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Removal of Radiation Damage from Graphite by Alternate Reirradiations and Low Temperature Anneals*

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Removal of Radiation Damage from Graphite

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ABSTRACT

Analyses of property changes during reirradiations and anneals show that the technique of alternate reirradiations and anneals can consistently be used to remove radiation damage from graphite. Laboratory experiments, monitoring studies, and reactor height measurements all correlate favorably. It is shown that the recovery for a reirradiation and annealing cycle is independent of irradiation temperature and total damage over wide ranges. The reciprocal of the recovery per cycle is a simple linear function of the exposure between anneals with a slope that is determined by the anneal temperature.

I. INTRODUCTION

The retained property changes in graphite after repeated cycles of reirradiations and anneals were described in other work.[1] It was concluded that alternate reirradiations and anneals can remove radiation damage from graphite. In this investigation the changes in properties that occur during reirradiations and anneals are shown to be consistent with the analysis of the retained property changes.

The dimensional expansion during a reirradiation and the contraction during an anneal were studied as functions of irradiation temperature, anneal temperature, total damage, and exposure between anneals. Experiments in which one of these variables at a time is changed are compared with experiments in which all of the variables change simultaneously. It is shown that the ratio of the growth during a reirradiation (G) to the recovery during an anneal ($|R|$) is independent of both irradiation temperature and retained damage over large ranges. The ratio $G/|R|$ is a linear function of exposure between anneals with a slope that is determined by the anneal temperature. The results are found to be applicable to changes in the Brookhaven reactor (BGRR) in spite of complexity in the irradiation and annealing procedure.

II. EXPERIMENTAL

The type of graphite used in these studies and the method for measuring changes in reactor height have been described in detail by R. W. Powell.[2] Apparatus and techniques used in measuring dimensional expansions were given in other work.[3] Exposure units are given in net fast exposure or MWD where 1 MWD (megawatt day per adjacent ton of uranium) $\sim 7 \times 10^{16}$ nvt for neutrons with energies above 0.6 Mev. 1 MWD is also approximately equivalent to a "net fast exposure" of 3×10^{17} nvt.

Samples that follow the irradiation and annealing history of the BGRR are referred to as monitor samples. Monitoring samples are dispersed throughout the reactor. Irradiation and annealing histories of samples in different positions vary considerably since the flux, the irradiation temperature and the anneal temperatures change with position. Temperature distributions also varied from anneal to anneal. At the time the measurements reported in this work were completed the reactor had been annealed

38 times. The first seventeen anneals occurred under conditions of variable annealing frequency. The reactor was then annealed every three weeks (~16 MWD). Representative data illustrating the history through the first twenty anneals were described in other work.[1] The monitoring studies given in this work (Fig. 3, Table I) pertain to the 18th through the 38th anneal.

Contractions occurring during anneals are described as either net recoveries or % recovery per reirradiation and annealing cycle. Net recoveries refer to the fraction of the total expansion removed by contraction. Recoveries per cycle are defined as the recovery during an anneal divided by the growth during the previous reirradiation i.e.

$$\text{recovery/cycle} = |R|/G \, .$$

In some experiments both $|R|$ and G were measured independently. In most of the monitoring studies only net changes in dimensions after a reirradiation and annealing cycle could be measured. In these cases the growth occurring during the reirradiation was estimated from growth rates of continuously irradiated BGRR graphite. The data necessary for these estimates were given as functions of exposure and irradiation temperature in other work.[3] The recovery for a reirradiation and annealing cycle under these conditions is defined as

$$\frac{[\text{estimated growth} + \text{measured contraction}]}{\text{estimated growth}} .$$

Reactor height measurements involve columns of graphite with varying degrees of radiation damage. Some of the graphite is essentially unirradiated since the fast flux in the periphery is zero. Recoveries for such measurements are defined relative to expansions that occurred during the continuous exposure before the onset of alternate reirradiations and anneals. It is assumed that the complicating factors due to inert graphite effect the expansion and contraction of the reactor contour in the same manner.

These studies are based on dimensional changes. It is expected, from other work,[1] that the results are also valid for c-axis and stored energy changes.

III. RESULTS AND DISCUSSION

A. Variation of $G/|R|$ with Exposure and Irradiation Temperature

Continuous neutron irradiations of graphite result in expansions that depend on exposure and irradiation temperature. When irradiated graphite is thermally annealed, the amount of contraction that occurs at a given anneal temperature is monotonic with the magnitude of the first exposure and inversely dependent on the irradiation temperature.[5] However, neither relationship is well defined.

In graphites that are alternately irradiated and annealed more dimensional damage can be removed by a reirradiation and annealing cycle than is produced by the reirradiation itself. The ratio of the growth to the recovery ($G/|R|$) will vary from less than unity to greater than unity with increasing exposure between anneals. Although the growth during the reirradiation and the recovery during the anneal are each dependent on irradiation temperature and magnitude of exposure between anneals, the ratio $G/|R|$ is independent of irradiation temperature and is a linear function of the exposure between anneals.

The ratio $G/|R|$ was measured on samples that were annealed at 350°C after each of a series of different exposures at a fixed irradiation temperature. The results of such experiments for irradiation temperatures between 20°C and 200°C are shown in Fig. 1. The samples studied had total exposures from 1200 MWD to 2000 MWD with an exposure before the first anneal of about 1000 MWD. It can be seen that for the range of exposures studied, the ratio of expansion during the reirradiation to the contraction occurring at 350°C is proportional to the exposure between the 350°C anneals and is independent of the temperature of irradiation.

The abscissa value for $G/|R| = 1$ corresponds to the exposure at which the 350°C anneal removes the same quantity of damage that the previous reirradiation produced. The results of studies relating the exposure necessary for 100% recovery ($G/|R| = 1$) to the anneal temperature are compared to a similar experiment of Woodruff's[4] in Fig. 2. It is seen that as the exposure between anneals increases, higher anneal temperatures are necessary to remove the expansion produced by the reirradiation.

B. Recoveries Under Conditions of Varying Anneal Temperatures

Net recoveries for all the 3 week reirradiation and annealing cycles of the BGRR are shown for 3 typical samples in Fig. 3. The samples were taken from different reactor positions[2] and represent a range of irradiation temperatures from 28°C to 125°C. The original damage present before the first cycle varies by a factor of three in the samples shown. Additional data for these and a fourth sample are given in Table I. (The plot of new recovery vs number of cycles for the fourth sample is almost identical to the sample with a 28°C irradiation temperature and is not shown.)

It is observed from Fig. 3 that the net recoveries are approximately linear with the number of cycles and show no trend with irradiation temperature. From Table I it is seen that while the net recoveries show no trend with original dimensional damage, the average recovery per cycle is the same for all samples.

Similar effects were also observed in reactor height measurements. Fig. 4 shows the data used to obtain the recovery per cycle in reactor height. The three linear plots with positive slopes on the left side of the figure represent growth before the first anneal given by R. W. Powell [Reactor Operations Division Monthly Report, April 1957, BNL 450 (T-96)]. The curves on the right of Fig. 4 give the recent data on reactor contractions for the 24th through the 37th anneal. The average recoveries obtained from the ratios of the slopes of the lines are 182%, 189%, and 188% for the three positions measured. These values compare favorably with the monitoring results.

C. Estimates of Effective Anneal Temperatures

The absence of any dependence of the recovery per cycle on the irradiation temperature is not surprising in view of the studies described in Section A. The consistent value of 185% obtained throughout the reactor from monitoring and reactor height measurements is, however, unexpected. Since the recovery (i.e. $|R|/G$) is dependent on anneal temperature, the data imply that a single effective anneal temperature characterizes each cycle at all positions.

It is possible to estimate such a temperature from the data given in Figs. 1 and 2. It is first assumed that the $G/|R|$ functions are linear with exposure between anneals for all anneal temperatures. The actual slopes of the lines can then be determined by connecting the origin with points obtained from Fig. 2 for $G/|R| = 1$. The results are shown in Fig. 5. It is observed that a recovery of 185% for a 16 MWD exposure between anneals is equivalent to an effective anneal temperature of 250°C. It is worth noting that the same effective anneal temperature can be obtained from a weighted average of the seventy thermocouples used to measure anneal temperatures. The numerical temperature distribution and weighted averages for the last seven anneals are given in Table II. The weighted average, therefore, appears to be a useful empirical tool for estimating the effectiveness of an anneal.

IV. CONCLUSIONS

Laboratory experiments, monitoring studies, and reactor height measurements all show that the recovery for a reirradiation and annealing cycle is independent of irradiation temperature and total damage over wide ranges. The reciprocal of the recovery per cycle is linear with exposure between anneals with a slope that is determined by the anneal temperature. When the exposure between anneals and the anneal temperature are fixed, the recovery per cycle remains constant and does not appear to decrease after many cycles. This effect was also observed in other work[(1)] and holds for wide ranges of dimensional damage. However, it is not expected to hold when most of the damage is removed.[(6)]

In spite of complexity in the irradiation and annealing program and fluctuations in pertinent variables, reactor height measurements correlate well with monitoring sample measurements.

It is concluded that the technique of alternate re-irradiations and anneals can be used consistently to remove radiation damage from graphite.

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FIGURE CAPTIONS

Fig. 1 - Ratio of growth to recovery (after 350°C anneals) vs exposure between anneals for various irradiation temperatures.

Fig. 2 - Anneal temperature for $G/|R| = 1$ vs exposure between anneals.

Fig. 3 - Net dimensional recoveries vs number of 3 week reirradiation and annealing cycles.

Fig. 4 - Reactor height expansions and contractions.

Fig. 5 - $G/|R|$ functions for various anneal temperatures.

RATIO OF GROWTH TO RECOVERY (AFTER 350°C ANNEAL) vs IRRADIATION INTERVAL BETWEEN ANNEALS FOR GRAPHITES IRRADIATED TO 2000 MWD

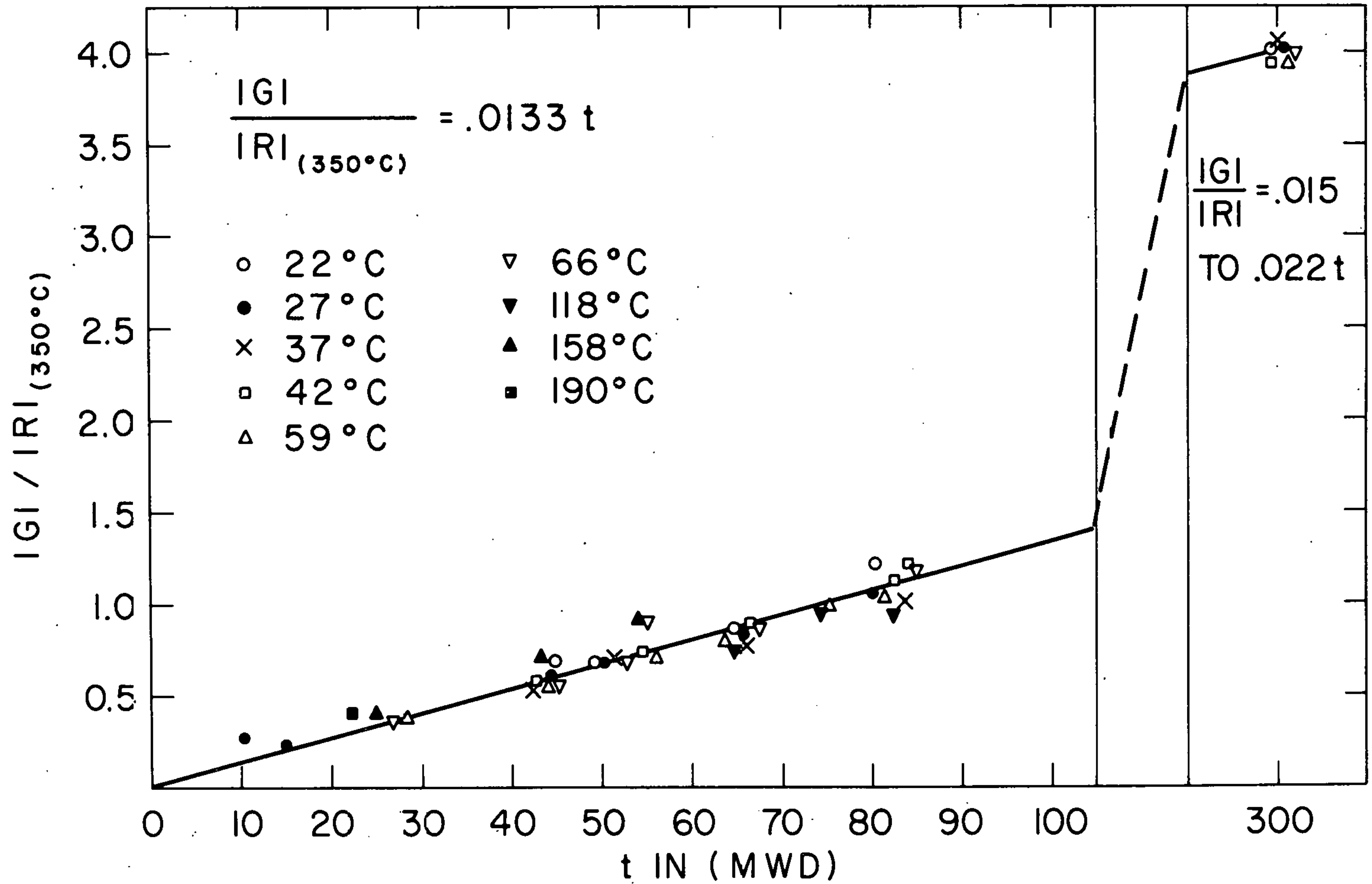


FIGURE 1

ANNEAL TEMPERATURE vs
IRRADIATION INTERVAL FOR G/IRI=1

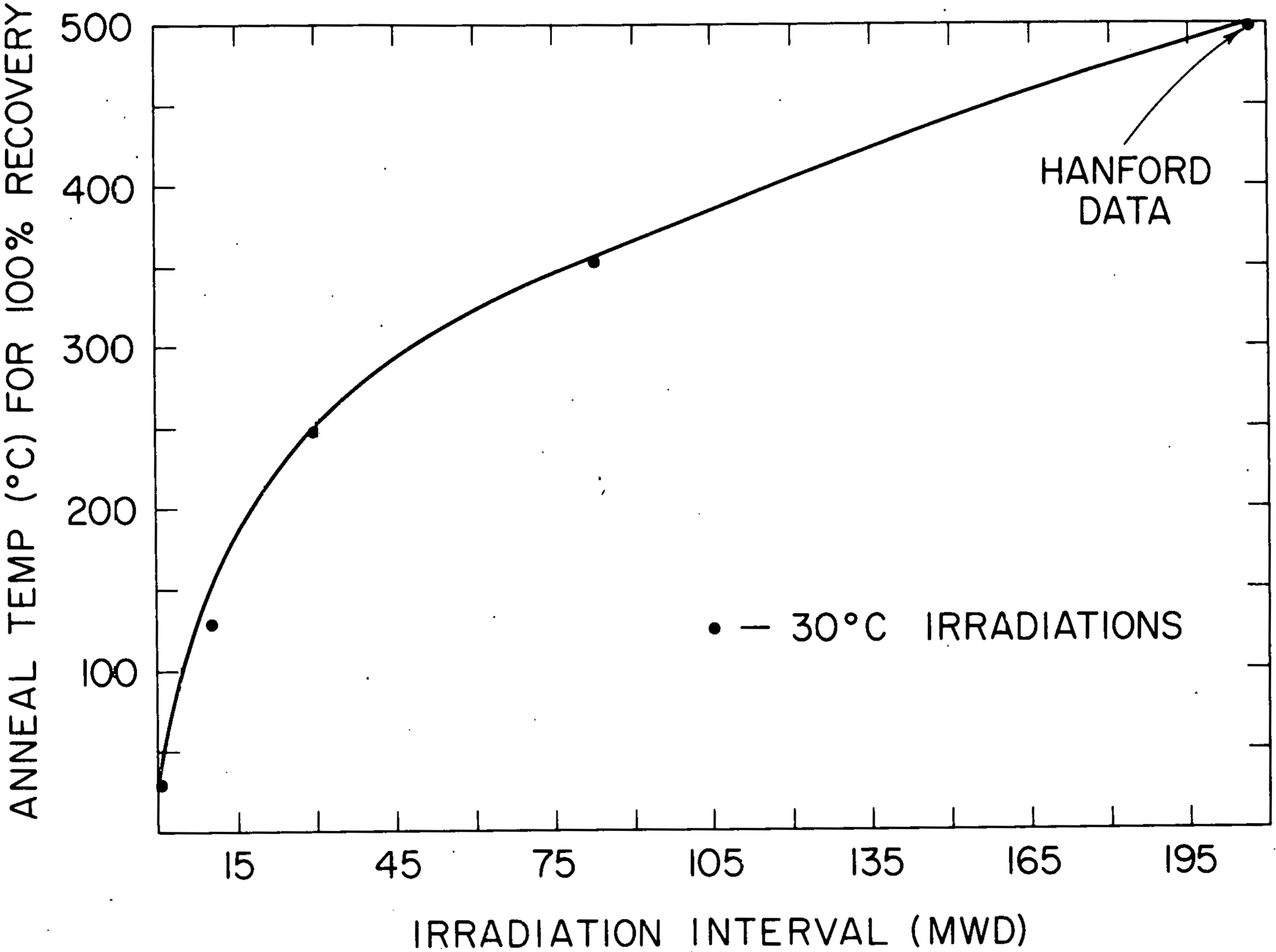


FIGURE 2

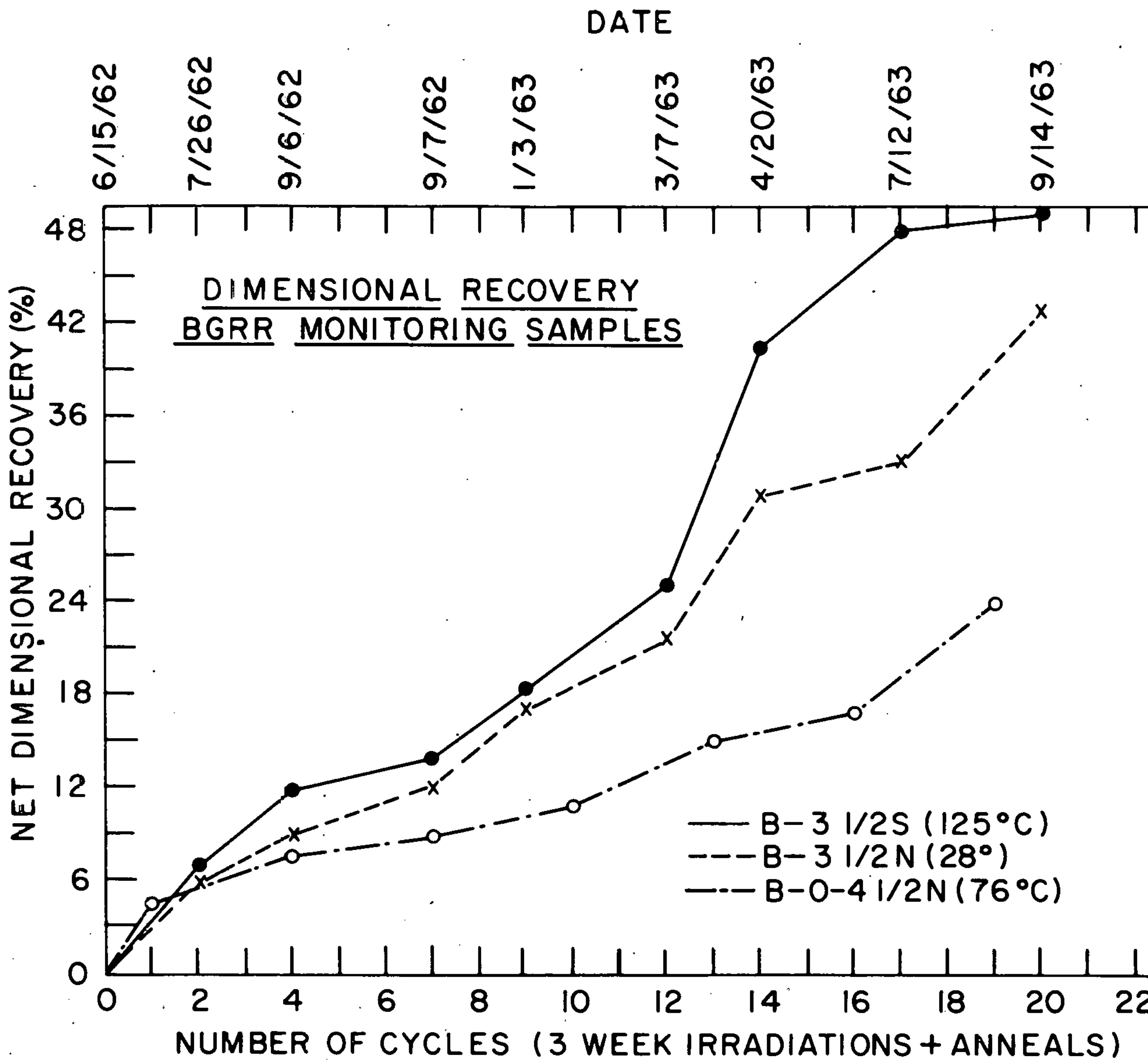

DATE
6/15/62
7/26/62
9/6/62
9/7/62
1/3/63
3/7/63
4/20/63
7/12/63
9/14/63
DIMENSIONAL RECOVERY
BGRR MONITORING SAMPLES
NET DIMENSIONAL RECOVERY (%)
0
6
12
18
24
30
36
42
48
B-3 1/2S (125°C)
B-3 1/2N (28°)
B-0-4 1/2N (76°C)
0
2
4
6
8
10
12
14
16
18
20
22
NUMBER OF CYCLES (3 WEEK IRRADIATIONS + ANNEALS)

FIGURE 3

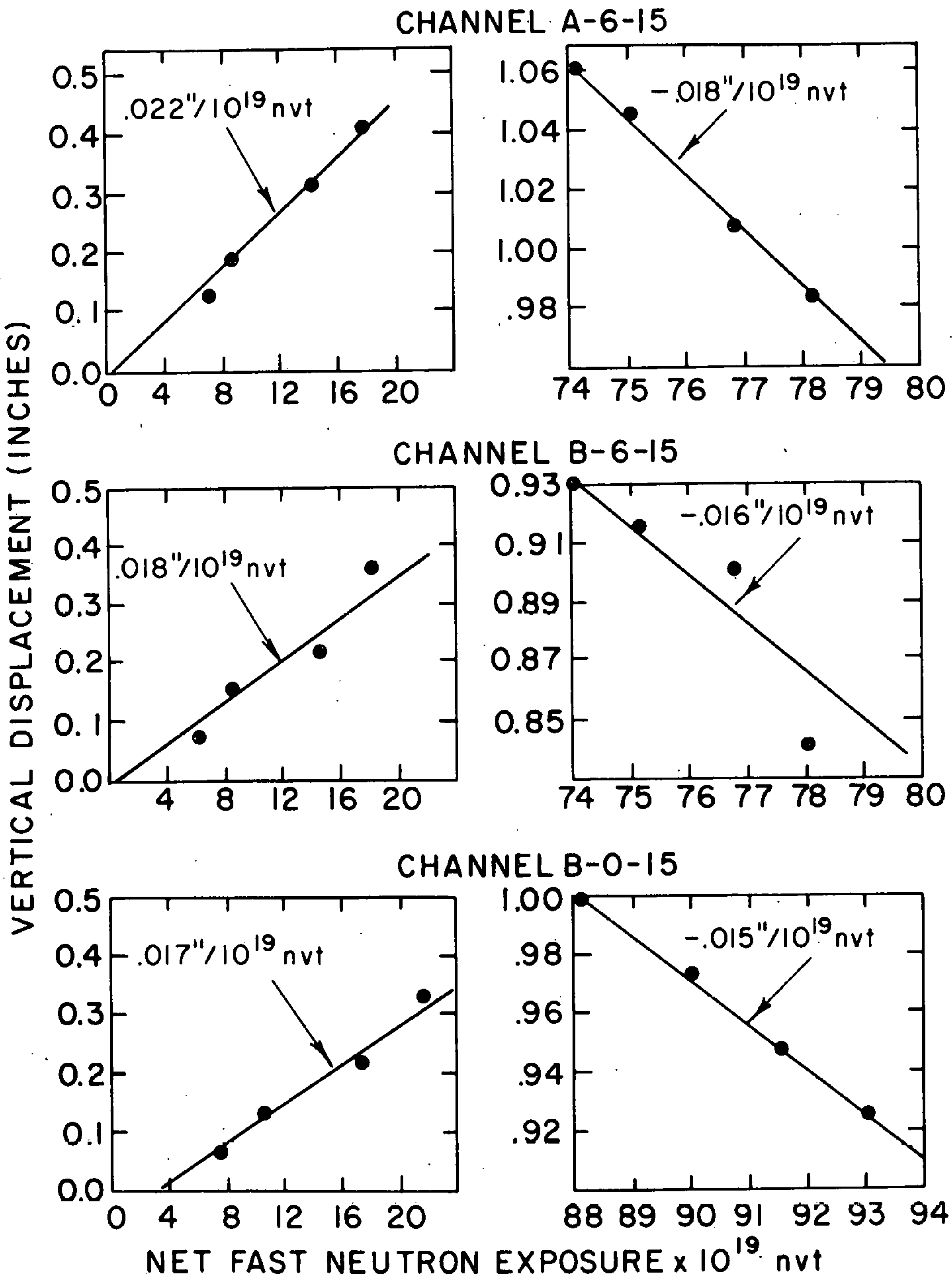
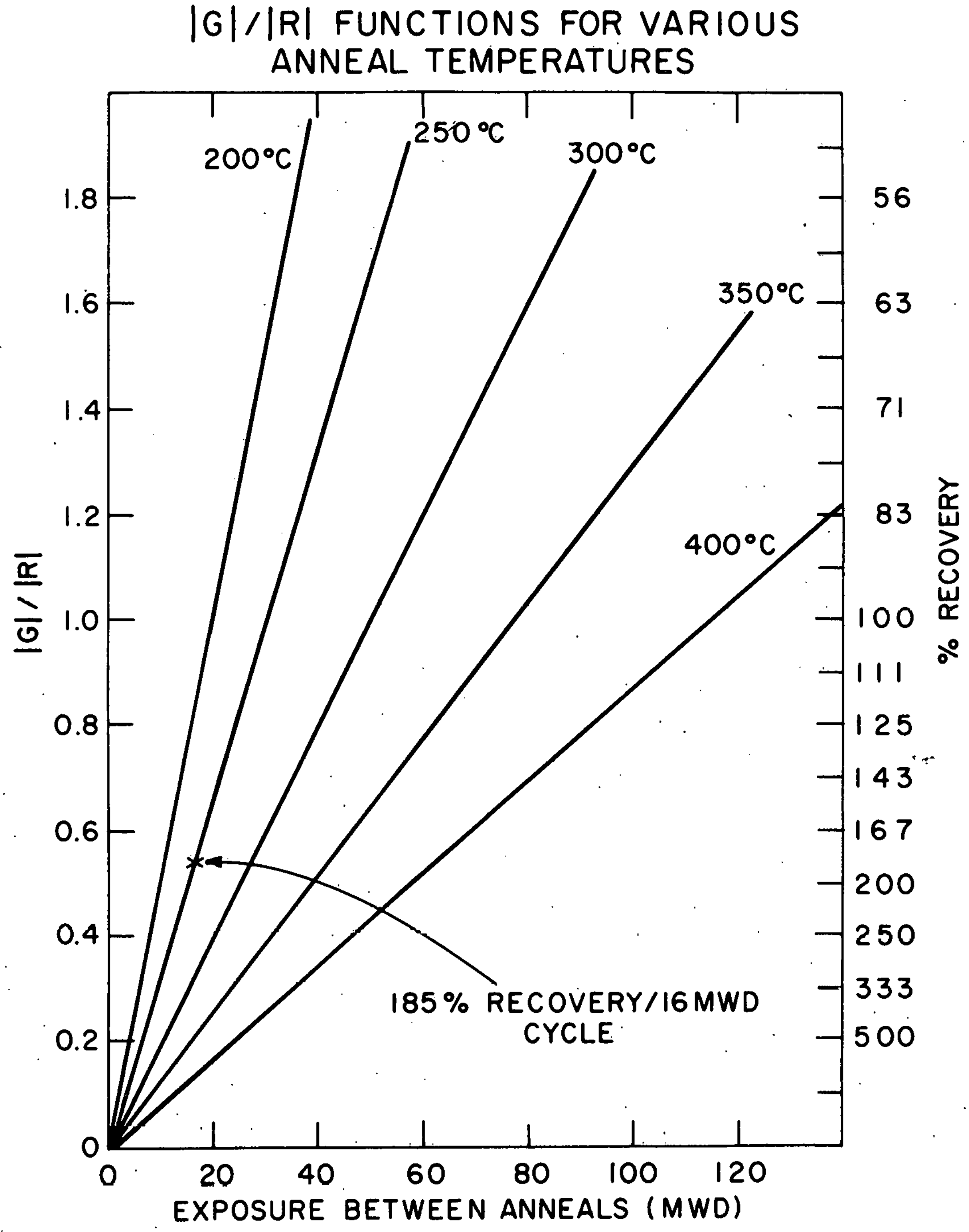

CHANNEL A-6-15
.022"/10^19 nvt
-.018"/10^19 nvt
CHANNEL B-6-15
.018"/10^19 nvt
-.016"/10^19 nvt
CHANNEL B-0-15
.017"/10^19 nvt
-.015"/10^19 nvt
VERTICAL DISPLACEMENT (INCHES)
NET FAST NEUTRON EXPOSURE x 10^19 nvt

FIGURE 4



|G|/|R| FUNCTIONS FOR VARIOUS ANNEAL TEMPERATURES
200°C
250°C
300°C
350°C
400°C
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
|G|/|R|
56
63
71
83
100
111
125
143
167
200
250
333
500
% RECOVERY
185% RECOVERY/16MWD CYCLE
0
20
40
60
80
100
120
EXPOSURE BETWEEN ANNEALS (MWD)

FIGURE 5

TABLE 1

Average Dimensional Recovery of BGRR Monitoring Samples

Channel	Irradiation Temp.	Total Growth at Beginning of 3 Week Program	Expected Growth During 3 Week Program	Contraction During 3 Week Program	Average Recovery Per Cycle
	°C	inches x 10^4	inches x 10^4	inches x 10^4	%
B-3½ S	125	54	31	26	185
B-3½ N	28	108	57	46	181
B-D-4½ N	76	169	48	41	185
B-3½ S	80	82	41	36	188

TABLE 2

Temperature Distribution in BGRR During an Anneal

Temp. (°C)	Number of Thermocouples*						
	38th Anneal (9/14/63)	37th Anneal (8/23/63)	36th Anneal (8/2/63)	35th Anneal (7/12/63)	34th Anneal (6/22/63)	33rd Anneal (5/10/63)	32nd Anneal (4/20/63)
100	1	3	1	1	1	3	1
125	5	2	4	5	6	4	6
150	4	3	3	4	3	4	4
175	1	3	3	1	1	0	0
200	1	0	0	1	2	0	2
225	7	8	3	3	3	4	4
250	8	8	11	11	20	15	17
275	15	9	8	16	8	13	18
300	11	16	22	13	17	13	13
325	12	11	9	11	9	9	5
350	5	5	6	4	0	5	0
375	0	2	0	0	0	0	0
Weighted Average	252°C	267°C	269°C	263°C	254°C	260°C	250°C

*Based on 70 thermocouples dispersed through the BGRR.