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ROLE OF IONIZATION IN RADIATION ANNEALING

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RADIATION EFFECTS ON
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ROLE OF IONIZATION IN RADIATION ANNEALING

[redacted]

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ABSTRACT

The role of ionization in the phenomenon of "radiation annealing" of graphite has been studied by using a 1-Mev electron beam. Changes in the c-axis of a sample with a Hanford irradiation of 460 mwd/ct were studied. Two thermal anneals of 4 hours each at 350° C proved sufficient to complete the thermal annealing at this temperature. The samples were then irradiated for 7-1/2 hours at a temperature of 340° C. The samples received an irradiation of 47 microampere-hours, equivalent in ionization to an exposure of 200 mwd/ct in a Hanford reactor. No changes were noted as a result of the electron bombardment. It is concluded that the ionization is not of major importance in radiation annealing.

This report is based on studies conducted for the Atomic Energy Commission under Contract AT-11-1-GEN-8.

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I. INTRODUCTION

When graphite irradiated in a nuclear reactor at one temperature is subjected to further reactor exposure at a higher temperature, annealing takes place beyond that resulting from thermal annealing alone. Such a phenomenon is termed "radiation annealing." The mechanism by which such an annealing process occurs is a matter of considerable interest. This paper reports the results of an experiment designed to determine the role played by ionization in radiation annealing. A 1-Mev electron beam was used which produced very few displaced atoms and hence was almost entirely ionizing in nature. The experiment was comparable to work done at the Hanford Works on c-axis changes both with regard to the total amount of energy dissipated in the samples and the temperature of exposure.[1] The rate of energy dissipation in the present experiment, however, was considerably greater than in the reactor experiments.

II. EXPERIMENTAL PROCEDURE AND DATA

The experiment was designed so that several samples could be simultaneously annealed in the same apparatus while some of the samples were exposed to the beam and some were not. Such a procedure ensured identical temperature histories for the samples and hence simplified the interpretation of the results.

Samples 1/8 inch by 1/8 inch by 0.005 inch were chosen and were mounted on a 1-1/4 inch square copper block in three parallel rows as shown in Fig. 1. National Carbon Company No. 15 graphite cement was used to secure the samples to the holder. A central section of the block (5/8 inch by 7/8 inch) was recessed 0.007 inch below the surface of the holder so that the surfaces of the samples and the surface of the copper were in the same plane. With this arrangement all the samples were in proper relative focus when placed in the X-ray machine. Copper X-radiation was used in conjunction with a nickel filter and in each case the samples not being studied were covered with lead sheeting so that only one specimen at a time was exposed to the X-ray beam. The instrument was calibrated before and after each set of runs by taking standard silicon curves. This calibration did not change appreciably throughout the experiment.

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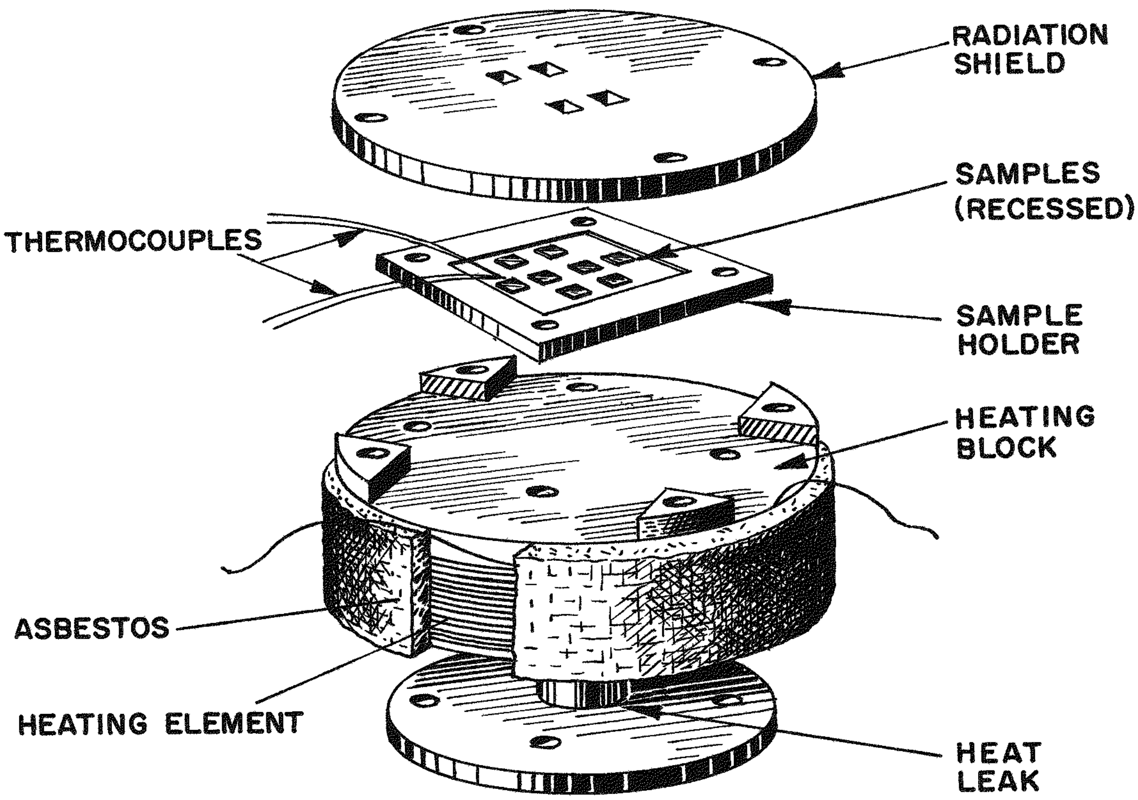

Fig. 1. Sample Holder and Heater

The copper holder was fastened to a copper heater block by means of four brass screws. Figure 1 shows an exploded view of the apparatus. The heater block was 2 inches in diameter with a thickness of 3/4 inch. Ten turns of nichrome wire, insulated from the copper block by a thin strip of asbestos, served as the heating element. Further layers of asbestos on the outside of the wire provided thermal insulation. A thermal leak for the block was provided through a post of stainless steel which was fastened to a water-cooled base plate. After the sample holder was screwed to the heater block, a copper shield was fastened to four posts on the block so that the shield would be at the same temperature as the block and would cover the samples without touching them. This shield was 1/8 inch thick and had four square holes 1/8 inch on edge cut in it as shown in Fig. 1. This arrangement permitted four of the samples to be irradiated while the adjacent samples remained unirradiated. The alignment of the samples and the holes in the radiation shield was done prior to the setting of the graphite cement. The temperature was measured by two chromel-alumel thermocouples which were also fastened to the sample holder with graphite cement. During the course of the experiment one of the couples was exposed to the beam, in order to guard against over heating of the samples being irradiated. The four thermocouple leads and the heater wires were brought through the base plate by means of Kovar seals. The entire unit was mounted on the North American Aviation Statitron by means of the base plate in the customary manner.[2] This base plate was water cooled by means of copper coils soldered to the back side.

A set of eight samples of AGOT-KC-type graphite was studied, consisting of two virgin pieces and six pieces with a previous irradiation of 460 mwd/ct at 30° C in a Hanford reactor. The six irradiated samples were taken from a single piece of graphite which had a Hanford sample designation of 87-206. During the electron bombardment one virgin and three damaged samples were exposed to the beam. Two separate measurements of the c-spacing were made several days apart prior to the statitron run in order to check the reproducibility of the X-ray measurements. The results of these two measurements are shown in the first two colums of Table I. The maximum deviation was within ± 0.005, comparable to the expected precision.

The holder was then placed on the heater block, the entire apparatus was pumped down to 5×10^{-5} mm mercury by using a bell jar which fit over the base plate, and the samples were annealed at 350° ± 3° C for a period of 4 hours. The holder was then removed from the heater block and placed in the X-ray unit. The results of the subsequent c-axis measurements are shown in column 4 of Table I. An average change of 0.088 A in the c-spacing was

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TABLE I

c-SPACING IN A

Sample	Run #1 as Received	Run #2 as Received	First Thermal Anneal	Second Thermal Anneal	Statitron
1†	6.936	6.938	6.865	6.871	6.865
2*†	6.738	6.739	6.743	6.747	6.747
3*	6.751	6.749	6.750	6.747	6.751
4	6.958	6.959	6.881	6.879	6.879
5	6.951	6.954	6.871	6.869	6.866
6	6.953	6.953	6.876	6.873	6.865
7†	6.945	6.946	6.900	6.886	6.885
8†	6.951	6.954	6.878	6.880	6.873

*Virgin graphite.

†Exposed to electron beam.

observed to result from thermal annealing. It will be noted that the value for Sample 7 did not agree at this point with the others. During the measurements following the first anneal, this sample was jarred from its place and was recemented to the copper holder. Unfortunately, the graphite was replaced at a slight angle which changed the relative reading for this sample. A second annealing identical to the first was then made. The values obtained after this anneal showed no appreciable change in c-spacing for any of the samples including Sample 7. It was assumed, therefore, that the thermal annealing was essentially complete for this temperature.

The entire apparatus was then placed in the North American Statitron where it was exposed for 7-1/2 hours. The temperature of the thermocouple not in the beam was 339° ± 3° C while the thermocouple in the beam ran 5 to 7° hotter. A total integrated flux of 47 μah/cm^2 was received by the holder. The results of the X-ray measurements made after this statitron run are given in column 6 of Table I. It is evident that no significant change in c-spacing has occurred after statitron irradiation of Samples 1, 7, and 8. The average observed change for this set was -0.005 A while the change for Samples 4, 5, and 6, which received no electron bombardment, was -0.004 A. These changes were well within the experimental error.

III. CONCLUSIONS

In order to compare the results of this experiment with results obtained from similar tests performed in a nuclear reactor, it is necessary to correlate the ionization produced in the statitron with that produced in a Hanford pile. The electron beam may be considered to produce only ionization. In traversing 5 mils of AGOT-KC graphite, one would expect[3] the electrons to lose 4 x 10^4 ev, and hence a total integrated flux of 1 μah/cm^2 corresponds to a loss of energy of 6600 joules per gram of graphite. Now the amount of heat generated in a Hanford reactor at a test hole is approximately 0.03 watt per gram,[4] while the number of calendar days is roughly half the number of megawatt days per central ton. The energy dissipation in one of the reactors is thus of the order of 1300 joules per gram for each megawatt day. If, as an upper limit, it is assumed that all of this heat goes into ionization, then an equivalence between the statitron and pile irradiations is obtained, namely, 5 mwd/ct is equal to 1 μah/cm^2. Thus the electron flux of 43 μah/cm^2 obtained here is equivalent to about 220 mwd/ct in a Hanford pile in terms of ionizing power.

The Hanford experiments were on several samples with various irradiation histories.[1] For comparison with the present experiment, we have chosen the data for one sample of AGOT-KC. This sample was exposed for 651 mwd/ct at 30° C, annealed at 375° C for 120 hours, and then further annealed in the reactor at 335° C. The behavior of the sample is shown graphically in Fig. 2. Since the thermal annealing should be almost complete after 120 hours, the large initial change observed on re-exposure to the neutron flux must result from radiation annealing. The results of the present experiment are also plotted in Fig. 2. The c-spacings measured in this experiment both before and after thermal anneal fall below the values noted for the Hanford sample as would be expected since the latter specimen has received considerably more neutron irradiation. If the Hanford data are extrapolated to an exposure of 460 mwd, but with a temperature and ionizing radiation history similar to that of the statitron samples, a change in the c-axis spacing of 0.080 A may be expected as a result of radiation annealing alone. In the statitron experiment any change was less than 6 per cent of this amount. Indeed, in the case of the 651 mwd/ct sample the radiation annealing caused the c-axis spacing to decrease substantially below the values for the present samples. It is also noteworthy that this large radiation annealing took place at only 325° C after a thermal annealing at 375° C, some 50° C higher. In the case of the electron-bombarded samples, thermal annealing took place at only 350° C, while the bombardment occurred at about 340° C. This lower thermal annealing temperature, as well as the increased bombardment temperature, should be much more conducive to showing radiation annealing, if present, than the pile experiments.

It should be pointed out, however, that a process might exist which could explain the negative results reported here without entirely ruling out ionization as a factor in radiation annealing. If the efficiency of this process is dependent on the intensity of the radiation, the statitron experiments could not be compared with the pile results since the rate of ionization is so different. Such a process might be similar in nature to the breakdown of the reciprocity law in photographic films. As an illustration let us consider the following model.

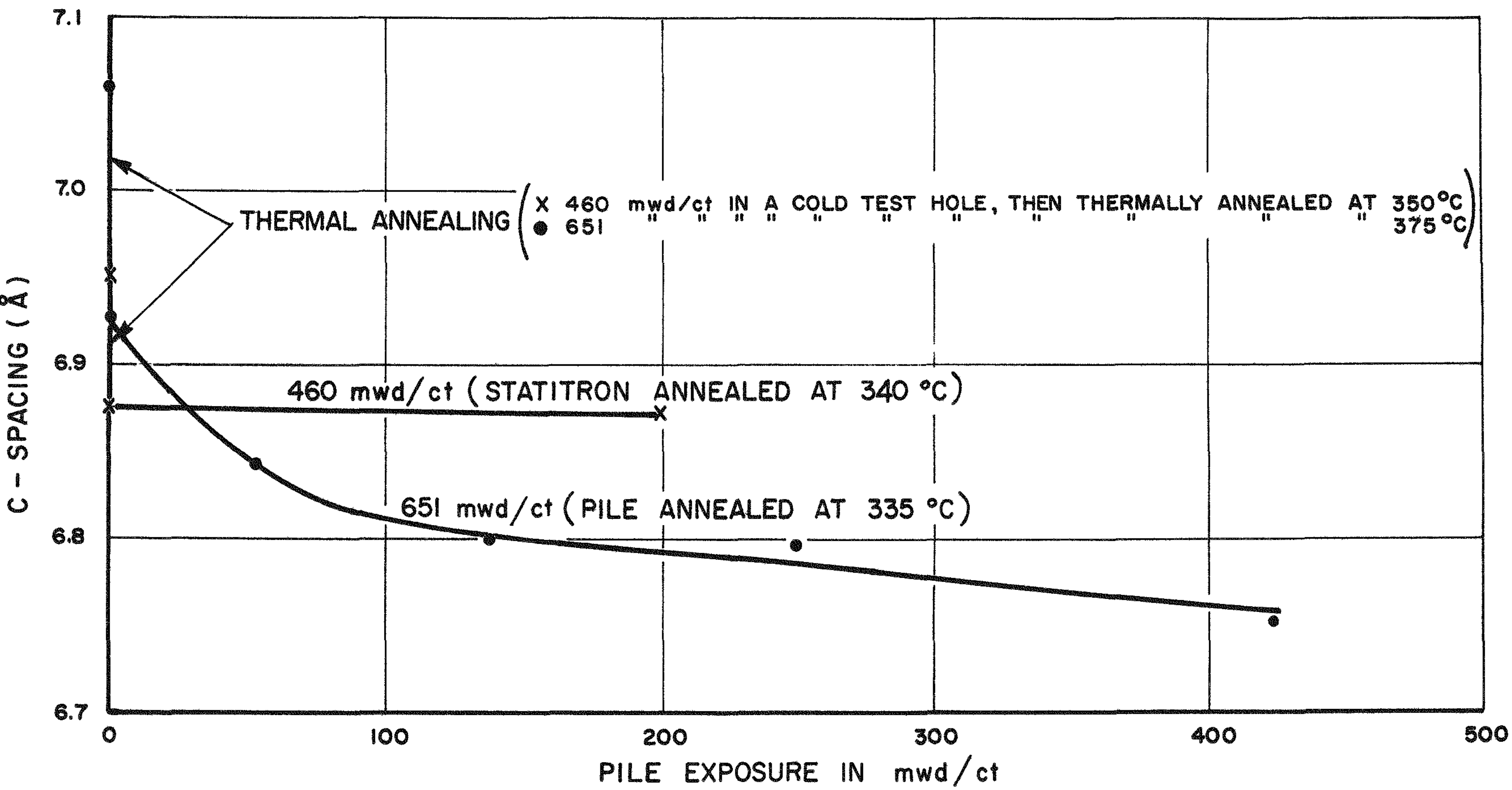

Fig. 2. Radiation Annealing

An interstitial complex C_n is excited by the ionizing radiation

$$C_n + \text{ionizing radiation} \longrightarrow C_n^* , \qquad \ldots(1)$$

the actual charge on the complex being unimportant here. The excited complex may then dissociate

$$C_n^* \longrightarrow C_{n-1} + C . \qquad \ldots(2)$$

The freed atom may then either react with a vacancy or reunite with the complex, that is,

$$C + \text{vacancy} \longrightarrow \text{annealed graphite} , \qquad \ldots(3)$$

$$C + C_{n-1} \longrightarrow C_n . \qquad \ldots(4)$$

Considering Eqs. (3) and (4), we note that the number of vacancies is independent of the radiation intensity while both the number of free atoms as well as the number of C_{n-1} complexes are dependent on the intensity. Therefore as the ionizing intensity is increased, there will be relatively more recombinations into complexes than annealing of vacancies. Thus the efficiency of annealing will decrease with increasing intensities so that in the relatively low intensity of the pile the effect of ionization may be substantially greater than in the case of the statitron.

Such a breakdown at high ionizing intensities should be small in view of the large number of vacancies available in the graphite in comparison with the number of complexes formed. Thus, it is felt that although the process described is a possible one, it is not a probable one. In order to resolve this point unambiguously, however, experiments have been planned with lower ionization intensities which are comparable to pile levels.

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