UNCLASSIFIED

Metallurgical Project

A. H. Compton, Project Director

• • •

Metallurgical Laboratory

S. K. Allison, Director

• • •

CHEMICAL RESEARCH - RADIATION CHEMISTRY

J. Franck, Division Director; W. H. Johnson, Associate Division Director
M. Burton, Section Chief; W. M. Garrison, Associate Section Chief

• • •

ERRATA FOR CC-918

October 16, 1943

673 001

UNCLASSIFIED

~~SECRET~~

CONFIDENTIAL

Errata for CC-918

Page 6, Table 1.2: Note should be made of the fact that the oxygen was determined by difference.

Page 7, Table 1.3: 2-hr/cc. should read ~~/~~ -hr/cc

Page 10, Section 3.1.1, Paragraph 2: Line 8: Figure 1 should read Figure 3.1

Page 14, Table 3.2: The "approximate percent change in E per 100,000 ~~/~~ a-hr" and "approximate percent change in R per 100,000 ~~/~~ a-hr" are extrapolated values.

" " " " The "extrapolated approximate percent change in R per 100,000 ~~/~~ a-hr" values for 4007 graphite is ~ 11.2%, and for natural ~ 9%.

" " " " The entry for natural graphite vs. average elastic modulus should read "not observable by resonance frequency methods", rather than "none observable".

" " " " In column 5 insert "x" signs before 10's.

Page 16, Table 3.4: The ⊡ points on the "% increase in E per 100,000 ~~/~~ a-hr" curve should be marked 5.3, 15, and 57, rather than 5.3, 10.7, and 15.

The point △ 0.0008 should be appropriately lowered.

Page 18; Line 4 above 3.1.3: Insert percentage before contributions.

Page 28, Table 4.2: 2nd column, 1st item - Make $Fe_2(SO_4)$ read $Fe_2(SO_4)_3$.

" " " " 3rd column, 2nd item - $2.25NH_2SO_4$ should be 2.25 N H_2SO_4

CONFIDENTIAL

673 cc2

~~SECRET~~

Report CC-918

Metallurgical Project

A. H. Compton, Project Director

• • •

Metallurgical Laboratory

S. K. Allison, Director

• • •

CHEMICAL RESEARCH - RADIATION CHEMISTRY

J. Franck, Division Director; W.H. Johnson, Associate Division Director
Milton Burton, Section Chief; W.M. Garrison, Associate Section Chief

• • •

REPORT FOR MONTH ENDING SEPTEMBER 11, 1943

• • •

Table of Contents

• • •

0.1 Abstract (W. M. Garrison)

Further work has been carried on with the infra-red spectrometer at Urbana. Biphenyl and an aliphatic polymer have been identifed in electron and deuteron bombarded benzene. The work is being continued and a complete infra-red analysis of benzene and n-octane will be presented later (1.1.1). Chemical tests show that biphenyl comprises ~7.5% of the total solids formed in β irradiated benzene (1.1.2). Apparatus has been constructed for measuring the average energy to produce an ion-pair in liquid hydrocarbons (1.1.3). Naphthene base lubricating oil on β irradiation shows less change in acid number and iodine number than similarly exposed paraffin oils. Viscosity changes are, however, comparable (1.1.4). A separate report on the effect of radiation on shielding materials is being prepared and will appear shortly (1.2.1).

A complete literature survey is being made on the effect of radiation on water (2.1.1.). Deuteron irradiation experiments at U of M are now under way. An unexpectedly high degree of corrosion found on the non-irradiated controls precludes any estimation of the radiation corrosion at this time. Efforts are being directed towards determining operating conditions under which the corrosion of the controls will be minimized. Construction of the system for the study of electron corrosion is being completed (2.1.2).

Additional data for the change of Young's Modulus and electrical resistance of AGOT graphite with neutron bombardment continue to show a trend toward saturation. The curves have been corrected for the variation of intensity with position of the samples on the cyclotron target (3.1.1). Experiments with special graphites in which there is a variation of crystal size and degree of graphitization, indicate that the greater the order of the crystallites, and the larger and more nearly perfect the graphite crystals, the greater is the Wigner effect caused by neutron bombardment, as measured by resonance frequency and electrical resistance changes (3.1.2). Experiments on the thermal healing of the Wigner effect have been inaugurated. Results indicate that healing of the effect, as measured by resistance changes, proceeds by at least two mechanisms, and that one process goes virtually to completion at 200°C. Acoustical measurements indicate that complete healing may take place at 500°C, but results of heating at 200°C, where a slight increase in resonance frequency occurred, do not support the electrical resistance data (3.1.3). A description of the method and the results of monitoring the fast neutron intensities over the target area of the Washington University Cyclotron are included (3.1.4). Additional data on the rate of diffusion of helium into the pores of graphite again demonstrate poor communication. Measurements have been made on the surface area of AGOT graphite (3.1.6).

Several additional samples of optical glass as well as various types of light bulbs have been subjected to X radiation at the N. D. generator. Lucite exposed to 50 kR exhibits no noticeable color change (4.1). Additional data on the solubilities of NO and Xe are presented. A solution of CrO_3 and H_2SO_4 seems to be the most satisfactory solution for removing NO (4.2).

The 1.25 Mv generator is operating satisfactorily in the production of β radiation up to a measurement of 50 μa at rated voltage (5).

Group I W. M. Garrison

Table of Contents

• • •

1.1. Organic Liquids

1.1.1. Infra-red Study of Irradiated Benzene and Octane (E. Schlegel)

Further work has been carried on with the infra-red spectrometer at Urbana. In CC-841 it was reported that electron bombardment of benzene results in the production of aliphatic bonds. This production is indicated by increased absorption at the aliphatic C-H frequency, i.e. at 3.43 μ. The same effect has been observed with deuterons, but the conversion is apparently not as efficient, in terms of energy input. For example, in benzene which was electron irradiated in an 8 cc. volume for fifty-five minutes at 1.5 Mev. and 5 μa, the log $\frac{I_o}{I}$ value at 3.43 μ was found to be .25. Benzene bombarded with deuterons in a 2 cc. volume for 7.5 minutes at 6.9 Mev. and 2 μa, (giving approximately the same energy of bombardment per benzene molecule as the electron bombardment), had a log $\frac{I_o}{I}$ value of .17 at 3.43 μ.

Absorption studies were made of electron bombarded and untreated benzene and n-octane, in the range 2.4-12.4 μ. The majority of molecular vibration spectra is within this range. There is no single

suitable solvent which is transparent to infra-red radiation throughout this range. Therefore, the substances were formed in thin films between NaCl plates, and the absorption of the films measured. The film technique is useful for getting an absorption spectrum through a large range; but it is of little value in making quantitative comparisons of the absorption of different substances, since films will vary considerably in thickness.

It is readily apparent from the spectrum of the electron irradiated benzene that biphenyl is produced by the irradiation. Biphenyl has characteristic absorption peaks at ~ 9.4 μ and ~ 11.0 μ which are not present in the benzene spectrum. The spectrum of the bombarded benzene shows absorption at those wave-lengths. Thus, the infra-red study corroborates the conclusion, previously established by chemical methods, that biphenyl is produced in irradiated benzene. CCl_4 does not absorb strongly at either 9.4 μ or 11.0 μ. It will be possible therefore to study the absorption of CCl_4 solutions of benzene at those regions in a cell of given thickness, and thereby make quantitative comparisons of the amount of biphenyl produced.

Further interpretation of the benzene and n-octane spectra is in progress, and will be discussed in a later report.

1.1.2 Identification of Products Formed by β Irradiation of Benzene (J.N. Burr)

In report CC-841 it was shown that β irradiation of benzene produces biphenyl in an amount equal to 7.5% of all solid radiation products, and a polymer which is at least in part aliphatic. The biphenyl was separated from the irradiated benzene solution by steam distillation. Table I.1 gives the carbon-hydrogen analysis on the material thought to be biphenyl. These results, together with the melting point and molecular weight data, firmly establish the presence of biphenyl in the irradiated product. It is interesting to note that Mr. Schlegel (Section 1.1.1. this report) has detected biphenyl in β bombarded benzene by infra-red absorption methods.

Table 1.1 Carbon-hydrogen Analysis of Biphenyl from Benzene

Sample	Radiation Product	Biphenyl (C.P. recryst)
Melting Point °C	66.2 - 67.4	67.2 - 69.0
Percent Carbon	93.08	93.07
" Hydrogen	6.88	6.64

The aliphatic polymer mentioned above as making up the major part of the solid radiation product possesses initially a comparatively low average molecular weight of about 400. However, with moderate heating

this material immediately polymerizes to a substance of very high molecular weight. This is a behavior which is not exhibited by aromatic hydrocarbons, but very frequently found in highly unsaturated aliphatic or cycloaliphatic hydrocarbons. The nature of the polymer has not as yet been ascertained but a determination of the carbon and hydrogen content has been made and the results are given in Table 1.2

Table 1.2 Carbon-hydrogen Analysis of Polymer from Benzene

Sample	A	B	C
Percent carbon	88.11	87.75	89.67
" hydrogen	7.90	7.17	7.78
" oxygen[1]	3.99	5.00	2.55

[1] The oxygen is present as a result of absorption from the atmosphere.

This polymer has been further separated by solution in acetone. The acetone soluble fraction contains material of low molecular weight. The acetone insoluble fraction has been found to have a molecular weight of ~ 6000. The isolation of this material shows that β irradiation of benzene produces, in addition to biphenyl, a whole series of compounds with molecular weights ranging from ~ 250 to ~ 6000. It is hoped that further work will disclose the nature of these products.

1.1.3 Apparatus for Measuring the Average Energy Required to Produce an Ion-Pair in Liquid Hydrocarbons. (C.Hochanadel and M. Wiedenbeck)

Calculations of the ion-pair yield Γ in our work have involved the assumption that the energy required to produce an ion pair in liquids and solids is the same as the values which are reported in the literature for gases. Experiments are now in progress, the primary purpose of which is to check the validity of this assumption. Also, since relatively few investigations have been conducted to determine the effect of ionizing radiations on highly insulating liquids, the problem promises to be of considerable interest from this standpoint.

The method to be used is simply to measure the number of ion pairs produced for a given energy input. An electron beam, of known energy, will be absorbed in a layer of liquid between two parallel plate electrodes. From the measured ion saturation current, the energy required to produce an ion pair will be calculated. To prevent a leakage current between the high voltage electrode and the grounded target of the generator, along the highly ionized air path, the cell is enclosed within a glass jacket in which an insulating vacuum is maintained. Insulating coatings, etc. will be used to prevent leakage between the electrode leads, along the glass surface and through the ionized air.

It may be found impossible to obtain saturation for any experimentally reasonable applied field. In that case it will be necessary to resort to a method of extrapolation such as that used by Jaffe and Chia-Shan Pao.[1] Preliminary experiments indicate that saturation can be obtained. Further work is now being delayed until the Notre Dame electrostatic generator is repaired. ([1] Phys. Review 64 65, 1943)

673

1.1.4 Naphthene Base Lubricating Oil (R. Scott)

Data have been published on the effect of radiation on paraffin type lubricating oils (CC-238, CC-389, CC-520, CA-265, CC-784). It is known from our work on pure hydrocarbons that in general cyclic and aromatic hydrocarbons are affected less by radiation than those of an aliphatic nature (i.e. paraffin type oil). Certain oils, for example the naphthene oil, are rich in aromatics and higher members of the series C_nH_n. It was thought, therefore, that a naphthene base lubricating oil would possibly be superior to a paraffin base oil when used in regions of high radiation levels.

Samples of Sun Oil Co. "Circo Heavy Medium", a naphthene base oil, were submitted to us for tests by Mr. J. A. Collins. Bombardments were made with 8 cc. samples as described in previous reports. The viscosity, acid number, and iodine number were measured before and after irradiation; the data are given in Table 1.3.

Table 1.3 Irradiation of Naphthene Base Oil

Sample	Exposure 2-hr/cc.	Viscosity at 27° Centipoise	Iodine No. mgI/g.sample	Acid No. MgNaOH/g.sample
1	unheated	203	11.5	.11
2	.13	235	11.2	.10
3	.13	230	11.6	.03

Comparison of the data in Table 1.3 with similar data for paraffin base oil (see report CA-265) shows that changes in acid number and iodine number are much smaller in the case of the naphthene oil. Viscosity changes on bombardment are practically the same for both oils.

1.2 Organic Solids

1.2.1 Materialsfor the Physiological Shield (J. Flanagan)

A separate report on the effect of radiation on shielding materials is being prepared and will appear shortly.

673 009

Group II A. O. Allen

Table of Contents

* * * *

2.1 Water (G. Jenks, R. A. Day, L. H. Treiman, S. Gordon, R. G. Larson, S. G. Davis, J. Ghormley)

2.1.1 Theoretical

A complete literature survey is being made on the action of radiation on water, and on such related topics as the chemistry of free radicals and the kinetics of hydrogen peroxide decomposition. The survey results will be used to aid in interpretation of irradiation experiments already done, in formulating a program for future fundamental studies, and in widening our basis for prediction of effects to be expected in water under pile conditions.

2.1.2 Radiation Corrosion of Aluminum

The deuteron irradiation experiments at the University of Michigan, described in report CC-841, are now under way. To simulate neutron recoil concentrations at N, the cyclotron current was held at 0.008 μa, of which about half is believed to penetrate the water, the other half being taken up by the grid which protects the aluminum foil window on the corrosion cell. Considerable difficulty was found in measuring this current, owing apparently to back conduction through the residual air in the evacuated space between the cyclotron window and the cell window. It is believed that this trouble is now largely overcome.

The water is made up to contain, in p.p.m., 35 HCO_3^-, 45 CO_2, 1 Cl^- and 70 $SO_4^=$, Na^+ to balance, pH at room temperature 6.5. It is circulated through the cell by a Worthite centrifugal pump at 90°C. Linear speed through the cell is about 22 ft./sec. The water is kept free of air and the CO_2 concentration is maintained

at the desired level by bubbling a properly adjusted mixture of nitrogen and CO_2 through the reservoir. Under these conditions, it was found that numerous pits were present in all samples, including the blanks placed before the beam. Analysis showed the water to contain less than 2 p.p.m. of chloride. Iron was present (probably from the stainless steel of the pump) but apparently in a concentration of only about 0.1 p.p.m. Because of the small amount of irradiation, no detectable amount of H_2O_2 was present. The unexpectedly high degree of corrosion found on the non-irradiated controls precludes any estimation of the radiation corrosion. Efforts will be directed towards discovering operating conditions under which the corrosion of the controls is minimized, so that the radiation corrosion can be determined as a separate effect.

Construction of the system for the study of corrosion in the electron beam is being completed, but operations are not expected to begin at once because of the demands for other work on the Chicago Van de Graaf.

Group III

T. J. Neubert

Table of Contents

• • • •

3.1.1. Resonance frequency and resistance measurements

The graphite bombardments at the St. Louis cyclotron are being continued (CC-649, 734, 784, 841). The data previously reported have now been corrected so that the values quoted for the neutron induced changes in the elastic modulus and electrical resistance are no longer dependent upon the position of the graphite samples on the cyclotron target during exposure. The details of the monitoring as well as a contour map showing the relative fast neutron flux through the target face will be found in section 3.1.4.

The bombardment of the elastic modulus samples has been extended to a total corrected value of 287,000 μa-hr. (nominal value 343,000 μa-hr) with an increase in the modulus of ~ 33%. The electric resistance test pieces (i.e. those with bismuth contacts, cf. CC-734) have now reached a corrected exposure value of 144,000 μa-hr. (nominal value 160,000 μa-hr). As a result of this treatment, the resistance has been increased ~ 14%. The data for these two tests have been presented in Figures 3.1 and 3.2. The line in Figure 1 has been drawn with consideration being given to the number and the reliability of the measurements. Vertical lines indicate the range in moduli values for a given bombardment; horizontal lines indicate the uncertainty in estimating the neutron exposure; and the small numbers beside each point give the number of sets of measurements which were averaged to give the point plotted. The saturation effect is not so evident in Figure 3.1 as it was in the corresponding figure in Report CC-841. We have observed, however, that new samples when exposed for the same length of time in the same tube with previously bombarded samples, in general, exhibited a larger Wigner effect than those which already had some bombardment. The data supporting this observation are listed in Table 3.1.1. In the case of the effect of fast neutron irradiation on the electrical resistance of AGOT, the curve is merely an extension of that given in CC-841. We wish at this time, however, to call attention to an error in labeling Figure 3.2 in Report CC-841. The abscissae should read 0; 40,000; 80,000; and 120,000 μa-hr instead of 0; 100,000; 200,000, etc.

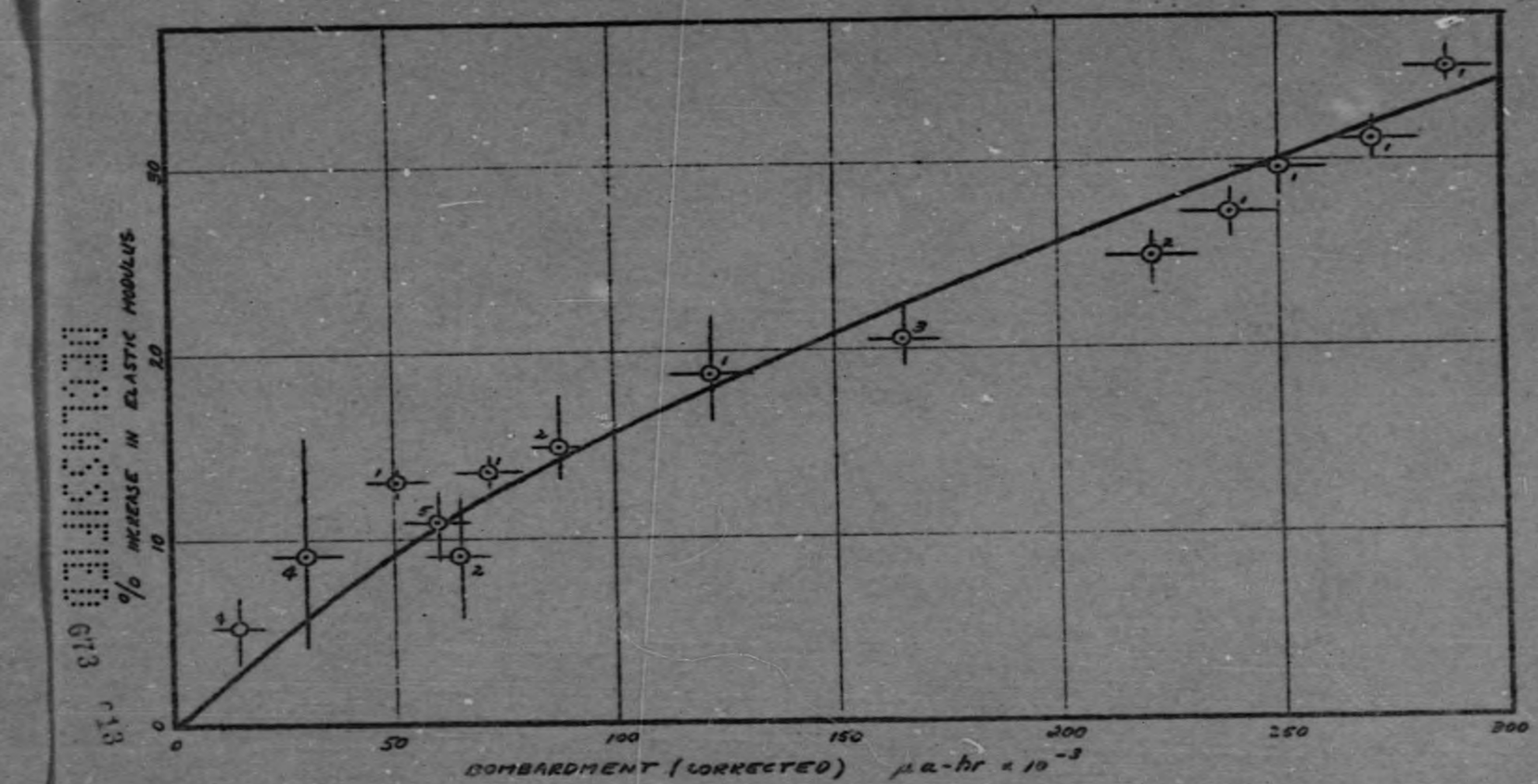

Fig. 3.1 Effect of fast neutrons on the elastic modulus of AGOT graphite.

Table 3.1 The Effect of Repeated Bombardment on the Elastic Modulus of AGOT Graphite

Tube C	Bombardment 36,000 μa hr. (corrected)	
Sample No.	Previous % change in E	Additional % change in E
1	27.8	3.6
3	29.5	5.4
5	13.3	5.6
11	14.6	7.7
12	6.5	5.2
16	5.4	7.4
29	2.7	3.7

Tube A	Bombardment 59,000 μa hr. (corrected)	
Sample No.	Previous % change in E	Additional % change in E
1	19.6	4.8
2	0	10.2
3	20.3	4.0
5	0	9.9
7	0	9.7
8	0	9.4

Tube D	Bombardment 28,000 μa hr. (corrected)	
Sample No.	Previous % change in E	Additional % change in E.
3	27.2	2.3
5	10.2	3.0
7	12.2	4.0
12	0	6.5

3.1.2. Wigner Effect in specially prepared graphite

In order to investigate the mechanism by which neutron changes occur, and to study the effect of varying the crystallite size, irradiations have been made on graphites of several different types which were prepared for us by Dr. V. C. Hamister.

Type 1. Pitch bonded lampblack. This was prepared from lamp black base flour, pitch bonded and fired to 3000°C. The graphite crystals in lampblack are very small and this is probably the reason that lampblack and pitch mixtures do not graphitize very satisfactorily.

Type 2. Pitch bonded natural graphite. This was prepared from natural graphite flour, pitch bonded and fired to 3000°C. The graphite crystals in this product should be large, but not so large as those of pure, bulk, natural graphite.

Type 3. Natural Graphite.

Type 4. Special 2100°C Graphite. This is a low density artificial graphite prepared from pitch and petroleum coke in the usual way, but fired at 2100°C. The pieces supplied us by N.C. Co. were cut so that the long dimension was perpendicular to the axis of extrusion or to the "grain" of the graphite.

Type 5. Special 2400°C Graphite. This is a low density artificial graphite prepared in the usual way but fired at 2400°C. The pieces supplied to us were cut with their long dimension perpendicular to the axis of extrusion.

Type 6. Special 3000°C Graphite. This graphite is similar in preparation and cutting to the 2100 and 2400° graphite. It was fired at 3000°C — the normal graphitizing temperature.

Graphites fired at 2100, 2400 and 3000°C were selected for tests on the basis of density data supplied by N.C. Co. From an examination of density vs. firing temperature data, see curve A, Figure 3.3, it appeared that a graphite prepared at 2100°C would represent a product in which graphitization was just beginning, in a 2400° graphite the graphitization should be partially completed; while a 3000° graphite should be more completely graphitized.

Samples for tests were cut from each of the different types of graphite. The measurements made include real density, apparent density, elastic modulus and electrical resistance. After the initial measurements the samples were sealed in vacuum and irradiated with fast neutrons at the St. Louis cyclotron. The procedures followed throughout were similar to those outlined for the AGOT work, CC-466, 520, 649, and 754. The results of the measurements on the six different types of graphite as well as average values for equivalent experiments on AGOT are summarized in Table 3.2 and in Figures 3.3 and 3.4.

673 015

Table 3.2 The Properties of Graphites of Various Types

Description	crystal size	apperent density	real density	average elastic modulus dynes/cm^2	average electrical resistance ohm cm x 10^{+4}	approximate % change in E per 100,000 n-v hr.	approximate % change in R per 100,000 n-v hr.
Pitch bonded lampblack	very small	1.45	1.88-1.97	5.2 10^{10}	63	5.3	---
Special 2100° graphite	2400° graphite crystals	1.64	1.96-2.06	3.7 10^{10}	41	24	---
Special 2400° graphite	medium	1.49	1.92-1.99	4.0 10^{10}	19	32	---
Special 3000° graphite	2400° graphite crystals	1.51	1.98-2.01	4.4 10^{10}	12	65	---
AGOT graphite	larger	1.65	2.07-2.12	10.7 10^{10}	8	15	---
Pitch bonded natural graphite	large	1.77	2.11	15 10^{10}	9-11	57	---
Natural graphite	very large	(2.28)	2.25	none observable	6.3	--	---

CONFIDENTIAL

CONFIDENTIAL

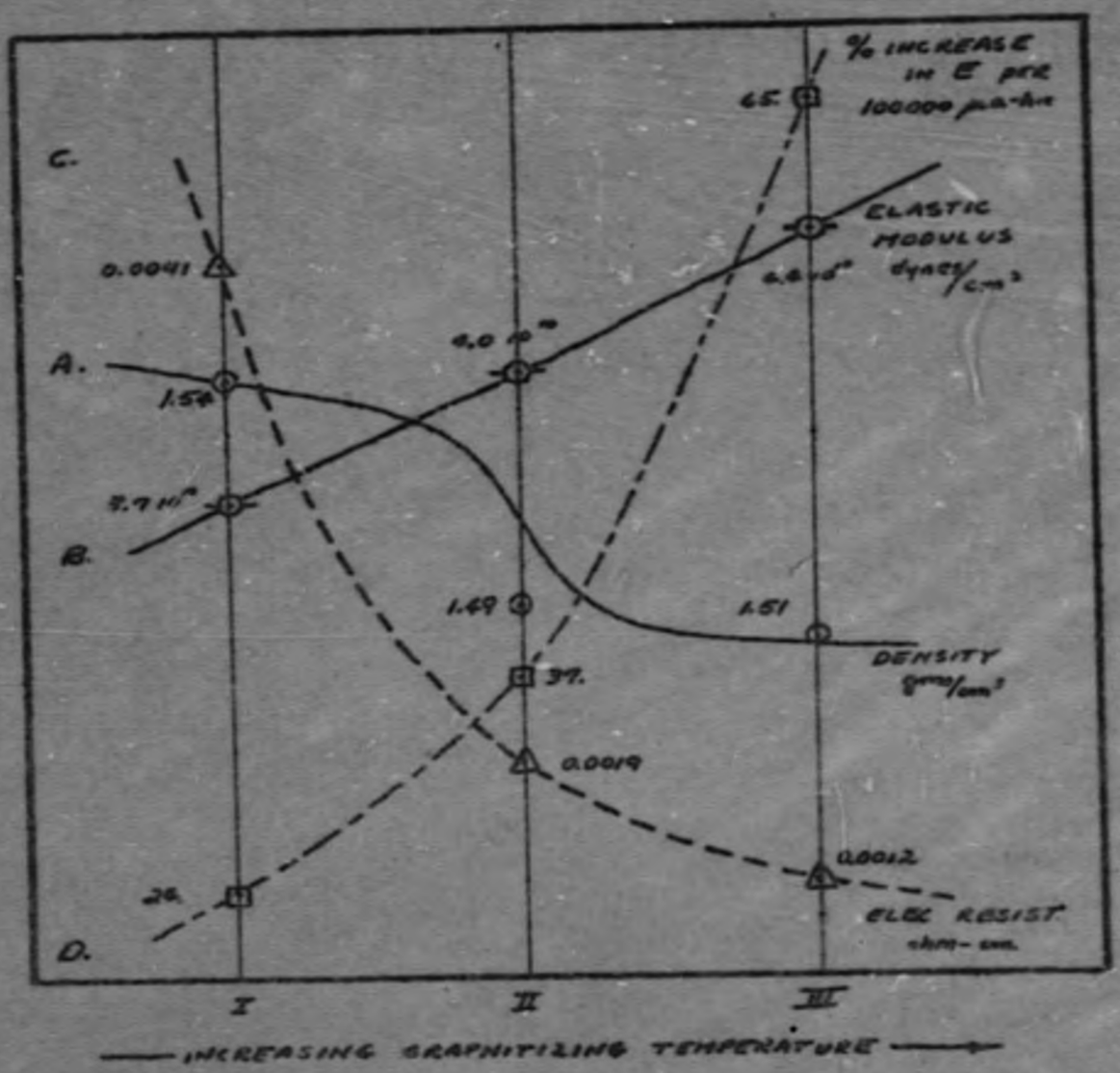

Figure 3.3 Physical properties of specially prepared graphites

I Graphite processed at 2100°C

II Graphite processed at 2400°C

III Graphite processed at 3000°C

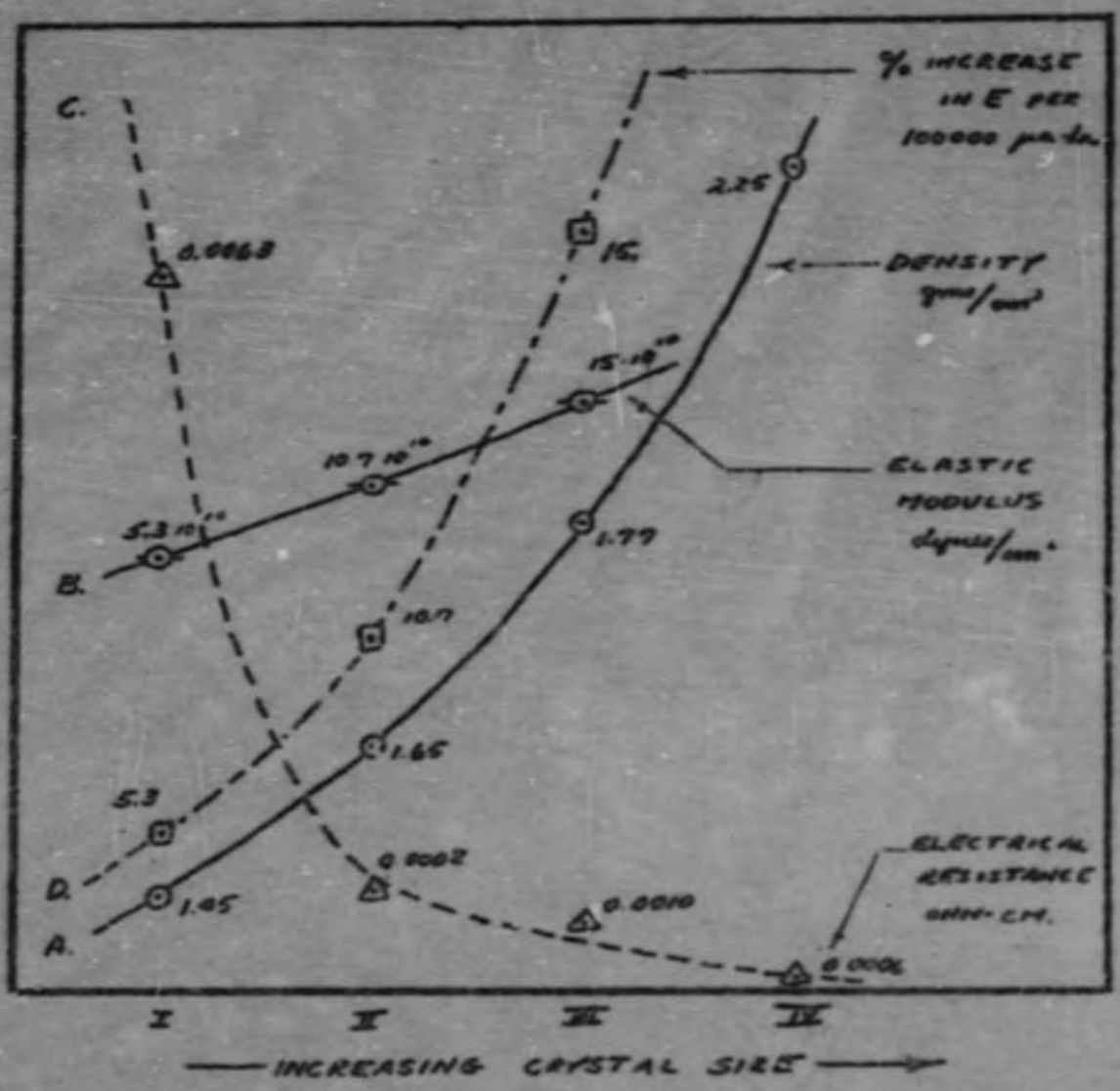

Figure 3.4 Physical properties of various graphites

I Pitch bonded lampblack

II AGOT graphite

III Pitch bonded natural graphite

IV Natural graphite

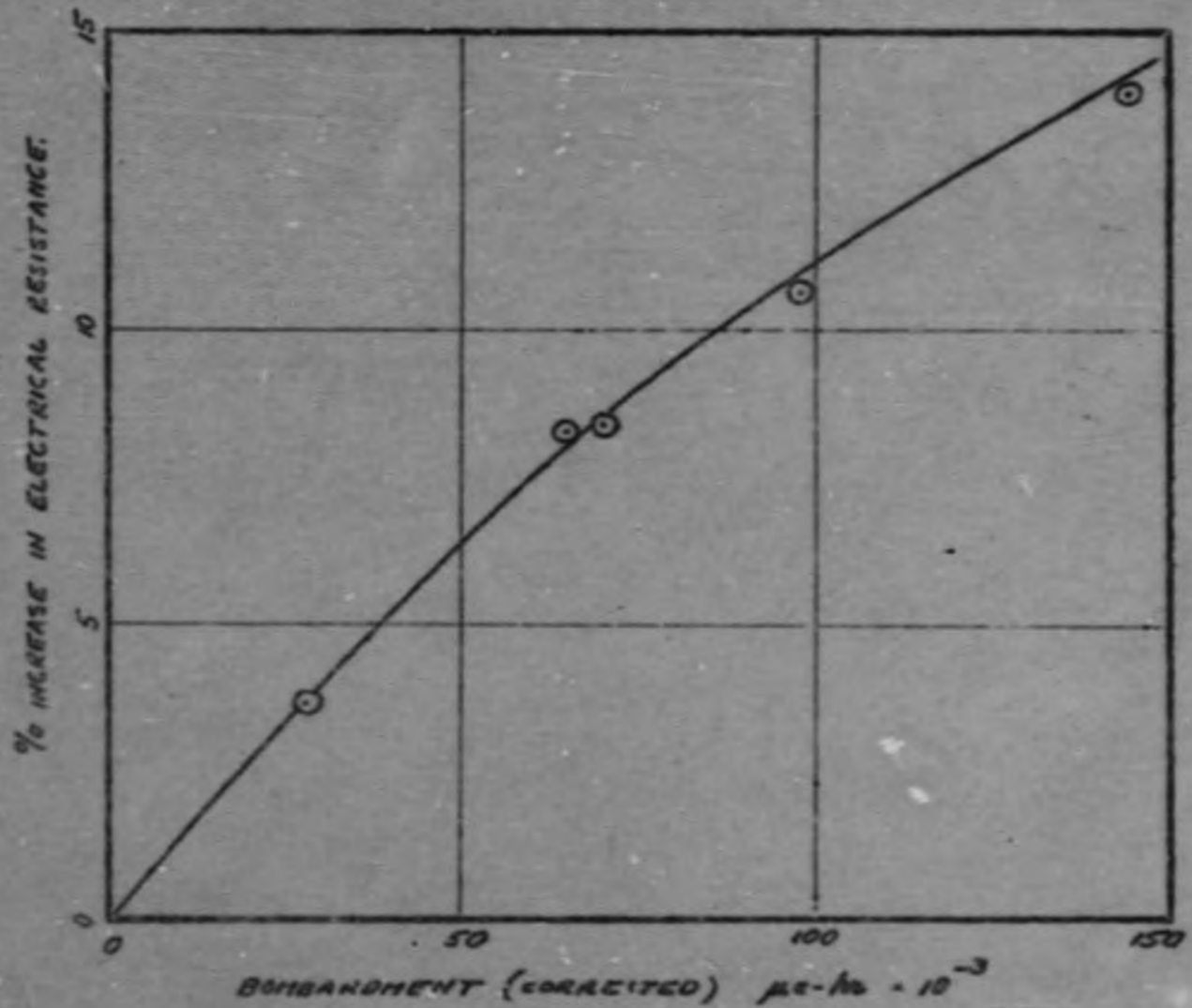

Figure 3.2 Effect of fast neutrons on the electrical resistance of AGOT graphite

673 719

From an examination of Figures 3.3 and 3.4 it is observed that as the crystal size or the crystal perfection increases, the elastic modulus increases and the electrical resistivity decreases. It is further seen that the Wigner effect as evidenced by elastic modulus measurements is correspondingly greater, the larger or the more perfect are the graphite crystallites.

The Wigner effect as it is now understood arises from the effects on the graphite lattice of the interstitial carbon atoms and the corresponding holes in the lattice which are produced in the process of moderating fast neutrons. The fate of a carbon atom when knocked loose from its normal position may be one of three possibilities:

1. It may return immediately to the position from which it was thrown or to an equivalent position.

2. It may lodge between the planes of the graphite lattice

3. It may come to rest at the edge of the crystal.

The existence of a neutron effect indicates that the first process is not the only one occuring. The distribution between the two remaining process appears to depend upon the relative surface to volume ratio of the individual crystallites. In a small crystal the chance of a knocked out carbon atom's reaching a crystal boundary is certainly greater than it would be in a larger crystal, where lodging between lattice planes would be expected to be the predominating process. Since a carbon atom coming on to the edge of the crystal produces only one disturbance, namely the hole in the lattice, while an interstitial carbon atom produces two disturbances centers, the process 2 is to be expected to cause a greater change in physical properties than process 3. Furthermore, in an imperfect lattice the effect of additional disorders caused by interstitial carbon atoms or the corresponding vacant lattice points are thought to make lesser contributions towards changes in physical properties than similar disorders in a more nearly perfect lattice. Both of these considerations correlate very well with the actual observations; see Figures 3.3 and 3.4.

3.1.3. Thermal healing of irradiated graphite

In CC-784 an experiment was described in which a bombarded test sample of AGOT graphite was heated at 3000°C in an inert atmosphere. Modulus measurements made before and after heating indicated that the treatment completely reversed the Wigner effect. The temperature, however, was so high that graphitization could occur and so there was very little chance that the graphite would not heal. In addition, Mulliken has pointed out, CP 773, that the binding energies of the interstitial carbons were sufficiently less than those of the normal atoms in the lattice that lower temperatures might be expected to be effective in healing. Furthermore, it was reported last month (CC-841) that β radiation exhibited an ability to heal neutron bombarded graphite.

673 020

Calculations based on beam dimension and sample size showed that the reversal was practically complete in the portion of the graphite exposed to the beam. From the current and the energy, further calculation indicated that this reversal was accomplished with an expenditure of 10 ev per carbon atom. More experiments are under way to determine how small the β irradiation can be and still reverse the neutron effect.

All these observations suggest that much lower annealing temperatures should be investigated. Accordingly, samples of AGOT and five of the special graphites were heated for one hour at 100, 200, and 500°C. Elastic modulus measurements were made before and after heat treatment. The resultant data are listed in Table 3.3. Experimental variation in measuring the change in elastic modulus is estimated to be ~± 3%.

In every case, the sample heated at 500°C was partially annealed. The extent of annealing is seen to be approximately proportioned to the magnitude of the neutron effect in the particular graphite sample. The heating at 200°C, in all but one case, caused the elastic modulus to increase. The cause of this change, if it is real, is not understood. The heating at 100° gave changes which were within the experimental variation.

Preliminary experiments, using bismuth-contact resistance samples to determine the mechanism and extent of the reversal of the Wigner effect by heating, have been inaugurated. Sample No. 10 was placed in an oven at 200°C, and sample No. 11 was similarly treated at 100°C. The samples were removed at intervals and cooled to room temperature for the resistance measurements. The percentage changes in resistance and the heating intervals are shown in Table 3.4. The resistance of sample No. 10 had increased 8.0% during neutron bombardment. Sample No. 11 had increased 8.2%.

Table 3.4 Thermal reversal of neutron induced resistance changes in AGOT graphite

Sample 10 (200°C)			Sample 11 (100°C)		
Heating Time	% Change	% of ine. lost	Heating Time	% Change	% of ine. lost
17 min.	-2.9	33	30 min.	-.6	7
32 "	-3.5	40	150 "	-.8	11
60 "	-3.7	43	336 "	-1.2	14
180 "	-4.0	46	1056 "	-1.5	18
365 "	-4.1	47	3755 "	-2.0	24
1086 "	-4.1	48			
3788 "	-3.9	45			

The data for sample No. 10 suggest that the process in the graphite that manifests itself by a decrease in resistance is approximately complete after 2½ hours. No change greater than experimental

Table 3.3 Heating Experiments
Effects on Elastic Modulus

Sample No.	Irrad. a-hr.	% Change in E	Temp. °C	% Change in E (heat)	Type of graphite
11	64	+22.5	500	-18.5	AGOT
12	64	+11.6	200	+ 1.6	
13	58	+12.3	100	+ 4.5	82½% annealed
14	0	~0	25	~ 0	after 1 hr. at 500° C.
28	59	+12.5	25	~0	AGOT(normalized)
30	58	+10.4	25	~0	78% annealed
32	58	+10.9	100	- .5	after 1 hr.
33	58	+ 9.7	200	+1.7	at 500° C.
34	58	+11.3	500	-8.8	
118	0	0	200	+ 7.0	Pitch-bonded
123	36	19.4	500	-17.4	Natural
124	36	22.1	200	+ 3.6	90% annealed
125	36	22.1	25	0	after 1 hr.
126	36	18.7	25	0	at 500° C.
104	36	+ 1.9	500	+ 4.6	Pitch-bonded
107	36	+ 1.9	200	+2.0	lampblack
111	0	0	25	~0	
90	36	+ 7.7	500	- 3.1	Special 2100°
91	36	+10.2	200	+ 3.7	graphite
92	0	0	25	+ 3.2	40% annealed.
63	36	+11.6	500	- 3.7	Special 2400°
64	36	+ 8.1	200	0	graphite
66	36	+ 16.0	25	0	
67	0	0	200	- 1.2	32% annealed
65	36	+ 10.2	25	0	after 1 hr. at 500° C.
48	36	(+33)	500	-15.2	Special 3000°
49	36	+ 20.1	200	—	graphite
50	36	18	25	—	
51	36	25.5	25	—	46% annealed
55	0	0	25	—	after 1 hr. at 500° C.

error occurs upon heating for an additional 60 hours. The fact that only about half of the neutron induced increase in resistance is lost during the 200° heating, suggests that there are, at least, two mechanisms necessary for a complete reverse, or healing.

Sample No. 2, with an increased resistance of 14.3% during neutron bombardment, was used in an attempt to measure resistance changes during the actual heating. Unfortunately, the temperature of the furnance used was not sufficiently uniform for the resistance versus time plot to have significance. The temperature was maintained for 19 hours at ~170°C. After cooling to room temperature it was found that the resistance of the sample had decreased 7.7%, or 54% of the neutron induced increase. An additional heating at 170°C for four hours caused no further change. This again suggests that the reverse of the effect as detected by resistance measurements, must proceed by at least two stages.

The resistance of sample No. 11 (heated at 100°C) is still dropping after 62.5 hours. It would be expected that if the observed increase in resistance were directly proportional to the number of displaced carbon atoms, that a treatment of the data by chemical kinetics methods would be possible. Unfortunately, the precision of the measurements ($\sim$.2%) is not great enough to permit these calculations for reversals of the magnitude thus far exhibited by sample No. 11. The heating of the sample is continuing.

It is hoped that "velocity constants" may be determined at different temperatures, and therefore permit an estimation of the activiation energy or energies for the reversal or healing of the Wigner effect. A correlation of the thermal energy and the minimum energy required for a reversal by electron bombardment should prove valuable in predicting the behavior of the graphite under pile conditions.

The heating experiments on the acoustical graphite samples do not support the data for the resistance experiments at 100° and 200°C. The conflicting results suggest the possibility that the changes which are detected by electrical resistance and resonance frequency measurements are not identical.

3.1.4. The monitoring experiments

The fast neutron monitoring of the targe area of the St. Louis cyclotron was done with aluminum. Aluminum, upon bombardment with fast neutrons, undergoes an (n, α) type reaction giving Na^{24} with a 14.8 hour half-life. The other possible reactions (n,p), (n, 2n) and (n, γ) give products with very short half-lives. It was therefore rather easy to count the activity of the Na^{24} without interference, and since the threshold for the (n, α) reaction is $\sim$ 2.5 Mev, the back scattering of slow neutrons from the metal and metal compound surrounding the target is not important.

A sheet of 2 mil aluminum foil 9 cm. x 18 cm. was placed directly against the brass back-plate of the cyclotron target, bombarded for 27,000 μa-hr and removed. The foil was then cut into pieces 1 cm^2

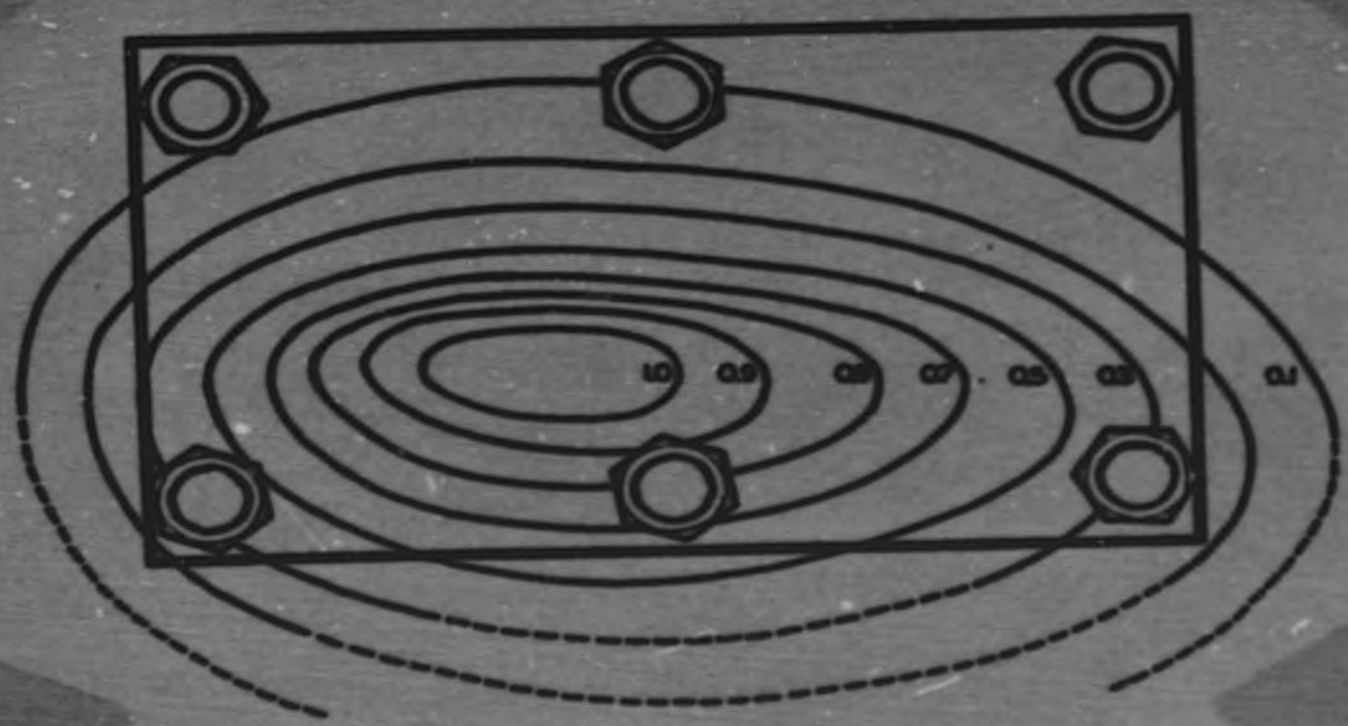

Figure 3.6 Relative fast neutron intensities on the target of the St. Louis cyclotron. (diagram in full size).

and the relative activities of the pieces determined by the use of a Lauritsen type electroscope with a thin window. From the activities of the foils and their relative positions, a contour diagram of relative fast neutron intensities was prepared. Figure 3.6. Making use of this diagram and the positions which the various tubes occupied during irradiation, the corrections mentioned in Section 3.1.1 were deduced.

3.1.5. Surface area and pore volume measurements

More precise measurements have been made of the rate of diffusion of helium into graphite. A cylinder of AGOT graphite, one centimeter in diameter and four centimeters in length, was degassed at 1500C at 10^{-6} m.m. Hg for twelve hours. The sample was, in effect, immersed in helium kept at constant pressure, and the volume change of the helium at this pressure determined as a function of time. The study was made at 37.0°C ± .05°C . Pressure was held constant to 0.1 mm. The table below illustrates the results obtained on one sample at three different pressures.

Table 3.5 Variation of the pore volume of AGOT graphite

Time	0 hr.	1 hr.	2 hr.	4 hr.	8 hr.	24 hr.	48 hr.	
Δ V	0	.010	.015	.021	.035	.048	.060	p = 251 mm.
Δ V	0	.010	.015	.018	.025	.043	.056	p = 339 mm.
Δ V	0	.010	.013	.018	.025	.037	.055	p = 478 mm.

Initial volume of the graphite = 2.410 cm^3
Weight of graphite = 5.064 gm.
D = weight of graphite / (initial graphite volume - ΔV)

It is hoped that data of this type can be used with adsorption data to determine the pore structure of graphite.

The nitrogen adsorption at liquid nitrogen temperature has been measured for one sample of AGOT graphite. S-shaped isotherms were obtained which give a surface area of 0.480 ± 0.008 square meters per gram by the Brunauer, Emmett, and Teller equation.

673 025

Group IV W. L. Kay

Table of Contents

• • • •

4.1 The Effect of Radiation on Optical Glasses (W.L.Kay, B.Bluestein)

Several additional samples of optical glass, as well as various types of light bulbs, have been subjected to X radiation at the M.D. generator. The light bulbs were exposed so that the front faces were completely covered with an average intensity of the amount shown below in Table 4.1. The bulbs were unlit during exposure, but no trouble was experienced in lighting them after exposure. These results are valid only for the bulbs tested as the glass in various bulbs of the same type might differ widely in properties.

Table 4.1 Effect of X Radiation on Optical Glasses and Electric Light Bulbs

Sample	Intensity Mv	Current μa	Dosage R/min.	Time Min.	Color	Remarks
Medium Barium Crown Index D line 1.572 Dispersion 57.4	1.3	15	5000 at target sample 0.5 mm from target	5 10 15	yellow dark yellow brown	⅛ in. penetration of col. " " " " 1 in. " "
Boro Silicate Crown Index D line 1.517 Dispersion 64.5	"	"	"	5 10 15	no color " " light yellow	 shallow penetration of col.
Extra Dense Flint (yellow at start) Index D line 1.909 Dispersion 21.5	"	"	"	5 10 15	brown " dark brown	1 in. penetration of col. " " " " " " " "
Dense Barium Crown Index D line 1.638 Dispersion 56.5	"	"	"	5 10 15	slight color sl. black gray black	on edges 1/8 in. penetration of col. ¼ in. penetration of col.
Special Short Flint Index D line 1.613 Dispersion 44.2	"	"	"	5 10 15	sl. brown " " " "	⅛ in. penetration of col. ¼ in. penetration of col. 1 in. " "
Extra Dense Flint Index D line 1.649 Dispersion 33.8	"	"	"	5 10 15	sl. brown brown dark brown	1 in. penetration of col. 1 in. " " 1½ in. " "
Light Barium Crown Index D line 1.572 Dispersion 57.2	"	"	"	5 10 15	sl. brown brown dark brown	⅛ in. penetration of col. 1 in. " " 1½ in. " "
Barium Crown X lense sample	1.5	14	"	5 10 15	only sl. col. yellow yel. brown	 1/8 in. penetrat. of col.
Barium Crown X lense sample	"	"	"	3	light yellow	
Barium Crown X lense sample	"	"	"	1	very sl. color	
Dense Flint X lense blank	"	"	"	6	dark brown	¼ in. penetra. of color
Dense Flint X lense blank	"	"	"	2½	"	¼ in. penetra. of color
Dense Flint X lense blank	"	"	"	½	"	1/8 in. penetra. of color
Achromat lens cemented with Canada balsam	1.7	30	5000 at 1 cm from target where sample located	5 10	yellow-brown brown	1/8 in. penetra. of col. ¼ in. penetra. of color
Lucite	"	"	1000 at 5 cm from target 5000 at 1 cm from target	20 10	no change no change	 sample moved up toward target
Westinghouse Reflection Spot	"	"	1000 at 5 cm from target	20	very slight yellow col.	exposed front end on
Westinghouse Projector Spot	"	"	"	"	"	"
Sunlamp - S-1 Type	"	"	"	"	"	"
Mercury Lamp Type A-H-2 (101-13-Q)	"	"	"	20 35	no change no change	exposed on side exposed on end
Mercury Lamp Type A-H-3	"	"	"	30	sl. yellow	mica internal support slightly brown.

4.2 Solubility of NO and Xe in Various Solutions

The problem of recovering radioactive Xenon from the gas obtained by solution of the pile metal in nitric acid was mentioned in Report CC-841, p-21 and various solubilities of air, Xe, and NO were listed for evaluating possible recovery schemes. Additional data are tabulated below and summarize the latest work concerning nitric oxide and Xenon solutilities.

Table 4.2 Solubility of Nitric Oxide and Xenon

Gas	Solution	Concentration	Solubility $\frac{V_{gas}}{V_{liquid}}$	Temp°C	Remarks
NO	$Fe_2(SO_4)_3 + H_2SO_4$	33g $Fe_2(SO_4)_3$ in 100 g 2.25 N H_2SO_4	~ 0.2	28	
NO	$FeSO_4 + H_2SO_4$	15 g $FeSO_4$ in 100 g 2.25N H_2SO_4	7.2 6.6 6.8	27 30 31	Warming or allowing solution to stand expelled all the dissolved NO, and the original solution could be obtained unchanged.
NO	$FeSO_4$	25 g $FeSO_4$ in 100 g H_2O	9.2	31	Behavior similar to $FeSO_4$ H_2SO_4, except recovered solution after expelling NO always contained some Fe^{+++}
NO	$CrO_3 + H_2SO_4$	20 g CrO_3 in 100 g 40% H_2SO_4	60 57	30 30	End solution was deep green showing complete reduction to Cr^{+++} No ppt was formed.
Xe	CCl_4	100%	2.4 2.9 2.4 2.3 1.3*	32 31 28 31 30	*This value reported earlier is evidently in error.

673 C28

Group V M. Burton

Table of Contents

* * * * *

5. 1.25 Mv electrostatic generator

The 1.25 Mv generator is operating satisfactorily in the production of β radiation up to a maximum of 60 μa at rated voltages.

END