IN—PILE LOOP IRRADIATION STUDIES OF ORGANIC COOLANT MATERIALS

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Department of Nuclear Engineering
Massachusetts Institute of Technology
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PROGRESS REPORT
OCTOBER 1, 1963 - DECEMBER 31, 1964

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1.0 SUMMARY

Irradiations of Santowax WR in the MITR at a fast neutron fraction of 40% at temperatures of 425°F to 800°F have been completed. The irradiations were carried out with steady-state distillation processing and recycle of the coolant. Samples of degradation gases have been analyzed and the gas generation rate determined for the irradiation of Santowax WR.

Analysis of the results on the decomposition of terphenyls using first-order kinetics indicates that the rate of pyrolysis of irradiated coolant is much greater than that of unirradiated coolant. The ratio of the first-order pyrolysis rate constant for irradiated coolant to the rate constant for unirradiated coolant varies with temperature but, for example, is approximately 10 at a temperature of 750°F. This indicates that the pyrolysis can have an important effect on the design and operation of organic-cooled reactor systems.

A series of six measurements of dose rates by calorimetry have been carried out; some difficulty with the use of polyethylene as an energy absorber has been experienced. Nine measurements of the fast neutron flux at the irradiation facility have been made using a number of epithermal resonance and fast neutron threshold foils.

Heat transfer measurements have continued and have again indicated no evidence of fouling over long periods of loop operations. A new temperature controller and trim heater have been installed in the loop and operated as a means of providing more constant temperature during the course of the irradiations, especially at high temperatures where pyrolysis is important. The mass of circulating coolant in the loop has again been checked using tritium dilution.

Work has been completed on fractionation of high boiler formed during irradiation of Santowax OMP and on determination of the molecular weight of the various fractions. Analyses by gas chromatography of reference samples submitted by Euratom have been completed and samples of M.I.T. irradiated coolant have been sent to Euratom for analytic comparison.

A new in-pile section has been designed, procured, delivered, and has been undergoing a series of acceptance tests and repairs. Calculations and measurements are currently underway for determining the feasibility of using a cadmium sleeve around the in-pile section as a thermal-neutron-to-gamma-ray converter in order to decrease the fast neutron fraction of the dose rate in the irradiation facility; a low fast neutron fraction is desired to permit a direct measurement of the fast neutron effect, G_N/G_γ.

Visits were made to the A.E.C.L. Chalk River Laboratories and to the principal European research centers engaged in studies of organic coolants for nuclear reactors and a report submitted to the AEC (1).

A report covering an analysis of the effects of radiolysis and pyrolysis during the Santowax OMP irradiations and the first four irradiations of Santowax WR has been prepared and distributed (2). A report on the molecular weight distribution in high boiler has been prepared and will be distributed in January, 1965 (3). A more detailed report on the results of Santowax WR irradiations is in preparation. Two papers on the results of the Santowax WR irradiations and an interpretation of the results were prepared and delivered at the San Francisco meeting of the American Nuclear Society held in December, 1964 (4, 5).

2.0 IRRADIATIONS OF SANTOWAX WR

Santowax WR was subjected to mixed fast neutron and gamma ray irradiation in the MITR under a variety of operating conditions during the period from July, 1963, through September, 1964. The principal experimental conditions and results of these irradiations are presented in Table 1. Figure 1 is a plot of the first-order G* values for OMP disappearance vs irradiation temperature and shows the marked increase in the rate of degradation with temperature; this increase begins at a temperature of about 350° to 375°C (see Reference (2) for discussion of apparent reaction kinetics and relation of

Table 1

Results of Santowax WR Irradiations in M.I.T. Reactor

$$G(-omp) = \frac{\text{molecules omp degraded}}{\text{100 ev absorbed}}$$

$$G^*(-omp) = \frac{G}{C_{omp}} = \frac{\text{molecules omp degraded/100 ev energy absorbed}}{\text{wt. fraction omp in coolant}}$$

$\bar{d}$ = 18.2 - 20.6 milliwatts/gm

$$f_N = 0.40 \pm 0.04 \frac{\text{watts from fast neutrons}}{\text{watts total dose}}$$

Run No.	Date	Method Operation[a]	Temp. Irradiation Zone		C, weight %			G(-omp)[c]	G*(-omp)[c]
			°F	°C	OMP	DP	Bottoms		
N	1/1/64 - 1/18/64	Tr	425	218	69-58	31-42	–	–	0.26±0.08
11	8/25/64 - 9/25/64	SS	610	321	86	14	10[b]	0.39±0.05	0.46±0.06
5	1/20/64 - 3/10/64	SS	700	371	55	45	31	0.23±0.02	0.40±0.03
3	7/25/63 - 9/26/63	Tr	750	399	78-45	22-55	–	–	0.58±0.05
3	10/2/63-11/27/63	SS	750	399	54	46	30	0.37±0.02	0.63±0.03
6	3/12/64 - 4/12/64	SS	750	399	69	31	15	0.32±0.03	0.46±0.04
7	4/20/64 - 5/8/64	SS	750	399	74	26	12	0.41±0.04	0.55±0.06
4	12/4/63-12/23/63	SS	780	416	62	36	25	0.60±0.06	0.97±0.10
8	5/11/64 - 6/12/64	Tr	780	416	69-52	31-48	–	–	to be analyzed
9	6/18/64 - 7/20/64	SS	800	427	51	49	28	0.86±0.04	1.70±0.08
10	7/21/64 - 8/25/64	SS	800	427	63	37	17	0.93±0.07	1.50±0.12

[a] SS = steady-state bottoms
Tr = transient

[b] High Boiler (lower temperature cut-off for distillate than Bottoms)

[c] Error limits are one standard deviation.

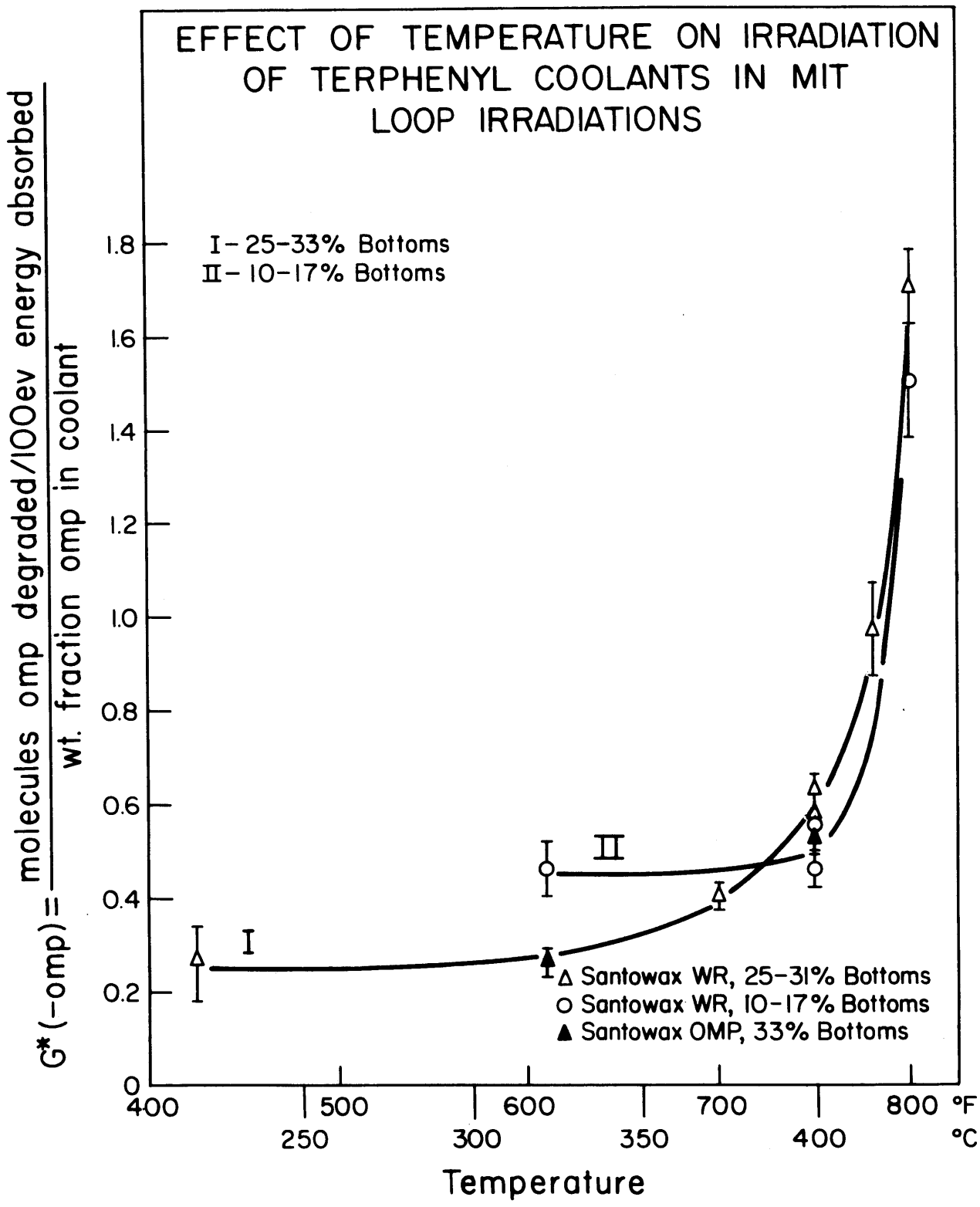

EFFECT OF TEMPERATURE ON IRRADIATION OF TERPHENYL COOLANTS IN MIT LOOP IRRADIATIONS
I - 25-33% Bottoms
II - 10-17% Bottoms
G*(-omp) = molecules omp degraded/100ev energy absorbed / wt. fraction omp in coolant
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
I
II
Δ Santowax WR, 25-31% Bottoms
○ Santowax WR, 10-17% Bottoms
▲ Santowax OMP, 33% Bottoms
400
500
600
700
800 °F
250
300
350
400 °C
Temperature

Figure 1

first-order rate constants and G* values). The steady-state concentration of degradation products (DP) was varied from irradiation to irradiation at two different temperatures in order to provide information concerning the effect of degradation product concentration on the rate of pyrolysis as well as to provide a reevaluation of the apparent reaction order for degradation. The somewhat higher degradation yields found at low temperatures and low concentrations of degradation products has indicated that the apparent mechanism for pure radiolysis may be greater than first-order. The lower degradation yields at high temperature and low DP concentration suggest that the rate of pyrolysis may be dependent on the concentration of DP or of some DP fraction. Additional irradiations using steady-state conditions at varying coolant compositions and temperatures will be made in the future in order to provide a basis for a more statistically significant quantitative analysis of these trends.

Euratom suggested in late 1963 that the rate of pyrolysis of irradiated coolant might be significantly higher than the rate of pyrolysis of unirradiated coolant (earlier data of the Canadians had also suggested this). Consequently the data of the Santowax OMP and Santowax WR runs have been evaluated using first-order kinetics in the following simplified model (2). The rate of degradation due to irradiation was assumed to be constant and equal to that found at irradiation temperatures of 320°C and below. The increase in the overall rate of degradation observed during irradiations carried out at higher temperatures is attributed to "radiopyrolysis", or pyrolysis of irradiated coolant. The first-order rate constants of radiopyrolysis were correlated according to an Arrhenius model.

The results of this analysis are plotted in Figure 2. Curve 1 presents the first-order pyrolysis constants for unirradiated coolant whereas Curve 2 shows the results of first-order pyrolysis constants for irradiated coolant containing about 30% Bottoms. Curve 3 also shows the results of a similar analysis by M.I.T. of irradiations reported by Euratom.

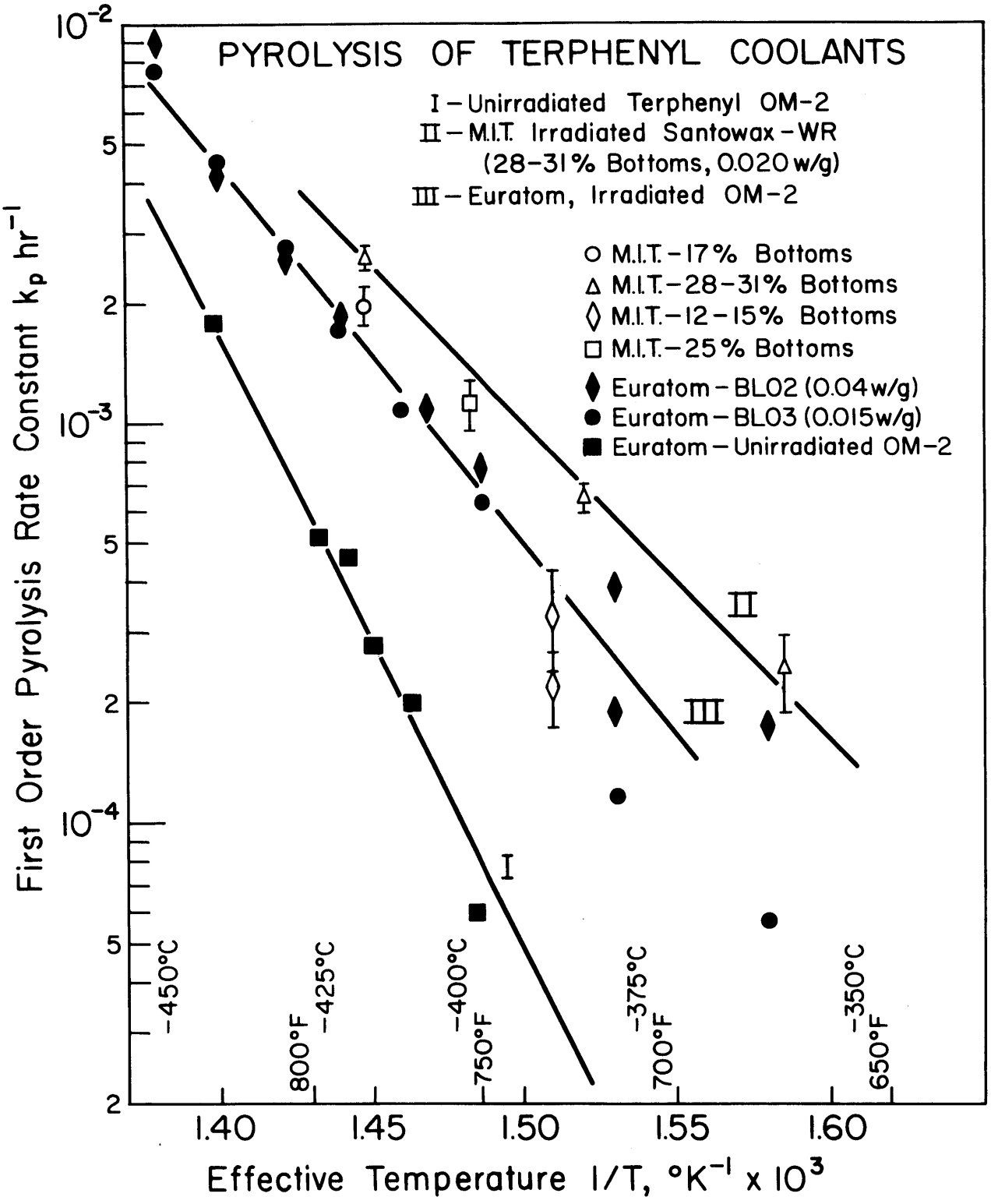

PYROLYSIS OF TERPHENYL COOLANTS
I – Unirradiated Terphenyl OM-2
II – M.I.T. Irradiated Santowax-WR
(28-31% Bottoms, 0.020 w/g)
III – Euratom, Irradiated OM-2
M.I.T. – 17% Bottoms
M.I.T. – 28-31% Bottoms
M.I.T. – 12-15% Bottoms
M.I.T. – 25% Bottoms
Euratom – BL02 (0.04 w/g)
Euratom – BL03 (0.015 w/g)
Euratom – Unirradiated OM-2
First Order Pyrolysis Rate Constant k_p hr^{-1}
10^{-2}
5
2
10^{-3}
5
2
10^{-4}
5
2
-450°C
800°F
-425°C
-400°C
750°F
-375°C
700°F
-350°C
650°F
1.40
1.45
1.50
1.55
1.60
Effective Temperature 1/T, $°K^{-1} \times 10^3$

Figure 2

The Euratom irradiations were made using the transient method of operation to about 20% DP, while the M.I.T. results were obtained using steady-state operation at about 40% DP. In addition to showing the large difference in the rates of radiopyrolysis and the rate of pyrolysis of unirradiated terphenyls and the variation of radiopyrolysis with temperature, the irradiations made at Bottoms concentrations of about 15% also indicate that the rate of radiopyrolysis is dependent upon coolant composition. The rate of radiopyrolysis increases for increasing concentrations of degradation products.

Table 2 shows the results of measurements of densities, viscosities and molecular weights made on samples of irradiated coolants taken during the Santowax WR irradiations. Figure 3 shows the temperature dependence of the density of Santowax WR irradiated under varying conditions of temperature and Bottoms concentration. The results of measurements of the density and viscosity of irradiated Santowax WR irradiated at a given temperature and concentration of degradation products agree closely with measurements made on Santowax OMP (7). The viscosity increased with increasing concentrations of degradation products, and the average molecular weight of the coolant increased as the concentration of degradation products in the coolant increased. The density of Santowax WR obtained for a given set of radiation conditions was found to be a linear function of the temperature of measurement and the temperature dependence was found to be the same for all irradiations (see Figure 3). The density generally increased with increasing concentration of degradation products.

Analyses of samples of gases taken during the Santowax WR irradiations were made using mass spectrographic and gas chromatographic methods of analysis and gas generation rates were estimated. Typical results are shown in Table 3 along with the results from Santowax OMP irradiations. Comparison of the results obtained during the 750°F irradiations of Santowax OMP and Santowax WR indicate that the composition of the gas and the gas generation rates are approximately the same for the two isomeric mixtures of terphenyls. However, increasing

Table 2

Summary of Viscosities, Densities and Molecular Weights of Irradiated Santowax WR

Temp. Irradiation Zone °F	Bottoms Concentration w/o	Temperature of Measurement 700°F μ cp	700°F ρ gm/cc	750°F μ cp	750°F ρ gm/cc	800°F μ cp	800°F ρ gm/cc	Molecular Weight Coolant	Molecular Weight Bottoms
Unirradiated	0	0.21	0.82	0.18	0.80	0.16	0.77	230	
700	31	0.51	0.87	0.44	0.85	0.38	0.82	280	
750	30	0.47	0.87	0.41	0.85	0.35	0.82	285	665
750	15	0.29	0.84	0.25	0.82	0.22	0.79	245	
750	11	0.29	0.83	0.25	0.81	0.22	0.79	235	
780	25	0.36	0.85	0.32	0.83	0.28	0.81	265	
800	28	0.26	0.86	0.22	0.84	0.20	0.82	274	
800	17	0.19	0.84	0.16	0.82	0.14	0.80	253	

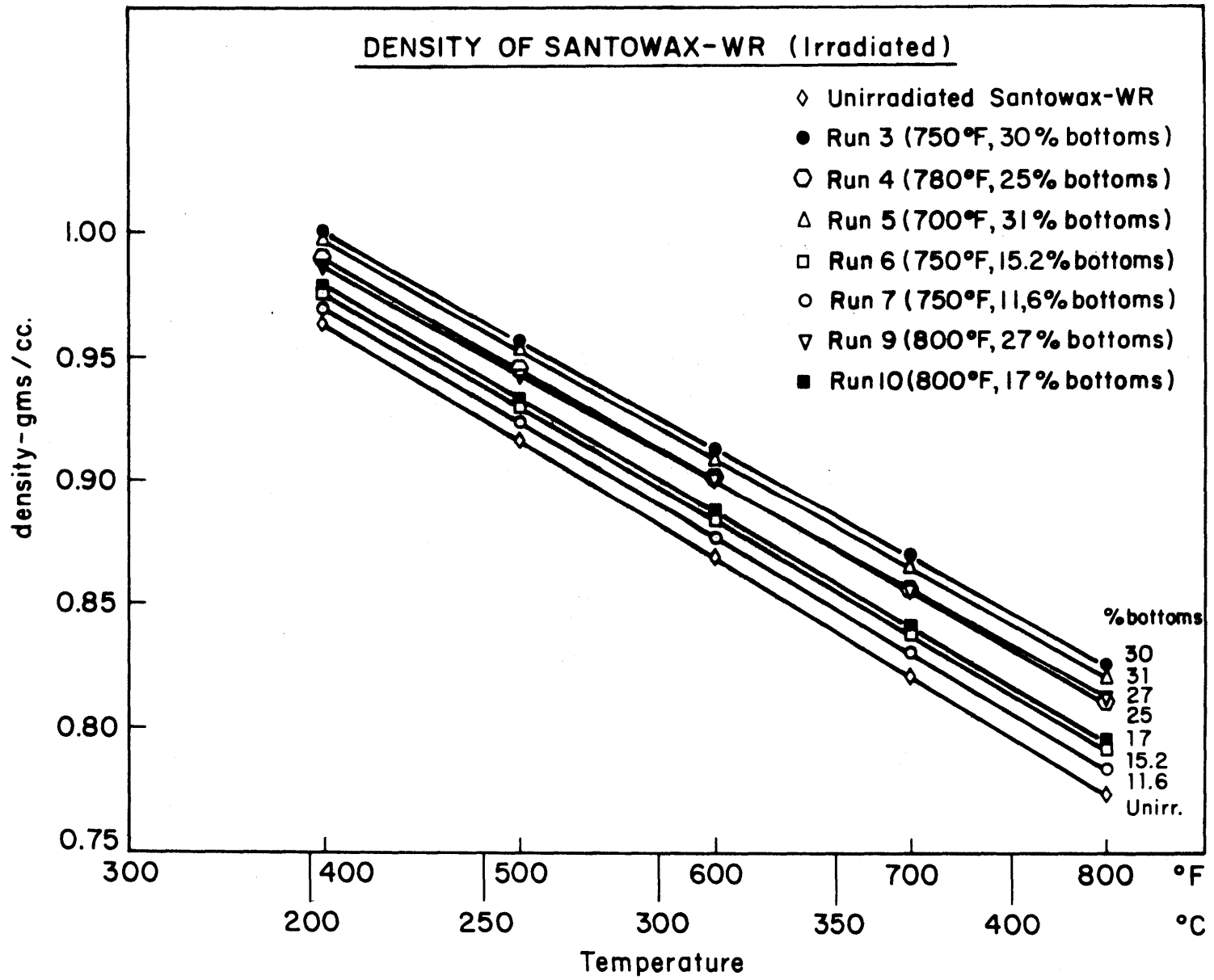

DENSITY OF SANTOWAX-WR (Irradiated)
◊ Unirradiated Santowax-WR
● Run 3 (750°F, 30% bottoms)
○ Run 4 (780°F, 25% bottoms)
△ Run 5 (700°F, 31% bottoms)
□ Run 6 (750°F, 15.2% bottoms)
○ Run 7 (750°F, 11,6% bottoms)
▽ Run 9 (800°F, 27% bottoms)
■ Run 10 (800°F, 17% bottoms)
density-gms/cc.
1.00
0.95
0.90
0.85
0.80
0.75
%bottoms
30
31
27
25
17
15.2
11.6
Unirr.
300
400
500
600
700
800
°F
200
250
300
350
400
°C
Temperature

Figure 3

Table 3

Gas Composition and Generation Rate - Irradiated Santowax OMP and WR

	Santowax OMP		Santowax WR	
	610°F - Run 1	750°F - Run 2	750°F - Run 7	800°F - Run 9
Composition: mol %				
Hydrogen	60.5	37.5	41.9	34.8
Methane	15.6	38.4	32.6	28.1
Ethane and Ethylene	14.9	16.5	15.3	11.6
Propane and Propylene	5.5	5.1	6.0	5.5
Butane and Butylene	2.0	1.1	1.6	2.0
Benzene, Hexene, Toluene and Xylene	0.5	1.1	1.7	16.5
Gas Generation Rate: (std. cm^3/watt-hr)	0.30	0.85	1.25	3.94

the temperature of irradiation from 610 to 750°F, a region in which the effects of radiopyrolysis begin to be important, caused a decrease in the relative production of hydrogen with an increased production of methane, as well as a significant increase in the rate of gas generation. Further increase in the temperature of irradiation from 750°F to 800°F maintained approximately the same ratio of the hydrogen-to-methane-to-ethane as was found at 750°F but caused a marked increase in the rate of production of aromatic species such as benzene, hexene, toluene and xylene as well as a marked increase in the overall gas generation rate.

To permit measurement of the dose rate and fast neutron fraction in the irradiation position utilized for the loop and to prepare for the next series of irradiations at a reduced fast neutron fraction, the in-pile section was removed from the MITR on September 25, 1964.

3.0 CALORIMETRY

A series of six calorimetric measurements have been made in the MITR since October, 1964. The first three series of measurements were made in the 10-plate element in the central position (Position No. 1) of the MITR from which the in-pile section used during the Santowax WR irradiations had been removed. The purpose of these measurements was to determine the change in dose rate and fast neutron fraction during the course of the Santowax WR irradiations. The results are shown in Table 4 and are compared with the calorimetry measurements made in June and July, 1963, before the in-pile section was inserted in this fuel element. During the course of these irradiations it became apparent that the results obtained with polyethylene (the energy absorber having the highest hydrogen concentration, and therefore the greatest fast neutron contribution) were not agreeing with the results obtained in the other energy absorbers (polystyrene, Santowax OMP, beryllium, and carbon) and were varying erratically from measurement to measurement, (see

Table 4

Summary of Calorimetry Results

Calorimetry Series	Date	Calorimeter Model	Core Position[a] Fuel Element[b] Status	PE, PS, SW, Be, C			PS, SW, Be, C		
				F_T,[e] watt-cc/MW-gm	f_N	f_γ	F_T,[e] watt-cc/MW-gm	f_N	f_γ
IV[a]	6/26/63	C-1	Pos. No. 1 2MR34-fresh	67.4 ±1.5	0.43 ±0.03	0.58 ±0.03	67.4 ±2.3	0.38 ±0.03	0.58 ±0.03
IV[b]	7/16/63	C-1	Pos. No. 1 2MR34-fresh	67.2 ±3.4	0.45 ±0.04	0.56 ±0.03	66.3 ±2.7	0.36 ±0.04	0.62 ±0.04
V	10/2/64	C-2	Pos. No. 1 2MR34-spent	55.7 ±3.0	0.55 ±0.05	0.53 ±0.06	58.8 ±3.0	0.38 ±0.04	0.61 ±0.04
VI	10/7/64	C-2	Pos. No. 1 2MR34-spent	60.7 ±3.5	0.59 ±0.06	0.43 ±0.05	58.8 ±2.5	0.42 ±0.04	0.60 ±0.03
VII	10/15/64	C-2	Pos. No. 1 2MR34-spent	54.7 ±3.3	0.64 ±0.08	0.46 ±0.09	60.4 ±2.3	0.40 ±0.04	0.58 ±0.03
VIII	11/17/64	C-2	Pos. No. 1 SA	36.7 ±3.0	0.52 ±0.07	0.48 ±0.07	34.7 ±1.2	0.30 ±0.04	0.70 ±0.01
IX[c]	12/15/64	C-2	Pos. No. 1 SA	—	—	—	34.3 ±2.2	0.32 ±0.04	0.68 ±0.03
X[d]	12/22/64	C-2	Pos. No. 23 SA	—	—	—	~9	~0.27	~0.73

[a]Position No. 1 - center of core
Position No. 23 - outer ring of elements, 20.9" from center of core

[b]2MR34 - partial plate fuel element (10 plates); contained 100 gms U-235 when fresh and 75 gms U-235 when spent
SA - sample assembly, contains no U-235 fuel

[c]Polyethylene calorimeter gave extraneous results in this series.

[d]This calorimetry series was a limited survey of Pos. No. 23, and results are therefore approximate.

[e]F_T represents the total dose delivered to terphenyl calculated from the calorimeter absorbers indicated in column heading.

Tables 4 and 5). A calorimeter with a fresh piece of polyethylene was made up and comparative measurements were made in Position No. 1 using the polyethylene used in Runs V-Xa and the new sample; the dose rate predicted using the new sample was 15 to 50% lower than that obtained from the older sample. Inspection of the calorimeter used in Series V-Xa indicated considerable discoloration (presumably due to radiation damage, although this had not been expected on the basis of the low total dose absorbed). New samples of polyethylene have been obtained for which the specific heat is known; the possible use of zirconium hydride as an energy absorber is also being considered.

Using the results of the polystyrene, Santowax, beryllium and carbon absorbers, the dose rate to the Santowax WR over the period of 14 months of irradiation was found to decrease by approximately 10% while the fast neutron fraction did not change appreciably. These are essentially the same trends which were found over the course of the Santowax OMP irradiation conducted during the period from summer of 1961 until spring of 1963 (7). The results shown in Table 1 allow for this decrease in dose rate during the period of the irradiations.

4.0 PLANS FOR OPERATION AT REDUCED NEUTRON FRACTION

4.1 New In-Pile Section and Irradiation Facility

Previous estimates of the fast neutron effect, G_N/G_γ, have been based on comparison of results obtained in different irradiation facilities under varying conditions of coolant composition, temperature, methods of dose measurement, and dose rates. These differences in experimental conditions, and the need to assume an apparent reaction methanism in order to compare the results, have prevented determination of G_N/G_γ with satisfactory precision. Operation of a loop under steady-state conditions at a given reactor facility in a series of irradiations carried out with radiation fields having different fast neutron fractions and with the same conditions of

Table 5

Dose Rates Measured by Polyethylene Calorimeter in Core Position No. 1

Calorimetry Series	Date	Element[a] Status	Calorimeter Model[b]	Dose Rate to Polyethylene, watts/gm-MW		
				6 inches above core center	core center	6 inches below core center
V	10/2/64	2MR34 spent	C-2	0.492	0.584	0.589
VI	10/7/64	2MR34 spent	C-2	0.595	0.636	0.523
VII	10/15/64	2MR34 spent	C-2	0.460	0.666	0.615
VIII	11/17/64	SA	C-2	0.317	0.354	0.296
IX	12/15/64	SA	C-2	0.583	0.601	0.423
Xa	12/30/64	SA	C-2	0.455	0.485	0.337
Xa	12/30/64	SA	C-3	0.240	0.300	0.290
Xb	1/6/65	SA	C-3	0.247	0.309	0.299

[a] 2MR34 - partial plate element (10 plates), 75 gms U-235 when spent

SA - sample assembly, contains no U-235 fuel

[b] Calorimeter Model Number: C indicates 3rd design model; 2 and 3 indicate absorber sample number.

analysis, dose measurement, and temperature should permit a direct, and thus probably more significant, measurement of G_N/G_γ than has been available.

Since it would not be possible to increase the fast neutron fraction in the M.I.T. loop facility enough from the 40% value employed to date to produce a significant measurement of the relative effects of fast neutrons and gamma rays, it has been decided to reduce the fast neutron fraction. The experimental errors involved in the measurement of the change of the chemical composition of the coolant, as well as the determination of the total dose rate and fast neutron fraction, require that a relatively large change in the fast neutron fraction must be realized before the ratio G_N/G_γ can be determined accurately. For instance, in the M.I.T. loop facility the fast neutron fraction must be decreased from 0.40 to approximately 0.15, or less, in order that a fast neutron effect ratio, G_N/G_γ, of 2 can be determined to be significantly different from a value of unity. The BL02 and BL03 loops operated by Euratom in the Melusine Reactor at Grenoble have experienced a fast neutron fraction of approximately 0.16 to 0.18 so that reduction of the fast neutron fraction in the M.I.T. irradiations to this level would facilitate direct comparison of the M.I.T. and Euratom results.

Consequently a new irradiation facility and in-pile section has been designed and procured. The irradiation facility consists of an aluminum tube (containing no uranium and referred to as a sample assembly) as a replacement for the 10-plate fuel element which had previously been used to surround the in-pile section and which provided a high fraction of fast neutron energy deposition in the organic. The aluminum tube acts as a flow guide for the D_2O coolant which enters at the bottom and flows upwards between the tube and the in-pile assembly. This facility can be located in any fuel position throughout the core by substituting it for a fuel element. Removal of the fuel from the element immediately surrounding the in-pile section should decrease the fast neutron fraction of the total energy absorbed by the organic coolant.

A new in-pile section having a simplified design and a larger in-pile volume was also designed for use in this new facility. The new in-pile section contains a central monitor tube which extends from the top of the reactor shield down into the in-pile section so that foils and other measuring devices having a diameter of less than 0.3 inches can be inserted into a region surrounded by the circulating coolant in the core during an irradiation. The design was sent out for bids and the vendor giving the low bid (of three) was selected. The order was placed on August 12 and delivery was expected October 7. The in-pile section was delivered on November 2. Upon inspection at M.I.T., it was found that the assembly was misaligned. The assembly was returned to the fabricator; upon return to M.I.T. leaks were found in the aluminum thimble which encloses the stainless steel loop section and serves to keep the reactor D_2O away from the hot stainless steel. The assembly was again returned to the vendor following complete X-raying; it was returned to M.I.T. on January 4, 1965, following a leak test at the fabricator's shops. Additional leaks have been detected at M.I.T. however, and plans for further action are now being formulated.

4.2 Use of Cadmium Gamma Ray Converter

Measurements of the fast neutron fraction and dose were made in the new sample assembly in both Position No. 1 (see Table 4 Calorimeter Series VIII and IX) as well as in a fuel position near the outer core tank of the MITR (see Calorimeter Series X in Position No. 23). Substitution of the new sample assembly for the 10-plate element did not result in as large a change in the fast neutron fraction as had been anticipated.

Therefore consideration is now being given to the use of a cadmium liner around the in-pile section to provide for absorption of thermal neutrons and production of gamma rays in order to increase the amount of gamma ray absorption in the organic coolant. Measurement of the reactivity worth of 24 inches of cadmium tubing in one of the outer positions of

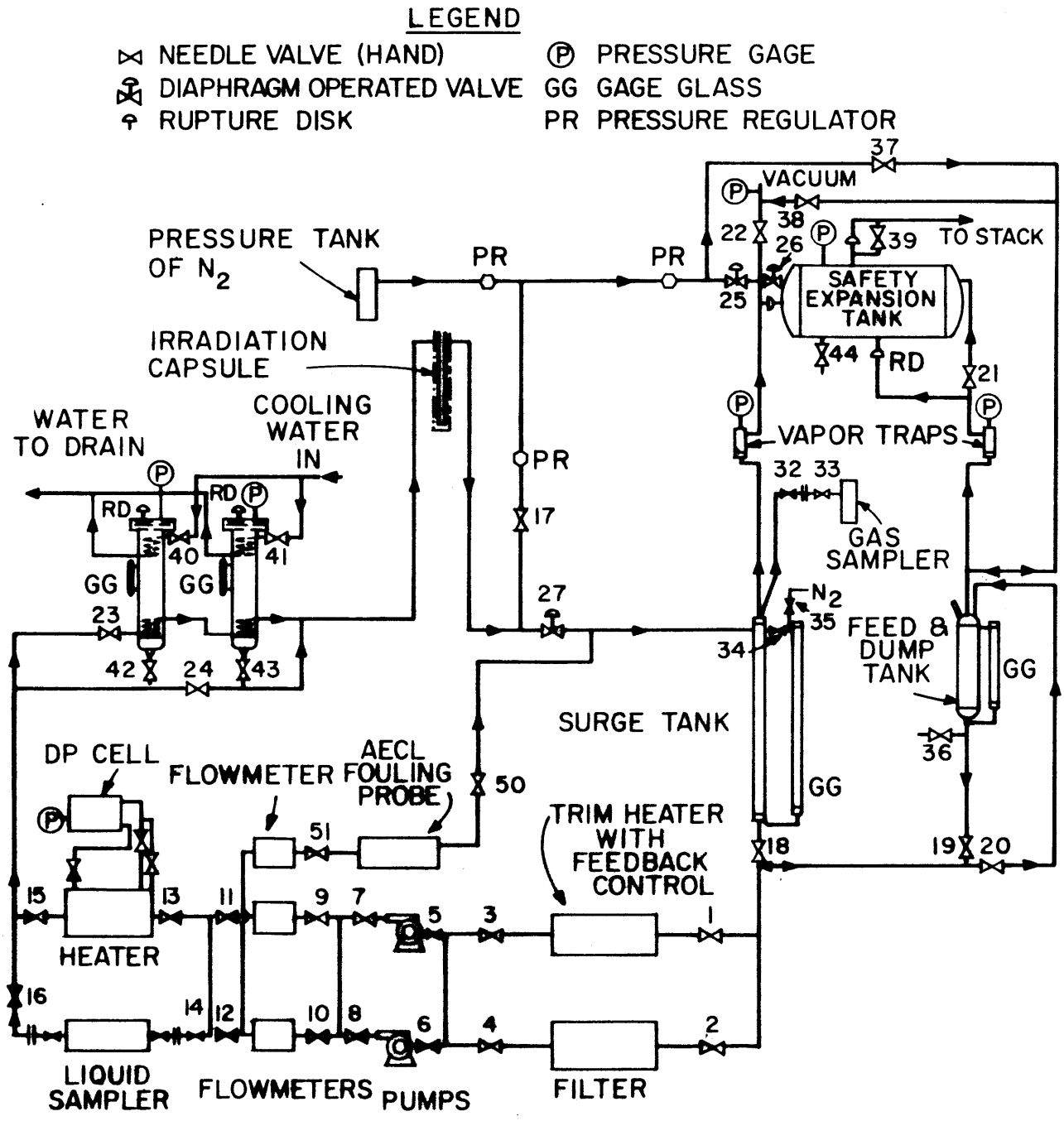
the reactor has shown the poison effect to be tolerable and calorimetry is now underway.

It is estimated that a fast neutron fraction of approximately 0.15 as well as an increased dose rate may be obtainable using a cadmium sleeve outside the in-pile section.

5.0 MODIFICATION OF OUT-OF-PILE EQUIPMENT

In August, 1964, an automatic temperature controller and trim heater were installed in the out-of-pile portion of the M.I.T. organic-cooled loop. The purpose of this modification was to provide a means for decreasing the slight temperature variations that had occurred during the course of previous irradiations. With use of this device in subsequent irradiations, the variation of temperature with time was held to within $\pm 3^{\circ}F$.

The test heater which has been in use since November, 1961, was replaced on November 30, 1964, by a new test heater (see Figure 4 for revised flow sheet of loop equipment showing location of new trim and test heaters). The test heater which had been in use showed no signs of fouling since its last inspection in June, 1963. The new test heater is equipped with pressure taps and a differential pressure cell arrangement for measuring the pressure drop across the heater. Experiments will be run in an attempt to correlate the fluid flow characteristics of the coolant with the heat transfer characteristics. Much of the previously reported data on the heat transfer of organic coolants had indicated a 0.9^{+} exponential dependence of the coefficient of heat transfer on the Reynolds number, while the bulk of the data reported in the literature for water, petroleum liquids, and gases indicates a 0.8 exponential dependence. The purpose of the present work is to determine whether the dependence of the forced convection coefficient of heat transfer on the Reynolds number for organic coolants is indeed significantly different than that of other coolants.



SCHEMATIC FLOW DIAGRAM OF MIT ORGANIC LOOP

Figure 4

6.0 DETERMINATION OF MASS OF THE LOOP

In transient experiments, the numerical value of the mass of the loop enters directly into the calculation of the G value. In steady-state experiments, the mass of the loop is needed only to obtain a minor correction term in calculating the G values in the event that the concentration of terphenyls changes slightly over the course of irradiation. The mass of the circulating loop in the MITR has been calculated from a knowledge of the measured volumes of the various parts of the system. From time to time experiments have been conducted in order to obtain an independent check of this value. In October, 1964, the mass of the circulating organic in the loop was again checked in a dilution experiment by the addition of terphenyls which contained tritium. The tritium counting was carried out by two independent laboratories and the mass of the loop obtained by the tritium dilutions agreed to within 3 and 6% of the value calculated using the normal M.I.T. method. This is considered to be very good agreement; the error introduced in the steady-state correction by an uncertainty in the mass of this magnitude is negligible.

7.0 ANALYSIS OF HIGH BOILER FRACTIONS

A study of the composition of high boiling components (HB) produced during the irradiation of Santowax OMP at 610°F and 750°F has been completed and a report prepared for distribution (3).

HB samples removed from the loop from the beginning and end of the steady-state irradiations were fractioned by sublimation and the fractions were compared in regard to number average molecular weights (MW_N) and the compositions qualitatively compared by high temperature gas chromatography. The analyses indicated that equilibrium HB composition had been achieved. The equilibrium values of MW_N for the HB from the 610°F and 750°F irradiations were 700 $\pm$ 35 and 580 $\pm$ 25

respectively. The sublimation yields for the 750°F HB indicated that "thermal cracking" and/or pyrolysis is an important process in the radiolysis of Santowax OMP.

8.0 VISITS TO OTHER LABORATORIES

In early October, 1964, E. A. Mason and T. H. Timmins of M.I.T. visited the Chalk River Laboratories of AECL for two days and held very complete and worthwhile discussions with personnel from the Chalk River and Whiteshell Laboratories concerning the organic coolant work being carried out at these laboratories as well as at M.I.T.

In the latter half of October, E. A. Mason made a trip to Europe to gather information on current results and plans in organic coolant research programs being carried out at the principal Euratom laboratories engaged in work on organic coolants. The laboratories visited were: the Central Research Laboratories of Euratom at Ispra, Italy, from which Euratom's ORGEL program is directed and at which research on pyrolysis, chemical analysis, coolant purification and out-of-pile coolant loop work as well as a very complete materials and reactor program in support of ORGEL are conducted; the C.E.A. Laboratories at Grenoble, France, where in-pile and electron irradiations of terphenyls using loops as well as out-of-pile heat transfer studies, and chemical and physical analyses are being carried out for Euratom; the Austrian Research Center at Seibersdorf where batch irradiations of samples of anthrancene oils (coal tar by-products) are being carried out to evaluate their potential as alternate coolants; the C.E.A. Laboratories at Saclay, France, where electron irradiations of terphenyls are underway; and the Harwell Laboratories of the A.E.R.E. in Great Britain where considerable research on the effects of radiation on terphenyl has been carried out in the past and more basic programs on heavy and charged particle irradiations of hydrocarbons are now underway. An extensive trip report has been prepared and submitted to the AEC (1).

Both of these trips proved valuable in providing an opportunity for mutual critical evaluation and inter-comparison of the work being done in Canada, Europe, and the U.S. as well as for planning of future complementary studies.

9.0 REPORTS

A report on the relative roles of pyrolysis and radiolysis on the degradation of terphenyls has been prepared and distributed (2). A report on the composition of High Boiler has been prepared and will be distributed in January, 1965 (3). A topical report covering the results obtained during all the irradiations of Santowax WR is also in preparation and is expected to be distributed in February, 1965. Two papers on the results of the irradiations of Santowax WR and on the relative roles of pyrolysis and radiolysis on the degradation of terphenyls were prepared and delivered at the San Francisco meeting of the American Nuclear Society in early December, 1964 (4, 5).

10.0 LITERATURE CITATIONS

1. E. A. Mason, Report on European Travel, October, 1964, (December 1964).

2. J. F. Terrien and E. A. Mason, "Relative Roles of Pyrolysis and Radiolysis in the Degradation of Terphenyls," M.I.T., Cambridge, Massachusetts, (June 1964), MITNE-48, SRO-87.

3. W. N. Bley and E. A. Mason, "The Nature of the High Boiler Degradation Products from Irradiated Santowax OMP," M.I.T., Cambridge, Massachusetts, (January 1965), MIT-334-11.

4. E. A. Mason, W. N. Bley, A. H. Swan, "Radiolysis and Properties of Santowax WR," Trans. Am. Nuc. Soc., 7, No. 2, 445, (Nov. 1964).

5. E. A. Mason, J. F. Terrien, T. H. Timmins, "Relative Roles of Pyrolysis and Radiolysis in the Degradation of Terphenyls," Trans. Am. Nuc. Soc., 7, No. 2, 441 (Nov. 1964).

6. "In-Pile Radiation Studies for Organic Reactor Coolants," Informal Letter Progress Report January 1, 1964 - April 15, 1964, M.I.T., Cambridge, Massachusetts, (April 23, 1964).

7. C. D. Sawyer, E. A. Mason, "The Effects of Reactor Irradiation on Santowax OMP at 610°F and 750°F," M.I.T., Cambridge, Massachusetts, (September 1963), MITNE-39, IDO-11,107.