

IN-SITU VERIFICATION TECHNIQUES FOR FAST CRITICAL ASSEMBLY CORES

by

S.B. Brumbach, P.I. Amundson, and C.T. Roche

MASTER**NOTICE**

EYES ONLY

PORTIONS OF THIS REPORT ARE ILLUSIBLE. It
has been reproduced from the best available
copy to permit the broadest possible avail-

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Prepared for

Topical Conference

on

Measurement Technology for Safeguards and Materials Control

American Nuclear Society

Kiawah Island, South Carolina

November 26-30, 1979

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

EB



U of C-AUA-DSDOE

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

Operated under Contract W-31-109-Eng-38 for the
U. S. DEPARTMENT OF ENERGY

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

The facilities of Argonne National Laboratory are owned by the United States Government. Under the terms of a contract (W-31-109-Eng-38) among the U. S. Department of Energy, Argonne Universities Association and The University of Chicago, the University employs the staff and operates the Laboratory in accordance with policies and programs formulated, approved and reviewed by the Association.

MEMBERS OF ARGONNE UNIVERSITIES ASSOCIATION

The University of Arizona
Carnegie-Mellon University
Case Western Reserve University
The University of Chicago
University of Cincinnati
Illinois Institute of Technology
University of Illinois
Indiana University
The University of Iowa
Iowa State University

The University of Kansas
Kansas State University
Loyola University of Chicago
Marquette University
The University of Michigan
Michigan State University
University of Minnesota
University of Missouri
Northwestern University
University of Notre Dame

The Ohio State University
Ohio University
The Pennsylvania State University
Purdue University
Saint Louis University
Southern Illinois University
The University of Texas at Austin
Washington University
Wayne State University
The University of Wisconsin-Madison

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States nor any agency thereof, nor any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use or the results of such use of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights. Mention of commercial products, their manufacturers, or their suppliers in this publication does not imply or connote approval or disapproval of the product by Argonne National Laboratory or the United States Government.

S.B. Brumbach and P.I. Amundson
Argonne National Laboratory, Idaho Falls, Idaho

C.T. Roche
Argonne National Laboratory, Argonne, Illinois

ABSTRACT

Active and passive autoradiographic techniques were used to obtain piece counts of fuel plates in fast critical assembly drawers and to verify the assembly loading pattern. Active autoradiography using prompt-fission and fission-product radiation was more successful with uranium fuel while passive autoradiography was more successful with plutonium fuel. A source multiplication technique was used to measure changes in reactivity when small quantities (2-2.5 kg) of fissile material were removed from a subcritical reference core of the Zero Power Plutonium Reactor. Efforts to compensate for the loss of reactivity by substituting polyethylene for fuel were largely unsuccessful. Some compensation was achieved by replacing U-238 with polyethylene. The sensitivity for detection of partially compensated fuel removed from minimum worth regions was approximately 2.5 kg (fissile) for a core containing 2600 kg (fissile). Substitution of polyethylene was detected with a spectral index which was the ratio of the rate of the In-115 (n, γ) reaction to the rate of the In-115 (n, n') reaction. This spectral index was sensitive to the presence of an 0.64-cm-thick, 5.08-cm-high polyethylene column 10-15 cm away from the indium foil. The reactivity worth of Pu-239 was also obtained as a function of location in the reactor core with the use of an inverse kinetics technique. Reactivity worths for Pu-239 varied from a maximum of 58.67 $\Delta k/k$ near the core center to a minimum of 14.86 $\Delta k/k$ at the core edge.

KEYWORDS: Nondestructive assay, plutonium, uranium, reactivity, autoradiography, spectral index, fast critical assemblies.

INTRODUCTION

Fast critical assemblies such as the Zero Power Plutonium Reactor (ZPPR) facility require large inventories of plutonium and enriched-uranium fuels. The ZPPR fuel is primarily in the form of thin metal or alloy plates (0.066-0.64-cm-thick, 5.08-cm high, and from 2.54 cm to 20.3 cm long) and small metal-oxide-containing rods (0.95 cm diam by 7.62-15.24 cm long). Uranium enrichments vary between 16% and 93% U-235, and plutonium concentrations vary from 15% to 99%. Fissile materials in these sizes and purities present an attractive target to a potential diverter. Because of this diversion potential, it is important that effective safeguards be implemented at fast critical assembly facilities. The subject of critical facility safeguards has been recently reviewed.^{1,2} Fuel elements at ZPPR are normally located in the reactor itself or are stored in canisters in a vault. Most of the time, a large majority of the fuel is in the reactor. The reactor has two halves, each of which consists of a square array of matrix tubes. The fuel is contained in drawers, 5.08 cm square by up to 91.4-cm long. The drawers also contain structural, coolant, and fertile materials. A typical drawer includes a core segment containing either one or two columns of fuel plates (~ 0.5 kg or 1.0 kg of fissile material) and a second segment containing typical axial blanket materials. The drawers are inserted into the matrix tubes, and the halves are brought together to form the final reactor configuration.

Because of the large number of fuel plates, and because the plates are normally located in the reactor, verification of the fissile material inventory of the facility is time-consuming. If individual plates, or even whole drawers are removed from the reactor for

verification, the efficiency of the experimental program will be adversely affected. A complete physical inventory at ZPPR, including visual inspection of each fuel element and nondestructive assay (NDA) measurements on 5% of the inventory, requires about 30 eight-hour shifts. Because of accumulated radiation exposure, individual participation is limited to 5 shifts. As a result, 36 persons are required to perform the inventory.

In order to reduce the impact of inventory verification, it is desirable to assay the fuel in the reactor core in the largest possible units. This is especially important during the long periods between major loading changes when fuel is not accessible to assay in small units. The most efficient inventory verification method would be able to examine the entire reactor core in a single measurement or combination of measurements. Ideally, the validity of the whole-core measurements should be verifiable by inspectors not normally part of the fast critical facility. This is especially important in international safeguards where institutional diversion is considered. Two potentially powerful assay methods, when used in combination, are autoradiography and the measurement of integral (whole core) reactivity.

EXPERIMENTAL METHODS

Autoradiography

When x-ray sensitive film is placed against the edge of a fast critical assembly fuel plate, the radiation emitted by the isotopes in that fuel plate form a clear image (autoradiograph) of the edge of the fuel plate. Two types of autoradiography can be used to obtain piece counts of fuel elements in reactor drawers. The first type is passive autoradiography in which spontaneously-emitted beta and gamma radiation form the fuel-plate image. The passive autoradiographic method was previously applied to plutonium-containing fast critical assembly fuels³ and to low-enriched uranium fuel in LWR assemblies⁴. The second type of autoradiography is active, in which prompt fission gamma radiation and fission-product beta and gamma radiation produced during and immediately after critical reactor operation form the fuel-plate image.

Passive autoradiographs of fuel plates in reactor drawers can be obtained by sliding a packet of x-ray film between the top of the contents of a drawer and the matrix element above that drawer. The clearance between the top of a fuel element and the matrix tube is 0.152 cm in the ANL fast critical assemblies. The film used in previous applications to plutonium fuel was Kodak Ready-Pack Type-M X-ray film. The film packet was 3.81 cm wide and was 0.089 cm thick where the ends of the packet were sealed with black tape. Typical exposure times were 15 min. The passive autoradiographic method allowed a fuel-plate piece count to be made in a large number of drawers in a short time. For plutonium fuels, passive autoradiographic images are predominantly formed by the 60 keV gamma ray from Am-241.

The passive autoradiography of uranium fuel in reactor drawers is less satisfactory than for plutonium fuels. One difficulty is that exposure times of many hours are required if ordinary x-ray film is used for uranium fuel. This problem is caused by the low gamma-ray emission rate for U-235 compared to the gamma-ray emission rates for the isotopes present in plutonium fuels. Intensifying screens cannot be used in reactor drawers because of their thickness. A second problem with the passive autoradiography of uranium is the large contribution from U-238 to the image density of a fuel plate. The U-238 daughter Pa-234 emits 2.29 MeV beta radiation which efficiently exposes film. The image densities from unclad depleted uranium plates and from unclad 93%-enriched plates are nearly equal. Uranium fuel plates used at ANL are not clad. Additional filtration is needed to reduce the contribution to image density from U-238, but additional filtration increases the thickness of the film packet. Further work is required to investigate possible methods for applying passive autoradiography to uranium fuel in reactor drawers.

The need to distinguish between U-235 plates and U-238 plates in an assembly core led to the development of active autoradiographic methods. In active autoradiography the prompt-fission gamma radiation and/or the short-lived fission-product beta and gamma radiation produces the fuel plate image. The formation of an image is the direct result of the critical operation of the assembly. Because the spontaneous radiation emission from uranium is small, two types of active autoradiography are applicable to uranium fuel. In the first type, the fuel plate images are formed by both prompt-fission and fission-product radiation. Operationally, there are three steps: (1) film is inserted into drawers, (2) the reactor

is brought to a critical configuration and operated at an appropriate power, and the film is removed and processed after shutdown and cooling. Relatively insensitive film is used in this first type of active autoradiography. In the second active autoradiographic method the fuel plate images are produced only by fission-product radiation. The steps are: (1) the reactor is operated at high power without film, (2) the reactor is shut down and allowed to cool over night, (3) the following day film is inserted into drawers and exposed for a few hours, and (4) the film is removed and processed. In this second method, more sensitive film is used. Active autoradiography is somewhat more complicated for plutonium than uranium because of the large spontaneous radiation component from plutonium fuel. Only at-critical active autoradiography is practical for plutonium.

Integral Reactivity Measurements

Reactivity Measurement Methods

A variety of reactivity measurement methods has been developed which are applicable to fast critical assemblies.^{5,6,7} The two basic types of methods are static (equilibrium) and dynamic (kinetic). Equilibrium methods are conceptually simple because details of the delayed-neutron behavior or the prompt-neutron lifetimes are not needed. One equilibrium technique in common use with subcritical systems is the source-multiplication method.^{6,8} For a plutonium-fueled reactor, the spontaneous-fission neutrons from the even plutonium isotopes provide a neutron source of known strength. The reactivity of the assembly can be deduced from the extent to which the fissile isotopes in the assembly multiply the spontaneous-neutron flux. The neutron flux can be monitored by detectors within the core or outside the core.

Dynamic techniques are also available which infer reactivity or changes in reactivity from the time-dependent changes in neutron flux. Knowledge is required of the delayed-neutron parameters and the prompt-neutron lifetime. Dynamic measurements are often made by measuring the change in neutron flux during and after the movement of a control rod or of other material within the reactor. Perhaps the conceptually simplest dynamic technique is the determination of reactivity from the measurement of the reactor period.⁵ For a supercritical reactor, the reactivity can be inferred from just the period and the prompt-neutron lifetime. Period measurements, however, suffer from many of the uncertainties and long measurement times encountered in the equilibrium methods. At ZPPR the more commonly used dynamic method involves perturbation techniques in which inverse reactor kinetics equations are used to derive values of time-dependent reactivity from measured time-dependent neutron fluxes.

Reference Configuration

The core configuration of a fast critical assembly may be designed to model a postulated reactor design or to address a generic reactor physics issue. For either type of experiment, it is useful to establish a reference core configuration for which various reactor physics parameters, including reactivity, are well characterized. As experiments progress, many modifications are made to this configuration. These modifications are very frequent, but often involve only a small fraction of the total number of drawers in the core. One way to verify the fissile content of the core is to periodically return to the reference configuration and experimentally verify that the reactivity is the same (within uncertainty limits) as it was when the reference core was initially established. When possible, such a reactivity verification could be scheduled to coincide with a time during the experimental program when the core was close to the reference configuration. The use of reference reactivity values from a reference configuration assumes that the initial reference core loading was independently verified. This initial verification can be accomplished by making NDA measurements on fuel elements or drawers as they are being loaded, by verifying the loading pattern with autoradiography, or by calculation of the reactivity expected for a particular core configuration and composition.

Reactivity Worth

The effectiveness of reactivity measurements as a diversion detection technique will depend upon the sensitivity with which these measurements can detect missing material. The factors influencing the diversion sensitivity are the uncertainty of the reactivity measurement and the reactivity worth of the fuel in the assembly. The reactivity worth of a

material is that reactivity effect, per unit mass (e.g., 1h/kg), produced by the addition of that material to the core. Reactivity worth can be either positive or negative, and both the magnitude and the sign of reactivity worth can vary as a function of position within the core.

Reactivity Compensation

The agreement between two measured reactivity values for a core in a reference configuration still does not necessarily mean that the fissile content has not changed in the time between measurements. The loss of reactivity by removal of fissile material can, in principle, be compensated for by addition of materials which also have a positive reactivity worth. Similarly, compensation might be accomplished by removing materials, such as absorbers, which have negative reactivity worth. A third compensation scheme could involve removal of fissile material from a region of relatively low worth with a simultaneous transfer of fissile material from a region of relatively low worth to a region of relatively high worth. This third scheme would, of course, constitute a deviation from the reference configuration.

EXPERIMENTAL RESULTS

Autoradiography

The ZPR-9 fast critical assembly was used to test the applicability of active autoradiography to uranium-fueled cores. The fuel drawers of the assembly contained a single column of 93%-enriched fuel plates. Each plate was 5.08 cm wide, 0.16 cm thick, and from 2.54 to 7.62 cm long. On either side of the uranium plate was a thin steel plate, and the remaining drawer volume was occupied by thick steel plates. The drawers contained only steel and uranium.

An example of an active autoradiograph produced by fission product radiation is shown in Fig. 1A. This autoradiograph was obtained approximately 18 hours after reactor shutdown. The reactor had been operated at a power of 255 watts for 66 min. Film exposure time was 66 min. The film used was the relatively sensitive Kodak AA Industrex X-ray film. The fuel column was readily visible. The individual fuel plates could not be distinguished because they were unclad and were fit tightly together in the column. Figure 1B shows a passive autoradiograph of an identical fuel column obtained with Kodak AA Industrex film and a 75-min exposure. The uranium plates in Fig. 1B had not been irradiated in the reactor for several weeks.

An example of an active autoradiograph obtained during a critical operation of the reactor is shown in Fig. 2. The film used was the relatively insensitive 3M QA-V graphic arts-type film. Sheets of film were cut into strips approximately 3.81 cm wide and were contained in Kodak Ready-Pack film holders. The autoradiograph of the fuel column segment in Fig. 2 was obtained during a 15-min reactor operation at a power of 85 watts followed by a three-hour cooling period. The film was in the reactor for a total of approximately four hours. The contribution to image density from spontaneous radiation during the four-hour period was very small. The 0.16-cm-thick fuel column can be clearly seen as the darkest part of the line in the center of Fig. 2.

The sensitivity of the active, at-critical autoradiographic technique to some simple drawer-loading defects was tested. Figure 3A shows an active autoradiograph of part of a fuel column from which a 7.62-cm-long, 93%-enriched uranium plate was removed. Figure 3B shows an active autoradiograph of part of a fuel column in which a 7.62-cm-long, 93%-enriched fuel plate was replaced by a 20%-enriched fuel plate. Both defects in Fig. 3 were easily detectable. In another case, a 0.080-cm-thick fuel plate was substituted for a 0.61-cm-thick plate. The presence of the thin plate was detectable, but only upon careful inspection of the autoradiograph. Smaller differences in plate thickness would probably not be detectable.

The film base and film packaging materials are hydrogenous, and in the ZPR-9 core described here, the film had a positive reactivity effect. A reactivity worth of 103 1h/kg was measured for Kodak 35 mm Ready Pack X-ray film randomly distributed through a quadrant of one-half of the ZPR-9 assembly. This reactivity worth meant that only approximately 30 pieces of film, each 102-cm long, could be placed in the 800-drawer assembly without violating safety regulations. More pieces of film could be added, but the assembly would be

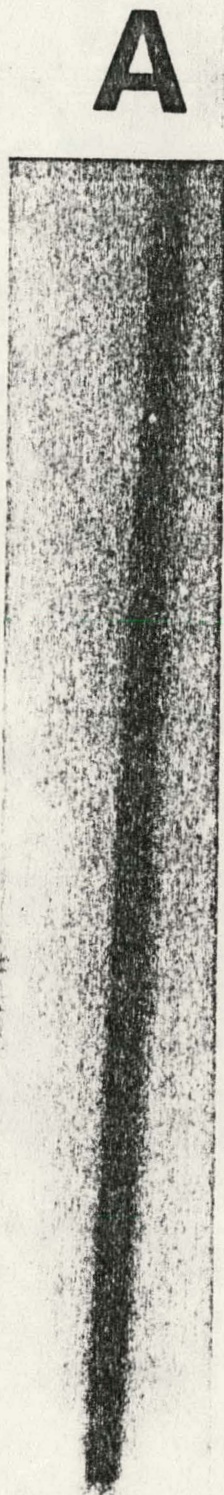


FIG. 1. A: Post-irradiation Active Autoradiograph of a Highly-enriched-uranium fuel column
B: Passive Autoradiograph of Highly-enriched-uranium Fuel Column



Fig. 2. At-critical Active Autoradiograph of
a Highly-enriched-uranium Fuel Column

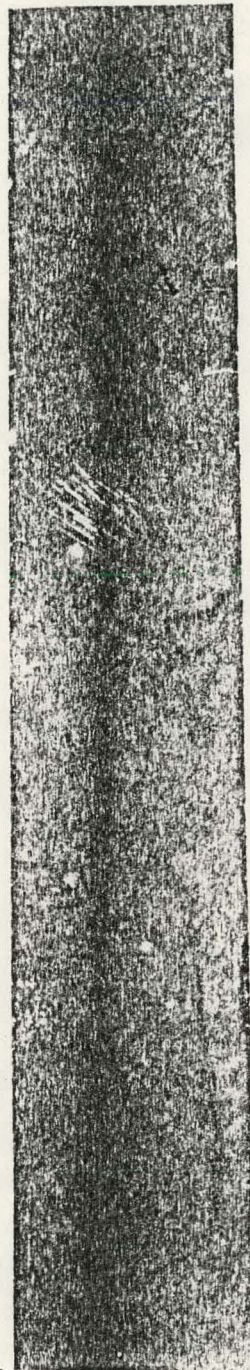


Fig. 3. At-critical Active Autoradiographs of 93%-enriched Uranium Fuel Columns With Defects
A: Missing Plate
B: 20% Enriched Plate

would have to be decreased by removing fuel drawers. In order to insure safe reactor operation, the reactivity effect of the film must always be considered when doing an at-critical autoradiographic verification.

The success of the active autoradiographic technique for uranium fuel in an assembly core led to a test of the applicability of the active technique to a plutonium-fueled assembly core. An example of an active autoradiograph obtained in the 6100-liter ZPPR assembly 10D is shown in Fig. 4A. The autoradiograph was produced during a 100 watt-hour operation of the reactor. The film was in the reactor core for 5.5 hours. This active autoradiograph shows portions of the edges of two 0.64-cm-thick plates in the center. The image at the right edge of the autoradiograph and parallel to the fuel plates corresponds to a column of U-238 plates. The space between the U-238 column and the fuel-plate column was occupied by a can containing sodium. Figure 4B shows a passive autoradiograph produced in a 5.5-hour exposure when the assembly halves were separated. Both autoradiographs were obtained with 3M QA-V film. The film was processed by hand at room temperature in Kodak X-Omat chemicals.

The 5.5 hours the film spent in the assembly core was greater than the minimum time necessary for autoradiography because other experiments were also conducted. The minimum time for an active autoradiographic irradiation is approximately 2.5 hours, which includes reactor startup, irradiation, and reactor cooling.

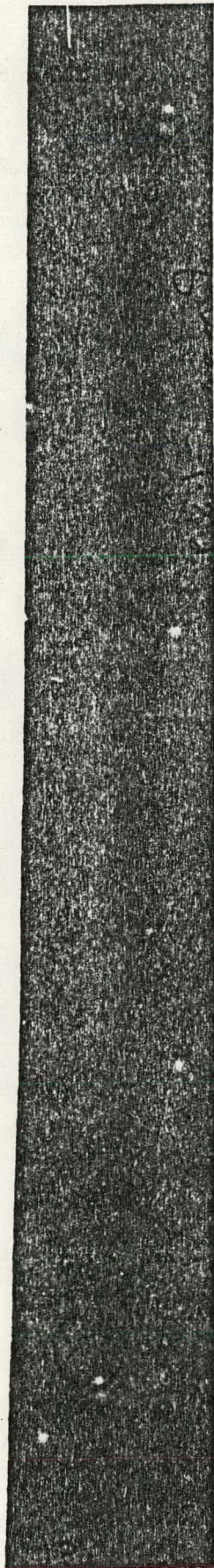
The fuel-column image is more dense in the active autoradiograph (Fig. 4A; 1.2 optical density units) than in the passive autoradiograph (Fig. 4B; 0.6 optical density units). However, the background density is also higher in the active autoradiograph. The density difference between active and passive fuel plate images may have been greater if the total time the film was in the core had been the minimum 2.5 hours, instead of the actual 5.5 hours.

The plutonium fuel-plate images in the active autoradiographs obtained in the assembly drawers had only very small contributions from the fuel columns in adjacent drawers. Active autoradiographs were obtained in drawers which contained a single fuel column and which also had drawers above containing a single fuel column. The two fuel columns were separated by 0.28 cm of steel in the two matrix tubes and in the bottom of one drawer. Active autoradiographs were also obtained in drawers which contained a single fuel column, but which had no fuel-containing drawer above. The density of the fuel column images were nearly identical in both cases. From this observation of nearly equal image densities, it was concluded that the 0.28-cm-thick steel separating the two fuel columns absorbed most of the radiation which produced the fuel column images. This, in turn, led to the conclusion that low-energy radiation produced the images. This final conclusion is not surprising, because low-energy photons dominate both the prompt-fission gamma ray spectrum⁹ and the fission-product spectrum¹⁰ even though some very high energy photons are produced. In addition, the beta radiation from fission products will be highly absorbed by the 0.28-cm-thick steel, but will still be able to expose film within the drawer.

Active autoradiographs were also obtained for blanket drawers which contained no fuel, but which had fuel-containing drawers above them. The fuel column in the drawer above was not visible on the blanket drawer autoradiographs. Images of the U-238 plates in the blanket drawers were visible. An active autoradiograph of a blanket drawer is shown in Fig. 4C. It was obtained simultaneously with the autoradiograph in Fig. 4A. In Fig. 4C, the light, central portion of the autoradiograph corresponds to an 1.27-cm-wide sodium can, and the darker regions correspond to U-238 plates.

In Fig. 4A, the density of the image of the plutonium-containing fuel plates is approximately equal to the density of the image of the U-238 plates. This equality of density may be surprising at first because the fission rate in the U-238 in the assembly was only 15% of the total fission rate. However, the plutonium fuel plates were clad in 0.03-cm-thick stainless steel, while the U-238 plates were unclad. It was earlier concluded that the fuel-plate images were produced primarily by low-energy photons and beta radiation. If this conclusion is correct, then it can be expected that the image density of the U-238 plates will be enhanced because these plates have no cladding to absorb low-energy photons or beta radiation. While the images of the clad fuel plates and of the unclad U-238 plates do not look the same, their equal image densities for unequal fission rates presents a potential complication for interpreting active autoradiographs of drawers containing both clad plutonium

A



B



C



200-11-2002

0

Fig. 4. A: At-critical Active Autoradiograph of Plutonium Fuel Column
B: Passive Autoradiograph of a Plutonium Fuel Column
C: At-critical Active Autoradiograph of a Blanket Drawer

176.76

plates and unclad U-238 plates. The density of the images of the U-238 plates in Fig. 4C is low compared to the density of the images of U-238 plates in Fig. 4A because the fission rates are much lower in the blanket region than in the core region.

Reactivity Worths

As a first step in assessing the sensitivity of reactivity measurements to diversion detection, reactivity worths were measured for Pu-239 as a function of position within the ZPPR core. These measurements were made on ZPPR assembly 10-A whose reference configuration is shown in Fig. 5. The assembly was a hexagonal 4590-liter mockup of an LMFBR which contained 19 mockup control-rod positions. The core had an axial length of 101.6 cm and was divided into two fuel-enrichment zones. The inner core, with the mockup control-rod positions, contained, primarily, drawers with a single column of Pu-alloy fuel. The outer core was loaded with single-column drawers and double-column drawers in alternate positions. The 5.08 cm by 5.08 cm cross section of the core zones of a single-column drawer and a double-column drawer are shown in Fig. 6. Outside the outer core was a blanket zone whose drawers contained fertile material and coolant. The outermost zone contained a steel reflector. The ZPPR-10A reference configuration was critical, with a total fissile mass of approximately 2100 kg.

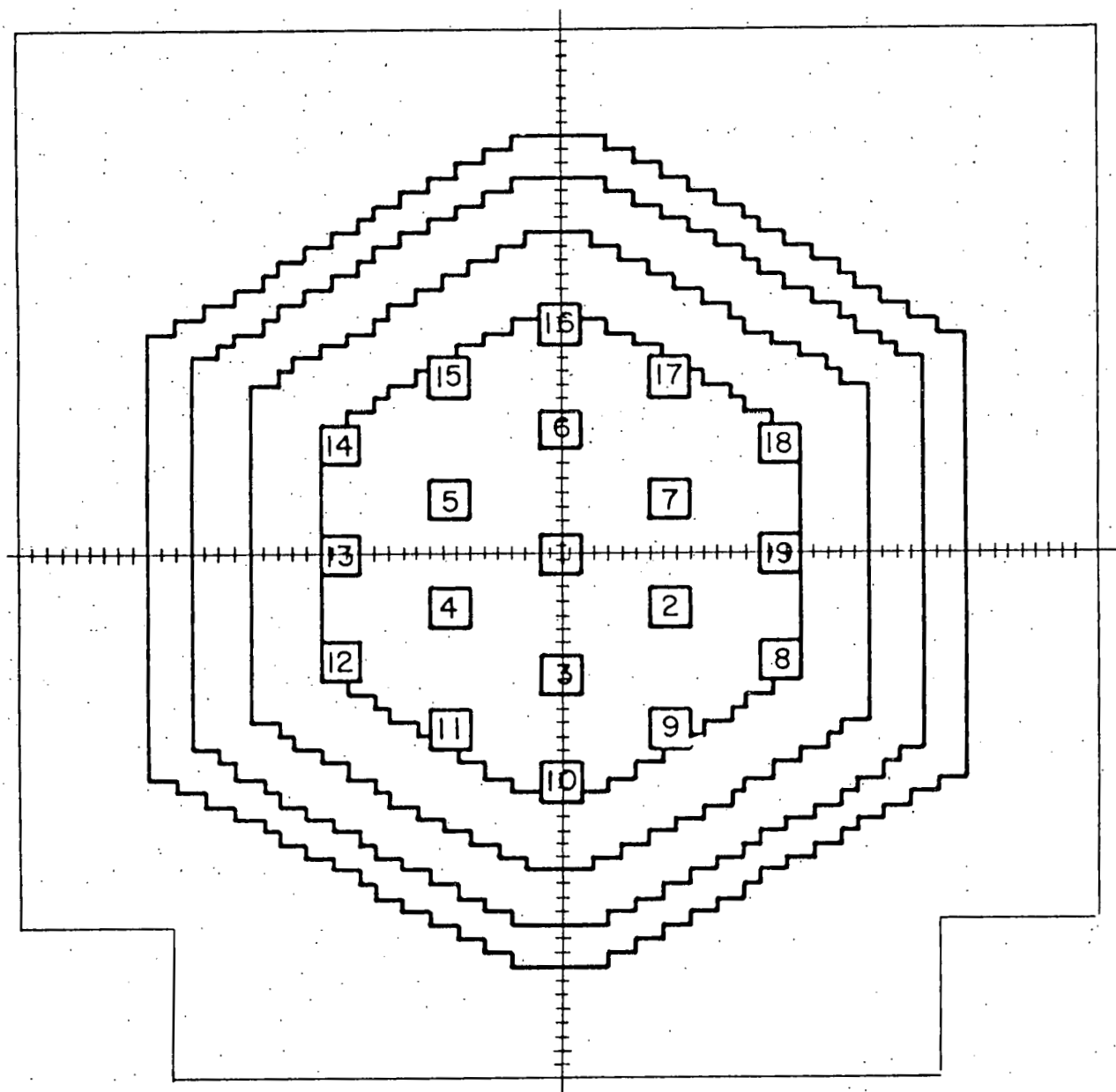
Reactivity worths were obtained with the use of inverse kinetics techniques. The neutron flux was measured as a function of time for a fissile sample which was moved across the center of the axial midplane of the core. The sample was moved in steps by remote control so that an entire traverse was performed during a single reactor run. A traverse was made with an empty sample holder to correct for the reactivity effect of the holder. Neutron flux measurements were made with BF₃ detectors located outside the core.

In addition to the experimental results, first-order perturbation reactivity worths for the isotopes Pu-239, Pu-240, and U-238 were obtained from a 28-neutron-energy-group diffusion calculation in two-dimensional geometry. Isotopic worths were combined in the proper weight fractions so that results could be compared to the experimental samples. A sample size (self-shielding) correction was also made.

The experimental and computational results for Pu-239 reactivity worths as a function of radial position at the centerline of the core axial midplane are presented in Table I. The uncertainties in the measured worths are 1 σ statistical variations. Graphical results are displayed in Fig. 7. The squares in Fig. 7 are the experimental data, while the solid line was obtained by applying a normalization factor to the calculated data over the fuel-bearing regions. The normalization factor was derived from a comparison of calculation and experiment.

The second factor required to determine the sensitivity of reactivity measurements to fissile material removal is the uncertainty of such measurements. One obvious contribution to this uncertainty is the simple experimental uncertainty in making any single reactivity measurement. However, because reactivity verification measurements will be compared to previously measured reference reactivities, the long-term reproducibility of reactivity measurements is also important.

In a fast critical assembly such as ZPPR, differences between reference reactivity measurements may arise from temperature variations, irreproducibility of table closure and control-rod positions, and unavoidable changes in the core composition, particularly the decay of Pu-241. The temperature of the ZPPR cell can be held constant to within $\pm 0.14^\circ\text{C}$. Therefore, if the approach to critical is always the same, uncertainties associated with temperature variations during reference measurements are small. Experience at ZPPR indicates that overall reproducibility of reactivity measurements on the same assembly over a short period (< 1 week) should be approximately ± 1 lh. However, over a longer time period, the decay of the Pu-241 (14.4-year half-life) present in small quantities in the ZPPR fuel alloy produces consistent and observable reactivity loss with time. Corrections can be made for the Pu-241 decay, but the decay still makes a contribution to the overall uncertainty. Assessment of these factors (for a range of critical assemblies containing 1.5% Pu-241 in Pu fuel) leads to a long-range (several month) uncertainty of ± 3 lh.



□ CONTROL ROD POSITION

Fig. 5. Core Configuration for ZEPHRA Assembly 10A.

U308
Fe2O3
Na2CO3
Na
ZPPR - Pu
Na
U238
U308

SINGLE COLUMN

Fe2O3
ZPPR - Pu
Na2CO3
Na
U308
Na
ZPPR - Pu
Fe2O3

DOUBLE COLUMN

Fig. 6. Cross Section Views of Single- and Double-column Fuel Drawers

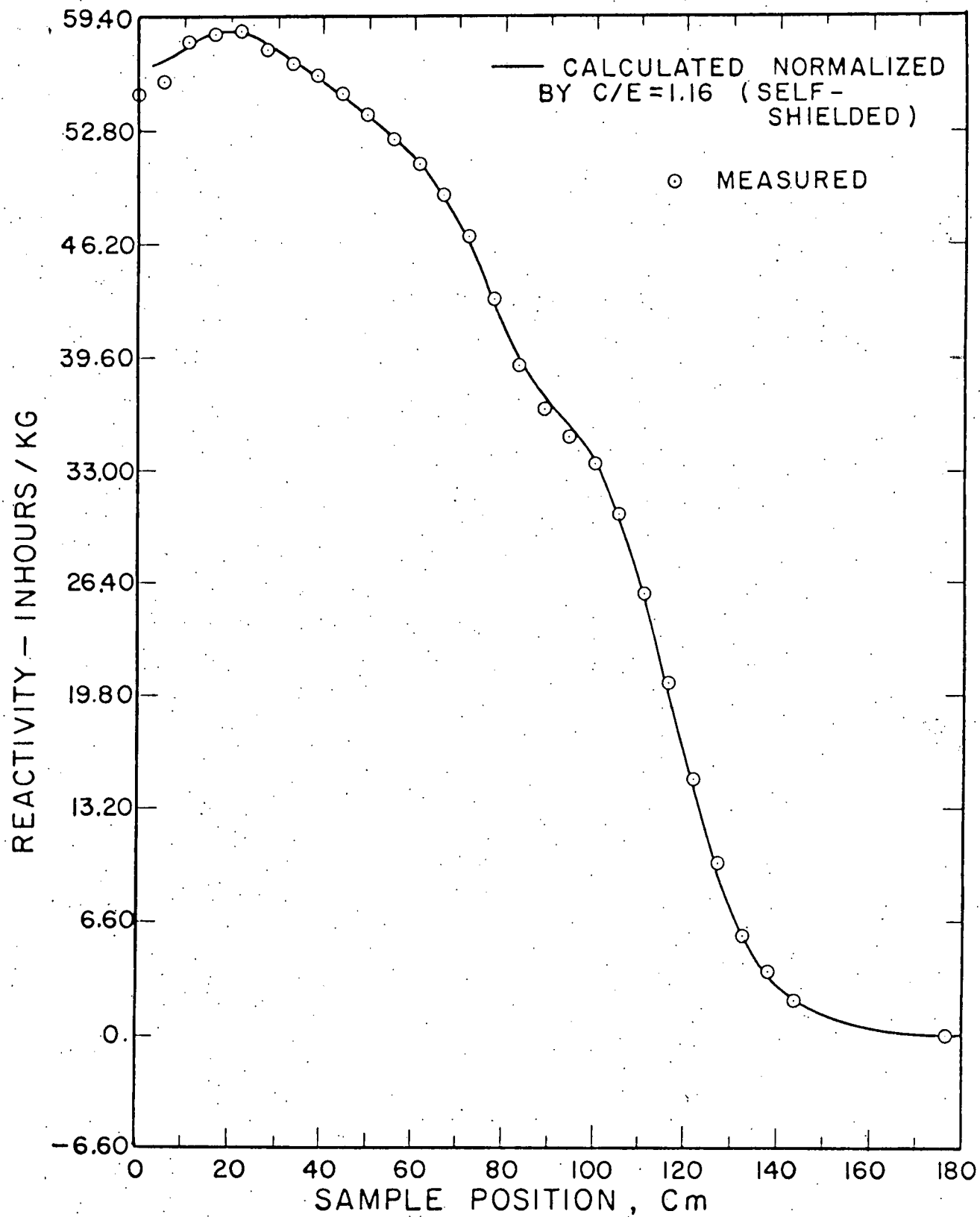


Fig. 7. Pu-239 Reactivity Worth as a Function of Position in Core

TABLE I. Calculated and Corrected Measured Radial Traverse for Pu-30, Sample in ZPPR Assembly 10A

Position, cm	Sample Worth, 1h/kg				
	Experiment	Calculated	Self-Shielding	Corrected Calculated	C/E
0.023	55.04 + 0.09				
5.522	55.67 + 0.12	63.66	1.04	65.97	1.19
11.052	57.95 + 0.10	64.39	1.04	67.06	1.16
16.576	58.48 + 0.12	65.00	1.05	67.94	1.16
22.095	58.67 + 0.09	64.96	1.05	68.00	1.16
27.625	57.49 + 0.09	64.26	1.05	67.29	1.17
33.147	56.68 + 0.11	63.12	1.05	66.11	1.17
38.671	56.01 + 0.11	61.82	1.05	64.82	1.16
44.201	54.94 + 0.12	60.52	1.05	63.54	1.16
49.720	53.71 + 0.10	59.33	1.05	62.22	1.16
55.245	52.33 + 0.10	58.03	1.05	60.83	1.16
60.772	50.77 + 0.07	56.42	1.05	59.22	1.17
66.294	49.01 + 0.12	54.14	1.05	56.97	1.16
71.818	46.55 + 0.11	51.28	1.05	53.76	1.15
77.346	42.91 + 0.09	47.53	1.04	49.63	1.16
82.867	39.05 + 0.11	44.11	1.04	45.90	1.18
88.389	36.57 + 0.11	41.70	1.04	43.42	1.19
93.914	34.95 + 0.09	39.59	1.05	41.39	1.18
99.444	33.37 + 0.09	37.08	1.05	39.09	1.17
104.965	30.32 + 0.14	33.24	1.06	35.14	1.16
110.490	25.65 + 0.13	28.02	1.06	29.66	1.16
116.017	20.46 + 0.11	21.88	1.06	23.09	1.13
121.539	14.86 + 0.13	15.82	1.05	16.57	1.12
127.066	9.90 + 0.12	10.87	1.01	11.04	1.12
132.588	5.66 + 0.13	6.90	0.97	6.73	1.19
138.115	3.71 + 0.08	4.25	0.93	3.95	1.07
143.634	2.04 + 0.13	2.73	0.87	2.38	1.18
176.789	0.00 + 0.06	0.00	0.67	0.00	

core edge

TABLE II. Fissile Removal Detection Sensitivity - ZPPR-10A (Using 1 1h short-term and 3 1h long-term Reactivity Sensitivity)

Radial Position, cm	Axial Position, cm	Isotope	Sensitivity, kg	
			Short Term	Long Term
0	0	^{239}Pu	0.017	0.05
124	0	^{239}Pu	0.067	0.2
124	50	^{239}Pu	0.267	0.8
0	0	^{235}U	0.023	0.07
124	0	^{235}U	0.09	0.27
124	50	^{235}U	0.36	1.09
0	0	^{233}U	0.012	0.036
124	0	^{233}U	0.05	0.15
124	50	^{233}U	0.20	0.60

These short- and long-term uncertainties can be combined with the reactivity worth results in Table I to obtain estimates of the fissile removal sensitivities. These sensitivities are presented in Table II for three core positions: the core center (0,0), the core edge at the axial midplane (124,0), and the minimum reactivity position at the radial and axial core edge (124,50). These sensitivities are only estimates and will later be compared with observed changes in reactivity when fuel was removed. Also, these sensitivities represent removals of almost pure fissile fuel material. Actual fuel plates which contain mixtures of fissile and fertile material will have reduced detection sensitivities at the core center and probably slight enhancement at the core edge due to the presence of the fertile material. This is expected because of the fertile material's negative reactivity effect at the core center and small positive reactivity effect at the core edge.

Effects of Fuel Removal

The next step in assessing the sensitivity of reactivity measurements to diversion was the measurement of reactivity change when fuel was removed from a reference core. These measurements were made on ZPPR assembly 10D. This assembly was a hexagonal 6100 liter mockup of an LMFBR which contained 31 control-rod positions. Though larger than the core in assembly 10A, the loading pattern was similar. Reference core 10D contained a fissile mass of roughly 2600 kg.

The reactivity of the subcritical 10D reference core was determined by the source-multiplication method. This reference reactivity was -0.29% . In the source-multiplication measurement, the neutron flux was measured by 64 in-core fission counters. Each fission counter consisted of a thin U-235 foil inside an ionization chamber filled with an argon-methane mixture. The fission counter was enclosed in a stainless steel chamber, 5.08 cm by 0.64 cm by 15.2 cm. The fission counters took the place of the fuel plate nearest the axial mid-plane in 32 different single-column fuel drawers in each reactor half. Reference assembly 10D is shown in Fig. 8. The x's indicate the location of fission counters in Half 1, while the o's indicate the location of fission counters in Half 2. One advantage in using the source-multiplication method to determine reactivity for diversion detection purposes is that one can simultaneously obtain both an integral reactivity for the entire core and a relative fission-rate distribution map from the 64 individual fission counters. The value of the integral reactivity is obtained from the weighted absolute fission rates of all 64 counters. This integral reactivity is then compared to the previous result for the reference core. In addition, the relative fission rate for each detector (fraction of the total fission rate in the core seen by one detector) in the verification measurement can be compared to the relative fission-rate obtained in the reference measurement. If a local diversion has occurred, the absolute fission-rate measurements will detect the reduced integral reactivity, and the fission-rate distribution map may be able to indicate the location of the diversion. If a diversion is made uniformly throughout the core, the integral reactivity will still decrease, but the fission-rate distribution could essentially be unchanged.

Operationally, fuel was removed from selected drawers of the reference configuration, and then the reactor halves were brought together and the control rods set to their reference positions. After a wait of a few minutes to assure equilibrium, the fission-counter outputs were read and stored by a digital computer. The computer then calculated the integral reactivity, the relative fission rates for each detector, and the ratio of these relative fission rates to the relative fission rates for the reference. The relative fission-rate ratios were displayed on a core map so that local variations could be seen.

The first simulated fuel diversion was a simple removal of all fuel plates from four single-column drawers near the center of the inner core. Approximately 2 kg of fissile material was removed. The fuel plates were not replaced with any other material. The results for this simulated diversion are shown in Fig. 9. The drawers with an "x" are those from which fuel was removed. This fuel removal resulted in a decrease in the integral reactivity of 0.16% . The numbers in Fig. 9 represent the changes in relative fission rates from the reference values. Each number appears at the approximate core location of its respective fission counter. The numbers, which we will call "Q", were computed from the expression

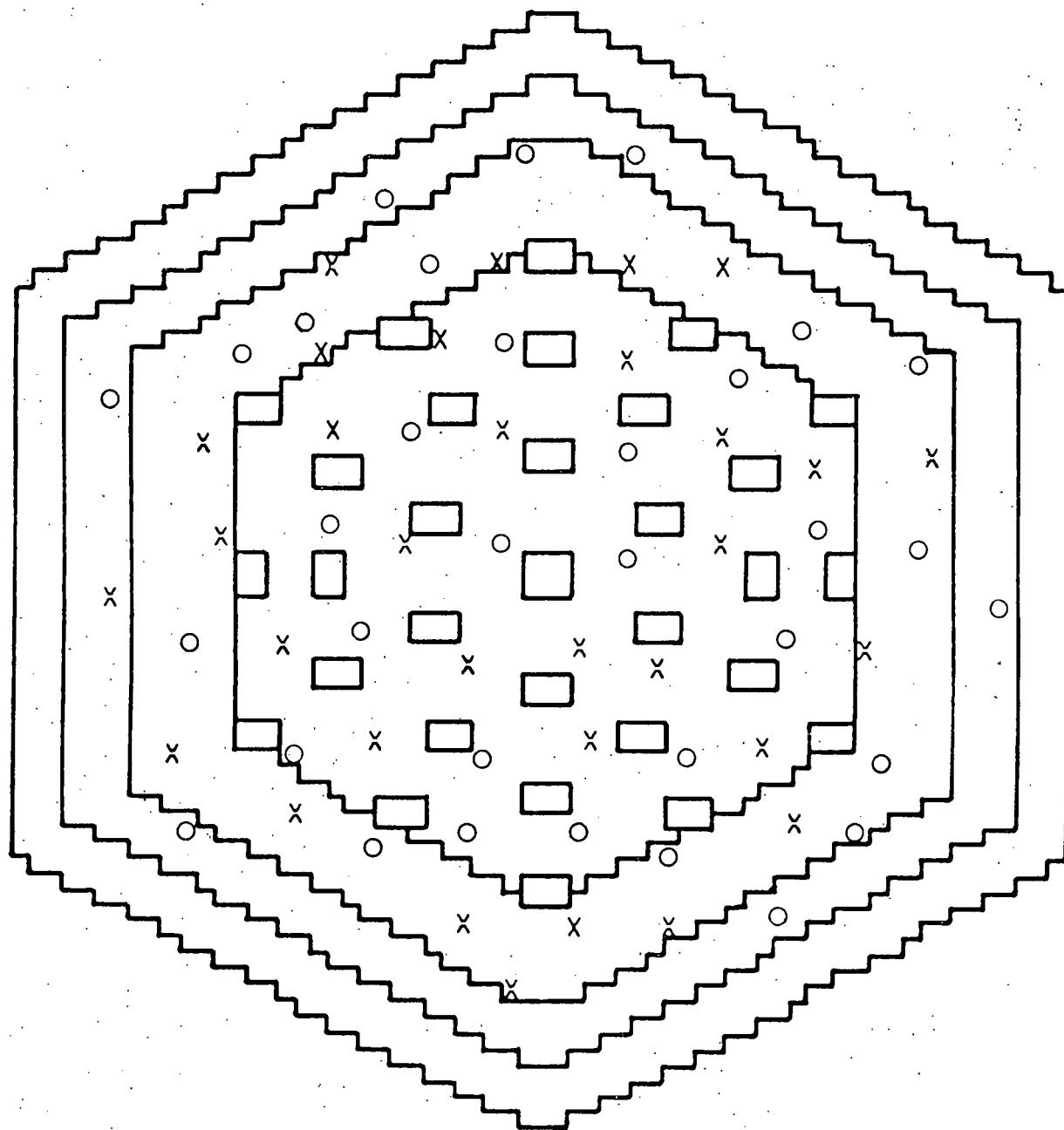


Fig. 8. ZPRR-10D Core Configuration Showing Locations of Fission Counters

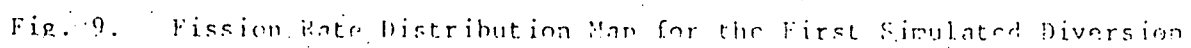


Fig. 9. Fission Rate Distribution Map for the First Simulated Diversion

$$Q = 1000$$

$$\left[\frac{\frac{a_i R_i}{\sum_i a_i R_i}}{\left(\frac{a_i R_i}{\sum_i a_i R_i} \right)_{\text{ref}}} - 1 \right]$$

where R_i is the fission-counter count rate and a_i is a weighting factor for that counter. The subscript "ref" indicates values for the reference configuration. Thus, a Q of -20 indicates a 2% decrease in the relative fission rate for that detector compared to the relative fission rate for that detector in the reference measurement. As indicated by the values in Fig. 9, there is a definite decrease in relative fission rates in the vicinity of the 2-kg localized fuel removal. The statistical uncertainties of the Q 's (1σ) were ± 3 for the inner core, ± 4 for the outer core, and ± 6 for the blanket. The uncertainty in the integral reactivity measurement was approximately 0.003% (1σ), although, when long-term instabilities are included (comparisons with reference reactivities made months previously), this uncertainty is likely to be 0.01% .

Table I gives a plutonium reactivity worth near the core center of ZPPR-10A of approximately 56 lh/kg. Using a conversion factor of 333 lh/\$, one would predict that a removal of 2 kg of Pu from the core center would reduce the reactivity by about 0.33\$. The observed reactivity reduction in the larger but similar ZPPR-10D was 0.16\$. This smaller reactivity decrease is expected because fuel was removed from the entire 50.8-cm fuel column, and because the reactivity worth of fuel decreases with increasing distance from the axial midplane. Axial worths at 50.8-cm from the midplane can be as much as a factor of four lower than at the midplane, so an 0.16\$ change in reactivity is reasonable.

One postulated method of compensating for a reactivity loss due to fissile removal is the substitution (or addition) of moderator material which may have a positive reactivity worth in a fast critical assembly. A very simple moderator substitution was made in the second simulated diversion. The voids created in the drawer by the fuel removal of the first simulated diversion were filled with polyethylene plates having the same dimensions as the fuel plates. Except for the presence of the polyethylene, the reactor core configuration was the same in the second simulated diversion as it was in the first. The source-multiplication measurement results for this second diversion are shown in Fig. 10. The integral reactivity was 0.18\$ less than the reference core, which was 0.02\$ less than the uncompensated diversion. Obviously, this effort to compensate for the reactivity loss was not successful. A possible explanation for this apparent negative reactivity worth for the polyethylene may be that the local shift of the neutron spectrum resulted in increased absorption in local fertile material (U-238), reducing the neutron flux available to more distant fissile material.

As in the first diversion, the map of Q values shows a marked reduction in relative fission rates in the vicinity of the diversion. These maps can be generated with any set of fission counter readings as a reference. When the first simulated diversion was used as the reference, there was no statistically significant difference between the relative fission rate distributions in the second and in the first diversions.

A more rigorous test of the integral reactivity method for detecting diversion (and the relative-fission-rate ratio method for locating diversion) is a distributed diversion in a low-worth region. In the third simulated diversion, fuel was removed from each of four single-column drawers along the core-blanket interface. The fuel plates were replaced with polyethylene plates. The location of the fuel removal and results for this third diversion are shown in Fig. 11. The integral reactivity was 0.02\$ less than the reference. This reactivity reduction is in reasonable agreement with the results of Table I when the low-worth locations of the fuel removals are considered. The total fissile material removed was again approximately 2 kg.

The most notable feature in the Q -value map is the very high reading for the fission counter directly adjacent to the drawer containing polyethylene near the bottom of the reactor. This is due to the counters' high sensitivity to low energy neutrons and the lack of intervening absorber. In the second diversion, the nearest fission detector was four

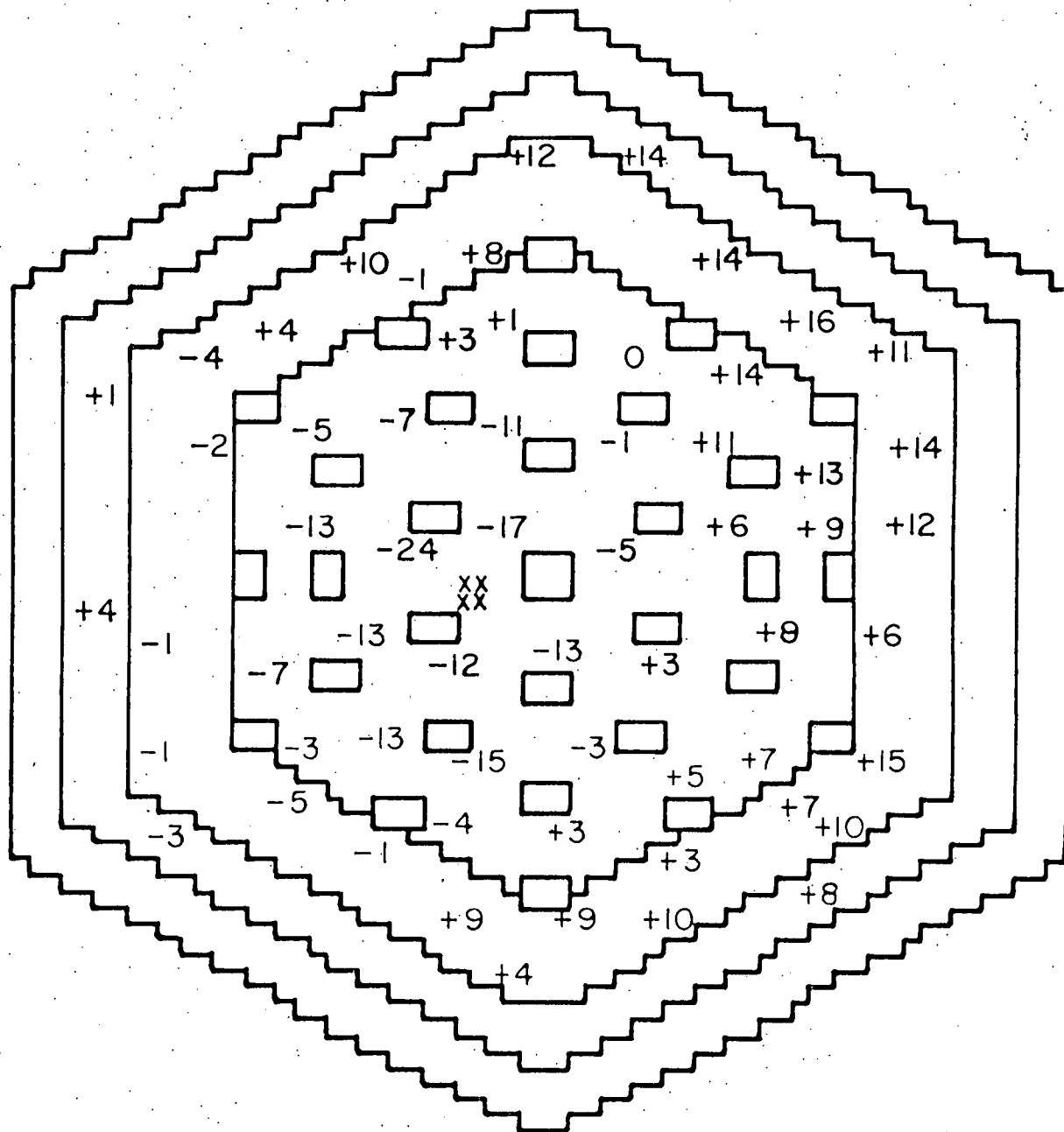


Fig. 10. Fission Rate Distribution Map for the Second Simulated Diversion

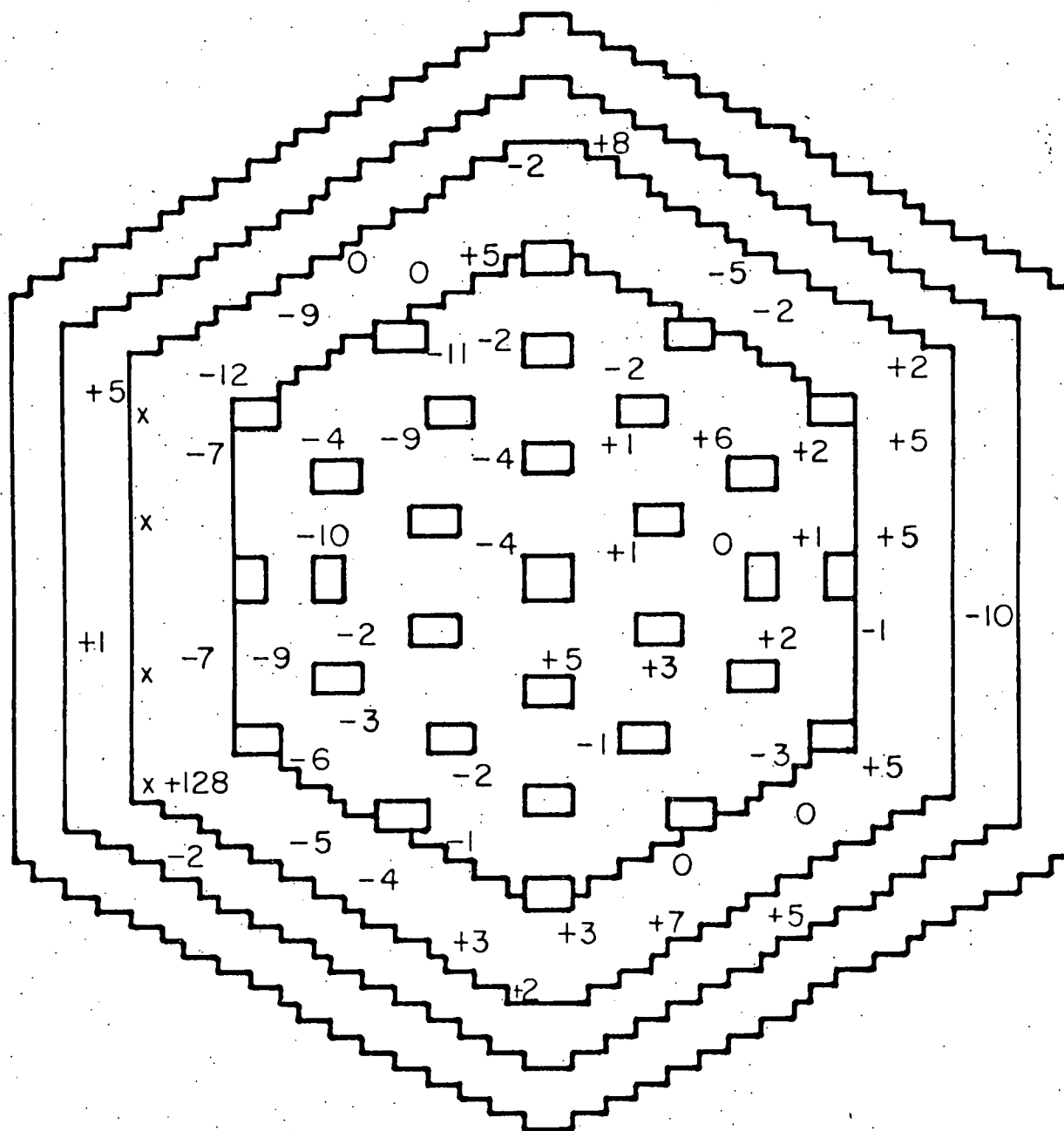


Fig. 11. Fission Rate Distribution Map for the Third Simulated Diversion

drawers away from the polyethylene. The other fission counters show a negative tilt toward the side where the fuel was removed, but these Q-values are not much greater than the statistical uncertainties.

A fourth simulated diversion was made which was localized in a minimum-worth region at a corner of the core. The fuel plates were removed from three single-column drawers and on double-column drawer, and polyethylene plates were substituted for the fuel. Total fissile material removed was 2.5 kg. Figure 12 shows the location of fuel removal and the Q-value map. The integral reactivity was only 0.01\$ less than the reference, a value which is close to the expected value for the uncertainty of long-term comparisons to reference measurements. The Q-value map successfully located the region of the diversion. The reason for the rather large positive reading from the fission counter in the blanket near the diversion is not obvious.

In order to test a slightly different reactivity compensation scheme, the configuration of the second simulated diversion was reproduced with the addition that polyethylene was substituted for fertile material (U-238) in two drawers in the outer core. Polyethylene once again replaced fuel removed from the inner core. This fifth simulated diversion is shown in Fig. 13. The x's indicate a drawer in which polyethylene replaced fuel, and the o's indicate a drawer in which polyethylene replaced U-238. The integral reactivity for this configuration was 0.14\$ less than the reference, which was 0.02\$ more than the case of fuel removal with no polyethylene (first diversion), and was 0.04\$ more than the case when polyethylene was substituted only for fuel (second diversion). It appears that replacing U-238 with polyethylene was a more successful reactivity compensation method than was replacing fuel with polyethylene. The Q-value map clearly located the missing fuel.

Figure 14 shows a Q-value map in which the configuration of the fifth simulated diversion was compared to the second simulated diversion. The only difference between these configurations is the polyethylene which replaced the U-238 in the outer core. This change is apparent from the map which shows a positive tilt to the side of the core with the extra polyethylene and the highest Q-values closest to the polyethylene.

Foil-Irradiation Experiments

Because the reactivity of a particular assembly-core configuration depends on the neutron-energy spectrum, it might be necessary to verify the spectrum as part of a reactivity verification. The addition of moderator to a fast reactor core can increase reactivity by increasing the relative neutron flux at low energies where the fission cross sections are relatively large. A typical neutron-energy spectrum for a ZPPR LMFBR mockup is shown in Fig. 15. As indicated, the relative flux is very small below a few hundred eV.

A verification of the neutron-energy spectrum of a fast critical assembly core can be obtained from experimentally determined rates of neutron-energy-dependent reactions. The types of measured data that are sensitive to neutron spectra are called spectral indices. One spectral index which has been used for many years in reactor experiments is the ratio of the fission rates in U-235 and U-238. The fission rate for U-235 generally decreases or is relatively flat with increasing neutron energy, while the fission rate for U-238 increases with increasing neutron energy, with a threshold near 1 MeV. Thus, the ratio of fission-product concentrations in U-238 and U-235 foils can be used as an index of the neutron energy distribution. The use of reaction-rate ratios in metal foils for safeguards purposes has been investigated in the highly enriched uranium-fueled SPECTR assembly in the USSR.¹¹ The foils should contain a material sensitive to low-energy neutrons in one reaction, and should contain a material sensitive to high-energy neutrons in another reaction. For low-energy sensitivity, (n,γ) reactions are useful, while (n,n') , $(n,2n)$, (n,p) and threshold fission reactions have good high-energy sensitivity.

The spectral index chosen for this study was the ratio of the rate of the In-115 (n,γ) reaction to the rate of the In-115 (n,n') reaction. This index has the advantage of requiring only a single foil of nonradioactive material. Natural indium contains about 96% In-115. The product of the (n,γ) reaction, In-116m₁, has a 54-minute half life and emits several gamma rays, including one at 417 keV. The product of the (n,n') reactions, In-115m, has a 4.5-hour half life and emits a single gamma ray of 335 keV. The ratio of the intensities

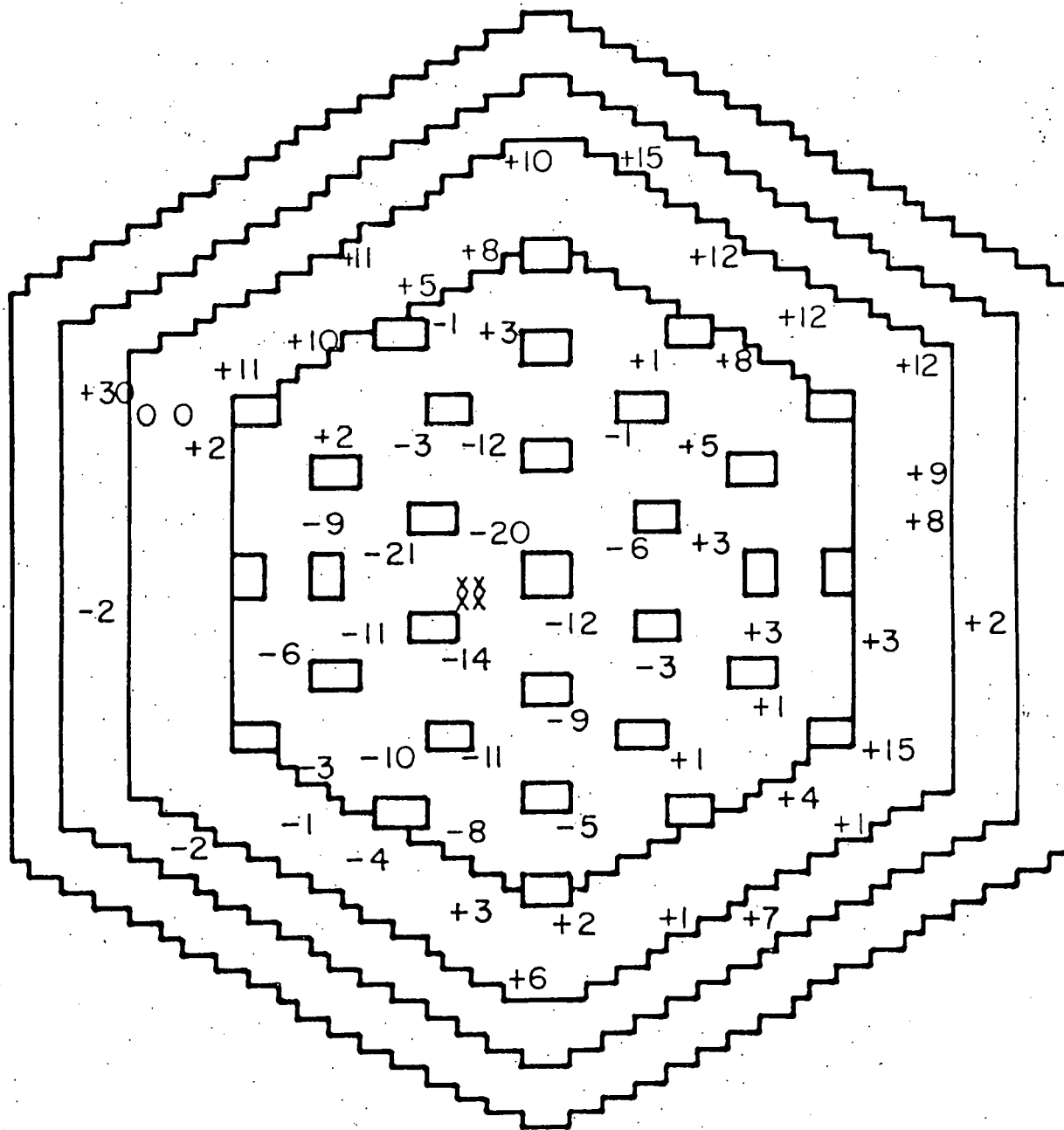


Fig. 13. Fission Rate Distribution Map for the Fifth Simulated Diversion

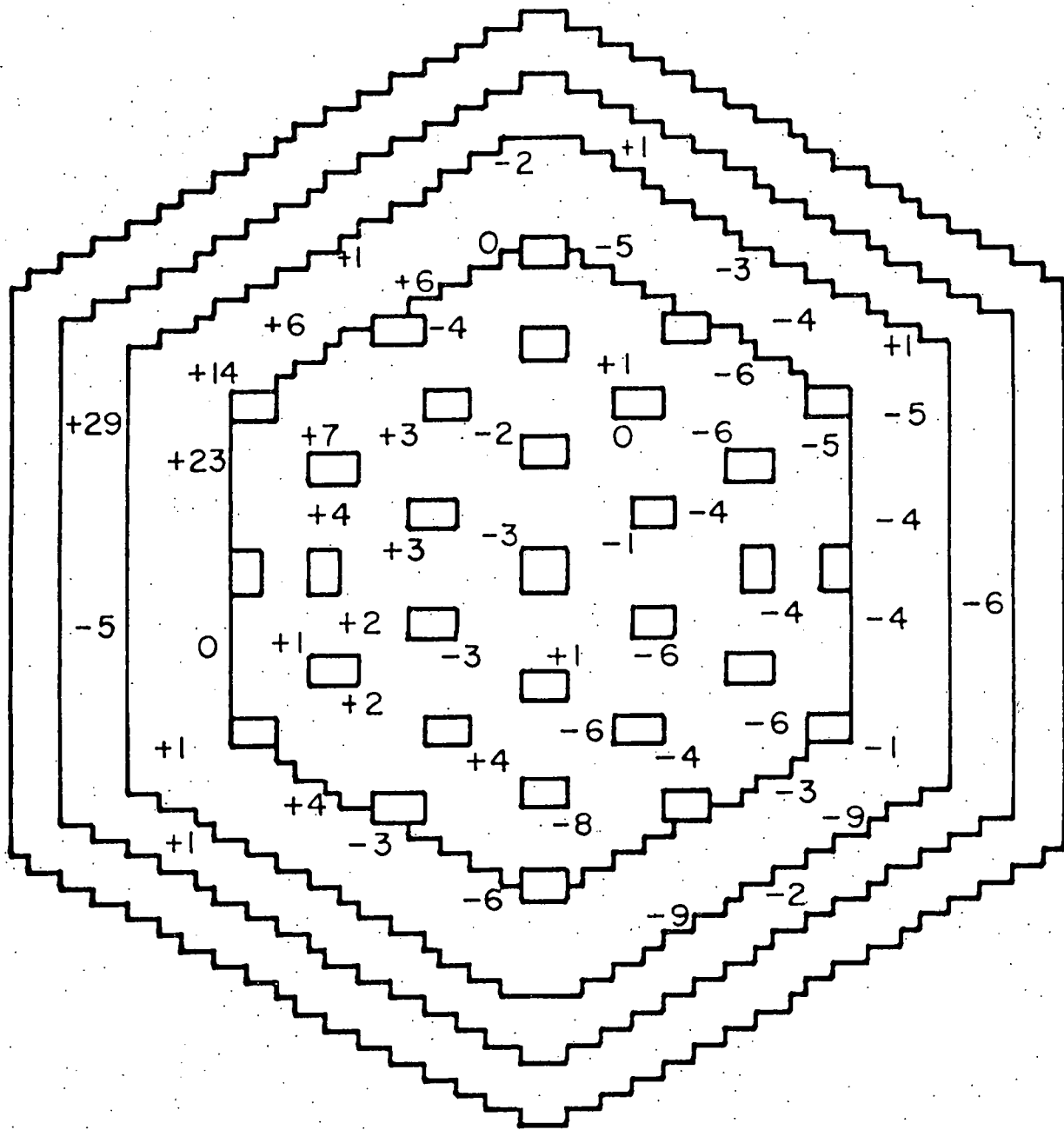


Fig. 14. Fission Rate Distribution Map Showing the Difference between the Fifth and Second Simulated Diversions

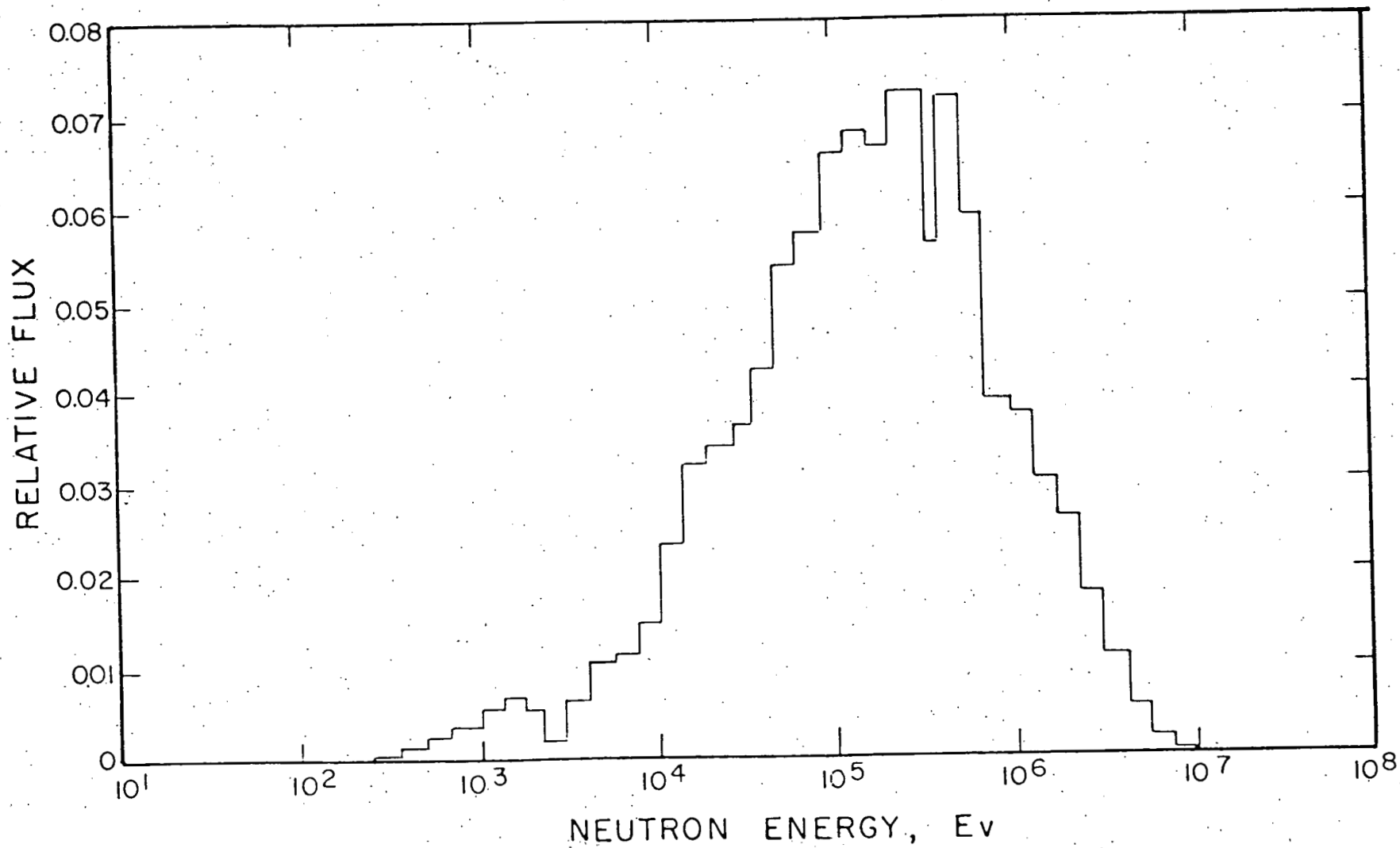


Fig. 15. Typical ZPPR Neutron Energy Spectrum

of the 417-keV and the 335-keV gamma rays as used as the spectral index. The neutron-energy dependence of the In-115 (n, γ) and In-115 (n, n') reactions is shown in Fig. 16.

The ability of the indium-foil spectral index to detect local spectral changes due to moderator substitution was tested by placing indium foils in several locations around the sites where polyethylene had been substituted for fuel. The foil-irradiation test was conducted with the reactor in a configuration which contained both the second and fourth diversion types. That is, polyethylene had been substituted for fuel in four drawers near the core center and in four drawers of a corner of the outer core. Foils were placed in and near the drawers containing polyethylene, and also in symmetrically equivalent drawers away from the diversion locations. These foils away from the diversion points provided reference spectral-index data points for essentially unperturbed regions of the core. Figure 17 gives the core-loading pattern for the foil-irradiation experiment. The x's indicate drawers in which polyethylene was substituted for fuel, and the o's indicate drawers which contain an indium foil.

The indium foils were 1.27 cm in diameter and 0.0127 cm thick. The foils were held in 0.051-cm-thick stainless steel holders. The holders were placed in the drawers parallel to the fuel or coolant plates. Each foil was placed adjacent to the central plate of the drawer, and each foil was 7.62 cm from the axial midplane. The foils were irradiated at a power of approximately 800 watts for 15 minutes. The reactor control rods were withdrawn from their positions during the reactivity measurements to permit the higher power operation. At 800 watts, the total neutron flux near the core center was approximately 8×10^9 neutrons $\text{cm}^{-2} \text{sec}^{-1}$. After about a two-hour cooling period, the foils were removed from the core. The foils were then placed in other holders for counting. Two Ge(Li) detectors with automatic sample changers were used to count the 41 foils. Total count time for the 41 spectra was approximately 20 hours. Several spectra were obtained for each sample.

The results for the simulated diversion near the core center are shown in Fig. 18. The horizontal cross-hatch marks indicate a drawer in which polyethylene was substituted for fuel. The numbers at the drawer locations are the ratios of the intensities of the (n, γ)-product gamma rays to the (n, n')-product gamma rays. The intensities were corrected for decay from the time of reactor shutdown. The larger the value of the spectral index, the greater the (n, γ) contribution, and hence, the softer the spectrum. The average value of the spectral index (s.i.) for the 13 foils in drawers in the relatively unperturbed region was 62.3, with a standard deviation (σ) of 3.0. The s.i. values of the foils located in the drawers with polyethylene were quite high. The third drawer removed (both horizontally and vertically) from the diversion site had an s.i. which was more than 2σ higher than the s.i.'s in the unperturbed region. The s.i. in the fourth drawer was not significantly higher than the average for the unperturbed region. One of the foils in the unperturbed region also had an s.i. value of 70, which was more than 2σ higher than the average for the region. This value may have been high due to its proximity to the diversion site and to the rather large amount of intervening sodium with its low absorption cross section.

The results for the simulated diversion at the edge of the outer core are shown in Fig. 19. The diagonal cross hatch marks indicate a drawer in which polyethylene was substituted for fuel. The average value of the s.i.'s for the unperturbed region was 49.7 with a standard deviation (σ) of 3.6. Again, the foil in a drawer containing polyethylene had a very large value of the s.i. The value of the s.i. is significantly higher than the unperturbed average value for only the two drawers adjacent to the diversion site, with the possible exception of the third drawer below the diversion. It appears that the foils are slightly less sensitive to the presence of the polyethylene at the core edge than at the core center, although that conclusion may not be justifiable from this small amount of evidence.

DISCUSSION

Autoradiography can provide a piece count of fuel elements in fast critical assembly drawers, and can also provide a simultaneous verification of the loading pattern in the reactor core. Autoradiography is an in-situ technique which requires no handling of fuel elements or reactor drawers, and a large number of drawers can be examined in a short time. Passive autoradiography, presently applicable only to plutonium fuels, can be used to examine any desired fraction of the reactor drawers. Because the passive autoradiographic images are produced mainly by radiation from Am-241, the verification of the fissile content of

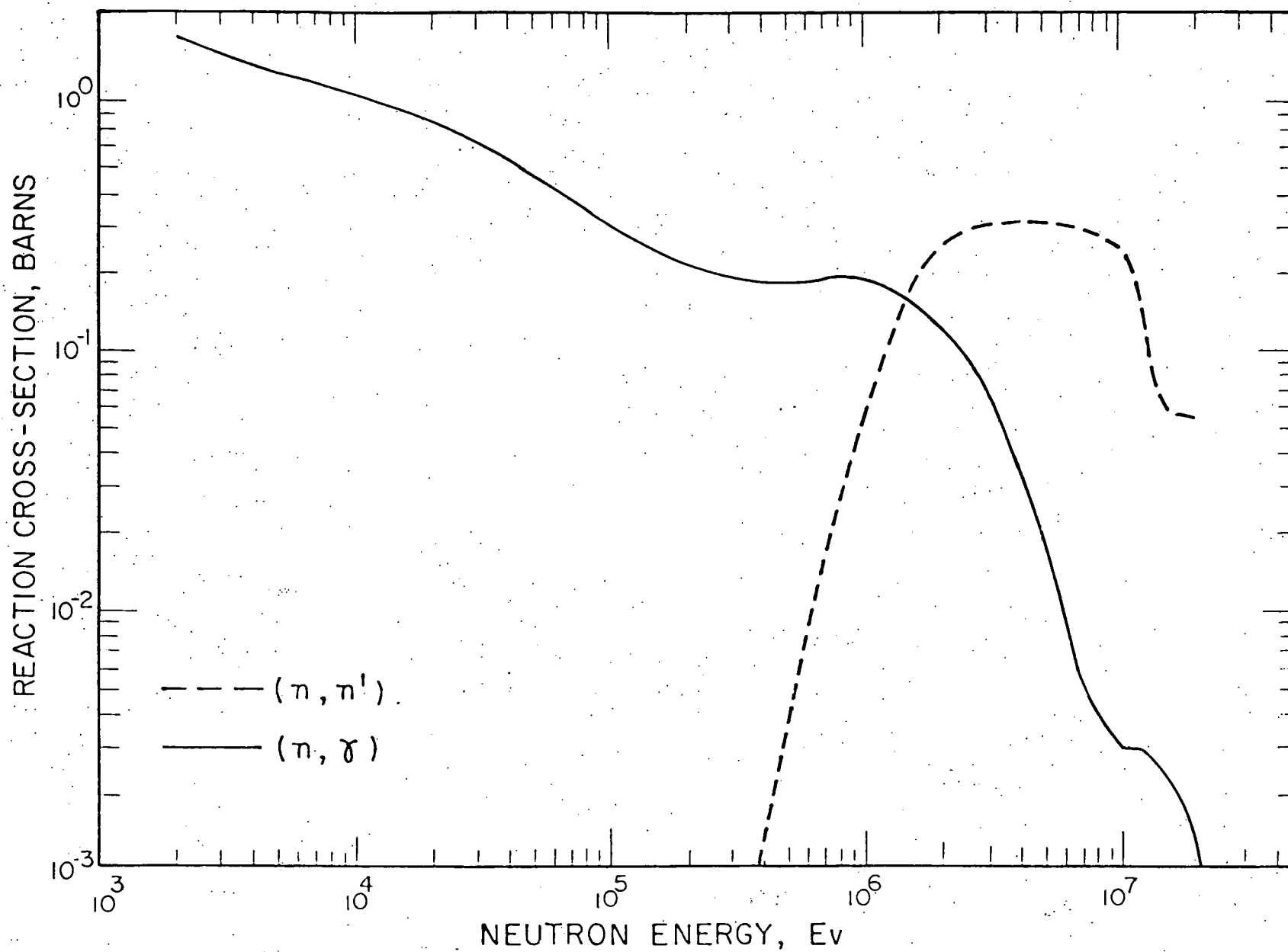


Fig. 19. Cross Sections of the In-115 (n, γ) and In-115 (n, n') Reactions as a Function of Neutron Energy

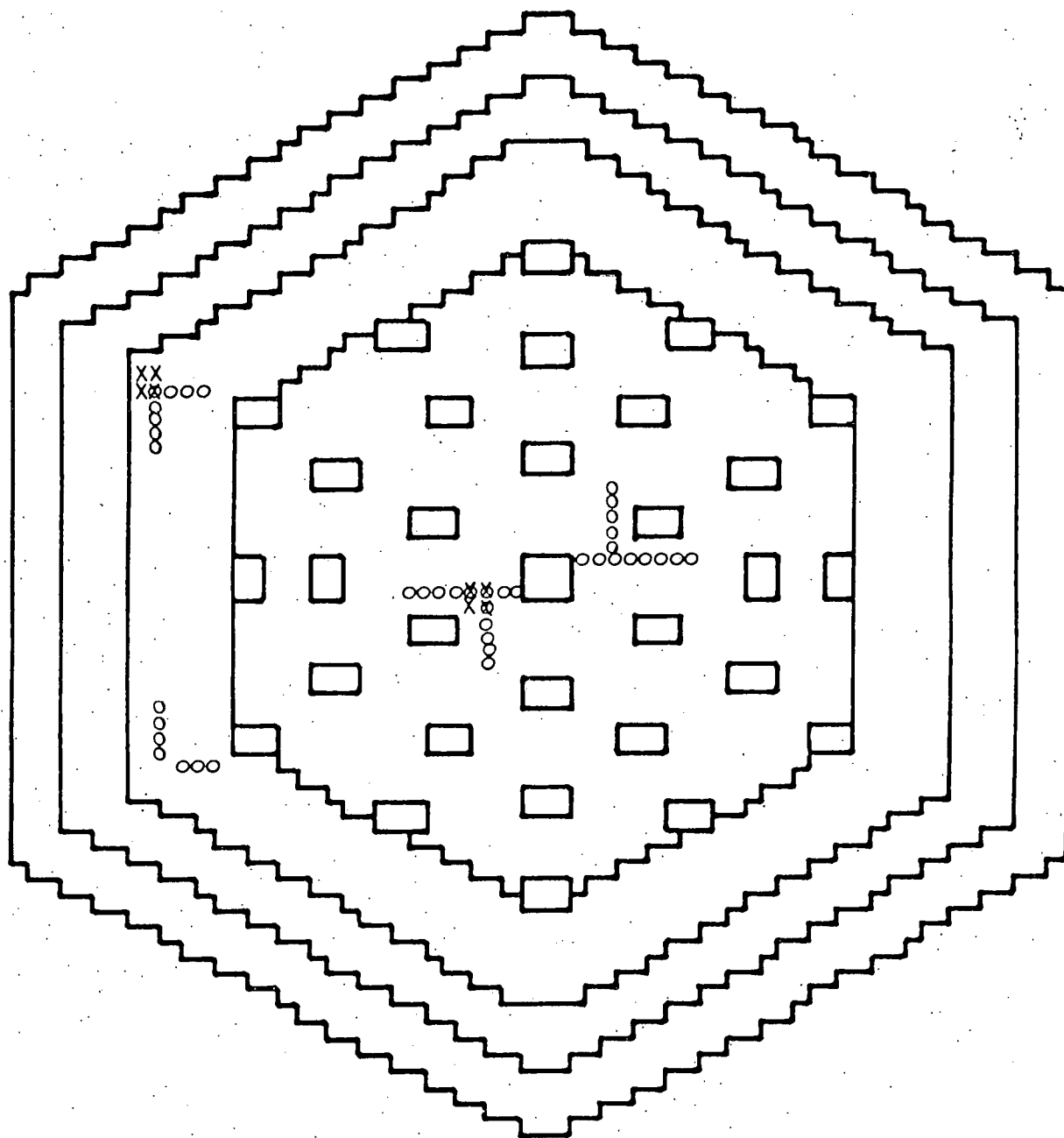


Fig. 17. Indium Foil Loading Pattern

fuel elements is indirect. However, for the active technique, the fuel element images are the direct result of the fissile content of the fuel elements and of the presence of a critical mass in the reactor. The film itself may have a positive reactivity effect that must be considered when planning an active autoradiographic inventory verification. Any reactivity effect must also be considered when interpreting the results of an active autoradiographic inventory.

Passive autoradiography is not presently practical for in-core uranium application. Further work is needed to explore the possibility of adapting existing passive autoradiographic techniques to the in-core uranium problem. The availability of a passive technique is important for verifications when the assembly cannot easily be brought into a critical configuration.

The application of active autoradiography to plutonium-fueled cores is more complicated than it is for uranium-fueled cores because of the approximately equal image density for the clad plutonium plates and the unclad U-238 plates and because of the large contribution to fuel-plate images from spontaneous radiation. More data is needed to better evaluate the active autoradiographic technique for plutonium-fueled cores. However, the present passive technique is quite adequate as a companion technique to reactivity measurements and foil irradiations.

The reactor power required for film exposure in an active autoradiographic verification is sufficient to activate indium foils. Both the products of the (n,γ) and (n,n') reaction are produced in sufficient concentration with approximately a 100-watt-hour irradiation of the 6100-liter ZPPR. The 100-watt-hour irradiation is recommended for active autoradiography in a core with a volume of 6100 liters. The simultaneous foil irradiation and film exposure is expected to be applicable to uranium-fueled cores.

The results of the integral reactivity measurements have indicated that these measurements are indeed sensitive to the removal of small quantities of fissile material from fast critical assembly cores. The great attractiveness of the integral reactivity measurement lies in its ability to provide a quantitative estimate of the fissile content of an entire core in a single measurement requiring approximately two hours. This measurement time assumes that the reactor is already in its reference configuration. Depending on the degree of deviation from a reference configuration, the time required for a reactivity verification could be significantly longer than a few hours. The use of reference reactivity measurements requires an independent verification of the core contents upon initial reactor loading. The detection sensitivity to the 2.0 - 2.5 kg (fissile) simulated diversion was well outside measurement uncertainties for all but the fourth simulated diversion which was made in a zone of minimum fissile worth. These results were particularly encouraging in view of the IAEA detection-sensitivity goal of 8 kg (fissile).

The attempt to compensate for reactivity loss from fuel removal by substitution of polyethylene was not successful. In the only case where both a compensated and an uncompensated diversion was made (diversions 1 and 2), the compensated diversion actually resulted in a lower reactivity than did the uncompensated case. All other diversions were made with polyethylene substituted for fuel. This reactivity decrease with the addition of moderator was attributed to increased neutron capture in nearby absorbing materials (e.g., U-238). This increased absorption resulted in a reduction in the total neutron flux and consequently in a reduction in the fission rate in fuel several drawers away from the substitution where the nearest fission counters were located. In the third simulated diversion, a fission counter was in a drawer adjacent to a drawer containing polyethylene, and this fission counter showed a much higher relative fission rate than it did in the reference case. This high fission rate was attributed to the high counter sensitivity to low-energy neutrons and to the presence of a relatively large number of low-energy neutrons close to the polyethylene. These low-energy neutrons were not absorbed because of the relatively small amount of absorber material between the polyethylene and the fission counter. Fission counters four drawers removed from polyethylene substituted for fuel showed reduced relative fission rates. These observations with fission counters were supported by the foil-irradiation experiments which indicated that the effects of the polyethylene had a short range. The polyethylene-induced effect on flux was not detectable four drawers away in the inner core, and was not detectable three drawers away at the core edge. In both regions, the effect was readily detected in the adjacent drawer.

In the fifth simulated diversion, polyethylene was substituted for U-238 in fuel-containing drawers. The reactivity effect for this substitution was positive and the effect was observed by fission counters more than four drawers away. The reactivity effect resulted from the removal of U-238 absorber and addition of moderator. No measurements were made to evaluate the effects of U-238 removal alone.

The combined results of the reactivity measurements and the foil-irradiation measurements do seem to suggest that the most effective way to conduct a reactivity-measurement-based verification is to place the indium foils in drawers directly adjacent to the fission counters. This tactic appears to have the best chance of detecting the substitution, or the addition, of moderator to compensate for the removal of fissile material. If a diverter wants to substitute moderator for fuel to cover up the removal of fuel, he must do so close to the fission counter. However, if this substitution is close enough to the fission counter to compensate for the fuel removal, it is likely to be close enough to the indium foil to have a detectable effect on the spectral index.

One other conclusion which can be drawn at this point is that it is not a trivial problem for a diverter to devise a reactivity-compensation scheme. An extensive series of test diversions and compensations would be needed to cover up for any significant fuel removal, especially if 64 fission counters with 64 accompanying spectral-index-measuring foils were used in a verification measurement. Such a diversion and cover-up may be possible for an entire facility working together, but would be exceedingly difficult for an individual, or even a few individuals in collusion.

The applicability of reactivity measurements and other reactor operating parameters to safeguards purposes was studied previously by Gryazev^{12,13} at the small, highly enriched uranium-fueled fast critical assembly SPEKTR. In their experiments, Gryazev, et al., made simulated diversions by removing uranium fuel and compensated for the reactivity loss by adding moderator in various parts of the core. They found that they could completely compensate for fuel removal by replacing uranium-containing elements with polyethylene elements. In these experiments, Gryazev measured the fundamental harmonic decay constant for a subcritical configuration whose reactivity was held constant by additions of moderator. Deviations of this decay constant from reference values were due to changes in the prompt neutron lifetime. Compensated diversions as small as 1.5 kg U-235 were detected.

The evaluation presented here of the reactivity measurement as a safeguards technique for fast critical assemblies has been limited to strictly technical considerations, such as detection sensitivities and measurement uncertainties. Another type of evaluation should be mentioned, at least in passing, when considering the application of reactivity measurements to international safeguards. This second type of evaluation concerns the practical application of the method by inspectors who are outsiders at a fast critical assembly, and it also concerns the degree to which reactivity measurements can be made truly independent of the facility and, hence, credible to an outsider. The experiments described here relied on in-core fission counters to obtain reactivity data. Fission-counter output was fed into scalars which were then read by a digital computer which, in turn, calculated reactivities. It would be extremely difficult for an international inspector to assure that all aspects of the fission-counter data-collection and -processing system were working properly and had not been subject to tampering. The practicability of inspectors' supplying their own instrumentation in the form of in-core or outside-core detectors should be considered, but is likely to be a difficult problem.

The foil irradiation techniques used to obtain spectral indices are relatively independent of the facility. The inspector can supply his own foils, can insert them into and remove them from the assembly drawers himself, and can use his own gamma-ray spectrometer to obtain the reaction-rate ratios. It is necessary to make reference spectral-index measurements throughout the reference core when it is first established because spectral indices will vary with location within the core. Foil-irradiation measurements require that the core be capable of producing relatively high fluxes ($10^9 - 10^{10}$ neutrons $\text{cm}^{-2} \text{sec}^{-1}$).

The indium reaction products used in these experiments had half-lives of 54 minutes and of 4.5 hours. Because of the gamma-detectors and automatic sample-changing equipment, the 41 foils could be counted quickly enough to make the 54-minute half-life isotope useful.

Inspectors may find it difficult to utilize this isotope because of its short half-life. Several other (n, γ) reactions can be substituted for the In-115 (n, γ) reaction.

Gryazev¹¹ also studied the application of spectral-index measurements to the detection of moderator-compensated diversions. His spectral index was the ratio of the Au-197 (n, γ) reaction rate to the In-115 (n,n') reaction rate. He found that he could detect polyethylene-compensated diversions 10 cm away from the point of diversion. This detection range of 10 cm was similar to that observed in the experiments reported here (diversion detectable at least two drawers away -- each drawer ~ 5 cm wide). Gryazev also used foil activation as a neutron flux measuring technique. A flux measuring technique using foils would probably require a prohibitively large number of foils in the large ZPPR cores.

The methods described here for measuring reactivity and for verifying the neutron-energy spectra are only examples of many methods available. Reactivities can be determined by measuring reactor period, or by such perturbation techniques as rod drop or rod oscillation. If external neutron sources are used, reactivities can be determined by a source-jerk technique or by fundamental-mode decay-constant measurements. The source-multiplication technique used in these experiments is applicable as described only for plutonium-fueled cores because the neutron source is the spontaneous fission in the even Pu isotopes. A similar technique could be applied to uranium-fueled cores by introducing some other neutron sources.

Several techniques are available for neutron-energy spectrum verification in addition to the foil-irradiation methods described here. Reactivity worths can also serve as spectral indices, and Gryazev¹² has successfully applied U-238 and boron worth measurements to the detection of compensated diversion in the SPEKTR fast critical assembly. The prompt neutron lifetime is also sensitive to changes in the neutron-energy spectrum. Estimates of prompt neutron lifetimes can be obtained from reactor-power noise measurements or from fundamental-mode decay-constant measurements at constant reactivity.

Each of the three techniques described here -- autoradiography, foil irradiation, and reactivity measurement -- have individual strengths which make them useful as inventory verification techniques, and have individual weaknesses which make them vulnerable to defeat by specific diversion scenarios. The roles of the various inventory verification techniques will be determined, in part, by the credibility of various postulated diversion strategies. For example, passive autoradiography of plutonium fuel in reactor drawers may possibly be vulnerable to defeat by radioactive "dummy" fuel elements which do not contain fissile material. A diversion scenario involving the fabrication of "dummy" fuel elements may be credible in international safeguards problems, and passive autoradiography would be combined with foil irradiation and/or reactivity measurements. However, fabrication of "dummy" fuel elements is much less credible when considering diversion by individuals or groups of individuals so that passive autoradiography may require less support in domestic safeguards applications.

Reactivity measurements have the advantage of great sensitivity to changes in the fissile content of a reactor. The entire core can be examined in a single measurement. However, because of the possibility of reactivity compensation by changes in core geometry, or by the substitution of non-fissile materials with positive reactivity worth, reactivity measurements must be supported by other techniques. If changes in core configuration or core composition are considered credible diversion scenarios, the autoradiography can verify the core loading pattern while foil irradiation measurements can, with some limitations, verify the core composition. Similarly, the foil irradiation technique is not very sensitive to core geometry and thus needs to be supported by autoradiography.

The potentially greatest assurance of diversion detection lies in a combination of all three inventory verification techniques. If the reactivity measurements are considered too facility dependent to be credible, then the relatively facility-independent autoradiographic and foil irradiation techniques should be used in combination. Only autoradiography can be applied to cores which are substantially subcritical.

REFERENCES

1. D.D. Cobb, J.L. Sapir, E.A. Kern, and R.J. Dietz, "Concepts for Inventory Verification in Critical Facilities", LA-7315 (1978).
2. D.O. Gunderson and J.L. Todd, "International Safeguards for Fast Critical Facilities", SAND 78-0168 (1978).
3. S.B. Brumbach and R.B. Perry, "Autoradiography as a Safeguards Technique for Plutonium Fuels", ANL-NDA-2, ISPO-49, (1979).
4. S.B. Brumbach and R.B. Perry, "Autoradiography as a Safeguards Inspection Technique for Unirradiated LWR Fuel Assemblies", ANL-78-27, ISPO-12 (1978).
5. G. Robert Keepin, Physics of Nuclear Kinetics, (Addison-Wesley, Reading, Mass., 1965).
6. W.G. Davey and W.C. Redman, Techniques in Fast Reactor Experiments, (Gordon and Breach, New York, 1970).
7. J.J. Duderstadt and L.J. Hamilton, Nuclear Reactor Analysis, (Wiley, New York, 1976).
8. S.G. Carpenter, H.F. McFarlane, M.J. Lineberry and C.L. Beck, "Conclusions Drawn from Subcritical Multiplication Results in ZPPR", Adv. React. Phys; Proceedings of ANS Topical Meeting, Gatlinburg, Tenn., April 10-12, 1978.
9. R.W. Peele and F.C. Maienschein, "Spectrum of Photons Emitted in Coincidence with Fission of U-235 by Thermal Neutrons", Phys. Rev. C, Vol. 3, p. 373 (1971).
10. R.L. Heath, "Gamma-ray Spectrum Catalogue", ANCR-1000-2.
11. V.M. Gryazev, "The Development of Methods and Procedures for Monitoring the Amount of Nuclear Materials in the Core of the SPEKTR Critical Assembly on the Basis of the Spectral Characteristics of the Neutron Flux", translated from Russian, IAEA unpublished data (1978).
12. V.M. Gryazev, "Development of Procedures and Techniques for Verifying the Quantity and Composition of Nuclear Materials at the SPEKTR Fast and Thermal Neutron Critical Facility", translated from Russian, IAEA unpublished data (1974).
13. V.M. Gryazev, "Experiments with a Nuclear Materials Accounting and Control System on the SPEKTR Fast-Thermal Critical Assembly", translated from Russian, IAEA unpublished data (1976).