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DETERMINATION
OF THE ABSOLUTE NEUTRON FLUX SPECTRUM
IN THE PCTR FAST NEUTRON CAVITY
FROM MULTIPLE FOIL
ACTIVATION MEASUREMENTS

D. F. Newman

August 1970

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By

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ABSTRACT

A large Fast Neutron Cavity (FNC), which permits the flexibility of altering the neutron spectrum, has been developed and operated in the central region of the Physical Constants Testing Reactor (PCTR). Analysis of multiple foil activation measurements in the center of the FNC was made to obtain a set of infinitely dilute saturated activities for twenty-six reactions. Activation responses of these foils included the entire range of possible neutron energies in the FNC. The SAND-II code was used for neutron spectrum determination from the experimental data. Neutron spectrum results unfolded from the multiple foil activation data were compared with proton recoil spectrometer measurements, in the center of the FNC, and with reactor theory calculations for this fast-thermal coupled reactor.

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CHAPTER 1

INTRODUCTION

Experimental determination of the energy distributions of the neutron flux in large dilute fast breeder reactors is currently of interest. These measurements are required to serve as benchmarks in testing computational methods used in fast reactor physics calculations. Proton recoil proportional counters are widely used for the measurement of fast neutron flux spectra. The relatively large size of the gas filled chambers, the counter's sensitivity to high gamma ray background radiation, and the necessity for signal cable leads out of the fast neutron flux environment are disadvantages of proton recoil counters which multiple foil activation techniques eliminate. Extremely detailed spatial variations in foil activities can be measured in fast breeder reactor sub-assemblies. If the energy dependent activation cross sections can be unfolded from the measured activation integrals to obtain the neutron flux spectrum with sufficient accuracy, the multiple foil activation measurements can be very useful.

One objective of this research project was to develop a facility in the Physical Constants Testing Reactor (PCTR) in which the neutron spectrum can be altered from a typical thermal reactor spectrum to a spectrum quite similar to that

in large dilute fast breeder reactors. A large Fast Neutron Cavity (FNC), which permits the flexibility of altering the neutron spectrum, has been designed, constructed and operated in the central region of the PCTR. This engineering phase of the project is described briefly in Chapter 2, Section II. Detailed information (including fuel geometry, dimensions, and compositions) necessary to perform reactor theory calculations for this fast thermal coupled reactor core are given in Appendix A.

The other major objective of this project was to perform multiple foil activation measurements in the center of the FNC. This experimental phase of the project is discussed in detail in Chapter 3. Analysis of multiple foil activation measurements was made to obtain a set of infinitely dilute saturated activities for twenty-six reactions. Six of these reaction rates were measured by threshold detectors that respond only to neutrons with energies in the fast range. The remaining twenty reaction rates were measured from activation of bare foils, foils covered by 0.1 cm thick cadmium, and foils covered by 1.0 cm thick ^{10}B filters. Activation responses of these foils included the entire range of possible neutron energies in the FNC. The infinitely dilute saturated activities measured and reported in this work are available for use in any study of neutron spectrum unfolding methods.

To illustrate the usefulness of these multiple foil activation measurements, one available method for neutron spectrum determination from the experimental data was exhibited. The neutron spectrum analysis and a technique developed to estimate the uncertainty in the unfolded neutron spectrum results are discussed in Chapter 4. In addition, the neutron spectrum results unfolded from the multiple foil activation measurements were compared with proton recoil spectrometer measurements and reactor theory calculations.

CHAPTER 2

EXPERIMENTAL APPARATUS

When results of an experimental study are reported, it is customary to give a brief description of the experimental apparatus used to obtain the measurements. Such a description is particularly appropriate for this work since the development of the Fast Neutron Cavity (FNC) in the PCTR facility was a major portion of this project. The successful development of the FNC, and subsequent neutron spectrum studies in the empty cavity provided experimental data to which reactor theory calculations can be compared. If analysis of these critical experiments reveals good agreement between theory and experiment, the analytical methods can be extended to predict the fast reactor core volume required to perform zoned fast reactor studies in the FNC.

In essence, zoned fast reactor experiments entail the insertion of a small volume of the fast reactor medium into the center of the test reactor being used. The fast reactor core volume in the experiments would be ~ 60 to 200 liters and is small enough that the majority of fissions in the reactor as a whole are caused by thermal neutrons. However, this volume should be large enough that the spectra of the neutron fluxes and adjoints are matched to within a few percent. In this manner, the saving of fast core material is achieved and at the same time a large measure of safety is provided since the neutronic characteristics to be controlled are mainly provided by the test reactor.

I. Description of the PCTR Facility

The PCTR graphite stack measures 86-1/4-inches by 86-1/4-inches by 90-inches deep. A cavity with maximum dimensions of 41-1/4-inches by 41-1/4-inches by 37-1/2-inches deep is centered on its horizontal axis. One face of the reactor, 86-1/4-inches by 86-1/4-inches by 26-1/4-inches, is built upon a motor driven carriage and can be moved back on tracks as far as six feet from the closed position. Easy access to the test cavity is provided when the face has been moved back in this manner. An 11-1/4-inch by 11-1/4-inch removable section is provided at the center of the moving face to allow access to the test cavity without opening the face. A horizontal section of the PCTR is shown in Figure 1, and the PCTR core face elevation is shown in Figure 2.

Surrounding the central cavity is a graphite moderated "driver" region. There are holes drilled in the graphite bars in this region to contain "driver" fuel and control rods. The fuel consists of highly enriched uranium rods extending the length of the sides. There are also many holes in the movable and stationary ends of the central region which contain short rods of highly enriched uranium fuel. Additional information on the specifications and description of the PCTR is given in references 1 and 2.

FIGURE 1

Negative No.
062 2324-2

PCTR Horizontal Section

Neg. No.
062 2324-1

FIGURE 2

PCTR Core Face Elevation

II. Design, Construction, and Operation of the Fast Neutron Cavity

A large Fast Neutron Cavity (FNC) assembly has been designed, constructed, and operated in the central region of the PCTR. This assembly transforms the thermal reactor neutron spectrum from the surrounding PCTR "driver" region into a spectrum typical of a fast reactor, by absorption of low energy neutrons and production of fast neutrons in a uranium metal region. The slightly enriched (0.85% ^{235}U) uranium buffer absorbs a large fraction of the incident thermal and epithermal neutrons while most of the incident fast neutrons pass through into the cavity region. Additional fast neutrons are produced in the neutron spectrum buffering process due to fission in the uranium.

The fast neutron spectrum in the central cavity is degraded from the fission spectrum due to inelastic scattering of fast neutrons in the uranium buffer. The neutron flux per unit lethargy, $\phi(u)$, peaks at a neutron energy about 300 keV in the central cavity, as shown in Chapter IV. This fast neutron spectrum is quite similar to spectra in large dilute fast breeder reactors.

This thermal to fast neutron spectrum conversion is accomplished without significant reduction of the integral flux^(a) in the central cavity, when compared to the integral flux in the driver region. A large depression of the integral flux in the central cavity would reduce the sensitivity of a fast thermal coupled reactor to small sample reactivity perturbations, and decrease the precision of fast reactor experiments in the central zone. Such a depression would occur if an absorber material were used instead of fissionable material for the neutron spectrum buffer.

On the other hand, the number of fissions in the neutron spectrum buffer and the central fast zone should be a small fraction of the fissions in the reactor as a whole. When most of the fissions are caused by thermal neutrons in the driver region, the neutronic characteristics of the fast thermal reactor are essentially unchanged from previous operating experience. Reactivity control is provided by the existing safety system in the thermal zone. A large measure of safety is provided since the prompt neutron lifetime in the PCTR ($\ell_p \approx 1.5 \times 10^{-3}$ sec)⁽³⁾ is several orders of magnitude longer than the prompt neutron lifetime in a full sized fast critical assembly ($\ell_p \approx 10^{-7}$ sec).⁽⁴⁾ Thus, the power level in the PCTR would rise with a period much longer than that of a fast reactor if the excess reactivity should exceed prompt criticality.

(a) Integral flux is defined by $\int_0^{\infty} \phi(E) dE$.

The Fast Neutron Cavity (FNC) is designed to utilize the space in the PCTR test region fully.⁽⁵⁾ A cutaway drawing (Figure 3) shows the assembly scheme using 1.5-inch diameter aluminum tubes stacked four rows thick enclosing a 1/2-inch thick stainless steel box (29.0-inches by 29.0-inches by 24.7-inches inner dimensions) on all sides. A removable stepped door in the front buffer provides access to the center of the FNC. The outside dimensions of this door (10.5-inches by 10.5-inches) will allow it to be removed through the 11-1/4-inch by 11-1/4-inch removable section in the moving face of the PCTR without opening the face. A 6-inch by 6-inch test sample can be removed through an access hole once the door has been withdrawn. The removable stepped door can also be removed when the movable face is fully opened (as shown in Figure 4). This mechanically stable rack of aluminum tubes containing uranium metal fuel is assembled as a liner of the central cavity in the PCTR. The inner two rows of tubes contain the uranium metal buffer fuel. The outer two rows of tubes contain high fissile content fuel and lucite rods. This undermoderated material ($H/^{239}\text{Pu} + ^{235}\text{U} \approx 24$) provides a transition zone in the reactor between the uranium buffer and the driver zone. This transition zone increases the efficiency of the thermal to fast neutron conversion due to its large neutron macroscopic fission cross section. In fact, it was not possible to drive the reactor critical without this thermal zone. The thermal neutron sink imposed by the large surface area of the

Neg 0680893-2

FIGURE 3. Cutaway View of the Fast Neutron Cavity

Neg 0681645-1

FIGURE 4. Removal of the Stepped Door to the FNC

4000 kg metallic uranium buffer could not be overcome by fully loading the PCTR driver fuel.

The design concept of the FNC is similar to that of the Karlsruhe STARK Fast Thermal Argonaut Reactor.⁽⁶⁾ However, the central fast neutron spectrum zone of the FNC is approximately five times larger than STARK. The concentric cubic design of the PCTR/FNC facility is illustrated in Figure 5, and the nuclei densities and volume in each region of the critical reactor are summarized. Based on previous calculational studies⁽⁷⁾ it is concluded that it would be possible to conduct meaningful reactivity experiments on fast reactor cores in the PCTR/FNC facility. This facility provides what is believed to be the largest fast neutron cavity in operation. It should provide the required volume to equilibrate the neutron spectrum in a zone of fast reactor medium inserted in the central cavity.

Zoned fast reactor experiments are most accurate for those cores for which full sized critical experiments would be exceedingly costly because of the large volume of core material needed⁽⁸⁾ (6030 liters for 1000 MWe capacity is perhaps typical). Thus, it is concluded that neutronics experiments for large dilute fast breeder reactor media could be conducted in the FNC.

Region	Region Volume, liters	Material	Nuclei Density, nuclei/cm ⁻³ × 10 ⁻²⁴
Cavity	376.2	N	3.98 × 10 ⁻⁵
		O	1.07 × 10 ⁻⁵
Stainless Steel	41.2	Fe	6.03 × 10 ⁻²
		Cr	1.73 × 10 ⁻²
		Ni	8.09 × 10 ⁻³
Uranium Buffer	274.6	²³⁸ U	3.36 × 10 ⁻²
		²³⁵ U	2.86 × 10 ⁻⁴
		Al	5.00 × 10 ⁻³
Transition Zone	353.4	²³⁹ Pu	3.72 × 10 ⁻⁵
		²⁴⁰ Pu	3.16 × 10 ⁻⁶
		²³⁵ U	1.18 × 10 ⁻⁴
		²³⁸ U	1.62 × 10 ⁻³
		O	5.14 × 10 ⁻³
		Al	1.05 × 10 ⁻²
		Zr	1.23 × 10 ⁻³
		C	6.58 × 10 ⁻³
Driver	1676.5	H	6.58 × 10 ⁻³
		²³⁵ U	3.53 × 10 ⁻⁵
		²³⁸ U	2.62 × 10 ⁻⁶
		Al	2.71 × 10 ⁻³
		O	1.12 × 10 ⁻⁴
		C	7.30 × 10 ⁻²
Reflector	8249.3	H	1.19 × 10 ⁻³
		C	8.28 × 10 ⁻²
		N	1.17 × 10 ⁻⁵
		O	3.15 × 10 ⁻⁶

FIGURE 5 Geometry and Composition of the PCTR Fast Neutron Cavity

III. Activation Foils and Counting System

The activation foils, neutron filters and holders, which are irradiated both inside the FNC and in the PCTR external thermal column were selected so that measured gamma ray activities could be related to the saturated activities of each foil reaction. Gamma ray scintillation spectroscopy techniques with NaI(Tl) counters⁽⁹⁾ were utilized to obtain activation data from the irradiated foils subsequent to the removal of the foils from the PCTR.

A. Gamma Ray Scintillation Counting System

The principal components of the counting system used to obtain foil activation data included:

(1) NaI(Tl) scintillation crystal; (2) photomultiplier tube; (3) high voltage power supply; (4) linear amplifier; (5) pulse height analyzer; and (6) scaler. A 2.5-inch diameter by 2.5-inch high Harshaw NaI(Tl) scintillation crystal (type 10 D 4) was used with a 2.0-inch diameter RCA photomultiplier tube (Model 6342). High voltage was obtained from a John Fluke Manufacturing Company Power Supply (Model 400 CE). When the counter was used as a gamma ray spectrometer, the signal from the photomultiplier tube was amplified by a Victoreen linear amplifier (Model 851A) and input to a multichannel analyzer. Use of the counter

as a single channel analyzer for relative activity measurements employed a RIDL scaler system including:

- (1) Model 31-21 preamplifier; (2) Model 30-23 linear amplifier; (3) Model 30-10B pulse height analyzer; (4) Model 40-28 scaler; and (5) Model 54-7 timer.

Preceding operation of the counting system for data acquisition each day, the Poisson Index of Dispersion Test⁽¹⁰⁾ was used. At least ten independent counts with a ^{137}Cs source were made and the quantity χ^2 ,

$$\chi^2 = \frac{\sum_{i=1}^N (\bar{n} - n_i)^2}{\bar{n}}$$

was calculated. This quantity is related to the probability that another set of data taken in the same fashion would have a dispersion leading to a larger value of χ^2 . Data was acquired only after these tests indicated that the counting equipment was functioning satisfactorily.

B. Multichannel Analyzer

A Nuclear Data 512 Channel Analyzer Computer (Series 130) with additional memory storage for spectrum integration and spectrum "stripping" was used in the gamma ray scintillation spectrometer

counting system. The counting time was in terms of analyzer "live" time because of a 10 microsecond dead time for data storage was required. The clock time at the beginning and end of each count was recorded so that the actual decay time to the median count of the counting time interval could be used in the data analysis. A Tektronix type RM 516 oscilloscope display was used and data was printed out by an IBM typewriter. The linearity of the multichannel pulse height analyzer was tested and the channel number versus gamma ray energy was calibrated at approximately 13.5 keV per channel using several standard gamma ray sources, as shown in Figure 6. The absolute efficiency of this counter was calibrated to determine the absolute activities of foils irradiated in these experiments.

It should be pointed out that the measurement of absolute activities is a necessity for neutron flux spectral determination by foil activation, i.e., one must measure the absolute neutron activation rate in every foil material that is employed. This procedure involves the determination of the absolute counter efficiency versus gamma ray energy calibration curve for the foil counting geometry, and the use of decay scheme branching ratios and radioactive

FIGURE 7

Automatic Foil Counting System

of the nominal distance between the foil and the crystal surfaces. The foil counting sequence, the foil counting time, and the maximum number of counts were programmed to obtain optimum accuracy in the relative activities. Each foil was counted three times for ten minutes or 50,000 counts, whichever came first, to allow all foil sets to be counted in a 24 hour period. The automatic counter output consisted of both a typewriter and a paper punched tape unit. The paper punched tape was read into the UNIVAC 1108 computer by means of a remote teletypewriter system. Thus, computer automated data reduction was obtained without requiring off line data preparation or system surveillance. This equipment was particularly useful for the foil self shielding measurements where varying thicknesses of a foil material were irradiated simultaneously in the FNC or the thermal column.

D. Foil Holders and Irradiation Facilities

The foil holders were designed to minimize the perturbing effects of nearby foils and neutron flux spatial variations. A graphite foil holder (3-inches in diameter) in the PCTR external thermal column⁽¹³⁾ positioned the foils 12.5-inches

above the lower surface on the thermal column centerline. Foil sized depressions were machined into the graphite foil holder to permit accurate foil placement. A vertical shaft connected an electrical drive motor to the foil holder which rotated the foils on its axis continuously during the foil irradiation. This rotator averaged any angular variation in the neutron fluence in the region of the thermal column foils.

The foil packet in the FNC was contained in an aluminum holder connected to a uranium plug (see Figure 8). This assembly was inserted into the FNC through a 1.0-inch diameter aluminum tube from the rear of the stationary section of the PCTR. After irradiation of the foil packet at the power level of 100 watts for two hours, the foil holder assembly was retrieved through the tube with a wire and the foils were immediately taken to the counting room. Some of the activation reactions measured in the FNC have short half lives, such as the $^{27}\text{Al}(n,p)^{27}\text{Mg}$ reaction with a 9.5-minute half life. Use of the foil removal tube allowed activation counting to start 15 minutes after the irradiation had finished.

FIGURE 8
Multiple Activation Foils and Holder

CHAPTER 3

MULTIPLE FOIL ACTIVATION MEASUREMENTS

I. Selection of Activation Foils

A review of methods for the determination of neutron spectra by foil activation was made by Zijp⁽¹⁴⁾ which lists the following requirements for selection of activation detectors:

- The material should contain only one or predominantly one stable isotope, so that possibilities for competing reactions with isotopes in the material are minimized.
- The material should be available with as high a purity as possible in order to minimize perturbing reactions with impurities.
- The material should be commercially available and not too expensive.
- The material should be simple to handle and safe to use; preferable in metallic form.
- The reaction should have a well known activation cross section, $\sigma_{\text{act}}(E)$, at all possible neutron energies.
- The reaction should have a convenient magnitude of the cross section so that excessive irradiation time and high flux levels are avoided.

- The product nuclide should have a well known and preferable a simple disintegration scheme.
- The product nuclide should have a suitable half life.
- The product nuclide should be the only remaining longer lived radioactive one produced by irradiation, so that counting should be possible without chemical separation.
- The product nuclide must have possibilities for rather simple absolute activity determination, preferably with gamma counting or coincidence counting techniques, so that corrections for absorption and self absorption of β particles are not required.

Due to this long list of requirements, the number of reactions in use is quite limited. The main requirements which limit the application of a number of reactions are connected to the half life, the order of magnitude of the cross section, and the availability of cross section data.

A. Foil Reactions

Transmutations induced by neutrons usually have radioactive species as product nuclides. The activities of the product radioisotopes can be used for measurement of the

absolute level and energy dependent shape of the neutron flux spectrum, which induced the radioactivity. Two primary considerations concerning the suitability of a foil reaction are: (1) the desired energy response, and (2) the properties of the activation product.

The activation reaction cross sections vary with neutron energy. The value of the activation cross sections must be known at all neutron energies, where the activation response functions are non-zero, for the activation measurements to yield useful information. An evaluated set of activation cross sections were obtained from the cross section library of the SAND-II computer code.⁽¹⁵⁾ A set of twenty-six different foil reactions, listed in Table I, were selected to obtain adequate coverage of all neutron energy ranges, since each reaction has a different activation response function. Six threshold reactions respond only to neutrons with energies in the fast range. Seven foil reactions are sensitive to thermal neutrons, but some reactions have differing non- $1/v$ cross section factors. The remaining thirteen foil reactions are primarily sensitive to narrow ranges of neutron energies because of resonance peaks in the activation cross sections.

TABLE I

MEASURED REACTIONS IN THE FAST NEUTRON CAVITY .

<u>Reaction</u>	<u>Filter Used</u>	<u>Lower Energy of Activation Response</u> ^(b)	
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	Bare	6.6	MeV
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	Bare	3.3	MeV
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	Bare	2.2	MeV
$^{238}\text{U}(n,f)\text{F.P.}$	Bare	1.5	MeV
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	Bare	720.0	keV
$^{103}\text{Rh}(n,n')^{103\text{m}}\text{Rh}$	Bare	300.0	keV
$^{239}\text{Pu}(n,f)\text{F.P.}$	Boron-10 ^(c)	23.0	keV
$^{232}\text{Th}(n,\gamma)^{\beta^-} \rightarrow ^{233}\text{Pa}$	Boron-10	13.5	keV
$^{235}\text{U}(n,f)\text{F.P.}$	Boron-10	12.0	keV
$^{238}\text{U}(n,\gamma)^{\beta^-} \rightarrow ^{239}\text{Np}$	Boron-10	5.8	keV
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Boron-10	2.0	keV
$^{197}\text{Au}(n,\gamma) \rightarrow ^{198}\text{Au}$	Boron-10	880.0	eV

(b) A lower neutron energy of activation response can be defined as the energy above which 95% of the activity for a particular neutron filter and foil combination is produced. This neutron energy, E_{lower} , is listed in Table I for each of twenty-six different reactions, based on the neutron spectrum results in Figure 18.

(c) Boron-10 covered foils irradiated separately from bare foils and Cd-covered foils. Bare foils in the thermal column normalize activities between separate irradiations.

TABLE I continued

<u>Reaction</u>	<u>Filter Used</u>	<u>Lower Energy of Activation Response</u> ^(b)
$^{238}\text{U}(n,\gamma)^{\beta}\text{-}^{239}\text{Np}$	Cd ^(d)	4.0 eV
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	Cd	2.2 eV
$^{232}\text{Th}(n,\gamma)^{\beta}\text{-}^{233}\text{Pa}$	Cd	1.8 eV
$^{238}\text{U}(n,\gamma)^{\beta}\text{-}^{239}\text{Np}$	Bare	1.8 eV
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Cd	1.35 eV
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	Cd	1.05 eV
$^{235}\text{U}(n,f)\text{F.P.}$	Cd	0.92 eV
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	Bare	0.92 eV
$^{239}\text{Pu}(n,f)\text{F.P.}$	Cd	0.80 eV
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	Bare	0.63 eV
$^{232}\text{Th}(n,\gamma)^{\beta}\text{-}^{233}\text{Pa}$	Bare	0.58 eV
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Bare	0.16 eV
$^{239}\text{Pu}(n,f)\text{F.P.}$	Bare	0.092 eV
$^{235}\text{U}(n,f)\text{F.P.}$	Bare	0.060 eV

(d) Cd-covered foils irradiated separately from bare foils and B^{10} -covered foils. Bare foils in the thermal column normalize activities between separate irradiations.

The activation product should be easily measured and interpreted correctly. Activity measurements were made by gamma ray spectroscopy⁽⁹⁾ to satisfy this requirement. Interpretation of activation data was simplified when there was a single radioactive species in the product. When two or more radioactive products were present, the half lives were sufficiently different so that the activity of the main product of interest could be measured. The gamma rays emitted by the product nuclides were of relatively high energy, with the exception of ^{103m}Rh , so that self absorption in the foils was not excessive.

B. Foil Materials and Specifications

Neutron activation detectors were chosen in the form of 1/2-inch diameter thin metal foils. This form was desirable because it was available without impurities having significant activation cross sections, and it could be handled conveniently as a detector. Introduction of these small foils in the PCTR was made without the introduction of voids or significant flux perturbations. The effect of neutron absorption was made negligibly small by the selection of aluminum alloy foils or by the application of a small correction. Activation self shielding was measured by irradiating several foil thicknesses

of the same material simultaneously. Foil materials were rolled to nominal thickness within a 10% variation. The foil detectors were punched in a die from the foil sheets after the thicknesses were verified. All plutonium aluminum alloy foils were clad with aluminum 0.005-inches thick to prevent the spread of contamination. A list of the foil materials and specifications used in the PCTR thermal column and the Fast Neutron Cavity is given in Table II. This foil set irradiated successively bare, cadmium covered, and boron covered in the FNC, and bare in the thermal column yielded sufficient activation data to determine the infinitely dilute saturated activities for the foil reactions listed in Table I.

TABLE II
FOIL MATERIALS AND SPECIFICATIONS

<u>Material</u>	<u>Thickness (Inches)</u>	<u>Description</u>
Copper Metal	0.0005 0.001 0.002 0.003 0.005	1/2" Diameter Foil
Gold Metal	0.0005 0.001 0.002 0.004	1/2" Diameter Foil
Al-8.3 wt% In	0.010	1/2" Diameter Foil
Indium Metal	0.005	1/2" Diameter Foil
Al-1.8 wt% Pu (8% ^{240}Pu)	0.005	1/2" Diameter Foil
Al-2.0 wt% Pu (8% ^{240}Pu)	0.035 dia.	1/4" Long Pin
Al-7.0 wt% U (93% ^{235}U)	0.004	1/2" Diameter Foil
Thorium Metal	0.002	1/2" Diameter Foil
Rhodium Metal	0.001 0.002 0.003	1/2" Diameter Foil
Nickel Metal	0.015	1/2" Diameter Foil
Aluminum Metal	0.250	1/2" Diameter Foil
Natural Uranium Metal	0.010	1/2" Diameter Foil
Depleted Uranium Metal {0.036% ^{235}U }	0.010	1/2" Diameter Foil

C. Neutron Filters for Activation Foils

The foil activation information from reactions sensitive to neutrons in both the thermal and resonance energy ranges were improved by surrounding the activation foils with neutron filters. Measurements of the foil activities induced by neutrons transmitted through 0.040-inch thick cadmium covers included only the activation due to epithermal neutrons. The absorption cross section of cadmium varies in such a way that the cadmium absorbed practically all of the neutrons with energy less than about 0.64 eV, but transmitted most of the epithermal neutrons. A higher effective threshold for neutron activation was obtained with foils inserted in 1.0 cm wall thickness boron (90% enriched ^{10}B) spheres. The very large $1/v$ absorption cross section of ^{10}B transmitted only those neutrons incident on the boron filter with energies greater than 1 keV. A boron sphere and cadmium cover are shown in Figure 9, with a list of their specifications.

CADMIUM COVER

0.040 in. Thick Cadmium Metal
 Cadmium Density = 8.65 gm Cd/cm^3
 Cd Surface Density = 0.004708

$$\frac{\text{Atoms Cd}}{\text{barn}}$$

BORON SPHERE

1.0 cm Wall Thick. Boron Powder
 90 at% Enriched ^{10}B
 ^{10}B Density = $1.60 \text{ gm } ^{10}\text{B/cm}^3$
 ^{10}B Surface Density = 0.09623

$$\frac{\text{Atoms } ^{10}\text{B}}{\text{barn}}$$

FIGURE 9

Cadmium and Boron Neutron Filters

The energy responses of the seven foil reactions sensitive to both thermal and resonance neutrons were altered by covering the foils with cadmium and boron. Thus, foil activation data spanning three neutron energy ranges was obtained from each foil reaction without requiring any additional activation cross section information. The filter neutron transmission factors can be accurately determined^(16,17) since the cross sections for the filter materials are well known.

An isotropic neutron flux is incident on a detector irradiated in a cavity whose dimensions are large compared with those of the detector. It is assumed that the perturbation of the detector on the reactor is negligible. The impinging flux spectrum is filtered by cadmium or boron-10, as the case may be, before activation of the covered foil takes place. Neutron absorption in the filter is the primary means by which the incident flux spectrum is attenuated. Calculations by Pearlstein and Weinstock⁽¹⁶⁾ for cadmium covered foils show that the effect of neutron scattering in the cover is to reduce the activation of the foil by 1% below what would be observed if there were no scattering. For an isotropic flux, the neutrons backscattered in the second cover tend to compensate for the loss of neutrons backscattered in the first cover.

The 90% enriched boron-10 sphere is nearly a purely absorbing filter. Calculations were performed for the boron sphere in the center of the FNC using the 26 group transport theory PCTR calculational model, described in Appendix C, to determine the magnitude of the scattering effects in boron. The multigroup neutron transmission factors, T_g , were obtained from ratios of the flux inside the boron sphere, centered in the FNC, to the flux in the center of the empty FNC. These transmission factors are compared with simple exponential absorption factors, $e^{-\Sigma_a \bar{X}}$, in Table III. Numerical results of this comparison show that simple exponential absorption approximates the transmission factors quite well. The SAND-II algorithm, described in Chapter 4, I.A., uses single exponential absorption factors to calculate neutron transmission through filters.

TABLE III

BORON SPHERE NEUTRON TRANSMISSION FACTORS

<u>Energy Interval</u>	<u>Group</u>	$\frac{T_g^{(e)}}{g}$	$\frac{\left(e^{-\Sigma_a \bar{X}}\right)_g^{(f)}}{g}$	$\frac{\left(T/e^{-\Sigma_a \bar{X}}\right)_g}{g}$
6.20 - 10.00 Mev	1	0.9310	0.9680	0.962
3.80 - 6.20	2	0.9120	0.9570	0.953
2.40 - 3.80	3	0.9290	0.9630	0.965
1.30 - 2.40	4	0.9290	0.9520	0.976
0.76 - 1.30	5	0.9140	0.9660	0.946
0.38 - 0.76	6	0.8400	0.9190	0.915
0.19 - 0.38	7	0.8370	0.8640	0.969
96.00 - 190.00 keV	8	0.8320	0.8150	1.021
44.00 - 96.00	9	0.7820	0.7700	1.015
21.00 - 44.00	10	0.6940	0.6760	1.026
9.50 - 21.00	11	0.5870	0.5680	1.032
4.40 - 9.50	12	0.4350	0.4330	1.004
2.00 - 4.40	13	0.2920	0.2960	0.985
0.94 - 2.00	14	0.1646	0.1647	0.999
0.44 - 0.94	15	0.0706	0.0714	0.990
0.20 - 0.44	16	0.0191	0.0207	0.922
< 0.20 keV	17 - 26	0.0000	0.0000	-----

(e) $T_g = \frac{\phi_g \left(\text{Inside boron sphere} \right)}{\phi_g \left(\text{centered in FNC} \right)}$, calculated using 26 group transport theory PCTR spherical model.

(f) $\left(e^{-\Sigma_a \bar{X}} \right)_g$ = simple exponential absorption factor for boron sphere

where, $(\Sigma_a)_g$ = energy interval averaged macroscopic neutron absorption cross section for group g.

$\bar{X}$ = average path length to the foil, through the 1.0 cm thick boron spherical shell = 1.129 cm

D. Irradiation Time and Power Level Considerations

A single power level and irradiation time interval was chosen for all foil irradiations to eliminate uncertainties due to power level calibration and variations of the fission product gamma ray activities in fission foils after different irradiation intervals. The maximum power level permitted by the PCTR Operating Specifications is 250 watts. Reactor instrumentation scale range and bypass switching required to achieve maximum power were not necessary at the usual power level for irradiation at 100 watts. The activities of several of the short half life products, from the $^{27}\text{Al}(n,p)^{27}\text{Mg}$, $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$, and $^{103}\text{Rh}(n,n')^{103\text{m}}\text{Rh}$ reactions, approached their saturated activities after a two hour irradiation. Additional irradiation time would have been prohibitive due to high radiation exposure to PCTR operations personnel, since the radiation dose rate between the stationary and moveable faces of the PCTR 24 hours after a two hour irradiation at 100 watts was approximately 600 mRem per hour. The 100 watt power level was approached with the PCTR on a positive period of ~ 30 seconds. When the reactor reached 100 watts, the control rods were moved rapidly to level out the power. During the 138 second time

interval required to increase the power level from 1 watt to 100 watts, less than 0.05% of the integral power was generated. At the completion time of the irradiation, all control rods were scrambled to decrease the power level as rapidly as possible. Thus, the integral power generated during the irradiation was determined very precisely. This procedure was repeated for successive irradiations of bare, cadmium covered, and boron covered foils in the Fast Neutron Cavity. Absolute activities of foils at the completion time of the irradiation can easily be extrapolated to absolute saturated activities, as shown in Section III.C., by a simple exponential saturation factor based on steady state irradiation for a finite time.

II. Activation Foil Counting Technique

A. Absolute Efficiency Calibration of the Counter

The absolute efficiency of the gamma ray scintillation spectrometer was calibrated as a function of gamma ray energy. This calibration was necessary to measure absolute activities of irradiated foils. The need for absolute activities of all reactions, with the same irradiation history, is fundamental to the activation technique for neutron spectrum measurements. Energy dependent neutron activation cross sections, $\sigma_i(E)$, are used as weighting functions for the neutron spectrum in activation integral equations of the form:

$$A_i = \int_0^{\infty} \sigma_i(E) \phi(E) dE \quad i = 1, 2, \dots, n. \quad (3-1)$$

The neutron spectrum, $\phi(E)$, can be unfolded from this set of n integral equations only if the activation rate for the i^{th} reaction, A_i , is known relative to all other activation rates in the set. Since different reactions have different residual nuclei, in many cases, it is necessary to measure the absolute activity for all different residual nuclei having the same irradiation history.

The absolute efficiency was measured by counting an IAEA set of standard sources of known activity (within $\pm 1\%$ standard deviation) to convert the number of counts registered in a peak above the Compton background to the number of gamma rays of a particular energy emitted from a source. Gamma ray yields were obtained from the Table of Isotopes⁽¹⁸⁾ for the ^{241}Am , ^{57}Co , ^{203}Hg , ^{22}Na , ^{137}Cs , ^{54}Mn , ^{88}Y , and ^{60}Co standard sources. Results of the absolute counter efficiency calibration are shown in Figure 10. The standard sources were positioned 0.643 centimeters above the scintillation crystal. Irradiated foils were also positioned at that height above the crystal to duplicate the counting geometry. The effect of source displacement from the axial centerline of the crystal was measured using a ^{137}Cs pinpoint source. Results of these measurements shown in Figure 11, indicate that the counter efficiency for 1/2-inch diameter distributed source (same as the foil geometry) is 0.6% less than a point source on the crystal centerline. This small correction was applied to the absolute counter efficiency calibration given in Figure 10. During the irradiated foil counting, the standard source with a gamma ray energy nearest the gamma ray emitted from the product nuclides in the foil was counted to monitor the counter efficiency drift in

FIGURE 10. GAMMA RAY SCINTILLATION COUNTER EFFICIENCY CALIBRATION
SOURCE PLACED 0.643 cm CRYSTAL JULY 15, 1969

FIGURE 11. COUNTER EFFICIENCY AS A FUNCTION OF SOURCE DISPLACEMENT FROM THE AXIAL CENTERLINE OF THE CRYSTAL

the gamma ray spectrometer. This data was used to correct the results in Figure 10 to the counter efficiency at the time of the irradiated foil counting.

B. Counting Intervals and Activity Decay Intervals

The simultaneous irradiation of the foils listed in Table I made it necessary to follow a precise schedule for irradiated foil counting. The schedule listed in Table IV was planned to insure that good counting statistics were obtained from the short half lived products and that no conflicts developed between counting the same foils on different counting systems. All foils were irradiated and counted within a 16 hour work period with the exception of the uranium foils. The uranium foils were counted on the gamma ray spectrometer the following day. The fission product activities from ^{239}Pu and ^{235}U result from several fission products of varying concentrations. The half life of the integral fission product activity above 400 keV is approximately equal to the decay time for the first hundred hours. The integral fission product activities were counted between 6 and 8 hours decay time so that good counting statistics were obtained and the half life (least squares fit to a single exponential) did not change rapidly during counting. The activity decay interval for the $^{27}\text{Al}(n,p)^{27}\text{Mg}$ reaction ($t_{1/2} = 9.5 \text{ min.}$) was reduced to the minimum time required to unload the foils from the reactor and transport them to the counting room. The activity of

TABLE IV
Gamma Ray Spectrometer Measurement Intervals

Reaction	Half Life	γ -Ray Energy Counted (keV)	% Yield of γ -Ray	Activity Decay Interval (hr)	Foil Counting Interval (min)
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	9.5 min	840	70%	0.25	10
$^{103}\text{Rh}(n,n')^{103\text{m}}\text{Rh}$	57.0 min.	40	0.4%	1.0	10
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	15.0 hr.	1369	100%	5.0	20
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	54.0 min.	1293	80%	7.0	2
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	4.5 hr.	335	50%	7.0	2
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	12.8 days	511	38%	8.0	10
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	2.7 days	412	95%	9.0	10
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	71.0 days	810	99%	10.0	80
$^{238}\text{U}(n,\gamma)^{\frac{238}{2}}\text{-}^{239}\text{Np}$	2.35 days	106	23%	20.0	4
$^{238}\text{U}(n,f)\text{F.P.}$	varies	510 $\rightarrow$ 675		20.0	4
$^{235}\text{U}(n,f)\text{F.P.}$	varies	510 $\rightarrow$ 675		20.0	4

TABLE IV continued
Relative Activity Measurement Intervals

Reaction	Half Life	γ -Ray Threshold Bias (keV)	Activity Decay Interval (hr)	Maximum Counting Interval (min)	Minimum Number Counts/ Interval
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	12.8 hr.	400	4	10	or 50,000
$^{239}\text{Pu}(n,f)\text{F.P.}$	Least Squares fit to single exponential	400	6	5	or 40,000
$^{235}\text{U}(n,f)\text{F.P.}$	+	400	7	5	or 40,000
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	54.0 min.	500	8	5	or 40,000
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	2.7 days	300	10	10	or 50,000
$^{232}\text{Th}(n,\gamma)^{\beta-233}\text{Pa}$	27 days	200	11	5	or 40,000

the indium foils was very high at the time of removal from the reactor. A decay interval of approximately 7 hours reduced the 54 minute half life ^{116m}In product to acceptable activity levels without excessive counter dead time. Time intervals for activity decay and counting were recorded for all measurements on the gamma ray spectrometer so that saturated activities could be calculated from the measured activities.

C. Multichannel Analysis of Gamma Ray Emission Spectra

One important property of scintillation detectors is the ability to relate the resulting voltage pulse height distribution to the incident gamma ray energy spectrum. Gamma ray emission spectra from foils irradiated in the FNC were analyzed using a calibrated NaI(Tl) scintillation detector and a 512 channel analyzer. The channel number versus gamma ray energy calibration, shown in Figure 6, related the voltage pulse height to the gamma ray energy. The absolute efficiency calibration discussed in Sections II.A converted the number of counts registered in a peak above the Compton background to the number of gamma rays of a particular energy emitted from the foil. At least three counts were taken on each foil after irradiation. A typical gamma ray emission spectrum from a nickel foil irradiated in the FNC is shown in Figure 12. The activity of the product nuclide counted above the Compton background in a dominant^(g) gamma ray peak was corrected to zero decay interval using the reference value half life:⁽¹⁸⁾

$$A(0) = A(t)e^{\lambda t}$$

where, $\lambda = \ln 2 / t_{1/2}$

t = the decay interval (t=0 at the time irradiation is terminated).

^(g) A dominant gamma ray peak occurs in the gamma ray emission spectra in which gamma rays are emitted with highest intensity at a particular energy, for the decay scheme of the product nuclide.

FIGURE 12

Typical Gamma-Ray Emission Spectrum

One criterion in the selection of the foil reactions was that the dominant gamma ray peak should be isolated in the gamma ray emission spectrum. Subtraction of the residual contributions from the counts registered in the interval of a gamma ray peak was simplified when the peak was isolated. The 335 keV peak from the $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ reaction was not totally isolated from the 417 keV peak due to the $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ reaction. The 1293 keV peak from $^{116\text{m}}\text{In}$, however, was isolated from the 335 keV peak. Using the ratio of the count rate in the 335 keV interval to the count rate in the 1293 keV interval for an indium foil irradiated in the thermal column, $(^{335}\text{A}/^{1293}\text{A})_{\text{th}}$, the count rate registered in the 335 keV interval for an indium foil irradiated in the FNC was corrected for the contribution from 417 keV gamma rays:

$$^{335}\text{A}_{\text{FNC corr.}} = ^{335}\text{A}_{\text{FNC}} - ^{1293}\text{A}_{\text{FNC}} \left(\frac{^{335}\text{A}}{^{1293}\text{A}} \right)_{\text{th}} \quad (3-3)$$

This correction was possible since no other gamma rays are emitted from indium above 192 keV and below 819 keV, as shown by decay schemes for $^{114\text{m}}\text{In}$, $^{115\text{m}}\text{In}$, and $^{116\text{m}}\text{In}$ in the Table of Isotopes.⁽¹⁸⁾ An estimate of the fraction of the reaction rate, for the indium foil irradiated in the thermal column, due to inelastic scattering was calculated from the gold cadmium ratio measured in the thermal column.

$$\left[\frac{A(n, n')}{A(n, \gamma)} \right]^{115\text{In}} = \frac{\sigma_o \left[{}^{115}\text{In}(n, n') \right]}{\bar{\sigma}_{\text{th}} \left[{}^{115}\text{In}(n, \gamma) \right]} \frac{\phi_{\text{fast}}}{\phi_{\text{th}}} \quad (3-4)$$

and

$$\frac{\phi_{\text{fast}}}{\phi_{\text{th}}} \approx \frac{\phi(u) \Delta u}{\phi_{\text{th}}} = \frac{\bar{\sigma}_{\text{th}} \left[{}^{197}\text{Au}(n, \gamma) \right] \ln \left(\frac{10\text{MeV}}{E_s^{\text{eff}}} \right)}{\text{RI}_{\text{eff}}^{197\text{Au}} [\text{CdR-1}]^{197\text{Au}}} \quad (3-5)$$

where, $\left[\frac{A(n, n')}{A(n, \gamma)} \right]^{115\text{In}}$ = The ratio of inelastic scattering activation rate to neutron capture activation rate in ^{115}In .

$\sigma_o \left[{}^{115}\text{In}(n, n') \right]$ = Inelastic scattering microscopic cross section for ^{115}In , high energy "plateau" value = 0.35 barns. (19)

E_s^{eff} = effective threshold energy of neutrons for inelastic scattering in ^{115}In = 1.65 MeV. (19)

$[\text{CdR-1}]^{197\text{Au}}$ = The gold cadmium ratio -1 for 0.005-inch thick foils irradiated in the thermal column = 217 ± 8

$\text{RI}_{\text{eff}}^{197\text{Au}}$ = Effective resonance integral for 0.005-inch thick gold foils above 0.6 eV = 270 barns. (20)

$\frac{\bar{\sigma}_{\text{th}} \left[{}^{197}\text{Au}(n, \gamma) \right]}{\bar{\sigma}_{\text{th}} \left[{}^{115}\text{In}(n, \gamma) \right]}$ = ratio of thermal neutron spectrum averaged microscopic capture cross sections for ^{197}Au and ^{115}In = 0.606 (21)

Combining equations (3-4) and (3-5) and substituting

numerical values, we get:

$$\left[\frac{A(n, n')}{A(n, \gamma)} \right]^{115\text{In}} = \frac{\sigma_o \left[{}^{115}\text{In}(n, n') \right] \ln \left(\frac{10\text{MeV}}{E_s^{\text{eff}}} \right) \bar{\sigma}_{\text{th}} \left[{}^{197}\text{Au}(n, \gamma) \right]}{\text{RI}_{\text{eff}}^{197\text{Au}} [\text{CdR-1}]^{197\text{Au}} \bar{\sigma}_{\text{th}} \left[{}^{115}\text{In}(n, \gamma) \right]} = 6.5 \times 10^{-6} \quad (3-6)$$

Thus, inelastic scattering contributes negligible activity in the foil irradiated in the thermal column. Equation (3-3) was used to correct the counts registered in the 335 keV peak, from the indium foil irradiated in the FNC, for contributions other than the ^{115m}In activity.

The assumption that the neutron flux per unit lethargy, $\phi(u)$, in the PCTR thermal column is constant from 5 eV to 10 MeV, in Equation (3-5), was investigated using the PCTR/FNC transport theory calculational model (see Appendix C). Calculations show that $\phi(u)$ decreases monotonically with increasing neutron energy above 5 eV for neutrons penetrating the graphite reflector and thermal column to depths greater than 30 cm. The assumption in Equation (3-5) leads to an over-estimate for $\left[\frac{A(n,n')}{A(n,\gamma)} \right]^{115}\text{In}$ in Equation (3-6). This confirms the statement that inelastic scattering contributes negligible activity in the indium foil irradiated in the thermal column.

D. Use of the PCTR Thermal Column for Activity Normalization

Absolute saturated activities of fission reactions were difficult to obtain directly because the flux level in the FNC was only about 1.5×10^9 neutrons/cm²-second at a power level of 100 watts. In addition, the uncertainties in the fission yields of product nuclides usually counted when direct measurement of the fission rate is attempted would limit the accuracy to about 10%. Integral gamma ray counting can be used to normalize activity data from a foil irradiated in the FNC to the same type foil irradiated in the PCTR thermal column, with the same irradiation history. These measurements, called "relative activity measurements," are useful for reactions having well known thermal neutron activation cross sections, but complex decay schemes. Absolute activation rates can be measured from foils with simple decay schemes (such as gold, copper, and indium) irradiated in the thermal column. If activation in the thermal column is entirely due to thermal neutrons, the infinitely dilute saturated activity per nucleus of target, $A_{\text{Sat}_0}^{\text{T.C.}}$, at a constant thermal flux level, ϕ_{th} , is given by:

$$A_{\text{Sat}_0}^{\text{T.C.}} = \bar{\sigma}_{\text{act}} \phi_{\text{th}} = \sigma_{\text{o act}} \left(\frac{\bar{\sigma}_{1/v}}{\sigma_{\text{o } 1/v}} \right) \phi_{\text{th}} g_{\text{th}} \quad (3-7)$$

where, $\bar{\sigma}_{\text{act}}$ = thermal neutron spectrum averaged microscopic cross section for the activation reaction.

$\sigma_{\text{o act}}$ = the 2200 m/sec microscopic cross section value for the activation reaction.

$\bar{\sigma}_{1/v}$ = thermal neutron spectrum averaged cross section for a neutron absorber whose velocity-dependent cross section inversely with neutron velocity.

$\sigma_{\text{o 1/v}}$ = the 2200 m/sec cross section value for the 1/v absorber.

The non-1/v factor, ϵ_{th} , for each reaction was calculated for the thermal column spectrum from transport theory using computer code THERMOS.⁽²²⁾

$$\epsilon_{\text{th}} = \frac{\hat{\sigma}_{\text{act}}}{\sigma_{\text{o act}}} = \left(\frac{\bar{\sigma}_{\text{act}}}{\sigma_{\text{o act}}} \right) \left(\frac{\sigma_{\text{o 1/v}}}{\bar{\sigma}_{1/v}} \right) \quad (3-8)$$

Using 2200 m/sec activation cross section values from BNL-325⁽²¹⁾ the thermal column flux level was determined from the expression:

$$\phi_{\text{th}} \left(\frac{\bar{\sigma}_{1/v}}{\sigma_{\text{o 1/v}}} \right) = \frac{A_{\text{sat}}^{\text{T.C.}}}{\sigma_{\text{o act}} \epsilon_{\text{th}}} \quad (3-9)$$

Absolute infinitely dilute saturated activities measured for the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$, and $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ reactions in the PCTR thermal column were used to determine the thermal flux level at a power level of 100 watts.

Consistent values were obtained from the thermal flux by this method, as shown in Table V. The average thermal column flux at 100 watts (from Equation 3-9) was used in Equation 3-7 to calculate the infinitely dilute saturated activities of foils irradiated in the thermal column for which direct measurement of absolute activities was difficult. The inferred activation rates (including $^{239}\text{Pu}(n,f)$, $^{235}\text{U}(n,f)$, $^{238}\text{U}(n,\gamma)$, and $^{232}\text{Th}(n,\gamma)$ reactions) are given in Table V.

The assumption that activation in the thermal column is entirely due to thermal neutrons can be evaluated by a simple calculation using the measured cadmium ratio and effective cross sections. The cadmium ratio for 0.005-inch thick gold foils irradiated in the thermal column with the FNC installed in the PCTR was 218 ± 8 . This value is in agreement with previous measurements made with a thermal test core installed.⁽²³⁾ A numerical example will be made for the $^{238}\text{U}(n,\gamma)$ reaction, since it has the largest epithermal resonance integral to thermal neutron cross section ratio of

TABLE V

PCTR Thermal Column Flux Level Determination

Thermal Column Reaction	Infinitely Dilute Saturated Activity	σ_0 (Barns)	ξ_{th}	$\phi_{th} \left(\frac{\bar{\sigma}_{1/v}}{\sigma_{01/v}} \right) \frac{\text{neutrons}}{\text{cm}^2 \text{ sec}}$
$^{63}\text{Cu}(n, \gamma)^{64}\text{Cu}$	$4.26 \times 10^{-16} \frac{\text{dps}}{\text{nucleus}} (+2.0\%)$ (h)	4.5 ± 0.1 (i)	1.00000	9.476×10^7
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$	$8.94 \times 10^{-15} \frac{\text{dps}}{\text{nucleus}} (+2.0\%)$	98.8 ± 0.3	1.00468	9.006×10^7
$^{115}\text{In}(n, \gamma)^{116m}\text{In}$	$1.56 \times 10^{-15} \frac{\text{dps}}{\text{nucleus}} (+2.8\%)$	162.0 ± 3.0	1.00622	9.549×10^7

Average Thermal Column Flux at 100 watts = $9.343 \times 10^7 (+2.6\% \text{ Standard Deviation})$

Thermal Column Saturated Activities Inferred from Cross Section and Flux Level Normalization

Thermal Column Reaction	σ_0 (Barns)	ξ_{th}	$\phi_{th} \left(\frac{\bar{\sigma}_{1/v}}{\sigma_{01/v}} \right)$	Infinitely Dilute Saturated Activity
$^{239}\text{Pu}(n, f)\text{F.P.}$	$740.6 \pm 3.5^{(i)}$	1.07281	9.343×10^7	$7.42 \times 10^{-14} \frac{\text{fissions}}{\text{sec-nucleus}} (+2.6\%)$ (h)
$^{235}\text{U}(n, f)\text{F.P.}$	577.1 ± 0.9	0.97483		$5.26 \times 10^{-14} \frac{\text{fissions}}{\text{sec-nucleus}} (+2.6\%)$
$^{238}\text{U}(n, \gamma)^{239}\text{U}$	273 ± 0.04	1.01342		$2.59 \times 10^{-16} \frac{\text{disintegr.}}{\text{sec-nucleus}} (+3.0\%)$
$^{232}\text{Th}(n, \gamma)^{233}\text{Th}$	7.4 ± 0.1	0.97370		$6.73 \times 10^{-16} \frac{\text{disintegr.}}{\text{sec-nucleus}} (+2.9\%)$

(h) All uncertainty percentages are 1 standard deviation.

(i) 2200 m/sec microscopic cross section values from Reference 21.

the reactions measured in the thermal column. The ratio of epithermal to thermal neutron activation in ^{238}U , $(A_{\text{epi}}/A_{\text{th}})^{238}$, can be related to the measured gold cadmium ratio assuming the epithermal flux varies as $1/E$ with neutron energy, E :

$$\left(\frac{A_{\text{epi}}}{A_{\text{th}}}\right)^{238} = \left(\frac{\sigma_{\text{o}g_{\text{th}}}}{\sigma_{\text{o}g_{\text{th}}}}\right)^{197} \left(\frac{F_{\text{th}}^{197}}{F_{\text{th}}^{238}}\right) \left(\frac{\text{RI}_{\text{eff}}^{238}}{\text{RI}_{\text{eff}}^{197}}\right) \frac{1}{(\text{CdR}-1)^{197}} \quad (3-10)$$

where $\left(\frac{F_{\text{th}}^{197}}{F_{\text{th}}^{238}}\right)$ = the ratio of thermal neutron flux depression factors for 0.005-inch thick gold foils to 0.010-inch thick depleted uranium foils irradiated in an isotopic thermal flux = 0.86. (20)

$\left(\frac{\sigma_{\text{o}g_{\text{th}}}}{\sigma_{\text{o}g_{\text{th}}}}\right)^{197}$ = the ratio of effective thermal neutron spectrum averaged microscopic cross sections for ^{197}Au to ^{238}U (from Table V) = 35.9.

$\left(\frac{\text{RI}_{\text{eff}}^{238}}{\text{RI}_{\text{eff}}^{197}}\right)$ = The ratio of the effective epithermal resonance integrals for 0.010-inch thick depleted uranium foils to 0.005-inch thick gold foils irradiated in an isotopic $1/E$ epithermal flux = 0.222. (20)

Substitution of numerical values in Equation 3-10

results in a value of 0.032 for $(A_{\text{epi}}/A_{\text{th}})^{238}$.

This small correction was applied to the $^{238}\text{U}(n,\gamma)$

activation rate measured in the thermal column.

Similar calculations for all other thermal column foil

reactions show that activation contributions due to

epithermal neutrons in the thermal column are negligible.

Since absolute saturated activities per target nucleus for infinitely dilute foils irradiated in the thermal column have been determined from Equation 3-7 or measured directly, relative activity measurements can be used to determine absolute saturated activities in the FNC from the expression:

$$\left(A_{\text{sat}}^{\text{FNC}} \right)_i = \left(A_{\text{sat}_o}^{\text{T.C.}} \right)_i \left(F_{\text{th}} \right)_i \left(\frac{SA^{\text{FNC}}}{SA^{\text{T.C.}}} \right)_i \quad (3-11)$$

where $\left(A_{\text{sat}}^{\text{FNC}} \right)_i$ = the absolute saturated activity per target nucleus for the i^{th} type foil irradiated in the FNC (disintegrations/sec - nucleus).

$\left(A_{\text{sat}_o}^{\text{T.C.}} \right)_i$ = The absolute infinitely dilute saturated activity per target nucleus for the i^{th} type foil irradiated in the thermal column (disintegrations/sec - nucleus).

$\left(F_{\text{th}} \right)_i$ = The thermal neutron flux depression factor for the i^{th} type foil irradiated in the thermal column.

$\left(\frac{SA^{\text{FNC}}}{SA^{\text{T.C.}}} \right)_i$ = the specific activity (counts/target nucleus) ratio for the i^{th} type foil irradiated in the FNC relative to the same type foil irradiated in the thermal column simultaneously.

The precision of these absolute saturated activity determinations was nominally within a $\pm 3\%$ uncertainty (1 standard deviation).

E. Relative Activity Measurements

Specific activities of foils irradiated in the FNC relative to foils of the same type irradiated simultaneously in the thermal column were accurately measured (within $\pm 0.5\%$) using both the automatic and manual single channel gamma ray scintillation counting system. The counting data was analyzed using a revised version of the computer code APDAC-1⁽²⁴⁾ which corrects the counting data for activity decay during counting, room background, foil mass variations, residual activity, counter drift, and dead time. The half life of the product nuclide being counted was forced, in the weighted least squares fitting method, to be equal to the appropriate tabulated value. A statistical analysis of the corrected counting data was performed by APDAC and the error in the quoted relative activities was expressed as standard deviation in percent.

F. Fission Product Activity Measurements

Determination of the relative fission rates in Al - 1.8 wt% ^{239}Pu and Al - 7 wt% ^{235}U foils irradiated both in the FNC and in the thermal column was similar to the relative activity measurements. The method used to correct the counting data for activity decay during counting was different. Since the half life of the integral fission product activity varies with the decay interval, the foils were permitted to cool until the half life was changing slowly. As shown in Table IV, the foils containing ^{239}Pu irradiated in the thermal column and the FNC were counted in succession, alternately, five times beginning at a time six hours after the irradiation was terminated. The counting data from the foil irradiated in the thermal column was fitted, by a weighted least squares method in APDAC-I, ⁽²⁴⁾ to a single exponential in time spanning the approximately 50 minute foil counting session. Knowing the activity of the thermal column foil at any time during the foil counting session from the fitted decay function, the activity of the FNC foil, counted at some time, t , in the foil counting session was divided by the activity of the thermal column foil at time, t , to obtain the

fission rate of ^{239}Pu in the FNC relative to the thermal column. A statistical analysis of the five relative fission product activity ratios obtained during the counting session was performed by APDAC-I and the error in the average relative activity was expressed in percent, one standard deviation. A similar 50 minute foil counting session, beginning seven hours after termination of the irradiation, was used to obtain the fission rate of ^{235}U in the FNC relative to the thermal column.

Fast neutrons, with energy greater than 0.5 MeV, are required to produce fission in ^{238}U . Thus, a ^{238}U foil irradiated in the thermal column, where the fast neutron flux is negligible (see Equation 3-5) with respect to the thermal flux, would not yield useful information concerning the ^{238}U fission rate. To avoid absolute fission product counting difficulties, the ^{238}U fission rate was measured relative to the ^{235}U fission rate in the FNC. The specific gamma ray activities of two thin uranium metal foils irradiated simultaneously in the FNC were used to measure the ratio, at time t after irradiation, of counts originating from ^{238}U

fission products to counts from ^{235}U fission products in natural uranium. One foil was composed of uranium depleted to 0.036 wt% ^{235}U ; the other foil was natural uranium (0.711 wt% ^{235}U). The gamma ray activities were counted on the multichannel gamma ray spectrometer between 510 keV and 675 keV in order to exclude the ^{239}Np pile up peaks at lower energies. This gamma ray energy interval was used in a previous study by Newman and Pervich⁽²⁵⁾ to determine $P(t)$. The function $P(t)$ is defined as the count rates in the energy interval (510 - 675 keV) at time t per ^{235}U fission, divided by the count rate in the energy interval (510 - 675 keV) at time t per ^{238}U fission, after a two hour uranium foil irradiation. The ^{238}U fission rate relative to the ^{235}U fission rate in the FNC per target nucleus, $\bar{\sigma}_f^{28} / \bar{\sigma}_f^{25}$, can be determined from relative integral fission product activities of the depleted uranium to the natural uranium foils, F_D/F_N , as a function of time, t , after irradiation, where

$$\frac{F_D(t)}{F_N(t)} = \frac{N_D^{28} \bar{\sigma}_f^{28} + P(t) N_D^{25} \bar{\sigma}_f^{25}}{N_N^{28} \bar{\sigma}_f^{28} + P(t) N_N^{25} \bar{\sigma}_f^{25}} \quad (3-12)$$

$\bar{\sigma}_f^{28}$ = The FNC neutron spectrum averaged ^{238}U microscopic fission cross section

$\bar{\sigma}_f^{25}$ = The FNC neutron spectrum averaged ^{235}U microscopic fission cross section.

N = The number of nuclei of the superscripted isotope in the subscripted foil type.

Superscripts 28 and 25 refer to ^{238}U and ^{235}U , and subscripts N and D refer to natural and depleted uranium, respectively. Solving Equation 3-12 for the ratio of spectrum averaged fission cross sections, $\bar{\sigma}_f^{28}/\bar{\sigma}_f^{25}$, we get

$$\frac{\bar{\sigma}_f^{28}}{\bar{\sigma}_f^{25}} = P(t) \left(\frac{N_N^{25}}{N_N^{28}} \right) \left[\frac{\frac{N_D^{28}}{N_D^{25}} \frac{F_D(t)}{F_N(t)} - \frac{N_D^{28}}{N_D^{25}} \frac{N_N^{28}}{N_N^{25}}}{1 - \frac{N_N^{28}}{N_D^{28}} \frac{F_D(t)}{F_N(t)}} \right] \quad (3-13)$$

The function $P(t)$ corrects for the time dependence of the ^{238}U integral fission product activity relative to the ^{235}U integral fission product activity, as indicated in Equation 3-12, to yield relative fission rates independent of decay time. Measurement of this function, $P(t)$, is based on a technique developed by J. R. Wolberg, et. al.,⁽²⁶⁾ using the 1.6 MeV gamma ray activity from ^{140}La and the total

^{140}La fission yields from ^{235}U and ^{238}U to calibrate the time dependent correction factor for the gamma ray energy interval (510 - 675 keV) used to measure $F_D(t)/F_N(t)$. The absolute saturated activity (fissions/ ^{238}U nucleus - sec) for the $^{238}\text{U}(n,f)$ reaction was determined from the absolute saturated activity of the $^{235}\text{U}(n,f)$ reaction in the FNC by use of the relative fission rate given in Equation 3-13 as the FNC spectrum averaged microscopic cross section ratio of ^{238}U fission to ^{235}U fission:

$$A_{\text{sat}}^{238\text{F}} = \left(\frac{\sigma_f^{28}}{\sigma_f^{25}} \right) A_{\text{sat}}^{235\text{F}} \quad (3-14)$$

where $A_{\text{sat}}^{238\text{F}}$ = the absolute saturated activity for the $^{238}\text{U}(n,f)$ reaction in the FNC with the PCTR at a 100 watt power level.

$A_{\text{sat}}^{235\text{F}}$ = The absolute saturated activity for the $^{235}\text{U}(n,f)$ reaction in the FNC with the PCTR at a 100 watt power level, from Equation 3-11.

III. Foil Activation Data Reduction

A. Self-Shielding Correction Measurements

The observed activations of foils used in the determination of the neutron flux spectrum must be corrected for the self shielding of the neutron flux in the foil and for flux depression caused by the presence of the foil. The specific activity of a foil, expressed in disintegrations per second per target nucleus, is referred to as the infinitely dilute specific activity, $SA(o)$, after correction for self shielding. Infinitely dilute specific activities were obtained from foils irradiated in the FNC and in the thermal column, by extrapolation of specific activities of different thickness foils (of the same type, irradiated in positions of equivalent neutron flux) to zero thickness. A series of gold, indium, and copper foils ranging from 0.0005-inches to 0.005-inches thickness were irradiated both bare and cadmium covered in the FNC and bare in the thermal column. Theoretical curves for the resonance integral self shielding⁽²⁰⁾ and the thermal absorption self shielding⁽²⁷⁾ in the FNC and the thermal column, respectively, were normalized to the experimental data by a least squares fit to obtain the extrapolation to zero thickness foil activities.

Resonance integral self shielding factors $G(t)$, were obtained from foil thickness dependent resonance integral data, $RI(t)$, measured in an epithermal flux that varied as $1/E$ with neutron energy, E .⁽²⁰⁾ Since the variation of the epithermal flux spectrum in the FNC is not $1/E$, the infinitely dilute specific activity, $SA(o)$, for each type foil was determined from a least squares fit normalization factor, C , using the expression:^(j)

$$SA(o) = SA(t) + C \left[RI(o) - RI(t) \right]_{1/E} \quad (3-15)$$

The self shielding factor, $G(t)$, is defined as the ratio of the specific activities from a foil of thickness, t , to an infinitely dilute value. By rearranging Equation 3-15 we get

$$G(t) = \frac{SA(t)}{SA(o)} = 1 - \frac{C}{SA(o)} \left[RI(o) - RI(t) \right]_{1/E} \quad (3-16)$$

As shown in Figure 13 for gold and copper foils, the extrapolation was nearly linear for thermal neutron self shielding. Resonance integral self shielding was considerably non linear, and a reduction of the uncertainty in the extrapolation was obtained by the use of very thin foils. Indium aluminum alloy foils were used for the self shielding measurements, along

^(j) Equation (3-15) assumes that the resonance integral self shielding in the FNC is nearly a linear function of the resonance integral self shielding in a $1/E$ neutron spectrum.

FIGURE 13 . SELF-SHIELDING FACTORS FOR GOLD AND COPPER FOILS

with indium metal foils because very thin indium metal was quite fragile to handle. Self shielding measurements in the thermal column were in agreement with the results of Klema and Ritchie.⁽²⁷⁾ Self shielding corrections for the Al - 1.8 wt% ²³⁹Pu foils, the Al - 7.0 wt% ²³⁵U foils, other dilute foils, and the threshold reaction foils were assumed to be unity since the effect of neutron absorption was negligible. The excellent correlation of the resonance integral self shielding data measured in the FNC and calculated values from Equation (3-16), shown in Figure 13, confirms the adequacy of the assumption that the resonance integral self shielding in the FNC is a linear function of the resonance integral self shielding in a 1/E spectrum.

B. Absolute Activity Determination for Foil Reactions

The activity of the product nuclide in an irradiated foil was obtained from the multi-channel analysis of the gamma ray emission spectra. The activity above the Compton background in a dominant gamma peak was corrected to zero decay interval by Equation 3-2. In order to convert the measured foil activity (counts/sec) into absolute activity per nucleus of target material, the absolute efficiency of the counter $\eta(E)$, at the energy of the counted gamma ray peak and the number of gamma rays emitted by the product nuclide per disintegration,⁽¹⁸⁾ $\gamma(E)$, were used:

$$A_{\text{abs}} \left(\frac{\text{disintegrations}}{\text{sec-nucleus}} \right) = \frac{A(\text{counts/sec})}{\eta(E)\gamma(E)N_T} \quad (3-17)$$

The number of target nuclei in the foil, N_T , can be expressed in terms of foil mass, M ; foil material atomic weight, A ; Avogadro's number, N_A ; and the isotopic abundance, I_A , of the target nuclei in the foil material:

$$N_T = \left(\frac{M}{A} \right) N_A I_A \quad (3-18)$$

C. Dilute Foil Saturated Activities

The absolute activities of foils at zero decay interval (from Equation 3-17) were corrected to infinitely dilute activities by the use of the measured self shielding factors (described in Section III.A.). Extrapolation of these absolute activities (obtained after a two hour irradiation) to their values at saturation (corresponding to an infinitely long irradiation time) was made using the reference values for the decay constants.⁽¹⁸⁾ Infinitely dilute saturated activities, A_{sat}^x , were obtained from the measured absolute activities, A_{abs}^x , by applying self shielding and saturation factors using the expression:

$$A_{\text{sat}}^x = \frac{A_{\text{abs}}^x}{(1 - e^{-\lambda_x T})} \left(\frac{SA_x(o)}{SA_x(t)} \right) \quad (3-19)$$

where, λ_x = decay constant for the x^{th} product nuclide;

T = the irradiation time interval = 2 hours;

$\left(\frac{1}{1 - e^{-\lambda_x T}} \right)$ = saturation correction factor

$\frac{SA_x(o)}{SA_x(t)}$ = self shielding correction factor

Infinitely dilute saturated activities are useful experimental data because they can be directly related to the integral of the energy dependent microscopic activation cross section neutron flux product.

$$A_{\text{sat}}^{ij} = \int_0^{\infty} T_j(E) \sigma_i(E) \phi(E) dE \quad (3-20)$$

where

A_{sat}^{ij} = infinitely dilute saturated activity for the i^{th} activation product covered by the j^{th} neutron filter (disintegration/sec - target nucleus);

$T_j(E)$ = the energy dependent neutron transmission factor for the j^{th} neutron filter ($T(E) = 1$ for bare foils);

$\sigma_i(E)$ = The energy dependent infinitely dilute microscopic activation cross section for the i^{th} target nucleus (cm^2);

$\phi(E)$ = the energy dependent neutron flux (neutrons/ cm^2 - sec - MeV)

Experimental results for the infinitely dilute

saturated activities for 26 foil reactions measured

in the FNC are presented in Table VI.

The average uncertainty (root - mean - square deviation)

in the measured values of the infinitely dilute

saturated activities for all 26 reactions was less

than $\pm 3\%$. One available method for determination of

the energy dependent shape and absolute level of the

neutron flux, from the experimental data in the FNC,

is exhibited in Chapter 4.

TABLE VI

Infinitely Dilute Saturated Activities

Reaction	Filter	Infinitely Dilute Saturated Activity	<u>disintegrations</u> <u>second-nucleus</u>	Uncertainty in ^(k) Measurement
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	Bare	5.6213×10^{-20}		3.3%
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	Bare	3.0650×10^{-19}		2.8%
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	Bare	1.6470×10^{-17}		3.9%
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	Bare	2.452×10^{-17}		2.3%
$^{103}\text{Rh}(n,n')^{103\text{m}}\text{Rh}$	Bare	4.7261×10^{-16}		3.9%
$^{238}\text{U}(n,f)\text{F.P.}$	Bare	$3.4801 \times 10^{-17(1)}$		3.4%
$^{235}\text{U}(n,f)\text{F.P.}$	Bare	$5.5208 \times 10^{-15(1)}$		2.8%
	Cadmium	$4.0348 \times 10^{-15(1)}$		2.9%
	Boron-10	$1.2715 \times 10^{-15(1)}$		2.7%
$^{239}\text{Pu}(n,f)\text{F.P.}$	Bare	$1.0695 \times 10^{-14(1)}$		2.9%
	Cadmium	$4.5987 \times 10^{-15(1)}$		2.7%
	Boron-10	$2.0486 \times 10^{-15(1)}$		3.7%
$^{238}\text{U}(n,\gamma)^{\beta-}^{239}\text{Np}$	Bare	5.3612×10^{-16}		3.8%
	Cadmium	5.3612×10^{-16}		3.8%
	Boron-10	1.5623×10^{-16}		4.2%
$^{232}\text{Th}(n,\gamma)^{\beta-}^{233}\text{Pa}$	Bare	1.5977×10^{-15}		2.6%
	Cadmium	1.5889×10^{-15}		2.6%
	Boron-10	9.0549×10^{-16}		2.6%
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Bare	1.2195×10^{-16}		2.2%
	Cadmium	1.1343×10^{-16}		2.2%
	Boron-10	0.4086×10^{-16}		2.2%
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	Bare	6.3127×10^{-15}		2.2%
	Cadmium	6.1824×10^{-15}		2.2%
	Boron-10	3.4678×10^{-16}		2.2%
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	Bare	2.783×10^{-14}		2.8%
	Cadmium	2.709×10^{-14}		2.8%

(k) All uncertainty percentages are 1 standard deviation.
 Root - Mean - Square
 deviation =

$$\sqrt{\frac{\sum_{i=1}^N \sigma_i^2}{N}} = 2.977\%$$

(1) Fissions
sec-nucleus

CHAPTER 4

NEUTRON FLUX SPECTRUM DETERMINATION FROM MEASURED
INFINITELY DILUTE SATURATED ACTIVITIESI. The SAND-II Computer Code

The description of the computer code SAND-II and the method of solution used in the code is the original work of W. N. McElroy, et. al.^(28,29) A summary of this work will be given to familiarize the reader with the method utilized to analyze the experimental data presented in this report. For a more detailed description, the reader is referred to their original work. The mathematical procedure for this method involves selection of an initial neutron spectrum approximation and its subsequent iterative perturbation to obtain a best fit simultaneous solution for a system of activation integral equations. Such a solution is not unique. Gold has stated that, in general, an exact or even approximate solution is not required, but that the solution should properly satisfy the set of activation integral equations and certain subsidiary conditions imposed by physical implications;⁽³⁰⁾ he calls such a solution an appropriate solution. As indicated by McElroy⁽³¹⁾ and Gold,⁽¹²⁾ exact solutions are not unique and, in some cases, do

not even exist depending upon the choice of detectors and on the errors which are always present in the experimentally determined activation cross sections and saturated activities.

A. The SAND-II Algorithm

The calculated saturated activities per target nucleus of a set of n foil detector reactions can be expressed by:

$$A_i^{(K)} = \int_0^{18 \text{ MeV}} \phi^{(K)}(E) \sigma_i(E) e^{-N_r X_r \sigma_r(E)} dE \quad (4-1)$$

($i = 1, 2, \dots, n$)

where, $A_i^{(K)}$ = the activity of the i^{th} foil calculated at the K^{th} iteration (disintegrations/sec - target nucleus);

$\phi^{(K)}(E)$ = the K^{th} iterative energy dependent neutron flux (neutrons/cm² - sec - MeV);

$\sigma_i(E)$ = the energy dependent activation cross section of the i^{th} foil reaction (cm²);

N_r = r^{th} foil cover nuclei density (nuclei/cm³);

X_r = average path length through r^{th} foil cover (cm);

$\sigma_r(E)$ = r^{th} foil cover removal cross section (for cadmium and boron-10 covered foils), (cm²).

$K = 0$ refers to the initial neutron spectrum approximation.

The true neutron flux spectrum, $\phi(E)$, satisfies the activation integral equations within experimental uncertainties. Thus, the comparison of the ratio of measured to calculated activity for each foil reaction,

$$R_i^{(K)} = \frac{A_i}{A_i^{(K)}} \quad (4-2)$$

is a measure of the "appropriateness" of the K^{th} iterative solution for the neutron flux spectrum. The energy range between 10^{-10} and 18 MeV is represented by 620 discrete contiguous intervals in the code. Then the iterative algorithm⁽³²⁾ for the j^{th} energy interval used in SAND-II can be written as:

$$\phi_j^{(K+1)} = \phi_j^{(K)} \exp \left\{ C_j^{(K)} \right\} \quad (4-3)$$

$$j = 1, 2, \dots, 621$$

where,

$$C_j^{(K)} = \frac{\sum_{i=1}^n w_{i,j}^{(K)} \ln \left\{ A_i / A_i^{(K)} \right\}}{\sum_{i=1}^n w_{i,j}^{(K)}} \quad (4-4)$$

A weighting function ($w_{i,j}$) for each energy interval was obtained, for each foil reaction, based on the sensitivity function (energy dependent cross section multiplied by differential flux] for that foil calculated at the current iteration,

$$w_{i,j}^{(K)} \equiv \left[E_{H,i} - E_{L,i} \right]^{(K)} A_{i,j}^{(K)} / A_i^{(K)} \quad (4-5)$$

where, $E_{L,i}$ and $E_{H,i}$ are the lower and upper energy limits, respectively, within which 90% of the activity is produced, for the i^{th} foil reaction. The foil weighting functions were applied to the averaging procedure in Equation 4-4 to obtain an average correction factor for each energy interval. The average correction factors were then applied to the current iterative flux values at each energy interval by Equation 4-3, to obtain the next iterative flux spectrum.

The criteria for achievement of an appropriate solution spectrum were based on the examination of the deviation from unity in the ratio of measured to calculated activities for each iteration, Equation 4-2. One criterion for termination of the iterative process was achieved when the average error (root - mean - square deviation) of the measured to calculated activity ratios was equal to the average experimental error (root - mean - square deviation) of the measured absolute saturated activities for all foils. To preclude the possibility of an inordinately large number of iterations caused by over optimism of the user as to expected precision, a secondary criterion

was included, based on the rate of iterative change of the average error (rms deviation) for the measured to calculated activity ratios. The activation cross section data, such as that tabulated in the SAND-II cross section library, may actually contain errors in both shape and magnitude substantially larger than the errors involved in the measurement of absolute saturated activities for all foils. Such mutual inconsistencies in cross section data will produce larger values than expected for the average error (rms deviation) of the measured to calculated activity ratios from the iterative neutron spectrum solution. These larger error values will become fairly invariant with successive iterations. Thus, the iterative process was terminated when successive iterations reduced the average error of activity ratios less than 0.3% (root - mean - square deviation).

B. Neutron Spectrum Coverage by Foil Reactions

The 26 foil reactions were selected so that the set of activities calculated by SAND-II after each spectrum iteration was sensitive to changes in the shape and magnitude of the energy dependent neutron flux, $\phi(E)$. It was important to consider the shape and magnitude of the energy dependent activation cross sections in selecting activation foils and neutron filters. The neutron spectrum shape can be unfolded from the set of activation integral equations with better resolution if the energy dependent activation cross sections are quite different. Energy dependent activation responses of foil reactions sensitive to both thermal and intermediate energy neutrons were altered by covering the foils with cadmium and boron-10 filters, as discussed in Chapter 3 Section I.C. The relative "uniqueness" of the neutron spectrum solution was dependent on the energy ranges covered by each foil reaction, and the overlap of the coverage among all 26 reactions. This relative uniqueness was observed as the width of the envelope of all SAND-II solutions obtained

from the measured activation integrals, using different initial trial functions for the neutron spectrum. For example, if all the reaction cross sections varied as $1/\sqrt{E}$ the set of activation integral equations would be ill conditioned. The resulting SAND-II neutron flux spectrum solutions would not change from the shape of the initial trial functions. Different initial trial function spectral shapes would merely result in different magnitudes for the absolute integral neutron flux, $\int_0^{\infty} \phi(E) dE$, required to satisfy all the absolute saturated activities for a given spectral shape. A solution for such a system of activation integral equations does not exist. Hence, an attempt was made to select a set of reactions with quite different energy dependent activation response functions that offer some possibility for a "relatively unique" absolute neutron spectrum solution.

The range of neutron energies covered by each of the 26 foil reactions was examined after each spectrum iteration by the SAND-II code. The energy region, within which 90% of the detector activity was produced, was used to provide an

indication of the detector sensitivity ranges. Since the energy dependent shape of the neutron spectrum is altered in the iterative process by the SAND-II algorithm, Equation 4-3, the neutron energy range covered by each foil reaction shifts slightly as required by the current iterative neutron spectrum solution. Coverage of the neutron spectrum by each of the 26 foil reactions measured in the FNC is listed in Table XII. The reactions are listed in order of decreasing high energy response. Nominal 90% activity limits were obtained from the best SAND-II solution representation of the neutron spectrum.

TABLE VII

Neutron Spectrum Coverage by Foil Reactions

<u>Reaction</u>	<u>Filter</u>	<u>Nominal 90% Activity Limits, MEV</u>	
		<u>Lower Energy</u>	<u>Upper Energy</u>
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	Bare	6.600	12.50
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	Bare	3.300	9.400
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	Bare	2.200	6.900
$^{238}\text{U}(n,f)\text{F.P.}$	Bare	1.500	5.500
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	Bare	0.720	4.900
$^{103}\text{Rh}(n,n')^{103\text{m}}\text{Rh}$	Bare	0.300	3.800
$^{239}\text{Pu}(n,f)\text{F.P.}$	Boron	0.023	2.100
	Cadmium	8.00×10^{-7}	0.690
	Bare	9.20×10^{-8}	0.320
$^{235}\text{U}(n,f)\text{F.P.}$	Boron	0.012	1.400
	Cadmium	9.20×10^{-7}	0.600
	Bare	6.00×10^{-8}	0.475
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	Boron	5.75×10^{-3}	0.760
	Cadmium	4.00×10^{-6}	0.400
	Bare	1.80×10^{-6}	0.400
$^{232}\text{Th}(n,\gamma)^{233}\text{Th}$	Boron	1.35×10^{-2}	0.660
	Cadmium	1.80×10^{-6}	0.425
	Bare	5.75×10^{-7}	0.400
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Boron	2.00×10^{-3}	0.500
	Cadmium	1.35×10^{-6}	0.300
	Bare	1.60×10^{-7}	0.280
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	Boron	8.80×10^{-4}	0.475
	Cadmium	2.20×10^{-6}	0.063
	Bare	6.30×10^{-7}	5.75×10^{-2}
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	Cadmium	1.05×10^{-6}	2.10×10^{-6}
	Bare	9.20×10^{-7}	2.00×10^{-6}

C. Numerical Test Case of the Neutron Spectrum Resolution

A numerical test case of the SAND-II iterative method was evaluated to estimate the uncertainty in the neutron spectrum results. The SAND-II analysis determines both the magnitude and the energy dependent shape of the neutron flux, from a set of infinitely dilute saturated activities that have been inserted into the computer code. A 621 group neutron flux spectrum, similar to that in the FNC, was chosen as the "exact" spectrum for the test case. Saturated activities were calculated from the "exact" spectrum for each of the foil reactions used in this study. The activation cross sections in the SAND-II cross section library were also assumed to be exact. All the foil reactions listed in Table 1 were used, including bare foil reactions, cadmium covered foil reactions, and boron-10 covered foil reactions. These saturated activities (calculated, exactly) were then inserted into SAND-II in an attempt to reproduce the "exact" neutron spectrum from which the foil reaction activities were calculated.

1. Iterative Spectrum Solution Criterion

A study of the sensitivity of the iterative solution spectrum to the initial flux shape approximation was made using several initial trial functions. This study was merely a test of the iterative method used in SAND-II without propagation of experimental errors in the activation integrals or the cross section data. The absence of experimental errors was considered in the specification of the iterative spectrum solution criterion. The iterated spectrum solution changed quite rapidly during the first five or six iterations, when the fractional error in the ratio of the measured to calculated activities was large. As the iterated spectrum was changed to minimize the fractional error in the measured to calculated activities, the spectrum changes per iteration were much smaller. The parametric study in Figure 14 shows that the average error (root - mean - square deviation) of the measured to calculated activity ratios was changing less than 0.2% per iteration after approximately twenty iterations.

The energy dependent spectrum correction factors, from Equation 4-4, were all near unity and subsequent iterations would not significantly change either the energy dependent neutron spectrum or the fractional error in the measured to calculated activity ratios. Thus, a twenty iteration limit was selected as a practical criterion for achieving a solution with the SAND-II code for this test case.

2. Sensitivity of Solution to the Initial Trial Function

A constant flux per unit lethargy was used as the initial trial function to obtain a SAND-II twenty iteration solution with an average rms deviation of the measured activity from the calculated activity of 2.07% error. Successive calculations using a Gaussian function, one energy decade full width at half maximum, centered at 10^{-3} MeV, 10^{-2} MeV, 10^{-1} MeV, and 1 MeV, respectively, as the initial trial functions were made to obtain SAND-II twenty iteration solutions.

As shown in Figure 14, the twenty iteration solution obtained using the Gaussian function centered at 10^{-1} MeV as the initial trial function had an average error of 1.93% rms deviation in the measured to calculated activity ratios. This solution over estimated the flux in the energy range of the "exact" spectrum maximum flux per unit lethargy, $\phi_{\max}(u)$. The twenty iteration solution obtained using a constant flux per unit lethargy as the initial trial function under estimated the flux in the energy range of the exact spectrum maximum flux per unit lethargy. These two solutions, found by trial and error to have the smallest average error in the measured to calculated activity ratios, were averaged and input to the SAND-II code to obtain a twenty iteration solution with an average error of 0.77% rms deviation in the measured to calculated activity ratios.

FIGURE 14. PARAMETRIC STUDY OF THE SAND-II SOLUTION CRITERION FOR DIFFERENT INITIAL TRIAL FUNCTIONS

3. Locating the Flux Peak

If neutron spectrum calculations for the FNC were not readily available from reactor theory computer codes, a neutron spectrum approximation obtained by trial and error (averaging two solutions having the smallest average error, as described previously) is one alternate. The neutron energy range in which the maximum flux per unit lethargy, $\phi_{\max}(u)$, occurs in the spectrum can be located by the trial and error method. The average error in the measured to calculated activity ratios was smallest, in the numerical test case solutions using Gaussian shaped initial trial functions, when the Gaussian function was centered on the peak of the exact spectrum, $\phi_{\max}(u)$. Successive SAND-II calculations were made using Gaussian functions centered at different neutron energies, to locate the flux peak. Iteration to a final solution was accelerated by inserting the average of two solutions having the smallest average error in the measured to calculated activity ratios for the neutron spectrum trial function, as shown in Figure 15. This averaging procedure redistributes the weighting

functions, W_{ij} , for the energy-dependent neutron flux correction factors, C_j , used in the iterative algorithm (Equation 4-3), and results in accelerated convergence. Gaussian functions were chosen for input neutron spectrum approximations since it was known that the flux per unit lethargy, $\phi(u)$, generally has a shape similar to this in fast reactors. Neutron spectra in other environments, such as that in intermediate reactors, would be poorly approximated by a single Gaussian function. Thus, the trial and error method used to locate the flux peak in the FNC would not be directly applicable for those environments. Generally, the success of the trial and error method depends to large degree on knowledge of the spectrum, and the rms error in the measured to calculated activity ratios gives a measure of the appropriateness of the trial function.

4. Error Correlations of the SAND-II Solutions

The error in the SAND-II absolute neutron flux spectrum, $\phi(E)$, as a function of neutron energy, E , in the Fast Neutron Cavity was estimated from an error correlation with the numerical test case. The absolute neutron flux spectrum SAND-II test case solution was compared with the exact test case spectrum, from which the saturated activities input to SAND-II were calculated. This comparison is displayed in Figure 15 by the percent error of the iterative solution to exact neutron spectrum as a function of neutron energy. The maximum width of the envelope of all test case solutions obtained using different initial trial functions, $\Delta\phi(E)_{\max}$, was compared with the average value of the test case solutions, $\phi(E)_{\text{ave}}$, as a function of neutron energy, E , as shown in Figure 16, where

$$\Delta\phi(E)_{\max} \equiv \phi(E)_{\max} - \phi(E)_{\min} = \text{maximum variation}$$

in value of the absolute neutron flux at energy, E , from all test case solutions. $\phi(E)_{\text{ave}} \equiv \sum_{i=1}^N \phi(E)_i / N =$

the average value of the absolute neutron flux at energy, E , from all test case solutions.

FIGURE 15. RELATIVE ERROR COMPARISON OF TEST CASE ITERATIVE SOLUTION SPECTRUM WITH THE EXACT NEUTRON SPECTRUM

FIGURE 16. SAND-II SOLUTION ENVELOPE WIDTH VARIATION WITH NEUTRON ENERGY

Neutron energy dependent SAND-II solution envelope widths are also shown in Figure 16 for the neutron spectrum obtained from multiple foil activation measurements in the FNC. The similarities in the variations of the SAND-II solution envelope width obtained from multiple foil activation measurements in the FNC and from the numerical test case are systematic. These variations are produced, in large degree, by the sensitivity of the SAND-II solution to the initial trial function. This systematic variation provides a means of correlating the solution envelope width and iterative solution error in the test case.

The test case error correlation was used to infer the error (root - mean - square deviation) in the measured FNC neutron spectrum, as shown in Figure 17. The error estimate for the measured FNC neutron spectrum assumes that the energy dependent SAND-II solution envelope widths in both the measured spectrum and the numerical test case are directly proportional to the root - mean - square error in the average spectrum solution, $\phi(E)_{ave}$,

$$\left[\begin{array}{c} \% \text{ Error (E)} \\ \text{FNC} \\ \text{Spectrum} \end{array} \right] = \frac{\left[\begin{array}{c} \% \text{ Error (E)} \\ \text{Numerical} \\ \text{Test Case} \end{array} \right] \left[\begin{array}{c} \Delta\phi(E)_{\text{max}} \\ \phi(E)_{\text{ave}} \end{array} \right]_{\text{FNC Spectrum}}}{\left[\begin{array}{c} \Delta\phi(E)_{\text{max}} \\ \phi(E)_{\text{ave}} \end{array} \right]_{\text{Numerical Test Case}}}$$

(4-6)

Root - mean - square error in the numerical test case solution (Figure 15), and the root - mean - square solution envelope widths (Figure 16) were averaged over each neutron energy decade to obtain the smoothed rms error variation, shown in Figure 17, using Equation (4-6). Similarities in the variations in the solution envelope widths suggest that this is a reasonable approach.

FIGURE 17. SAND-II WEIGHTED AVERAGE NEUTRON SPECTRUM SOLUTION ERROR VARIATION WITH NEUTRON ENERGY

II. Neutron Flux Spectrum Determination in the FNC

A. Iterative Neutron Flux Spectrum Solution From Measured Dilute Foil Saturated Activities

The neutron flux spectrum was unfolded from the infinitely dilute saturated activities for 26 foil reactions, listed in Table VI, using the SAND-II computer code.⁽²⁸⁾ Appropriate solutions which satisfy the measured activation integral equations simultaneously, within experimental uncertainties in the cross sections and activation measurements, were obtained. Initial trial functions for the neutron spectrum in the SAND-II analysis included: diffusion⁽³³⁾ and transport⁽³⁴⁾ theory code calculations; a constant $\phi(u)$; a Gaussian function centered at 100 keV; and a Gaussian function centered at 1 MeV. The Gaussian functions were one energy decade full width at half maximum. The best SAND-II representation for the neutron spectrum, shown in Figures 18 and 19, was obtained from a weighted average of the iterative neutron spectrum solutions resulting from different initial trial functions. A weighting factor proportional to the inverse variance in the measured to calculated activity ratios was used in the averaging procedure. The standard deviation in the multiple foil activation neutron spectrum was inferred from the width $\Delta\phi(E)_{\text{max}}/\phi(E)_{\text{ave}}$, of the envelope of all SAND-II solutions obtained from measured activities using different initial trial functions.

FIGURE 18. NEUTRON FLUX SPECTRUM IN THE PCTR FAST NEUTRON CAVITY

FIGURE 19. COMPARISON OF PROTON-RECOIL SPECTROMETER AND MULTIPLE FOIL ACTIVATION RESULTS

B. Comparison of the Multiple Foil Activation Results with Proton-Recoil Counter Measurements and Reactor Theory Calculations

An intercomparison between two experimental methods for determining the neutron spectrum was obtained using a proton recoil spectrometer and a multiple foil activation method.⁽³⁵⁾ These neutron spectrum studies made in the PCTR Fast Neutron Cavity show that information obtained from each method supplements the results of the other.

Proton recoil counter measurements utilizing both hydrogen gas and methane gas filled detectors were made by L. C. Davenport⁽³⁶⁾ following the method of E. F. Bennett.⁽³⁷⁾ The neutron spectrum obtained from the proton recoil counter data, shown in Figure 19, is the composite of several measurements at the same reactor flux level. Count rates during all measurements were kept below 920 counts/sec. The proton recoil spectrum was observed in consecutive overlapping energy intervals, obtained by varying the gas multiplication in each proportional counter. The methane detector spanned energies between 1.4 MeV and 160 keV in three intervals. Neutron energies between 335 keV and 9.6 keV were observed in six intervals using the hydrogen detector. Corrections for systematic errors in the recoil proton spectrum including wall and end effect, inhomogeneous electric field effect

(tip effect), and heavy nuclei recoil distortion were made in the data analysis. The fast neutron flux spectrum was unfolded by differentiating the proton recoil spectrum using computer program PSNS.⁽³⁸⁾ The spectrometer was calibrated using the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction which releases ionization equivalent to a proton of 615 keV when thermal neutrons are incident. Both detectors, containing 4.0% nitrogen, were calibrated in the PCTR thermal column⁽²³⁾ with the reactor at a 10 watt power level. The averaged composite neutron spectral results from the proton recoil measurements consist of inverse variance weighted data from the individual measurements within the neutron energy interval 1.4 MeV to 9.6 keV.

The neutron spectrum obtained from the proton-recoil counter measurements was integrated over the interval (9.32 keV to 1.17 MeV) of neutron energies spanned. The integral flux in this same neutron energy interval was obtained from the SAND-II analysis of multiple foil activation measurements. The root - mean - square deviation of the SAND-II integral flux values from all iterative solutions was less than $\pm 3\%$. The proton - recoil spectrum integral neutron flux was normalized to the multiple foil activation spectrum integral neutron flux value (in the interval 9.32 keV to 1.17 MeV) for comparison of spectral shapes in Figures 18 and 19.

The envelope of all SAND-II solutions exhibited poor spectral definition in the interval where the proton recoil counter results were obtained. The activation cross sections were small and similar in shape over this neutron energy interval. Activation integrals were relatively insensitive to the shape of the neutron spectrum the region of the flux peak. Large flux depressions in the SAND-II neutron spectrum at neutron energies of 7, 20, and 40 eV, shown in Figure 18, were the result of ^{238}U resonance absorption in the FNC buffer.

Comparison of a one dimensional diffusion theory PCTR Fast Neutron Cavity calculation⁽³³⁾ using the 26 group Russian cross section set⁽³⁹⁾ with the multiple foil activation spectral results, in Figure 18, shows good agreement at most neutron energies. The detailed spectral structure corresponding to ^{238}U absorption resonances, however, was not shown in the calculation. A comparison of the energy interval averaged microscopic ^{238}U absorption cross sections in the SAND-II and Russian cross section libraries, shown in Figure 20, illustrates the reason for absence of detailed structure in the calculation.

FIGURE 20. INTERVAL-AVERAGED MICROSCOPIC ^{238}U ABSORPTION CROSS SECTION

The Russian cross sections are averaged over intervals (.77 lethargy units per group) which are too broad to show detailed resonance structure. The apparent depression in the SAND-II spectrum at ~ 2 MeV is believed to be due to errors in the cross sections and/or the activation measurements for the $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ reactions.

C. Conclusions and Sources of Uncertainties

Energy degradation of fission spectrum neutrons into the few hundred keV energy region, as shown both by experimental results and calculations was the result of ^{238}U inelastic scattering in the uranium buffer zone. This 5.4-inch uranium buffer zone completely surrounded a 1/2-inch thick stainless steel box (29.0-inches by 29.0-inches by 24.7-inches inner dimensions), forming the large Fast Neutron Cavity. The uranium fuel in the FNC absorbed incident thermal neutrons and produced fast neutrons by the fissioning process such that a flux of predominantly fast neutrons entered the central cavity.

Neutron spectrum results from multiple foil activation measurements were obtained over the entire energy spectrum, 10^{-10} to 18 MeV. Detailed neutron spectrum structure was measured by proton recoil spectrometer methods in the energy interval, 9.6 keV to 1.4 MeV, comprising 80% of the integrated neutron flux. Thus, each of these two experimental methods supplemented the information obtained from the other. Using both methods permitted the shape and absolute value of the neutron flux spectrum to be obtained in the center of the FNC with the PCTR operating at a 100 Watt power level.

A trade off between spectral definition and the range of neutron energies covered was observed when comparing the two methods. The proton recoil spectrometer nominally spans neutron energies from a few keV to about 1 MeV. The range of recoil protons in the counter gas should be small, with respect to counter size, to prevent escape of protons from the effective counting region. At higher neutron energies, the high gas pressure and detector volume necessary to obtain sufficient stopping power is a limiting factor. At lower energies the energy expended per ion pair, W ,

increases rapidly and terminates detector response. The ability of the proton recoil proportional counter to resolve the energy distribution of the neutron flux was very good. Gaussian shaped neutron spectra resulting from incident monoenergetic neutrons had a full width at half maximum narrower than 15% of the neutron energy over the energy interval 1 MeV to 40 keV, increasing to about 23% at 10 keV.

The entire range of possible neutron energies was spanned by the multiple foil activation method. Careful selection of a set of reactions with different cross section shapes and spans of neutron energy responses was necessary to insure relatively unique spectrum solutions. The width of the envelope of neutron spectrum solutions, $\Delta\phi(E)_{\max}/\phi(E)_{\text{ave}}$, obtained using different initial flux shape approximations gave an indication of the relative uniqueness of the spectrum. Spectral definition from the activation method was generally poor in the range 1 keV to 1 MeV, where proton recoil measurements were obtained.

Although the proton recoil spectrometer will have gamma ray discrimination capabilities, gamma discrimination was not used in these initial experiments. Neutron spectrum information was obtained over a wider energy range than would be normally expected from a proton recoil spectrometer not employing gamma discrimination. This was possible because the Fast Neutron Cavity has an extremely low gamma field.

The supplementary nature of the two methods, illustrated with respect to spectral definition and span of neutron energy response, also includes absolute flux level normalization. The absolute integral flux in the span of neutron energies covered by the proton recoil spectrometer was determined from the activation method within 3% uncertainty (root - mean - square deviation) from all iterative SAND-II solutions. Normalization of the integral flux from the proton recoil spectrometer to the multiple foil activation results provided an absolute flux normalization and a direct comparison of the results from both methods.

Calibration of the absolute neutron flux spectrum^(m) in the Fast Neutron Cavity with the PCTR power level makes this facility a valuable tool for calibration of fast reactor instrumentation. The high neutron to gamma ray tissue equivalent absorbed dose ratio (~ 35 to 1) in the Fast Neutron Cavity provides a unique environment for the irradiation of relatively large objects (29-inch by 24.7-inch) in a uniform field of fast neutrons without gamma ray interference.

Sources of experimental error in the activation data included: uncertainties in determination of efficiency vs. gamma ray energy calibration curves, errors in decay scheme branching ratios, errors in corrections for activation self shielding, and statistical uncertainties in the counting data. Uncertainties for the measured infinitesimally dilute saturated activities (presented in Table VI) are standard deviations accumulated by the law of propagation of statistical errors. The average error (rms deviation) in this data is less than 3%. Uncertainties in the activation cross section data are much larger than uncertainties in activation measurements. Studies have been made on the accuracy of foil detector cross section data

^(m) The absolute neutron flux calibration is based on the activation cross section data in the SAND-II library.

by Liskien and Paulsen,⁽⁴⁰⁾ Barrall and McElroy,^(41,42) and others. A review of the more recent of these studies supports the conclusion that the integrated accuracy (defined as the root - mean - square error in the microscopic cross section integral, $\int_0^\infty \sigma(u)du$) of present evaluated cross section data in the SAND-II library is within an uncertainty of $\pm 10\%$ ⁽ⁿ⁾ for a number of foil detector reactions in current use. Errors in the shape and magnitude of energy dependent activation cross sections in the SAND-II library produce errors in the spectrum averaged cross section ratios, $\bar{\sigma}_m/\bar{\sigma}_n$, for the m^{th} and n^{th} reactions, where

$$\frac{\bar{\sigma}_m}{\bar{\sigma}_n} = \frac{\int_0^\infty \sigma_m(E)\phi(E)dE}{\int_0^\infty \sigma_n(E)\phi(E)dE} \quad (4-7)$$

$\sigma(E)$ = energy dependent microscopic activation cross section

$\phi(E)$ = energy dependent neutron flux in the FNC.

These neutron spectrum averaged cross section ratios are equal to the infinitely dilute saturated activity ratios as defined by Equation 3-20,

$$\frac{\bar{\sigma}_m}{\bar{\sigma}_n} = \frac{A_m^{\text{sat}}}{A_n^{\text{sat}}} \quad (4-8)$$

⁽ⁿ⁾ The error in cross section data is expressed in percent at one standard deviation.

Significant errors in the cross section data will produce inconsistencies between the measured activity ratios and cross section ratios averaged over the exact neutron spectrum.

Mutual inconsistencies in cross section data propagated highly oscillatory flux spectrum solutions and negative fluxes when a series expansion method was used to unfold activation integrals.⁽⁴³⁾ This ill conditioning was reduced by careful selection of detectors in an attempt to avoid pairs with similar cross sections. In contrast, the SAND-II algorithm used in this study, did not exhibit similar instability due to mutual inconsistencies in cross section data. Addition of detectors with similar cross sections aided the understanding of the results because the weighting factor for each reaction was reduced in the least squares fitting method. This reaction duplication had an identifying effect on inconsistent activation cross section data, and pointed out particular reactions requiring further cross section evaluation.

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APPENDIX A

Fuel Loading in the Reactor

1. FNC Fuel Loading

For this series of experiments, it was decided to load half the tubes of the FNC with uranium metal buffer fuel. The inner row of tubes in the radial buffer was filled with natural uranium metal tubular cores as shown in Figure 21. The next two rows of tubes were filled with 0.95 wt% enriched uranium metal tubular cores. Approximately 9400 pounds of uranium metal buffer fuel were loaded in the FNC. The row of tubes outside the 0.95 wt% enriched uranium contained alternate tubes of 26.5 wt% enriched UO_2 rods and Al 5.0 wt% Pu rods. The outer row of tubes in the radial buffer was loaded with Al 33.5 wt% uranium (93% enriched in ^{235}U) rods. The two outer rows of tubes are a transition zone between the PCTR "driver" fuel region and the uranium metal buffer region.⁽⁶⁾ In order to achieve criticality, it was necessary to insert hydrogenous moderator in the transition zone around the high fissile content fuel. Details of the radial buffer fuel loading are listed in Table VIII.

(6) The reason many types of high fissile content fuels were used in the transition zone is that the PCTR fuel inventory of any one type of fuel was quite limited.

FIGURE 21. FNC RADIAL BUFFER LOADING

TABLE VIII

Radial Buffer Fuel Loading

Fuel Type	Number	Description
UA1	96	<p>5/8-inch diameter x 28-inch long plexiglass rod 33.5 wt% U alloy (93% enriched in ^{235}U); Rods 0.30-inch diameter = 59.6 gm ^{235}U Plexiglass tube 5/8-inch diameter x 28-inch long, 0.030-inch wall thickness 1/4-inch diameter x 28-inch long plexiglass rod</p>
PuAl	50	<p>5/8-inch diameter x 28-inch long plexiglass rod. Al-5.0 wt% Pu (8% ^{240}Pu) rods 24-inch long; 1/2-inch fuel diameter = 33.0 gm Pu clad with Zircalloy-2, 0.565-inch O.D.</p>
UO ₂	50	<p>3/4-inch diameter x 28-inch long plexiglass rod 26.5% enriched UO₂ rod 20-inch long; 0.548-inch fuel diameter = 180 gm. ^{235}U clad with Inconel 0.625-inch O.D.</p>
E	92	<p>0.947% enriched U metal core; 1.420-inch O.D. 0.545-inch I.D. 0.916 lb Uranium/inch 28.2-inches long</p>
N	84	<p>Natural uranium metal cores; 1.432-inch O.D. 0.533-inch I.D. 0.944 lb Uranium/inch 25.0-inch long</p>
N	8	<p>Natural uranium metal core 0.50-inch diameter x 28-inch long, aluminum clad, 0.60-inch O.D.</p>

Radial buffer tubes are 1.5-inch O.D., 0.028-inch wall thickness of 6061T6 aluminum, 28.3-inches long.

The loading of the FNC end buffers (shown in Figure 22) was similar to the loading in the radial buffer in order to preserve symmetry. The inner row of tubes was left empty. This allows the volume inside the uranium metal buffer to be nearly a cube, 30-inches on a side. The second row of tubes from the inside contained natural uranium metal tubular cores; and the third row of tubes contained 0.95 wt% enriched uranium metal tubular cores. The outer row of tubes in the end buffers contained UO_2 - PuO_2 rods and homogeneous moderator. This transition zone on the outside of the end buffers helped preserve symmetry with the radial buffer and increased the fissile fuel loading near the region of the flux leveling fuel. Details of the fuel loading in the FNC end buffers are listed in Table IX.

The FNC rear buffer had a 1.0-inch diameter aluminum tube installed through the rear of the reactor to allow immediate removal of a packet of activation foils after irradiation. A rod of 0.8% enriched uranium metal 0.867-inch diameter by 4.050-inches long was used to plug the hole in the rear buffer, as shown in Figure 22. The foil packet was contained in an aluminum capsule connected to the removable uranium rod, as shown in Figure 8.

FIGURE 22. FNC END BUFFER LOADING

TABLE IX

FNC End Buffer Loading Details

<u>Fuel Type</u>	<u>Number</u>	<u>Description</u>
52		0.95 wt% enriched uranium metal core (36.0-inch long) 1.420-inch O.D. 0.545-inch I.D. 0.916 pounds uranium/inch
54		Natural uranium metal core (39.3-inches long) 1.432-inch O.D. 0.533-inch I.D. 0.944 pounds uranium/inch
46		4 rods 36-inches long UO ₂ - 2.0 wt% PuO ₂ (8% ²⁴⁰ Pu) 2 0.500-inch diameter fuel; 0.565-inch Zr clad O.D. 5 rods 18-inch long lucite, 0.25-inch diameter
7		<u>Located on columns aligned with</u> <u>removable door</u> 12 rods 8-inches long UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu) 0.500-inch diameter fuel; 0.575-inch Zr clad O.D. 10 rods 12-inches long and 5 rods 10-inches long lucite 0.25-inch diameter
1		<u>Column aligned with foil removal</u> <u>tube</u> 8 rods 20-inches long UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu) 0.500-inches diameter fuel; 0.565-inch Zr clad O.D. 10 rods 18-inches long lucite 0.25-inches diameter

2. PCTR Driver Fuel Loading

Since there were only 116 driver rod holes in the graphite when the FNC was installed (see Figure 23), it was necessary to add hydrogenous moderating material to the driver fuel to increase its effectiveness. It was possible to double the reactivity worth of a tubular uranium aluminum alloy driver rod by the addition of a 3/4-inch diameter by 36-inch long Plexiglass rod in the center of the fuel and by wrapping 157.5 grams of polyethylene sheet around the outer surface of the driver.

The hollow lead uranium oxide drivers have been designed to provide an inherent shutdown mechanism for the PCTR. In a power excursion, the lead in the fuel mixture will melt and the slugs assume the shape of collapsed cylinders. In this geometry, the self shielding of the uranium is greater than it is in the cylindrical slug configuration, and therefore, the reactor will lose some reactivity when the fuel melts. Plexiglass rods were not inserted in the center of twenty of these meltable fuel rods because their presence might interfere with this safety feature.

FIGURE 23

PCTR Critical Loading with FNC Installed

Additional driver fuel of solid uranium aluminum alloy rods were loaded into the PCTR graphite driver hole locations until criticality was achieved. A diagram of the critical fuel loading is shown in Figure 24, and a description of the fuel types listed on the diagram is given in Table X. With the cadmium shutter to the thermal column closed and the inner and middle leveling ring positioned 3.0-inches from the FNC graphite interface, criticality was achieved with all of the 116 available driver hole locations loaded with fuel. Calculations have shown that when a test core is inserted in the FNC, the amount of fuel loaded in the thermal region of the PCTR at criticality is decreased because the fraction of total fissions in the fast region is increased.⁽⁸⁾ Thus, sufficient fuel can be loaded in the PCTR driver region to achieve criticality in the planned experiments in the FNC.

FIGURE 24. PCTR DRIVER FUEL LOADED IN GRAPHITE, 2" THICK PLEXIGLASS SHEETS ON THE SIDES OF THE PCTR

TABLE X

Driver Fuel Loaded in PCTR Graphite

<u>Fuel Type</u>	<u>Number</u>	
(X)	20	Pb-UO ₂ (93% enriched) PCTR driver = 147.6 gm. ²³⁵ U wrapped with 157.5 gm. Polyethylene sheet.
(XL)	32	Al 93% enriched U alloy PCTR driver = 149.7 gm. ²³⁵ U wrapped with 157.5 gm. Polyethylene sheet 3/4-inch diameter by 36-inch long Plexiglass rod in center of driver.
(J)	43	Al 7 wt% U alloy (93% enriched in ²³⁵ U) rod 1.36-inch diameter containing 153.2 gm. ²³⁵ U, wrapped with 78.75 gm. Polyethylene sheet.
(JC)	6	Al 7 wt% U alloy (93% enriched in ²³⁵ U) rod 1.36-inch diameter containing 131.3 gm. ²³⁵ U, wrapped with 78.75 gm. Polyethylene sheet.
(UA1)	15	<p>Al 33.5 wt% alloy (93% enriched in ²³⁵U) rods, 0.30-inch diameter = 117.34 gm. ²³⁵U.</p> <p>Plexiglass tube 5/8-inch diameter by 37-inch long, 0.030-inch wall thickness.</p> <p>Plexiglass rods (3 ea.) 1/4-inch diameter by 36-inch long.</p> <p>157.5 gm. Polyethylene sheet wrapped around bundle.</p>

Total of 116 driver rod holes loaded.

3. Critical Approach Procedure

The fuel loaded in the PCTR to achieve criticality included approximately 23 kg of ^{235}U in the graphite moderated "driver" region and 16 kg of ^{235}U plus 4 kg of ^{239}Pu in the transition zone between the "driver" region and the uranium metal buffer region. This large quantity of fissile fuel was required for production of sufficient neutrons to compensate for the neutrons absorbed in the uranium metal buffer. Since it was possible to increase reactivity by selective unloading of fuel from such a reactor configuration, care was taken to insure that conservative step wise fuel additions were made in compliance with the PCTR Operating Specifications.

The critical approach was initiated by the addition of 20 meltable lead uranium oxide driver fuel rods while the central test cavity was empty. The next step was the addition of a 3/4-inch diameter by 4-inch long plexiglass rod in the center of each flux leveling fuel tube in both PCTR end reflectors. The rear buffer of the FNC was assembled in the empty test cavity in the next three fuel loading steps. The lower fourth of the FNC radial buffer was inserted in the test cavity

in three fuel loading steps before the stainless steel FNC liner was installed. The remaining FNC radial buffer fuel was loaded in the next nine fuel loading steps. Completion of the FNC fuel loading was accomplished in the next three fuel loading steps as the front buffer was added. Fuel loading in the FNC at various stages is shown in Figure 25. Subsequent fuel loading steps to achieve criticality consisted of the addition of highly enriched uranium in aluminum fuel in the graphite moderated PCTR driver region as prescribed in the PCTR Operating Specifications.

After each fuel addition step, the reactor room was cleared and closed, the movable reactor face was closed, and subcritical multiplication measurements were made. The control rods were opened, the subcritical multiplication was measured and the reactor face was reopened. Plots of inverse multiplication versus the amount of fuel in the reactor were made for both control rod configurations (opened and closed) to monitor the control worth. Each fuel loading step took approximately one hour.

a. FNC Rear Buffer Installed

b. Lower Quarter of Radial Buffer and the Stainless Steel FNC Liner Installed

c. Completion of Radial Buffer Installation

d. Lower Two-Thirds of Front Buffer and the Removable Stepped Door Installed

FIGURE 25. Fuel Loading in the FNC at Various Stages

APPENDIX B

Criticality Safety Measurements

Since an unusually large quantity of fissile fuel was loaded in the PCTR to achieve criticality when the FNC was installed in the central test cavity, a number of criticality safety measurements were made to confirm operational safety. This data was also obtained as a benchmark to predict the operational safety of future PCTR experiments with fast reactor test cores installed in the FNC. Operation of the PCTR with the FNC installed was accomplished within the operating specifications.

1. Reactor Control System

Control-safety rods and safety disks provide rapid reactivity control for reactor shutdown conditions. The control-safety rods provide fine control of the reactor during startup and operation. Opening of the movable face provides a large reactivity loss.

The eight control rods run the length of the test cavity parallel to its axis, spaced symmetrically on an 80-centimeter radius from the axis. The rods are of shutter type and consist of a fixed outer tube and a sliding inner tube. The outer tube is 1.5-inches in diameter and has alternate bands of aluminum and cadmium with each being 4.166-inches long. The inner tube consists of bands of cadmium and ^{235}U aluminum alloy pieces with each being 4.166-inches long. A rod

of unusually uniform worth per inch results because the change from an effectively solid cadmium rod to one of alternate bands of cadmium and ^{235}U introduces a fission source which is closely proportional to the amount of ^{235}U exposed. The worth of each control rod was measured after criticality was achieved with the FNC installed. The total reactivity worth of the eight control rods was 2.92\$.

A slower but stronger safety system consists of the cadmium aluminum assemblies that fall vertically, under the force of gravity, in aluminum thimbles located just outside the driver rods. These circular safety disks are made of layers of 1/8-inch aluminum, 0.022-inch cadmium and 1/8-inch aluminum. The two safety disks provided are 21.5-inches in diameter, and the sandwich is surrounded by a steel ring with a 24-inch O.D. During normal operation, they are held out of the reactor, but still in their respective thimbles, by electromagnets. The safety disk strength was measured after criticality was achieved with the FNC installed. The total reactivity worth of the two safety disks was 3.21\$.

2. Reactor Operational Safety Testing

When the control rods or safety disks scram the reactor due to the interruption of the safety circuit, power is applied to the motor driven carriage to open the movable face. Source multiplication measurements were made with the reactor slightly subcritical and the face initially closed to determine the reactivity worth of the face as a function of distance from the closed position. Increasing the face distance from the closed position provided a corresponding increase in the reactivity loss. When the movable face was moved back to 6 feet from the closed position, the reactivity loss was $\sim 10\%$.

The flux leveling slugs were moved to find the optimum position for maximum reactivity. As the inner and middle leveling rings were moved from 0.0-inches (at the graphite - FNC interface) to 7.5-inches, the reactivity increased; further movement outward decreased reactivity. Movement of the outer leveling rings from 0.0-inches outward decreased reactivity. The optimum leveling ring positions with the FNC installed are: inner and middle leveling rings 7.5-inches and outer leveling rings at 0.0-inches. The increase in reactivity due to removal of the cadmium shutter below the thermal column was $\sim 60\phi$.

The effects of changes in ambient temperature and pressure on the reactivity of the PCTR were measured with the FNC installed. The pressure reactivity coefficient was measured by pressurizing the reactor room with compressed air and measuring the change in the overall reactor excess reactivity as a function of the reactor room pressure. The reactor temperature coefficient was measured by heating and cooling the reactor with the reactor room lights and air conditioner, respectively. The reactivity coefficients due to temperature and pressure changes were $-0.49\phi/\text{degree centigrade}$, and $-0.021\phi/\text{millibar}$, respectively.

Reproducibility of reactor measurements was about the same as that experienced with thermal test cores inserted in the PCTR previously. When the movable face was opened and reclosed for insertion or removal of a sample from the FNC, the nominal uncertainty in a single measurement was $\pm 0.04\phi$. If the face was locked closed, and the graphite was removed from the movable face to allow removal of the stepped FNC door through the face port, the nominal uncertainty after reassembly was $\pm 0.02\phi$. Removal of the graphite blocks and the nine leveling slugs in the face port caused a loss of reactivity. Subsequent removal of the FNC stepped door further reduced the reactivity. Small samples of fast reactor fuel can safely be inserted or removed through this face port within the Operating Specifications.

APPENDIX C

PCTR/FNC Computational Model

A one dimensional spherical calculational model of the PCTR was developed as an aid in understanding the spectral buffering requirements in the FNC. One dimensional diffusion theory code, 1 DX,⁽³³⁾ was used for generating effective nuclear cross sections using data in the 26 group Russian format. These cross sections were input to the S_n multigroup transport theory program, DTF-IV,⁽³⁴⁾ using the P_0 scattering approximation and S_n order 4. Equivalent concentric spherical radii were obtained from the actual concentric cubic dimensions (Figure 5) using the geometric buckling equivalence relation:

$$R = \left(\frac{\sqrt{3}}{3} \right) S$$

where R is the radius of a sphere, and S is the side of a cube.

The thermal to fast neutron spectrum conversion was accomplished without significant reduction of the integral flux in the central cavity, when compared to the integral flux in the driver region, as shown in Figure 26. A large depression of the integral flux in the central cavity would reduce the sensitivity of a fast-thermal coupled reactor to small sample reactivity perturbations, and decrease the precision of fast reactor experiments in the central zone. Calculations show that such a depression would occur if an absorber material were used instead of fissionable material for the neutron spectrum buffer.

FIGURE 26 Calculated Region Averaged Neutron Spectra in the PCTR/FNC Critical Facility

The calculated spatial variation of the neutron spectrum when the FNC was empty is shown in Figure 26. The distribution of fissions calculated for the empty FNC was 70.4% in the driver zone, 20.1% in the transition zone, and 9.5% in the uranium buffer. When a UAL fast core containing 13.3 kg ^{235}U was installed in the center of the FNC (see Figure 27), the distribution of fissions in the driver zone (69.8%), the transition zone (19.9%), the uranium buffer (9.6%), and the fast core (0.7%), showed little change. The total reactivity worth of the eight PCTR control rods, measured in this critical loading, was 2.92\$. These results indicate that fast reactor cores large enough to equilibrate the neutron spectrum in the central region of the fast zone can be operated in the FNC without reducing the total control rod worth below 2.0\$ minimum required by the PCTR Operating Specifications.

FIGURE 27 Aluminum-30 wt% Uranium (93% ^{235}U) Fast Core
Installed in the Center of the FNC (Shown with
the FNC Front Buffer Removed).

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