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I. INTRODUCTION

During the last quarter, members of the Chemical Development Group have been concerned with six major projects: (1) thermal and radiation properties of organic materials as moderators and coolants, (2) thermal and radiation stability of zirconium hydride, (3) reactor accessibility (transport of radioactive materials in cooling systems), *and spent lead*

There have been, in addition, a number of smaller service projects, mostly of an analytical nature. Project (3) is the responsibility of other groups and will be reported in NAA-SR-284

II. ORGANIC MATERIALS AS REACTOR COOLANTS AND MODERATORS

A. Objectives

This program, which began in Fiscal Year 1953, has the following objectives for Fiscal Year 1954:

1. To determine the vapor pressure, density, specific heat, thermal conductivity, and viscosity of terphenyls up to 500° C,
2. To determine heat transfer and fouling factor information, using a pump loop in which decomposition occurs,
3. To determine the extent of purification with respect to decomposition products, necessary to maintain the viscosity at a reasonable value,
4. To adapt the pump-loop to study the effect of gamma irradiation by means of an MTR in-canal experiment,
5. To conduct Van de Graaff generator dosimetry tests to calibrate the generator beam,
6. To design and construct an irradiation cell for irradiation of organic compounds on the Van de Graaff generator, and
7. Using this cell, to irradiate p-terphenyl, biphenyl, and naphthalene at temperatures near 400° C to determine,



- (a) temperature effect on decomposition,
 - (b) radiation effect,
 - (c) catalytic influence of metal surfaces, and
 - (d) G values (the molecules of gas produced per 100 ev of radiation absorbed),
8. To continue cooperative irradiations for the California Research Corporation, and
 9. Through the capsule test method, to measure the thermal stability of sulfur and sulfur-aromatic compounds, triphenylamine, diphenylamine, triphenylphosphine, etc.

B. Summary of Experimental Results

The following results include not only data obtained during the July-October quarter, but also data on previous experiments which have been analyzed during this quarter:

1. Statitron irradiations indicate that the most desirable compounds for application in the region 200 to 400° C are the terphenyls, biphenyl, and naphthalene.
2. Pyrolytic studies, in the absence of radiation, were made on 17 organic compounds in the temperature range 400 to 500° C. Visual comparisons of these samples indicate that para-terphenyl is the most stable, with meta- and ortho-terphenyl less stable in that order. However, further quantitative analyses will be made.
3. Five convection loops of terphenyls in stainless steel or Vycor, covering a temperature range of 140 to 530° C, have also shown that only 2 per cent decomposition of para-terphenyl occurred at 440° C in 8-2/3 days.

C. Effect of Radiation on Organic Materials

1. Biphenyl (M. Feldman) - Samples of biphenyl (C₁₂H₁₀) have been irradiated with 1 Mev electrons at temperatures in the range from 60 to 260° C. Power densities in the biphenyl were of the order of 2 x 10¹⁷ Mev/cm³-hr or 4 x 10⁹ r/hr. Results at 100° C indicated little decomposition after a 20 hour run in which the biphenyl received a total of about 1000μ ah of radiation. The



gaseous decomposition products were hydrogen, with some acetylene, benzene, and hydrocarbon gases. The irradiated liquid contains decomposition products of higher viscosity than biphenyl, which may be p-terphenyl and p-quaterphenyl, among others.

The extent of decomposition of biphenyl was evaluated by mass spectrometric analysis of the gases evolved, and by ultraviolet and infra-red absorption analysis of the decomposition products remaining in the solid biphenyl phase. "Yield-values" of 0.0014 for G_{gas} and 0.11 for G_{liquid} were calculated from the analyses, and were compared with the value of 0.05 for G_{gas} given by Hochanadel³ for 2.0 Mev irradiations. Recent calibration runs on the Van de Graaff generator show that our recorded integrated electron beam current readings are high. Power losses might explain, at least in part, the lower G_{gas} values obtained. The absolute magnitude of the G values depends on whether values are based on liquid phase or gas phase analysis, as well as on the extent of energy absorbed.

2. n-Butylbenzene (E. L. Colichman, H. Gercke) - Several samples of n-butylbenzene were irradiated with electrons in a helium atmosphere. These samples were sent to the California Research Corporation for comparison with reactor-irradiated samples.

3. MTR Irradiations - Eight aluminum capsules, each containing 14.4 grams of para-terphenyl, are being prepared for irradiation at the MTR. After the material is irradiated, the extent of purification required to lower the viscosity to an acceptable value will be determined. Each capsule is to be irradiated to an nvt of 10^{19} n/cm² in the hydraulic-rabbit facility. It is estimated that the maximum temperature at the center of the para-terphenyl will be about 60° C during the irradiation. The material will remain as a solid at this temperature. Each capsule contains 6 cm³ of free space; and on the basis of data in TID 5132,⁴ the maximum pressure build-up will be less than 200 psia.

D. Pyrolysis Studies (S. Nakazato and D. J. Zigrang)

Three types of pyrolysis experiments have been made as follows: first, a number of compounds have been screened; second, decomposition rate tests have been run on the most promising compounds; and finally, attempts have been made to ascertain what effect, if any, the presence of different metals have on rates of decomposition.



Some two hundred pyrolysis tests have been made on seventeen different compounds. Compounds investigated include the three terphenyls, para-quaterphenyl, hydrogenated para-terphenyl (Monsanto HB-40), alpha-alpha binaphthyl, beta-beta binaphthyl, pyrene, chrysene, and Santowax-p (impure para-terphenyl). The polyphenyls are more desirable than benzene as heat transfer agents, because of their considerably lower vapor pressures. The basic stability of the polyphenyls lies in the stability of the phenyl group itself. The stability of the phenyl-phenyl bond comes from the double bond characteristics which it gains at the expense of the double bonds of the attending phenyl groups. Figure 1 shows the structural formulas for the four polyphenyls investigated plus the structural formulas of the other organic compounds under consideration.

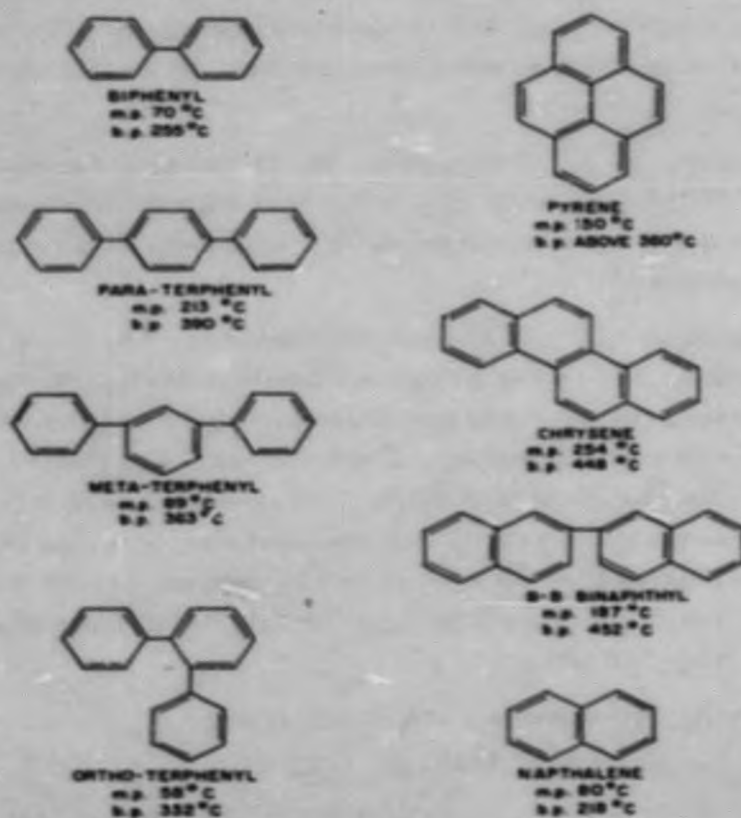



Fig. 1. Structural Formulas, Melting and Boiling Points of Polyphenyls

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The type of reaction, polymerization or de-polymerization, undergone in the pyrolysis of p-terphenyl is dependent upon temperature. A thermal loop, operated at 440° C for 8-2/3 days, showed only 2 per cent decomposition with one principle product, biphenyl. Polyphenyls were not noted, but some quantities of hydrogen and carbon were formed. A thermal loop, operated for just one day at variable temperatures up to 530° C, gave both polymers and biphenyl as products. Quartz tubes, which were maintained at 471° C for 8-2/3 days, gave both polymerization products and biphenyl. The polymer product increased with time, from 0 to 6 days.

At 475° C, a yellow solid is formed from p-terphenyl after several days. It is this solid which contributes to the increase in viscosity. The absorption spectrum of m-xenyl, (P-phenyl) xenyl benzene (which consists of two p-terphenyl groups joined in the m-positions) closely matches that of the yellow solid. The polymerization apparently does not occur in the para or ortho positions.

1. Experimental Procedures - The compounds were sealed in Vycor capsules under pressures of 10 microns or less. The dimensions of the capsules were 9 millimeters outside diameter, 9 inches long, and had inside surface areas of about 45 cm². Many of the capsules contained strips of Type 347 stainless steel having surface areas on the order of 6 cm². Occasionally, other materials were substituted for the stainless steel.

The furnace had an inside diameter of 3 inches and a heated length of 12 inches. It contained a thermal equalizer fabricated from a 3 by 10 inch rod of Type 2S aluminum. Ten holes, 1/2-inch in diameter, were drilled longitudinally through the equalizer. One hole was located on the axis, and the other nine were equally spaced on a circle 2-1/8 inches in diameter. A 1-2 inch plug of glass wool was placed at the bottom of each hole in the equalizer. Capsules were placed in nichrome capsule holders before being placed in the equalizer. Nominal furnace temperature settings were maintained to ± 2° C.

2. Experimental Results - The compounds that were screened, together with their melting points, boiling points, and observed degree of thermal stability, have been tabulated in Table I. The degree of thermal stability reported is, at present, based on visual observation; and, in most cases, is based on more than one capsule, and often on more than one temperature. Stability of the terphenyls has been investigated at temperatures as high as 500° C, while HB-40

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TABLE I
PYROLYSIS TESTS ON AROMATIC ORGANIC COMPOUNDS

Sample No.	Compound	Temp °C	Test Duration Hours	Remarks
11	para-quaterphenyl	500	16	Good cond. - somewhat decomposed
12	para-quaterphenyl	500	16	Good cond. - somewhat decomposed
13	para-quaterphenyl	500	16	Good cond. - somewhat decomposed
14	para-quaterphenyl	500	16	Good cond. - somewhat decomposed
15	para-terphenyl	500	16	Good cond. - light tan color
16	para-terphenyl	500	16	Good cond. - light tan color
17	para-terphenyl	500	16	Good cond. - light tan color
18	para-terphenyl	500	16	Good cond. - light tan color
19	meta-terphenyl	500	16	Tan color
20	meta-terphenyl	500	16	Fair cond. - light tan color
21	meta-terphenyl	500	24	Poor cond. - dark brown
22	ortho-terphenyl	500	24	Fair cond. - dark brown
23	ortho-terphenyl	500	24	Poor cond. - dark brown
25	ortho-terphenyl	500	24	Fair cond. - dark brown
27	Santowax P (97% para-terphenyl)	500	24	Good cond. - dark brown
28	Santowax P (97% para-terphenyl)	500	24	Fair cond. - dark brown
29	Santowax P (97% para-terphenyl)	500	24	Fair cond. - dark brown
31	para-terphenyl	500	72	Poor cond. - charred
32	para-terphenyl	500	72	Fair cond. - charred
33	para-terphenyl	500	72	Fair cond. - charred
34	meta-terphenyl	500	72	Fair cond. - charred, some light liquid
35	meta-terphenyl	500	72	Poor cond. - charred, some light liquid
36	meta-terphenyl	500	72	Poor cond. - charred, some light liquid
37	ortho-terphenyl	500	72	Poor cond. - charred, some light liquid
41	Santowax P	500	41	Fair cond. - charred
42	Santowax P	500	42	Good cond. - medium tan
43	Santowax P	500	65	Poor cond. - charred
44	Santowax P	500	138	Poor cond. - charred
45	para-terphenyl	500	41	Fair cond. - dark brown
46	para-terphenyl	500	19	Good cond. - slightly off white
47	para-terphenyl	500	65	Poor cond. - very dark brown
48	para-terphenyl	500	138	Poor cond. - charred
52	para-terphenyl	450	168	Good cond. - off white
53	para-terphenyl	400	192	No apparent decomposition
61	meta-terphenyl	450	72	White
63	ortho-terphenyl	450	72	Resembles liquid soap



TABLE I (Continued)

Sample No.	Compound	Temp °C	Test Duration Hours	Remarks
65	para-quaterphenyl	450	72	Good cond. - off white
67	$\beta\beta$ -Binaphthyl	450	72	Good cond. - med. brown color
68	$\beta\beta$ -Binaphthyl	450	168	Good cond. - med. brown color
70	HB-40	450	16	Brown liquid
71	para-quaterphenyl	475	168	Fair cond. - dark brown
72	para-quaterphenyl	475	245	Fair cond. - dark brown
73	ortho-terphenyl	475	168	Medium brown
76	meta-terphenyl	475	245	Good cond. - tan color
77	Pyrene	475	17	Good, reddish brown liquid
78	Chrysene	475	7	Poor, dark brown liquid
79	Chrysene	475	18	Poor, dark brown liquid
103	para-terphenyl	476	164	Dark brown liquid
104	para-terphenyl	476	164	Dark brown liquid
105	meta-terphenyl	476	164	Dark brown liquid
106	meta-terphenyl	476	164	Dark brown liquid
107	ortho-terphenyl	476	164	Dark brown liquid
108	ortho-terphenyl	476	164	Dark brown liquid
109	para-terphenyl	476	120, 168	Pt present - little change 5 days
110	para-terphenyl	476	120, 168	Cu present - little change 5 days
111	para-terphenyl	476	120, 168	303 stainless steel - little change 5 days
112	para-terphenyl	476	120, 168	Silver solder present - little change 5 days
114	para-terphenyl	475	0	Gradual transition from a white solid to a medium brown solid.
115	para-terphenyl	475	24	
116	para-terphenyl	475	48	
117	para-terphenyl	475	72	
118	para-terphenyl	475	96	
119	para-terphenyl	475	120	
120	para-terphenyl	475	144	
121	para-terphenyl	475	168	
122	para-terphenyl	475	192	
123	para-terphenyl	475	216	
124	para-terphenyl	475	240	
125	meta-terphenyl	475	0	Gradual transition from white solid to dark brown solid, the decomposition rate is somewhat faster than for the para-terphenyl samples.
126	meta-terphenyl	475	24	
127	meta-terphenyl	475	48	
128	meta-terphenyl	475	72	
129	meta-terphenyl	475	96	
130	meta-terphenyl	475	120	

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TABLE I (Continued)

Sample No.	Compound	Temp °C	Test		Remarks	
			Duration Hours			
131	meta-terphenyl	475	144		Gradual transition from white solid to dark brown solid, the decomposition rate is somewhat faster than for the para-terphenyl samples.	
132	meta-terphenyl	475	168			
133	meta-terphenyl	475	192			
134	meta-terphenyl	475	216			
135	meta-terphenyl	475	240		Transition from clear liquid to dark brown liquid. Discoloration and viscosity increase with pyrolysis time.	
136	ortho-terphenyl	475	0			
137	ortho-terphenyl	475	24			
138	ortho-terphenyl	475	48			
139	ortho-terphenyl	475	72			
140	ortho-terphenyl	475	96			
141	ortho-terphenyl	475	120			
142	ortho-terphenyl	475	144			
143	ortho-terphenyl	475	168			
144	ortho-terphenyl	475	192			
145	ortho-terphenyl	475	216			
146	ortho-terphenyl	475	240			
150	meta-terphenyl	462	240			Transition from a white to a tan solid.
153	meta-terphenyl	462	216			
155	meta-terphenyl	462	224			
159	meta-terphenyl	462	192			
161	meta-terphenyl	462	120			
164	meta-terphenyl	462	72			
167	meta-terphenyl	462	168			
170	meta-terphenyl	462	144			
173	meta-terphenyl	462	96			
176	meta-terphenyl	462	48			
179	meta-terphenyl	462	0			
151	meta-terphenyl	462	240		Transition from a clear to a dark liquid.	
154	meta-terphenyl	462	216			
186	meta-terphenyl	462	24			
160	meta-terphenyl	462	192			
163	meta-terphenyl	462	120			
165	meta-terphenyl	462	72			
169	meta-terphenyl	462	168			
171	meta-terphenyl	462	144			
175	meta-terphenyl	462	96			
178	meta-terphenyl	462	48			
180	meta-terphenyl	462	0		Transition from a white to a light tan solid.	
149	para-terphenyl	462	240			
152	para-terphenyl	462	216			
157	para-terphenyl	462	24			



TABLE I (Continued)

Sample No.	Compound	Temp °C	Test Duration Hours	Remarks
158	para-terphenyl	462	192	Transition from a white to a light tan solid.
162	para-terphenyl	462	120	
166	para-terphenyl	462	72	
168	para-terphenyl	462	168	
172	para-terphenyl	462	144	
174	para-terphenyl	462	96	
177	para-terphenyl	462	48	
181	para-terphenyl	462	0	

was heated to only 450° C. Based on visual observation, a compound characterized as "good" might be expected to be substantially unchanged after heating for ten days at 465° C, while a compound called "poor" might be completely decomposed after four hours at 465° C.

Biphenyl, terphenyl, naphthalene, and triphenylamine appear to be the most promising compounds investigated so far. Figure 2 is a plot of vapor pressure vs temperature for the terphenyls. Naphthalene and biphenyl have unreasonably high vapor pressures above 450° C; and therefore, studies have been confined to the terphenyls, and to triphenylamine.

Twenty-two samples of each of the terphenyls were prepared for use in decomposition rate experiments. Two of each were retained as blanks and the other twenty of each were subjected to pyrolysis. Ten capsules of each of the terphenyls were heated at 475° C for periods of one to ten days. This experiment was then repeated at 462 ± 2° C. Figures 3 and 4 are pictures of the samples heated at 462 ± 2° and 475° C respectively. Comparison of the 462 ± 2° C run with the 475° C run indicates that the decomposition rate at 475° C is approximately double that at 462 ± 2° C. Meta and para-terphenyls show evidence of decomposition; but they are considerably lighter in color, and are probably less decomposed than ortho-terphenyl under equal thermal exposures. A similar

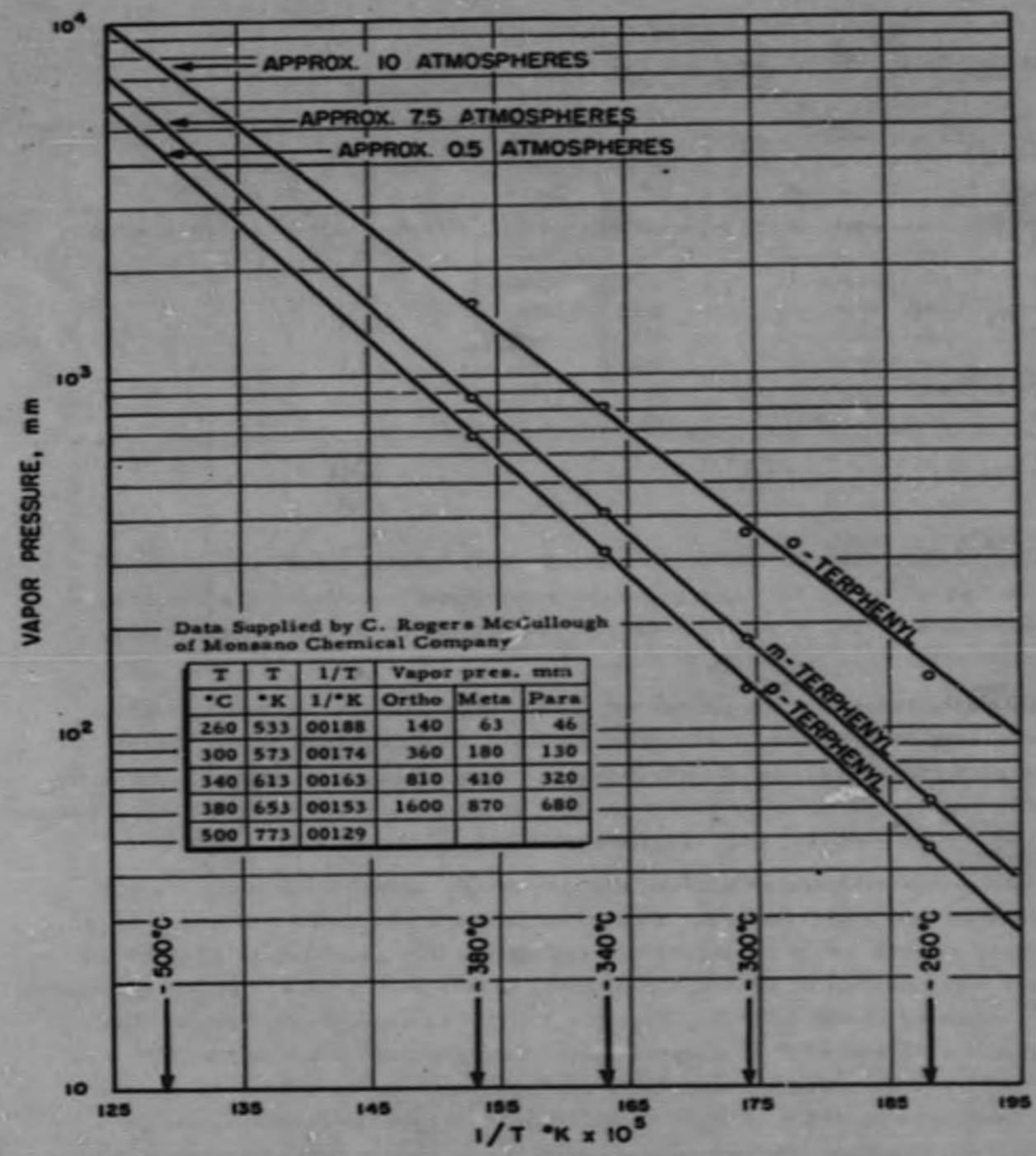


Fig. 2. Vapor Pressure vs 1/T in Degrees Kelvin for the Terphenyls

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O-TERPHENYL

M-TERPHENYL

P-TERPHENYL

TIME, DAYS

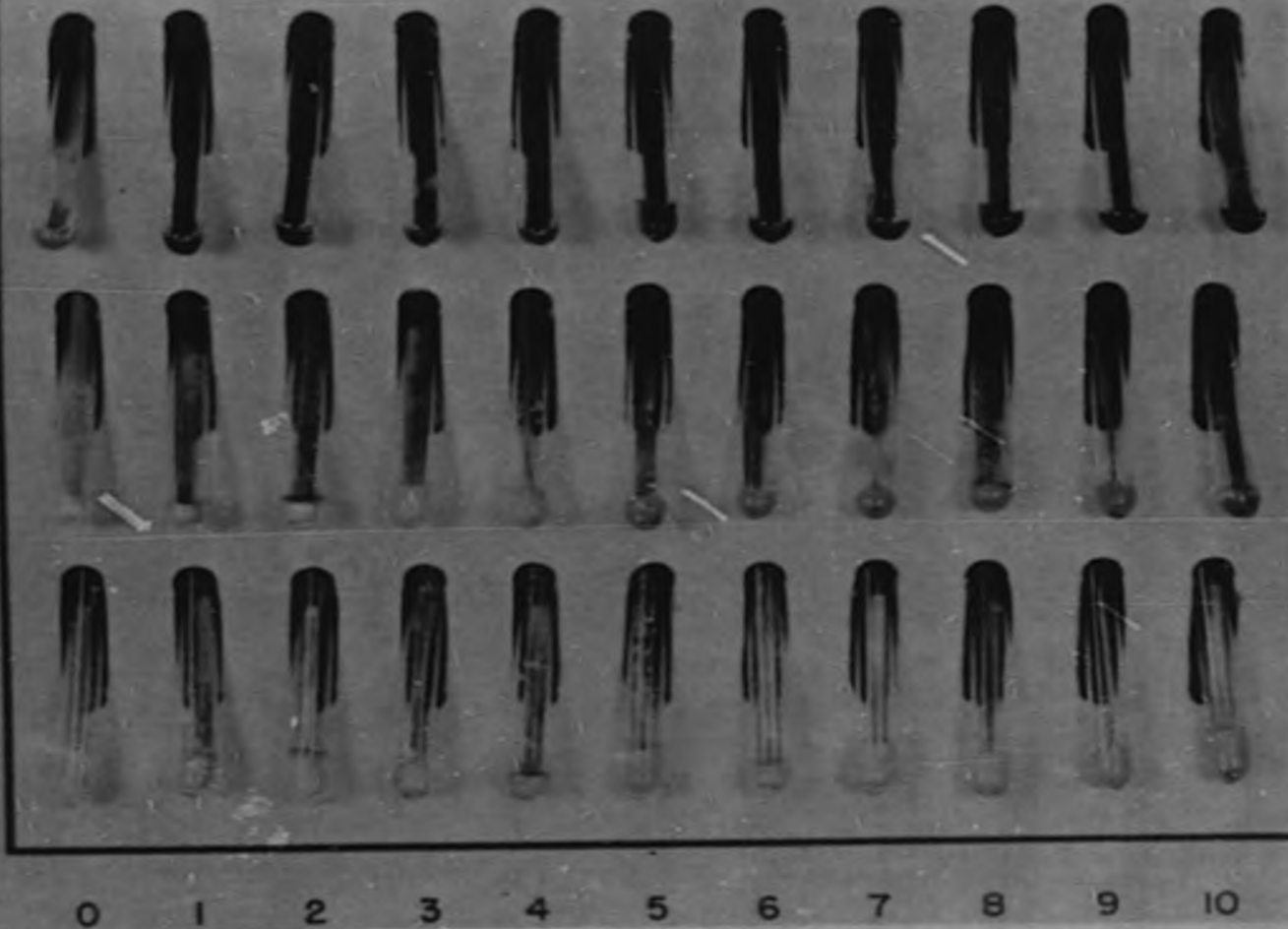


Fig. 3. Pyrolysis of Terphenyls at $462 \pm 2^\circ \text{C}$

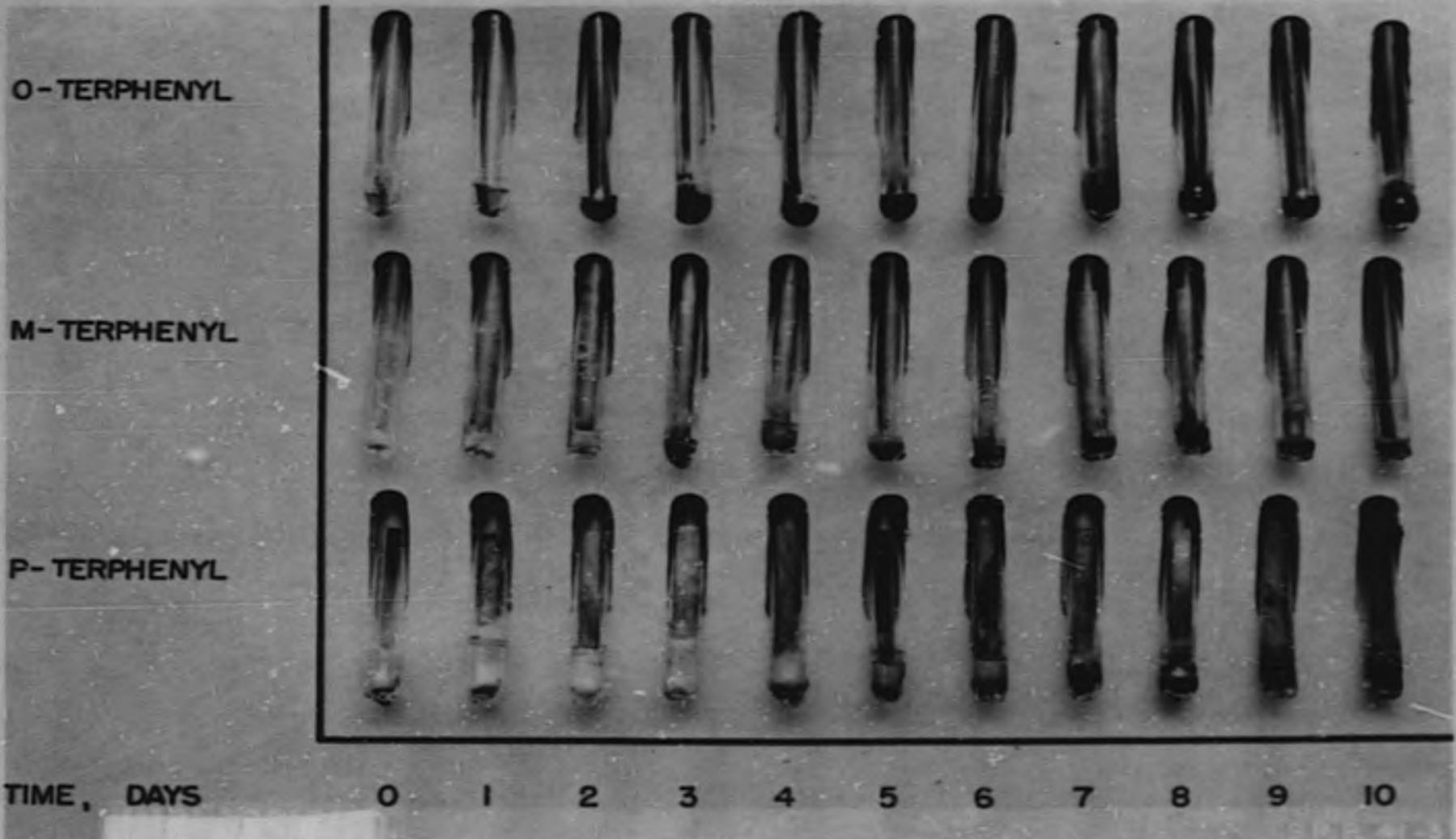
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TIME, DAYS

Fig. 4. Pyrolysis of Terphenyls at 475° C

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experiment at $462 \pm 2^\circ \text{C}$ with triphenylamine indicates that it has about the same degree of thermal stability as have the terphenyls. More detailed analyses are being made.

Several capsules have been prepared containing para-terphenyl and metals other than Type 347 stainless steel. The presence of platinum, copper, silver solder, or Type 302 stainless steel apparently did not affect the decomposition rate.

E. Thermal Loop Tests (S. Nakazato and D. J. Zigrang)

In addition to capsule experiments, boiling heat transfer loops were constructed to study the terphenyls as heat transfer media under dynamic conditions. Figure 5 shows the construction of a typical loop. Experiments to date are preliminary, leading up to the design and construction of a pumped loop. Loops were run with ortho, meta, and para-terphenyls as heat transfer media.

1. Stainless Steel Loop - Figure 6 illustrates the configuration of the loop. It was made of 1-inch (outside diameter) Type 347 stainless steel tubing. This loop had a pressure gage mounted on top of the boiler leg. An Ashcraft, 30-inch, vacuum to 30 psi pressure gage was used. The temperature of the boiler section was controlled with a Simplytrol controller.

Before the loop was charged with p-terphenyl, it was pickled with HNO_3 - HCl - HF solution, and degassed under vacuum at red heat. After loading, the p-terphenyl was melted down and then connected to a vacuum system to remove air from the loop and terphenyl.

The loops were operated with a fixed boiler temperature of 440°C . As decomposition proceeded and as the pressure due to non-condensable gases increased, the boiling point of the heat transfer media increased and heat transfer rate decreased. Eventually, the boiling point passed 440°C , and circulation of the fluid virtually ceased. This was indicated by the temperature of the liquid in the liquid return line. Figure 7 is a plot of the temperature in the liquid return line vs operating time. It should be noted that dT/dt is substantially greater for ortho-terphenyl than for meta and para-terphenyls. This agrees with the decomposition rate findings at 460 and 475°C as previously described.

The loops used for ortho- and para-terphenyls were disassembled and examined. The heat transfer surfaces were found to be unfouled, and generally

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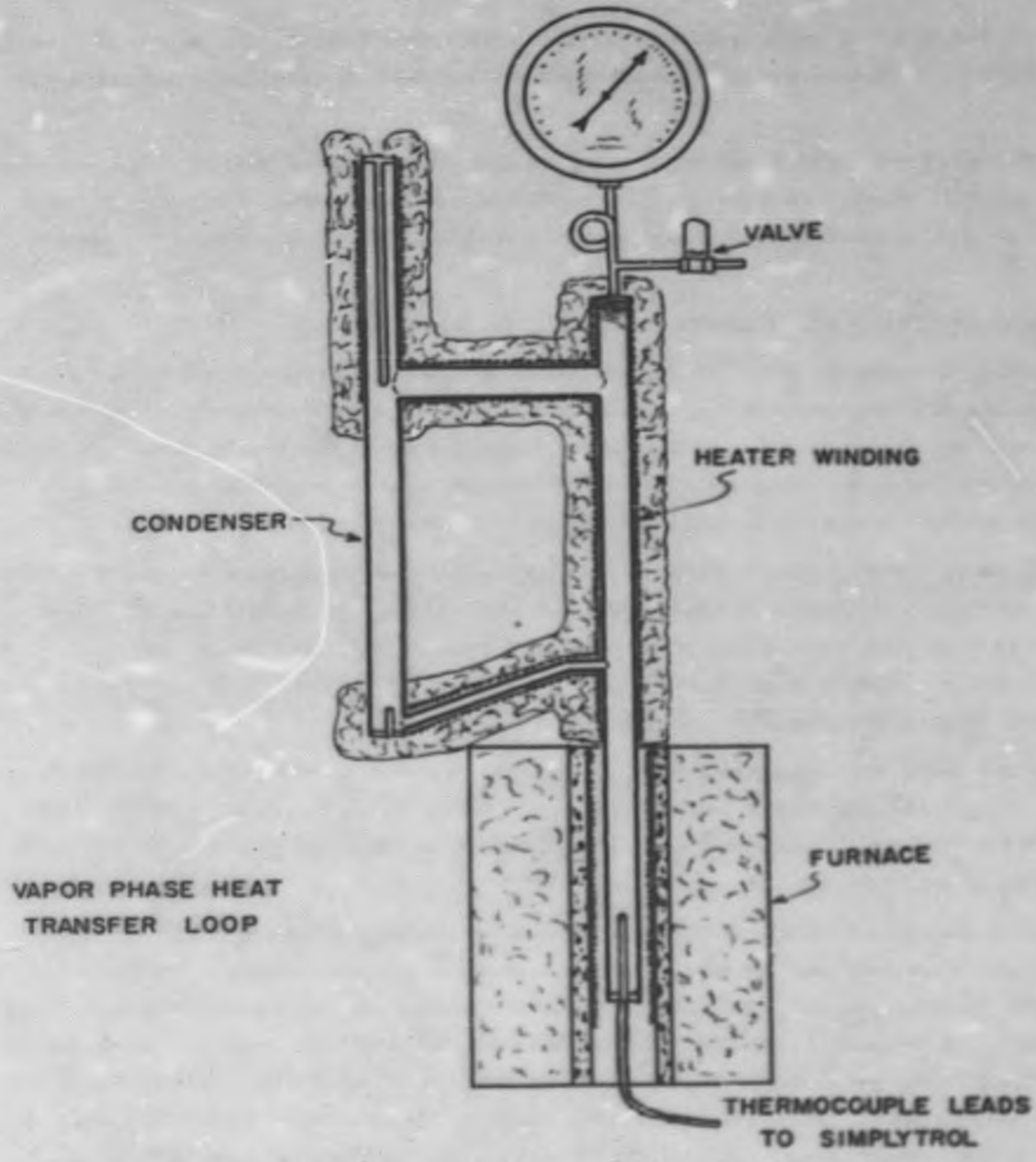


Fig. 5. Two Phase Loop

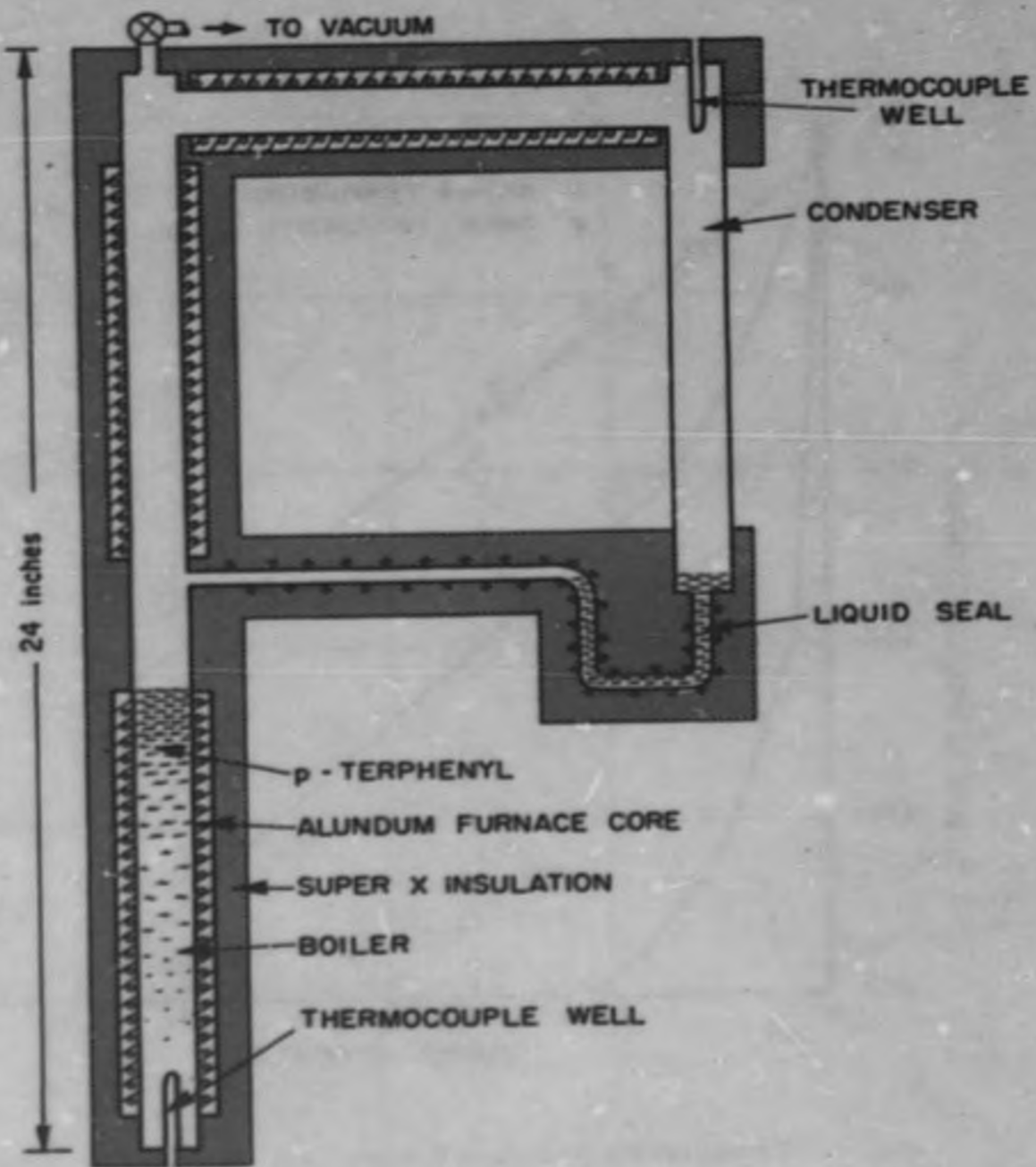


Fig. 6. Stainless Steel Loop

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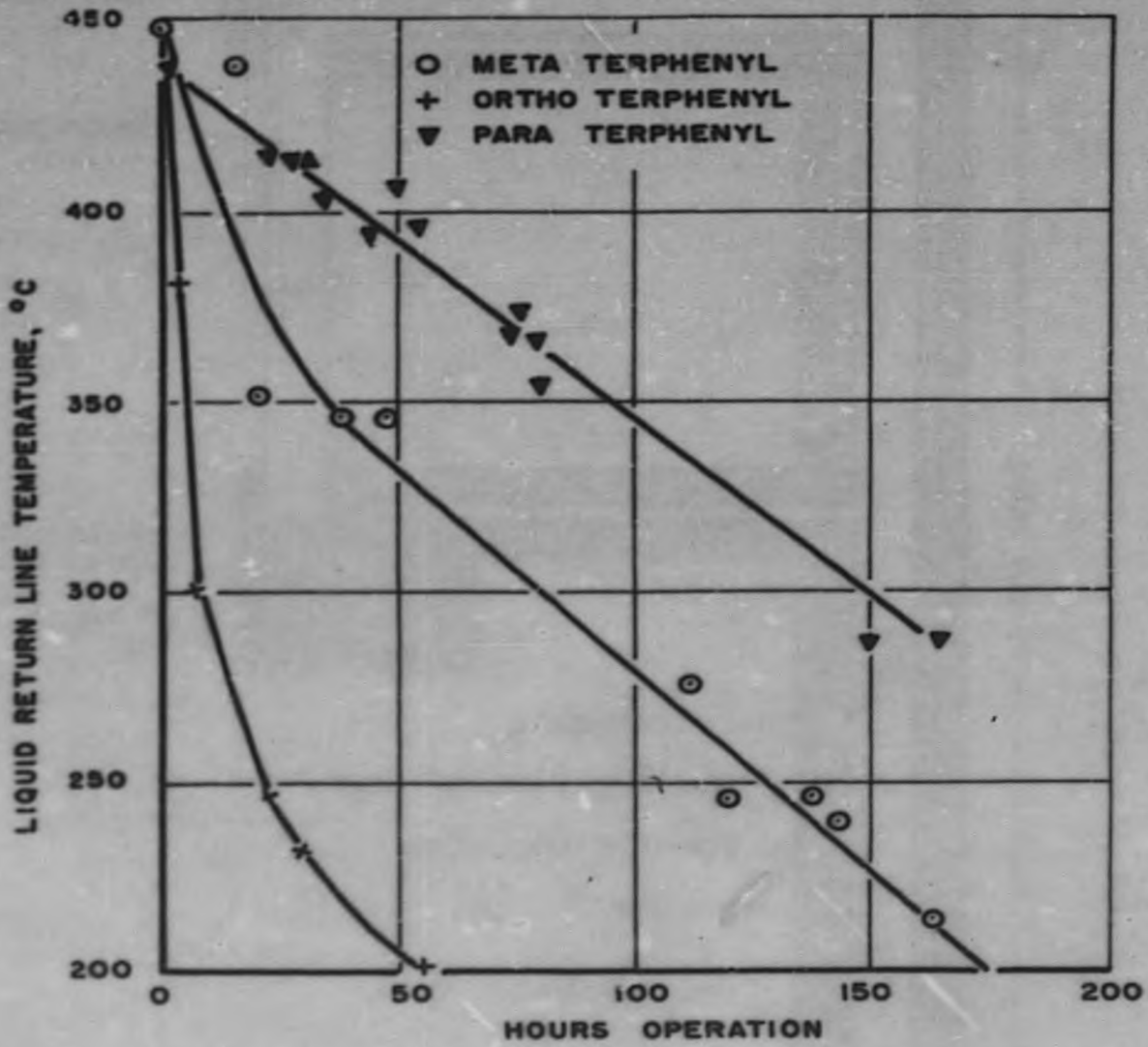


Fig. 7. Temperature in Liquid Return Line vs Operating Time

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in excellent condition. Figure 8 is a photograph of the p-terphenyl loop after it was opened at the end of 8-2/3 days at 440° C. The terphenyl was in a good condition with very little discoloration. The lower part of the boiler had a trace of black compound. The compound in the boiler section had a yellowish tint compared to the pure white compound found in the condenser section. There was no tar formation, and the inside of the loop was clean.

Three samples were analyzed with a Beckman absorption spectrophotometer; and Figure 9 shows the results of the analysis. The sample from the bottom of the boiler section, Sample A, had black specks in it, and had an absorption curve which duplicated the curve for the yellowish solid from the boiler section, Sample B. The curve for Samples A and B closely parallel the absorption curve of the original sample of p-terphenyl. The white solid from the condenser section, Sample C, gave an absorption curve similar to that of biphenyl. (M-terphenyl, also, has a similar absorption curve).

A compound was purified from the condensate by crystallization, and analyzed as biphenyl on the basis of melting point, boiling point, and the wave length at which maximum absorption occurs in the ultraviolet.

2. Vycor Loop - A loop similar to the stainless steel loop was constructed of Vycor. This loop served as a model which could be observed while in operation. It was cleaned with pickling solution, rinsed with distilled water, and degassed under vacuum at high temperature before loading. Two strips of Type 347 stainless steel were placed in the loop, one in the boiler section and the other in the condenser section. The loop was loaded with 48 grams of liquid terphenyl connected to a vacuum and then sealed. Figure 10 illustrates this loop.

The boiler temperature was maintained at 450° C by use of a Simplytrol. Circulation rate was about 14 lb/hour. The loop was run about 28 hours at this temperature, and the test was terminated by the rupture of the loop. Before this occurred, the condensate was a clear liquid, flowing rapidly and indicating a low viscosity fluid. Apparently the decomposition products increased the total pressure within the loop, causing the rupture. The largest Vycor tubing used in this loop was 19 millimeters outside diameter by 1.2 millimeters wall thickness.

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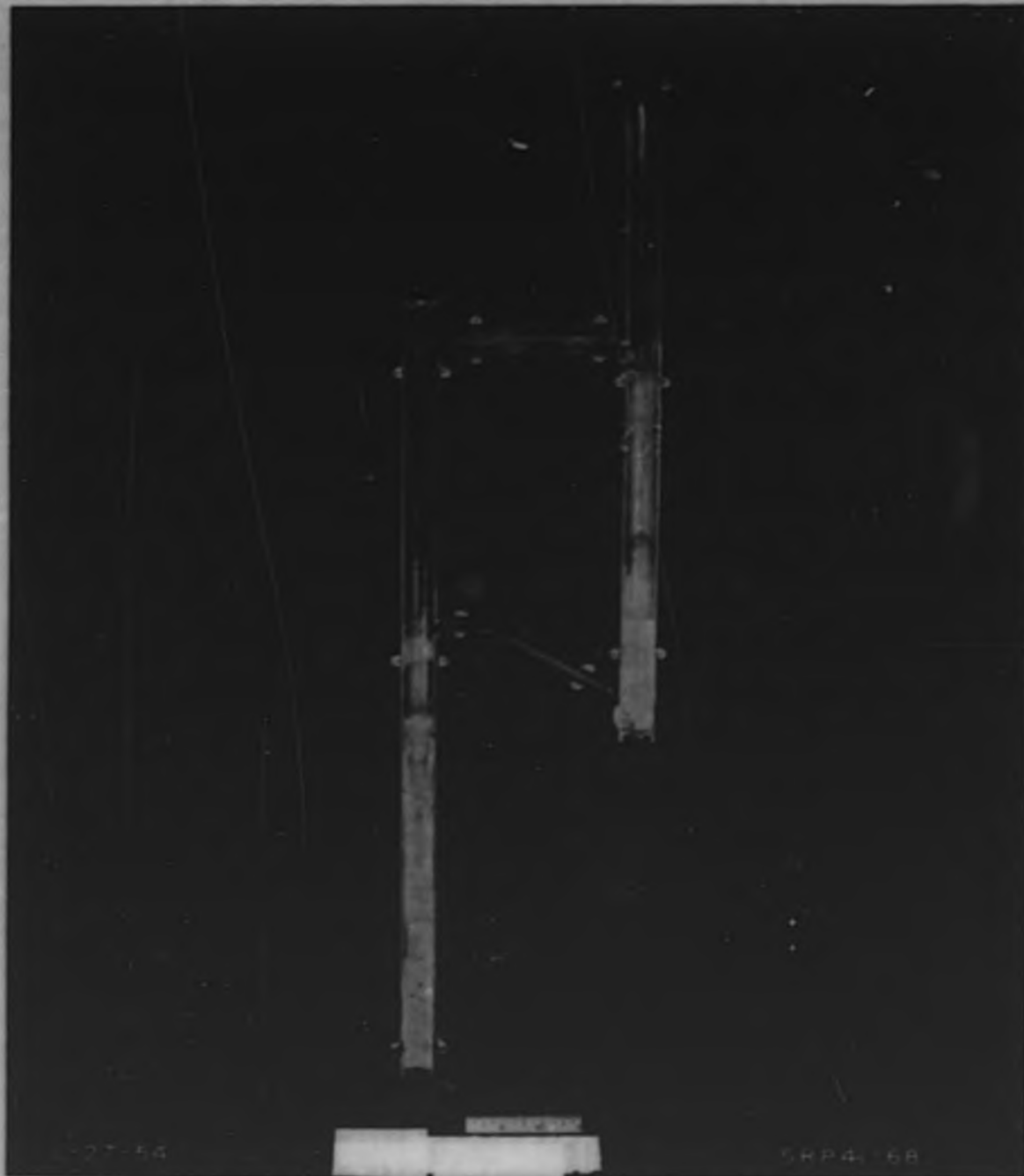


Fig. 8. The p-Terphenyl Loop after 8 2/3 days at 440° C

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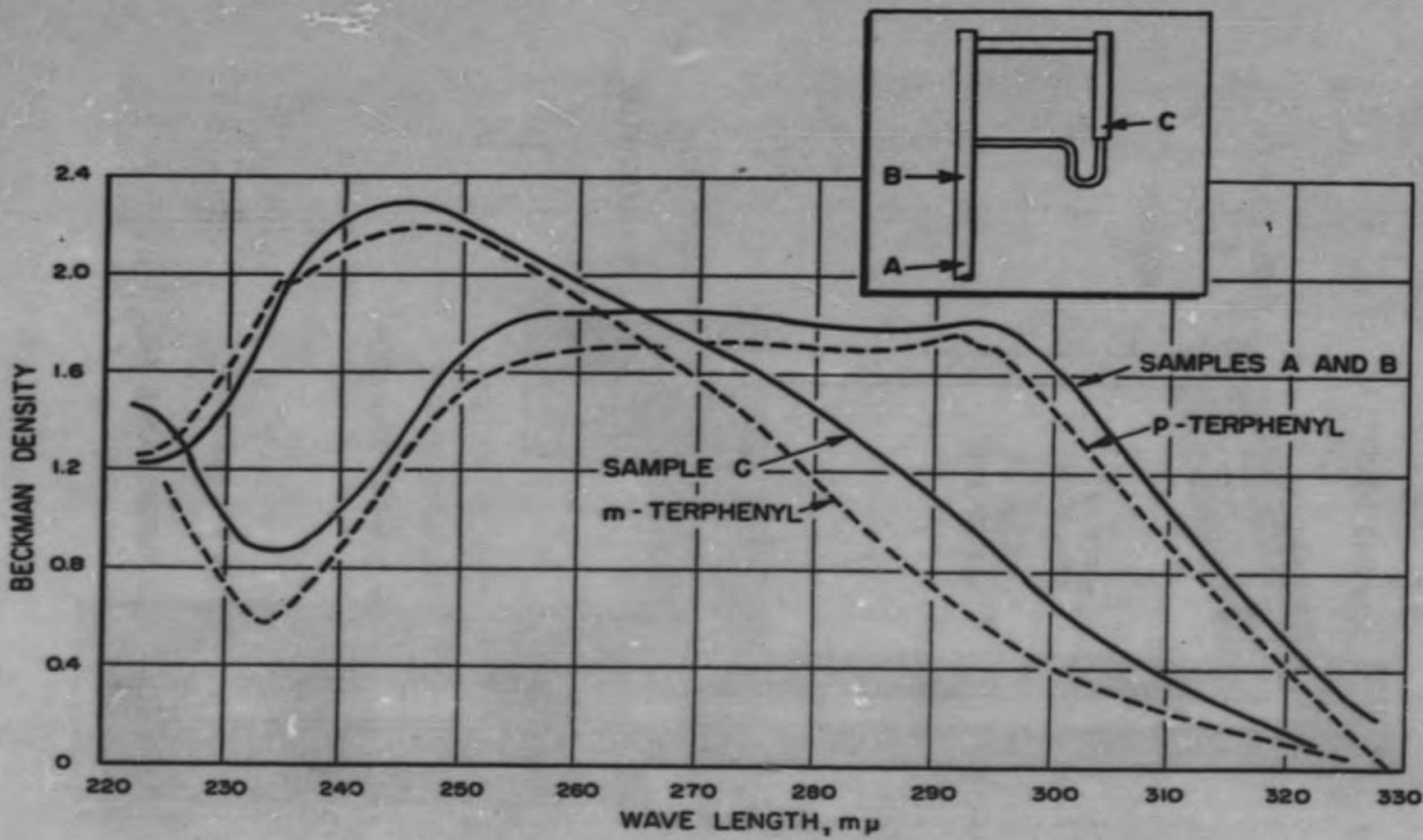


Fig. 9. Spectrophotometric Analysis of Samples from Stainless Steel Loop

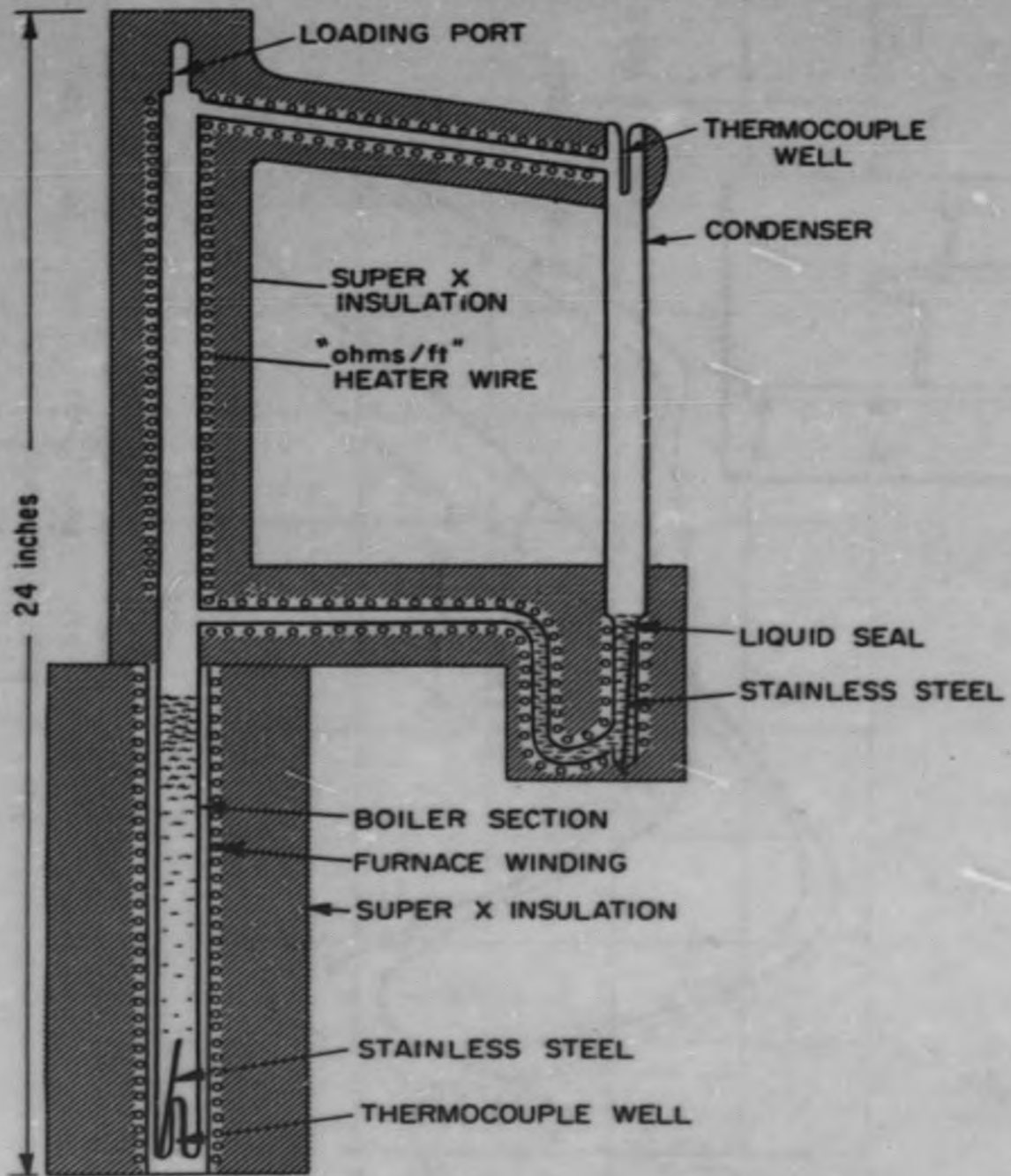


Fig. 10. Vycor Loop



F. Pump Loop Experiments (S. Nakazato and D. J. Zigrang)

A pump loop has been designed to study the heat transfer characteristics of p-terphenyl under dynamic conditions. Figure 11 is the flowsheet of this loop. The heater portion will be constructed so that the heat transfer coefficients can be measured to detect the change in fouling factor. It is planned to make initial runs without the purification system shown on the flowsheet. The purification system will be added to the system for later runs.

Figure 12 shows the equilibrium diagram for the biphenyl--para-terphenyl system. It can be seen from this diagram that the separation of these two compounds can be accomplished by distillation. Although the system to be purified will be a mixture of many decomposition products, the design was made considering it as a binary system. The components lighter than biphenyl will probably be non-condensable at this temperature, while the components heavier than terphenyl will remain in the still pot in a small concentration.

1. In-pile Pump Loop Discussions - On July 9, 1953, an informal discussion was held at the MTR site to write the specifications for in-pile and in-canal tests of organic coolants. Personnel of the Phillips Petroleum Company will make the final design and construct the test equipment. The following specifications were outlined:

Test Materials: para-terphenyl, ortho-terphenyl, meta-terphenyl, biphenyl, naphthalene, biphenyl-naphthalene eutectic, other organics tested by California Research Corporation, North American Aviation, Inc.

Temperature: 500-900° F

Pump: 2-5 gallons/min, 100 psi discharge pressure, 500 psi working pressure

Flux: 3×10^{14} n/cm² sec thermal
 5×10^{13} n/cm² sec fast

Flow Velocity: 1 to 20 ft/sec

Test Hole: HB-3 or HB-1 (MTR)

Capacity: less than 10 pounds of material

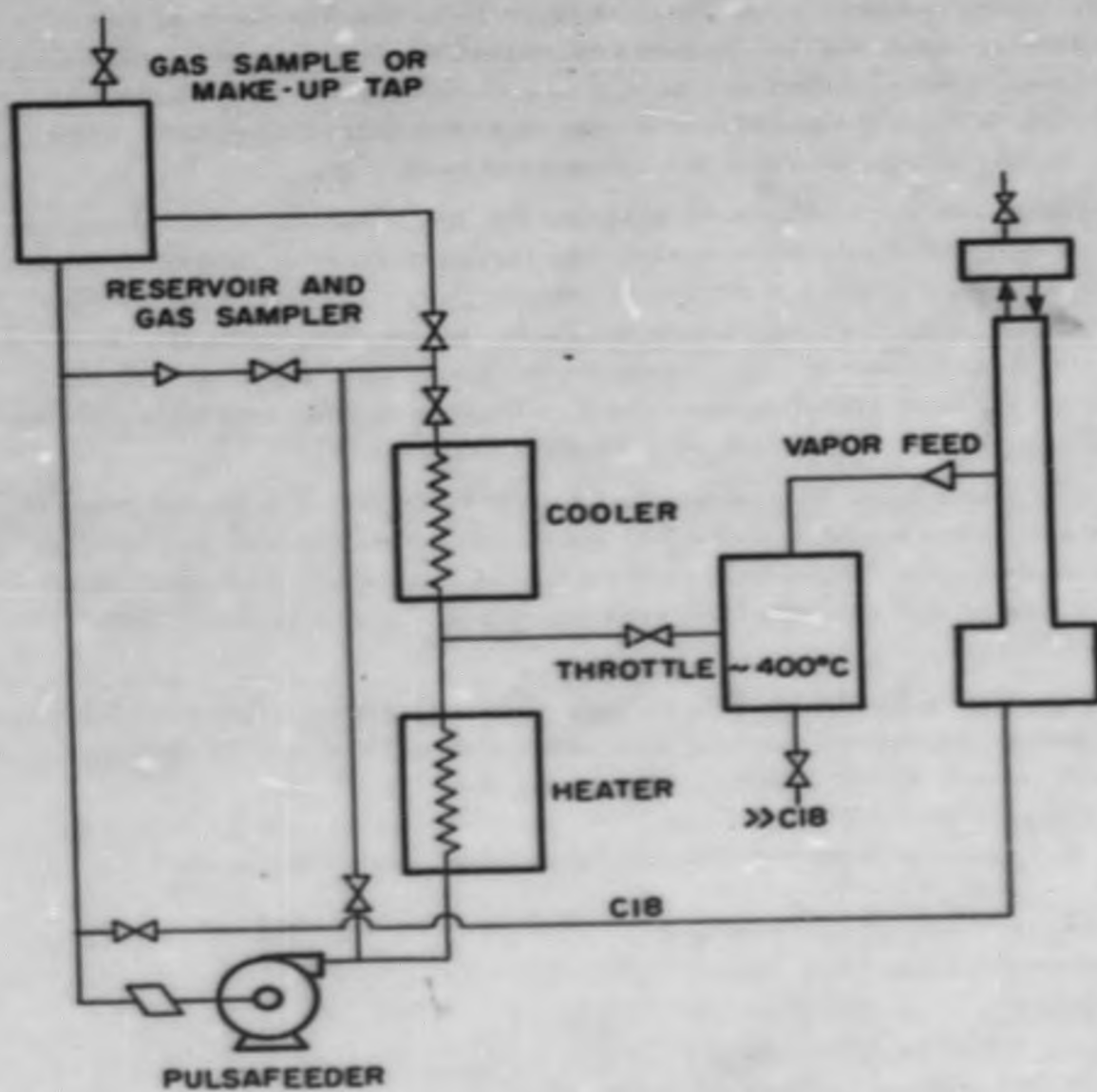


Fig. 11. Terphenyl Pump Loop with Purifier

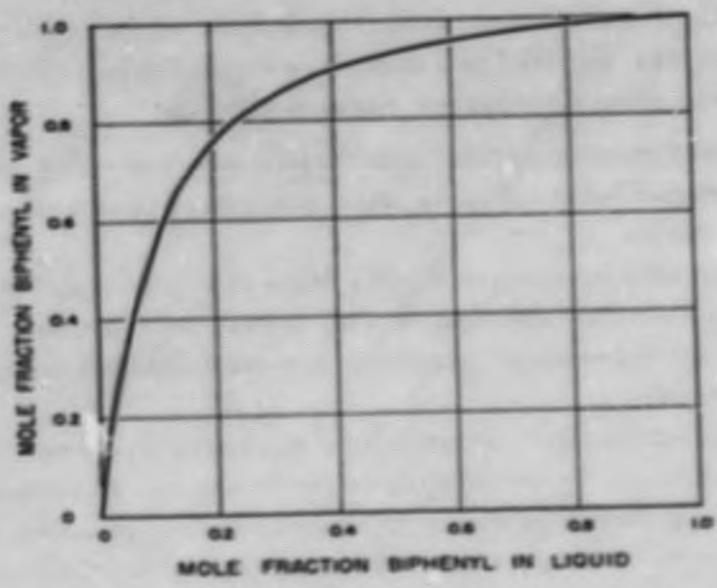


Fig. 12. Mole Fraction Biphenyl in Liquid

The loop will be fabricated from 3/8-inch (outer diameter) tubing of Type 347, or other 18-8 stainless steel. The volume holdup in the irradiation zone will be increased by using a coil. The ratio, volume irradiated per total volume = 1/2, desired by North American Aviation, Inc. cannot be attained for the pile experiment, because of the short attenuation length of the neutron flux which limits the coil height to 6 - 8 inches.

G. Organic Materials Conference

A conference on the use of organic materials for coolants and moderators in nuclear reactors, sponsored by the Engineering Division of the AEC, was held on August 5-6, 1953 at the Atomic Energy Department, North American Aviation, Inc., Downey, California.

The conference, organized by Dr. Harry Pearlman of North American Aviation Inc., was divided into three sessions covering the thermal stability of organic compounds, radiation damage in organic compounds, and the engineering analysis of organic compounds for reactor applications. The complete proceedings of this conference appear in NAA-SR-280.⁵



Many of the results and conclusions reported at the conference were of a tentative nature, and are subject to further confirmation or rejection. As reported by various authors, these tentative results are:

1. The simplest polyaromatic structures are the most thermally stable: biphenyl seems best for long thermal exposures and naphthalene for short exposures.
2. The polyphenyls are more stable than the condensed ring aromatics.
3. Thermal instability increases with molecular weight.
4. The use of additives and structural modifications have not given improved stability.
5. There is an inhibition period in the thermal decomposition of 1 to 3 days.
6. Radiation damage is a function of total energy absorbed.
7. Due to their greater ionizing characteristics, gamma rays and fast neutrons cause far more radiation damage than thermal neutrons.
8. Radiation damage appears to be proportional to the amount of ionization produced by the secondary radiation from fast neutron and gamma rays.
9. The presence of oxygen appears to accelerate the radiation damage.
10. A meta-terphenyl moderated reactor has been estimated to produce electric power at about 11.5 mils/kwh.
11. The make-up and purification costs for diphenyl in a diphenyl cooled reactor are estimated at 0.53 mils/kwh.

The results of the conference indicated that there was still much work to be done. The required data includes:

- A. Data on radiation and pyrolysis, as follows:
 1. The effect of additives,
 2. The effect of structure modification,
 3. New compounds,
 4. Eutectic mixtures,
 5. All compounds at lower temperatures and for longer periods of time to evaluate the apparent inhibition period, and
 6. The effect of various types of radiation such as gamma, neutron, electron, and so forth.
- B. Engineering data on the following:
 1. Heat transfer,



2. Viscosity changes,
3. Fouling factors,
4. Purification cycles,
5. Make-up requirements, and
6. Cost analyses.

III. ZIRCONIUM HYDRIDE IRRADIATION STUDIES

(W. B. Goedel and P. T. Gilbert)

Zirconium hydride has several properties which make it a potentially good neutron moderator for high temperature reactors. These are:

1. High hydrogen density (Up to 240 cc of hydrogen can be absorbed by 1 gram of zirconium. The calculated atomic density of hydrogen in zirconium is about 6.8×10^{22} atoms per cm^3 at 1200°F . Water contains 6.2×10^{22} hydrogen atoms per cm^3 at 300°F .),
2. Low thermal neutron absorption cross section ($.04 \times 10^{-24}$ cm), and
3. Satisfactory thermal stability (The equilibrium thermal dissociation pressure of $\text{ZrH}_{1.75}$ at 600°C is about one atmosphere).

However, few data are available on the effect of radiation on zirconium hydride. The present work was undertaken to obtain more complete information.

A Type 321 stainless steel irradiation cell, as shown in Fig. 13, was used to contain the zirconium hydride sample. This cell was mounted on the Van de Graaff generator for irradiation with 1 Mev electrons, the electron beam entering the cell through a 1.25 mil aluminum window. Pressure was measured by means of a Statham absolute pressure transducer. A vacuum valve permitted evacuation of the system prior to heating.

The procedure involved heating the sample to a given temperature, and holding this temperature constant until an equilibrium pressure was obtained. This was done for several increasing temperatures, and then repeated on cooling. The whole procedure was then repeated with irradiation. By following this scheme, it was possible to compare directly the pressure vs temperature curves obtained with and without irradiation.

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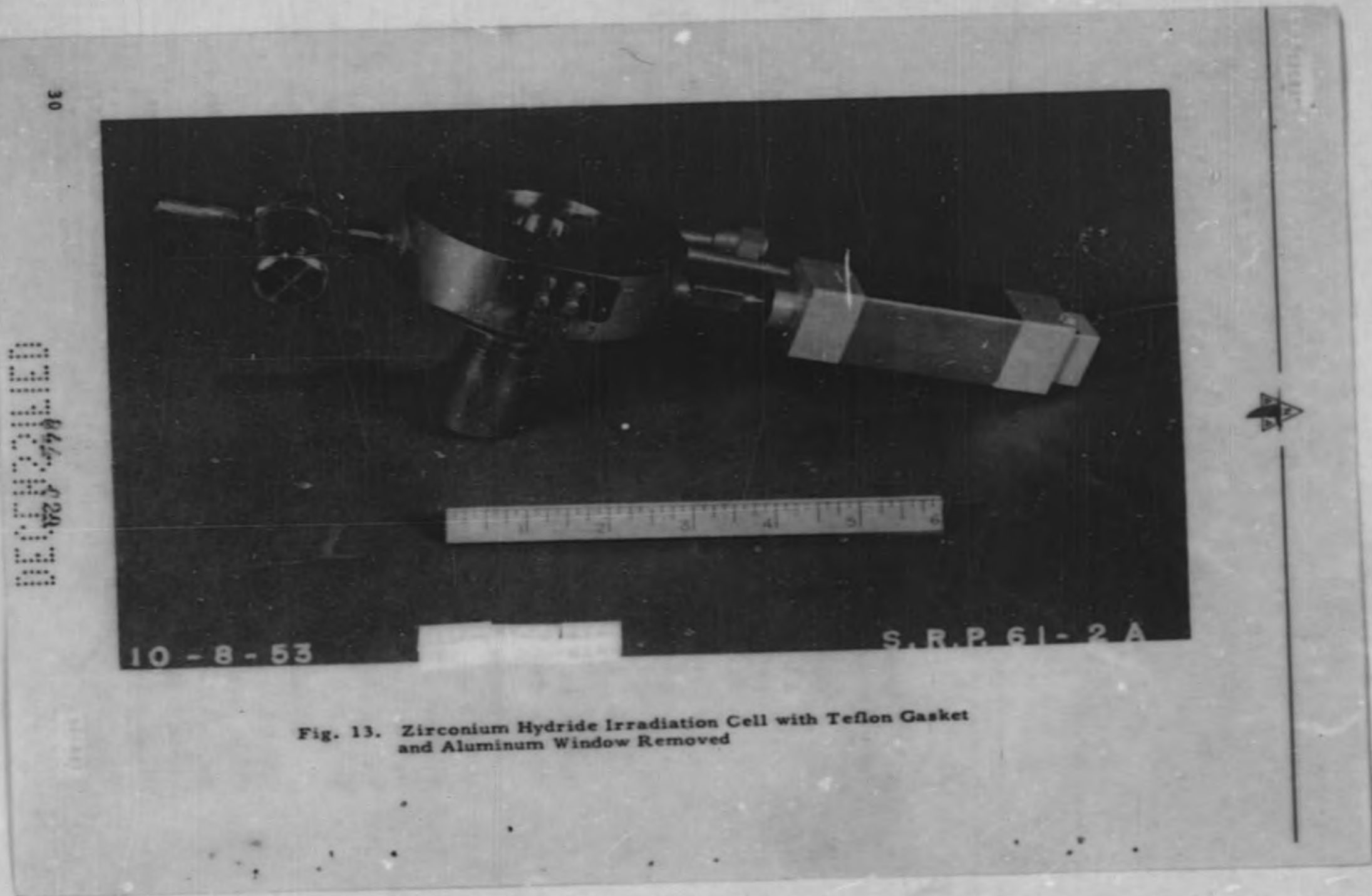


Fig. 13. Zirconium Hydride Irradiation Cell with Teflon Gasket and Aluminum Window Removed



In another type of run, the zirconium hydride was held at constant temperature under irradiation for several hours to determine if there was any change in the decomposition pressure with long term irradiation.

IV. DEVELOPMENT OF ANALYTICAL METHODS

Development is in progress on methods for plating PbO_2 on palladium, the determination of boron in uranium, the determination of uranium in pure bismuth, the determination of uranium in thorium, the conversion of $UO_2(NO_3)_2$ to UO_2SO_4 , the determination of O_2 in gases, the determination of the thermal and radiation decomposition products of terphenyls, and the determination of iron in zirconium.

A. Ion Exchange Method for Purification of D_2O

For use in exponential experiments, a 600 gallon batch of D_2O was contaminated with boric oxide at a concentration of 150 milligrams of B_2O_3 per liter. After completion of the experiments, the boron was removed by ion-exchange, using Rohm and Haas Amberlite resins IRA-400 and 410. The commercial resins were converted to the hydroxide form, and then deuterated by upflow of D_2O , prior to processing the borated solution. Final concentration of boron in the D_2O was less than 0.01 milligrams of boron per liter. A report on this method will be published shortly.

B. Determination of Terphenyl-Biphenyl Mixtures by Dielectric Constant and Refractive Index Measurements

Methods have been developed for determining the amount of biphenyl in terphenyl, using dielectric constant and refractive index measurements. Dielectric constant measurements were made with a Q-meter (type 160-A) at a frequency of 55 KC. For measurements at 80 to 100° C, a 100 micromicrofarad radio condenser was used; and at 220° C, a condenser made of concentric

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copper cylinders (3/8 and 5/8 inch outside diameters) was used. Accuracies of one to two per cent were obtained.

C. Articles Published and Reports Issued

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V. MISCELLANEOUS

A. Reactions of Water with Rapidly Heated Metals

Wires and foils of several metals used in reactor construction were dispersed under water by passing large currents through them from a condenser discharge. The metals were: 2S aluminum, aluminum-uranium alloy, uranium, and zirconium. Substantial amounts--up to 100%--of the metal samples reacted with water under the experimental conditions. From the short reaction times and the gas pressure developed (analyzed as hydrogen), it was concluded that explosive chemical reactions occurred. Details of this work appear in NAA-SR-197.⁷

PLATE
B. TOP

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7. Ruebsaman, W. C., F. J. Shon, and J. B. Chrisney, "Chemical reaction between Water and Rapidly Heated Metals," NAA-SR-197, October 27, 1952.

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