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AUTORADIOGRAPHIC INVENTORY METHODS

by

S. B. Brumbach and R. B. Perry

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ABSTRACT

Described are autoradiographic techniques which can verify the number and SNM content of plutonium- and uranium-containing fuel elements. These techniques are applied to fast critical assembly fuel elements. These techniques are applied to fast critical assembly fuel and to low-enriched uranium in LWR fuel assemblies. Autoradiographic images are formed by the spontaneously emitted X- and gamma-rays from the fuel elements striking X-ray film in contact with the fuel elements or their containers. Autoradiography allows a large number of items to be examined in a minimum inspection time and with minimum facility impact. Results are presented for fast critical assembly fuel in a variety of storage modes as well as in fast critical assemblies themselves. Results are also presented for low-enriched uranium rods in unirradiated LWR fuel assemblies. In all cases, missing fuel elements or substitution of elements containing inert material or depleted uranium were detected.

I. INTRODUCTION

Effective safeguards require frequent verification of special nuclear material (SNM) inventories. Inventory techniques are needed which can verify the piece count and can provide an SNM attribute check for large numbers of SNM-containing items while minimizing inventory time, personnel radiation exposure, and interference with normal operations of the facility under inspection. Such inventory techniques are particularly important at power reactors, fuel-fabrication facilities, and fast critical assemblies. One method which addresses these inventory requirements is autoradiography. In this method, spontaneously emitted radiation from the various isotopes in urnaium- and plutonium-containing fuels exposes film in contact with fuel elements or their containers. With favorable source-film geometry, clear images of individual SNM-containing elements can be obtained *in situ*, without handling fuel elements or opening containers. Counting the images on the film provides a piece count of fuel elements in containers or arrays. Visual inspection of the image provides an attribute check that the fuel element contains SNM. Autoradiography is a qualitative, rather than a quantitative technique, and does not provide a direct measure of the mass of fissil material contained in a fuel element. Autoradiography does provide an indication that a fuel element contains the expected concentration of radioactive material and that the emitted radiation qualitatively duplicates that from SNM-containing fuel elements. Recently, autoradiographic methods have been applied to inventory verifications of plutonium-containing fast critical assembly fuels.[1] The technique has also been applied to uranium in LWR fuel assemblies.[2,3]

II. APPLICATIONS TO PLUTONIUM

A. General

The photon radiation from the various isotopes in plutonium fuels can efficiently expose X-ray film. The photon energy spectrum from most plutonium fuels is dominated by the 60 KeV gamma-ray from Am-241. Because the principal fissil isotope, Pu-239, contributes only a minor part of the total photon flux, autoradiographic verifications of fissile content are indirect.

B. Fast Critical Assembly Fuel in Storage

The autoradiographic method has been sucessfully applied to inventories of plutonium-containing fuel plates at fast critical assemblies operated by the Argonne National Laboratory. Most of the fuel plates in the Argonne inventory contained an alloy of plutonium, depleted-uranium, and molybdenum. The plates were typically 5.08-cm wide, 0.64-cm thick, and from 2.54- to 20.32-cm long; they were clad in stainless steel, and were stored in two types of aluminum canisters. Fuel plates at the Argonne-Idaho site were stored on their edges in flat, rectangular canisters. Plates were separated by aluminum posts. Autoradiographs of the contents of a sealed canister were obtained by simply placing a cassette containing X-ray film under the canister. The film was exposed, removed and finally processed. Since the film can be quickly placed and removed, many autoradiographs can be obtained simultaneously. For Kodak Industrex-AA film and plutonium containing 11.5% Pu-240, exposures of 45 minutes were adequate. Shorter exposures were used for material containing larger fractions of

Pu-240. Fuel plates in the canister were easily counted by the counting of their images on film. Missing plates or the substitution of plates made of steel, lead, or uranium were easily detected. Figure 1 shows an autoradiograph of a canister containing 14 plates, each 10.1-cm long. Thirteen plates contained plutonium, while the plate at the center right contained only stainless steel. An image of the stainless steel plate was formed by scattered radiation from adjacent plates, but it is readily distinguished from the plutonium plates. Autoradiographic images were also used to verify plate length. A missing element in an array of plates was easily detected.

The contents of sealed fuel-plate storage canisters at the Argonne-Illinois site were also verified by autoradiography. The Illinois canisters had cylindrical geometry, and autoradiographs were obtained by wrapping X-ray film cassettes around the outer circumference of the canisters. Plates in the canisters could be counted, and missing plates, or substituted inert material or uranium could be readily detected.

The Proteus critical assembly at the Reactor Research Institute in Switzerland uses mixed-plutonium- and depleted-uranium-oxide fuel rods in its fast core. These rods are 0.67 cm in diameter, 5.6-cm long, and are clad in 0.01-cm thick aluminum. Aluminum block drawers, drilled with a square array of holes to accommodate the rods, are used for storage. A sheet of X-ray film placed under a storage drawer showed autoradiographic images of the individual fuel rods, as seen in Fig. 2. The film used was Kodak AA, and exposure was for 7 minutes for the 17% Pu-240 material. The pale image in the fifth row from the bottom of Fig. 2 is from a rod containing depleted uranium, which has a much lower photon emission rate than does a rod containing plutonium. A void rod position was characterized by a very faint circular image due to radiation from neighboring rods. Void positions were easily detected by visual inspection.

It is more difficult to interpret autoradiographs when the fuel elements are stored very close together, as is done at some facilities. An autoradiograph of a stack of plates is shown in Fig. 3. The plates contained a 99% Pu, 1% Al alloy and were clad in stainless steel. They were 5.08-cm wide, 7.62-cm long, and 0.32-cm thick. An 0.1-cm thick sheet of stainless steel between the plates and the film simulated a storage container wall. One feature of Fig. 3 common to all autoradiographs of thin plates is the greater optical density of the film around the plates compared to the optical density of the film directly under the plate edge. This feature is attributed to the greater surface area of a side of a plate compared to the surface area of an edge. This effect was much less important for fuel plates 0.64-cm thick. The autoradiograph in Fig. 3 shows a stack of 8 plutonium-containing plates, with one depleted-uranium plate in the center. The depleted-uranium plate was easily detected by its less dense image and by the reduced density of the images of the adjacent plutonium plates. Voids in plate stacks were also easily detected. Because of the small surface area of the plate edges, the exposure time for the autoradiograph in Fig. 3 was 4 hours.

Autoradiography cannot be used for piece-count verification if the fuel-plate storage containers are made of thick steel. Attempts were made to obtain images of a stack of plates through 1.2 cm of steel. The gamma-ray attenuation resulted in long exposures, while the combination of large source-film distance and the large amount of scattering in the

steel resulted in very diffuse images. Individual plates could not be distinguished.

C. *In Situ* Verification of Fuel in Assembly Cores

A large fraction of the SNM inventory at a fast critical assembly is normally contained in the assembly core. Verification of this in-core material is an important part of fast critical assembly safeguards. Autoradiography has been successfully applied to in-core verification at the split-table type assemblies operated by Argonne National Laboratory.

The Argonne assemblies consist of pairs of arrays of fuel-containing drawers held in place by a steel matrix. Fuel plates are loaded into a drawer on their edges, normally forming one or two columns. The remaining space in the drawers is filled with such diluent materials as depleted uranium, sodium, graphite, stainless steel, iron oxide, or aluminum. Autoradiographs of the contents of an assembly drawer were obtained by slipping an X-ray film packet between the top of the contents of a drawer and the steel matrix. Exposures of 15 minutes were adequate with Kodak-AA film for 0.64-cm plates and plutonium containing 11.5% Pu-240.

Examples of autoradiographs of fuel plates in reactor drawers are shown in Fig. 4. The autoradiograph on the top shows six plutonium-containing plates, each 7.62-cm long and 0.32-cm wide, arranged in two rows. Kodak-AA film and approximately one-hour exposures were used. The plate images are characterized by well-defined outlines of the plates resulting from scattering by the diluent material adjacent to the plutonium-containing plates. When a plate made of uranium or inert material was substituted for a plutonium plate, the dummy plate formed virtually no image and was immediately obvious. Less obvious, but still detectable, was a missing plate. An image of a void is shown in the center of the lower row of the autoradiograph on the bottom of Fig. 4. The void does not have the sharp outline characteristic of plutonium plates. An image is formed because, it it believed, the void acts like a collimator for radiation from the neighboring plates.

In addition to split-table type fast critical assemblies, some vertical assemblies are also in use. In contrast to the split-table assemblies in which fuel in the core is readily accessible for autoradiographic verification, the in-core fuel elements of a vertical assembly are inaccessible to interrogation by film. Access from the sides of the assembly is not possible because of blanket materials. Access through the top or bottom of the assembly is normally blocked by horizontal positioning plates which support the vertical fuel elements.

D. Large-Scale Inventory Experience

Autoradiographic methods have been applied to several large-scale inventory verifications at the Argonne fast critical assembly facilities. Two inventories of the plutonium fuel at the ZPPR storage vault in Idaho were conducted with the use of autoradiography. On the more recent occasion, the contents of 800 storage canisters were verified. One day was required for film exposure, one day for film processing, and one day for film analysis. The maximum radiation exposure to any person participating in the verification was 50 mr. Two autoradiographic inventory

verifications have also been conducted at the Argonne-Illinois site at the storage vault for the ZPR-6 and ZPR-9 assemblies. On one occasion, 172 fuel-plate canisters were verified with a total of four man-days of effort. Radiation exposure was 160 mr for the one individual who obtained the autoradiographs. A full-scale, *in situ* inventory verification was also performed for the core of the ZPR-9 assembly. Approximately 400 fuel drawers were examined, requiring two people working in the reactor cell for one shift. Radiation dose to each of those people was about 80 mr. Another man-day was required to process the film. In all cases, these autoradiographic inventories resulted in significant reductions in facility interruption, in total effort, and in radiation exposure when compared to previous inventory methods.

E. Discussion

Autoradiography provides a simultaneous piece count and attribute check for plutonium-containing fuel elements. The technique is not quantitative and does not provide a direct measure of the mass of fissile plutonium. It does provide assurance that an item of the approriate size, shape, and location contains radioactive material with the specific activity of some reference material.

Autoradiography can also provide a verification that the energy distribution of the radiation forming the image is qualitatively like that of the reference material. This energy-distribution verification can be improved by the use of thin absorber foils placed between the film and the radiation source. The ratio of the image density with and without the foil depends on the photon energy profile of the sample radiation. As an example, by using 0.05-cm thick copper foil, 0.05-cm thick lead foil, and 0.025-cm thick cadmium foil, it was very easy to distinguish between images formed by plutonium-containing fuel elements and images formed by the higher-energy gamma radiation from Co-60 or Ir-192 sources. In addition, the image-density ratio method was able to distinguish between fuel elements containing 11.5% Pu-240 and elements containing 27% Pu-240.

The specific activity of the radiation source can be assured by using an exposure calibration source when making inventory-verification exposures. Such a calibration requires that a small source of known activity, which produces a known image density for a specific exposure time, be attached to a piece of film used in an inventory. After film processing, this reference image can give assurance that film sensitivity and film-processing conditions were correct, and that the fuel elements under investigation had the expected activity.

Because of the qualitative nature of autoradiographic methods, autoradiography should be supported by quantitative, nondestructive assay (NDA) measurements in any inventory system. One appropriate companion technique to autoradiography is high-resolution gamma-ray spectroscopy to assure the presence of fissile plutonium in the expected concentrations, at least in the surface region of the fuel element.[4] Because of self-absorption, all gamma-ray techniques, including autoradiography, suffer from the problem of sensitivity only to plutonium in the surface region. Active-neutron techniques can provide added assurance that fissile material is distributed throughout the fuel element.[4] A potentially powerful companion technique to autoradiography for *in situ* assembly-core verification is a measurement of the excess reactivity for that core.[4]

The appropriate role of autoradiography, and its coordination with other NDA techniques, must be determined by the credible diversion paths and by the specific safeguards objectives applicable to any safeguards system. Inventory verifications performed by international organizations must consider diversion strategies where wide-scale institutional efforts can be made to conceal SNM diversions. Such a diversion strategy could include the fabrication of fuel elements which contain little or no fissile material but which closely duplicate the response of genuine fuel elements when examined by autoradiography or NDA techniques. The effort to detect such substitution would obviously require the application of several techniques. In other cases where diversion might be by subnational groups, the fabrication of sophisticated replacement fuel elements would be less likely. In this instance, autoradiography would require less support by other measurement methods. In any case, the value of the autoradiographic method is in its ability to provide qualitative assurance that a large number of items are present in the quantity and location expected and have the specific radiation activity appropriate for those items. This assurance can be provided with a minimum of inspector effort and of facility impact.

III. APPLICATIONS TO URANIUM

A. General

Much of the SNM in world commerce is in the form of low-enriched uranium in fuel assemblies for light water reactors (LWR). Effective safeguards of the uranium in unirradiated assemblies requires a verification of the U-235 content of all the rods in an assembly. At present, several NDA techniques can be used to verify the content of isolated, individual LWR fuel rods or the rods on the periphery of fuel assemblies. The only methods which can examine fuel rods in the interior of an assembly are autoradiography and an active-neutron technique. [5] There are some important differences between autoradiography as applied to uranium and as applied to plutonium. One difference is the longer exposure time caused by the lower gamma-ray emission rates for uranium isotopes. If the methods used to obtain images of plutonium-containing fuel elements were applied to uranium-containing fuel elements of equal fissile content, exposures of several days would be required to obtain equal image density. This exposure-time problem can be partially overcome by using intensifying screens with high-speed films. A survey of films and intensifying screens indicated that Kodak Lanex screens (coated with gadolinium and lanthanum oxides) and Kodak-XR film minimized the exposure time for uranium-containing fuels.[3] The slightly slower Kodak Ortho-G film was normally used because of its compatibility with automatic industrial processing equipment. Exposure times in fuel assemblies were typically 3-4 hours with Ortho-G film and 2 hours with XR film. Polaroid film was also evaluated.[3] Although easier to process, image quality was not as good as conventional X-ray films.

A second problem encountered in the autoradiography of U-235 in LWR fuel is the contribution to image density from U-238. The most intense gamma ray from low-enriched LWR fuel is the 185 KeV gamma ray from U-235. Decay daughters dominate the gamma-ray spectrum from U-238. The largest contribution to the total photon flux from both U-235 and U-238 are X-rays resulting from alpha decay. These X-rays are more intense from U-235 because of its shorter half-life. However, measurements made with

a multichannel analyzer and an NaI(Tl) detector showed that, in the 66-283 KeV photon-energy range, the total photon flux from a rod containing depleted uranium was about 65% of the photon flux from a rod containing 3%-enriched uranium.[3]

In addition to gamma radiation, the Pa-234m daughter of U-238 also emits 2.29 MeV beta radiation and associated bremsstrahlung photons which penetrate common fuel-element cladding and which efficiently expose film. These contributions from Pa-234m radiations were significantly reduced by putting an additional 0.046-cm thick stainless steel absorber between the film packet and the fuel rod. The resulting U-235 sensitivity with Ortho-G film, Lanex screens, and a total of 0.076 cm stainless steel absorber is shown in Fig. 5, and was the greatest sensitivity to U-235 enrichment achieved.

B. Low-Enriched Uranium: LWR Assemblies

A model boiling water reactor (BWR) fuel assembly containing 3%-enriched uranium fuel rods was constructed to evaluate autoradiographic methods for use in actual LWR assemblies. Autoradiographs were obtained by simply inserting a film cassette between two rows of rods. Autoradiographs obtained in this model assembly indicated an additional problem: background contributions from other rods in the assembly. The density of an autoradiographic image of any single rod in an assembly has a major contribution from all other rods in that assembly. Lead shielding can be used to reduce this effect. The shielding configuration used in this study incorporated approximately 0.076-cm thick lead foil in the film packet between the film and the row of rods not under investigation. This reduced background contributions from rods on the "back" side of the film. A second lead foil was placed parallel to the film, one row removed on the "front" side, to further reduce the background.

The cassettes used to obtain these autoradiographs normally contained Kodak Ortho-G film at the center. On either side of the film was a Kodak Lanex intensifying screen. On the "back" side of the screen was lead foil for background reduction. On the "front" side was stainless steel beta absorber. All these items were enclosed in 3.5- or 7-cm wide lighttight Kodak ready-pack film cassettes. These cassettes and the lead shielding for the "front" side of the film were then enclosed in polyethylene envelopes to prevent contamination of the fuel rods. Depending on the thickness of beta and gamma absorbers, the total cassette thickness varied between 0.25 and 0.36 cm. This provided a loose fit between rows of rods, and there was no rod displacement.

Even with the lead shielding, the contribution to image density of a particular rod position by other rods in the assembly was large. This contribution was seen in an autoradiograph of a row of 7 rods with the center rod position vacant. The densities of the rod images on either side of the void were 0.77 optical-density units (3.5-hour exposure). The density at the center of the void was 0.60.

Since 65% of the photon flux from a 3%-enriched uranium rod was attributed to U-238, it was anticipated that 65% of the image density would also be contributed from the U-238. When a rod containing depleted uranium occupied the central position of a row in the model assembly (3.5-hour exposure), its image density was 0.71. This is 65% of the difference between

the density of the adjacent 3%-enriched rods and the density of the vacant position. Thus, the density difference between a 3%-enriched rod and a depleted rod was only 0.06 optical density units. Even though an enrichment change from 0.2% to 3% U-235 represents a very large relative change in U-235 content, it is associated with a small relative change in image density.

The uncertainty in making an autoradiographic U-235 enrichment measurement depends on the uncertainties in reproducing image densities for equal enrichment and on the reproducibility of the density measurement. Typical optical densitometers for X-ray film have repeatability uncertainties of $\pm$ 0.01 optical-density units. The standard deviation of image densities for 8 autoradiographs of the same row of rods in the model assembly was 0.012. At a confidence level of 95% (2 standard deviations), this represents 40% of the density difference between a depleted-uranium rod and a 3%-enriched rod. This means that for a 3%-enriched rod, an autoradiographic enrichment measurement would be 3 $\pm$ 1.2% U-235. A rod enriched to 2% would have overlapping confidence intervals with a 3%-enriched rod, and this enrichment difference could not be reliably detected.

In some cases, the image density difference for rods of equal enrichment can be greater than 0.012 optical-density units. These differences have been attributed to nonuniform contact between the X-ray film and the intensifying screens. Since reactor operators and fuel fabricators do not allow fuel rods to be put under stress, the film cassettes must fit rather loosely between the rows of an assembly. Some other mechanism must be found to provide good screen-film contact in order to reduce this source of uncertainty.

Several autoradiographs were obtained in the model assembly in which one or more 3%-enriched uranium rods were replaced by rods containing depleted uranium, or in which rod positions were simply left vacant. In all cases, thse simple defects were detected by visual inspection of the autoradiographs. In all cases, an accurate count of enriched fuel rods could be made.

The autoradiographic technique was also field tested at a boiling water reactor (BWR) site and at a pressurized water reactor (PWR) site. The technique was approved by the reactor operators for use with their fuel assemblies. An autoradiograph of a row of rods in a BWR assembly and its scanning-densitometer trace are shown in Fig. 6. The numbers above the rod images refer to the percent U-235 enrichment in each rod. "W" refers to a water rod which contains no uranium. Figure 6 is very typical of autoradiographs of rows of low-enriched uranium rods. The images of the end rods have less density and are less well-defined than the images of interior rods. The total image density increases towards the center of the assembly. This was attributed to higher background at the center. The 2.06%-enriched rod at the left appears to have a greater image density than the 2.73%-enriched rod on the right. The rods adjacent to the end rods, both 2.73% enriched, do not have equal image densities.

Autoradiographs were also obtained for PWR assemblies. A PWR assembly presents a problem because of the presence of control-rod thimbles distributed throughout the fuel-rod matrix. These control-rod thimbles have larger diameters than the fuel rods. Consequently, it is difficult

to fit a film cassette between rows of rods and still maintain uniform film-screen contact. In the PWR autoradiographs which were obtained, all fuel rods were visible and could be distinguished from the control-rod thimbles. However, rods of equal enrichment did not always produce images of equal density. This was attributed to the nonuniform film-screen contact.

C. Highly Enriched Uranium

Tests have recently been initiated to evaluate the applicability of autoradiography to highly enriched uranium. Preliminary results indicate that autoradiography will be very useful for verifying highly enriched uranium in the core of split-table type fast critical assemblies. Images of highly enriched uranium plates were obtained on film which was in reactor drawers during and shortly after a period of reactor operation. Some of the image density was caused by radiation from fission products. The contribution to image density from fission radiation during reactor operation has not yet been determined. This in-core uranium technique has the advantage over the in-core plutonium technique of providing a more direct indication of the presence of fissile material in the fuel elements.

D. Discussion

Autoradiography provided a means of verifying that rods in the interior of an unirradiated fuel assembly did contain U-235. This indication of U-235 content was only relative and was not an absolute-enrichment measurement. Autoradiographs need to be supported by quantitative, absolute-enrichment measurements on the exterior rods of the assemblies. Once enrichment of exterior rods is established, autoradiographs can verify enrichment consistency throughout the assembly.

The uncertainty in even relative enrichment determinations of 3% U-235 fuel rods was typically $\pm$ 1% U-235. These uncertainties precluded the reliable detection of the small enrichment variations normally found in rows of BWR assemblies. These uncertainties were due to poor film-screen contact, to the large radiation background present in the interior of fuel assemblies, and to the large contribution to image density from U-238.

Despite its lack of sensitivity to small enrichment changes, the autoradiographic technique was consistently able to detect dummy rods, missing rods, and rods containing depleted uranium. These defects were detected by visual inspection of processed film. The technique has the advantage of being simple to apply and of requiring simple, portable equipment. The technique was acceptable to the utility operating the reactors at which field tests were performed. Since interior rods in assemblies are not subject to verification under present international safeguards procedures, the autoradiographic technique may prove useful in providing more effective safeguards of the U-235 contained in unirradiated fuel assemblies deployed internationally.

Acknowledgement

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Fig. 2: Autoradiograph of plutonium-containing fuel rods in a storage drawer at the Reactor Research Institute, Switzerland.

Fig. 3: Autoradiograph of a stack of eight plutonium-containing plates with a depleted uranium plate in the center.

Fig. 4: *In-situ* autoradiographs of plutonium-containing plates in a reactor drawer. Above: Six plutonium-containing plates. Below: Five plutonium-containing plates and one void.

Fig. 5: Single-rod image density as a function of U-235 enrichment.

Fig. 6: Autoradiography and scanning densitometer trace of a row of rods in a BWR fuel assembly. Rod W contains no uranium.

Figure 1: Autoradiograph of thirteen plutonium-containing plates and one solid stainless steel plate in an Argonne storage canister.

Figure 2: Autoradiograph of plutonium-containing fuel rods in a storage drawer at the Reactor Research Institute, Switzerland.

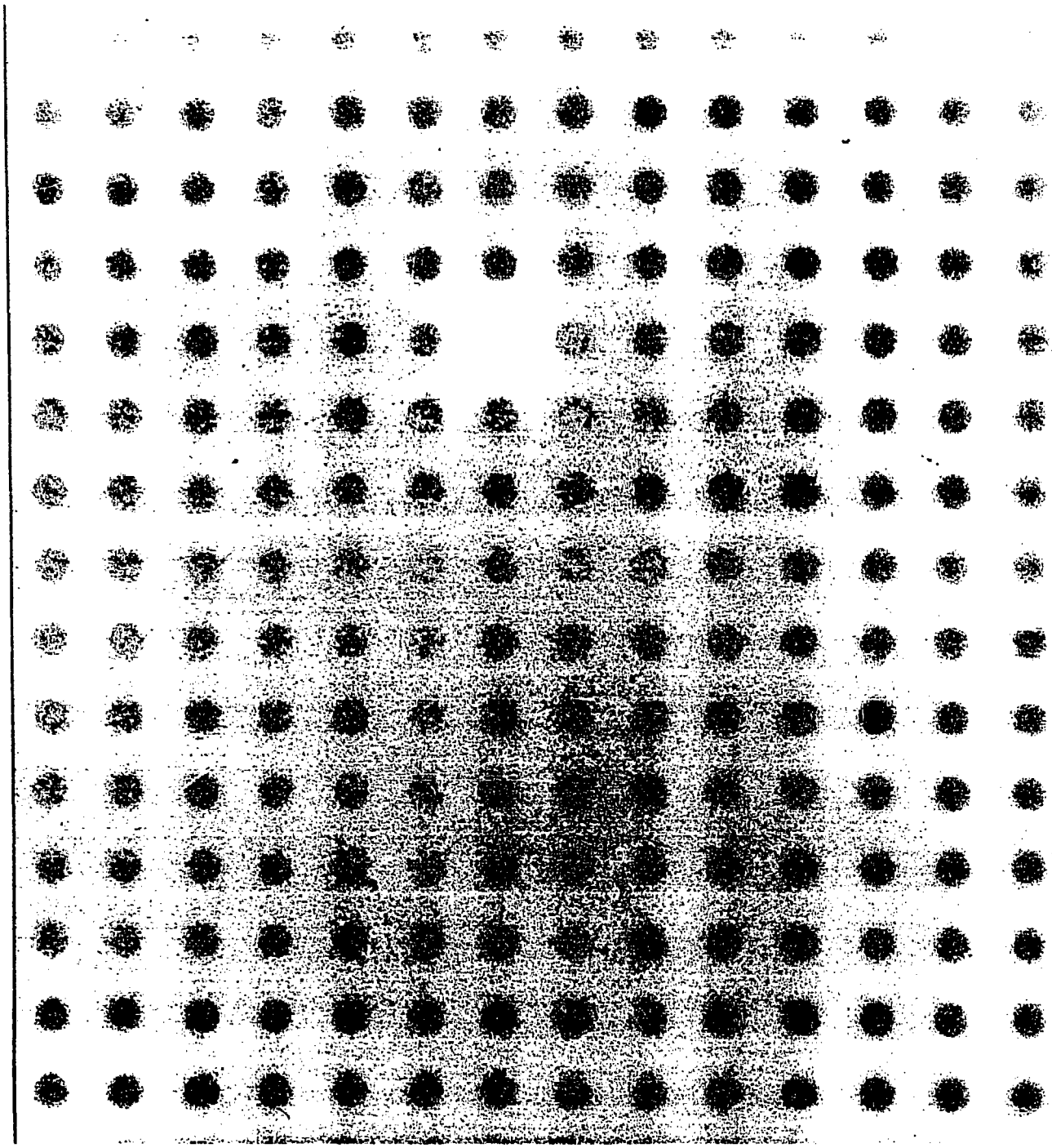

Figure 3: Autoradiograph of a stack of eight plutonium-containing plates with a depleted uranium plate in the center.

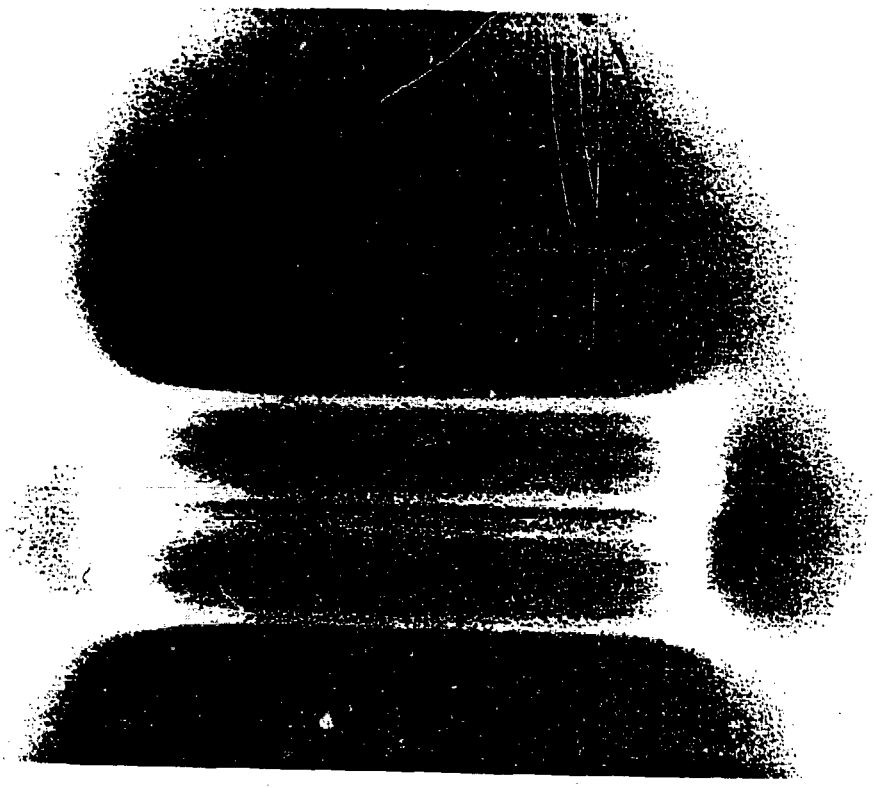

Figure 4: *In-situ* autoradiographs of plutonium-containing plates in a reactor drawer. Above: Six plutonium-containing plates. Below: Five plutonium-containing plates and one void.

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[redacted]

[redacted]

Figure 5: Single-rod image density as a function of U-235 enrichment.

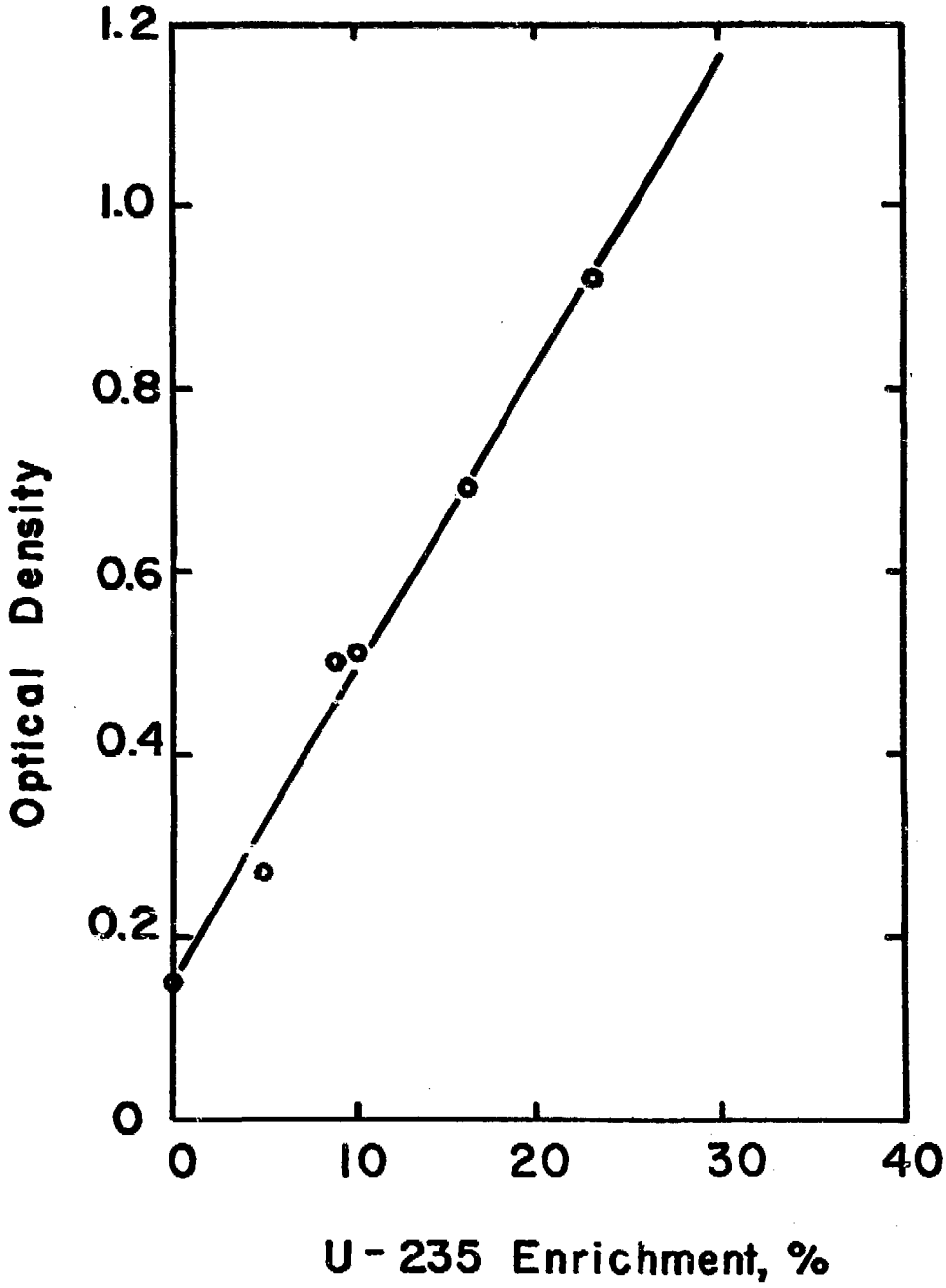
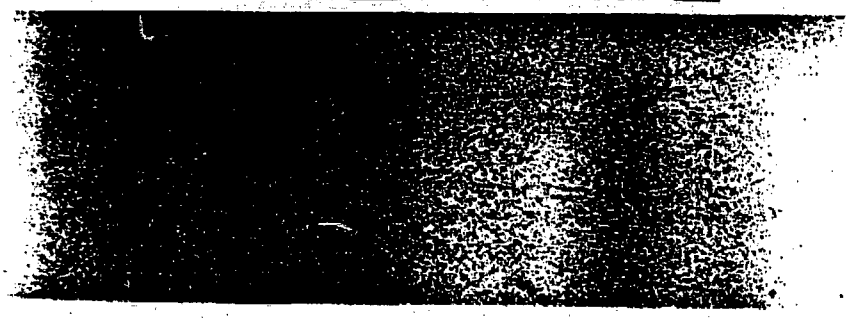
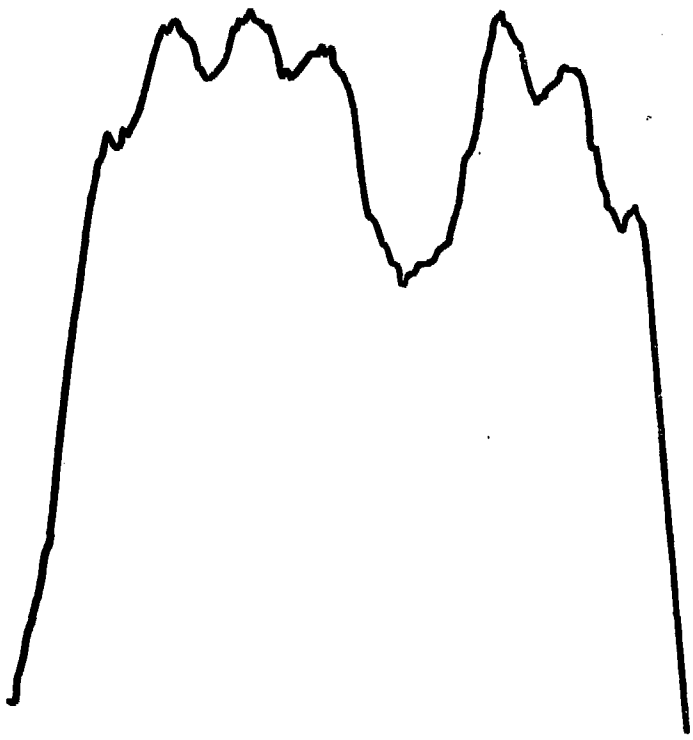

1.2
1.0
0.8
0.6
0.4
0.2
0
Optical Density
0
10
20
30
40
U - 235 Enrichment, %

Figure 6: Autoradiograph and scanning densitometer trace of a row of rods in a BWR fuel assembly. Rod W contains no uranium.

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