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TECHNICAL ACTIVITIES REPORT--JULY 1952

GRAPHITE DEVELOPMENT - PILE GRAPHITE

By

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GRAPHITE DEVELOPMENT - PILE GRAPHITE

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SUMMARY AND DISCUSSION

Physical data are presented for transverse CSF samples with capsule exposures of 568, 1049 and 1617 MD/CT. The higher exposures indicate a sharper damage gradient toward the front of the pile. Additional casings of various types of graphite were loaded into test holes during this month.

Average values of the thermal conductivity and electrical resistivity for several types of virgin graphites are presented. Data of this nature will be a regular portion of this report henceforth.

Process tube channel 2677-H was mined and traversed for bore diameter. Although several of the tube block junctions were obscured, the channel was quite uniform. Examination of all previously mined graphite powder samples for aluminum oxide corrosion product has been completed and the results are reported.

Study of damaged graphite specimens subjected to various thermal annealing temperatures has been initiated. It is planned to investigate the effect of the annealing temperatures well above 1000°C.

X-ray diffraction studies have been completed on several specimens of graphite subjected to chemical oxidation. A diffraction peak corresponding to a lattice spacing of approximately 12 angstroms appears with the formation of graphitic oxide. With increasing oxidation, intensity of the graphite 002 reflection decreases but does not shift in magnitude. This feature of the formation of graphitic oxide is unlike the formation of irradiation damage, and within the limits of this experiment. It appears that the radiation damage phenomena cannot be likened to chemical attack.

The reaction kinetics at constant volume for the system graphite - carbon dioxide have been studied at 962°C. The data from these runs fit the previously determined rate law for carbon monoxide concentration between 9 and 30 per cent. The extent of surface oxide formation seems essentially the same at this temperature as it was at 905°C.

Experimental difficulties have delayed the study of the reaction kinetics at constant pressure of the system graphite-carbon-dioxide-helium.

Samples of gases and graphite in sealed quartz containers are being prepared to replace those which will be discharged from tube 0776-H in August. This second charge will consist of six tubes of pure carbon monoxide, five tubes of pure carbon dioxide plus weighed samples of powdered GSF graphite and five containing weighed solid samples of GSF graphite in vacuum.

Results of mass spectrometric analysis of the gases evolved from a sample of irradiated GSF graphite and a sample of interlamellar graphitic oxide are presented without comment.

Texas coke graphite samples have been removed after capsule exposure in a process tube. These samples, which were cut from an electrode 6 inches in diameter, indicate that the expansion of transverse samples in the pile is comparable to other pile

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graphites. Parallel specimens from this exposure also expanded. This behavior is unique and additional information will be available from TS samples cut from square extruded bars and should offer a check on these measurements.

A series of graphite specimens produced by Battelle Memorial Institute has been received. Of these, two lots were graphites with exceptionally low thermal conductivity. The irradiation stability of these specimens will be obtained as rapidly as possible. There appears to be some discrete advantages associated with the use of carbon moderators having low initial thermal conductivity and associated poorly defined crystal structure.

Exposure of graphite samples in tube 1684-B continues at essentially constant temperatures.

Seven additional runs have been completed on the gas conductivity apparatus to explore the precision of the method and establish systematic behavior. The effect of convection is being investigated, using air as the conducting gas. Once these variables have been determined, the thermal conductivity of gases at 0°C will be investigated.

Special graphite exposures in the MTR at Arco have been approved and samples are being prepared for shipment. Standard CSF graphite samples are being loaded in aluminum containers and will be exposed in two locations in the MTR. These experiments are designed to explore the effect of various neutron energy spectra on irradiation damage to graphite. The specific flux at the MTR will be greater than the Hanford flux and these exposures may prove to be valuable in predicting future Hanford pile graphite conditions.

A study has been initiated to examine the surface characteristics of the stringer of graphite removed from the C test hole at the B Pile in December, 1950. In addition to this, additional controlled specimens of virgin graphite have been measured for surface characteristics after thorough outgassing and have been sealed in quartz tubes for pile irradiation. Simultaneously, quartz tubes containing graphite in atmospheres of oxygen and carbon monoxide will be exposed to pile irradiation, and surface characteristics measured after discharge.

Virgin and damaged graphite specimens were removed from the pile basin after receiving two months' exposure to intense gamma irradiation adjacent to discharged uranium slugs. No detectable changes in the properties of these specimens were noted. A second exposure is in progress to run for a six-month duration.

The National Bureau of Standards has examined the heat of combustion of several samples of KC, CSF, and CSGHF graphite. The weighted mean of the new measurements is 7833.1 ± 1.6 cal/gram. This new value is 6 cal/gram less than the previous standard and will be adopted for the future total stored energy calculations.

Completion of project C-410, the facility for the in-pile controlled atmosphere experiment, continues to be delayed by mechanical difficulties. Evaluation of the gamma activity at a distance of one foot from the heater-cooler assembly twelve hours after pile shut-down gives a value of 650 r/hr.

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PHYSICAL PROPERTIES STUDIES

Process Tube Exposures - PT-105-269-P Supplement B - D. H. Curtiss

A series of transverse samples of CSF graphite canned in four inch capsule carriers have been irradiated in selected positions along the length of a uranium loaded process tube under Production Test 105-269-P Supplement B. (1) The data from the initial exposure were previously reported in Technical Activities Report, June, 1951. (2) Physical measurements of the samples for the initial exposure (568 MD/CT capsule) of the carriers placed in slug positions 4, 12, 23, 42, 53, and 61 of a normal 64 slug process tube indicated no damage dependence on exposure temperature as evidenced by physical length and crystal expansion. Of the three samples initially charged in each capsule carrier, only one was recharged in the above slug positions and each previously irradiated sample placed in its original position. These samples were discharged after an additional exposure of 1049 MD/CT. Measurements of physical length expansion and crystal expansion now indicate a definite temperature coefficient of damage.

The physical length expansion data are shown graphically in figure 1. Length measurements on three of the six high exposure samples were not obtained because of mechanical difficulties. Calculations of the rate of physical length expansion corrected for flux distribution indicate that the expansion of samples in position 61 and 53 (near front face) is equivalent to cold test hole expansion. The crystal expansion data are shown in figure 2. These data also exhibit a sharper gradient and more damage for samples near the front face. This is evidence of the effect of the cooler inlet water as compared to the warmer effluent water.

The rates of damage for the various positions along the process tube for physical length and crystal expansions is somewhat complicated by the non-linear relations of these two properties with exposure. However, the study of these rates and their interpretation is continuing.

A summary of physical length and crystal expansion data and in addition, thermal conductivity and electrical resistivity ratios are shown in Table I.

- (1) HW-17964, "Production Test 105-269-P Supplement B, Special Exposure of Graphite", D. H. Curtiss
- (2) HW-21562, "Technical Activities Report, June 1951", L. P. Bupp

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Figure 1

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X - RAY CRYSTAL EXPANSION
PRODUCTION TEST -105-269-P- SUPPL.-B

Figure 2

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TABLE I

SUMMARY OF PHYSICAL PROPERTIES

Irradiated Transverse CSF Graphite
PT-105-269-P Supplement B

Position Number	4	12	23	42	53	61
Distance Front Face	31'5"	28'3"	24'7"	17'9"	14'1"	11'0"
Distance from C _L of Pile	10'3"	7'2"	3'6"	3'4"	7'0"	10'2"
<u>Physical Expansion in %</u>						
Exposure 568	.17	.35	.42	.49	.34	.17
1049	.28	.49	.73	.88	.70	.37
1617		.78	.97		1.12	
<u>Crystal Expansion - C₀ - A</u>						
*Exposure 568 MD/CT	6.83	6.94	7.02	7.06	6.95	6.82
1049 MD/CT	6.86	7.02	7.16	7.24	7.19	6.95
1617 MD/CT	6.97	7.13	7.36	7.45	7.42	7.08
<u>Thermal Conductivity K₀/K</u>						
*Exposure 568 MD/CT	13.3	23.3	20.5	23.8	18.3	14.0
1049 MD/CT	18.1	28.4	28.9	31.6	30.6	24.7
1617 MD/CT	21.7	29.1	27.4	30.7	25.5	19.2
<u>Electrical Resistivity ρ_{θ}</u>						
*Exposure 568 MD/CT	3.75	3.59	3.57	3.80	3.84	3.88
1049 MD/CT	3.32	3.31	3.25	3.41	3.56	3.66
1617 MD/CT	3.46	3.48	3.55	3.82	3.77	3.81

*Exposures are calculated as capsule exposures.

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Test Hole Exposures: General — D. H. Curtiss

A number of casings of various types of graphite were loaded into test holes during this month. A memorandum is being prepared that will summarize the existing loadings and expected date of discharge.

Virgin Graphite — E. M. Carr

As a portion of the graphite monitoring and graphite experimental programs, a total of 209 virgin graphite samples of various types were measured this month for initial thermal conductivity (K_0), and initial electrical resistivity (ρ_0).

The various types of graphite on which the above measurements were made include the following:

1. Bars #1 and #2 of batch #N11 obtained from the National Carbon Company. This is a Texas Company coke, standard pitch graphite, impregnated, graphitized and purified at 2500° C.
2. Bar #2 of batch #N6 obtained from the National Carbon Company. The specifications are the same as those for batch #N11, with the exception that batch #N6 is non-impregnated.
3. Battelle Experimental Graphite
 - a. Lot #E34, consisting of a Standard Oil Company #4 Korite petroleum asphalt binder and a Standard Oil Company "B" coke filler, graphitized at 2500° C.
 - b. Lot #E36, consisting of the same binder as in Lot #E34, with a filler of coke made at Battelle from binder asphalt, and graphitized at 2500° C.
 - c. Lot #E39, consisting of a Bakelite Corporation BU 1600 phenol-formaldehyde resin binder and a sugar carbon filler, graphitized at 2500° C.
 - d. Lot #E42, consisting of a Barrett Company coal tar pitch (145° F. M. P.) binder and a filler of coke made at Battelle from binder pitch, and graphitized at 2500° C.
 - e. Lot #E46, consisting of an Ironsides Company 79 IHS-CP phenol-formaldehyde resin binder, a filler of coke made from binder resin, and graphitized at 2500° C.
4. Bar #104, a stock Kendall coke, Chicago pitch graphite, graphitized at 2800° C.

The tabulated results of the thermal conductivity and electrical resistivity measurements are presented in Table II.

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TABLE II

Thermal Conductivity and Electrical Resistivity of Virgin Graphite

<u>Source of Sample</u>		<u>Orientation to Extrusion Axis</u>	<u># Samples Run</u>	<u>Average K_0 cal/cm.sec.deg.C.</u>	<u>Average ρ_0 ohm - cm</u>
N 11	Bar # 1	Transverse	15	0.165 $\pm$ 0.010	18.61 $\pm$ 1.33
N 11	Bar # 2	Transverse	42	0.215 $\pm$ 0.013	14.38 $\pm$ 0.68
N 11	Bar # 1	Parallel	6	0.197 $\pm$ 0.007	14.53 $\pm$ 0.48
N 6	Bar # 2	Transverse	45	0.160 $\pm$ 0.008	20.03 $\pm$ 0.89
N 6	Bar # 2	Parallel	15	0.221 $\pm$ 0.014	13.53 $\pm$ 0.65
E 39		Parallel	1	0.0314	54.31
E 34		Parallel	6	0.0531 $\pm$ 0.0019	42.68 $\pm$ 2.29
E 36		Parallel	6	0.0186 $\pm$ 0.0009	67.13 $\pm$ 0.77
E 42		Transverse	15	0.0760 $\pm$ 0.0088	36.89 $\pm$ 1.84
E 42		Parallel	7	No average taken	
E 46		Parallel	6	0.0203 $\pm$ 0.0013	69.04 $\pm$ 2.37
# 104		Transverse	23	0.237 $\pm$ 0.013	11.36 $\pm$ 0.66
# 104		Parallel	22	0.400 $\pm$ 0.021	5.49 $\pm$ 0.19

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Physical Property Correlations -- W. C. Riley

The empirical relationship, (3) $E = .19\alpha \times 10^6 - .12$ where E = initial rate of expansion in a cooled test facility in %/1000 MD/CT and α = average coefficient of thermal expansion from room temperature to 700°C in units of per °C, has been reexamined with respect to TS graphite. Expansion rates for TS graphite cut from an extruded six inch cylinder as determined on irradiation in a water cooled process tube are as follows:

$$\begin{aligned} E \text{ transverse} &= .60\%/1000 \text{ MD/CT} \\ E \text{ parallel} &= .25\%/1000 \text{ MD/CT} \end{aligned}$$

Average thermal expansion of TS graphite from room temperature to 700°C (4) has been determined as follows:

$$\begin{aligned} \alpha \text{ transverse} &= 4.0 \times 10^{-6} \text{ per } ^\circ\text{C} \\ \alpha \text{ parallel} &= 2.1 \times 10^{-6} \text{ per } ^\circ\text{C} \end{aligned}$$

The deviation from the experimental physical expansion values found by using thermal expansion values and the empirical relationship is less than 7% for the transverse value and about 12% for the parallel value. The deviation of 7% represents the largest deviation yet found on transverse material. This is the first graphite measured that exhibits an irradiation damage expansion in the parallel direction that can be approximated by the empirical relationship. It should be kept in mind that the experimental values are the results of only one irradiation and the average expansion rates may deviate considerably from this first result. The empirical relationship will be carefully checked against all types of experimental graphite receiving cooled test facility irradiations.

PILE GRAPHITE MONITORING

Process Channel 2677-H Bore Traverse and Mining: PT 105-458-P and 105-477-P
J. F. Fletcher, D. B. Lovett

On July 15, 1952, a bore diameter traverse was made of tube channel 2677-H, and mining samples were taken using the new centrifugal cutter.

The wire driving the recorder chart broke during the traverse. This resulted in considerable inaccuracy in the measured distances into the channel. Diameter measurements, however, are accurate to 0.001 inch.

Several of the tube block junctions in this channel were obscured. The channel itself was quite uniform in diameter; the diameters recorded ranged from 1.736 to 1.749 inches. The channel tends toward a slight ellipticity with the long axis in the vertical direction.

A plot of the vertical inside diameter of the process channel through the pile is shown in figure 3. Table III shows the inside diameter of the process channel in horizontal, vertical and two intermediate directions at several positions in the pile. The maximum, minimum and difference between these diameters at these positions are also included.

An examination of the powder samples showed the presence of only 1 particle of corrosion product. This was observed in the sample removed from the position 8 feet from the front Van Stone flange.

- (3) Bupp, L. P., Technical Activities Report, Graphite Studies, August 1951, HW-22145.
(4) Music, J. F., Technical Activities Report, Graphite Studies Group, Jan. 1952, HW-22145.

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INSIDE DIAMETER OF GRAPH

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TE BORE IN PROCESS CHANNEL 2677-H, 15 JULY 1952

DISTANCE FROM FRONT VAN STONE FLANGE (feet)

Figure 3

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TABLE III

Graphite Process Channel Inside Diameters
Process Channel 2677-H
(7/15/52)

Distances from front (feet)	INSIDE DIAMETERS						Difference Max. - Min.
	Vertical 0°	45°	Horizontal (90°)	135°	Maximum	Minimum	
10	1.740	1.741	1.745	1.743	1.745	1.739	0.006
14	1.744	1.741	1.737	1.740	1.745	1.736	0.009
17	1.739	1.742	1.741	1.741	1.742	1.739	0.003
21	1.743	1.742	1.740	1.741	1.743	1.740	0.003
22	1.743	1.741	1.738	1.741	1.743	1.738	0.005
27	1.741	1.740	1.741	1.741	1.742	1.740	0.002
29	1.742	1.745	1.742	1.741	1.745	1.741	0.004
31	1.744	1.743	1.743	1.745	1.746	1.743	0.003

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Presence of Aluminum Oxide in Mined Graphite Samples: J. F. Fletcher, D. B. Lovett

Microscopic examination of all previously mined graphite powder samples for the presence of the aluminum oxide corrosion product has been completed. In addition to the large concentration of oxide found in the samples mined from F File on 3-14-52 ('), traces of oxide were observed in samples removed at earlier dates from process channels of other piles. These channels in which traces of the aluminum oxide corrosion product were found and the dates when the samples were mined are listed in Table IV.

A report is being prepared to summarize the data of these observations.

TABLE IV

Process Channels in Which Aluminum Oxide Corrosion Product Traces Were Found in Addition to Those Found at 100-F on 3-14-52.

<u>Process Channel</u>	<u>Date Mined</u>
3467-B	11-2-51
1585-D	12-13-51
2291-D	5-24-50
2581-DR	5-9-51
0284-F	12-16-51
2584-F	7-12-50
3169-F	2-19-48
2677-H	7-15-52

GRAPHITE BURNOUT AND CHEMICAL STUDIES

X-Ray Studies of Graphitic Oxides -- J. R. Townsend

Figure 4 shows the most interesting X-ray diffraction effect found in the recent preliminary work on graphitic oxide (see Technical Activities Report, Graphite Studies Group - May 1952, HW-24676). The figure shows the effect of treating KSN graphite with increasing concentrations of potassium chlorate. The actual treatments of 2½ gram samples of KSN graphite are given in Table V.

TABLE V

Sample Treatment

<u>Sample</u>	<u>Treatment</u>
24	50 cc fuming HNO ₃ , 7 g KC1O ₃ , 20 minutes @ 59°C
22	50 cc fuming HNO ₃ , 9 g KC1O ₃ , 20 minutes @ 59°C
10	50 cc fuming HNO ₃ , 20 g KC1O ₃ , 10 minutes @ 55°C

(5) HW-24375 "Process Tube and Graphite Bore Measurements of F File," D. B. Lovett

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X-RAY INTENSITY, RELATIVE

Figure 4

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The principal feature of figure 4 is the change in relative peak intensities as the $KClO_3$ concentration increases. As mentioned in the earlier report, this is evidence that the reaction leading to the introduction of oxygen atoms between the graphite layers is heterogeneous. There is no detectable shift in the peak corresponding to $C_0 = 6.71$ Å. Such a shift would be expected if all crystallites were mixtures of planes with the regular spacing and planes with the larger spacing. The small shifts in the $C_0 \sim 11.8$ Å peak are not understood, especially in view of the fact that longer treatment results in a $C_0 = 13.3$ Å or more. It may arise from a very small number of graphite planes with the regular spacing in those crystallites which have been attached. This would also help explain the double peak at $C_0 \sim 11.5$ Å found on sample 10 when a high resolution tracing was taken.

Graphite - Carbon Dioxide Reaction — R. E. Woodley

A series of runs at a temperature of 962° C has been made on the graphite-carbon dioxide reaction kinetic study at constant volume. All runs were made at approximately the same initial pressure. After a definite interval of time, a sample of the gaseous products was removed and analyzed mass spectrometrically to give the results shown in Table VI.

TABLE VI
Graphite-Carbon Dioxide Reaction, Gas Composition

Run No.	Approximate initial pressure (cm.)	Time of sample removed.	%CO ₂	%CO
1	38	1 min. 44.6 sec.	90.2	9.18
2	38	6 min. 56.4 sec.	75.4	23.9
3	39	10 min. 37.9 sec.	69.6	30.3
4	38	13 min. 52.0 sec.	66.8	33.2
5	37	22 min. 5.4 sec.	62.4	37.1

Pressure measurements during the run were recorded only for Run 5 above. Using these pressure measurements and the percentages of CO₂ given above, the pressure of CO₂ at the time of product removal can be calculated. The log of this pressure plotted against the square root of the time gives a straight line between CO percentages of 9 to almost 30 percent as shown in Figure 5. At this temperature, it is virtually impossible to obtain any points at lower CO percentages.

Using the method outlined in HW-24836, the extent of "surface oxide" formation or adsorbed CO has been calculated for this series of runs. It was found that the relative amount of CO in the gas phase to that adsorbed on the graphite increases with time as shown in figure 6. The average equation that the reaction follows during the first 22 minutes at 962° C is

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Figure 5

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Figure 6

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This is essentially the same equation that was found at 905° C, i.e.

The extent of CO adsorption is therefore seemingly independent of temperature in this temperature range.

Graphite-Carbon Dioxide-Helium System: Reaction Rates — H. H. Burton

Difficulty has been encountered in the attempt to obtain a supply of helium sufficiently pure to allow heating a sample of graphite to constant weight at 1000° C. At present the weight losses of a graphite sample after successive heatings in purified helium are of such a magnitude (0.0003 gm/gm/hr @ 1000° C) as to prevent comparison between relative burnout rates in atmospheres of 100% CO₂ and CO₂ with 0 - 20% He added. Current work is directed toward the construction of a more efficient helium purification train and a more reliable form of gas flow indicating device.

Radiation Induced Reactions - PT 105-504-E — R. E. Woodley

Samples of gases and graphite in sealed quartz containers are being prepared to replace those which will be discharged from 0776-H in August. This second charge will consist of six tubes containing pure CO, five containing pure CO₂ plus weighed samples of powdered GBF graphite, and five containing weighed solid samples of GBF graphite in a vacuum. The distribution of these tubes along the process tube will be as follows: a tube containing pure CO, then one containing graphite in a vacuum, then pure CO₂ plus powdered graphite, etc. This cycle will be repeated until the entire process tube is lined.

The tubes containing pure CO are being used in each run as a direct measure of the amount of irradiation and the distribution of radiation along the process tube.

If a radiation induced reaction does occur between pure CO₂ and graphite, the effect of increasing the surface area will be of importance. Since tubes containing pure CO₂ and solid graphite pieces are now being exposed, the use of tubes containing pure CO₂ and powdered graphite in the second charge will allow this surface effect to be measured.

A test of the "knocked on" atom theory will be made by exposing samples of graphite in a vacuum and comparing the resulting properties with those of graphite exposed in various gas atmospheres.

The method of preparation of samples is described in HW-24028.

Degassing of Irradiated Graphite and Graphitic Oxide — R. E. Woodley

As reported in HW-24676, the 2000°C stored energy peak of both graphitic oxide and graphite irradiated in a cold test hole can be completely annealed out by extended heating at 1550°C. In a further attempt to compare the oxygen-graphite lamellar compounds with irradiated graphite, the gases evolved upon heating the two substances were collected and analyzed. A 0.1057 g. sample of graphitic oxide released 16.8 cc. of gas (STP) upon heating to 2000°C. A sample of irradiated CSF graphite, IRR # . . ., with 640 MD/CT cold test hole exposure was heated to 500°C and 0.53 cc. of the evolved gases collected. The mass spectrometric analyses of both gas samples are given in Table VII.

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TABLE VII

Analysis of Gases Evolved on Heating Graphitic Oxide and Irradiated Graphite

<u>Gas</u>	<u>Percentage</u> (for graphitic oxide)	<u>Percentage</u> (for irradiated graphite)
CO ₂	44.3	62.2
CO	44.6	14.0
N ₂	9.37	17.7
O ₂	2.06	4.66
H ₂	0.51	0.56
CH ₄	—	0.56
A	0.10	0.24
H ₂ O	High	High

Other peaks were found in the spectra of the sample evolved from irradiated graphite but could not be identified. The above analysis is on the basis that the sample is free from these unknown constituents. An explanation of these results will be attempted after a more thorough study has been made.

EXPERIMENTAL GRAPHITE PROGRAM

TS Graphite Program -- W. C. Riley, D. H. Curtiss

Eight capsules containing twelve transverse and twelve parallel samples of TS graphite have been discharged from process tube 0963-D. These samples were cut from an extruded cylinder six inches in diameter and six inches long. The temperature of graphitization was 3300°C. Tube 0963-D operated as a water cooled test facility with no metal charge under PT 105-269-P. The samples received an equivalent cold test hole exposure of about 420 MD/CT. Physical property changes are presented in Table VIII.

TABLE VIII

<u>Type of cut</u>	<u>% Change in Length</u>	<u>Thermal Conductivity Ratio</u> k _o /k	<u>Electrical Resistivity Ratio</u> P/P _o
transverse	+ .254	26.0	3.59
parallel	+ .104	27.2	3.89

The expansion noted in the transverse direction corresponds closely to the physical expansion of transverse CSF graphite under similar irradiation conditions. The expansion in the parallel direction is somewhat surprising. This is the only graphite irradiated to date that exhibits appreciable expansion in the parallel direction. On a percent per thousand MD/CT basis, this expansion amounts to .25% - just slightly less

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than half the transverse expansion of CSF graphite. Thermal conductivity and electrical resistivities were found to be quite comparable to those measured on CSF graphite of similar exposure history. It is possible, however, that production grade graphite may behave somewhat differently under irradiation than does the six inch cylinder which these samples were cut.

National Carbon Company Experimental Graphites -- W. C. Riley

Careful evaluation of the effect of impregnation of TS graphite physical properties has been continued. Additional experimental bars of impregnated and non-impregnated TS graphite have been sampled to obtain better averages for thermal conductivity measurements and to provide sufficient samples for an extensive exposure program.

Six casings of transverse and four casings of parallel samples of TS graphites, each containing four impregnated and four non-impregnated samples have been charged in water cooled test facilities. One casing of transverse material and one casing of parallel graphite were discharged on the July 23 shutdown at B Area and several casings of transverse TS graphite were loaded at that time. Appropriate hot test hole irradiations have begun.

Battelle Memorial Institute Graphites -- W. C. Riley

Shipment OGT-11282 of experimental graphites has been received from Battelle Memorial Institute. Physical properties are being measured. Measurements of electrical resistance taken at Hanford are in good agreement with similar measurements made at Battelle.

These graphites have a density range from 1.23 to 1.62, thereby providing a means of studying the effect of density on various irradiation characteristics of graphite.

Lots E-46 and E-34 have been measured for thermal conductivity and have been found to have the lowest thermal conductivities measured on any unirradiated graphite to date. Lot E-46 is produced from a coke calcined from binder resin with Ironsides Company phenol-formaldehyde resin for binder, while Lot E-34 consists of a Standard Oil Company B coke and a binder of Standard Oil Company No. 4 Korete petroleum asphalt. Both lots were graphitized at 2500°C. The average conductivity of parallel samples of lot E-46 was about 0.02 cal/cm/sec/°C. This is equivalent to the conductivity of a parallel sample of CSF graphite that has been irradiated in a cold test hole for 600 MD/CT. The advantage of using a graphite of low thermal conductivity as a pile moderator would be reflected in higher graphite temperatures and thus increased reactivity for typical Hanford pile behavior. This advantage would be particularly pronounced just after "startup," when there is difficulty in maintaining high graphite temperatures. Therefore, E-46 transverse samples will be exposed in a cold test hole to determine conductivity changes with irradiation and the maximum dimensional instability. Parallel samples have been loaded in a test facility running at ambient pile temperature to determine if any decrease in thermal conductivity on irradiation is possible at these temperatures.

CONTROLLED TEMPERATURE EXPOSURE OF GRAPHITE: PT-105-403-P -- D. B. Lovett

The exposure of graphite samples to pile irradiations in tube 1684-B continues. The controlled temperature remains at 135° C. The average temperatures for the month of the other three samples are 122°C, 115°C, and 112°C. These show no appreciable change from those of last month.

The control apparatus for the series III exposure is being assembled in the instrument shop.

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THERMAL CONDUCTIVITY OF GASES — J. M. Davidson, J. F. Music

Seven runs were made during the period covered by this report. Of these, five were made on air at various pressures. These were made to determine the reproducibility of measurement and to examine the effect of convection. A plot of $\log (T_1 - T_2)$ vs time is a straight line as shown in figure 7, where T_1 and T_2 are the temperature of the inner and outer cylinders respectively. The slope of this line is obtained by the method of least squares and is related to the thermal conductivity of the gas in question because

$$(1) \quad \alpha_{\text{gas}} = m \times 2.303 = \frac{KA}{Mcd}, \quad \begin{array}{l} K - \text{gas cond. in g-cal/cm.}^\circ\text{C. sec.} \\ A - \text{mean area of interior surfaces} \\ M - \text{mass of central cylinder} \\ c - \text{specific heat of central cylinder} \\ d - \text{gas skin thickness} \\ m - \text{slope of line } \log (T_1 - T_2) \text{ vs } t \\ t - \text{time after temp. diff. established} \end{array}$$

This is neglecting the heat leakage through the sapphire positioning beads and the thermocouple. If we incorporate this leakage, equation (1) becomes

$$(2) \quad \alpha_{\text{gas}} = \alpha_t - \alpha_v = \frac{KA}{Mcd} \quad \text{where } \alpha_t \text{ is the measured } \alpha \text{ with the gas in the apparatus and } \alpha_v \text{ is the measured } \alpha \text{ with the apparatus run in } \dots$$

Because there exists an uncertainty of approximately 10% in the spacing, d , and minor uncertainty in A , c , and m , the conductivity of the gas in question is best found by the relation

$$(3) \quad \frac{\alpha_{\text{gas}_1}}{\alpha_{\text{gas}_2}} = \frac{K_1}{K_2} \quad \text{where } K_1 \text{ is the conductivity of a particular gas, e.g., air whose conductivity is known to some precision and } K_2 \text{ is the conductivity in question.}$$

Hercus and Laby (6), as well as Johnston and Taylor (7) have made refined measurements on air at 0°C and all relative calculations made in this experiment are to be based on their work.

Also, by measurements on air, equation (2) can be modified to the following:

$$\alpha_{\text{gas}} = \alpha_t - \alpha_v = kC_1 \quad \text{where } C_1 = \frac{A}{Mcd}$$

By this means, the uncertainty in d may be diminished by calculation knowing C_1 , A , c and m .

(6) Hercus and Laby - Pro. Roy. Soc. A 95 191 (1919)

(7) Taylor and Johnston - Journal of Chemical Physics 14, 220, (1946)

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Figure 7

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The values for α_t and α_v that have been obtained thus far are as follows:

Values for α_t Air at a Temp. of 0° C.

Run #	Pressure	α_t
2	760 mm	-0.01946
4	760 mm	-0.01939
14	200 mm	-0.01899
15	98 mm	-0.01985
16	50 mm	-0.01905
17	28 mm	-0.02038
18	17 mm	-0.01872

Values for α_t Vacuum at a Temp. of 0° C.

Run #	Pressure	α_t
9	25 microns	-0.00808
10	25 microns	-0.00861
11	25 microns	-0.00860
12	8 microns	-0.00637
13	8 microns	-0.00574

The 25% difference between run 9 and 13 indicates a need for a lower pressure to have a valid vacuum run.

Values for α_t Helium at a Temp. of 0° C.

Run #	Pressure	α_t
6	760 mm	-0.08164
19	48 mm	-0.09109

It should be noted that an error of 0.15° C. in one point in the temperature measurement (corresponding to 0.1 μ v on a platinum to platinum - 10% rhodium thermocouple) will cause an error of approximately 2% in the least square slope calculated on 20 points.

The limit of error for a K-2 type potentiometer is 0.2 μ v due to thermal and parasitic electromotive forces of unknown magnitude. The error in the thermal conductivity caused by an error in temperature measurement will be greater for the gases and gas mixtures of low conductivity than for those of higher conductivity.

The mean absolute error and the mean square error, as well as the precision constant, h, the standard deviation and probable error are calculated for air using the following

The mean absolute error -

$$|\bar{x}| = \frac{2h}{\sqrt{\pi}} \int_0^{\infty} x e^{-h^2 x^2} dx = \frac{1}{h\sqrt{\pi}} = \frac{0.5643}{h}$$

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The mean square error -

$$\overline{x^2} = \frac{2h}{\sqrt{\pi}} \int_0^{\infty} x^2 e^{-h^2 x^2} dx = \frac{1}{2h^2}$$

both of which reduce to

$$|\overline{x}| = \frac{\sum_{i=1}^n m_i - n}{n} = \frac{\sum_{i=1}^n x_i}{n} = \frac{1}{h\sqrt{\pi}}, \quad h_1 = \frac{1}{|\overline{x}| \sqrt{\pi}}$$

and

$$\overline{x^2} = \frac{\sum_{i=1}^n (m_i - n)^2}{n} = \frac{\sum_{i=1}^n x_i^2}{n} = \frac{1}{2h^2}, \quad h_2 = \frac{1}{\sqrt{\overline{x^2}} \sqrt{2}}$$

From these we have

$$\begin{aligned} |\overline{x}| &= .00042 \\ h_1 &= 1.344 \times 10^3 \\ \overline{x^2} &= 2.732 \times 10^{-7} \\ h_2 &= 1.379 \times 10^3 \end{aligned}$$

The value for h_1 and h_2 are not identical and should not be expected to be, but agreement is such to indicate the data are reliable.

The standard deviation

$$\sigma = \sqrt{\overline{x^2}} = 51.26 \times 10^{-5}$$

and the probable error in measurement is

$$0.6745 \sigma = 34.58 \times 10^{-5},$$

approximately 2% of the arithmetic mean.

More investigations are to be made with the gases in question at 0° C before measurements are made at elevated temperatures.

Work is being continued on the new vacuum rack to be used for mixing and low pressures.

GRAPHITE EXPOSURES IN THE MTR -- A. T. Whatley

AEC Form 320, "Request for Irradiation in the Materials Testing Reactor," was completed and issued as a document, HW-24830. During a trip to the MTR site at Arco, Idaho,

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A. B. Carson learned that our requests had been approved by the AEC, and we would be given the requested space in the MTR. The latter has been confirmed by a telephone conversation with Dr. W. B. Lewis, of the Phillips Petroleum Company, who is at the MTR site.

The graphite samples will be CSF material, transverse cut, 4" long and .425 inches in diameter. Four of these will be contained in each sample holder which will consist of four 7/16 in. ID holes drilled in a 2S aluminum rod 1 1/4 in. OD. This holder will be made water tight by a crimp seal, as described in HW-20722. Samples and containers will then be sent to Arco for exposure.

Present plans call for exposure to be arranged so that at the end of approximately ninety days we will have eight graphite samples with about ten days in the MTR, eight with about thirty days and eight with about ninety days. One set such as this will have been exposed in the highest attainable flux, and another in a position with a thermal flux equivalent to the former but with a much lower energetic flux. From the above results, it is hoped to obtain an empirical correlation between exposures in the MTR and exposures in the Hanford piles. Some qualitative results on the energy dependence of damage should also be obtained. The sample carriers have been designed, and are at present being fabricated. It is planned to get the samples to Arco between August 15 and September 1.

Inquiries are being made as to the various methods available for determining the energy spectrum of the neutron flux in the MTR, such knowledge being a prerequisite to any quantitative study of the dependence of graphite damage on neutron energy.

SURFACE STUDIES -- C. N. Spalaris

The surface characteristics of several samples of irradiated graphite were investigated this month. These samples are part of a stringer of graphite cylinders removed from the C test hole of B Pile in December 1950 (8). The surface area values of these samples varies with the distance from the edge of the reflector. There are about twenty-one samples representing a distance of 2 to 172 inches from the reflector to the center of the pile. The surface area of fourteen of these samples has been determined. The values will be given in the next month's report along with other details and a graph indicating the change of surface area with the distance from the reflector.

A new series of graphite samples has been prepared for irradiation in various gas atmospheres. Three WSF graphite samples have been enclosed in quartz tubes and their surface characteristics will be determined before being placed in the pile. Three more samples of graphitized lampblack will be enclosed in quartz tubes, their surface characteristics determined and then exposed in a temperature controlled process tube in H Pile. Details of preparation and exposure will be presented in a later report.

The pore size distributions of two samples of KC graphite were determined and the results agree with those obtained and reported previously for this type of graphite(9).

(8) Curtiss, D. H., "Pile Thermal Conductivity," HW-24174, pp. 5, 18

(9) Technical Activities Report - Graphite Studies, February 1952, HW-23720.

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The two samples are #87-92 QP O₂ and #87-88 QP CO, outgassed at 1000° C for 45 minutes and enclosed in quartz tubes with oxygen and carbon monoxide gasses respectively. At the present, both are placed in process tube #0776 of H Pile for irradiation. The pore size distribution of the above samples will be compared with that obtained for the same samples after exposure to radiation.

DAMAGE MECHANISM STUDIES — A. M. Corners

The study of electrical conduction in solids is being continued. While the projected in-pile conductivity experiment is justified on a change-in-measurable property basis, it is felt that the full value of the experiment may be realized only if the factors causing a change in resistivity are known.

The completion of the experimental setup has been delayed several times by the unavailability of materials. For example, the aluminum tubing which is to contain the lead wires proved to be too small. A search is under way for larger tubing which is corrosion resistant.

In accordance with previous discussion that pile gamma radiation may produce detectable graphite damage, negative theoretical predictions notwithstanding, two containers of both damaged and undamaged graphite were subjected to intense gamma radiation by being covered with freshly discharged slugs in the pile basin. Neither the damaged nor the undamaged graphite showed any significant change in C₀ spacing, thermal or electrical conductivity after a two-months' exposure. A six-months' exposure will soon be completed and the effects noted.

STORED ENERGY STUDIES — J. C. Ballinger

Heat of combustion results have been received from the National Bureau of Standards for three common types of unirradiated graphite. These samples were sent to the Bureau to be used as replacement standards against which the heats of combustion of irradiated samples could be compared. The results are as follows:

TABLE IX
Heat of Combustion of Virgin Graphites

<u>Sample Type</u>	<u>Cal/gm</u>	
KC	7833.8	Mean of 5 determinations
CSF	7832.9	Mean of 3 determinations
CSGBF	7833.0	Mean of 4 determinations
Unknown	7839.1	Previously used standard

Included for comparison is the previously used standard.

The weighted mean of the new measurements is 7833.1 cal/gm with a total estimated uncertainty of 0.02 per cent. This new value will be adopted for future total stored energy calculations. The reason for the 6 cal/gm difference between the new and old values is not clearly understood but an effort is being made to account for the discrepancy.

A visit was made to the Bureau of Standards for the purpose of discussing the progress of the total stored energy program. It was indicated by the Bureau that their part in the program would be speeded up in the very near future to handle approximately 12 samples per month.

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Construction is under way at this laboratory on a new calorimeter which has been designed to eliminate some of the inherent problems encountered with the one presently in use. A detailed discussion of these problems and the proposed improvements is presented in Report HW-24692.

ANNEALING STUDY OF X-RAY PEAK SHAPES — C. A. Lund and J. R. Townsend

Eight irradiated graphite samples showing typical line shapes have been annealed at 100° intervals up to 600°C. Annealing temperatures of 1000°C are anticipated using regular laboratory furnaces available. Higher temperatures will be reached using an induction furnace now being constructed. Two samples showing an 002 diffraction peak with two maxima seem to be shifting as a whole rather than annealing first one peak component and then the other.

FLUX AND HEAT CALCULATIONS — K. J. Tauer

The request to the Theoretical Physics Group to calculate neutron fluxes at various positions in the Hanford piles is currently at the position of evaluating the improvement possible over similar calculations done three or four years previously. Significant improvement in these calculations will require improved fission spectrum determinations as well as better information on the inelastic cross section of uranium. The fission spectrum is now reasonably well known so that the inelastic cross section datum is the major consideration determining whether a new calculation is profitable.

The success of a heat generation experiment continues to depend on the magnitude of unknown spurious e.m.f.'s developed in in-pile leads which may mask an expected output of approximately 15C microvolts. However, the proposed experiment will give a continuous measure of the relative changes of thermal conductivity with exposure, so that the experiment is being continued cognizant of the possible lack of applicability to heat generation determinations.

PHYSICAL EXPANSION ANNEALING — E. M. Woodruff

Samples irradiated at 175°C and 225°C under Production Test 105-403-P have been re-run with the new thermocouple installed in the interferometer. No annealing resulted below an annealing temperature of 700°C on the sample irradiated at 225°C. The sample irradiated at 175°C began to anneal at 625°C.

New samples have been cut for the lower temperatures of irradiation and the data obtained from runs to be made on them will be presented next month in graph form along with the higher temperature of irradiation data.

IN-PILE CONTROLLED ATMOSPHERE PROGRAM — D. M. Knott

Liaison with Pile Services Group personnel concerned with the fabrication of the in-pile controlled atmosphere facility continued during this month. Calculation of the gamma radiation intensity and decay rate of the heater-cooler assembly after discharge has been made by the Pile Physics Group on the basis of the weights of elements present per linear foot of assembly. Table X indicates the results of their calculations.

TABLE X
Gamma Radiation From the C-410 Heater-Cooler Assembly After Discharge

Time After Pile Shutdown	0	0.5	12
Radiation Intensity r/hr at one foot from center of heater	75,000	2,000	650

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The following mechanical difficulties continue to plague the fabrication of the fac-

- (1) Leaks in the water jacket between the water jacket and the electrical heating elements.
- (2) Leaks in the copper and copper alloy components of the gas recirculation and analysis system. This problem is aggravated by amalgamation with mercury from the Toepler pump.
- (3) Failure of the gas recirculation pumps after limited service. All pumps thus far tested have become inoperative after less than 48 hours of service. The plastic liners have expanded and taken a permanent set which prohibits the rotor from exercising its compressive action. In one case the plastic liners ruptured.
- (4) Sample containers develop pin holes or active blow-outs during high temperature leak testing.

Pile Graphite personnel is actively assisting Pile Services personnel in the development of a satisfactory sample container.

G. F. Music

In-Charge, Graphite Development
PILE TECHNOLOGY UNIT
ENGINEERING DEPARTMENT

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