

# REACTOR PHYSICS EXPERIMENTS WITH GRAPHITE MODERATED SUBCRITICAL ASSEMBLY

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## I. INTRODUCTION

A graphite moderated subcritical assembly, designed by Fuji Electric Mfg. Co., Ltd., was constructed at the Nuclear Laboratory of the same company which is in Miura Peninsula in Kanagawa Prefecture. The subcritical assembly is built with the Government Subsidy for the Peaceful Uses of Atomic Energy for the Fiscal Year of 1960.

A series of experiments, among which the exponential experiment was the major one, had been completed by the end of March, 1962.

The subcritical assembly, which was the first one of this type built in Japan, used natural uranium as the fuel and graphite as the moderator in order to study nuclear characteristics of the Tōkai Nuclear Power Reactor and Calder Hall Reactor. The primary purpose of making the subcritical assembly, i.e., to obtain more reliable reactor physics data as well as to improve the techniques for reactor physics experiments and to train the reactor personnel, has successfully been fulfilled. It should, however, also be mentioned that through this experiment it was confirmed that Japanese-made reactor core materials are appropriate for practical reactor use from the viewpoint of reactor physics.

It had generally been believed that graphite moderated subcritical assemblies have difficulties in changing the lattice spacings in comparison with water moderated ones. Special considerations were given in this respect in designing the assembly so that the lattice spacings can easily be changed for wide range. The diffusion and slowing down length of graphite, cadmium ratios in the assembly, material bucklings and neutron flux fine structure in unit cell were measured and the results were satisfactory.

Fuji Electric Mfg. Co., is now constructing another subcritical assembly for High Temperature Gas Cooled Reactor with the same kind of government subsidy for fiscal year of 1961. It is expected that many interesting results will be obtained by the beginning of 1963.

This report describes the outline of the experimental facilities constructed with the subsidy of 1960 and the results of the experiments.

## II. EXPERIMENTAL FACILITIES

The experimental facilities consisted of

- (1) Neutron source and its remote handling device.
- (2) Graphite assembly (graphite pedestal and core assembly).
- (3) Fuel elements.
- (4) Cadmium sheets.
- (5) Nuclear radiation measuring equipments.
- (6) Remote handling device for  $\text{BF}_3$  counter.

(Fig. 1)

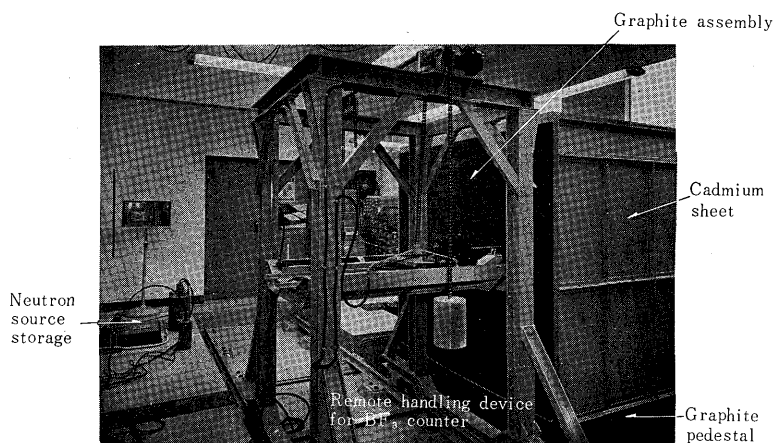


Fig. 1 General view of assembly

### 1. Neutron Source

The neutron source was placed in the center of the graphite pedestal which was laid under the core assembly. The characteristics of the neutron source used were as follows:

Material :	Po-Be, 5 curie, 1 piece.
Dimension :	Approx. 15 mm dia. $\times$ 20 mm
Neutron yield :	Approx. $1 \times 10^7$ n/sec.
Half life :	130 days.

The steps which the remote handling mechanism of the neutron source followed are: i) remove the cover of the neutron source storage container, ii)

hook up the neutron source and transfer it to the graphite pedestal, iii) place it in the proper position in the graphite stringer which has previously been pulled out of the pedestal, iv) put the stringer back into the pedestal. The mechanism was operated from the room adjacent to the assembly room.

## 2. Graphite Assembly

The core assembly was a cube of 1,800 mm on a side and was consisted mainly of horizontal layers of graphite blocks. The graphite pedestal, placed under the core assembly, had the dimension of  $1956 \times 1956 \times 535$  (h) mm. Each graphite block had a right circular cylindrical hole in the center in axial direction as shown in Fig. 2 so that a graphite sleeve or a

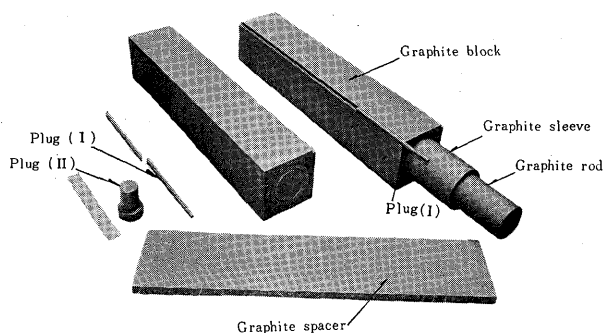


Fig. 2 Graphite block

graphite filler pieces could easily be inserted in or taken out of the hole. In order to carry out the exponential experiment, the graphite filler pieces were taken out of the hole and fuel elements were inserted instead. The channel diameters were changed by placing graphite sleeves of suitable thickness in the hole. One corner of each graphite block was cut off along the longitudinal direction as shown in Fig. 2 so that a miniature  $\text{BF}_3$  counter or foils for neutron flux distribution measurement could be inserted. It was possible, furthermore, to obtain the various lattices shown in Table 1 by inserting graphite spacers shown in the lower part of Fig. 2 in between every graphite blocks. The schematic diagram of each lattice arrangement and spacings used are shown in Fig. 3.

The total amount of graphite used for assembly was 22.026 kg and had been manufactured by Tōkai Electrode Mfg. Co., Ltd.

Its characteristics were:

Bulk density:  $1.650 \sim 1.700 \text{ g/cm}^3$   
 Boron content: 0.10 ppm  
 Ash content: 0.01 ppm  
 Specific electric resistance:  $9 \times 10^{-4} \Omega\text{-cm}$

## 3. Fuel Element

The fuel element had the dimension of  $34 \text{ mm (dia.)} \times 450 \text{ mm}$  and was clad with 3 mm thick aluminum. Four of these fuel elements were laid axially in a channel. They were placed in the hole of the graphite in such a way that the fuel and the hole

would form coaxial cylinders by suspending the both ends of the fuel element with aluminum holder. The fuel element had been fabricated by Furukawa Electric Co., Ltd. The specifications were:

Material: Natural uranium

Density:  $18.9 \text{ g/cm}^3$

Chemical composition: According to the information given by Genshi Nenryō Kōsha who had supplied the natural uranium ingot, the impurities contained were:

Al < 50 ppm      H < 5 ppm

B < 0.2          Mn < 10

C < 600          N < 50

Cd < 0.2          Ni < 80

Cr < 30          Si < 60

Fe < 150          V < 10

Total weight: 1,810 kg

Dimension:  $28 \text{ mm (dia.)} \times 440 \text{ mm}$

Clad thickness: 3 mm (Al)

## 4. Cadmium Sheets

The top and sides of the assembly were covered with 1.5 mm thick cadmium sheets clad with 3 mm thick aluminum sheets to absorb all thermal neutrons leaking out of and reflected back into the assembly.

## 5. Nuclear Radiation Measuring Instruments

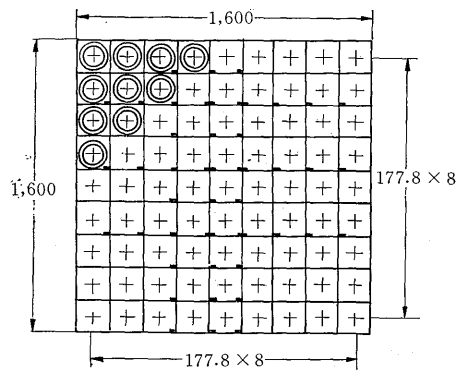
The nuclear radiation measuring instruments used in this experiment, which were made by Kōbe Kōgyō Corp., were as follows:

- (1) Thermal neutron detection instrument.
- (2)  $\beta$  ray counter.
- (3) Fast neutron monitor.
- (4) Thermal neutron monitor.
- (5)  $\gamma$  ray monitor.

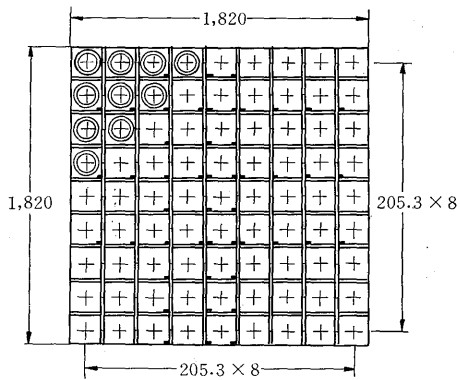
(1) Thermal neutron detection instrument: The purpose of this instrument was to measure the neutron flux in the assembly. It consisted of a  $\text{BF}_3$  counter, a pre-amplifier, an amplifier, a scaler and a timer. The size of the  $\text{BF}_3$  counter had to be small in order to be inserted in the assembly. The diameter of the  $\text{BF}_3$  counter used was 8 mm. The operating range of neutron flux was  $10^2 \sim 10^5 \text{ n/cm}^2 \text{ sec}$ . The theoretical sensitivity was  $0.24 \text{ cpm/n. cm}^{-2} \text{ sec}^{-1}$ , and the  $\gamma$  ray background was negligible for the  $\gamma$  ray level of less than 0.1 r/hr.

(2)  $\beta$  ray counter: The neutron flux distribution in the assembly was also measured by counting the induced  $\beta$  activity of metal foils which had been activated in the assembly. The  $\beta$  counter used for this purpose was an ordinary Geiger-Müller counter with dead time of approximately  $400 \mu \text{ sec}$ .

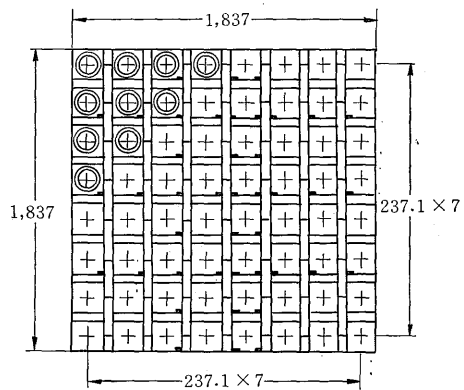
(3) Fast neutron monitor, thermal neutron monitor and  $\gamma$  ray monitor:  $\text{BF}_3$  counters were used for neutron monitors. The sensitive ranges for fast and thermal neutrons were below  $10^3 \text{ n/cm}^2 \text{ sec}$ . and  $10^2 \sim 10^5 \text{ n/cm}^2 \text{ sec}$ ., respec-



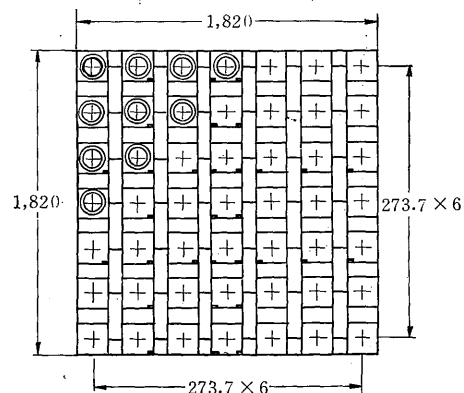
(a)  
Square lattice with 177.8 mm spacing



(b)  
Square lattice with 205.3 mm spacing

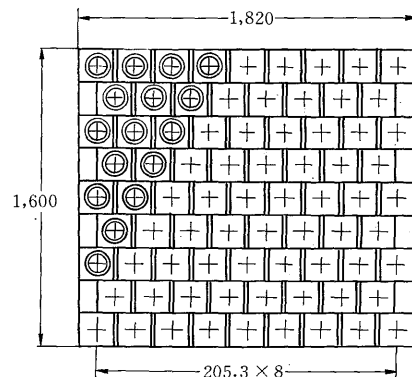


(c)  
Square lattice with 237.1 mm spacing

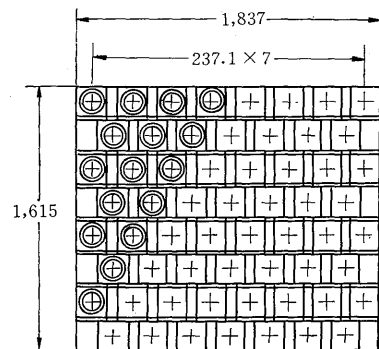


(d)  
Square lattice with 273.7 mm spacing

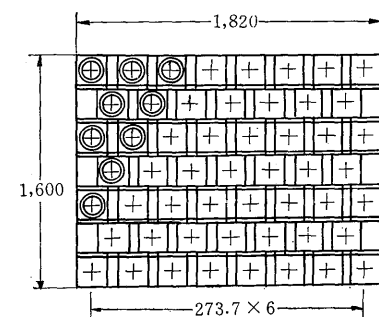
Fig. 3 Various lattices used



(b')  
Hexagonal lattice with 205.3 mm spacing



(c')  
Hexagonal lattice with 237.1 mm spacing



(d')  
Hexagonal lattice with 273.7 mm spacing

tively. Alarm was expected to be given out when any one of the three monitors received abnormally high radiation.

Besides those mentioned above, there were a scintillation counter for  $\alpha$  and  $\beta$  ray measurements, and a hand-foot monitor.

## 6. Remote Handling Device for $\text{BF}_3$ Counter

This device was designed so that a  $\text{BF}_3$  counter could be inserted in the assembly even when the radiation levels around the assembly were high. The device was remotely operated from the room adjacent to the assembly room. Although the device was prepared in consideration for the possible high levels of radiation, it has never been used as the actual radiation level in the room has been so low.

## III. EXPERIMENTS WITH GRAPHITE ASSEMBLY

Graphite assembly means, here, the core graphite assembly of which the fuel rods are replaced by graphite filler pieces. The purpose of the experiment with this assembly was to investigate nuclear characteristics of graphite; i. e., to measure diffusion length and slowing down length of graphite.

### 1. Diffusion Length of Graphite

Diffusion length is a quantity which is proportional to the average length travelled by thermal neutrons from the points of their birth to the points of being captured. It can be obtained from the measurement of neutron flux distribution in the assembly. In general, the neutron flux distribution in a rectangular parallelepiped assembly with horizontal cross section of  $a \times b$  is given by the following expression when there is a neutron source at the bottom:

$$\phi(x, y, z) = \sum_{l, m} K_{l, m} \sin \frac{l\pi x}{a} \cdot \sin \frac{m\pi y}{b} \cdot \sinh \frac{z}{L_{lm}} \quad \dots\dots(1)$$

Diffusion length,  $L$ , is given by

$$\frac{1}{L^2} = \frac{1}{L_{lm}^2} - \pi^2 \left( \frac{l^2}{a^2} + \frac{m^2}{b^2} \right) \quad \dots\dots(2)$$

The high mode can be neglected at large distances from the neutron source. Therefore,  $L$  can be calculated from the fundamental term  $L_{11}$  which is obtained from neutron flux distribution measurements in the region sufficiently far from the neutron source.  $L_{11}$  was obtained here by fitting the measured neutron flux distribution to the following equation by least square method:

$$\phi = \frac{A \sinh \frac{(c-z)}{L_{11}}}{\sinh \frac{c}{L_{11}}} \quad \dots\dots(3)$$

where  $c$  is the height of the assembly, and  $A$  is a constant. The digital computer used for this calculation was FACOM128, a relay type computer, and the convergence of  $L_{11}$  took approximately 15 minutes

for each case. As the density of the graphite used for the assembly varied from  $1.678 \text{ g/cm}^3$  to  $1.692 \text{ g/cm}^3$ , average value of  $\rho = 1.68 \text{ g/cm}^3$  was used. The diffusion length,  $L$ , obtained by this method was  $51.7 \text{ cm}$ , while ANL-5800 gives  $L = 54.4 \text{ cm}$  for graphite of density  $1.6 \text{ g/cm}^3$ . The discrepancy between these values can be eliminated by applying suitable density correction.

### 2. Slowing Down Length in Graphite

Slowing down length is a quantity which is proportional to the average distance travelled by neutrons from the points of their birth to the points where they become thermal. According to Fermi-age theory, which is a good approximation for graphite, the slowing down density,  $q$ , at the distance  $r$  from a neutron source emitting  $s$  neutrons per second is given by

$$q = \frac{s \cdot e^{-\frac{4L_s^2}{r^2}}}{(4\pi L_s^2)^{3/2}} \quad \dots\dots(4)$$

Where  $L_s$  is the slowing down length. The plot of the logarithm of neutron slowing down density distribution against the square of the distance from the source gives  $L_s$  from the slope of the resultant straight line,  $-\frac{1}{4L_s^2}$ . Following metal foils were inserted in the assembly by the method mentioned above.

	Au-197	In-115
Natural abundance:	100%	96%
Half life:	2.7 day	54 min.
Shape:	square	square
Dimension:	$8 \times 8 \times 0.08 \text{ mm}$	$8 \times 8 \times 0.12 \text{ mm}$
Mass:	100mg	50 mg

Fig. 4 is the plot of the saturation activity of

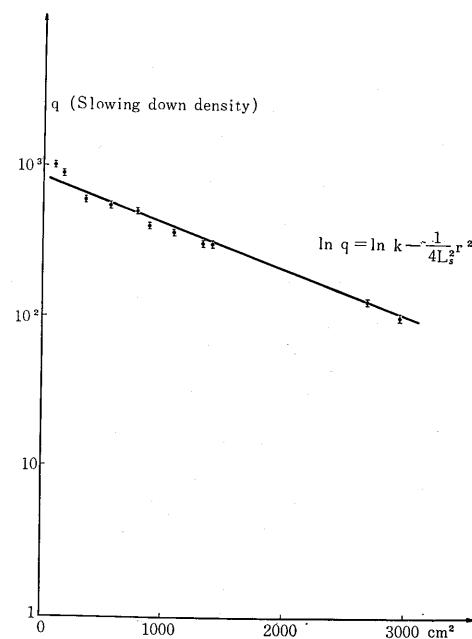


Fig. 4 Slowing down density distribution in (Cd) foil

In-115 against the square of the distance from the source. As the saturation activity is proportional to the slowing down density,  $q$ ,  $L_s^2$  was found to be 330 cm<sup>2</sup>. According to ANL-5800, Hill of ORNL gives  $L_s^2=329$  cm<sup>2</sup> for graphite, which is in good agreement with our result.

#### IV. EXPERIMENT WITH SUBCRITICAL ASSEMBLY

In this experiment, cadmium ratios in the assembly, material bucklings, neutron distributions in the assembly were measured with fuel elements loaded in the core assembly.

##### 1. Cadmium Ratio

Cadmium ratio is a quantity which is related to the proportion of slow neutron flux to the fast neutron flux in the assembly. It is an important parameter in studying such things as neutron spectrum, end effects and the effect of the neutron source. Cadmium ratio is generally defined as,

$$\text{Cadmium ratio} = \frac{A_{\text{bare}}}{A_{\text{res}}} \propto \frac{\phi_{ep} + \phi_{th}}{\phi_{ep}} = 1 + \frac{\phi_{th}}{\phi_{ep}}$$

where  $A_{\text{res}}$  is the induced activity of cadmium covered indium or gold foils, and  $A_{\text{bare}}$  is the induced activity of the foils without cadmium covers. As the cadmium absorbs neutrons of energy lower than its cut off energy, about 0.4 eV,  $A_{\text{res}}$  is proportional to fast neutron flux, while  $A_{\text{bare}}$  is proportional to the sum of fast and thermal neutron flux.

Cadmium ratios were measured in the vertical direction for the lattice spacing of 205.3 mm with the indium foils similar to the ones used in slowing down length measurement. Fig. 5 shows the result obtained. It should be noted here that the low average thermal neutron flux in the assembly caused considerably large statistical error in the result. Considerations on this aspect will be given in future experiments.

##### 2. Measurement of Material Buckling

Material buckling, which is a quantity related to the critical size of the multiplying assembly, was

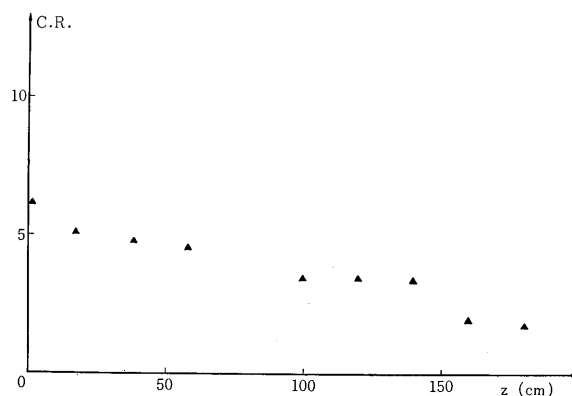


Fig. 5 Cadmium ratio distribution

one of the prime quantity to be measured in this experiment. It can be obtained from neutron flux distribution measurement in the assembly in the similar manner as diffusion length or slowing down length measurement. Thermal neutron flux distribution in an assembly with fuel elements loaded can be expressed as

$$\phi(x,y,z) = \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} A_{m,n} \cos \frac{m\pi x}{a} \cos \frac{n\pi y}{b} \sinh \frac{(c-z)}{b_{m,n}} \quad \dots (5)$$

where  $a$ ,  $b$  and  $c$  are lengths of the sides of the assembly with extrapolation lengths included. The material buckling,  $B^2$ , is now given by

$$B^2 = \left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{b}\right)^2 - \left(\frac{1}{b_{m,n}}\right)^2 \quad \dots (6)$$

The values of  $b$  and hence the values of material bucklings are obtainable from the slope of the plots of measured neutron flux distributions along the axial direction of the assembly. However, it should be mentioned here that points of flux measurements should be sufficiently far from the neutron source in order to avoid the effects of higher order harmonics of neutron flux.

In this experiment neutron flux distribution in the assembly was measured by the miniature BF<sub>3</sub> counter mentioned before. Lattice shapes and spacings were changed and bucklings for respective lattices were measured. Table 1 shows the lattice conditions for seven cases (No. C-0~C-6) which had been taken up in the experiment. As can be seen from this table, four measurements were taken on square lattices and three measurements on hexagonal lattices. The square lattice with 205.3 mm spacing corresponds to Calder Hall Reactor, and the hexagonal lattice with 237.1 mm spacing corresponds to Tokai Nuclear Power Reactor. Fig. 6 and Fig. 7 show the longitudinal and lateral thermal neutron flux distributions for square lattice

Table 1. Dimensions of various lattices

Lattice spacing	Square lattice		Hexagonal lattice	
	No.	Outline dimension	No.	Outline dimension
177.8 mm (approx. 7")	(C-0)	width, height, length (mm) 1,600×1,600×1,800	—	—
205.3 mm (approx. 8.1")	(C-1)	1,820×1,820×1,800	(C-4)	1,820×1,600×1,800
237.1 mm (approx. 9.35")	(C-2)	1,837×1,837×1,800	(C-5)	1,837×1,615×1,800
273.7 mm (approx. 10.4")	(C-3)	1,820×1,820×1,800	(C-6)	1,820×1,600×1,800

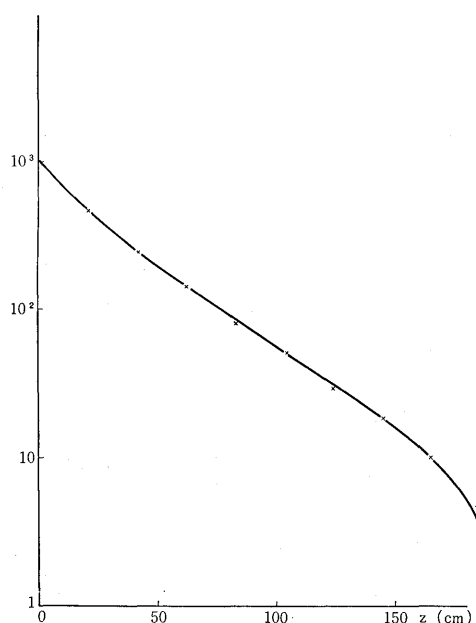


Fig. 6 Axial thermal neutron flux distribution

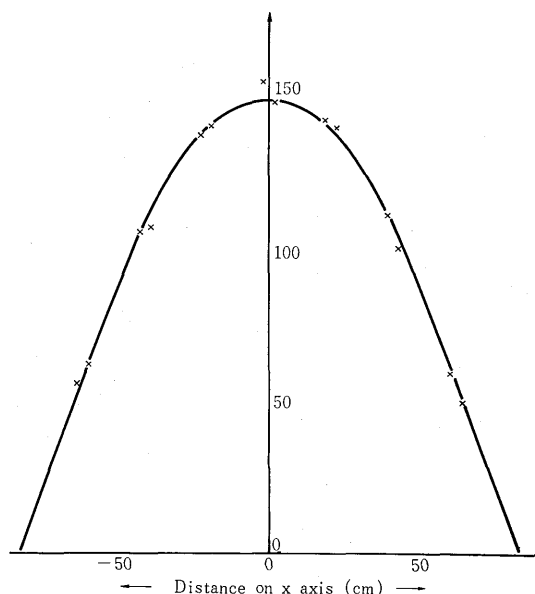


Fig. 7 Radial thermal neutron flux distribution

of 205.3 mm spacing. The variation of material buckling with lattice spacing and type of lattice is shown in Fig. 8.

Incidentally, the first order term of  $b_{m,n}$  in eq. 5, i.e.,  $b_{11}$ , was obtained from thermal neutron flux distribution in  $z$  direction by fitting the thermal neutron distribution with following equation:

$$\phi = \frac{A \cdot \sinh\left(\frac{c-z}{b_{11}}\right)}{\sinh \frac{z}{b_{11}}}$$

The curve was fitted by least square method using FACOM 128. Comparison of the measured values of material bucklings with theoretical values was

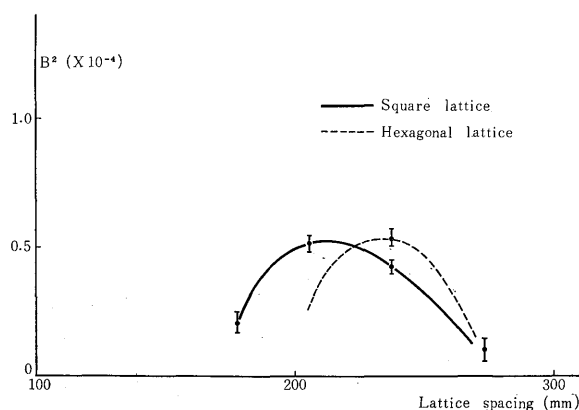


Fig. 8 Material buckling

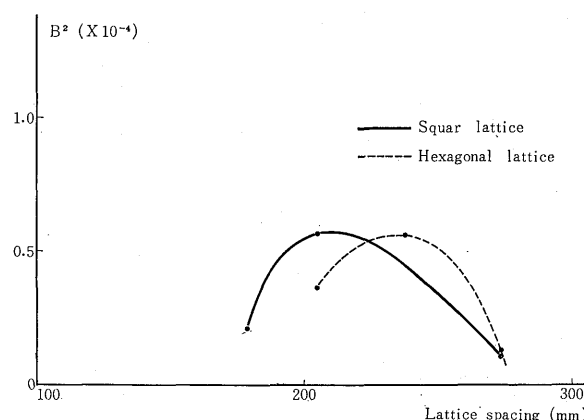


Fig. 9 Material buckling

done by calculating the bucklings from the equations used for core calculation of Calder Hall Reactor. The results are shown in Fig. 9. The discrepancies between theoretical and experimental values which are shown in Fig. 8 and 9, are thought to be resulted from

- (1) Inaccuracy of the various nuclear constants used for the calculation,
- (2) Statistical error in experimental data.

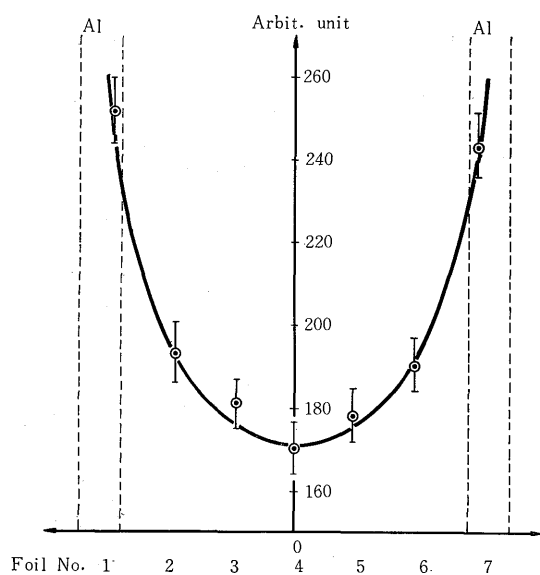
Especially, it is necessary to increase neutron flux intensity in the assembly in order to lessen the statistical error.

### 3. Measurement of Neutron Flux Distribution within a Fuel Rod

Thermal utilization,  $f$ , is one of the most important parameters in the criticality calculation of a reactor. Determination of thermal utilization requires the knowledge of thermal neutron flux fine structure within a unit cell. When considerable accuracy is required for this value, ordinary simple diffusion approximation is no longer applicable, and extremely complicated equations based on transport theory are generally used for this purpose. In many cases, however, the thermal neutron flux fine structure is measured by experiments and the obtained values are used in calculations of  $f$ . It was tried in this

**Table 2. Flux fine structure measurement data**

Distance from the center of the fuel (mm)	Relative intensity of activation	Deviation (%)
14.75	252	3.1
9.75	193	4.0
4.75	181	3.7
0	171	3.7
-4.75	179	3.6
-9.75	190	3.4
-14.75	243	3.2



**Fig. 10 Neutron distribution in a fuel rod**

experiment, also, to measure the neutron flux distribution within a fuel rod. The common way of measuring flux fine structure is to irradiate a thin Mn (90%)—Ni (10%) alloy wire of approximately 0.005" diameter in a hole penetrating the fuel radially, and the induced activity of the wire gives the flux distribution. However, as the neutron flux level in the assembly used was low, indium wire which had greater absorption cross section than Mn—Ni alloy was used in this experiment. What was actually done was that an indium wire of diameter less than 1 mm was irradiated for approximately 4 hours in a hole 3 mm in diameter which was made along the diameter of the fuel. In measuring the induced activity the wire was cut into pieces and was rolled to a thin foil of about 8 mm × 8 mm square in order to decrease the self-absorption of  $\beta$  rays within indium. The measurement was repeated five times under the same condition and the results were summed up in order to increase the statistical accuracy. The neutron flux distribution in the fuel was calculated from the counts obtained on G—M counter unit by applying suitable self-absorption correction, weight

correction and other necessary corrections. The relation used for self-absorption correction is

$$f = \frac{\mu \cdot s}{1 - e^{-\mu \cdot s}} \quad \dots\dots(9)$$

where  $f$  is self-absorption correction coefficient,  $\mu$  is self-absorption coefficient, and  $s$  is the thickness of the foil. The values obtained from this method are given in Table 2, and are plotted in Fig. 10.

## V. MEASUREMENT OF RADIATION LEVELS AROUND SUBCRITICAL ASSEMBLY

The subcritical assembly is covered on all sides by 3 mm thick cadmium sheets clad with aluminum in order to decrease thermal neutron leakage into the room. Radiation levels around the assembly were continually measured during the experiment and the alarm was to be given if the level exceeds permissible value.

Radiation shielding always presents some serious problems when one tries to design some experimental facilities involving radio-active materials. Excessive shielding is undesirable from economical point of view as well as from the consideration on experimental flexibility. Especially, as more subcritical assemblies are liable to be built in universities and laboratories in the future, it was thought to be desirable to obtain as many informations on shielding of this assembly as possible. Radiation levels around the bottom of the assembly were measured with 1.7 ton of natural uranium loaded, after several hours since Po—Be neutron source of 5 curies was inserted in the pedestal. It was found that,

- (1) The maximum thermal neutron flux around the assembly measured with thermal neutron monitor was 50 n/cm<sup>2</sup> sec.
- (2) The maximum fast neutron flux around the assembly measured with fast neutron monitor was 10 n/cm<sup>2</sup> sec.
- (3) The maximum  $\gamma$  radiation intensity around the assembly measured with scintillation counter, type PR-300, was 0.9 mr/hr.

From these results it can be seen that the radiation dose to be received cannot exceed maximum permissible dose in any position as the sum of these maximum values is approximately 2 mrem/hr. It was, therefore, possible to handle BF<sub>3</sub> counter without using its remote handling device, which had been prepared for unexpectedly high radiation levels around the assembly. Consideration on the  $\gamma$  activity of the fuel after irradiation was also given.  $\gamma$  radiations from fuel rods, placed in two positions where the neutron flux is the maximum and minimum, were measured after every experiment. The level on the surfaces were found to be 1.1 mr/hr~1.2 mr/hr, and no hazard was, therefore, expected from this source. It has, thus, been found that this subcritical assembly is quite safe from the viewpoint of radiation hazard.