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THERMAL ANNEALING OF NEUTRON INDUCED
DISCOMPOSITION IN ARTIFICIAL GRAPHITE.
II. ASYMPTOTIC ANNEALING EXPERIMENTS

By
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July 7, 1950

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2

ARGONNE NATIONAL LABORATORY
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THERMAL ANNEALING OF NEUTRON INDUCED DISCOMPOSITION
IN ARTIFICIAL GRAPHITE

II. ASYMPTOTIC ANNEALING EXPERIMENTS

Report written
by
T. J. Neubert

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Work done by: Aaron Novick, A. R. Van Dyken,
J. Royal, R. C. Boe, W. R. Burns,
H. Petrzilka, E. H. Phillips,
E. H. Smith, G. Steel, M. Stewart,
L. Weil and T. J. Neubert

CHEMISTRY DIVISION

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ABSTRACT

An attempt has been made to discover the mechanism for the neutron induced discomposition of graphite by means of experiments in which irradiated specimens were heated for long periods at successively higher temperatures. The properties studied were the electrical resistivity and elastic modulus. The measurements were designed to show

- (a) the effect of the amount of irradiation,
- (b) the differences between the various types of pile graphite,
- (c) the dependence upon the extrusion-produced crystallite orientation,
- (d) the dependence upon degree of graphitization and crystallite size.

Extension of annealing experiments to 2000°C is described. It was found that whereas annealing at temperatures less than 1000°C caused the electrical resistivity effect and the elastic modulus effect to decrease, annealing in the region 1100° to 1330°C caused the electrical resistivity effect to increase. Annealings above 1350°C caused the electrical resistivity effect to decrease again until at ca. 2000°C the specimens exhibited their prebombardment resistivities.]

1. Introduction

A previous paper has discussed the thermal annealing of the fast neutron induced changes in graphite from the viewpoint of rate of annealing experiments.⁽¹⁾ In most cases, when irradiated graphite samples are heated at temperatures higher than the irradiation temperature, the neutron induced

increases in electrical resistivity⁽²⁾ and elastic modulus⁽³⁾ are seen to decrease. The changes, upon heating at constant temperature, are rapid in the early stages of heating, but as the heating is continued the rate of change decreases until times are reached after which further changes are difficult to measure. The rapid initial decrease and the long slow decrease are seen each time the heating is repeated at a new higher temperature.

In order to be able to make quick crude appraisals of the character of the neutron induced disturbances many annealing experiments were performed in which the limiting residual neutron induced change, after a long-time anneal at constant temperature, was investigated as a function of temperature. These experiments have been called "asymptotic annealing experiments." Strictly speaking, neutron induced changes should be expected to anneal completely at any temperature, but practical experimental problems of precision of measurement and time of heating make asymptotic experiments necessary.

The present paper discusses asymptotic annealing experiments which had for their purpose determination of the dependence of the Wigner effect on the extent of neutron bombardment, the type of artificial graphite, the orientation of specimen axis relative to the axis of extrusion, and the degree of graphitization.

2. Description of Experimental Techniques

In all the experiments to be discussed in this paper, the residual neutron induced effects were determined by measurements at room temperature. Such measurements, of electrical resistivity and elastic modulus, have been described in earlier papers.^(2,3)

For temperatures up to 303°C samples were heated, in groups of approximately thirty, in a graphite block. The graphite block had holes bored in it; each hole held one sample; and the hole diameter was chosen so as to assure good contact between the sample and the graphite block. This graphite magazine, containing the samples, was then slipped into a tight fitting nickel tube (ca. 2" O.D. x 1/32" wall x 20" long) closed at the lower end. The assembly was then heated by immersing in a boiling liquid bath which was heated electrically. Glass wool packed into the nickel tube above the sample magazine decreased convection losses and a closure and pumping system at the outer upper end was provided so that, for heating at temperatures of 230°C and above, the system could be evacuated and filled with helium. Table I lists the boiling baths used for the various temperatures and gives the approximate time of heating at each temperature. The entries for the anneal at 37°C have been included for orientation purposes only. No annealing experiments at room temperature will be described in this paper; see, however, the preceding paper of this series.⁽¹⁾

Table I

Summary of heating arrangements for the asymptotic annealing experiments

Temp. °C	Approximate Heating Time	$E_{\max}$ (ev)	Heating Arrangement	Atmosphere
(37)	(400 days)	(1.27)	(room temperature)	(air)
131	230 hrs.	1.52	boiling isoamyl alcohol	air
156	130 hrs.	1.59	boiling bromobenzene	air
183	85 hrs.	1.67	boiling o-dichlorobenzene	air
207	75 hrs.	1.76	boiling tetralin	air
230	50 hrs.	1.83	boiling butyl carbitol	helium
260	50 hrs.	1.93	boiling Dowtherm	helium
303	30 hrs.	2.07	boiling diphenylamine	helium
450	3 hrs.	2.45	electric furnace	helium
1000	20 min.	4.07	electric furnace	helium

For temperatures above 303°C samples were heated in two smaller graphite magazines in an electric furnace. In order to decrease oxidation, an atmosphere of helium was used. The temperatures and approximate annealing times are listed in Table I. For temperatures above 1000°C a completely different heating technique was employed. This will be described later, in Section 7.

3. Discussion of Annealing Experiments

Samples were heated at each annealing temperature for such times that the rate of healing was practically insignificant at the time heating was discontinued. Times of heating were chosen in accordance with the rate of annealing experiments described in the previous paper⁽¹⁾ of this series. Figure 1 displays typical data for two samples of approximately the same amount of irradiation (ca. 3.7 L-units). The abscissa gives the annealing temperature; the ordinate gives the residual fractional increase in electrical resistivity, r_T , and elastic modulus, e_T , i.e.,

FIG. 1. TYPICAL ASYMPTOTIC ANNEALING DATA FOR TWO SAMPLES OF PARALLEL-CUT AGOT-K GRAPHITE; IRRADIATED CA. 3.7 L-UNITS AT $T = 32 \pm 5^\circ\text{C}$.

$$r_T = (R_T - R_0)/R_0 \quad \text{and} \quad e_T = (E_T - E_0)/E_0 \quad (1)$$

where the subscript T indicates that the value has been obtained by measurement after a long time anneal at temperature T, and the subscript 0 indicates the initial pre-irradiation value of the appropriate physical property. Occasionally the designations r_x and e_x will be used to indicate the fractional increases in electrical resistivity and elastic modulus for irradiated samples (as received from the pile) prior to any annealing; the designation r_M and e_M will be used to indicate the residual fractional increase (as defined in equation 1) after an anneal at 1000°C.

Figure 1 is typical of all the asymptotic data to be reported. It can be seen that the electrical resistivity effect is particularly sensitive to annealing at low temperature whereas the principal decrease in the elastic modulus effect occurs at relatively higher temperatures. After about 400°C there is little further change in either electrical resistivity or elastic modulus. The significant decreases in the region of temperatures up to 400°C are usually considered to be related to the removal by healing to edges, cracks, or lattice vacancies (holes) of the interstitial disturbances produced during neutron irradiation. According to such a model, the residual effects at 1000°C are the result of holes.

It is not necessary to presume that only holes remain after a 1000°C anneal. It is reasonable that the interstitial disturbances are C-atoms and/or C₂-molecules, existing singly and/or in clusters and that the holes are single lattice vacancies and/or groups of adjacent lattice vacancies. Thus, many "mechanisms" can be conceived. So far no really satisfactory model for the Wigner effect has been invented. The problem would be simpler if there were reasons to expect some kind of interstitial complex to be stable at 1000°C, but even if such were the case the differences between the behaviors of the electrical resistivity and the elastic modulus would be difficult to rationalize.

Further examination of Figure 1 indicates reasonably good agreement between the results for the two samples. The way in which the points for the two samples are distributed suggests that sample KP-367 was irradiated somewhat more than sample KP-316 and at a slightly higher temperature. Considering either set of points it is seen that the electrical resistivity curve (solid line) and the elastic modulus curve (dashed line) could be drawn with bends and kinks such that the lines would pass through all points. There would, furthermore, be quite good correlation between the positions of the wiggles in the curves for both electrical resistivity and elastic modulus. In reporting the present data, however, all such effects have been disavowed because of the imprecisions of measurement of physical properties, the temperature fluctuations during annealing, the dependence of results on time of heating, the sample to sample variation always troublesome in the graphite work and because of uncertainties about bombardment conditions. The precision of measurement of electrical resistivity and elastic modulus is such

that the residual fractional increases are considered to be reliable to ± 0.01 in most cases, leaving deviations larger than this to be explained otherwise.

From the discussion of the rates of annealing of neutron induced changes in irradiated graphite,⁽¹⁾ it has been possible to compute distribution functions $R_0(E)$ and $E_0(E)$ for the electrical resistivity and elastic modulus effects which describe the concentration and importance of disturbances in changing the properties of graphite as a function of their activation energy for thermal annealing. The computations involve several crude approximations, principally that of a first-order-rate equation for the healing process, but nonetheless the distribution spectra are useful for recording some of the differences between the electrical resistivity and elastic modulus effects.

Figure 2 shows the distribution spectra for the thermal annealing of disturbances in AGOT-K graphite irradiated 10 L-units at about 35°C. The solid curves $R_0(E)$ and $E_0(E)$ are shown decomposed into two curves each (dotted curves). The disturbances appear to fall into two principal classes; those with characteristic activation energy 1.4 ev seem to be most important so far as electrical resistivity effects are concerned, while those with activation energy 1.8 ev are more important in changing the elastic modulus. Each of these principal kinds of disturbance, cf. curves R_R and E_E , has however a certain effect on the other property of which it is not most characteristic, cf. curves R_E and E_R .

Asymptotic annealing experiments give integrals of the distribution functions shown in Figure 2. Thus

$$r_T = \int_{E'}^{\infty} R_0(E) dE \quad \text{and} \quad e_T = \int_{E'}^{\infty} E_0(E) dE, \quad (2)$$

where

$$E' = kT \ln At. \quad (3)$$

These relationships state that as a specimen is annealed the disturbances heal in order of their characteristic activation energies so that after an anneal for t seconds at T degrees Kelvin all disturbances of energy less than E' ev have disappeared. (k is Boltzman's constant, 8.62×10^{-5} ev deg⁻¹ and A is a frequency factor taken to be ca. 10^{13} sec⁻¹.)

Using the distribution functions of Figure 2 and the times and temperatures of heating listed in Table I, the integrals of equation (2) were calculated. Values of E' , the lower integration limit, are included in Table I. The results, shown in Figure 3, have been plotted against annealing temperature in order to emphasize the analogical equivalence to the data and curves of Figure 1. The bumps and wiggles discussed previously, in connection with Figure 1, are not evident in Figure 3, because the R_R peak is so large compared

FIG. 2. DISTRIBUTION SPECTRA FOR THE THERMAL ANNEALING OF DISTURBANCES IN IRRADIATED AGOT-K GRAPHITE (10 L-units).

FIG. 3. FORM OF ASYMPTOTIC ANNEALING CURVES
COMPUTED BY INTEGRATION OF $R_0(E)$
AND $E_0(E)$ DISTRIBUTION FUNCTIONS.

with the R_E peak, and because the E_E and E_R peaks are so nearly the same size.

4. Dependence of Neutron Induced Effects on the Amount of Irradiation

Table II summarizes the available data of asymptotic annealing experiments on samples of parallel-cut AGOT-K graphite with varying amounts of neutron irradiation. Specific details of sample history have been entered in an appendix, Table X. The samples used were bombarded either in the Clinton pile or in the CP-3 pile. Samples were, for the most part, irradiated at temperatures close to room temperature. However, in order to take maximum advantage of the data at hand, it was necessary to include some samples for which the irradiation temperature was above the usual temperature for a part of the irradiation. Irradiation magnitudes, in L-units, were assigned by means of the curve for elastic modulus change versus bombardment for parallel-cut AGOT-K samples irradiated in the CP-3 pile.⁽⁴⁾

Figures 4, 5, 6, and 7 indicate the results of the annealing experiment plotted as functions of the temperature of annealing and amount of neutron irradiation. Plotted points are not shown because of insurmountable difficulties in designating the points for various samples and because there is a considerable scattering. Instead an attempt was made to prepare self-consistent curves which would depict the trends but at the same time would not violate the experimental numbers too seriously. Figures 4, 5, 6, and 7 are such curves. In order to demonstrate the agreement or lack of agreement between the curves and the experimental data, two sets of numbers have been entered in Table II. The (a) entries are the results of measurements; the (b) entries are appropriate values read from the original graphs of Figures 4, 5, 6, and 7 before they were redrawn and reduced in size. The estimated precision in the residual fractional increase is about ± 0.01 . The (a) and (b) entries of Table II are for the most part consistent with this standard of precision. Some samples, for example KP-428, show large deviations between the (a) and (b) entries for the electrical resistance effect at the lower temperatures, indicating that the irradiation temperature was higher than usual. The deviations in the electrical resistivity values for two samples, KP-185 and KP-212, are probably attributable to bad measurements of the initial electrical resistivity, i.e., R_0 .

The trends in the asymptotic curves in Figures 4 and 6 are the same as those in Figure 1. In fact, Figure 1 and curve 6 of Figures 4 and 6 are equivalent. Curves 1 through 5 are the data for the first five samples in Table II; curves 6 through 9 are averaged results of pairs of samples having nearly the same amount of irradiation, i.e., curve 6: KP-316 and KP-367; curve 7: KP-380 and KP-222; curve 8: KP-171 and KP-419; curve 9: KP-428 and KP-184. Samples KP-185 and KP-212 have been omitted.

Table II

Asymptotic annealing data for parallel-cut AGOT-K graphite samples with various amounts of fast neutron irradiation

($\bar{r}$ and $\bar{e}$ denote the residual fractional increases in electrical resistivity and elastic modulus. The entries, $\bar{a}$, are the experimental results, the entries, $\bar{b}$, are values self-consistent with Figures 4, 5, 6, and 7.)

Sample	Irrad. L-units		As rec'd		131°C		156°C		183°C		207°C		230°C		260°C		303°C		450°C		1000°C	
			r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e
KP-273	0.35	a	0.052	0.098	0.014	0.100	0.011	0.102	0.007	0.098	0.006	0.096	0.008	0.087	0.005	0.080	0.006	0.055	0.006	0.029	0.012	0.014
		b	0.060	0.100	0.013	0.100	0.010	0.100	0.007	0.100	0.006	0.099	0.005	0.095	0.004	0.084	0.004	0.048	0.004	0.019	0.004	0.007
KP-257	0.47	a	0.081	0.137	0.019	0.139	0.019	0.139	0.015	0.129	0.004	0.121	0.010	0.100	0.006	0.092	0.005	0.060	0.002	0.023	0.006	0.002
		b	0.080	0.137	0.019	0.134	0.015	0.132	0.012	0.129	0.010	0.124	0.009	0.116	0.007	0.097	0.006	0.060	0.006	0.020	0.006	0.008
KP-219	0.80	a	0.141	0.214	0.036	0.186	0.035	0.188	0.033	0.172	0.011	0.153	0.016	0.129	0.027	0.106	0.011	0.064	0.017	0.021	0.016	0.012
		b	0.130	0.214	0.039	0.198	0.028	0.190	0.021	0.175	0.017	0.156	0.016	0.136	0.013	0.110	0.012	0.066	0.011	0.026	0.011	0.010
KP-240	1.33	a	0.212	0.302	0.089	0.258	0.055	0.247	0.044	0.228	0.033	0.195	0.036	0.167	0.036	0.139	0.023	0.081	0.021	0.032	0.025	0.016
		b	0.204	0.302	0.068	0.263	0.054	0.247	0.042	0.225	0.036	0.196	0.030	0.165	0.026	0.133	0.022	0.079	0.020	0.032	0.020	0.013
KP-305	2.00	a	0.322	0.383	0.103	0.314	0.085	0.295	0.075	0.267	0.055	0.228	0.054	0.184	0.052	0.147	0.040	0.088	0.036	0.036	0.041	0.012
		b	0.292	0.383	0.103	0.327	0.084	0.306	0.070	0.275	0.060	0.238	0.052	0.300	0.047	0.152	0.030	0.086	0.029	0.039	0.028	0.016
KP-185	3.13	a	0.398	0.494	0.132	0.406	0.111	0.385	0.094	0.351	0.080	0.270	0.069	0.220	0.072	0.138	0.048	0.062	0.032	0.020	0.031	0.013
		b	0.425	0.494	0.156	0.412	0.132	0.380	0.114	0.343	0.098	0.282	0.086	0.234	0.076	0.176	0.062	0.093	0.057	0.045	0.055	0.021
KP-316	3.70	a	0.491	0.544	0.163	0.464	0.148	0.431	0.123	0.379	0.104	0.295	0.098	0.238	0.087	0.180	0.069	0.087	0.056	0.045	0.056	0.025
		b	0.490	0.544	0.180	0.451	0.153	0.415	0.131	0.356	0.115	0.305	0.098	0.246	0.089	0.177	0.073	0.099	0.066	0.047	0.062	0.022
KP-367	3.75	a	0.465	0.547	0.192	0.480	0.164	0.439	0.143	0.398	0.119	0.328	0.116	0.259	0.103	0.205	0.083	0.108	0.066	0.059	0.068	0.029
		b	0.494	0.547	0.183	0.452	0.155	0.416	0.133	0.357	0.116	0.306	0.099	0.247	0.090	0.178	0.074	0.099	0.066	0.047	0.062	0.022
KP-212	4.33	a	0.545	0.594	0.177	0.475	0.151	0.434	0.132	0.389	0.110	0.316	0.087	0.262	0.094	0.198	0.061	0.102	0.043	0.051	0.038	0.026
		b	0.555	0.594	0.206	0.489	0.176	0.450	0.153	0.402	0.130	0.327	0.116	0.268	0.106	0.197	0.088	0.102	0.074	0.051	0.069	0.025
KP-380	5.30	a	0.647	0.662	0.222	0.556	0.201	0.498	0.175	0.446	0.142	0.352	0.138	0.282	0.134	0.218	0.129	0.108	0.111	---	0.099	0.030
		b	0.652	0.662	0.250	0.548	0.214	0.503	0.182	0.446	0.160	0.358	0.143	0.290	0.125	0.213	0.107	0.107	0.089	0.057	0.080	0.029
KP-222	5.30	a	0.632	0.662	0.248	0.544	0.217	0.491	0.192	0.438	0.153	0.337	0.142	0.285	0.138	0.215	0.097	0.100	0.084	0.057	0.076	0.031
		b	0.652	0.662	0.250	0.548	0.214	0.503	0.182	0.446	0.160	0.358	0.143	0.290	0.125	0.213	0.107	0.107	0.089	0.057	0.080	0.029
KP-263	6.35	a	0.688	0.730	0.278	0.602	0.244	0.552	0.213	0.484	0.174	0.377	0.157	0.313	0.149	0.235	0.117	0.113	0.094	0.065	0.079	0.030
		b	0.752	0.730	0.289	0.605	0.247	0.555	0.213	0.489	0.191	0.390	0.167	0.314	0.148	0.226	0.126	0.113	0.102	0.060	0.088	0.031
KP-171	7.55	a	0.778	0.801	0.324	0.665	0.277	0.600	0.242	0.532	0.196	0.402	0.183	0.328	0.172	0.241	0.130	0.107	0.100	0.058	0.082	0.035
		b	0.861	0.801	0.338	0.665	0.289	0.609	0.249	0.534	0.219	0.424	0.195	0.339	0.172	0.240	0.148	0.120	0.117	0.067	0.102	0.034
KP-419	8.00	a	0.853	0.823	0.370	0.709	0.320	0.640	0.280	0.580	0.239	0.451	0.215	0.351	0.202	0.262	0.167	0.130	0.134	0.074	0.123	0.038
		b	0.900	0.823	0.356	0.686	0.304	0.628	0.262	0.550	0.231	0.437	0.205	0.348	0.182	0.245	0.156	0.122	0.123	0.070	0.107	0.036
KP-428	11.8	a	1.013	1.006	0.480	0.835	0.418	0.775	0.379	0.685	0.306	0.498	0.291	0.406	0.269	0.281	0.215	0.124	0.170	0.084	0.136	0.071
		b	1.180	1.006	0.509	0.843	0.434	0.779	0.376	0.685	0.326	0.510	0.287	0.425	0.263	0.287	0.225	0.143	0.174	0.086	0.145	0.047
KP-184	13.7	a	1.116	1.086	0.581	0.945	0.506	0.849	0.419	0.751	0.370	0.596	0.335	0.465	0.316	0.350	0.264	0.206	0.204	0.146	0.177	0.078
		b	1.206	1.086	0.587	0.926	0.503	0.855	0.437	0.758	0.372	0.596	0.322	0.468	0.304	0.310	0.262	0.155	0.198	0.095	0.165	0.053

13

FIG. 4. DEPENDENCE OF THE RESIDUAL FRACTIONAL INCREASE IN ELASTIC MODULUS, e_T , UPON ANNEALING TEMPERATURE, T , FOR PARALLEL-CUT AGOT-K GRAPHITE WITH VARIOUS AMOUNTS OF IRRADIATION.

FIG. 5. DEPENDENCE OF THE RESIDUAL FRACTIONAL INCREASE IN ELASTIC MODULUS, e_T , UPON THE AMOUNT OF IRRADIATION, IN L-UNITS, FOR VARIOUS ANNEALING TEMPERATURES.

FIG. 6. DEPENDENCE OF THE RESIDUAL FRACTIONAL INCREASE IN ELECTRICAL RESISTIVITY, r_T , UPON ANNEALING TEMPERATURE, T , FOR PARALLEL-CUT AGOT-K GRAPHITE WITH VARIOUS AMOUNTS OF IRRADIATION.

FIG. 7. DEPENDENCE OF THE RESIDUAL FRACTIONAL INCREASE IN ELECTRICAL RESISTIVITY, r_T , UPON THE AMOUNT OF IRRADIATION, IN L-UNITS, FOR VARIOUS ANNEALING TEMPERATURES.

Examination of Figures 4 and 6 shows that while the elastic modulus effect in samples with low bombardments is quite unaffected by annealing temperatures below 200°, the electrical resistivity effect is very nearly completely removed by the time that temperature is reached. As the amount of irradiation is increased, however, the elastic modulus effect becomes more sensitive with respect to low temperature annealing and the electrical resistivity effect becomes less sensitive. These differences are demonstrated in another way in Figures 5 and 7.

It should be noted that the curves of residual electrical resistivity effect versus bombardment, Figure 7, exhibit a slight concavity in the region of small bombardment. The available data are, however, not of sufficient precision to make this concavity certain, but if true, it would not be unreasonable to suggest that some of the disturbances responsible for the electrical resistivity effect are complex disturbances, involving more than one carbon atom. Correspondingly, the shape of the e_T curves, Figure 5, might indicate that the elastic modulus effect is caused by the primary disturbances produced by neutron impacts, i.e., C-atoms. It is also possible to presume that whereas the effect of disturbances on elastic modulus is always such as to increase the elastic modulus, different kinds of disturbances effect the electrical resistivity either positively or negatively. Thus, for a suitably devised model, if the rates and activation energies of thermal annealing were properly chosen, bombardment and annealing curves could be calculated which would be concave for small bombardments.

5. Dependence of Neutron Induced Effects on the Type of Artificial Graphite and upon the Orientation of the Specimen Axis Relative to the Axis of Extrusion

Asymptotic annealing experiments have been carried out on samples of each of the three pile graphites AGOT-K, AGOT-C and AGOT-W. The preparation and properties of these graphites have been described in an earlier paper.⁽⁵⁾ Table III summarizes the data for several sets of samples of the three artificial graphites, both parallel-cut and perpendicular-cut. In the sample designation, the first letter indicates the kind of graphite; the second letter indicates the orientation of the specimen axis, relative to the axis of extrusion of the stock bar, P denoting parallel-cut, N denoting perpendicular-cut. Thus KN-166 is the 166th AGOT-K sample and was cut with its axis perpendicular to the extrusion axis.

Examination of the data of Table III has revealed no particularly significant differences between K, C and W graphites, other than those which can be dismissed on the basis of the known differences in behavior under irradiation. It has been found that, to within the precision with which observations can be made, the neutron induced effect, for any graphite, is related to the effect in AGOT-K graphite by a multiplicative constant.⁽⁴⁾

Table III

Summary of asymptotic annealing data for irradiated samples of AGOT-K, AGOT-C and AGOT-W artificial graphite

(Sample designations K, C, or W denote the kind of graphite, and P or N denotes the orientation of the specimen axis relative to the axis of extrusion, i.e., parallel-cut and perpendicular-cut, respectively.)

Sample	Irrad. L-units	Irrad. Temp.	As rec'd		131°C		156°C		183°C		207°C		230°C		260°C		303°C		450°C		1000°C	
			r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e
KP-428	12	~100°C	1.013	1.006	0.480	0.835	0.418	0.775	0.379	0.685	0.306	0.498	0.129	0.406	0.269	0.281	0.215	0.124	0.170	0.084	0.136	0.071
CP-130	12	~100°C	0.930	0.964	0.585	0.851	0.513	0.772	0.465	0.654	0.397	0.485	0.368	0.440	0.353	0.311	0.280	0.161	0.227	0.110	0.190	0.079
WP-150	12	~100°C	0.939	0.818	0.557	0.717	0.497	0.651	0.447	0.558	0.380	0.451	0.343	0.371	0.333	0.269	0.271	0.137	0.222	0.102	0.200	---
KP-380	5.3	~ 25°C	0.647	0.662	0.222	0.556	0.201	0.498	0.175	0.446	0.142	0.352	0.138	0.228	0.134	0.218	0.129	0.108	---	---	0.063	0.030
CP-117	5.3	~ 25°C	0.604	0.581	0.213	0.465	0.192	0.441	0.176	0.393	0.145	0.300	0.134	0.250	0.131	0.187	0.098	0.098	0.084	0.050	0.072	0.033
WP-133	5.3	~ 25°C	0.578	0.534	0.209	0.451	0.175	0.420	0.157	0.378	0.135	0.287	0.118	0.237	0.118	0.180	0.087	0.104	0.070	0.072	0.065	0.041
KP-300	5.0	~115°C	0.341	0.672	0.252	0.652	0.216	0.610	0.177	0.527	0.156	0.403	0.148	0.378	0.129	0.285	0.102	0.150	0.070	0.097	0.051	0.072
CP-108	5.0	~115°C	0.332	0.585	0.279	0.557	0.240	0.513	0.219	0.452	0.186	0.341	0.169	0.296	0.161	0.217	0.122	0.112	0.106	0.074	0.091	0.039
WP-128	5.0	~115°C	0.312	0.507	0.260	0.467	0.227	0.446	0.202	0.401	0.173	0.301	0.157	0.252	0.155	0.184	0.122	0.100	0.095	0.068	0.091	0.038
KM-459	3.1	~ 25°C	0.399	0.584	0.126	0.492	0.102	0.461	0.090	0.410	0.074	0.332	0.073	0.267	0.072	0.200	0.058	0.105	0.043	0.058	0.032	0.067
CN-211	3.1	~ 25°C	0.348	0.433	0.114	0.360	0.088	0.346	0.081	0.301	0.065	0.234	0.072	0.193	0.064	0.151	0.047	0.083	0.030	0.046	0.028	0.035
WN-181	3.1	~ 25°C	0.339	0.382	0.122	0.322	0.093	0.312	0.087	0.299	0.083	0.212	0.087	0.165	0.076	0.152	0.061	0.112	0.038	0.071	0.034	0.031
KP-367	3.8	~ 35°C	0.465	0.547	0.192	0.480	0.164	0.439	0.143	0.398	0.119	0.328	0.116	0.259	0.103	0.205	0.083	0.108	0.066	0.059	0.068	0.029
KN-166	3.8	~ 35°C	0.462	0.681	0.179	0.590	0.168	0.531	0.148	0.482	0.114	0.392	0.117	0.302	0.114	0.236	0.085	0.123	0.068	0.066	0.073	0.042

Table IV summarizes these multiplicative constants and the initial elastic modulus and electrical resistivity for various graphites. In the present annealing experiments the curves obtained for K, C, and W graphite seem to be superposable if first properly normalized using the factors given in Table IV.

Table IV

Summary of relative neutron induced changes
in various graphites irradiated at ca. 30°C

(E_0 = initial elastic modulus, R_0 = initial electrical resistivity,
 $e_i = \Delta E/E_0$, $r_i = \Delta R/R_0$ for any graphite; the subscript k denotes
that the change is that for AGOT-K(II) graphite.)

Designation	E_0 (10^{10} dynes/cm ²)	e_i/e_k	R_0 (10^{-4} ohm cm)	r_i/r_k
AGOT-K(I) ()	12.9	0.96 ± 0.02	6.5	0.89 ± 0.01
AGOT-K(II) ()	13.7	1.000	7.8	1.000
(⊥)	5.7	1.24 ± 0.05	10.5	0.96 ± 0.02
AGOT-C(II) ()	10.6	0.91 ± 0.03	7.6	0.92 ± 0.02
(⊥)	6.4	0.92 ± 0.03	11.9	0.75 ± 0.04
AGOT-W(II) ()	10.0	0.79 ± 0.03	8.4	0.86 ± 0.02
(⊥)	6.6	0.79 ± 0.02	11.8	0.78 ± 0.01
Sp 3000 ()	7.0	1.03 ± 0.03	9.4	0.85 ± 0.03
(⊥)	5.4	1.09 ± 0.01	11.4	0.83 ± 0.02
Sp 2400 ()	6.6	0.50 ± 0.02	21.8	0.25 ± 0.01
(⊥)	5.5	0.61 ± 0.03	21.7	0.35 ± 0.02
Sp 2100 ()	6.4	0.23 ± 0.03	37.7	0.00 ± 0.01
(⊥)	4.9	0.24 ± 0.03	44.9	0.00 ± 0.01
PBNG	15.6	1.16 ± 0.09	9.7	0.61 ± 0.02
PBL	5.8	0.24 ± 0.03	65.3	0.07 ± 0.01

The differences in behavior under annealing between parallel-cut (KP) and perpendicular-cut (KN) specimens of irradiated AGOT-K graphite are also describable in terms of the multiplicative constants of Table IV. The data, for all the samples measured, are listed in Table III. Figure 8 shows the results for two samples, KP-367 and KN-166, irradiated for 3.8 L-units at about 35°C.

FIG. 8. ASYMPTOTIC ANNEALING DATA FOR ONE SAMPLE OF PARALLEL-CUT AND ONE SAMPLE OF PERPENDICULAR-CUT AGOT-K GRAPHITE IRRADIATED 3.8 L-UNITS AT CA. 35 °C.

6. Dependence of Neutron Induced Effects on the Degree of Graphitization and Upon Crystallite Size

Table V summarizes the available data for asymptotic annealing experiments on irradiated special-graphites and pseudo-graphites which differ in degree of graphitization and in crystallite size. The various samples are as follows:

- (a) QP-12 and QN-23 were cut from AGOT-W stock which had been graphitized at 3000°C (Q = quite graphitized).
- (b) PP-11 and PN-22 were cut from AGOT-W stock which had been fired at 2400°C (P = partly graphitized).
- (c) JP-10 and JN-23 were cut from AGOT-W stock which had been fired at 2100°C (J = just a little graphitized).
- (d) PBNG-116 was cut from a block of pseudo-graphite made of natural graphite bonded with pitch and fired at 3000°C.
- (e) PBL-103 was cut from a block of pseudo-graphite made of lamp-black bonded with pitch and fired at 3000°C.

The special graphites Q, P, and J differ in the degree of graphitization; the differences in the asymptotic annealing curves are shown in Figure 9. The pseudo-graphites PBNG and PBL differ in crystallite size; the differences in behavior under asymptotic annealing are shown in Figure 10.

7. Description of High Temperature Experiments

In discussion of annealing experiments Franck at one time suggested that, if in healing to edges, an interstitial C-atom required a smaller activation energy than would be required for it to force an entry into a hole; then it might happen that different 1000°C residuums would obtain for samples heated slowly to 1000°C and for samples heated quickly to 1000°C. To test this idea an apparatus was constructed in which one of the ordinary graphite specimens could be heated to 1000°C in ca. 1/3 second. Comparisons of samples heated to 1000°C in the course of about 45 minutes with samples heated to 1000°C in ca. 1/3 second showed, however, no differences beyond the usual scattering of results. This experiment was, therefore, inconclusive, but the apparatus had been designed also to provide means for investigation of healing in temperature regions above those attainable with ordinary electric furnaces.

For heating samples either quickly, or to high temperatures, the specimen was mounted between two clamps and heated by suitable direct

Table V

Asymptotic annealing data for irradiated special graphites and pseudo-graphites

(Sample designations: Q = graphitized at 3000°C, P = fired at 2400°C, J = fired at 2100°C; the second letters denote whether the sample is parallel-cut (P) or perpendicular-cut (N); PBNG = pitch bonded natural graphite, PBL = pitch bonded lampblack.)

Sample	Irrad. L-units	As rec'd		131°C		156°C		183°C		207°C		230°C		260°C		303°C		450°C		1000°C	
		r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e	r	e
QP-12	3.1	0.377	0.483	0.109	0.437	0.099	0.433	0.083	0.398	0.087	0.381	0.072	0.333	0.068	0.259	0.057	0.155	0.058	0.111	0.033	0.094
QN-23	3.1	0.362	0.552	0.124	0.471	0.095	0.439	0.107	0.401	0.088	0.324	0.092	0.271	0.076	0.216	0.058	0.126	0.057	0.082	0.045	0.065
PP-11	3.1	0.122	0.237	0.051	0.211	0.043	0.200	0.046	0.194	0.038	0.149	0.034	0.125	0.026	0.110	0.021	0.085	0.022	0.057	0.019	0.032
PN-22	3.1	0.139	0.265	0.052	0.221	0.032	0.200	0.039	0.209	0.036	0.155	0.040	0.135	0.028	0.014	0.022	0.083	0.022	0.062	0.018	0.025
JP-10	3.1	0.000	0.078	0.000	0.066	0.000	0.066	0.000	0.063	0.000	0.061	0.000	0.055	0.000	0.053	0.000	0.047	0.000	0.035	0.000	0.030
JN-23	3.1	0.000	0.094	0.000	0.079	0.000	0.075	0.000	0.075	0.000	0.061	0.000	0.047	0.000	0.041	0.000	0.057	0.000	0.043	0.000	0.028
PBNG-116	~18	0.873	1.284	0.436	1.114	0.380	1.031	0.335	0.919	0.290	0.707	0.287	0.639	0.275	0.570	0.228	0.470	0.197	0.349	0.108	0.227
PBL-103	~25	0.080	0.361	0.071	0.332	0.067	0.307	0.060	0.288	0.051	0.246	0.054	0.207	0.048	0.165	0.036	0.150	0.031	0.138	0.010	0.130

FIG. 9. ASYMPTOTIC ANNEALING CURVES FOR PERPENDICULAR-CUT SAMPLES OF SPECIAL GRAPHITES WITH DIFFERING DEGREES OF GRAPHITIZATION; IRRADIATED 3.1 L-UNITS AT $25 \pm 5^\circ\text{C}$.

24

FIG. 10. ASYMPTOTIC ANNEALING CURVES FOR IRRADIATED SPECIMENS OF PITCH BONDED NATURAL GRAPHITE (PBNG), PITCH BONDED LAMPBLACK (PBL), AND PILE GRAPHITE (AGOT-K).

currents from a bank of storage batteries. The clamps were made of graphite and had small recessions accurately machined to fit firmly over the end of the specimen to be heated. The clamps were copper plated and then silver soldered into large threaded pieces of copper which were fastened with good thermal and electrical contact to water cooled current leads. An attempt was made to decrease heat losses along the clamps by having the clamp diameter only about three-fourths of the specimen diameter for a distance of about $3/8$ " between the cup which fitted the sample and the silver solder joint to the threaded copper bar. A platinum radiation shield 1 cm in diameter was provided to decrease radiative losses. The entire heating assembly was designed to fit freely into a cylindrical pyrex envelope (3" in diameter, 20" in length) which could be flushed with helium and then evacuated to about 10^{-4} mm Hg.

The storage batteries were connected and disconnected by means of an automobile starter switch. Currents were adjusted by a water-cooled variable resistor made of 4 ft. of $1/8$ in. O.D. stainless steel tubing. Variability was attained via sliding metal clamps. An ammeter was included in the circuit. The technique in heating a specimen was to adjust the variable resistor to a value commensurate with the voltage and internal resistance of the storage batteries and with the sample temperature desired. The activator switch for the starter was then closed and as the heating occurred the sample temperature was observed with an optical pyrometer. When the temperature reached the desired value, the activator switch was opened. In this way annealings of several seconds were accomplished. Longer times could, of course, be obtained by repetitions.

In the construction of the high temperature apparatus it was discovered that, unless one of the sample clamps was spring loaded, differential expansions would be such as to bend the samples. Unfortunately no records are available of what the temperatures were in these preliminary experiments. In the final apparatus, a simple spring mechanism was included and samples could be heated very nicely and without bending.

8. Results of High Temperature Annealing Experiments

Electrical resistivity and elastic modulus measurements were made on a number of irradiated and unirradiated samples of parallel-cut AGOT-W graphite. Table VI summarizes some of the data for electrical resistivity changes; Table VII summarizes the corresponding elastic modulus data. The electrical resistivity data are given as fractional changes calculated as described previously, i.e., $r_T = (R_T - R_0)/R_0$; the elastic modulus data are given as ratios to the elastic modulus measured after an anneal at 1000°C , i.e., as (E_T/E_M) . Samples designated EP are specimens cut from a bar of AGOT-W which had been irradiated as part of the structure of the Clinton pile (epsilon bar from foil slot 59) for about 20 L-units.

Table VI

Effect of high temperature annealing on the electrical resistivity of AGOT-W graphite

Sample	WP-206	WP-174	EP-11	EP-12	EP-21	EP-22	EP-23	WP-209	WP-286	WP-126	WP-150
L-units irradiation	none	none	~ 20	~ 20	~ 20	~ 20	~ 20	~ 40	~ 35	~ 30	~ 10
r_x (as rec'd)	0.000	0.000	1.189	---	---	---	---	1.540	1.568	1.516	0.939
r_M (1000°C)	0.000	0.000	0.241	0.241	0.241	0.241	0.241	0.434	0.394	0.337	0.200
r_T T (°C)	0.005 1400	0.003 1630	0.261 1140	0.303 1480	0.330 1530	0.248 1650	0.013 2180	0.668 1360	0.521 1580	0.024 1850	0.322 1400
r_T T (°C)	0.009 1600	0.014 2150	0.333 1440	0.167 1600	0.005 2130			0.019 1850	0.005 2025	0.008 2210	0.012 2190
r_T T (°C)			0.314 1460								
r_T T (°C)			0.039 1725								
r_T T (°C)			0.010 2180								

27 Table VII

Effect of high temperature annealing on the elastic modulus of AGOT-W graphite

(The elastic modulus, E , is given in units of 10^{10} dynes cm^{-2} .)

Sample	WP-206	WP-174	EP-11	EP-12	EP-21	EP-22	EP-23	WP-209	WP-286	WP-126	WP-150
L-units irradiation $E_M(1000^\circ\text{C})$	none 10.81	none 10.19	~ 20 9.99	~ 20 9.95	~ 20 9.80	~ 20 10.04	~ 20 9.77	~ 40 11.40	~ 35 10.30	~ 30 10.63	~ 10 10.14
E_T (E_T/E_M) T ($^\circ\text{C}$)	10.60 0.981 1400	10.16 0.997 1630	9.68 0.969 1440	9.51 0.956 1480	9.53 0.972 1530	9.73 0.969 1650	8.68 0.888 2180	10.92 0.958 1360	9.74 0.946 1580	9.63 0.906 1850	10.21 1.007 1400
E_T (E_T/E_M) T ($^\circ\text{C}$)	10.62 0.982 1600	9.51 0.933 2150	9.60 0.961 1460	9.50 0.955 1600	8.81 0.899 2130			10.12 0.888 1850	8.90 0.893 2025	9.20 0.865 2210	8.97 0.885 2190
E_T (E_T/E_M) T ($^\circ\text{C}$)			9.38 0.939 1725								
E_T (E_T/E_M) T ($^\circ\text{C}$)			8.91 0.892 2180								

Inspection of Table VI shows that as irradiated samples are heated to higher and higher temperatures, the residual fractional increase in electrical resistivity passes through a maximum and drops back close to its pre-bombardment value only when the annealing temperatures exceed 1600°C. Unirradiated graphite increases slightly in electrical resistivity upon heating at high temperature, cf. the data of samples WP-206 and WP-174. No such interesting phenomena are seen in the elastic modulus data. Figure 11 shows the values of E_T/E_M , plotted against annealing temperature, for both irradiated samples (solid points) and unirradiated samples (open points). The two lines which have been drawn are intended to indicate that the changes in irradiated samples should be expected to be larger because of the final removal of the neutron induced changes. Whatever disturbances cause the electrical resistivity change to increase upon high temperature annealing seem to have little or no effect on the elastic modulus.

Because of the curious behavior of electrical resistivity further measurements were made. Table VIII summarizes a more exhaustive set of measurements on two EP samples, similar to the EP samples of Tables VI and VII. No elastic modulus measurements were made. In computing the residual fractional increases for EP samples it was necessary to choose some reasonable value for R_0 since the specimens were cut from an already irradiated bar. The value selected in each case was that which would give a r_T of ca. 0.01 for $T > 2000^\circ\text{C}$ and mutually consistent values of r_M (1000°C) for all EP samples. The R_0 values were finally calculated by defining $r_M = 0.241$ for all EP samples. Figure 12 shows the complete asymptotic annealing curve for EP graphite. Table IX gives the averaged low temperature asymptotic annealing data for two EP samples.

The data of Tables VI and VIII have been plotted in Figure 13 in such a way as to indicate the manner in which the asymptotic high-temperature resistivity effect depends upon the amount of irradiation. The maximum at ca. 1330°C is seen to increase with irradiation but no further conclusions are justifiable with the present data.

9. Concluding Remarks

It has not been the intention of this paper to make any definite statements about possible mechanisms for the Wigner effect in irradiated graphite. The data presented are those taken in the course of experimentation which it was hoped might point the way to a suitable model. Such hopes, however, went unrealized. There seem to be few salient facts which are at all clearly established by asymptotic experiments. Some of these may be summarized as follows.

FIG. II. DEPENDENCE OF ELASTIC MODULUS OF AGOT-W GRAPHITE UPON ANNEAL AT TEMPERATURES ABOVE 1000 °C.

Table VIII

30

High temperature asymptotic annealing data for two samples of
AGOT-W graphite, irradiated ca. 20 L-units

Sample EP-39		Sample EP-40	
Temp. (°C)	r_T	Temp. (°C)	r_T
(As rec'd)	1.189	(As rec'd)	1.196
1100	0.241	1047	0.241
1168	0.258	1199	0.297
1272	0.366	1217	0.338
1316	0.398	1268	0.408
1350	0.384	1300	0.414
1388	0.359	1335	0.404
1402	0.360	1358	0.401
1437	0.337	1408	0.387
1489	0.280	1504	0.308
1597	0.146	1586	0.174
1655	0.096	1668	0.094
1724	0.056	1711	0.039
1783	0.055	1858	0.063

31

FIG. 12. ASYMPTOTIC ANNEALING CURVE FOR AGOT-W GRAPHITE IRRADIATED CA. 20 L-UNITS.

Table IX

32

Asymptotic annealing data for the neutron induced electrical resistivity changes in samples EP-11 and EP-12, AGOT-W graphite, irradiated ca. 20 L-units (values given are averaged values)

(R_T = resistance after heating at temperature, T ,
 R_x = resistance "as rec'd," i.e., before any anneal,
 r_T = residual fractional increase in electrical resistivity)

	R_T/R_x	r_T
As Rec'd	1.000	1.189
131°C	0.804	0.756
156°C	0.760	0.666
183°C	0.727	0.594
207°C	0.683	0.497
230°C	0.676	0.484
260°C	0.645	0.416
303°C	0.615	0.351
475°C	0.583	0.283
1000°C	0.561	0.241

1. The electrical resistivity effect is very sensitive with respect to annealing for low-bombarded samples, but becomes progressively less sensitive as the amount of bombardment is increased (Figure 6).

2. The elastic modulus effect is very insensitive with respect to annealing for low-bombarded samples, but becomes progressively more sensitive as the amount of bombardment is increased (Figure 4).

3. The electrical resistivity effect remaining after a 1000°C anneal is relatively large and appears to be increasing approximately linearly with bombardment (Figure 7), while the elastic modulus effect after a 1000°C

FIG. 13. HIGH-TEMPERATURE ASYMPTOTIC ANNEALING CURVES FOR AGOT-W GRAPHITE WITH DIFFERENT AMOUNTS OF IRRADIATION.

anneal is relatively small and gives some evidences of approaching a "saturation value" for high bombardments (Figure 5).

4. The initial slopes of elastic modulus effect versus bombardment (Figure 5) are large and quite independent of temperature while the initial slopes of the corresponding curves for the electrical resistivity effect (Figure 7) are smaller and very temperature-dependent. The concavity of the initial portions of the electrical resistivity curves may be very important.

5. The differences between parallel-cut and perpendicular-cut samples (Table IV and Figure 8) might provide hints as to which kinds of disturbances in irradiated graphite cause positive changes in properties measured and which kinds cause negative changes.

6. That some kinds of disturbances must cause changes with negative coefficients is clear from the high-temperature annealing results (Figure 12 and Tables VI and VII).

This summary is primarily a series of questions from which it can be seen that further work is necessary. In that work it will be important to place heavy emphasis on precision of measurement, on numbers of samples used, on control of bombardment variables and on the choice of the properties to be studied. The present work has indicated the gross annealing behavior of irradiated graphite so far as the electrical resistivity and elastic modulus effects are concerned and that there are no large differences between the three principal (K, C, and W) pile graphites. The differences between graphites having varying degrees of graphitization (Figure 9) and varying crystallite sizes (Figure 10) are large but at the moment do little toward suggesting a suitable model for the "graphite disease." Indeed, all of the models proposed to date fall short of qualitatively describing these differences or the various points summarized above. The "nature" of artificial graphite is not an insignificant factor in the difficulty of constructing an over-all picture of the effects of fast neutron bombardment on graphite.

10. Appendix

35

Table X

Irradiation histories of samples used in asymptotic annealing experiments

Sample designation:

- (a) K, C, and W denote AGOT-K, AGOT-C, and AGOT-W, respectively.
 (b) Q, P, and J denote special graphites, fired at 3000°C (quite well graphitized), 2400°C (partly graphitized) and 2100°C (just slightly graphitized).
 (c) The second letter, P or N, indicates the orientation of the specimen axis relative to the axis of extrusion. P = parallel-cut, N = perpendicular-cut.
 (d) PBNG and PBL denote pitch bonded natural graphite, and pitch bonded lampblack, respectively.

Sample	Irrad. L-units	Irrad. Pile Units	Treatment
KP-273 KP-257 KP-219 KP-240 KP-305 KP-185 KP-316 KP-212	0.35 0.47 0.80 1.33 2.00 3.13 3.70 4.33	3.1 Mwd 5.1 Mwd 9.0 Mwd 14.1 Mwd 24.5 Mwd 32.9 Mwd 42.4 Mwd 56.4 Mwd	Irradiated in hole 1867 of the Clinton pile (8 inches from uranium metal); 2 to 4 feet from the center of the pile; sample temperature $32 \pm 5^\circ\text{C}$.
KP-222 KP-263 KP-171	5.30 6.35 7.55	65.9 Mwd 75.4 Mwd 89.4 Mwd	Same as above, except for an unfortunate increase in temperature (to probably $50 - 70^\circ\text{C}$) for the period of irradiation above ~ 60 Mwd.
KP-419 KP-184	8.00 13.7	33.0 Mwd 73.0 Mwd	Irradiated in the uranium metal "doughnuts" in hole No. 1867 of the Clinton pile; sample temperature, $35 \pm 5^\circ\text{C}$.
KP-428 CP-130 WP-150	12 12 12	70.0 Mwd 70.0 Mwd 70.0 Mwd	Irradiated in the uranium metal "doughnuts" in hole No. 1867 of the Clinton pile; pile conditions were such that sample temperatures were $\sim 100^\circ\text{C}$ for the last 25 Mwd.
KP-380 CP-117 WP-133	5.3 5.3 5.3	126.0 Mwd 126.0 Mwd 126.0 Mwd	Irradiated in the central thimble of the CP-3 pile; sample temperature $25 \pm 5^\circ\text{C}$.
KP-300 CP-108 WP-128	5.0 5.0 5.0	100.0 Mwd 100.0 Mwd 100.0 Mwd	Irradiated in experimental hole No. 21 of the Clinton pile; sample temperature $\sim 115^\circ\text{C}$.
KN-459 CN-211 WN-181	3.1 3.1 3.1	76.6 Mwd 76.6 Mwd 76.6 Mwd	Irradiated in the central thimble of the CP-3 pile; sample temperature $25 \pm 5^\circ\text{C}$.
KP-367 KN-166	3.8 3.8	24.2 Mwd 24.2 Mwd	Irradiated in charging channel No. 2064 of the Clinton pile, within 1 ft. of the center of the pile, and adjacent to one uranium metal slug; temperature $\sim 35^\circ\text{C}$.
QP-12 QN-23 PP-12 PN-23 JP-10 JN-23	3.1 3.1 3.1 3.1 3.1 3.1	76.6 Mwd 76.6 Mwd 76.6 Mwd 76.6 Mwd 76.6 Mwd 76.6 Mwd	Irradiated in the central thimble of the CP-3 pile; sample temperature $25 \pm 5^\circ\text{C}$.
PBNG-116	~ 18	108.0 Mwd	Irradiated in the uranium metal "doughnuts" in hole No. 1867 of the Clinton pile; sample temperature, $35 \pm 5^\circ\text{C}$.
PBL-103	~ 25	178.0 Mwd	Same as above, except for an unfortunate increase in the temperature (to probably $90 - 100^\circ\text{C}$) for the last 25 Mwd.

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36

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