is capable of imparting energy to materials at rates as high as 100 watts, the possibility of producing equivalent damage to liquids within a much shorter time than that needed for irradiation testing seemed useful, particularly with respect to testing materials at high temperatures and high dosages.

The test cell used in the Van de Graaff electron irradiation of organic liquids was fabricated of stainless steel having such dimensions that with a liquid volume of 30 cm^3, a depth of 2.2 cm was maintained. This depth was approximately twice that necessary to absorb 2-Mev electrons. The cell flange was sealed with a 0.0006-inch aluminum window and a Buna N rubber gasket. The 0.0006-inch aluminum window was selected because it resulted in negligible loss due to absorption and negligible scattering of the impinging electrons. A thermocouple well provided a means of measuring the liquid temperature during the test. The volatile gases formed during irradiation were exhausted through a water-cooled condenser and then metered through a wet-test meter. All samples were magnetically stirred to give equal distribution of damage throughout the material. A sectional

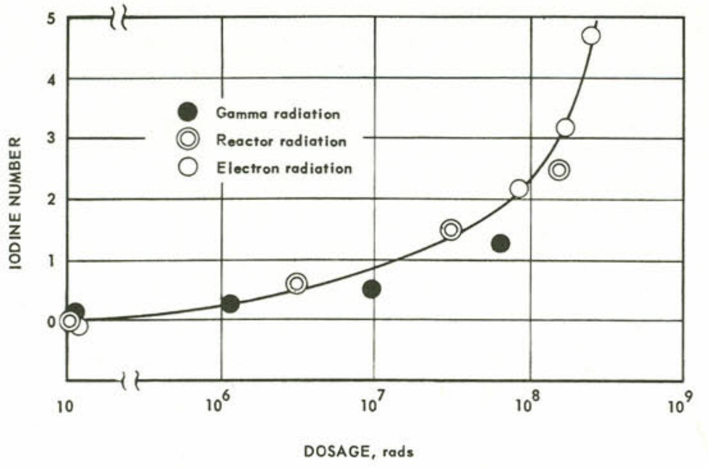

Fig. 13 – Comparison of reactor, gamma, and electron radiation damage to benzene

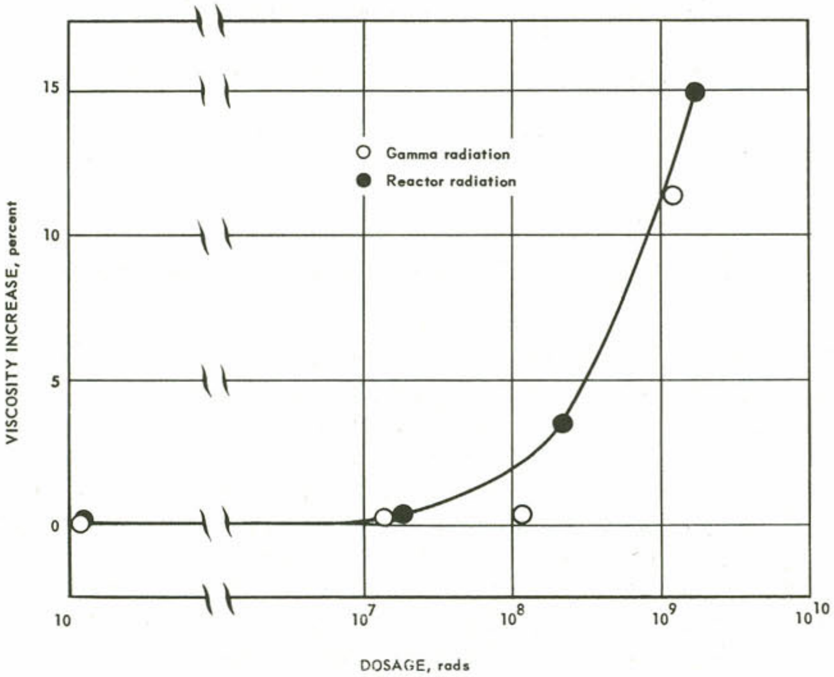

Fig. 14 – Change in viscosity of toluene as a function of absorbed energy

drawing of the test system is shown in Figure 4. The tests using the Van de Graaff accelerator are described in the following sections.

Correlation of Electron Irradiation with Various Particle Type Irradiations - The extent of electron irradiation damage to a material is based on the amount of energy absorbed by a specific material. Since the beam current and energy of the electrons are known, it is possible to calculate the amount of energy absorbed by a material during a definite exposure. The unit of energy absorption is termed a rad and is defined as 100 ergs absorbed per gram. Similarly, with respect to reactor irradiations, if the intensity and energy spectra of the fast and thermal neutrons are known, it is possible to calculate the amount of energy absorbed by a specific material.

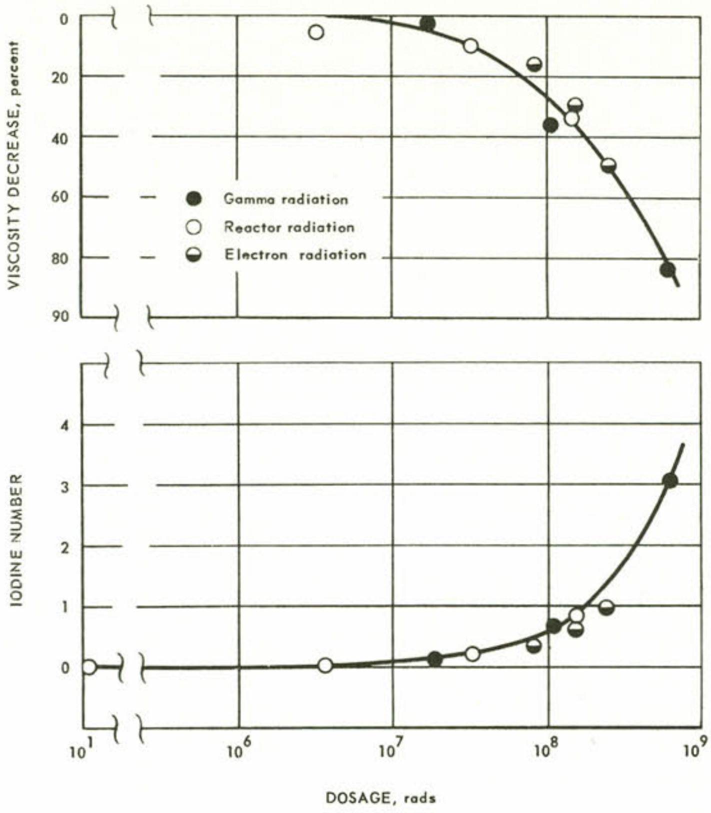

Fig. 15 – Comparison of reactor, gamma, and electron damage to glycerin

Experimental work on the correlation of various particle types on the basis of energy absorption was devoted to the irradiation of benzene, toluene, and glycerin. The data shown in Table 7 and Figures 13, 14, and 15 indicate that for dosages that are equal on the basis of energy absorption the damage from electron irradiation matches within a factor of 2 the damage from gamma and reactor irradiations. Further correlation has been established on the comparison of electron and LITR irradiation of two alkylbenzene materials. Figure 16 compares these results. The accuracy of calculations of dosages may be in error by a factor of 2 because of uncertainty of flux and sample position during the reactor irradiation test. However, reasonably good agreement of the results was obtained. The relative stability of the two materials was the same for both types of exposure.

This comparison of the effects of electron and reactor irradiations indicated that electron irradiations could be used to advantage in studying damage to organic liquids.

Screening of Organic Liquids for Possible Moderator Materials - In the screening of organic liquids using 2-Mev electrons, several types of materials were tested. These consisted of hydrocarbons, ethers, ketones, alkylated benzene ring structures, and alkylated naphthalene structures. Of these materials evaluated, the longer chain compounds were found to be less radiation stable than the shorter or branched chain compounds. Tests of such alkylated benzene ring compounds as alkylbenzene 250 and alkylbenzene 350 showed the alkylbenzene 250 to be more radiation stable than alkylbenzene 350 at temperatures in the range from 300° to 500°F. Since the napthalene ring is more radiation stable than the benzene ring, a mixture of amylnaphthalenes was evaluated for radiation and temperature stability. The amylnapthalenes tested were mixtures of mono-, di-, and polyamylnaphthalene. Studies were continued in finding a naphthalene ring having temper-

TABLE 7

CHANGES IN ORGANIC LIQUIDS IN REACTOR, GAMMA, AND ELECTRON IRRADIATIONS[a]

Material	Reactor Radiation			Gamma Radiation			Electron Radiation[b]		
	Dosage, rads	Viscosity,[c] centistokes	Iodine Number	Dosage, rads	Viscosity,[c] centistokes	Iodine Number	Dosage, rads	Viscosity,[c] centistokes	Iodine Number
Benzene	0		0.0	0		0.3	0		0
	3×10^6	d	0.615	1.1×10^6	d	0.28	8.4×10^7	d	2.2
	3×10^7	-	1.47	9.9×10^6	-	0.48	1.7×10^8	-	3.25
	1.5×10^8	-	2.49	6.8×10^7	-	1.32	2.5×10^8	-	4.60
Toluene	0	d	0.68	0	d	0.58	0	d	0.51
	3.2×10^6	-	0.61	1.9×10^7	-	0.483	8.4×10^7	-	0.90
	3.2×10^7	-	0.79	1.1×10^8	-	0.64	1.7×10^8	-	1.59
	1.6×10^8	-	1.45	1×10^9	-	-	3.4×10^8	-	2.54
Glycerine	0	1064	0.08	0	1329	0.08	0	1259	0.0
	3.2×10^6	1013	0.12	1.9×10^7	1312	0.16	8.4×10^7	1079	1.06
	3.2×10^7	959	0.24	1.1×10^8	838	0.64	1.7×10^8	843	2.86
	1.6×10^8	965	0.84	6.4×10^8	212	3.06	2.5×10^8	615	4.2

[a]Reactor and gamma irradiation data have been reported previously in APEX 11.

[b]Electron irradiation was made with 30-gram samples in Van de Graaff electron beam. Operating conditions of the Van de Graaff were 50 μa at 2 Mev; the non-scattered beam impinging on the sample was 42 μa.

[c]At 68°F

[d]Viscosity changes in benzene and toluene at the indicated dosages were within the experimental error of measurement and therefore were not included.

TABLE 8

EXPERIMENTAL PHYSICAL PROPERTY CHANGES OF ORGANIC LIQUIDS WITH VAN DE GRAAFF IRRADIATION

	Pentalene 95		Pentalene 195		Alkylbenzene 250		Mixture Alkylbenzene 250 and Dowtherm-A 1:1 by weight	
	Original	Irradiated	Original	Irradiated	Original	Irradiated	Original	Irradiated
Dosage, rads		4.28×10^9		4.28×10^9		1.2×10^9		4.86×10^9
N_H	4.8[a]	5.3[b]	5.2[a]	5.1[a]	5.7[b]		4.8[b]	5.0[b]
N_H/density ratio	5.9	6.0	6.4	6.1	7.2	7.1	5.6	5.5
Viscosity, centipoises at 100°F	14.2	51.0	18.1	111.4	6.29	7.82	3.12	39.1
300°F	1.45	2.34	1.44	0.889	0.864	0.75	0.634	2.34
Gas Evolution, liters/liter		30.0		42.0		30.7		24.6
Temperature of irradiation, °F		500		400		500		300

[a]Determined at 400°F

[b]Determined at 300°F

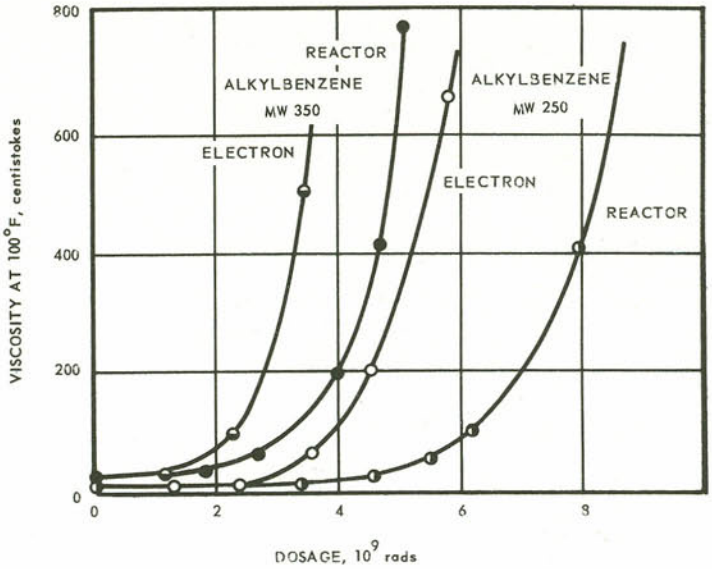

Fig. 16 – Comparison of reactor and electron irradiation of two alkylbenzenes

ature stability to 600°F. A mixture of polyamylnaphthalenes having a boiling point of 625°F was tested for radiation stability.

High Temperature, High Dosage Evaluations of Some Feasible Moderators - Since the reactor irradiation results obtained previously on alkylbenzene could be duplicated by Van de Graaff irradiation, the Van de Graaff was used for additional studies of organic liquids.

To take partial advantage of the higher N_H* of alkylbenzene 250 (5.4 at 450°F) and the stability of Dowtherm-A, a mixture of equal parts of the two materials was irradiated in the Van de Graaff accelerator. The N_H of this mixture was approximately 4.3. Figure 17 compares the gas evolution from alkylbenzene 250, from Dowtherm-A, and from a mixture of equal parts of the two liquids as a function of LITR and Van de Graaff radiation dosage. Figure 18 compares the viscosity at 100°F of the three liquids as a function of dosage, and Figure 19 compares the viscosities at 300°F. The curves suggest that the mixture is more promising than alkylbenzene 250 as a possible moderator liquid since viscosity change in the mixture is less by a factor of approximately 2.

Two selected organic liquids that are stable at high temperatures were evaluated using 2-Mev electrons from the Van de Graaff accelerator. The liquids are mixtures of mono-, di-, and polyamylnaphthalenes and are commercially known as Pentalene 95 and Pentalene 195. The Pentalene 95 is an undistilled mixture, whereas the Pentalene 195 is a distilled mixture of amylnaphthalenes. Since the naphthalene ring is more stable to radiation than the benzene ring, the Pentalenes should be quite radiation stable.

Experimental data obtained from the electron irradiation of these organic materials are given in Table 8. Gas evolution and changes of viscosity measured at 100°F and 300°F are graphically compared in Figures 20, 21, and 22 respectively.

Pentalene 95 showed damage comparable to that of the 1:1 by weight mixture of alkylbenzene 250 and Dowtherm-A. The viscosity increase of Pentalene 195 was greater than

$$^*N_H = \frac{\text{number of hydrogen atoms/cc}}{10^{22}\ \text{hydrogen atoms/cc}}$$

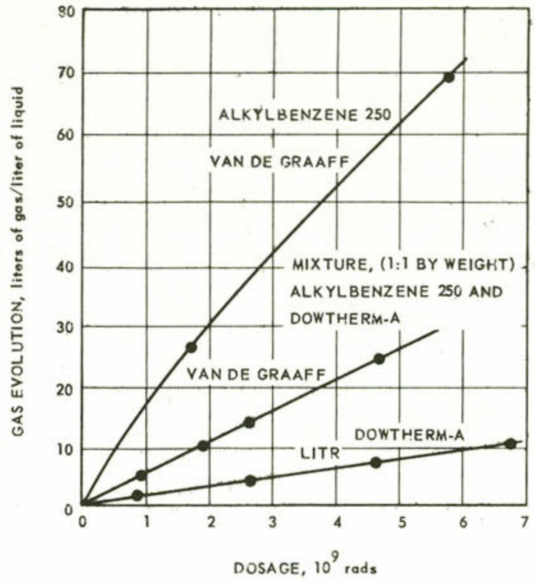

Fig. 17 – Gas evolution from alkylbenzene 250, Dowtherm-A, and a mixture of alkylbenzene 250 and Dowtherm-A as functions of LITR and Van de Graaff radiation

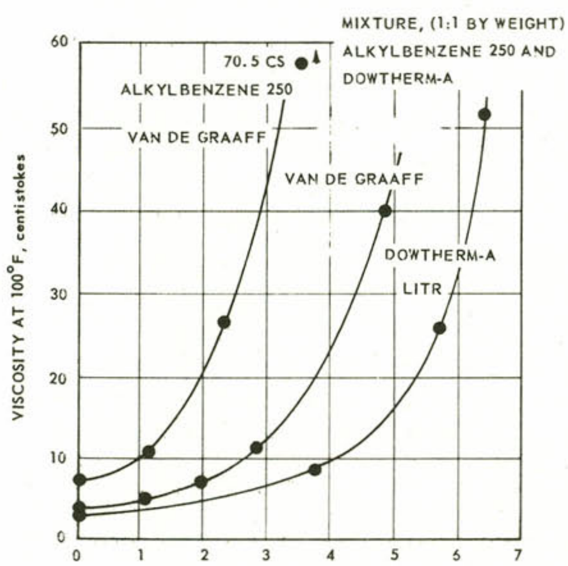

Fig. 18 – Viscosity at 100°F of alkylbenzene 250, Dowtherm-A, and a mixture of alkylbenzene 250 and Dowtherm-A as functions of LITR and Van de Graaff radiation

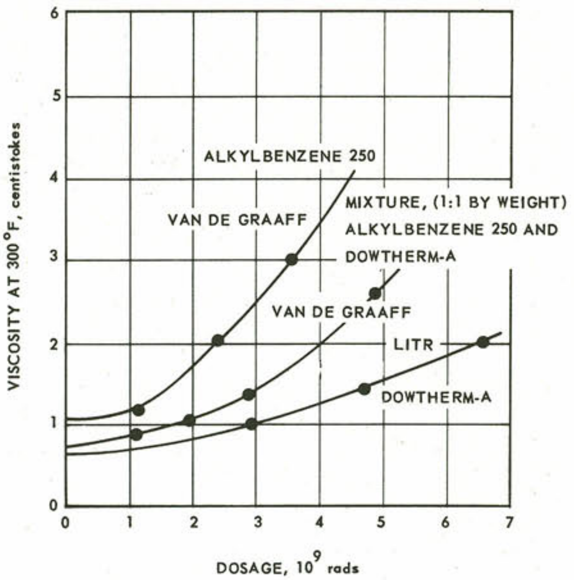

Fig. 19 – Viscosity at 300°F of alkylbenzene 250, Dowtherm-A, and a mixture of alkylbenzene 250 and Dowtherm-A as functions of LITR and Van de Graaff radiation

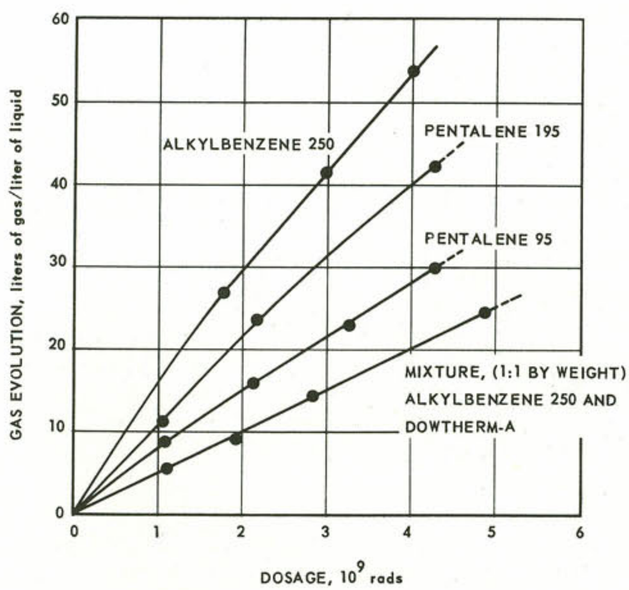

Fig. 20 – Gas evolution from alkylbenzene 250, a mixture of alkylbenzene and Dowtherm-A, Pentalene 95, and Pentalene 195 as functions of Van de Graaff radiation

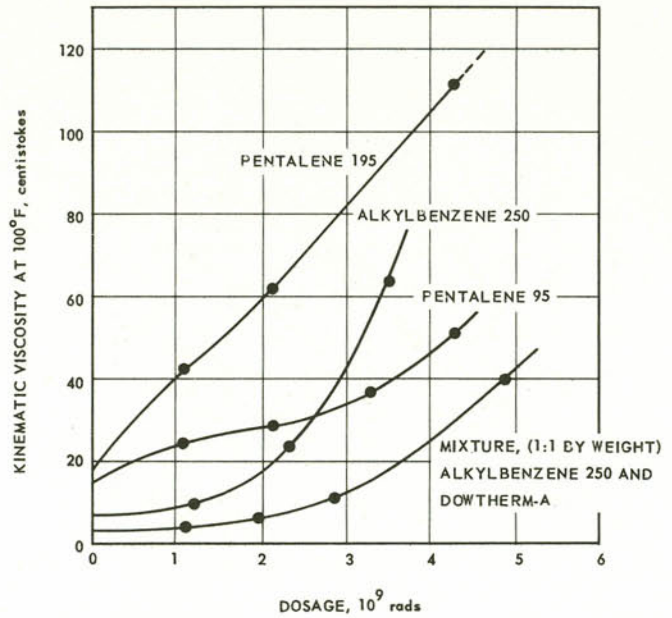

Fig. 21 – Kinematic viscosity at 100°F of alkylbenzene 250, a mixture of alkylbenzene 250 and Dowtherm-A, Pentalene 95, and Pentalene 195 as functions of Van de Graaff radiation

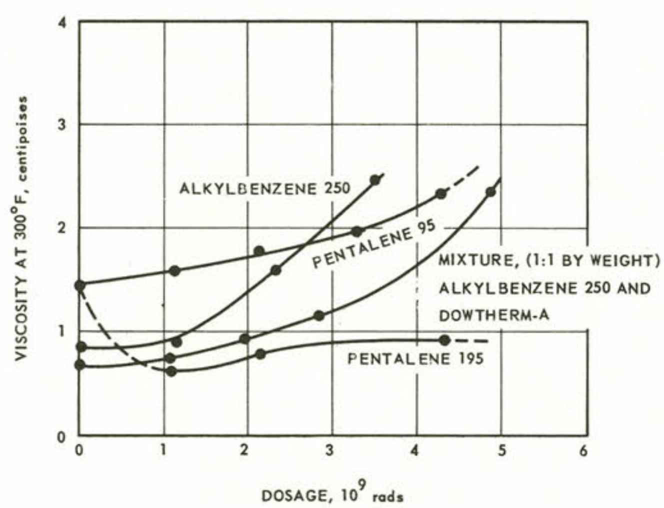

Fig. 22 – Absolute viscosity at 300°F of alkylbenzene 250, a mixture of alkylbenzene 250 and Dowtherm-A, Pentalene 95, and Pentalene 195 as functions of Van de Graaff radiation

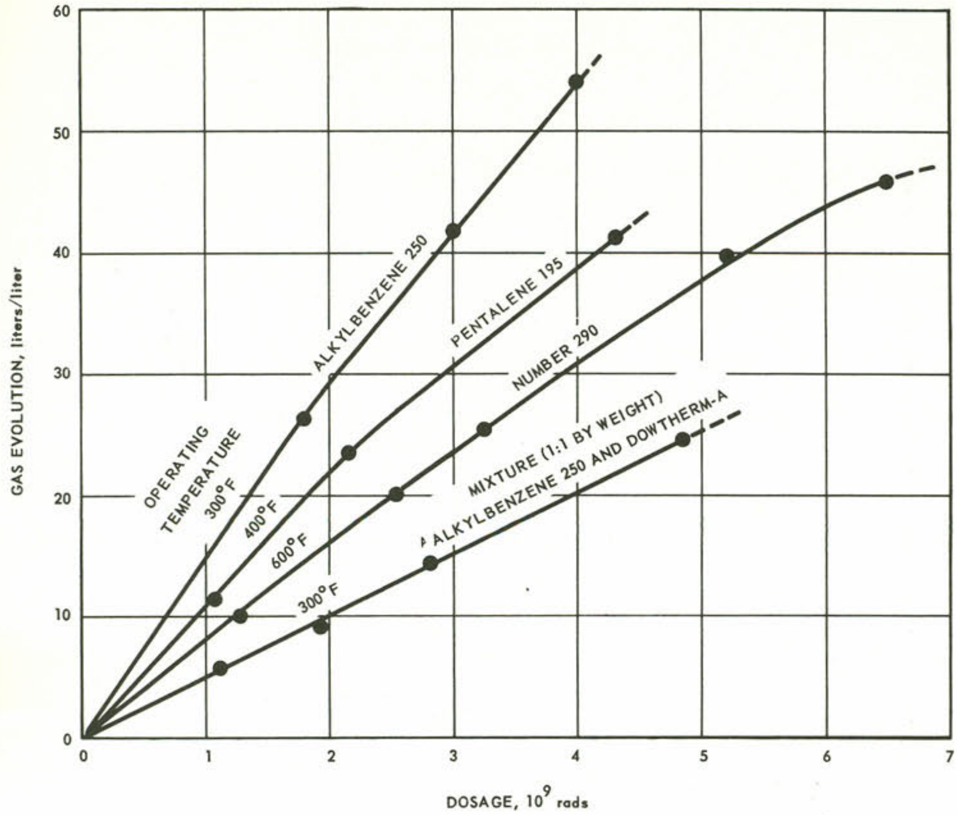

Fig. 23 – Gas evolution from alkylbenzene 250, Pentalene 195, number 290, and a mixture of alkylbenzene 250 and Dowtherm-A as functions of Van de Graaff radiation

that of Pentalene 95 by a factor of 2 at 100°F; however, at 300°F the viscosity of Pentalene 195 was less than that of Pentalene 95 by a factor of approximately 3.

The need for materials that can be used at temperatures up to 600°F has necessitated irradiation studies of many new high temperature organic liquids. Pentalene 195 was distilled at 625°F so that the lower boiling fractions from the original material were removed. The residue, termed number 290, had an initial boiling point of 625°F. This liquid was irradiated at 600°F in the Van de Graaff accelerator to determine the pertinent physical property changes as a function of 2-Mev electron radiation. The N_H of number 290 at the operating liquid moderator temperature of 600°F is 4.5 and is comparable to that of alkylbenzene 250 (N_H = 4.8) and water (N_H = 4.5) at this temperature. At a temperature of 400°F, at which the main bulk of the shield liquid would be operating, the N_H of Pentalene 290 is 5.06, that of alkylbenzene 250 is 5.4, and that of water is 5.7.

Experimental data obtained from the Van de Graaff electron irradiations of number 290 are given in Table 9. Viscosities measured at 100°F and 300°F, and gas evolution are compared in Figures 23, 24, and 25 with those of alkylbenzene 250, Pentalene 195, and the 50:50 mixture (by weight) of alkylbenzene 250 and Dowtherm-A as functions of radiation dosage. Figure 26 is a plot of viscosity increase of number 290 at 600°F as a function of Van de Graaff radiation dosage.

A detailed study of Figures 23, 24, 25, and 26 reveals that the evolution of gas from number 290 under irradiation is less than that from Pentalene 195 or alkylbenzene 250. The viscosity measured at 100°F of non-irradiated and irradiated 290 is higher than the

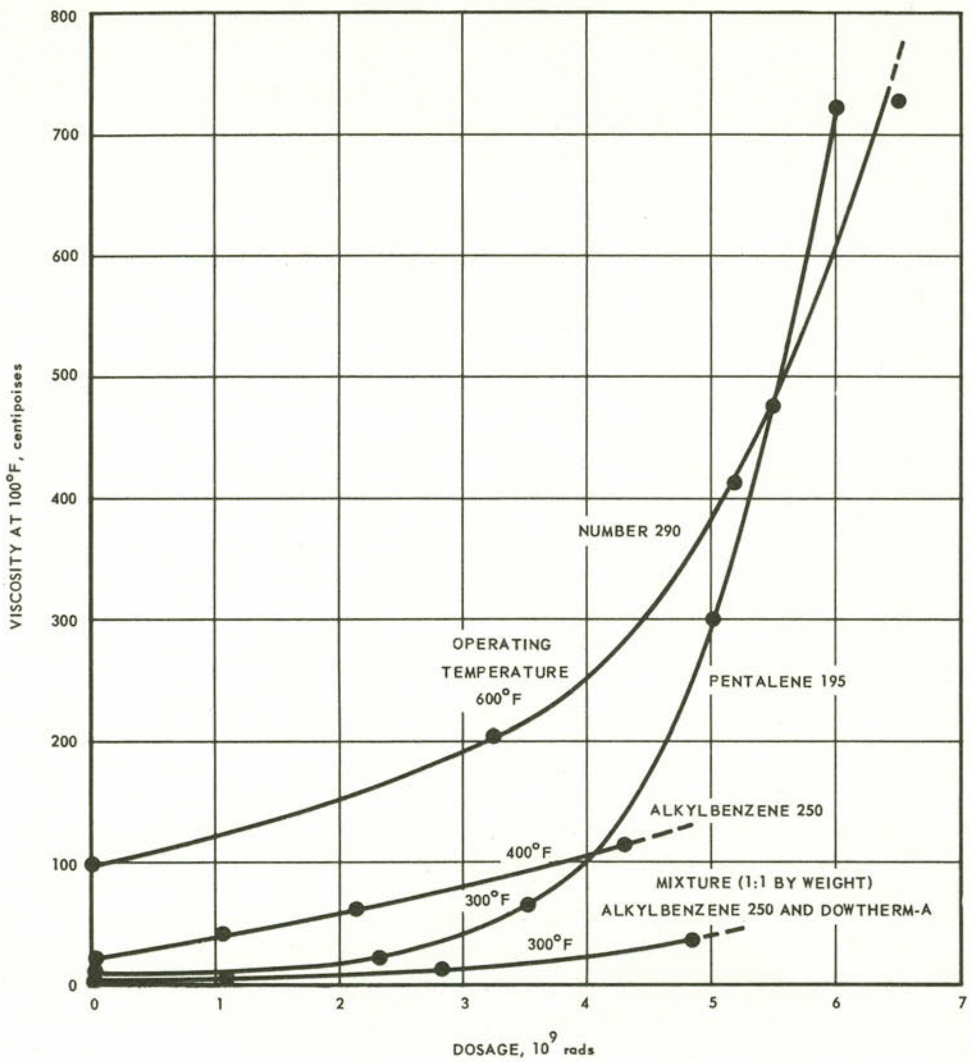

Fig. 24 – Absolute viscosity at 100°F of number 290, Pentalene 195, alkylbenzene 250, and a mixture of alkylbenzene 250 and Dowtherm-A as functions of Van de Graaff radiation

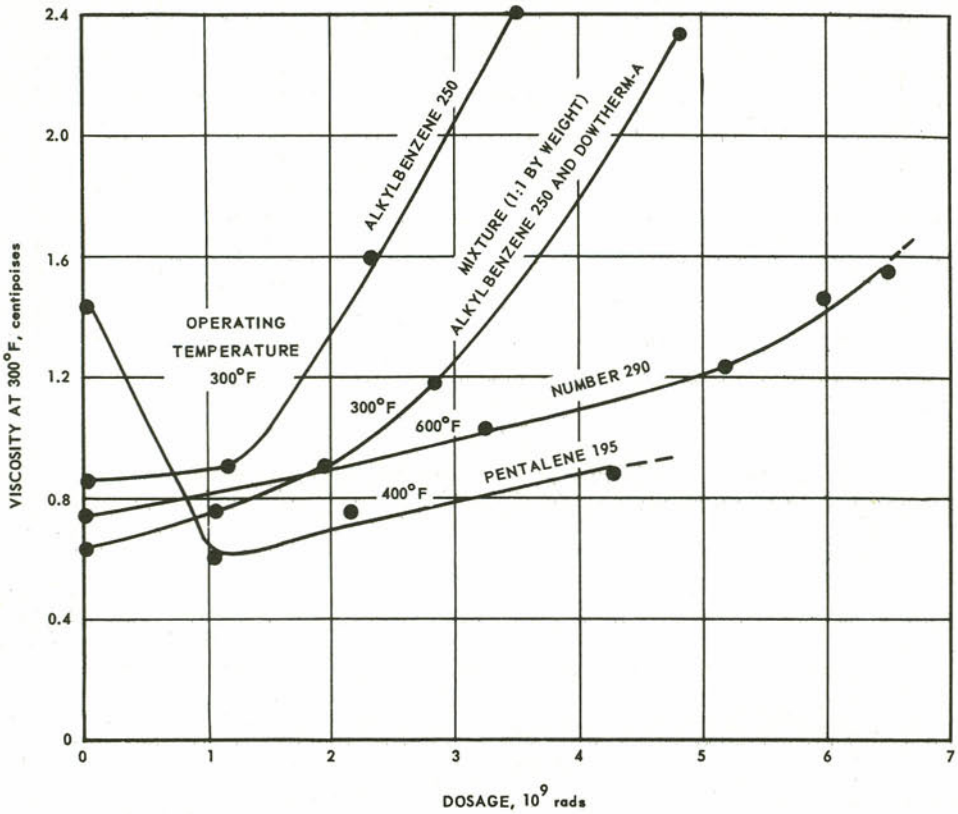

Fig. 25 – Absolute viscosity at 300°F of number 290, Pentalene 195, alkylbenzene 250, and a mixture of alkylbenzene 250 and Dowtherm-A as functions of Van de Graaff radiation

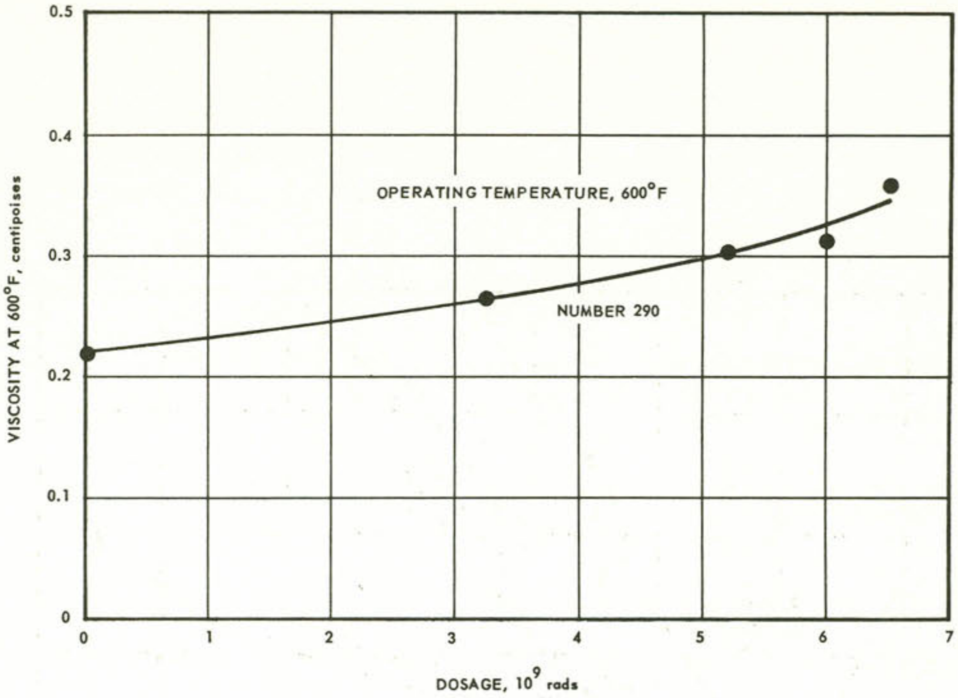

Fig. 26 – Absolute viscosity at 600°F of number 290 as a function of Van de Graaff radiation

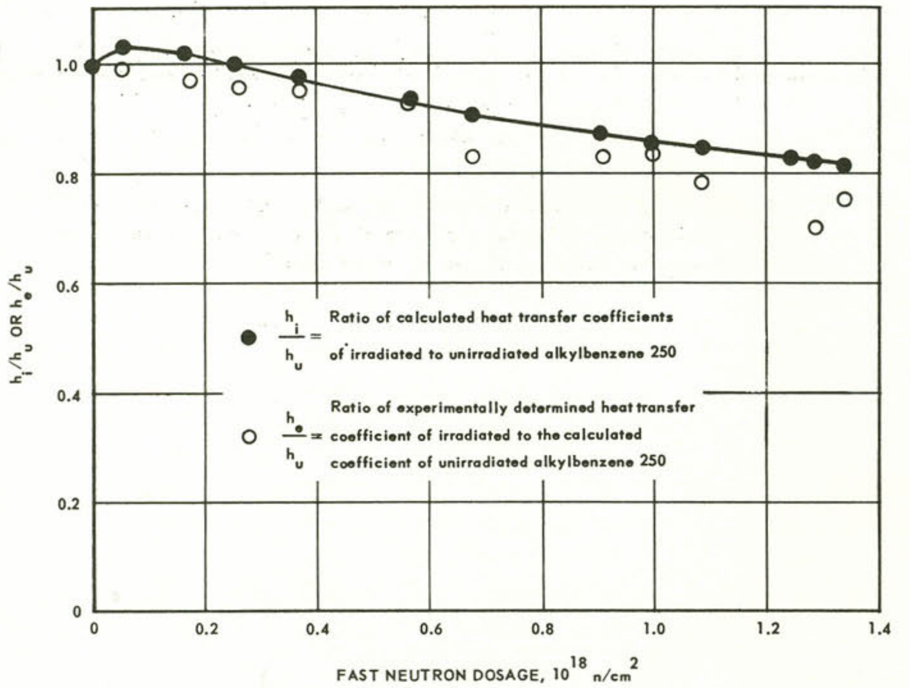

Fig. 27 – Comparison of the ratio h_e/h_u to the ratio h_i/h_u versus fast neutron dosage

TABLE 9

EXPERIMENTAL PHYSICAL PROPERTY CHANGES OF NUMBER 290 WITH VAN DE GRAAFF IRRADIATION

		Original	Irradiated
Dosage, rads			6.5×10^9
N_H value at:	100^oF	6.4	
	300^oF	5.3	
	600^oF	4.5	
Density, grams/cc at:			
	100^oF	0.919	0.956
	300^oF	0.835	0.878
	600^oF	0.713	0.750
Viscosity, centipoises at:			
	100^oF	87.5	726.4
	300^oF	0.74	1.55
	600^oF	0.22	0.36
Gas evolution, liters/liter			46.6
Boiling point, ^oF		625	670
Temperature of irradiation, ^oF			600

TABLE 10

EFFECT OF IRRADIATION ON THE HEAT TRANSFER CHARACTERISTICS OF ALKYLBENZENE 250

Fast Neutron Dosage, fn/cm^2	Density (ρ), lb/ft^3	Viscosity (μ), lb/ft-hr	h_u	h_i	h_i/h_u	h_e	h_e/h_u	Heat Balance[a] $\left(\frac{Q_1 - Q_2}{Q_1}\right)100\%$
0 (Control)	45.5	1.09			1.0	227		
5.6×10^{16}	45.7	1.01	228	235	1.03	227	0.99	+3
1.65×10^{17}	45.9	1.04	231	236	1.02	224	0.97	+3
2.57×10^{17}	46.1	1.09	230	230	1.0	221	0.96	+3
3.71×10^{17}	46.4	1.17	223	217	0.973	212	0.95	+3
5.65×10^{17}	46.8	1.31	226	210	0.924	210	0.93	+2
6.78×10^{17}	47.0	1.39	228	207	0.908	190	0.83	-8
9.09×10^{17}	47.4	1.54	235	205	0.871	195	0.83	-4
9.93×10^{17}	47.6	1.60	223	191	0.858	187	0.84	-4
1.09×10^{18}	47.8	1.66	242	205	0.847	189	0.78	-15
1.24×10^{18}	48.1	1.76	233	192	0.826	186	0.80	-6
1.29×10^{18}	48.2	1.79	232	190	0.820	162	0.70	-20
1.34×10^{18}	48.2	1.82	231	191	0.815	172	0.75	-23

Temperature = 450^oF

Average Velocity $\cong$ 2fps

[a]Q_1 = Electrical power input; $Q_2 = WC_p \Delta T$ as determined from measured flow and thermocouple readings.

viscosities of the three other liquids studied. However, the viscosity of 290 is lower than the viscosities of the alkylbenzene liquids after initial irradiation when measured at temperatures above $300^{o}F$.

A preliminary survey of the infrared absorption spectrum of the irradiated number 290 indicated that no unsaturation was present in the sample. This was later confirmed by a bromine number determination. The spectrum indicated further that the functional groups appeared to be unaffected by irradiation except for a slight change to the 2-mono-substituted naphthalene.

Analysis of Heat Transfer Data

The heat transfer coefficient of alkylbenzene 250 was evaluated using the following relation:

$$h = A \frac{K}{D} \left(\frac{DG}{\mu} \right)^{0.8} \left(\frac{C_p \mu}{K} \right)^{0.4}$$

where

h	= heat transfer coefficient	$Btu/hr\text{-}ft^2\text{-}^oF$
D	= tube inside diameter	ft
G	= fluid mass velocity	$lb/ft^2\text{-}hr$
C_p	= fluid specific heat	$Btu/lb\text{-}^oF$
μ	= fluid viscosity	lb/ft-hr
K	= fluid thermal conductivity	$Btu/hr\text{-}ft\text{-}^oF$
A	= constant	

Values of the heat transfer coefficient h were determined for the unirradiated and irradiated liquid throughout the heat transfer test described earlier in this report. Ratios of the heat transfer coefficients of the irradiated liquid h_i to the unirradiated liquid h_u were computed and are shown in Table 10.

Values for the experimentally determined heat transfer coefficient h_e were determined using the relation:

$$h_e = \frac{Q}{(A_H)(\Delta T_M)}$$

where Q = heat dissipated in test section
A_H = heated surface to test section
ΔT_M = mean wall-to-liquid temperature difference in the test section.

The ratio h_e/h_u, based on experimental values, shows a cumulative effect of fouling and viscosity on the heat transfer coefficient.

Values of h_e and the ratio are given in Table 10.

Figure 27 is a plot of the h_i/h_u and h_e/h_u ratios as a function of fast neutron dosage. A study of the curves indicates that there is no serious degradation of the heat transfer coefficient of alkylbenzene 250 for fast neutron dosages up to 1.4×10^{18} n/cm^2. The data indicate that the heat transfer coefficient was decreased by irradiation to approximately 80 percent of the unirradiated value when subjected to a dosage of 1.4×10^{18} fast neutrons/cm^2.