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FAST CRITICAL ASSEMBLY SAFEGUARDS
Summary Report
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Nondestructive Assay Section
SPECIAL MATERIALS DIVISION

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ACTIVE AND PASSIVE AUTORADIOGRAPHIC METHODS
IN FAST CRITICAL ASSEMBLY SAFEGUARDS

ABSTRACT

Nuclear material inventory verification techniques for large split-table type fast critical assemblies are being studied under this program. Emphasis has been given to techniques that minimize fuel handling in order to reduce facility down time and radiation exposure to the inventory team. The techniques studied include autoradiography, reactivity, and spectral index measurements. Autoradiographic techniques capable of verifying the core loading diagram for a uranium or plutonium core have been demonstrated and examples of results are given. Results of reactivity and spectral index measurements for detection of simulated diversion of fuel from a large plutonium core are presented. These measurements indicate that the sensitivity of reactivity measurements to fuel removal is sufficient to detect a removal of 2 kg in a 2600 kg core. Reactivity compensation by the addition of polyethylene was readily detected by spectral index measurements using indium foils, if the foils were located within 10 cm of the moderator. The measurements made in this study were relative to a known reactor configuration and were made using facility equipment. Independent verification of reactivity and spectral index measurements by a safeguards inspector may be difficult.

GENERAL INTRODUCTION

Fast critical assemblies, such as those operated by the Argonne National Laboratory (ANL), require large inventories of plutonium and enriched-uranium fuels. The fuel elements which constitute this large inventory are considered to be attractive targets for diversion. Thus, it is important that effective safeguards be implemented at fast critical assembly facilities.

Three fast critical assemblies are operated by ANL: ZPPR in Idaho, and ZPR-6 and ZPR-9 in Illinois. Each is the split-table type, consisting of two halves. Each half is a square array of square cross-section matrix tubes. Fuel is contained in drawers 2" square and up to 36" long. The drawers also contain structural, coolant, and fertile materials. A typical drawer includes a core segment with either one or two columns of fuel plates (0.5 kg or 1.0 kg of fissile material), and a second segment with typical axial blanket materials. The drawers are inserted into the matrix tubes, and the halves are brought together to form the final reactor configuration.

The plutonium-fuelements in use at ANL are in the form of small, stainless steel-clad plates or rods. The plates contain plutonium metal alloys and are 2" wide, 0.25" or 0.125" thick and from 1" to 8" long. The rods contain a mixture of plutonium oxide and depleted uranium oxide and are 0.375" in diameter and 6" long. The uranium fuel elements are in the form of unclad (but coated) metal plates or stainless steel-clad rods. The rods contain uranium oxide and are 0.375" in diameter and 6"

long. The plates are 2" wide, from 1" to 3" long, and 0.026", 0.031", 0.062", or 0.125" thick. Uranium enrichments are 46%, 16%, and depleted for the rods, and 93%, 31%, 20%, and depleted for the plates.

Fuel elements at a fast critical assembly are normally in one of two locations: the assembly itself, or a storage container in a vault. Most of the time, a large majority of the fuel elements are in the assembly. Because of the large number of fuel elements, and because many of the elements are in the assembly, verification of the fissile-material inventory of the facility is time-consuming. If individual elements or even whole drawers are removed from the assembly for verification, the efficiency of the experimental program is 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 verification.

Any safeguards system for fast critical facilities must have minimization of operational impact as an important system goal. This report will describe some components for the inventory-verification part of a fast critical facility safeguards system. These components were selected because they have the potential to provide a high degree of assurance of diversion detection while not impacting severely on the facility. The general aspects of fast critical facility safeguards systems were recently reviewed by Gunderson and Todd.¹ Cobb, et al,² have described a model fast critical facility, its fissile material, and its operating procedures,

and have also studied the problem of inventory verification in such facilities.

Because the fuel elements at ANL facilities are very numerous and in the form of discrete, well-characterized plates and rods, item-control methods are used by the facility in its material-control system. Fuel elements are stored and loaded into drawers according to fissile element, size, manufacturer, and isotopic composition. Facility records provide the number of fuel elements of each type in any given storage container or assembly drawer. Responsibility for the material at the facility is shared between the operating division (Applied Physics) and the custodial division (Special Materials). Custody of the material is transferred from the custodial division to the operating division when the material is transferred between the vault and reactor. Each division maintains records of what is in the vault and what is in the reactor. Vault and reactor inventories are normally verified at the same time so that the total number of items of each type must equal the sum of the vault and the reactor inventories. Some of the inventory-verification techniques discussed in this report, such as autoradiography, will be aimed at verification of the facility's stated item count, while other techniques, such as active and passive neutron counting, will be aimed at verifying that the items contain the stated amounts of fissile material. Because combinations of inventory verification techniques provide greater assurance of diversion detection than does a single technique, the techniques discussed here are evaluated both for their individual capabilities and for their ability to combine with and complement other techniques.

PART I

ACTIVE AND PASSIVE AUTORADIOGRAPHIC METHODS IN FAST CRITICAL ASSEMBLY SAFEGUARDS

by

S. B. Brumbach, C. T. Roche and R. B. Perry

I. INTRODUCTION

In order to reduce the impact of inventory verification at fast critical assembly facilities, it is desirable to have the capability to perform in situ verifications of the fuel in the assembly core. It is also desirable to assay fuel in vault storage quickly and with a minimum of fuel handling and fuel movement. For fuel in the assembly core and in vault storage, a promising in-situ assay technique is autoradiography. Two types of autoradiography will be discussed. The first type is passive autoradiography in which spontaneously emitted beta and gamma radiation form an image of a fuel element on photographic film. The passive-autoradiographic method was previously applied to both plutonium-containing fuels³ and to low-enriched uranium-containing fuels.⁴ The second type is active autoradiography, in which prompt fission gamma radiation and fission-product beta and gamma radiation form an image of a fuel element on photographic film. Also described are foil irradiation and track-etch techniques which can use fast neutrons to form fuel-element images.

II. FUEL IN STORAGE

A. Plutonium

When X-ray sensitive film is placed on the outside surface of a fuel-storage container, the spontaneously emitted radiation from the various isotopes in plutonium fuels form clear images of the fuel elements inside the container. The autoradiograph of the storage container provides a piece count of the contents with a simultaneous visual verification that the fuel elements contain the expected concentration of radioactive material, and that the emitted radiation qualitatively duplicates that from plutonium-containing fuel elements. The autoradiographic piece count is then compared to the item count in the facility records.

The application of passive autoradiography to plutonium fuel in storage at ANL and at other facilities has been discussed in detail in previous reports.^{3,5} Only passive autoradiographic methods are applicable to fuel in storage. The speed of the autoradiographic method has allowed 100% piece-count verifications to be made for large populations of fuel elements without opening or moving the fuel storage containers. The autoradiographic piece-count verifications resulted in significant savings in time and radiation exposure compared to previous inventories which used visual piece counting on the entire population and nonautoradiographic NDA methods on a sample of the population. Commercially available industrial X-ray film was used in the verifications. Exposure times varied from 10 to 90 minutes, depending on the geometry and composition of the samples.

In some applications, the verification of fuel-element piece count alone may not provide sufficient assurance that diversion has not occurred. If the credible diversion scenarios for a facility include the fabrication of "dummy" fuel elements that contain radioactive, but nonfissile, material, then the autoradiographic piece count should be accompanied by other NDA measurements on a fraction of the inventory. The neutron-counting methods to be discussed in Part III can be used to verify that the fuel elements which produced the autoradiographic images also have the neutron-emission rates characteristic of plutonium-containing fuel elements. Gamma-ray spectroscopy is also an effective method for verifying that the fuel elements which produce autoradiographic images do contain plutonium. A random sample of fuel storage containers can be selected for gamma-ray spectrum verification or neutron counting after obtaining autoradiographs of their contents.

B. Uranium

Active neutron interrogation is the most effective technique for verifying the enriched-uranium fuel plates in vault storage. Results of active neutron interrogation measurements for uranium fuel plates are discussed in Section IV.

Passive autoradiography of uranium fuel elements is possible, and results have been reported for low-enriched uranium fuel rods in LWR assemblies.⁴ Images of highly enriched uranium plates can also be obtained. However, several problems are associated with obtaining passive autoradiographs of the uranium fuel plates stored at ANL which make autoradiography

a less desirable verification technique than active neutron interrogation.

One difficulty in obtaining an autoradiographic image of a U-235-containing fuel element is the exposure time required for useful image density. Exposures of many hours are required if ordinary X-ray film is used. 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. The problem can be partially overcome by using a combination of fast film and intensifying screens, but there would be a corresponding deterioration in image resolution. A second problem with the passive autoradiography of uranium is the large contribution from U-238 to the image density of a fuel element. The U-238 daughter, Pa-234m, emits 2.29 MeV beta radiation which efficiently exposes film. Consequently, additional filtration is needed to reduce the contribution to image density from this U-238 daughter. A third problem in applying autoradiography to uranium fuel plate verifications in ANL vault storage areas is the storage configuration. Plates are stored in stacks with each unclad plate touching its neighbors. This configuration makes it difficult to obtain a piece count from the image of a stack of plates.

III. FUEL IN ASSEMBLY CORES

A. Passive Methods1. Plutonium

Because a large portion of the population of fuel elements at a fast critical assembly is normally in the assembly core, it is very important that methods be available for in-core inventory verifications. Because the fuel elements in the core are not amenable to measurement by conventional NDA methods, the potential role for autoradiography is quite important. The application of passive autoradiography to plutonium fuel in the cores of fast critical assemblies has been described previously.^{3,5} Images of the edges of fuel plates in a reactor drawer 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 the drawer. The clearance between the top of a fuel element and the matrix tube is 0.060" in the fast critical assemblies operated by ANL. The film packet used in previous applications was Kodak Ready-Pack Type-M X-ray film. The film packet was 1.50" wide and was 0.035" thick where the ends of the packet were sealed with black tape. The passive autoradiographic method allowed a fuel-plate piece count to be made in a large number of drawers in a short time.

If a piece count alone is not considered sufficient, it may be possible to follow a passive, in-core autoradiographic verification with a reactivity measurement or with a foil irradiation which can verify the

presence of a critical mass in the core. If reactivity measurements, or any critical-mass-verification methods are used, autoradiography will be required to verify the core-loading pattern of the assembly. Reactivity measurements and foil-irradiation measurements will be discussed in Part II.

2. Uranium

The contributions to uranium-plate images from U-238 daughters makes it difficult to distinguish between unclad U-238 plates and unclad U-235 plates in assembly drawers. The beta-radiation filtering methods used for low-enriched uranium fuel rods⁴ are not presently practical for in-core applications because the additional filters make the packets too thick to fit into the drawers. Further work is required to investigate possible methods for applying passive autoradiography to uranium fuel in assembly cores.

B. Active Methods

1. Uranium

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 the short-lived fission-product beta and gamma radiation produce the fuel-plate image. The formation of an image is the direct result of the critical operation of the assembly. Film is loaded into drawers just as

it was in the case of passive autoradiography. With film in the drawers, the assembly is brought into a critical configuration and is operated at a power which produces a prompt-fission and fission-product gamma-ray flux much greater than the spontaneous gamma-ray flux from the uranium.

The ZPR-9 fast critical assembly was used to test the applicability of active autoradiography in uranium-fueled cores. The fuel drawers of the assembly contained a single column of 93%-enriched fuel plates. Each plate was 2" wide, 1/16" thick, and from 1" to 3" 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.

Because the spontaneous radiation emission from uranium is small, two types of active autoradiography are possible. 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 the drawers; (2) the reactor is brought to a critical configuration and operated at an appropriate power; (3) 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 overnight; (3) the following day, film is inserted into the drawers and is exposed for a few hours; (4) the film is removed and processed. In this second method, relatively sensitive film is used.

An example of an active autoradiograph obtained after critical operation of the reactor (the second method) is shown in Fig. 1-A. This autoradiograph was obtained approximately 18 hours after reactor shutdown. The reactor had been operated at a power of 255 watts for 66 minutes. Film exposure time was 66 minutes. 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 1-B shows a passive autoradiograph of an identical fuel column obtained with Kodak AA Industrex film and a 75-minute exposure. The uranium plates in Fig. 1-B had not been irradiated in the reactor for several weeks.

An example of an active autoradiograph obtained during a critical operation of the reactor (the first method) 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 1.5" 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-minute 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 1/16"-thick fuel column can be clearly seen in Fig. 2 as the darkest of the three lines in the center.

The sensitivity of the active, at-critical, autoradiographic technique to some simple drawer-loading defects was tested. Figure 3-A shows an

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.

active autoradiograph of part of a fuel column from which a 3"-long, 93%-enriched-uranium plate was removed. Figure 3-B shows an active autoradiograph of part of a fuel column in which a 3"-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 1/32"-thick fuel plate was substituted for a 1/16"-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 lh/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 about 30 pieces of film, each 40" long, could be placed in the 800-drawer assembly without violating safety regulations. More pieces of film could be added, but the assembly reactivity would have to be decreased by removing fuel drawers. In order to ensure 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. Active autoradiography is somewhat more complicated for plutonium than for uranium because of the large spontaneous radiation component from plutonium fuel. Only at-critical, active autoradiography is practical for plutonium.

An example of an active autoradiograph obtained in the ZPPR assembly is shown in Fig. 4-A. 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 0.25"-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 fission rate in the U-238 in the ZPPR core was 15% of the total fission rate. The space between the U-238 column and the fuel-plate column was occupied by a can containing sodium. Figure 4-B 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 that 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 start-up, irradiation, and reactor cooling. In Fig. 4, the fuel-column image is more dense in the active autoradiograph (Fig. 4-A; 12 optical-density units) than in the passive autoradiograph (Fig. 4-B; 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 might 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

Fig. 4. A. At-critical active autoradiograph of a plutonium fuel column.
 B. Passive autoradiograph of a plutonium fuel column.
 C. At-critical active autoradiograph of a blanket drawer.

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 0.110" 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 0.110"-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 spectrum⁶ and the fission-product spectrum,⁷ even though some very high energy photons are produced. In addition, the beta radiation from fission products will be highly absorbed by the 0.110"-thick steel but will still be able to expose film within the drawer.

Active autoradiographs were also obtained from 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. 4-C. It was obtained simultaneously with the autoradiograph in Fig. 4-A. In Fig. 4-C, the light center portion of the autoradiograph corresponds to an 0.5"-wide sodium can, and the darker regions correspond

to U-238 plates.

In Fig. 4-A, 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 was only 15% of the total fission rate. However, the plutonium fuel plates were clad in 0.012"-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 plates and unclad U-238 plates. The density of the images of the U-238 plates in Fig. 4-C is low compared to the density of the images of U-238 plates in Fig. 4-A because the fission rates are much lower in the blanket region than in the core region.

IV. NEUTRON TRACK-ETCH TECHNIQUES

Track-etch techniques have long been used to record the passage of ionizing radiation. The basic principles of track formation in solids has been recently reviewed.⁸ Neutron-track detectors have been used to record images in both thermal-neutron radiography and in fast-neutron radiography.^{9,10} Because we were interested in techniques applicable to fast critical assemblies, only track-etch techniques sensitive to fast neutrons were considered. The materials commonly used for detecting fast neutrons are polycarbonate plastics. Such materials register fast neutrons by registering the mechanical damage to the plastic caused by carbon and oxygen recoil ions from neutron elastic collisions.¹¹ Some track-recording plastics also register alpha particles and recoil protons. The mechanical damage produced by the passage of recoil ions can be made visible--at least with the aid of an optical microscope--by chemical etching of the damaged material. Areas of local damage (the particle track) can be dissolved at a higher rate than nearby undamaged material, so that the fast-neutron interaction point is revealed as an etch pit in the recording material.

Various materials were evaluated for their ability to record the passage of fast neutrons. These materials were cellulose nitrate, cellulose acetate butyrate (CAB), cellulose triacetate (CTA), Lexan, Makrofol, and CR-39. The sensitivity of the various detector materials to fast neutrons was measured with a Cf-252 spontaneous-fission source of known strength (3.2×10^6 neutrons $\text{cm}^{-2} \text{sec}^{-1}$ at the sample). All six of the track recorders studied did form etch pits following fast-neutron irradi-

ation. A few basic properties of these track recorders are summarized in Table I. The most sensitive material to Cf-252 fission neutrons was CR-39. The background pit density was also lowest for CR-39. The background pit density was the density of etch pits in etched, unirradiated samples. Some of the unirradiated materials, particularly Lexan, Makrofol, and cellulose nitrate, had quite rough surfaces after etching. The rough background often made pit counting very difficult. Pit counts were obtained from photomicrographs. The low background density of pits in CR-39 had been reported earlier.¹² Unfortunately, CR-39 was available only in cast sheets with a minimum thickness of 1/32", which was too thick to fit readily into assembly drawers. These sheets of CR-39 were quite brittle. Figure 5-A shows a photomicrograph of etched, unirradiated CR-39, while Fig. 5-B shows an etched CR-39 sample which was irradiation with 1×10^9 neutrons cm^{-2} from the Cf-252 source. The elongated pits in Fig. 5-B were attributed to recoil ion trajectories that were not normal to the surface. A series of Cf-252 irradiations of CR-39 samples with various exposure times showed that the etch-pit density was proportional to the neutron fluence.

Because of the high sensitivity of CR-39 to fast neutrons, and because of the high neutron fluxes present during the operation of fast critical assemblies, CR-39 was easily saturated. Saturation results when the individual track-etch pits intersect to form a rather uniformly rough surface with no individually distinguishable pits. The less sensitive track recorders such as CTA and CAB can be irradiated in fast critical assemblies and not be saturated. Figure 6-A shows a photomicrograph of unirradiated etched CAB, while Fig. 6-B shows an etched CAB sample which

TABLE I. Properties of neutron-track recorders.

	CR-39	Lexan	Makrofol	Cellulose Nitrate	Cellulose Triacetate	Cellulose Acetate Butyrate
Fission neutron (Cf-252) sensitivity pits/neutron	2×10^{-4}	1×10^{-5}	a	2×10^{-6}	2×10^{-6}	2×10^{-6}
Am-Li neutron sensitivity	9×10^{-5}	$(< 10^{-5})^a$	a	--	very small	very small
Etching conditions	3 hr in 6.5N KOH (aqueous) 65°	2 hr in 10% KOH (Methanol) 22°	3 hr in 6.5N KOH (aqueous) 65°	1-2 hr in 6.5N KOH (aqueous) 40°-50°	3 hr in 6.5N KOH (aqueous) 65°	3 hr in 6.5N KOH (aqueous) 65°
Background pit density, pits/cm ² (no irradiation, standard etch)	5×10^4	3×10^5 surface is quite rough	surface is quite rough	2×10^5 surface is quite rough	1×10^5 surface is rough	b

^aPits too difficult to count due to rough background

^bPit background is low; surface roughness makes background counting difficult

A

B

100 μ m

Fig. 5. A. Etched, unirradiated CR-39.
B. Etched CR-39 after 10^9 neutrons cm^{-2} .

Fig. 6. A. Etched, unirradiated CAB.
B. Etched, ZPPR-irradiated CAB.

was irradiated during a 1500 watt-hour run in ZPPR. Results for CTA were similar to those for CAS, except that CTA had a somewhat rougher etched background which made pit counting more difficult.

The most used track-etch recorder in neutron radiography is cellulose nitrate. Figure 7-A shows an etched, unirradiated cellulose-nitrate sample, and Fig. 7-B shows an etched cellulose-nitrate sample which was irradiated with 2×10^{11} neutrons cm^{-2} from the Cf-252 source. The cellulose nitrate was Kodak LR 115 Type II. As shown in Fig. 7, the background pit density was quite high. Lexan and Makrofol also exhibited high background pit densities.

Because track-etch detectors can record the passage of fast neutrons, they can be used as imaging devices to record the spatial variation within a fast-neutron flux. Thus, a sheet of polycarbonate plastic in a neutron flux can be viewed as analogous to the sheet of X-ray-sensitive film in an X- or gamma-ray flux. For fuel-column imaging in the core of a fast critical assembly, a fast-neutron track recorder would be used instead of photon-sensitive film and would record the fast-neutron flux gradient in the assembly drawer. The work of Berger⁹ indicated that, with a sufficiently high fast-neutron flux, images of neutron-flux gradients could be made visible to the unaided eye.

In a fast critical assembly, the fissile material is localized in the column of fuel plates in the assembly drawer, so that a discrete, local source of fast neutrons does exist. It is this local source which is to be imaged. Fuel columns are normally separated by 1" to 2".

Fig. 7. A. Etched, unirradiated cellulose nitrate
 B. Etched cellulose nitrate after 2×10^{11} neutrons cm^{-2} .

However, the fast neutrons produced in the discrete sources penetrate the material between sources much more readily than the photons produced in these sources, so that the fast-neutron flux gradient across an assembly drawer is much smaller than the photon-flux gradient. Consequently, it is expected that fast-neutron images will have poorer resolution than photon images. Calculations which were based on a drawer-loading pattern and on the appropriate reaction cross sections for the materials in the drawer were used to generate the neutron flux and the neutron-energy spectrum at various points within a drawer.¹³ The results of this calculation are shown in Fig. 8 for a drawer which contains a single column of 0.25"-wide plutonium fuel plates. Along the bottom of Fig. 8 is a cross section of the drawer showing the locations of the various materials in the drawer. The drawer is 2" wide. The open circles show the normalized total neutron flux as a function of position within the drawer. The solid circles show the normalized average energy of the neutrons as a function of position. As indicated, the total neutron flux has a slight maximum at the position of the fuel plate, and the average neutron energy also has a maximum at the fuel plate. The gradient in the total flux is quite small--only a 1.75% difference between maximum and minimum flux--while the gradient in average energy is 10% between maximum and minimum.

Fortunately, fast-neutron track recorders are sensitive to both flux and energy gradients. Etch pit production in plastics is a threshold process.¹¹ The minimum neutron energy required to form an etchable track is different for different materials and was reported to be 1.2 MeV for Makrofol.¹¹ The probability of neutron-induced recoil track formation increases with increasing neutron energy, although the amount

Fig. 8. Neutron flux and energy gradients in a ZPPR drawer.

of experimental data is limited.¹⁴ Table I shows that, for the materials studied here, the production of etch pits was much lower with the low-energy Am-Li neutron source compared to the fission-spectrum Cf-252 source. As mentioned earlier, the number of etchable tracks increases with increasing neutron flux for constant neutron energy. Fuel-column imaging should be possible in assembly drawers if the fast-neutron flux and energy gradients are large enough to provide a visible gradient in etch-pit density on the plastic track recorders.

An example of a fast-neutron-induced fuel-column image is shown in Fig. 9. Shown is a piece of CAB, approximately 1.5" wide, 2" long, and 0.005" thick, which was irradiated in a 1500 watt-hour run in ZPPR (5×10^{13} neutrons cm^{-2}). The sample was etched in 6.5-n KOH at 65° for approximately 3 hours. The very pale line in the center of the CAB sample corresponds to the position of the fuel column and is attributed to the fast-neutron flux and energy gradient. The image is quite faint and was only observable when held in front of a dark background with bright side illumination. The visibility of the image was attributed to light scattering from the etch pits. Similar images were observed after identical irradiations of CTA and Lexan.

Attempts were made to correlate the fuel-column image with a maximum in the etch-pit density. Etch-pit densities were measured by counting the pits in a known area on a photomicrograph of an etched surface. Unfortunately, except for CR-39, pit counting was very difficult because of the rough background of the etched materials. The uncertainty in any pit density was large, possibly $\pm 20\%$. In some samples there was a

Fig. 9. Fuel-column image in CAB.

trend toward higher pit density at the fuel column, but there were also samples which showed no significant trend across the recording plastic. Because the neutron flux and energy gradients in the drawer were expected to be small, and because the uncertainties in the pit densities were large, it was not surprising that there was no clear maximum in pit density at the fuel-column.

Because track recorders are only sensitive to fast neutrons, and because the resulting pit density is proportional to the neutron flux, it is theoretically possible to use track recorders as fast-neutron flux monitors in an assembly core. Some track recorders are currently in use as fast-neutron dosimeters in radiation monitoring applications.⁸ However, the uncertainty in the pit densities was quite large, and the counting procedure was quite cumbersome, so that the use of track recorders as flux monitors was not considered practical.

V. FOIL-IMAGING TECHNIQUES

Metal foils can record the passage of neutrons by the formation of radioactive, neutron-induced reaction products. The concentration of reaction products depends on both the neutron flux and the neutron energy. The concentration of reaction products can be determined by measuring the intensity of radiation, normally gamma, from the foil. Gradients in neutron flux and neutron energy result in gradients in the concentration of reaction products in the detector foils. Thus, an irradiated metal foil can be considered analogous to an undeveloped photographic film or to a track recorded before etching. In order for the latent image on the foil to become visible to the eye, the image must be transferred to some other medium. Foil-image transfer techniques incorporating X-ray sensitive film have been used in thermal neutron radiography.¹⁵ Alternatively, a concentration map for a specific reaction product can be generated by counting the gamma rays emitted from different regions of the foil with gamma-ray spectroscopic techniques.

Again, because we were interested in techniques applicable to fast critical assemblies, we considered fast-neutron-induced reactions. Such reactions include the types $(n,2n)$, (n,p) , (n,n') , and for some materials (n,f) . Indium was the material chosen for imaging the fast neutron flux and energy gradients in a fast critical assembly fuel drawer. Natural indium contains approximately 96% In-115 which undergoes a high-energy-induced (n,n') reaction. The reaction product, In-115m, has a 4.5-hour half-life and emits a 335 keV gamma ray. The (n,n') reaction cross section increases with increasing neutron energy up to 5 MeV. In addition

to the (n,n') reaction, In-115 undergoes an (n,γ) reaction to produce In-116m₁ which has a 54-minute half-life and emits several high-energy gamma rays. The cross section of the (n,γ) reaction decreases with increasing neutron energy. The (n,γ) reaction cross section is larger than the (n,n') cross section for most of the neutrons in a typical fast critical assembly spectrum. (The neutron-energy dependence of the cross sections of the In-115 (n,n') and (n,γ) reactions, and the neutron-energy spectrum of a typical ZPPR core are shown in Figs. 24 and 23, respectively, in Part II.) Because the (n,n') and (n,γ) reactions occur simultaneously, the In-115m concentration profile (the image) will be superimposed on a background of In-116m₁.

The ability of indium foil to form a fuel-column image was tested in a 1500 watt-hour irradiation in the ZPPR assembly. An indium foil, 0.010" thick, 1.875" wide, and 1" long, was used. The foil was placed in a drawer with the same loading pattern as that shown in Fig. 8, in which there was a single fuel column in the drawer center. The foil was inserted between the top of the drawer contents and the matrix tube. After removal from the assembly, the foil was cut into 0.125" wide pieces. A Ge(Li) spectrometer system with automatic sample changer was used to count the activity in the 15 individual pieces of foil. The 335-keV gamma ray from In-115m, and the 417-keV gamma ray from In-116m₁ were counted. Each piece was counted several times. Total counting time was approximately 20 hours. Counts were corrected for background and for decay from the time of assembly shutdown. After counting, each piece of foil was weighed. Figure 10 shows the variation in In-115m activity as a function of foil position in the drawer. Error bars indicate the 1 σ

Fig. 10. Variation in In-115m activity as a function of position in a ZPPR drawer.

uncertainties from counting statistics. As indicated, there is a small maximum in the In-115 activity near the drawer center. This activity profile is approximately that expected from the neutron flux and neutron energy profiles in Fig. 8. The profile of In-116m₁ activity showed no statistically significant variation from a horizontal line.

An attempt to transfer fuel-plate images from other irradiated foils to X-ray film was not successful. The induced activity level was quite low, and long exposure times were required. Because most of the activity in the foil was from the In-116m₁, which showed no significant spatial variation, the faint foil image that was obtained on film had no visible density gradient.

The irradiated foil was cut into pieces for counting because experiments on samples irradiated earlier indicated that a collimated detector could not provide both the spatial resolution and counting rates required.

VI. DISCUSSION

The strength of the autoradiographic method is its ability to provide a piece count and simultaneous attribute check for a very large number of fast critical assembly fuel elements in a short time and with very little handling of fuel elements. In the techniques described for autoradiographic verification of fuel in storage,^{3,5} fuel element containers were not removed from their vault locations, and the containers were not opened. In the techniques described for autoradiographic verification of fuel in assembly cores, the fuel drawers were not removed from the assembly matrix. These techniques allowed inventory verifications to be performed with minimum impact on facility operations and with minimum radiation exposure to personnel. The greatest disadvantage of the autoradiographic method is that it is not quantitative in the sense that the mass of fissile material is not directly obtained when the image of a fuel element is recorded on film. The primary problem to be resolved is how to incorporate the advantages of the autoradiographic method into a total inventory verification system for a fast critical assembly facility.

The important considerations when selecting techniques to be incorporated into an inventory verification system are the capabilities and limitations of the individual techniques, the way in which individual techniques complement one another, and the credible diversion scenarios against which the system must defend. The consideration of credible diversion scenarios is important when deciding the role of autoradiography. If the potential diverters are considered capable of fabricating "dummy" radioactive fuel elements which produce images on film similar to the

images produced by fissile material-containing elements, then autoradiography should be supported by other techniques. Passive autoradiography is more susceptible to defeat by "dummy" elements than is active autoradiography in which image formation depends on the fissile content of fuel elements and on the presence of a critical mass in the assembly. The credibility of fabricating "dummy" fuel elements is low in the case of individual diverters, but may be possible for institutional diverters. For fuel in storage, where only passive autoradiography is possible, some fraction of the storage containers examined by autoradiography should be selected for gamma-ray analysis or should be subject to active or passive neutron-counting measurements. In the case of plutonium-fueled assembly cores, passive autoradiography can be supported by reactivity measurements or some other verification technique which depends on the critical operation of the assembly. The combination of passive autoradiography and foil irradiation is potentially a very effective verification scheme with low facility impact. It is important to keep in mind that the existence of a postulated scenario for defeating an autoradiographic verification does not mean that autoradiography is not a useful technique; it only means that autoradiography should be used in combination with other techniques which can easily detect diversion by the postulated scenario.

Active autoradiography provides a piece-count verification which depends directly on the fissile content of the fuel elements. Active autoradiography is only applicable to fuel in assembly cores. In addition, the formation of an active autoradiographic image is contingent upon the achievement of criticality and thus provides an assurance that an assembly

core contains at least a critical mass for its particular composition and configuration. For fast critical assemblies, this is equivalent to verifying that the core contains at least 99% of its fissile material, because these assemblies normally contain less than 1% excess fissile material. The film itself may have a positive reactivity effect that must be considered when interpreting the results of an active autoradiographic inventory.

Active autoradiography appears to be a very promising technique for verification of the contents of a uranium-fueled assembly core. A powerful combination of techniques for a uranium-fueled core may be active autoradiography plus foil irradiation. A foil irradiation and subsequent gamma-ray counting assures that the assembly has achieved criticality, and the foils can be used to monitor neutron flux at various core locations. In addition, foils can also serve as spectral indices to assure that the neutron-energy spectrum is that expected for the stated assembly composition. One foil useful as a spectral index is indium. The spectral index in this case is the ratio of the In-115 (n, γ) reaction rate to the In-115 (n, n') reaction rate. Spectral-index measurements are discussed more thoroughly in Part II.

In the case of uranium-fueled cores, autoradiographic verification will be by fuel-column length rather than by plate count, because individual plates are not generally distinguishable.

Passive autoradiography is not presently practical for in-core uranium applications. Further work is needed to explore the possibility

of adapting existing passive autoradiographic techniques for 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 densities 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, at present, the passive autoradiographic technique followed by a foil irradiation measurement can provide a very effective inventory verification for a plutonium-fueled core.

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') reactions are produced in sufficient concentrations with an approximately 100 watt-hour irradiation of the ZPPR. The 100 watt-hour irradiation is recommended for active autoradiography in a core with a volume of 6,100 liters. The simultaneous foil irradiation and film exposure is expected to be applicable to uranium-fueled cores.

If reactivity measurements are to be applied as an inspection technique for fast critical assemblies, then autoradiography will be a required companion technique. Autoradiography will be needed to verify the loading

pattern of the core. In addition, foil irradiation will be required to verify the core composition through spectral-index measurements. In the case of uranium-fueled cores, the core-loading verification will necessarily be with active autoradiography. In the case of plutonium-fueled cores, the core-loading verification can be made with either active or passive autoradiography. The passive technique will probably prove somewhat simpler.

Although of some academic interest, neither the foil-imaging technique nor the track-etch imaging technique is practical for inspector use in obtaining images of fuel columns. The foil-imaging technique is much too slow and the spatial resolution is poor. As mentioned earlier, foil irradiation techniques do play an important role in fast critical assembly safeguards and will be discussed more fully in Part II. The spatial resolution of the track-etch images is also poor, and the etching process requires the use of corrosive solutions. Track-etch techniques are probably not as useful as foil irradiation techniques for monitoring the neutron flux or neutron energy spectrum because pit counting is less convenient and less precise than gamma-ray counting.

PART II

REACTIVITY AND SPECTRAL INDEX MEASUREMENTS

by

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I. INTRODUCTION

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. One potentially powerful assay method is the measurement of integral (whole core) reactivity. A variety of reactivity measurement methods have been developed which are applicable to fast critical assemblies.¹⁶⁻¹⁸ The potential applicability of reactivity measurements for safeguards purposes will be discussed in the following sections.

II. INTEGRAL REACTIVITY MEASUREMENTS

A. The Definition of Reactivity

The ability of a reactor or critical assembly to sustain a neutron

chain reaction is characterized by the multiplication factor, k , defined as the ratio of the number of neutrons in any one generation to the number of corresponding neutrons in the immediately preceding generation. The value of k determines the behavior of the neutron population within the assembly. If k (associated with both prompt and delayed neutrons) is equal to unity, a chain reaction will be exactly maintained, and the neutron population will remain constant with time. Such a system is said to be at delayed critical. If $k < 1$, the system is said to be subcritical, and any neutron-induced chain reaction will eventually die out. If $k > 1$, the assembly is said to be supercritical, and the neutron population will rise exponentially with time.

A quantity called reactivity is used to measure the deviation of the multiplication factor from its critical value $k = 1$. The reactivity, ρ , is defined as $\rho = \frac{k - 1}{k}$. Changes in a reactor due to a system perturbation, such as the addition or removal of fuel, are measured in terms of a change in reactivity. Two common units of reactivity are the inhour (Ih) and the dollar (\$). The inhour is defined as the amount of reactivity required to produce a reactor period of one hour. The reactor period is the time required for the neutron population to increase by a factor of e . The dollar measures reactivity in units of the delayed neutron fraction β . That is, $\rho = \beta$ is one dollar of reactivity, corresponding to a prompt critical condition. The calculation of reactivities in these units requires knowledge of the delayed neutrons from fission and/or the reactor average neutron lifetime and effective neutron-source strength. For typical plutonium-fueled cores presently being studied at ZPPR, a 1% change in k is equal to about 3\$, or about 1000 Ih.

B. Reactivity Measurement Methods

Several methods are available for determining the reactivity of a fast critical assembly.¹⁷ 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.^{17,18,19} 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. Results of source-multiplication measurements on ZPPR will be reported in subsequent sections.

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.¹⁶ For a supercritical reactor at steady state, 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 most 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.

C. 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 (see Part I), or by calculation of the reactivity expected for a particular core configuration and composition.

D. 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., $\Delta k/k$), 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.

E. Reactivity Compensation

The agreement between two reactivity values for a core in a reference configuration still does not necessarily mean that the fissile content has not changed. The loss of reactivity by removal of fissile material can, in principle, be compensated for by addition of materials which 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.

III. EXPERIMENTAL RESULTS

A. Reactivity Worths

As a first step in assessing the sensitivity of reactivity measurements to diversion detection, reactivity worths were measured for Pu-239, U-235, and U-233 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. 11. The assembly was a hexagonal 4590-liter mock-up of an LMFBR which contained 19 mock-up control-rod positions. The core had an axial length of 40" and was divided into two fuel-enrichment zones. The inner core, with the mock-up 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 2" by 2" cross section of the core zones of a single-column drawer and a double-column drawer are shown in Fig. 12. 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 chamber reactivity effect of the holder. Neutron flux measurements were made with BF_3 detectors located outside the core.

Fig. 11. Core configuration for ZPPR 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. 12 Cross-section views of single- and double-column fuel drawers.

In addition to the experimental results, first-order perturbation reactivity worths for the isotopes Pu-239, Pu-240, U-235, U-238, and U-233 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, U-235, and U-233 reactivity worths as a function of radial position at the centerline of the core axial midplane are presented in Tables II-IV. The uncertainties in the measured worths are 1σ statistical variations. Graphical results are displayed in Figs. 13-15. The squares in Figs. 13-15 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

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

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

core edge

TABLE III. Calculated and Measured Radial Traverse for U-6,
Sample in ZPPR Assembly 10A

Position, cm	Sample Worth, Ih/kg				
	Experiment	Calculated	Self- Shielding	Corrected Calculated	C/E
0.025	42.13 + 0.05				
5.527	42.34 + 0.09	54.50	0.96	52.14	1.23
11.049	43.40 + 0.10	53.89	0.97	52.07	1.20
16.576	43.44 + 0.08	53.50	0.97	52.08	1.20
22.098	43.13 + 0.08	53.10	0.98	51.85	1.20
27.622	42.98 + 0.07	52.42	0.98	51.22	1.19
33.144	42.19 + 0.07	51.44	0.98	50.27	1.19
38.669	41.22 + 0.07	50.18	0.98	49.10	1.19
44.199	40.61 + 0.10	48.83	0.98	47.89	1.18
49.720	39.51 + 0.09	48.07	0.98	47.07	1.19
55.245	38.66 + 0.09	47.10	0.98	46.09	1.19
60.772	37.41 + 0.09	45.54	0.98	44.66	1.19
66.291	35.76 + 0.08	43.26	0.98	42.59	1.19
71.818	34.05 + 0.08	41.62	0.98	40.71	1.20
77.343	31.66 + 0.07	39.31	0.97	38.15	1.20
82.870	29.21 + 0.10	37.18	0.96	35.80	1.23
88.392	27.20 + 0.07	35.12	0.96	33.81	1.24
93.916	25.55 + 0.07	32.75	0.97	31.74	1.24
99.441	23.93 + 0.09	29.35	0.99	28.91	1.21
104.968	21.36 + 0.08	25.66	0.99	25.47	1.19
110.490	17.98 + 0.08	21.39	1.00	21.32	1.19
116.012	14.20 + 0.09	16.98	0.99	16.86	1.19
121.542	10.93 + 0.09	12.73	0.98	12.50	1.14
core edge					
127.061	7.90 + 0.09	9.40	0.94	8.86	1.12
132.591	5.20 + 0.09	6.29	0.90	5.66	1.09
138.118	3.29 + 0.07	3.99	0.85	3.41	1.04
143.637	1.66 + 0.08	2.54	0.79	2.02	1.22
176.784	0.00 + 0.03	0.00	0.60	0.00	

TABLE IV. Calculated and Measured Radial Traverse for WR-2,
Sample in ZPPR Assembly 10A

Position, cm	Sample Worth, Ih/kg				
	Experiment	Calculated	Self- Shielding	Corrected Calculated	C/E
0.025	82.85 $\pm$ 0.26				
5.522	82.49 $\pm$ 0.23	98.63	0.99	97.69	1.18
11.054	82.91 $\pm$ 0.32	97.14	0.99	96.51	1.16
16.573	82.95 $\pm$ 0.26	96.30	1.00	95.88	1.16
22.098	82.78 $\pm$ 0.33	95.56	1.00	95.23	1.15
27.625	81.22 $\pm$ 0.30	94.35	1.00	94.04	1.16
33.147	80.89 $\pm$ 0.29	92.65	1.00	92.35	1.14
38.674	78.23 $\pm$ 0.27	90.56	1.00	90.29	1.15
44.196	77.27 $\pm$ 0.33	88.34	1.00	88.12	1.14
49.723	74.45 $\pm$ 0.32	86.80	1.00	86.55	1.16
55.245	73.41 $\pm$ 0.31	84.92	1.00	84.67	1.15
60.775	70.81 $\pm$ 0.33	82.22	1.00	82.02	1.16
66.291	67.64 $\pm$ 0.27	78.40	1.00	78.28	1.16
71.821	64.87 $\pm$ 0.32	75.15	1.00	74.91	1.15
77.343	61.45 $\pm$ 0.31	70.98	0.99	70.60	1.15
82.870	57.37 $\pm$ 0.28	67.49	0.99	66.95	1.17
88.392	52.91 $\pm$ 0.28	64.03	0.99	63.52	1.20
93.919	50.85 $\pm$ 0.35	59.91	0.99	59.53	1.17
99.441	45.68 $\pm$ 0.25	53.74	1.00	53.64	1.17
104.968	40.33 $\pm$ 0.30	46.81	1.00	46.83	1.16
110.490	34.06 $\pm$ 0.32	38.91	1.00	38.97	1.14
116.017	26.01 $\pm$ 0.26	30.58	1.00	30.60	1.18
121.539	20.00 $\pm$ 0.34	22.53	1.00	22.48	1.12
core edge					
127.061	14.32 $\pm$ 0.29	15.91	0.99	15.71	1.10
132.591	8.50 $\pm$ 0.28	10.25	0.98	10.00	1.18
138.115	6.44 $\pm$ 0.24	6.29	0.96	6.05	0.94
143.637	2.35 $\pm$ 0.25	3.94	0.93	3.68	1.57
176.784	0.00 $\pm$ 0.13	0.00	0.80	0.00

Fig. 13. Pu-239 reactivity worth as a function of position in core.

Fig. 14. U-235 reactivity worth as a function of position in core.

Fig. 15. U-233 reactivity worth as a function of position in core.

the core composition, particularly the decay of Pu-241. The temperature of the ZPPR cell can be held constant to within $\pm 0.25^\circ\text{F}$. 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.

These short- and long-term uncertainties can be combined with the reactivity worth results in Tables II-IV to obtain estimates of the fissile removal sensitivities. These sensitivities are presented in Table V 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

TABLE V. Fissile Removal Detection Sensitivity - ZPPR-10A
(Using 1 Ih short-term and 3 Ih 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

the core edge.

B. 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 10-D. This assembly was a hexagonal 6100-liter mock-up of an LMFBR which contained 31 mock-up control-rod positions. Though larger than the core in assembly 10-A, the loading pattern was similar. Reference core 10-D contained a total fissile mass of approximately 2600 kg.

The reactivity of the subcritical 10-D 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, 2" by 1/4" by 6". The fission counters took the place of the fuel plate nearest the axial midplane in 32 different single-column fuel drawers in each reactor half. Reference Assembly 10-D is shown in Fig. 16. 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. The control-rod positions contained only sodium and stainless steel. 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

Fig. 16. ZPPR-10D core configuration showing locations of fission counters.

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 reference 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. 17. 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. 17 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

$$Q = 1000 \left[\frac{\frac{a_i R_i}{\sum_i a_i R_i}}{\left(\frac{a_i R_i}{\sum_i a_i R_i} \right)_{\text{ref}}} - 1 \right]$$

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 Q values in Fig. 17, 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 II gives a plutonium reactivity worth near the core center of ZPPR 10-A of approximately 56 Ih/kg. Using a conversion factor of 333 Ih/\$,

Fig. 17. Fission-rate distribution map for the first simulated diversion.

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 10-D was 0.16\$. This smaller reactivity decrease is expected since fuel was removed from the entire 20" fuel column, and since the reactivity worth of fuel decreases with increasing distance from the axial midplane. Axial worths at 20" 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. 18. 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.

Fig. 18. Fission-rate distribution map for the second simulated diversion.

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. 19. The integral reactivity was 0.02\$ less than the reference. This reactivity reduction is in reasonable agreement with the results of Table II 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 counter's high sensitivity to low-energy neutrons and to the lack of intervening absorber. In the second diversion, the nearest fission detector was four drawers away from the polyethylene. The other fission counters show a negative tilt toward the side where the fuel was

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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 one double-column drawer, and polyethylene plates were substituted for the fuel. Total fissile material removed was 2.5 kg. Figure 20 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. 21. 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.

Fig. 20. Fission-rate distribution map for the fourth simulated diversion.

Fig. 21. Fission-rate distribution map for the fifth simulated diversion.

Figure 22 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.

C. Foil-Irradiation Experiments

Because the reactivity of a particular assembly-core configuration depends on the neutron-energy spectrum, it may 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 mock-up is shown in Fig. 23. 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

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Fig. 23. Typical ZPPR neutron-energy spectrum.

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.^{2C} 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-116m, has a 54-minute half life and emits several gamma rays, including one at 417 keV. The product of the (n,n') reaction, In-115m, has a 4.5-hour half life and emits a single gamma ray of 335 keV. The ratio of the intensities of the 417-keV and the 335-keV gamma rays was 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. 24.

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.

Fig. 24. Cross sections of the In-115 (n, γ) and In-115 (n, n') reactions as a function of neutron energy.

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 25 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 contained an indium foil.

The indium foils were 0.5" in diameter and 0.005" thick. The foils were held in 0.020"-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 3" from the axial midplane. Except for the foils, the core loading was identical to that used in the reactivity measurements. 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 is approximately 8×10^9 neutrons $\text{cm}^{-2} \text{sec}^{-1}$. After about a 2-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

Fig. 25. Indium foil-loading pattern.

shown in Fig. 26. 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 (1σ) 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. 27. 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 (1σ) 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

Fig. 26. Spectral-index values near the core center.

Fig. 27. Spectral-index values near the core edge.

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.

IV. DISCUSSION

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 measurements 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 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.

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). The 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 showed a much higher relative fission rate than it did in the reference case. The 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. Because of the relatively small amount of absorber material between the polyethylene and the fission counters, these low-energy neutrons were not absorbed. Fission counters four drawers removed from polyethylene substituted for fuel showed reduced relative fission rates. The 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^{21,22} 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 subcrit-

ical 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.

As presented in this report, evaluation 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.

Another difficulty faced by international inspectors making reac-

tivity measurements is the assurance that a reactor core under inspection is indeed in its reference configuration. This verification of configuration presents a problem both at the initial formation of the reference core and at the reestablishment of the reference core for a verification measurement. The core is first vulnerable to diversion during its loading phase. Inspectors must verify that the fissile content is as stated. This can be accomplished by observing fuel-element loading and randomly selecting drawers for nondestructive assay. Inspectors must also verify the core geometry. Autoradiographic methods can be used to verify the loading pattern (see Part I). Finally, inspectors should verify that the reactivity of the reference core is reasonable for its composition and geometry. Calculations can be used to provide an estimate of reactivity. Assurance is also required that a reactor core has been returned to its reference configuration when a reactivity verification measurement is made. Verification of correct loading patterns can be obtained by using autoradiography. Autoradiography can be particularly helpful in verifying the core loading in low-reactivity-worth areas near the core-blanket boundary. Verification of control-rod positions will be very difficult. In order to collect and interpret reactivity data, it will be necessary for inspectors to thoroughly understand the reactivity measurement techniques and to be familiar with the operational details of the facilities under inspection.

A technique which is relatively independent of the facility is the foil-irradiation method for obtaining spectral indices. 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 foils used in these experiments required only 200 watt-hours irradiation to produce sufficient reaction products for satisfactory counting statistics. After a 200 watt-hour run and about a 2-hour cooling period, the fission-product concentration in the fuel was low enough to allow personnel to enter.

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.

Gryazev²⁰ 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 (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 $\sim$ 5-cm wide). Gryazev also used spectral-index results to estimate the magnitude of simulated diversions.

The foil-irradiation technique has one additional significant attribute. The technique can be used as a qualitative reactivity test. The neutron-induced reactions will only occur if the reactor has enough excess reactivity to operate at a power of a few hundred watts. The foil-irradiation reactivity test will not be as sensitive to fuel removal as quantitative reactivity measurements. However, most fast critical assemblies contain only small excess reactivity--normally less than 1% of their critical mass. Thus the detection of the neutron-induced reaction products provides assurance that the core contained 99% of its stated critical mass. The validity of the foil-activation test is subject to the same requirements concerning the reestablishment of the reference configuration as is the quantitative reactivity measurement. The foil-irradiation method, of course, has the built-in test for moderator substitution. As in the case of quantitative reactivity measurements, the core-loading pattern can be established with the use of autoradiography. A combination of foil-activation and autoradiography methods has the advantage of being completely independent of facility-supplied equipment.

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

in 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 Gryazev²¹ 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.

V. CONCLUSIONS

The integral reactivity of a fast critical assembly core is very sensitive to changes in the fissile content of that core. Fissile removals of 2 kg from a core containing 2600 kg fissile material can be detected even when removal is from a region of low worth. The general area of a local diversion can be indicated by relative-fission-rate maps. The measurement of integral reactivity alone is not sufficient to assure that the fissile content has not deviated from some reference value because reactivity losses can be compensated for by moderator addition and/or absorber removal. Precise reactivity compensation in large, multicomponent assemblies, such as ZPPR, is probably feasible, but is not a simple task. Compensation by moderator addition alters the neutron-energy spectrum in the core. Spectral indices can be used to detect spectral changes. A spectral index, the ratio of the rate of the In-115 (n, γ) reaction to the rate of the In-115 (n,n') reaction, was able to detect polyethylene at distances of 10-15 cm from the polyethylene. In addition to reactivity measurements and spectrum verification measurements, assurance is required that the core is in its reference configuration. While technically sound, reactivity measurements made by inspectors may be less credible if facility instruments are used.

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