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RLD-CG-3-REV1

March 29, 1949

REPORT ON GRAPHITE TESTING FOR DR PILE

This document consists of 12 pages. No. of copies. Series

Purpose

When the testing program for DR was started in November, 1947, considerable difficulty was experienced in finding literature describing procedures, results, and analyses of the testing done for prior construction. This report is written in detail to provide a basis for any future testing, as well as to furnish a compilation for current use.

Summary

The graphite testing was accomplished by means of a series of functional tests in the 305 pile, accompanied by pertinent chemical analyses. One of the first characteristics of a graphite heat that became apparent was the relatively poor quality of the bottom layer of heat in the furnace. As a result, each heat was divided into an "O" fraction and an "N" fraction for testing and allocation. The "N" fraction consisted of the more inferior section of the heat.

Early in the testing program, United Carbon Products Company of Fay City, Michigan, developed a method of removing a considerable percentage of the impurities remaining in graphite after the graphitization process. The purification process was modified and used to produce large amounts of purified graphite at the Morganton, North Carolina plant of the National Carbon Company for use in the DR pile.

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The purity reached by the graphite treated in the "F", or purification, furnaces, was considerably higher than was expected. Calculations showed that other impurities than boron and vanadium were present in the unpurified graphite and were removed by the "F" process. Subsequent investigation showed that these impurities were rare earths, specifically samarium, europium, and gadolinium.

The graphite allocated to the green zone of DR pile consisted entirely of purified graphite. The average  $\Delta ih$  of this material was + 0.946 with an average density of 1.718 gm/cm<sup>3</sup>.

The white zone had an average  $\Delta ih$  of + 0.281 and an average density of 1.687 gm/cm<sup>3</sup>.

The blue zone had an average  $\Delta ih$  of + 0.121 and an average density of 1.666 gm/cm<sup>3</sup>.

The red zone had an average  $\Delta ih$  of -0.030 and an average density of 1.672 gm/cm<sup>3</sup>.

Calculations based on the above values show that the DR pile should have a reactivity approximately 400 inhours greater than the average of the D and F piles at startup. The average density of graphite in DR is 1.678 gm/cm<sup>3</sup>.

#### Details

#### Testing Methods and Standards

The graphite used in the construction of DR pile was segregated into various quality grades by means of functional tests. The sample bars were compared with bars of known quality to find their relative effect upon the reactivity of the 305 test pile. A group of two hundred bars of CS graphite was chosen as representative of the graphite used in the 1943 and 1944 construction, and of these, two bars of about average density and purity were selected as standards. These were used as master standards and designated as 3-S-3 and 3-S-4. All functional test results made on graphite for the B, D, and F piles in 1944 as well as the recent tests on DR graphite are relative to these same master standards. All  $\Delta ih$  values can therefore be compared directly. In order to preserve the master standards they were only used for periodic checks against another pair of "working" standards known as 4-S-3 and 4-S-4.

A functional test consists of placing the pair of standard bars in the center of the pile by means of a removable graphite stringer, and calculating the reactivity of the pile in inhours. Then the two standard bars are replaced by the sample bars, and the new reactivity is ascertained. The difference between the two inhour readings is recorded as the  $\Delta ih$  of the two sample bars. A correction is then made for the density of the bars, and for the difference in purity and density between the working and master standards. The final result, or  $\Delta ih$  of the two bar sample, is the difference in inhours between the reactivity of the 305 test pile with the master standards in place, and the reactivity of the pile with a pair of bars in their place whose purity is indicated by the  $\Delta ih$ , but whose density is the same as that of the master

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standards.<sup>2</sup> In order to obtain the effective  $\Delta ih$  for use in 105 type piles it is necessary to add a density correction different from the one used in the 305 pile (because the effect of changes in density is not the same in the two types of piles<sup>3</sup>). The effective  $\Delta ih$  is the measure of quality used in segregating graphite.

In the course of the testing program, it became necessary to test bars one, three, and four at a time as well as in pairs. For the single bar tests, the sample bar replaced one of the standard bars, and correction factors were calculated so that the  $\Delta ih$ , if multiplied by two, would give the correct result for two bars of equal purity. The four bar test necessitated the choosing of two more working standards, 5-8-3 and 5-8-4. Two graphite stringers, side by side, were used in four bar testing. Correction factors again were calculated so that the final result could be divided by two, furnishing a  $\Delta ih$  equal to the average of two double bar tests.

The three bar tests were a combination of single and two bar tests, using two stringers. Four more working standards, 1F, 2F, 3F, and 4F, were chosen when testing of purified graphite became routine. These standards are also purified bars, and new correction factors were calculated for their use. An additional refinement for single bar testing was the use of two half-length standards for centering the sample bar in the pile<sup>4</sup>.

#### Sampling Procedures, Unpurified Graphite

At the beginning of the testing program, the decision was made to use a systematic sampling system, rather than a random method such as that used in prior construction. With a wise choice of sample bars, any systematic trend in purity should be discovered.

The graphite was manufactured by the National Carbon Company, Inc. of Cleveland, Ohio, at a plant in Morganton, North Carolina. The unit of graphite manufacture, produced in one furnace, is known as a heat, and consists of 342 bars.

The graphitization furnace is loaded with seven layers of graphite bars, each layer containing forty-eight bars. There are three extra bars at each end, two bars on stacks one and forty-eight, and one each on stacks two and forty-seven.

The first sampling system used consisted of ten sample bars per heat. The locations of the sample bars are given in the following table.

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TABLE I

FIRST SAMPLING SYSTEM

<u>Sample Bar No.</u>	<u>Stack</u>	<u>Layer (numbered from top)</u>
I	1	1
II	12	1
III	12	4
IV	12	7
V	24	1
VI	24	4
VII	36	1
VIII	36	4
IX	36	7
X	48	1

This sampling system was used on the first ninety heats of CS and KS graphite. The bars were tested in pairs: I and X, II and VII, III and VIII, IV and IX, V and VI. Samples IV and IX gave consistently lower  $\Delta$ ih values than did the other pairs, with the possible exception of I and X. On the basis of these tests, the decision was made to divide the heats into "W" and "O" fractions. The "W" fraction at first included the three stacks of bars on either end, and the lower three layers from each furnace heat. Later, after a complete heat had been tested one bar at a time, (heat CS-100), the division of the heat was changed so that the "W" fraction contained only the bottom layer of each heat. The upper six layers constituted the "O" fraction. This zoning system was continued until graphite testing for DR was completed.

The results of testing each bar from heat CS-100 showed that every bar in the bottom layer of the heat was distinctly inferior in purity to the bars above it (Table II). A gradual decrease in purity was noted from top to bottom in the upper six layers also, but it did not compare in magnitude with the decrease apparent in the bottom layer<sup>2</sup>.

Fifty bars were tested singly from heat CS-132, and the averages for the bars from each layer are given in Table II. This heat was raised to a higher temperature in the graphitization furnace than was normal, and held for a longer time at this elevated temperature. The total energy input was approximately double the normal amount. The same general trend of purity apparent in CS-100 is in evidence in this heat.

Fifty bars of KC-244 were tested singly and average values by layers are shown in Table II. Approximately the same purity pattern was found in this heat as in CS-100 and CS-132. The pattern does not seem to be influenced by the type of graphite.

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TABLE II

ASH VALUES BY LAYERS

<u>Layer from Top</u>	<u>CS-100</u>	<u>CS-132</u>	<u>KC-244</u>
1	0.235	0.283	0.240
2	0.215	0.301	0.273
3	0.204	0.293	0.292
4	0.201	0.270	0.267
5	0.177	0.270	0.251
6	0.152	0.227	0.235
7	-0.101	-0.118	0.036

The inadequacy of ten sample bars per heat became apparent, so after the first ninety heats had been tested, a new sampling system was put into effect. The number of sample bars was increased to twenty-two. The following table gives the positions of the new samples in the furnace:

TABLE III

REVISED SAMPLING SYSTEM

<u>Stack</u>	<u>Layer (numbered from top)</u>	<u>Sample Number</u>
1	3, 5	1, 2
6	1, 4, 7	3, 4, 5
12	3, 5	6, 7
18	1, 4, 7	8, 9, 10
24	3, 5	11, 12
30	1, 4, 7	13, 14, 15
36	3, 5	16, 17
42	1, 4, 7	18, 19, 20
48	3, 5	21, 22

Heats up to and including CS-147 were tested using all twenty-two samples. At this point the manufacture of CS graphite was discontinued in favor of KC production. When production of CS was resumed at heat #216, the heats were being divided into "O" and "N" fractions. Statistical analyses showed that the number of samples could be safely reduced, so only sixteen of the twenty-two samples were tested. Twelve samples were from the "O" fraction and four from the "N" fraction. This system was used for testing all subsequent unpurified heats for DR construction.

Chemical Analyses

At various times throughout the testing program, chemical analyses were made on graphite bars. In the majority of cases, the elements analysed were boron and vanadium, since they were the only important neutron absorbers known to be in graphite. Ash analyses were made with each boron and vanadium analysis.

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A formula was developed for use in calculating the  $\Delta ih$  of a pair of bars from the amount of boron and vanadium present. The formula is as follows:

$$\Delta ih = 0.42 - (1.08)(\text{ppm. boron}) - (0.00131)(\text{ppm. vanadium})$$

(It was later found that 0.50 works better than 0.42 in this formula.)

Samples taken from a slice fifteen inches from one end of graphite sample bars of the first few heats were analyzed both at Hanford and at Y-12 (Oak Ridge, Tennessee). The  $\Delta ih$  values calculated from results of these tests did not correlate very well with test pile results. Suspicion that the boron content of a bar is non-uniform was confirmed by results of analyses on representative samples from bars which had been completely ground up and mixed thoroughly. The values of calculated and test pile  $\Delta ih$  then checked closely. The formula cannot be expected to give correct results on purified graphite, however.

### Purified Graphite<sup>6</sup>

The average  $\Delta ih$  (weighted according to the effectiveness of the various zones) of the graphite used in piles D and F is + 0.15. The purest graphite used in these piles had a  $\Delta ih$  value of about + 0.32. The main impurities known to be present in graphite at the beginning of DR testing were boron and vanadium. A method for removing these impurities was developed by the United Carbon Products Company, of Bay City, Michigan. In brief, the method consisted of adding freon gas at a high temperature. It decomposes into chlorine and fluorine gas, which unite with the metallic ions in the graphite. The volatile halides are swept out, as the furnace cools, by a stream of nitrogen gas. The first bars of graphite produced by this method gave  $\Delta ih$  values as high as + 0.40.

The purification process was modified and used by National Carbon Company in the manufacture of large amounts of purified, or "finished", graphite. The usual graphite type letters were followed by an F to denote finished graphite. Thus, CG graphite which had been subjected to the purification process was labelled CSF.

In detail, the original procedure used by National Carbon Company was as follows: Acheson graphitizing furnaces were converted to purify sixty-eight full length bars in a single layer. Gas distribution tubes were placed under each bar, and the tubes and bars were covered by powdered coke. The heat was supplied by an electric current passing through the furnace. The furnace was sealed from the atmosphere by a steel sheet on top and masonry on the sides and ends. On the heating cycle, which required from 4 to 5 hours, carbon tetrachloride vapor was swept into the furnace by nitrogen gas until the temperature reached 2000° C. Freon was substituted for CCl<sub>4</sub> and nitrogen from 2000° C until the peak temperature of about 2400° C was reached and until the furnace cooled off again to 2000° C. CCl<sub>4</sub> and nitrogen were again added until the furnace cooled to 1000° C. The nitrogen was allowed to flow for six hours as a further sweep. The entire cooling cycle required about thirty-six hours<sup>7</sup>.

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Each bar in the furnace was numbered consecutively from one end to the other. Sample bars were taken systematically from certain positions, depending on the information desired.

The first heats of F material produced by the National Carbon Company were below expectations. Induced radioactivity studies at Hanford showed that residual chlorine in the graphite was causing the trouble. Apparently the chlorine, which formed by decomposition of the freon and carbon tetrachloride at high temperatures, was adsorbed on the graphite or united chemically with impurities and remained behind after the cooling cycle. The purification procedure was changed to omit the  $CCl_4$  on the cooling cycle. The maximum temperature reached was increased to about  $2500^{\circ}C$ . On the cooling cycle, the freon addition was stopped at  $2000^{\circ}C$  and nitrogen was used as a sweeping gas until the furnace cooled to  $1000^{\circ}C$ . At this point, all gas addition was stopped.

Subsequent experiments during the graphite testing schedule for DR brought to light several more facts about the purification process. To obtain the purest possible graphite, it is necessary to add chlorine in some form during the heating period while the temperature is between  $1000^{\circ}C$  and  $2000^{\circ}C$ . Chlorine is as good as carbon tetrachloride and nitrogen in this respect. If chlorine is substituted for freon as a purifying agent, the purity of the graphite is reduced slightly. The amount of freon used is not critical. If nitrogen gas is used throughout, with no other gases added, about one-third of the neutron absorbing impurities are removed that are removed by the regular process. The value of the final sweep by nitrogen was in doubt at the end of DR testing. It was also discovered that purer graphite can be produced by using two layers instead of one in the purification furnace.

#### Rare Earths in Graphite

At the beginning of the testing schedule, the only impurities with high capture cross-sections known to be present in the graphite were boron and vanadium. Quantitative chemical analyses were made on these impurities, and the maximum  $\Delta_{ih}$  values to be expected upon their removal were computed. The maximum  $\Delta_{ih}$  for a two bar sample was about + 0.50 corresponding to an atomic capture cross-section for carbon of an estimated 4.06 mb. However, as the purification process progressed, it became apparent that a  $\Delta_{ih}$  of greater than + 1.00 could be achieved. This gave carbon a maximum atomic cross-section of around 3.6 mb., and required the presence of other high cross-section impurities in the unpurified graphite.

In various analyses of induced radioactivities in unpurified graphite at Hanford rates of decay were discovered which corresponded closely to published values of half lives for certain rare earth elements. A series of experiments was undertaken to determine which rare earths were present if any, and in what amounts. At the end of the DR testing period, it was apparent that europium, samarium, and possibly gadolinium were the rare earths present, and their removal by the purification process resulted in the unexpectedly high  $\Delta_{ih}$  values.

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Quality of Graphite Zones in DR

DR pile was built of graphite whose quality had previously been determined by the functional tests as explained in previous sections of this report. Since the neutron flux is greater in the center of the pile than around the edges, and the loss in reactivity due to impurities is proportional to the square of the flux, it is mandatory that the best graphite be placed in the center of the pile. Accordingly, the graphite of the best quality was placed in the so-called "green" zone, the innermost core of the pile. Around the green zone in concentric layers are the white (including the W-4 zone), blue, and red zones. The red zone is the outer shell, in which the least pure graphite is used.

Summaries of the types, quantities, and qualities of graphite allocated to the green, W-4 (subdivision of the white zone), white, blue, and red zones of the DR pile are shown in Tables IV, V, VI, VII, and VIII, respectively.

TABLE IV

GREEN ZONE\*

<u>Type</u>	<u>No. of Heats</u>	<u>Average Density</u>	<u>Average Δlh (effective)</u>
KCF	194	1.724	0.97
CSF	33	1.666	0.79
KCNF	20	1.728	0.94
KSF	2	1.702	0.73
Total	249	1.718 <sup>***</sup>	0.946 <sup>**</sup>

\* The green zone allocation included only "finished" or purified graphite. The reason for the high density was a double pitch impregnation, to offset the low coking value of Chicago pitch.

TABLE V

W-4 ZONE

<u>Type</u>	<u>No. of Heats</u>	<u>Average Density</u>	<u>Average Δlh (effective)</u>
CSF	14	1.670	0.72
CSNF	1	1.667	0.32
KCF	5	1.730	0.66
KCNF	2	1.735	0.84
KSF	1	1.693	0.73
Total	24	1.691 <sup>**</sup>	0.715 <sup>***</sup>

\*\*\* Weighted averages in which the different sizes of the heats have been taken into account.

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TABLE VI

WHITE ZONE

<u>Type</u>	<u>No. of Heats</u>	<u>Average Density</u>	<u>Average Δih (effective)</u>
KS	16	1.708	0.22
KSO	2	1.698	0.16
KCO	28	1.675	0.29
KCN	8	1.665	0.21
CS	13	1.671	0.22
Total	67	1.687**	0.238**
W-4 plus white	91	1.687**	0.281**

TABLE VII

BLUE ZONE

<u>Type</u>	<u>No. of Heats</u>	<u>Average Density</u>	<u>Average Δih (effective)</u>
CS	67	1.662	0.13
KS	7	1.704	0.12
CSO	27	1.657	0.12
KSO	10	1.705	0.09
KCO	9	1.670	0.10
KCN	22	1.712	0.12
Total	142	1.666**	0.121**

TABLE VIII

RED ZONE

<u>Type</u>	<u>No. of Heats</u>	<u>Average Density</u>	<u>Average Δih (effective)</u>
CS	24	1.668	0.03
CSO	8	1.664	0.01
CSN	39	1.664	-0.07
KCO	3	1.660	-0.05
KCN	14	1.670	-0.09
KS	2	1.703	-0.05
KSO	4	1.702	-0.09
KSN	6	1.710	-0.17
Total	100	1.672**	-0.030**

\*\*Weighted averages in which the different sizes of the heats have been taken into account.

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Estimated Initial Reactivity of Pile

The reactivity of the DR pile at startup can be estimated from the functional tests on the graphite used to build it. Weighting factors for the different zones are necessary for this calculation. These factors are taken from calculations made for simply shaped piles such as spheres, cubes, and infinite cylinders<sup>9</sup>. The zones have the same shapes as the piles themselves. The weighting factor of a central zone is expressed as a function of the ratio of the size of the zone to the size of the entire pile.

Two complications arise in attempting to obtain weighting factors for the zones of the DR pile. The first is that the DR pile is a rectangular parallel-piped and therefore does not fit any of the special cases exactly. It is apparent from the graphs used, however, that the factors for a cubic pile of the same volume as DR are sufficiently accurate. The second complication lies in the fact that the zones of DR are not of the same shape as the pile itself. Since the pile is made of blocks four feet long laid with the end of any one bar adjacent to the centers of the two parallel bars on either side, the boundaries of the zones are necessarily very irregular. The effect of the imperfect boundaries is to reduce the importance of the green zone slightly, since some of this high quality material extends into regions where its effectiveness is reduced. Similarly, the importance of the red zone is increased.

Calculated weighting factors for ideal geometry zones and estimated values for the actual zones in DR are shown in Table IX. The estimated contribution of each zone to the total  $\Delta ih$  of the entire pile is also shown.

TABLE IX

Zone	Percentage of Graphite Included	Weight Factors		Weighted $\Delta ih$	Excess Inhours*
		Ideal	Estimated Actual		
Green	12.46	.735	.70	0.662	475
White	24.72	.171	.17	0.048	35
Blue	45.94	.092	.11	0.013	9
Red	16.88	.002	.02	-0.001	- 1
Total				0.722	518

\* In calculating these values 1% K was assumed equivalent to 417 inhours.

A decrease in reactivity of the test pile of one inhour was obtained by adding 1.70 cm<sup>2</sup> of thermal absorption cross-section, in the form of cadmium wire, to two central graphite test blocks. This amount of absorber per pair of blocks has been calculated to decrease the reproduction factor by 1.72 percent. Thus a  $\Delta ih$  of one inhour in the test pile is equivalent to 1.72% K in a 105 pile. A value of 417 inhours for 1% change in K was obtained during the startup of the B, D, and F piles by combining calculated increases in K (due to loading additional tubes) with measured increases in reactivity<sup>10</sup>. More recently, an indirect method involving measurements of the yields and half lives of delayed neutrons from fission of U<sup>235</sup> produced a value of 385 ± 30 inhours for 1% K<sup>11</sup>.

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If the value of 417 is used, DR is calculated to have a reactivity 518 inhours greater than a pile made of graphite with average  $\Delta ih$  equal to zero. If the 385 value is used, however, the calculated number of excess inhours is reduced to 478.

The weighted average  $\Delta ih$  of graphite used in the D and F piles (records are incomplete on B pile) is + 0.15. Thus DR pile exceeds the average of D and F by 0.572 in effective average  $\Delta ih$  or 0.98% K. If 1% K corresponds to 417 inhours, then DR should have a reactivity 410 inhours greater than the average of D and F at startup. If 385 inhours for 1% K is correct, then DR should have 378 inhours more than the average of D and F.

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