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GRAPHITE INTERFACE STUDIES

TECHNICAL SUMMARY REPORT

May 16, 1963 through May 15, 1964

Part I

Contract AT(04-3)-167
Project Agreement No. 22
U.S. Atomic Energy Commission

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May 15, 1964

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DIVISION OF
GENERAL DYNAMICS

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I. INTRODUCTION

This Technical Summary Report is published in two parts, each under separate cover, in order to facilitate handling of the unclassified portion of the document, which is contained in this Part I.

Before the work on this project was started, considerable time and effort had been expended at General Atomic to develop a realistic thermalization model for graphite. This included both theoretical as well as experimental work. (1-5)

However, it was known that additional work should be performed for systems having rather severe design limitations in order to insure justification of any conclusions drawn from an analysis based on crystalline graphite kernels calculated from the SUMMIT code. Reactors of the Kiwi class appear to be just such systems because of their low C/U ratios in the core and their rather thick graphite-beryllium reflectors. In addition to the sharp discontinuity in absorption cross section, there is a tremendous temperature difference between the core and the reflector when a Kiwi reactor is at power. This is due to the extremely high core temperature and the unusually low reflector temperature arising from the passage of liquid hydrogen through the latter. In these reactors the power generation near the core-reflector interface has been adjusted by extensive zoning. Power distribution measurements have been made on room temperature assemblies; however, to our knowledge measurements have not been made on the hot assemblies. Hence, the hot power distribution must rely heavily on calculations. The zoning is an expensive procedure and the extrapolation to the hot-core cold-reflector core is a long one so that it was felt that an additional effort was warranted to check out the graphite thermalization

model, both experimentally and theoretically.

The program chosen to perform this work was based mainly on the facts that the budget was limited and that the experimental difficulties in mocking up a section of a Kiwi-type core were rather formidable.

It was felt that a better approach would be to perform a series of "clean" one-dimensional experiments of increasing complexity that would check various features of the problem. A careful comparison of experimental and analytical results should then answer the question: Are the SUMMIT type crystalline kernels used in a multigroup S_n transport formulation capable of sufficient accuracy to predict thermal neutron spectra near absorption and temperature discontinuities? We believe that the answer is encouraging, and this report is submitted in support of that belief.

II. EXPERIMENTAL METHODS

Since all of the experiments performed under this contract were of a similar nature, a general discussion of the facilities and the experimental techniques employed introduces this section.

Briefly, fast neutrons are thermalized in a graphite assembly and the 90° angular flux spectrum is determined by measuring the time it takes for neutrons to travel from the assembly to a detector located approximately 50 meters away.

The neutrons used in these experiments were obtained by bombarding a uranium target with a beam of electrons from the General Atomic Linear Accelerator. Figure 1 shows the Linac facility, and some of its operating parameters are shown in the inset. As the electron beam slows down in the target its kinetic energy is converted to bremsstrahlung which in the case of uranium then produces fast neutrons primarily by photofission. The fast neutron spectrum obtained in this way is quite similar to neutron induced fission spectra. Typical operating values for the Linac for this series of experiments were: Energy: 28 Mev., Current: 0.4 amp, Pulse Width: 4.5×10^{-6} sec. and, Repetition Rate: 15 pulses per second. This results in an energy deposition rate of approximately 0.76 kilowatts and since the energy is deposited over a small area the target was cooled by circulating water.

Figure 2 shows a typical arrangement used for measuring the thermal neutron spectra in graphite assemblies. The lead shielding between the graphite and target is used to reduce the γ -flash that is seen by the detection equipment at the 50-meter station and also to improve

ELECTRON LINEAR ACCELERATOR

Fig. 1--Schematic diagram of General Atomic 45-Mev electron linear accelerator

Fig. 2--Typical Experimental Layout

the distribution of fast neutrons on the edge of the assembly. Further details on each individual assembly will be given later.

At this point it seems desirable to answer three questions. First, how was the size of the assembly selected; second, what perturbation does the re-entry hole have on the measured spectrum; and third, is the time average of the time-dependent spectrum equivalent to the spectrum that would be obtained from an identical constant source?

The selection of an assembly size is based on a compromise after making the following considerations. A large assembly has the advantage that neutron leakage is reduced. This of course makes the spectrum more dependent on the absorption and scattering properties of the medium and less dependent on leakage. The disadvantages of the larger assembly are: (1) The flux shapes in the directions perpendicular to the beam tube axis depart from the fundamental or cosine mode and are more difficult to handle analytically; (2) The mean emission time for a neutron to leave the assembly becomes longer in lightly poisoned systems and, hence, necessitates a sizable correction to the flight time; and (3) The problems of heating, cooling and maintaining thermal equilibrium of the system become more severe.

With respect to the first question, an estimate of the higher modal effects in an assembly of the size chosen can be roughly estimated as follows. For simplicity, consider a rectangular prism of graphite of sides $2a$, $2b$ and $2c$. A point source of fission neutrons is placed at the origin. The particular solution of the age equation that satisfies the boundary condition at the origin and at the extrapolated boundaries X_0 , Y_0 , Z_0 .

$$Q(X=0, Y=0, Z=0, \tau=0) = Q_0 = \text{Constant}$$

$$Q(X=X_0, Y=Y_0, Z=Z_0, \text{all } \tau) = 0$$

is

$$Q(X,Y,Z,\tau) = \frac{Q_0}{X_0 Y_0 Z_0} \sum_{\ell, m, n \text{ odd}} \exp \left[e^{-\frac{\pi}{4} \left(\frac{\ell^2}{X_0^2} + \frac{m^2}{Y_0^2} + \frac{n^2}{Z_0^2} \right) \tau} \right] \\ \cdot \cos \left(\ell \frac{\pi}{2} \frac{X}{X_0} \right) \cdot \cos \left(m \frac{\pi}{2} \frac{Y}{Y_0} \right) \cdot \cos \left(n \frac{\pi}{2} \frac{Z}{Z_0} \right)$$

The amplitude of the higher modal terms is determined by the factor

$$\exp \left[e^{-\pi^2/4 \left(\frac{\ell^2}{X_0^2} + \frac{m^2}{Y_0^2} + \frac{n^2}{Z_0^2} \right) \tau} \right]$$

The terms with $\ell = m = n = 1$ represent the fundamental mode. The next larger terms will be those with $\ell = 3, m = 1$ and $\ell = 1, m = 3$. (The Z direction can be handled explicitly in the one dimension transport analysis used:) For the assembly chosen $X_0 = Y_0 = Z_0 \approx 30$ cm. and $\tau(2.38 \text{ ev}) = 309 \text{ cm}^2$. The ratio of the amplitudes for each of the next harmonic terms to the amplitude of the fundamental terms is therefore,

$$\frac{e^{-\frac{\pi^2}{4} \left(\frac{9}{900} + \frac{1}{900} + \frac{1}{900} \right) \tau}}{e^{-\frac{\pi^2}{4} \left(\frac{3}{900} \right) \tau}}$$

$$\approx e^{-6.8} = .0011$$

It is then apparent that for the assembly size chosen, the higher modal terms will give a negligible contribution in the transverse directions. In the actual case, because the source is located at the edge of the assembly, the result should be somewhat different than the above; however, this is believed to be a good estimate in the X and Y directions.

The question as to how much perturbation the re-entry hole introduces can only be answered from past experience.⁽⁷⁾ This has shown that fairly large, up to 2 x 2 inches re-entry holes can be used without distorting the thermal spectrum in polyethylene assemblies. In view of the high age of graphite and the moderately long diffusion length in these assemblies it is believed that no appreciable perturbation is introduced by the 1 x 1 inch re-entry hole.

With respect to the third question, it can easily be shown that if the time dependent Boltzmann equation with a delta function source at $t = 0$ is integrated between times zero and infinity, the equation of the corresponding stationary problem is obtained. This of course assumes that the assembly is not heated by the pulse. The time of flight technique does indeed perform this integration although care must be exercised when comparing experimentally measured spectra with an analysis based on a stationary calculation since the neutrons do not all have the assembly at time zero. That the source has a finite burst width is a negligible correction for the neutron energies considered here. This matter is discussed in detail in Section III.

Thermally black boundaries were obtained on each assembly by a 0.020 in. cadmium cover or 1/4 in. boral plate if any heating was involved. The poisoning was introduced by stacking poisoned foils between alternate layers of graphite instead of a homogeneous dispersion. This was done for convenience in exchanging poison materials and concentrations, but since

spectra are calculated for homogeneous systems, the calculations must take into account the disadvantage factors associated with such lumping. In some experiments, flux plots were taken along the central source axial direction and along a horizontal center axis in the flight path direction by means of cadmium covered indium foils. The purpose of these plots was to provide a basis for the distributed source to be used in the calculations and to allow the transverse buckling of the system to be determined. However, in experiments involving hot graphite, this could not be done since the cadmium would melt and vaporize. In these cases, the assumption was made that the room temperature flux plots in the assembly were the same as those at temperature. The indium resonance energy of 1.4 ev is sufficiently high that the neutron flux there is not affected much by the moderator temperature, and the assumption is therefore reasonable. Calculations have shown this to be true.

The neutrons leave the assembly through the re-entry hole and travel down the fifty meter flight path. The entrance to the flight path consists of a 1/2 in.-diameter precollimator of borated epoxy resin. Intermediate collimation along the flight path is obtained by borated paraffin inserts. The collimation defines a beam diameter of 29 in. which is slightly smaller than the sensitive area of the detector bank.

At the beginning of this program the flight path was filled with helium gas to reduce the attenuation of neutrons and to reduce the changes in flight path transmission that would arise from variations in water vapor content if air were used. Before the last two experiments were performed a new flight tube was installed which could be evacuated to pressures below one micron, and which had a higher transmission.

The neutron detection system located at the far end of the flight path consists of a bank of 36 BF_3 counters. This bank has been energy

calibrated by comparing its response to a given flux to that of a standard detector for the same flux. The sensitivity of the detector bank is known to within 15% over the energy range 0.001 to 100 ev.

Signals from the BF_3 counters are amplified and used as signal input to a 1024 Channel, TMC Model 212 time-of-flight analyzer. The time zero signal for this analyzer was obtained, from a small NaI photomultiplier system in the assembly room. This system responds to the γ -flash after each electron burst and thus corresponds to the generation of neutrons in the target. A modified RCL time-of-flight analyzer was also used as a backup unit.

The neutron production is monitored in several ways. First, by locating a small fission counter in the assembly room, the short time-averaged neutron production rate was monitored on a count rate meter. This same signal was also used to drive a scaler which gave the total integral count for each run. In addition, gold foils were activated to obtain a further check of the integral neutron count.

In making spectrum measurements by the pulsed technique, there was a nonnegligible background of neutrons that reach the detector bank from locations and paths that are different than those desired. In order to correct for this background and any other background effects that have a constant time average, a B^{10}C plug was placed in the re-entry hole at the centerline of the assembly. The assembly was then pulsed under the same conditions present when making an actual spectrum measurement so that the energy independent background could be obtained. Figure 3 shows a block diagram of the electronic units comprising the recording system.

The experimental data obtained in these runs were reduced using the code ECTOPLASM. This code performs the following functions:

Fig. 3-- Block diagram of electronic units for spectral measurements.

1. Converts the measured time spectrum to an energy spectrum
2. Subtracts the measured background spectrum
3. Applies the mean emission time correction
4. Applies corrections for counting rate losses in the detector and monitor systems.
5. Applies the detector sensitivity correction combined with a flight path transmission function.
6. Groups the channels into a more convenient energy mesh
7. Computes the statistical error.

III. ANALYTICAL METHODS

All of the assemblies in which thermal neutron spectra were measured were analysed in order to obtain a comparison between theory and experiment.⁽⁸⁾ This was done by using the multigroup one-dimensional S_n transport code GAPLSN which is a modified version of the Los Alamos developed code DSN. GAPLSN solves by a numerical integration procedure the time-independent form of the neutron transport equation for a one-dimensional problem. The code is written for rectangular, spherical and cylindrical coordinate systems. In the code, the scattering cross section and the external source may have anisotropy represented by an arbitrary number of angular moments. The energy range of the problem is determined by the cross sections fed in as input. Provision is made for a variety of boundary conditions and for distributed or external sources. The code will calculate the flux distribution, the multiplication factor K , or it will adjust some other parameter until K takes on a prescribed value. GAPLSN is written principally in the FLOCO II language and its versatility lies in its ability to vary its data storage array.

The fundamental question that must be answered when using a one-dimensional code to analyse a three-dimensional assembly is: Are the approximations which are made to incorporate the effects of the other two dimensions in the one-dimensional structure valid? In our assemblies the problems were primarily those of incorporating transverse leakage and of using the scalar flux to approximate the 90° angular flux.

The flux shape in the transverse directions has been measured experimentally on several of the assemblies by activating cadmium covered indium foils. This has shown the transverse fluxes to be in a fundamental cosine mode throughout most of the assembly. Deviations do occur near the source end;

however, they are relatively small. This being the case, the diffusion theory approximation has been used so that the leakage is given by $D(E) \nabla^2 \phi(E)$ giving macroscopic leakage cross sections $D(E)B^2$ along the center line of the assembly where $D(E)$ is the diffusion coefficient and B^2 the transverse buckling. These cross sections were added to the total cross section in each energy group. This would seem to be a satisfactory procedure, however, some difficulty was encountered near the interface region. Since most of the leakage occurs in the energy range where the absorption cross section is low, 0.5 to 2.38 ev, there is no problem as long as the graphite number density is approximately the same on both sides of the interface even though there may be an appreciable difference in $\sum_a$ at the low energies. However, in cases where there is a gap of low density material such as carbon felt between the assembly halves it is not clear as to how one assigns a value to $D(E)$ in this region. In particular for $N \rightarrow 0$, $D(E) = \frac{1}{3 \sum_{TR}}$ becomes infinite giving an infinite macroscopic leakage cross section. It is therefore necessary to use some average value of D based on average number densities of the materials in the neighboring regions out to perhaps a diffusion length. Because the gap was quite small in most of this series of experiments this has not been carried out but rather the average D for the materials in the adjacent halves was used without attempting to volume weight on the basis of diffusion lengths. In any event the uncertainty is believed to be a small one.

That the scalar flux is a good approximation to the 90° angular flux may be seen by expanding the latter as

$$\Phi(\vec{r}, e, \vec{\Omega}) = \frac{1}{4\pi} \left[\phi(\vec{r}, E) + 3\vec{\Omega} \cdot \vec{J}(\vec{r}, E) \right] + \sum_{\ell \geq 2, m} \left(\frac{2\ell+1}{4\pi} \right) f_{\ell m}(\vec{r}, E) P_{\ell m}(\vec{\Omega})$$

Since $J_x = J_y = 0$, $\vec{\Omega} \cdot \vec{J} = 0$ at the 90° angle. The higher order terms in the summation do not have any clear physical definition. That they must be small can be seen from the following argument. When GAPLSN calculations were run with a higher order S_n approximation there was very little change in the scalar flux as compared with the lower order calculations and the angular fluxes near 90° differed from the scalar flux only slightly in magnitude.

Another difficulty that is encountered in setting up these calculations lies in providing a source of neutrons. This problem cannot be adequately handled in one-dimensional calculations by starting with a plane fission source at the assembly boundary **because the neutron wave** originates from a point source and is still essentially spherical when it strikes the assembly. In addition, the fast leakage corrections would be very difficult to make. Instead, the calculation is started at a lower energy using a source shape obtained by activating cadmium covered indium foils placed along the Z axis of the assembly. In some of the early analysis this source shape was placed in the top energy group and a GAPLSN Q calculation performed. This seemed to be a satisfactory procedure in view of the fact that the maximum energy transfer αE was less than the group width in this energy region. This procedure was refined later in the following manner. After having run a GAPLSN calculation in the manner described, the indium activation was calculated from the results and an adjustment was made in the Q source so that on re-calculating, the measured and calculated indium activation would be nearly identical. The lumped indium cross sections used were obtained from a fine mesh GAPLSN cell calculation that included the cadmium covers. Since the changes in Q necessary to obtain agreement in the indium activation were small and primarily near the source side boundary where one dimensionality is at its poorest it is believed that this procedure

was justified especially since the resulting spectrum changes were small.

In the experiments where there was a carbon felt heat barrier between the halves, the source term was reduced by the ratio of the carbon number density in the felt to the carbon number density in the adjoining graphite. This produced only a slight change in the spectrum. Finally, in order to obtain the P_1 component of the source the diffusion approximation was invoked and by differentiating the measured curve a term of the form $D\nabla^2\phi$ was obtained. This term was small and produced only a very slight effect on the calculated spectra.

Cross sections for these calculations were obtained using the GGC II code⁽⁹⁾ which contains the GATHER II thermalization code⁽¹⁰⁾ and which automatically punches the cross sections for GAPLSN.

The GATHER II code allows for the calculation of the regular and/or adjoint fluxes using the B_1 , B_0 , P_1 or P_0 approximations to the static Boltzmann transport equation. In our case, the B_1 approximation is usually used. The code makes use of a library data tape of cross sections and scattering matrices for various materials over 101 energy points ranging from 0.001 to 2.38 ev. The library data tape is derived from a master tape and contains in our case kernels for graphite at various temperatures calculated from the SUMMIT⁽⁶⁾ code plus all of the necessary absorber cross sections. The code calculates a spatially independent flux spectrum over these energy points taking into account the local total buckling. Cross sections of interest in the problem, both P_0 and P_1 components, are averaged over this spectrum into a fewer number of groups. This fewer number of groups is necessitated by the storage limitations of the computer in the calculation of the space-dependent problem, and generally 25 to 32 were used depending on whether the poisoning materials were $1/v$ or resonance

absorbers. Because of the large number of groups the dependence of the cross sections on group structure is minimized.

SUMMIT is a computer code written to calculate the differential energy transfer cross section for scattering by a crystalline material. It accounts for the inelastic incoherent components. To obtain the total differential scattering cross section, the elastic coherent component determined from the code ELASTIC⁽¹¹⁾ is added to the output from SUMMIT. In the earlier versions of SUMMIT, generation of the scattering kernel was lengthy and expensive, and the practice was to attach differential cross sections based on a free gas kernel at some appropriate temperature to the SUMMIT output above ~ 1 ev. This procedure has appeared to work quite well.

In cases where the poison foils contained enough absorber to depress the flux, cell calculations were run to obtain disadvantage factors. These factors were then used in the GATHER calculations when obtaining macro cross sections.

The justification for the pulse-time of flight procedure of measuring thermal neutron spectra is dependent first on the assumption that the neutrons slow down, thermalize, and leave the assembly in a relatively short time compared to the flight time and second that an adequate correction can be made to the flight time of account for the fact that all of the neutrons do not leave the assembly at the same time. The selection of a modest size assembly with an appreciable poison content assures that the correction will be a small one. The method of making this correction has been examined by Koppel.⁽¹²⁾ Without going into all the details of his procedure suffice it to say that for the assemblies discussed in this report, the mean emission time provides a small correction, (less than 3-1/2%). For the semi-infinite medium cases this correction was obtained from the GATHER II code, which

incorporates the following equations of the order P_0 . Defining the mean emission time as

$$T(E) = \frac{\int_0^{\infty} t \cdot \Phi(E, t) dt}{\int_0^{\infty} \Phi(E, t) dt} = \frac{\Phi_1(E)}{\Phi_0(E)}$$

The solution $\Phi_0(E)$ is obtained as a solution to the steady state balance equation

$$(DB^2 + \sum_T) \Phi_0(E) = \int \sum_0^0 (E' \rightarrow E) \Phi_0(E') dE' + Q_0(E)$$

The solution $\Phi_1(E)$ is obtained by multiplying the time dependent equation for a pulsed source at $t = 0$

$$-\frac{1}{v} \frac{d\Phi(E, t)}{dt} = (DB^2 + \sum_T) \Phi(E, t) - \int \sum_0^0 (E' \rightarrow E) \Phi(E', t) dE' + Q_0(E) \delta(t)$$

by t and integrating from $t = 0$ to $t = \infty$.

The resulting equation is then

$$(DB^2 + \sum_T) \Phi_1(E) = \int \sum_0^0 (E' \rightarrow E) \Phi_1(E') dE' + \frac{\Phi_0(E)}{v}$$

which is of the same form as the preceding equation with the source replaced by $\Phi_0(E)/v$. Since the GATHER energy mesh goes only to 2.38 ev, a constant slowing down time correction must be added to the $T(E)$ calculated in this way. This correction is essentially the time it takes a neutron to travel its last mean free path before reaching 2.38 ev, and is very well approximated by

$$t = \frac{2}{\xi \sum_0 (2.38 \text{ ev}) v (2.38 \text{ ev})}$$

with the usual notation. The slowing down time is about 16 μ secs so that the total mean emission time amounts to a correction of a few percent in the semi-infinite medium cases.

When the first experiment with an absorption discontinuity was analyzed, consideration was given to the possibility of distorting the spectrum because of the longer lifetime of neutrons in the unpoisoned graphite side. For this case the GAPLSN equations were used in the same manner as described above to determine the spatially dependent mean emission time. The results indicated that there was indeed a spatial dependence but that the deviations from the average were not large enough to warrant the calculational expense of proceeding in this manner.

IV. EXPERIMENTAL AND ANALYTICAL RESULTS

This section discusses the experiments and the analyses performed to test the crystalline scattering kernel for graphite, leading up in a step by step manner to measurements of spectra at an absorption and temperature discontinuity.

The first series of experiments performed was made to measure neutron spectra in poisoned graphite under quasi-infinite medium conditions. The purpose of these tests was to resolve a discrepancy between spectral measurements made here at General Atomic and those made at Harwell in England. It was also intended to check our calculational methods in a simple geometry with experimental techniques which were refined since earlier measurements.⁽²⁾ The graphite was poisoned by placing 0.010 in. of 1% wt. samarium loaded aluminum between layers of graphite 0.525 in. thick, stacked into a 2 ft. cube. Samarium was selected since it was anticipated that its 0.096 ev resonance would provide a clear test of the bound kernel vis-a-vis the free gas kernel for graphite. The assembly was designed to be cooled with liquid nitrogen down to 80°K and heated electrically to 600°K. Figure 4 illustrates the over-all arrangement of the assembly and Fig. 5 which shows the corresponding calculational geometry reveals the basis for calculations of the later experiments. Table 1 details the parameters for the first set of experiments shown in the Figure.

Table 1

CONFIGURATION PARAMETERS FOR CASES A,B, AND C

Region	Number densities Atoms/barn-cm	Case No.	Temp. °K	Hole No.	d ₁ Inches	d ₂ Inches
I and II	C 0.0775 Al 2.23x10 ⁻³ Sm 2.08x10 ⁻⁶	A	300	1	4	--
				2	0	--
		B	80	1	0	--
				1	0	--
III	t = 0	C	600	1	0	--

Fig. 4--Construction details of low and high temperature graphite assembly.

GRAPHITE STACK HEIGHT = 24 IN.

RE-ENTRANT HOLE AT POSITION d_1 OR d_2

PULSED SOURCE ON LEFT HAND SIDE OF ASSEMBLY

Fig. 5--Calculational geometry for spectral measurements in graphite

Spectrum measurements were made at 90° to the pulsed neutron source at the temperatures and positions listed in Table 1. The results for these three cases are shown in Figures 6 through 9, and each measured spectrum shows good agreement with the spectrum calculated using the bound crystalline kernel for graphite. Figure 7 also shows the spectrum calculated using the free gas kernel and hardly any evidence of the samarium resonance is observed in that spectrum.

The good agreement between theory and measurement in this first series of experiments gave us confidence for proceeding to an absorption discontinuity. Half of the samarium was removed from the assembly and spectra were measured at three locations. Table 2 details the parameters of this set of measurements.

Table 2
CONFIGURATION PARAMETERS FOR CASE D

Region	Number densities Atoms/barn-cm	Temperature $^\circ\text{K}$	Hole No.	d_1 Inches	d_2 Inches
I	C 0.0775 Al 2.23×10^{-3} Sm 2.08×10	300	1	3-1/2	--
II	C 0.079		2	1	--
III	$t = 0$		3	--	1

The measured and calculated spectra are shown in Figure 10; they show excellent agreement in this spatially dependent situation.

The success at predicting spectra at this absorption discontinuity allowed us to proceed to the next experiment in which the poison concentration was increased by placing samarium-loaded aluminum foils at every 0.230 in. instead of every 0.525 in. It was expected that this would

Fig. 6--Neutron spectra in poisoned 300°K graphite, case A, at $d_1 = 4$ inches, hole 1.

Fig. 7--Neutron spectra in poisoned 300°K graphite, case A, at $d_1 = 0$ inches, hole 2.

Fig. 8--Neutron spectrum in poisoned 80°K graphite, case B, at $d_1 = 0$ inches, hole 1.

Fig. 9--Neutron spectrum in poisoned 600°K graphite, case C, at $d_1 = 0$ inches, hole 1.

Fig. 10--Neutron spectra in graphite interface experiment D at room temperature.

substantially enhance the positional dependence of the neutron spectra. The assembly was then modified so that the unpoisoned side was nearest to the source. It was anticipated that this arrangement would correspond more closely to the core reflector interface problem in which the gradient of the thermal neutron flux is from the unpoisoned reflector towards the heavily poisoned core. In addition, a 1/4 in. sheet of graphite felt was inserted between the two halves of the assembly. The felt would be needed when a temperature difference was incorporated and it was desirable to check whether it affected the calculations at this stage. The parameters of this experiment relative to Figure 5 are given in Table 3.

Table 3

CONFIGURATION PARAMETERS FOR EXPERIMENT E

Region	Number densities Atoms/barm-cm	Temperature °K.	Hole No.	d ₁ Inches	d ₂ Inches
I	C 0.079	300	1	1	--
II	C 0.077 Al 4.40×10^{-3} Sm 4.12×10^{-6}		2	--	1
III	t = 0.25 inches C 0.0039		3	--	3

The results of this experiment, shown in Figures 11, 12, and 13, were not as good as had been observed in earlier experiments. It was not clear what the error was but, at the time, segregation of the samarium in the aluminum seemed a possibility. It was therefore decided to return to the lower samarium concentration in the next experiment. The error was probably due to the felt not being treated properly in the calculations for reasons which are discussed later. It has not been considered worthwhile to repeat the

Fig. 11--Neutron spectrum in room temperature interface experiment, case E, at $d_1 \approx 1$ inch, hole 1,

Fig. 12--Neutron spectrum in room temperature interface experiment, case E, at $d_2 = 1$ inch, hole 2.

Fig. 13--Neutron spectrum in room temperature interface experiment, case E, at $d_2 = 3$ inches, hole 3.

calculations of this experiment because of the expense involved and the small return of improving the agreement between theory and measurement. The disagreement is greatest at hole 3 which is furthest into the poisoned side, and reduction of the leakage cross section in the carbon felt would probably soften the calculated spectrum only slightly at this location

For the next experiment (F) an assembly was constructed, see Figure 14, in which the poisoned side could be heated electrically, and the unpoisoned side could be cooled by circulating cold alcohol through outer cooling tanks. In this assembly two sides were also separated by a $1/4$ in. sheet of graphite felt. Unfortunately the cooling system proved unsatisfactory since it did not keep the temperature low enough. In addition, and far more serious was the fact that the hydrogenous material in the cooling tank greatly affected the flux plot shape and effectively provided a boundary source of neutrons that could not be measured. Figure 15 shows the flux plot shape of the experiment as compared with the shape of a later experiment in which the outer cooling tanks had been removed. It was decided to modify the cooling system for subsequent experiments. Despite the distortion caused by these tanks spectra were measured with the unpoisoned side cooled to 274°K and the poisoned side heated to 600°K . Table 4 details the configuration parameters of this experiment. The results of these measurements did not agree with the calculated values as well as previous experiments had agreed. The discrepancies were attributed primarily to the poor boundaries of the assembly, but also to incorrect treatment of the graphite felt in the calculations. It was not believed justified to recalculate the spectra in view of the difficult experimental geometry. Spectra for this experiment are shown in Figures 16 through 22 and indicate the extent of the disagreement.

Fig. 14--Experimental arrangement for cold-hot interface experiment in samarium poisoned graphite, case F

Fig. 15--Flux plot shapes for interface experiments, cases F and H.

Fig. 16--Neutron spectrum in cold-hot interface experiment, case F, at hole 1.

Fig. 17--Neutron spectrum in cold-hot interface experiment, case F, at hole 2.

Fig. 18--Neutron spectrum in cold-hot interface experiment, case F, at hole 3.

Fig. 19--Neutron spectrum in cold-hot interface experiment, case F, at hole 4,

Fig. 20--Neutron spectrum in cold-hot interface experiment, case F, at hole 5,

Fig. 21--Neutron spectrum in cold-hot interface experiment, case F, at hole 6.

Fig. 22--Neutron spectra across the cold-hot samarium poisoned graphite interface experiment, case F, calculated with the bound kernel,

Table 4

CONFIGURATION PARAMETERS FOR EXPERIMENT F

Region	Number densities Atoms/barn-cm	Temperature °K	Hole No.	d ₁ Inches	d ₂ Inches
I	C 0.079	274	1 2 3 4 5 6	3-1/2	--
II	C 0.0775	600		2-1/16	--
	Al 2.23 x 10 ⁻³			1	--
	Sm 2.08 x 10 ⁻⁶			---	1
				---	2-1/16
				---	3-1/2
III	t = 0 C 0.0039	300			

The next experiment (G) performed had an absorption and temperature discontinuity with an improved geometry. The cooling tanks of Figure 14 were replaced by 1/4 in. copper plates to which cooling coils had been brazed and then heavily cadmium plated. The graphite felt thickness was increased to 1/2 in. to make the temperature discontinuity more abrupt. In addition it was decided to use boron poisoning to achieve heavier absorption. The poisoning was obtained by inserting 0.005 in. 1% wt. boron loaded stainless steel foils between 0.230 in. layers of graphite. Neutron spectra were measured at six positions in this assembly. See Table 5.

Table 5

CONFIGURATION PARAMETERS FOR EXPERIMENT G

Region	Number densities Atoms/barn-cm	Temperature °K	Hole No.	d ₁ Inches	d ₂ Inches
I	C = 0.0765	274	1 2 3 4 5 6	3-1/2	--
II	B = 0.726 x 10 ⁻⁴	600		2-1/16	--
	C = 0.07587			1	--
	Ch = 3.063 x 10 ⁻⁴			--	1
	Fe = 1.0408 x 10 ⁻³			--	2-1/16
	Ni = 1.996 x 10 ⁻³			--	3-1/2
III	t = 0.500 in C = 0.0039	300			

When the theoretical spectra corresponding to these measurements were calculated and reported previously in Ref. 13 several difficulties were encountered in their analysis; these are discussed fully here. An incorrect boron concentration of $0.968 \pm 0.064\%$ wt in the stainless steel was used previously when a later analysis gave $0.9067 \pm 0.0375\%$ wt boron. In addition, the possibility that the boron was segregated in the stainless steel was raised, and this could not be determined due to the difficulty of making accurate chemical analyses of boron in that matrix. The axial flux plot with cadmium covered indium foils was not done well and some uncertainty was attached to its shape. Then it was discovered that the source density in the felt had not been reduced on the basis of its lower density. It was also decided that the leakage correction made in the felt was wrong. The leakage DB^2 was related to the geometrical buckling B^2 and to the diffusion coefficient D characteristic of the felt. However, the felt was so thin that the effective D was very much less. Hence, the spectra shown in Figures 23 through 29 are calculated with the leakage correction in the felt being made by a D characteristic of pure graphite. In addition, a somewhat smaller value of B^2 was used in these later calculations. In these figures, the agreement between theory and measurement is greatly improved over the results reported in Ref. 13 and indeed is excellent on the poisoned side. That the measured neutron spectra on the unpoisoned side of the assembly are slightly softer than calculated spectra tends to support the possibility that there was a source of thermal neutrons at the cold boundary.

In the final experiments, the basic heating and cooling system of the previous experiment G was retained. In these experiments, the hot side was poisoned by both samarium loaded and boron loaded aluminum foils to

Fig. 23--Neutron spectrum in cold-hot interface experiment, case G, at hole 1

Fig. 24--Neutron spectrum in cold-hot interface experiment, case G, at hole 2

Fig. 25--Neutron spectrum in cold-hot interface experiment, case G, at hole 3.

Fig. 26--Neutron spectrum in cold-hot interface experiment, case G, at hole 4

Fig. 27--Neutron spectrum in cold-hot interface experiment, case G, at hole 5

Fig. 28--Neutron spectrum in cold-hot interface experiment, case G, at hole 6

Fig. 29--Neutron spectrum across the cold-hot boron poisoned graphite interface experiment, case G, calculated with the bound kernel.

obtain the heaviest absorption of all. The boron loaded aluminum was used since its boron content was more certain and it was believed to exhibit no segregation. The samarium poisoning was re-employed since being a resonance absorber it shows the effects of crystal binding much more clearly than does a plain $1/v$ absorber. The graphite felt thickness was reduced to $1/4$ in. since the thicker felt of experiment (G) had shown no obvious improvement in the temperature discontinuity. Copper poisoning was added to the cold side by means of 0.010 in. sheets placed at each 0.525 in. layer of graphite. The purpose of the copper was to reduce the diffusion length of the cold side to about 10 cms and so lessen the dependence of the neutron flux on the cold side boundary conditions. In addition, the magnitude of the transverse leakage relative to absorption was greatly decreased thereby improving the approximation of the problem to a one dimensional calculation in the GAF13N code. Flux plots at room temperature across the assembly were taken in the axial and horizontal directions with cadmium covered indium foils. Furthermore, an axial flux plot across the interface was made with uranium foils, both bare and cadmium covered. Figure 30 illustrates the over-all geometry of this last test, and Figure 31 is a photo of the arrangement. Table 6 provides details of the corresponding configuration parameters relative to Figure 5. Neutron spectra were measured at six positions across the interface with the assembly at room temperature and then with the appropriate heating and cooling as shown in Table 6. Figure 32 shows a typical plot of the temperature across the interface in the cold-hot experiment K, indicating an excellent temperature discontinuity. The spatial variation of the activity of the cadmium covered indium is given in Figure 15 for comparison with the curve from experiment (F) and illustrates the sensitivity of the indium activity to the geometry of the coolant system.

Fig. 30--Experimental geometry for cases H and K

Fig. 31--Experimental arrangement for interface experiments H and K

Fig. 32--Temperature distribution in interface experiment, case K

Table 6

CONFIGURATION PARAMETERS OF EXPERIMENTS H AND K

Region	Number density Atoms/barn-cm	Case No.	Temperature °K	Hole No.	d ₁ Inches	d ₂ Inches
I	C = 0.07753	H	300	1	3-1/2	--
	Cu = 1.620x10 ⁻³	K	274	2	2	--
II	B = 1.030x10 ⁻⁴	H	300	3	1	--
	C = 0.07572	K	600	4	--	1
	Al = 2.421x10 ⁻³			5	--	2
	Sm = 2.242x10 ⁻⁶			5'	--	2-1/2
III	t = 0.25"	H	300			
	C = 0.0039	K	300			

Figure 33 shows the same curve of the cadmium covered activity compared with the calculated curve. In effect the distributed source fed into the top energy group in the GAPISN code has been adjusted until the measured indium activity agrees as well as possible with the calculated activity. Figure 33 indicates the degree to which this objective was realized. This same figure also shows the shape of the distributed source and indicates how adjustment near the boundaries is necessary to obtain agreement between the measured and calculated activities. The adjustment is not made on a good physical basis due to the presence of higher harmonics at the source edge and due to the possibility of a boundary source, but it does provide a consistent approach.

The calculated and measured spectra for these experiments are shown in Figures 34 through 49. On the lightly poisoned copper side, the agreement between measurement and calculation based on the bound kernel for graphite is excellent for both room temperature H and cold-hot K cases. The contrast with the calculation based on the free gas kernel for graphite is very distinct. On the poisoned side, the picture is much less clear. Again the

Fig. 33--Source distribution and calculated and measured distribution of cadmium-covered indium activity

Fig. 34--Neutron spectrum in room temperature interface experiment, case H, at hole 1.

Fig. 35--Neutron spectrum in room temperature interface experiment, case H, at hole 2

Fig. 36--Neutron spectrum in room temperature interface experiment, case H, at hole 3

Fig. 37--Neutron spectrum in room temperature interface experiment, case H, at hole 4

Fig. 38--Neutron spectrum in room temperature interface experiment, case H, at hole 5

Fig. 39--Neutron spectrum in room temperature interface experiment, case H, at hole 5

Fig. 40--Neutron spectrum across the room-temperature, boron-samarium poisoned graphite interface experiment, case H, calculated with the bound kernel.

Fig. 41--Neutron spectrum across the room temperature, boron-samarium poisoned graphite interface experiment, case H, calculated with the free gas kernel

Fig. 42--Neutron spectrum in cold-hot interface experiment, cases K, at hole 1

Fig. 43--Neutron spectrum in cold-hot interface experiment, case K, at hole 2

Fig. 44--Neutron spectrum in cold hot interface experiment, case K, at hole 3

Fig. 45--Neutron spectrum in cold-hot interface experiment, case K, at hole 4

Fig. 46--Neutron spectrum in cold-hot interface experiment, case K, at hole 5.

Fig. 47--Neutron spectrum in cold-hot interface experiment, case K, at hole 5¹

Fig. 48--Neutron spectrum across the cold-hot, boron-samarium poisoned graphite interface experiment, case K, calculated with the bound kernel.

Fig. 49--Neutron spectrum across the cold-hot, boron-samarium poisoned graphite interface experiment, case K, calculated with the free gas kernel

calculated spectra for the bound kernel are somewhat harder than the measured spectra. There is no obvious pattern in the discrepancy either spatially or with temperature and further examination of these data is in order. In the K case, it is certain however, that the calculated spectra based on the free gas kernel are a poorer fit on the hot side to the measured spectra.

The cause of these discrepancies is not known and we believe that it may lie either in experimental error, or possible uncertainties in the poison concentrations. It is possible that some of the differential energy transfer cross sections $\sigma_g (E' \rightarrow E)$ may be underestimated for the bound kernel, but more detailed experimentation and analysis are required to prove this. A plot of the thermal activity of the uranium foils across the interface, shown in Figure 50 is interesting in this aspect. The measured activities are higher on the poisoned side than the calculated values. This fact is consistent with the measured flux being more thermal on the poisoned side. Figure 51 compares the thermal activities of uranium across the interface based on spectra from both bound kernel and free gas kernel calculations. It is obvious from this figure that the free gas model overestimates the thermalization properties of the graphite. That model would also give poorer agreement with the measurements of Figure 50. This point is most relevant to the rocket core reflector problem. One can conclude from these experimental results that for simple configurations, the bound kernel for graphite is clearly superior to the free gas model. The measurements do not demonstrate this as clearly at an absorption and temperature discontinuity, but even there the evidence is still strong enough to merit the use of the crystalline model in design limited cases.

Fig. 50--Calculated and measured thermal uranium activities in interface experiment H

Fig. 51--Spatial distribution of uranium thermal activity calculated with both bound and free gas kernels.

V. THEORETICAL METHODS AND RESULTS

Further studies have been made of the basis from which the bound kernel for graphite was derived. The detailed theory is given below so that it may be seen how the new work performed under this contract appropriately extends the state of the art.

Introduction

One of the fundamental properties of an harmonic solid is the frequency spectrum of the lattice vibrations. The frequency spectrum is important not only for describing such macroscopic properties as the specific heat, but is important in the description of the interaction of thermal neutrons with the atoms of the solid. It has been shown that for a crystal having simple cubic symmetry, the incoherent scattering cross-section depends on the lattice dynamics only through the frequency spectrum. This same dependence has been shown to be an adequate description for more complicated crystals. Then if one is concerned about the thermalization of neutrons in a solid, the problem is to find the frequency spectrum of the lattice vibrations of that solid.

Graphite is extremely important as a moderator for thermal neutrons. A theory of the lattice vibrations of graphite has been given by Yoshimori and Kitano,⁽¹⁴⁾ (YK) which has been very successful in predicting the specific heat especially at low temperatures. Since one of the force constants was chosen to fit the specific heat data this agreement is not too surprising. The method (Houston's method) used by the above authors to obtain the frequency spectrum from their theory is generally known to be quantitatively incorrect except for predicting low temperature specific heats, and to lead to spurious singularities in the frequency spectrum.

In our new work we have used the theory of YK to calculate the frequency spectrum of graphite using the root sampling technique. The procedure is to sample the frequencies $\omega(\vec{k})$, where $\vec{k}$ is the phonon wave vector, for a large number of equally spaced points in an irreducible segment of the first Brillouin zone of a graphite

crystal. The frequency spectrum is found by constructing a histogram of the number of frequencies $\omega(\vec{k})$ falling into a frequency interval $\Delta\omega$.

The results of this calculation are compared with YK and with measurements of the frequency spectrum using the extrapolation technique in the theory of neutron scattering. In addition specific heat calculations are compared with experiment.

Since the paper of YK has some misprints, and the notation is not clear, a review of the theory is given here for clarification.

Review of the Theory of Yoshimori and Kitano

Graphite has the hexagonal structure and tends to form into weakly coupled hexagonal planer sheets. The unit cell of the graphite crystal contains four atoms. The basis vectors for the lattice are designated by $\vec{a}_1, \vec{a}_2, \vec{c}$, with $\vec{a}_1 \cdot \vec{c} = 0$ such that $|\vec{a}_1| = |\vec{a}_2| = \sqrt{3} a, |\vec{c}| = c, \vec{a}_1 \cdot \vec{a}_2 = -3a^2/2$ where a is the equilibrium separation of two atoms in the same plane. The position of any particle is given by the vector:

$$\vec{x}(k, l) = \vec{x}(k) + \vec{x}(l),$$

where for the lattice translational vector $x(l)$ we write

$$\vec{x}(l) = l_1 \vec{a}_1 + l_2 \vec{a}_2 + l_3 \vec{c},$$

and $k = A, B, C$, or D . For the cell vectors $\vec{x}(k)$ we have

$$\vec{x}(A) = (\vec{a}_2 - \vec{a}_1)/3,$$

$$\vec{x}(B) = 0,$$

$$\vec{x}(C) = (\vec{a}_3 - \vec{a}_1)/3 + \vec{c}/2$$

$$\vec{x}(D) = (\vec{a}_2 - \vec{a}_1)/3 + \vec{c}/2.$$

The vector $\vec{a}_3$ is defined by: $\vec{a}_3 = \vec{a}_1 + \vec{a}_2$. It is also useful to define unit vectors $\hat{i}_m, \hat{j}_m$ perpendicular and parallel respectively to the vectors $\vec{a}_m$,

and these are also shown in Fig. 52. The wave vector of a lattice wave is given in terms of the reciprocal lattice vectors $\vec{b}_1, \vec{b}_2, \vec{b}_3$ by:

Fig. 52--Two neighboring net planes of the graphite lattice showing the lattice vectors and the four particles A, B, C, and D of the unit cell

$$\vec{k} = k_1 \vec{b}_1 + k_2 \vec{b}_2 + k_3 \vec{c}$$

where $\vec{b}_n \cdot \vec{a}_m = \delta_{mn}$ ($m = 1, 2$), $\vec{d} \cdot \vec{a}_m = 0$, $\vec{d} \cdot \vec{c} = 1$. This identification gives the result $\vec{b}_1 \cdot \vec{b}_2 = 2/9a^2$, and

$$\vec{k} \cdot \vec{x}(\ell) = k_1 \ell_1 + k_2 \ell_2 + k_3 \ell_3.$$

In the harmonic approximation, the equation of motion of an atom in the graphite crystal is given by

$$Mu_\alpha(k, \ell) = - \sum_{\ell' k' \beta} \phi_{\alpha\beta} \left(\begin{smallmatrix} \ell \ell' \\ k k' \end{smallmatrix} \right) u_\beta(k', \ell'), \quad (1)$$

where M is the mass of a carbon atom, $u_\alpha(k, \ell)$ the α -th component of the displacement from equilibrium of the particle at $\vec{x}(k, \ell)$. The quantity $\phi_{\alpha\beta} \left(\begin{smallmatrix} \ell \ell' \\ k k' \end{smallmatrix} \right)$ is defined by

$$\phi_{\alpha\beta} \left(\begin{smallmatrix} \ell \ell' \\ k k' \end{smallmatrix} \right) = \left. \frac{\partial^2 \phi}{\partial u_\alpha(k, \ell) \partial u_\beta(k', \ell')} \right|_{\text{equil.}},$$

and ϕ is the potential energy of the lattice. Expanding in plane waves we arrive at the following determinantal equation,

$$\left| \sum_{\ell'} \phi_{\alpha\beta} \left(\begin{smallmatrix} \ell \ell' \\ k k' \end{smallmatrix} \right) e^{2\pi i \vec{k} \cdot (\vec{x}(k', \ell') - \vec{x}(k, \ell))} - \delta_{\alpha\beta} \delta_{kk'} M \omega^2 \right| = 0, \quad (2)$$

for the frequencies $\omega(\vec{k})$ as a function of $\vec{k}$. Since $\phi_{\alpha\beta} \left(\begin{smallmatrix} \ell \ell' \\ k k' \end{smallmatrix} \right)$ depends only upon $\ell - \ell'$, we put $\ell = 0$ for convenience.

The potential energy of the graphite lattice is assumed to consist of the sum of four different sources: (a) changes in the bond angles proportional to a force constant μ , (b) changes in the bond length between nearest neighbors in the same plane, proportional to a force constant κ , (c) a central force due to changes in the bond length between nearest neighbors in two adjacent planes, proportional to a force constant κ' , and (d) a force due to the bending of the planes and proportional to the displacement in the $\vec{c}$ -direction of any atom relative to its nearest neighbors in

the same plane. The force constant for this last type of restoring force is denoted by μ' . The change in any angle θ is given by

$$a\Delta\theta = \vec{u}(A;o,-1,o) \cdot \hat{j}_1 - \vec{u}(A;o) \cdot \hat{j}_3 - \sqrt{3} \vec{u}(B;o) \cdot \hat{i}_2,$$

and the corresponding change in potential energy by $\mu(a\Delta\theta)^2/2$. The change in the bond length ℓ , is:

$$\Delta\ell_1 = (\vec{u}(A;o) - \vec{u}(B;o)) \cdot \hat{i}_3,$$

and the potential energy change associated with this is $\kappa(\Delta\ell_1)^2/2$. A representative contribution due to the bending of the planes is $\mu'(\Delta h_1)^2/2$, where

$$\Delta h_1 = \hat{k} \cdot \left[\vec{u}(A;o) - \frac{1}{3} (\vec{u}(B;o) + \vec{u}(B;o,1,o) + \vec{u}(B;-1,o,o)) \right].$$

The interaction between planes contribute terms like $\kappa'(\Delta c)^2/2$ where for Δc we write

$$\Delta c = \hat{k} \cdot (\vec{u}(D;o) - \vec{u}(A;o)).$$

Here the unit vector $\hat{k}$ is parallel to $\vec{c}$. The total potential energy of the lattice is the sum of all terms like those above, but since $\phi_{\alpha\beta} \left(\frac{\ell\ell'}{kk'} \right)$ depends only on $\ell - \ell'$ we need consider only those contributions involving atoms of the zero cell ($\ell = 0$). The matrix elements can be readily calculated once the potential has been evaluated. The results are:

$$\phi \left(\begin{smallmatrix} o;o \\ A;A \end{smallmatrix} \right) = \left(\frac{15}{2} \mu + \frac{3}{2} \kappa \right) (\hat{i}_1 \hat{i}_1 + \hat{j}_1 \hat{j}_1) + (2\kappa' + 4\mu'/3) \hat{k} \hat{k},$$

$$\phi \left(\begin{smallmatrix} o;1,o,o \\ A;A \end{smallmatrix} \right) = -\mu \hat{j}_3 \hat{j}_2 + \mu' \hat{k} \hat{k}/9$$

$$\phi \left(\begin{smallmatrix} o;o,-1,o \\ A;A \end{smallmatrix} \right) = -\mu \hat{j}_3 \hat{j}_1 + \mu' \hat{k} \hat{k}/9$$

$$\phi \left(\begin{smallmatrix} o;1,1,o \\ A;A \end{smallmatrix} \right) = -\mu \hat{j}_1 \hat{j}_2 + \mu' \hat{k} \hat{k}/9$$

$$\phi \left(\begin{smallmatrix} o;-1,-1,o \\ A;A \end{smallmatrix} \right) = -\mu \hat{j}_2 \hat{j}_1 + \mu' \hat{k} \hat{k}/9$$

$$\phi \left(\begin{smallmatrix} o;o,1,o \\ A;A \end{smallmatrix} \right) = -\mu \hat{j}_1 \hat{j}_3 + \mu' \hat{k} \hat{k}/9$$

$$\phi \left(\begin{smallmatrix} o;-1,o,o \\ A;A \end{smallmatrix} \right) = -\mu \hat{j}_2 \hat{j}_3 + \mu' \hat{k} \hat{k}/9$$

$$\phi \left(\begin{smallmatrix} o;o \\ A;o \end{smallmatrix} \right) = -\kappa' \hat{k} \hat{k}$$

$$\phi \left(\begin{smallmatrix} o;o,o,-1 \\ A;D \end{smallmatrix} \right) = -\kappa' \hat{k} \hat{k}.$$

$$\phi_{A;B}^{(0;0)} = -\kappa \hat{i}_3 \hat{i}_3 - 6\mu \hat{j}_3 \hat{j}_3 - 2\mu' \hat{k} \hat{k} / 3$$

$$\phi_{A;B}^{(0;0,1,0)} = -\kappa \hat{i}_1 \hat{i}_1 - 6\mu \hat{j}_1 \hat{j}_1 - 2\mu' \hat{k} \hat{k} / 3$$

$$\phi_{A;B}^{(0;-1,0,0)} = -\kappa \hat{i}_2 \hat{i}_2 - 6\mu \hat{j}_2 \hat{j}_2 - 2\mu' \hat{k} \hat{k} / 3$$

These last three matrix elements and the factor in κ' in the first matrix element differ from those of YK by a factor two, but appear correctly later in their paper. The remaining matrix elements are formed in an identical fashion since the plane containing A,B is dynamically equivalent to that containing C,D. Since there are no terms involving products like $\hat{i}_m \hat{k}$ or $\hat{j}_m \hat{k}$, it is possible to reduce the 12×12 determinant into three 4×4 determinants. Two of these smaller determinants will involve only vibrations in the planes and will be identical, and one will involve vibrations between the planes.

Thus, for vibrations in the plane we have

$$\left| \sum_{\ell'} \phi_{mn}^{(0,\ell')} \left(\frac{k,k'}{\ell'} \right) e^{2\pi i \vec{k} \cdot (\vec{x}(k';\ell) - \vec{x}(k;0))} \cdot M\omega^2 \delta_{mn} \delta_{kk'} \right| = 0 \quad (3)$$

where $m,n = 1$ or 2 , and k,k' are A or B, but could equivalently be the pair C,D. The vectors $\hat{i}_1, \hat{j}_1$ are taken as the x and y coordinates respectively. Since the matrix in Eq. (3) is Hermitian, we find

$$\begin{vmatrix} X_{11}(AA) - M\omega^2 & X_{12}(AA) & X_{11}(AB) & X_{12}(AB) \\ X_{12}^*(AA) & X_{22}(AA) - M\omega^2 & X_{12}(AB) & X_{22}(AB) \\ X_{11}^*(AB) & X_{12}^*(AB) & X_{11}(AA) - M\omega^2 & X_{12}^*(AA) \\ X_{12}^*(AB) & X_{22}^*(AB) & X_{12}(AA) & X_{22}(AA) - M\omega^2 \end{vmatrix} = 0 \quad (4)$$

where

$$x_{11}(AA) = \frac{15}{2} \mu + \frac{3}{2} \kappa + \frac{3}{2} \mu \cos 2\pi k_1$$

$$x_{22}(AA) = \frac{15}{2} \mu + \frac{3}{2} \kappa - \frac{\mu}{2} \cos 2\pi k_1 + \mu \cos 2\pi k_2 + \mu \cos 2\pi(k_1+k_2)$$

$$x_{12}(AA) = (\sqrt{3} \mu/2) \left[1 \sin 2\pi k_1 - e^{-2\pi i k_2} + e^{-2\pi i(k_1+k_2)} \right]$$

$$= x_{21}^*(AA)$$

$$x_{11}(AB) = -\frac{(\kappa+18\mu)}{4} \left[e^{2\pi i(k_1-k_2)/3} + e^{-2\pi i(2k_1+k_2)/3} \right] - \kappa e^{2\pi i(2k_2+k_1)/3}$$

$$x_{12}(AB) = -(\sqrt{3}/4) (\kappa-6\mu) \left[e^{2\pi i(k_1-k_2)/3} - e^{-2\pi i(2k_1+k_2)/3} \right]$$

$$= x_{21}(AB)$$

$$x_{22}(AB) = - (3/4) (\kappa+2\mu) \left[e^{2\pi i(k_1-k_2)/3} + e^{-2\pi i(2k_1+k_2)/3} \right] - 6\mu e^{2\pi i(2k_2+k_1)/3}$$

For the vibrations between the planes, the determining equation is:

$$\left| \sum_{\ell'} \phi_{33} \begin{pmatrix} 0 & \ell' \\ k & k' \end{pmatrix} e^{2\pi i \vec{k} \cdot (\vec{x}(k', \ell') - \vec{x}(k; 0))} - M \omega^2 \delta_{kk'} \right| = 0, \quad (5)$$

for k, k' equal to A, B, C, or D, or in detail the equation becomes:

$$\begin{vmatrix} z_{11} - M\omega^2 & z_{12} & 0 & z_{14} \\ z_{12}^* & z_{22} - M\omega^2 & 0 & 0 \\ 0 & 0 & z_{22} - M\omega^2 & z_{12} \\ z_{14} & 0 & z_{12}^* & z_{11} - M\omega^2 \end{vmatrix} = 0 \quad (6)$$

with the elements given by

$$Z_{11} = 2\kappa' + 4\mu'/3 + (2\mu'/9) (\cos 2\pi k_1 + \cos 2\pi k_2 + \cos 2\pi(k_1+k_2))$$

$$Z_{22} = Z_{11} - 2\kappa' = Z_{33}$$

$$Z_{12} = Z_{21}^* = -(2\mu'/3) \begin{bmatrix} e^{2\pi i(k_1-k_2)/3} & e^{+2\pi i(2k_2+k_1)/3} & e^{-2\pi i(2k_1+k_2)/3} \end{bmatrix}$$

$$Z_{14} = Z_{41} = -2\kappa' \cos \pi k_3$$

If we call

$$\lambda = M\omega^2 - 4\mu'/3 - (2\mu'/9) (\cos 2\pi k_1 + \cos 2\pi k_2 + \cos 2\pi(k_1+k_2)),$$

the four solutions to Eq. (6) are:

$$\begin{aligned} \lambda_{1,2} &= -\kappa' + Z_{14}/2 \pm \sqrt{(\kappa' + Z_{14}/2)^2 + |Z_{12}|^2} \\ \lambda_{3,4} &= \kappa' - Z_{14}/2 \pm \sqrt{(\kappa' - Z_{14}/2)^2 + |Z_{12}|^2} \end{aligned} \quad (7)$$

It is now possible, given the values of the force constants, to compute the dispersion relations $\omega(\vec{k})$ vs $\vec{k}$ for all twelve branches, - 6 optical and 6 acoustical. From measurements of specific heat and compressibility, the force constants κ' , μ' were determined by YK to be: $\kappa = 5.57 \times 10^3$, and $\mu' = 2.92 \times 10^5$ in units dyne/cm. The value of κ' is in excellent agreement with a recent measurement by Brockhouse.⁽¹⁵⁾ The values of the lattice constants are: $a = 1.42 \text{ \AA}$ and $c = 6.70 \text{ \AA}$.

The force constants κ and μ were taken by YK from the benzene molecule and are $\kappa = 6.71 \times 10^5$, $\mu = 0.48 \times 10^5$ in units dyne/cm.

Frequency Distribution of Graphite

All the possible values of the wave vector $\vec{k}$ are obtained by restricting $\vec{k}$ to lie in the first Brillouin zone of the crystal. For graphite this zone is a hexagonal cylinder bound by the planes normal to the vectors:

$$\pm \vec{b}_1/2, \pm \vec{b}_2/2, \pm (\vec{b}_1 - \vec{b}_2)/2, \pm \vec{b}_3/2.$$

The plane $\vec{b}_3 = \text{const.}$ of the first Brillouin zone is shown in Fig. 53. It is clear that we can restrict our solutions for $\omega(\vec{k})$ to the shaded portion of Fig. 53 which is $1/24$ of the zone, ($k_3 \geq 0$).

The frequency spectrum $f(\omega)$ is defined such that $f(\omega)d\omega$ gives the number of frequencies between ω and $\omega + d\omega$. The best means of finding $f(\omega)$ seems to be to solve Eqs. (4) and (7) on a large computer for a great number of values of the wave vector $\vec{k}$ in the irreducible segment ($1/24$) of the first Brillouin zone, carefully weighting the points as to whether or not they fall on the edges faces, etc. of the irreducible segment, and then to form a histogram for $f(\omega)$.

Due to the separation of the vibrational modes in and out of the basal planes (Fig. 53) one can construct two separate vibrational spectra corresponding to these modes: $f_{\parallel}(\omega)$ and $f_{\perp}(\omega)$. The total frequency spectrum is then given by

$$f(\omega) = \frac{2}{3} f_{\parallel}(\omega) + \frac{1}{3} f_{\perp}(\omega).$$

The spectra are normalized such that

$$\int_0^{\omega_{\perp \text{ max}}} f_{\perp}(\omega) d\omega = \int_0^{\omega_{\parallel \text{ max}}} f_{\parallel}(\omega) d\omega = 1.$$

Fig. 53--The plane $\vec{b}_3 = 0$ of the first Brillouin zone of graphite

An IBM 7044 computer program CAFS has been written which solves Eqs. (4) and (7) for equally spaced points in the irreducible segment of the Brillouin Zone. This program also constructs the histogram of the frequency spectrum, and calculates the specific heat at constant volume C_v by means of the relation

$$C_v/3R = \int_0^{\omega_{\max}} \left(\frac{\omega}{T}\right)^2 \frac{e^{\omega/T}}{(e^{\omega/T} - 1)^2} f(\omega) d\omega \quad (7)$$

where R is the gas constant, and T the temperature in electron-volts.

The only measurements on graphite which are useful in determining the force constants are the compressibility and the specific heat. The constant κ' was determined by YK from the observed compressibility, and μ' from the observed low temperature specific heat. Instead of using the same values of κ and μ as YK, these force constants were obtained by adjustment to the high temperature (300° - 1000° K) specific heat. CAFS calculations for 2413 equally spaced points in the irreducible segment of the Brillouin Zone show that in terms of the YK values of the force constants, the values $\kappa = .65$, $\mu = .75$, $\kappa' = 1.0$, $\mu' = 1.0$ give excellent agreement with the specific heat over the whole range. This agreement is presented in Fig. 54, and in Table 7.

It should be mentioned that the set of force constants cannot be made unique from specific heat comparisons alone, however only small changes from the set given above will still give good agreement with the specific heat. This is particularly true of the constant κ which, because of its size, determines the maximum frequency of the lattice.

With the set of force constants given above a CAFS calculation was

Fig. 54-- Specific heat of reactor type graphite.

Table 7

OBSERVED AND CALCULATED VALUES OF THE SPECIFIC HEAT OF
 REACTOR TYPE GRAPHITE ($C_V/3R$)

<u>Temp. °K</u>	<u>Observed Value(3)</u>	<u>Root Sampling Fig. ()</u>	<u>YK (Mod)</u>
100	.068613	.07284	.0786
125		.1029	
150	.13052	.1350	
175		.1684	
200	.201	.2026	.2103
225		.2374	
250		.2725	
275		.3075	
300	.3439	.3422	.3471
400		.4719	.4701
500	.582	.5800	.5725
600		.6645	.6541
700		.7293	.7198
800	.7817	.7787	.7673
900		.8167	.8060
1000	.8455	.8462	.8364

performed for a very large number of equally spaced points in the Brillouin zone (47,788). The resulting vibrational spectrum is shown in Fig. 55. Also shown in Fig. 55 is the spectrum obtained by YK using Houston's method for the vibrations in the plane which is known to lead to spurious singularities, while for vibrations between the basal planes YK replaced the Brillouin zone by a circular cylinder, and used approximate formulas for Eq. (7). Table 7 lists also the specific heats calculated by YK.

The measurements listed in Table 7 were made for Acheson graphite, CS grade (reactor type). Another major type of graphite is Ceylon natural graphite which has a lattice constant c which is roughly twice as large as Acheson graphite. Measurements for Ceylon graphite are given in Ref. (5). The measurements for this type of graphite can be better fit with the force constants given by YK. Table 8 lists the specific heats of Ceylon graphite compared with both the YK values and the values calculated by the root sampling technique presented here. Since the force constant μ' was chosen by YK to fit the low temperature specific heat, Table 8 gives an indication of the size of the errors incurred for integral quantities from using the approximation methods of YK. In Table 8 also are the specific heats obtained by choosing the force constants to give a better fit. Since the specific heat for Ceylon graphite has not been measured over a wide enough range it is far from possible to choose a unique set of force constants from specific heat data alone.

It is interesting to note that although the major difference between these two types of graphite is the lattice constant in the $\vec{c}$ -direction, the specific heats indicate that the force constants κ , μ are very different, rather than the constants κ' , μ' . This conclusion could be changed with more specific heat measurements on Ceylon graphite.

Fig. 55 -- Frequency Spectra of Graphite

TABLE 8

Comparison of Observed and Calculated Specific Heats of Ceylon Natural Graphite ($C_v/3R$)

Temp. °K	Observed Value (4)	YK (constant μ' chosen, to fit C_v)	Root Sampling (YK force constants)	Root Sampling $\kappa = .8, \mu = 1$ $\mu' = 1, \kappa' = 1$	Root Sampling $\kappa = 1, \mu = 1$ $\mu' = 9, \kappa' = 1$
100	.067254	.06739	.06635	.06640	.06957
125	.09334	.09409	.09270	.09339	.09678
150	.12233	.1226	.1205	.1220	.1252
175	.15291	.15266	.1492	.1519	.1547
200	.1845	.1835	.1788	.1827	.1848
225	.2162	.2151	.2088	.2142	.2153
250	.2488	.2466	.2389	.2461	.2460
275	.28166	.2796	.2691	.2781	.2764
300	.3159	.3156	.2990	.3099	.3065

Application to Slow Neutron Scattering

With the neglect of interference effects, the double differential cross-section $d^2\sigma/dE d\Omega$ can be calculated from the expression

$$\frac{d^2\sigma}{dE d\Omega} = \frac{\sigma_b}{4\pi} \frac{k}{k_0} \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i\epsilon t} X(\kappa^2, t) \quad (7)$$

where

$$X(\kappa^2, t) = \exp \left[\frac{\kappa^2}{2M} \int_{-\infty}^{\infty} d\omega \frac{f(\omega) e^{-\omega/2T}}{2\omega \sinh(\frac{\omega}{2T})} (e^{-i\omega t} - 1) \right] \quad (8)$$

where $\kappa = \vec{k} - \vec{k}_0$ is the neutron momentum transfer, and M the atomic mass. Eqs. (7) and (8) can be evaluated by means of the IBM-7044 code SUMMIT, and with these cross-sections one can calculate thermal neutron spectra under a variety of conditions. It has been shown⁽⁵⁾ that the vibrational spectrum of YK with an adjusted $\omega_{\max}$ is very successful in the prediction of measurements relevant to reactor physics.

In order to test the frequency spectrum calculated in this report the following comparison problem was done. A semi-infinite medium neutron spectrum, normalized to the source terms derived using a $1/E$ spectrum, was calculated containing resonance absorbers. A free gas was fitted to the calculations above 1.0 ev. In units of atoms per barn-cm the number densities of samarium, indium, Pu^{239} , Pu^{240} and graphite were respectively 5×10^{-6} , 8×10^{-6} , 1.6×10^{-4} , 2.0×10^{-6} and 8×10^{-2} . The buckling is 10^{-4} cm^{-2} . Fig.56 gives the comparison between the different frequency spectra. One can conclude from Fig.56 that for cases of interest to reactor physics the frequency spectrum obtained by the root sampling method will give very good agreement with experiment.

FIG. 56 COMPARISON OF VARIOUS FREQUENCY SPECTRA IN PREDICTING THE NEUTRON SPECTRUM IN A GRAPHITE MODERATOR WITH RESONANCE ABSORBERS.

In reference 5 the statement is made that reactor physics calculations are sensitive only to the maximum frequency $\omega_{\max}$, and not on the structure of $f(\omega)$, but Fig. 56 shows the statement to be untrue, since both YK (modified) and the more exact spectrum will give almost identical results for reactor problems, and they differ considerably in both cut-off and structure.

VI. CONCLUSIONS

The agreement between the spectra measured experimentally and the analyses based on kernels generated with the SUMMIT code and using the Yoshimori and Kitano frequency spectrum is generally quite satisfactory. However, there still exists a large enough discrepancy to warrant continuation of the work on this problem.

Examination of the results of the last two experiments indicates that the calculated spectra are too hard on the poisoned side of the interface. This is believed to be due to the heterogeneous nature of the assembly. Because of the target location and the increase in the absorption cross section as the interface is crossed, there is a net current flow from left to right across the interface in all energy groups. This current constitutes an appreciable fraction of the total source of thermal neutrons particularly for energies below 0.1 ev. In the calculations, the heterogeneous structure of graphite and poison is homogenized when generating macroscopic cross sections. Hence a neutron, on crossing the interface immediately "sees" an appreciable absorption cross section. Experimentally however, many of the neutrons that cross the interface must scatter several times before they pass through the poison sheets. Since the scattering mean free path is of the order of one inch in graphite this implies that it might take several inches before equilibrium is reached and homogenized cross sections are valid. It is therefore recommended that this program be continued as follows:

Experimental Work

A modified assembly should be fabricated that will enhance the neutron population near the interface in the region between 0.01 and 0.1 ev. This can be done by using the copper poisoning only in the first four inches

of the assembly, where the largest boundary perturbations exist. Six inches of unpoisoned graphite will then be used to the left of the interface to increase the thermal neutron intensity at the interface; this represents an overall reduction of two inches of graphite. On the right side of the interface it is recommended that a small amount of specially prepared graphite having resonance poisons homogeneously mixed in be used. This should be advantageous not only with respect to removing the heterogeneity but also, if careful quality control is exercised, the ratio of poison atoms to carbon atoms should be precisely known.

Analytical Work

So far, none of the space dependent problems have been analyzed using the J. A. Young frequency spectrum. Although no major changes are expected, this should not be left open.

Work is now in progress, under another contract, on a new code which will generate the scattering kernels by different numerical techniques than presently those used in SUMMIT. The new code has the advantage of being a more straightforward approach to the problem. The double differential cross section will be integrated directly instead of by the present method in which several different approximate expansions are used. In addition, it is hoped that the integrations can be performed in such a way as to remove the sensitivity of the results to the selection of the energy mesh. This will undoubtedly be of advantage to other groups who do not have the extensive experience with kernel calculations necessary to make the optimum choice of parameters and mesh for a SUMMIT calculation. It is expected that this code will be in operation in the near future. It should then be possible to pin down the problem of predicting thermal neutron spectra in graphite to within five percent, which is adequate for most reactor applications.

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