

EXPERIMENTAL PROGRAM FOR
DEVELOPMENT AND EVALUATION OF
NONDESTRUCTIVE ASSAY TECHNIQUES
FOR PLUTONIUM HOLDUP

by

Stephen B. Brumbach

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Nondestructive Assay Section
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ABSTRACT

An outline is presented for an experimental program to develop and evaluate nondestructive assay techniques applicable to holdup measurement in plutonium-containing fuel fabrication facilities. The current state-of-the-art in holdup measurements is reviewed. Various aspects of the fuel fabrication process and the fabrication facility are considered for their potential impact on holdup measurements. The measurement techniques considered are those using gamma-ray counting, neutron counting, and temperature measurement. The advantages and disadvantages of each technique are discussed. Potential difficulties in applying the techniques to holdup measurement are identified. Experiments are proposed to determine the effects of such problems as variation in sample thickness, in sample distribution, and in background radiation. These experiments are also directed toward identification of techniques most appropriate to various applications. Also proposed are experiments to quantify the uncertainties expected for each measurement.

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I. INTRODUCTION

An important aspect of the special nuclear material (SNM) control system in a mixed-oxide fuel fabrication facility is the use of book-physical inventory difference determinations. In making such determinations, the SNM in various phases of the fabrication process is inventoried by physical measurement and compared to the book value for SNM. If the physical inventory value, including corrections, is less than the book value by an amount greater than the uncertainty in this difference, then there is a possibility that a diversion has occurred.

The major categories of material inventoried are:

1. Starting material and all additional inputs to the process stream.
2. All finished, fabricated products both shipped and in storage.
3. All waste and scrap and other material removed during the fabrication process.
4. Material remaining as holdup in the process equipment.

The sensitivity of this test for diversion is determined by the uncertainty in the book-physical inventory difference. This, in turn, is a function of the uncertainties in the measurements of material in the various categories. Most of a facility's inventory is in categories 1 and 2, and these are the easiest to measure and have the smallest relative uncertainties. Categories 3 and 4 have a small fraction of the inventory but have large relative uncertainties and thus can contribute significantly to the overall uncertainty.

A recent analysis of the proposed Westinghouse-Anderson mixed-oxide fuel fabrication plant¹ estimated that holdup after cleanout would amount to about 1% of monthly throughput, assuming 8 kg plutonium per shift, 3 shifts

per day, and 22 days per month. In-process holdup (before cleanout) was estimated to be 4% of monthly throughput. This same analysis¹ also estimated that errors in holdup measurements would be $\pm 50\%$. Thus, the uncertainty in the physical inventory of holdup alone would be 0.5% of monthly throughput if a cleanout had been performed and 2% without cleanout. These are large uncertainties, and the aim of the following sections is to outline a program leading to holdup measurement techniques with reduced uncertainties. This will include the evaluation of existing techniques and the modification of such techniques where necessary. Because of the nature of the material and facilities being analyzed, only nondestructive assay (NDA) methods will be discussed.

II. OVERALL PLAN

This section presents a brief, general overview of the major topics which will be considered in a program to develop and evaluate improved in situ holdup measurement techniques. These are shown schematically in Fig. 1. The goal of the program is to obtain holdup measurements procedures and instruments with minimum uncertainty, hopefully better than the previously mentioned 0.5% of monthly throughput. It is important that these uncertainties be well characterized for various holdup applications.

Any analysis of NDA techniques for measurements in a fuel fabrication facility must consider the operations, apparatus, and material found in that facility, since these will have an impact on the measurements. Similarly, a measurement procedure will have some impact on the operation of the facility. This interaction is discussed in Section III.

The various NDA techniques applicable to holdup are considered in Section IV. Results of holdup measurements using these techniques are reviewed. Problem areas for applying these techniques to holdup situations and sources of uncertainties are identified. Suggestions are made where further research will be done. Specific experiments are proposed to evaluate these techniques, as are other experiments to consider the feasibility of alternate procedures and instruments.

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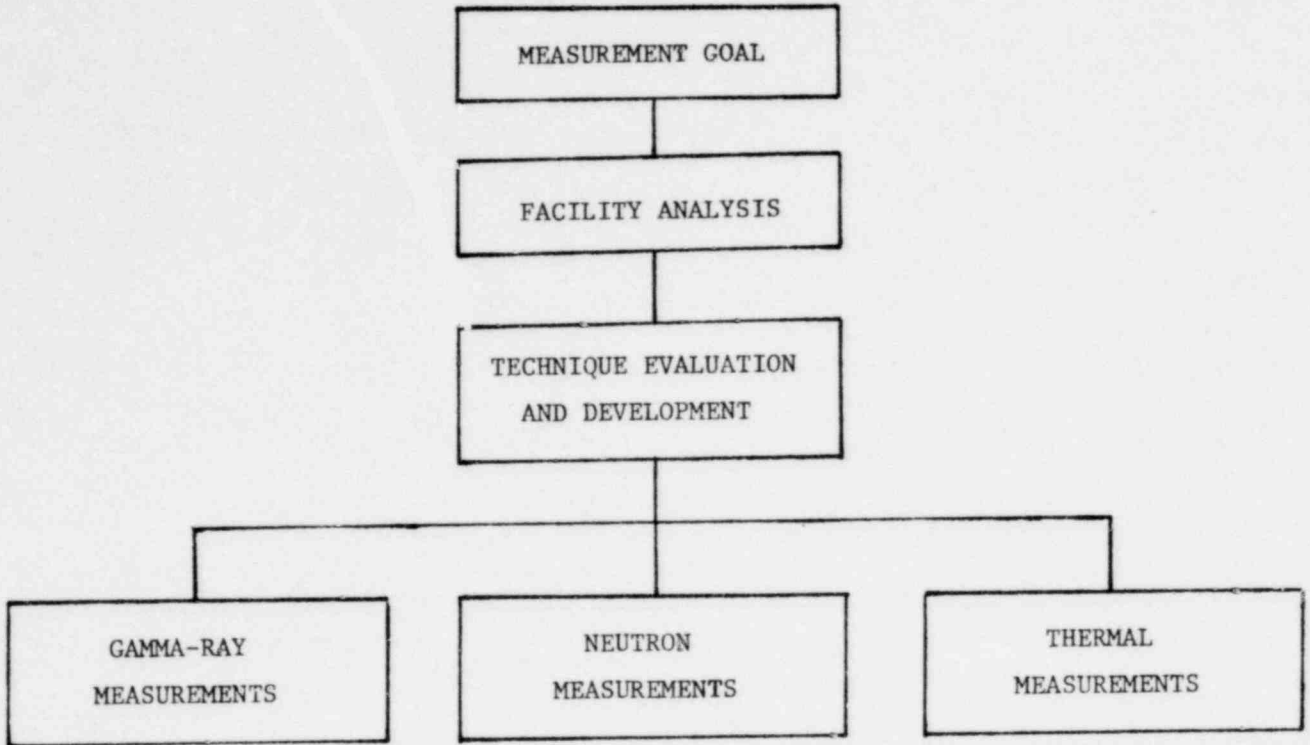


Fig. 1. Overall sequence of steps in program for holdup measurement technique evaluation and development.

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III. FACILITY ANALYSIS

A. Overall Process

The details of the processes, process equipment, and material processed in any facility will have a great impact on the instruments and procedures used to measure holdup in that facility. The major considerations for analysis of the facility-measurement interaction are outlined in Fig. 2. The basic processing steps in a mixed-oxide fuel-fabrication facility are shown in Fig. 3, which is a diagram of a plant flow sheet. This particular diagram pertains to the Westinghouse-Anderson plant² but is typical of most such plants.^{3,4} This flow sheet is not complete, since there are branches from this stream into various waste recovery, analysis, and inspection systems.

B. Materials

The SNM present in a mixed-oxide plant will primarily be solid PuO_2 , much of it in the form of finely divided powder. After blending, the solid PuO_2 will be mixed with a much larger amount (approximately 95%) of UO_2 . Some of the SNM may also be in solution form, especially in the scrap recovery system. It is possible, but not likely, that the plant input could be Pu nitrate solution and the solid oxide precipitated at the plant.

The plutonium present in the mixed oxide can have various isotopic compositions. Isotope 239 will be the major component, but isotopes 238, 240, 241, and 242 will also be present, as will be americium-241. The isotopic composition is a function of the irradiation history of the material. The presence of these various isotopes will allow NDA measurements to be made using emitted gamma rays, neutrons, and heat due to alpha decay. A precise knowledge of the isotopic composition is essential for such measurements, and this information should be available from chemical analysis.

C. The Occurrence of Holdup

Holdup is that amount of SNM remaining in and around processing equipment at the cessation of operation of that equipment. Holdup can be

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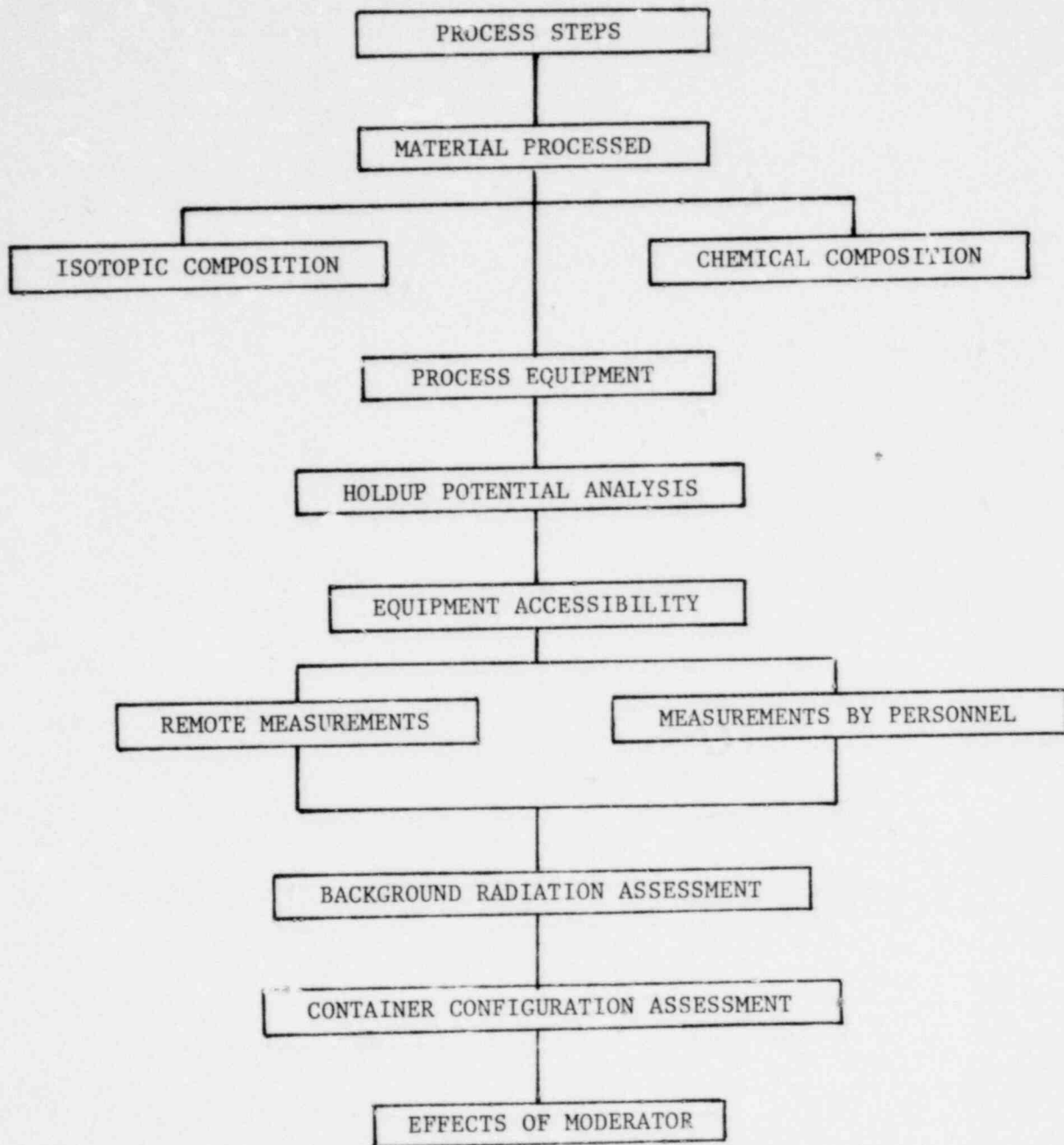


Fig. 2. Sequence of steps in the facility analysis for holdup measurement procedure evaluation.

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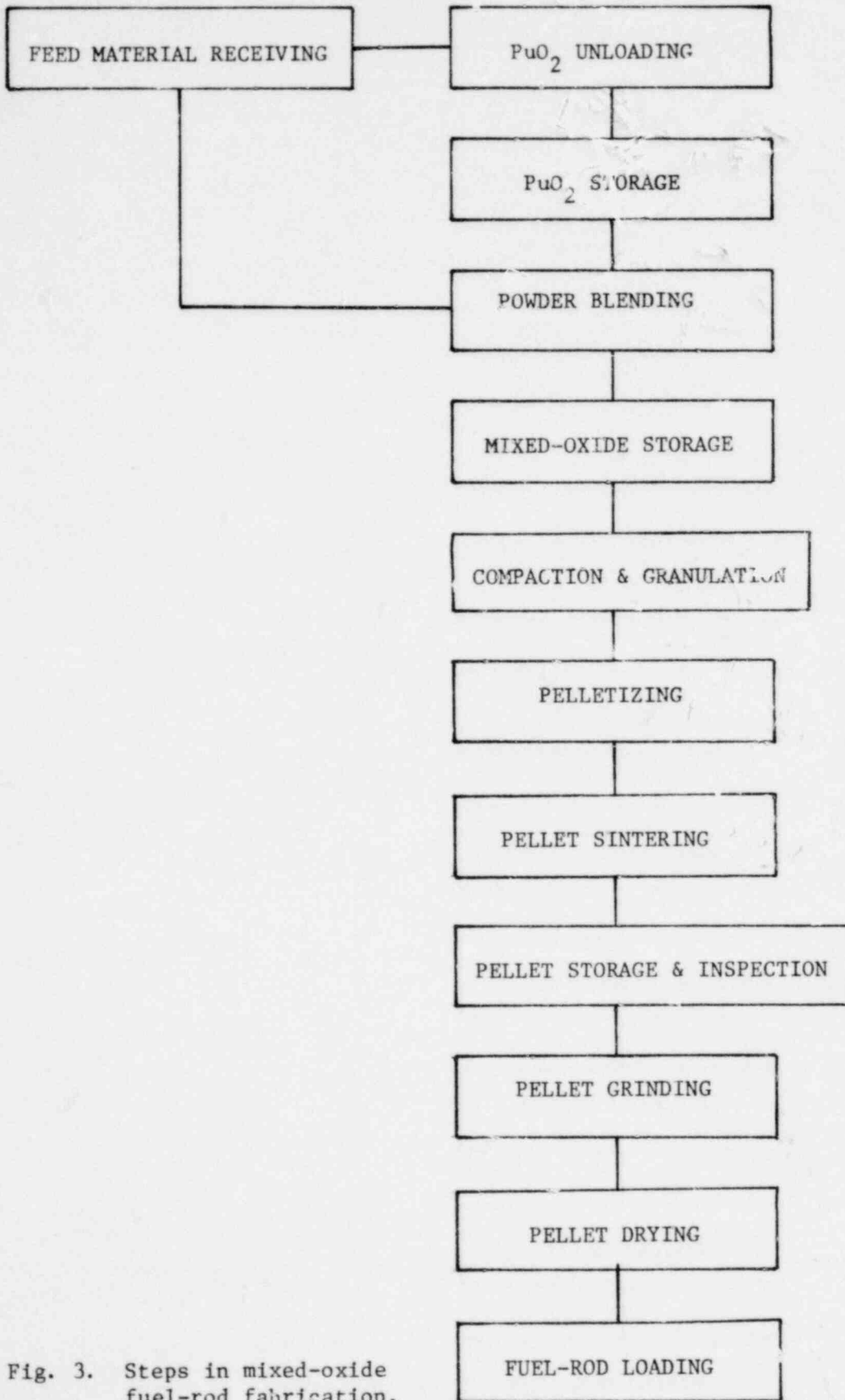


Fig. 3. Steps in mixed-oxide fuel-rod fabrication.

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characterized as that remaining:

1. After shutdown. Normally, that remaining at the end of processing a batch of material.
2. After draindown. That remaining after operating equipment specifically to remove remaining material.
3. After cleanout. That remaining after a series of non-production operations designed to remove material.

The amount of material contained in holdup decreases in going from category 1 to 3.

Where, and in what form, can holdup occur? Where finely divided solid powders are handled, dusts can be a problem. The insides of storage and transfer equipment can become coated with dusts, as can such confinement areas as glovebox walls and floors. Dusts can also accumulate in air ducts, on filters, at surface irregularities of components, and along transfer paths. The incomplete emptying of storage vessels and transfer devices is a potential holdup source. Spills and overflows, especially where mechanical transfer is used, are also possible contributors. Operations such as pellet grinding can significantly add to holdup, since material is carried along with liquid coolant. There is the possibility of material accumulation at valves or pipe elbows. Holdup can be uniformly distributed over large surface areas or localized in large concentrations. In summary, holdup can occur, and probably will occur, in almost every component of SNM-handling equipment and every SNM-containment device.

Holdup estimates made for the Westinghouse-Anderson facility¹ indicated that major contributions to holdup will be from the powder-blending and storage stages of operation. Total in-process holdup was estimated to be 21.8 kg of plutonium, with 6.6 kg remaining after cleanout.

D. The Measurement-Facility Interaction

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One major consideration affecting both the operation of a plutonium-

handling facility and the conduct of holdup measurements is the accessibility of facility apparatus to measurement personnel. It is likely to be easier to conduct a holdup assay if access is available to various components so that instrumentation can be positioned to make measurements of optimum sensitivity and accuracy for a given material configuration. However, such access may be undesirable from health physics or security considerations. A possible solution is to build NDA instruments into a facility so that measurements can be done remotely. Contamination and maintenance then become problems. The physical dimensions of the process equipment itself will affect holdup measurements. The thickness of a container and any additional shielding will affect the intensity of gamma rays transmitted. The regularity of the geometry of individual components can affect the ease of defining the sampling area in any measurement. Also, the relative location of various components will affect the background radiation at any component and hence affect a radiation measurement. The neutron-moderation characteristics of the equipment and its enclosures will affect the performance of any assay experiment using neutrons. The presence of moderator material in neutron detectors may be undesirable due to criticality requirements. Finally, the holdup assay measurements should be made quickly enough so that plant operations are not excessively disrupted.

IV. MEASUREMENT TECHNIQUES

A. Gamma-Ray Techniques

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1. Introduction

Gamma-ray techniques for quantitative determination of plutonium content in a sample have been successfully used for many years.⁵⁻⁹ Results with high precision and accuracy have been obtained for uniformly distributed homogeneous material with regular geometry such as reactor fuel elements.^{8,10,11} For such conditions, one can correlate the number of gamma rays in a particular energy interval with the amount of material by calibrating with similar standard samples of known plutonium content.

The application of gamma-ray methods to holdup problems is a much more difficult problem and only a few measurements have been reported.

Heagerty¹⁰ conducted a plutonium holdup survey of the Kerr-McGee mixed-oxide fuel plant using gamma-ray techniques, while Augustson and Walton¹¹ described gamma-ray techniques to make holdup measurements of the same plant. Reed, Andrews, and Keller¹² developed gamma-ray techniques for measuring U-235 holdup in ventilation duct work and incinerator systems. Their estimated measurement errors in two assays were $\pm 23\%$ and $\pm 33\%$, and the agreement with the amounts of U-235 actually recovered were high by 32% and 3%. Kindle¹³ used both neutron and gamma-ray methods to assay plutonium holdup in a scrap recycle plant. These measurements agreed with the amounts of plutonium eventually recovered to $\pm 16\%$. Anderson, James, and Morgan¹⁴ also analyzed a mixed-oxide fuel plant for holdup using gamma-ray methods and reported agreement with amount of recovered material of $\pm 20\%$. In these measurements, a typical apparatus was a small (2-inch diameter) NaI(Tl) scintillation detector with collimation. The associated electronics were never more complicated than a dual-channel analyzer. The use of gamma-ray techniques for holdup measurement is also discussed by Augustson and Reilly.¹⁵

2. Applications to Holdup Measurements

A schematic outline of the factors which must be considered in using gamma-ray techniques to measure holdup is given in Fig. 4. For most applications, NaI(Tl) detectors will be sufficient, but the following discussion will be equally pertinent to measurements using Ge(Li) detectors. The high-resolution Ge(Li) and low-resolution NaI(Tl) detectors will both be included in comparative evaluations of various gamma-ray techniques.

One problem in making gamma-ray holdup measurements is the contribution to the gamma-ray flux at a component of interest from material in other nearby components. Normally, this effect is minimized by the use of collimators and shields to define the detection region. The problem is complicated if two components are on the same line of sight. An example of this is the making of a measurement on a component inside a glovebox when the glovebox wall also has SNM holdup on its surface. In future experiments, collimator design will receive attention so that various components, such as pipes, tanks, glovebox walls, material-transporting devices, or furnaces can be isolated from one another and from background radiation. Collimators can and will be designed for special sample geometries such as cylindrical

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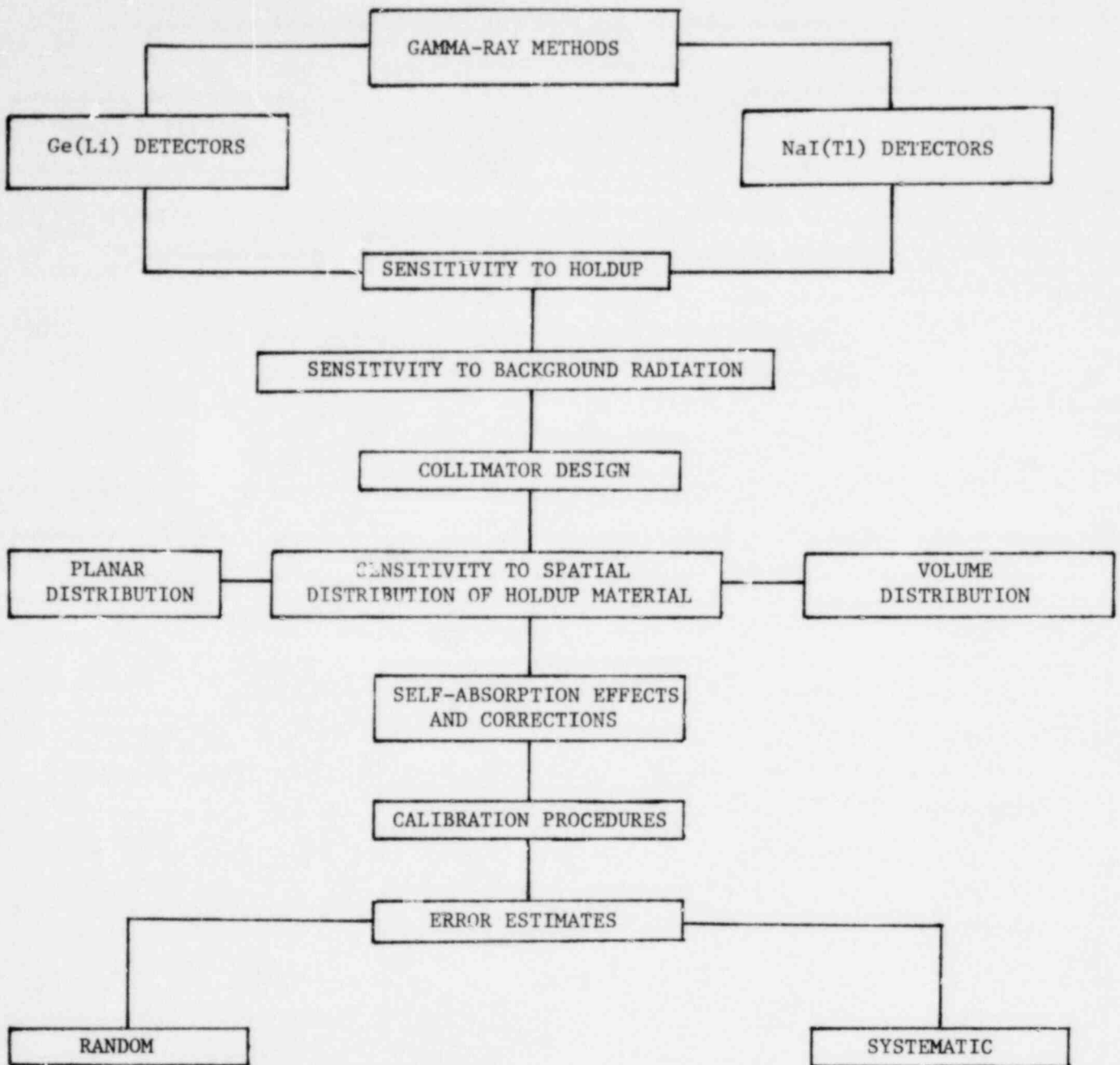


Fig. 4. Steps in the evaluation and development of gamma-ray holdup measurement techniques.

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storage containers or pipes. Multiple-hole collimators will be considered, since these can give high spatial resolution yet have good sensitivity.^{16,17}

A large source of uncertainty in gamma-ray measurements of holdup is the uncertainty in the location of the SNM within the container. The SNM can also be distributed nonuniformly, in either the surface coverage or the thickness, or in both. The uncertainty in source-detector distance is particularly severe, since the gamma-ray flux of a detector varies as $1/R^2$, where R is the source-detector separation for a point source. Such a problem could arise if a large container were assayed by a detector close to the container. If the SNM were localized (approximating a point source) on the side of the container closer to the detector, a much larger gamma-ray flux will be measured than if the same source were on the far side of the container. Future experiments will study the response of source-detector-collimation systems to variations in source location within containers encountered in holdup measurements. The use of detectors at different locations with respect to SNM holdup will be considered as a means of identifying the holdup locations. Such experiments will allow error estimates to be made for particular detector-holdup geometries.

The case of nonuniform area coverage occurs when the sampling region is only partially covered by SNM deposits. Such a situation can occur on a glovebox wall or on the inner wall of a storage tank. Figure 5 can be used to understand the effect of sample area distribution on the gamma-ray flux at a hypothetical detector. Here A is a point detector located a distance R from point O in the plane of the sample. The gamma-ray flux, ϕ_0 , at point A due to a point SNM source located at O is:

$$\phi_s = \frac{S_0}{4\pi R^2} \quad (1)$$

where S_0 is the source strength in units of gamma rays per second. Following the treatment of Reed, Andrews, and Keller,¹² the gamma-ray flux, ϕ_s , at point A due to the same amount of SNM distributed over a disc of radius r , is:

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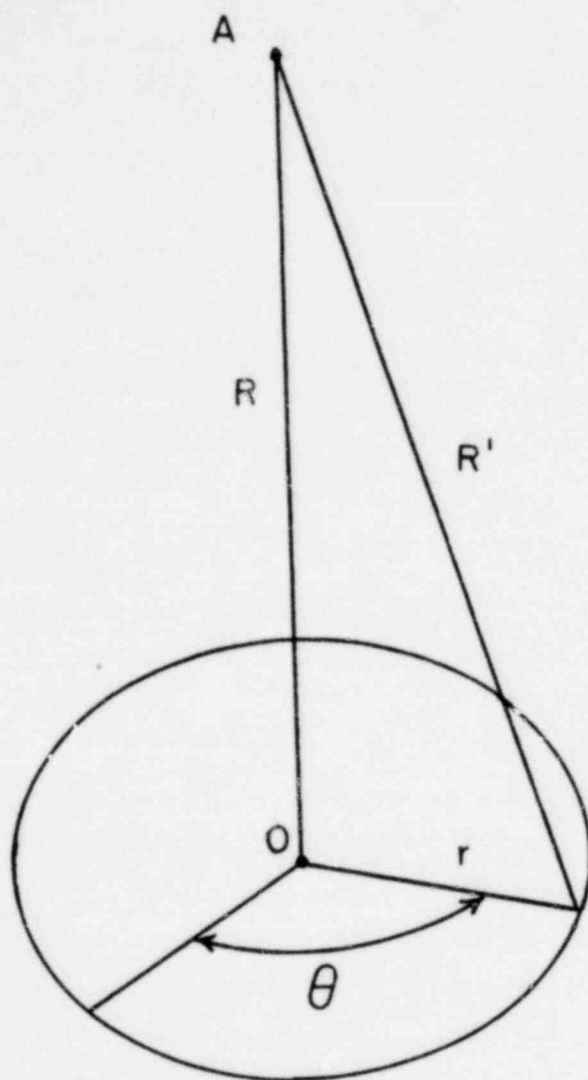


Fig. 5. Schematic diagram of source-detector arrangement for gamma-ray counting.

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$$\phi_s = \frac{S_0}{2\pi r^2} \int_0^{2\pi} \int_R^{\sqrt{r^2 + R^2}} \frac{1}{4\pi R'} dR' d\theta \quad (2)$$

where $S_0/2\pi r^2$ is now the unit area source strength.

Upon integration,

$$\phi_s = \frac{S_0}{4\pi r^2} \ln\left(1 + \frac{r^2}{R^2}\right) \quad (3)$$

In order to compare these two fluxes, consider the ratio ϕ_s/ϕ_0 :

$$\frac{\phi_s}{\phi_0} = \frac{R^2}{r^2} \ln\left(1 + \frac{r^2}{R^2}\right) \quad (4)$$

In Eqs. 1-4, the simplifying assumption of no gamma-ray absorption has been made. From Eq. 4, it is seen that, as R becomes large compared to r , ϕ_s/ϕ_0 approaches 1, as is expected. For $r = R$, $\phi_s/\phi_0 = 0.69$, and this ratio decreases as r becomes large compared to R . Thus it is evident that the way in which a given amount of SNM is distributed over the area viewed by a gamma-ray detector affects the observed gamma-ray flux.

Another type of distribution problem is that of nonuniform material thickness. Since gamma-ray experiments normally measure number of events and relate this to grams of material via calibration, sample thickness

would not need to be considered. However, for thick samples, self-absorption will occur. Plutonium holdup can possibly occur in thick deposits, and thus self-absorption effects can result in measurement errors. The errors introduced by self-absorption can be examined by considering the treatment by Evans.¹⁸ The ratio of transmitted gamma rays, I , to those emitted, I_0 , is given as a function of absorption coefficient, μ , and sample thickness, $2t$, in Eq. 5.

$$\frac{I}{I_0} = e^{-\mu t} \int_{-t}^t \frac{e^{\mu x}}{2t} dx \quad (5)$$

Here x is the unit differential thickness. Equation 5 includes the simplifying assumption that the source-detector distance is large compared to the material thickness. The solution to this equation is

$$\frac{I}{I_0} = \frac{1}{2\mu t} [1 - e^{-2\mu t}] \quad (6)$$

These self-absorption errors increase with increasing sample thickness and decreasing gamma-ray energy. The fraction of gamma rays self-absorbed ($1 - I/I_0$) for 413 keV and 129 keV gamma rays and for various thickness of metallic plutonium are shown in Table I.

Table I. Fraction of Gamma Rays Self-absorbed

<u>Plutonium Thickness, Inch</u>	<u>413 keV</u>	<u>129 keV</u>
0.001	0.007	0.09
0.002	0.014	0.17
0.015	0.10	0.33
0.060	0.33	0.91
0.120	0.55	0.96

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These values were calculated from Eq. 6.

In order to reduce errors caused by self-absorption, gamma-ray measurements can include an intensity ratio determination for high- and low-energy gamma rays or group of gamma rays in the plutonium spectrum. Such intensity ratio measurements have shown that a thickness determination can be made.¹⁶ Results showed that thickness could be determined with 1% accuracy with an NaI(Tl) detector and foils up to 0.060 inches thick.¹⁶ An alternative method for making self-absorption corrections is to measure the attenuation of gamma rays transmitted through the container and contents. Knowing the material and dimensions of the container, one can then calculate the attenuation due to the contents and estimate its thickness. This method has been discussed by Parker and Reilly.¹⁹ Further experiments with both techniques for making self-absorption corrections will be carried out for SNM in holdup configurations.

An important class of experiments which need to be developed and performed are measurement calibrations. The response of various sample-detector-collimator geometries must be characterized in experiments realistically simulating anticipated SNM holdup configurations. Such configurations as uniform coatings (thick and thin), nonuniform coatings, and localized high concentrations of SNM (lumps of material) need to be simulated. Potential locations such as glovebox walls, storage tanks, pipes, grinders, blenders, presses, furnaces, and transfer devices should be considered. These experiments can be used to select optimum procedures and configurations, and can also serve to estimate the accuracy of the measurements. Repetitive measurements on any given sample will allow random errors to be assessed (measurement precision) while comparison with known standards will allow bias estimates and hence systematic error estimates to be made. Estimates can be obtained for worst-case errors for extremely unfavorable geometries.

B. Neutron Techniques

1. Introduction

Neutrons emitted in α, n reactions and in spontaneous or induced fission of plutonium isotopes have long been used as a basis of well-established nondestructive assay methods.^{7,20} There have been very few examples of neutron assay techniques applied to holdup measurement. In one case, Tape Close, and Walton²¹ used total neutron-counting methods

to measure total room holdup of plutonium at the Kerr-McGee plant. Their reported measurement uncertainties were $\pm 50\%$. Kindle¹³ also reported using neutron techniques in some holdup determinations.

There are several neutron-assay methods available. Total neutron-counting methods measure all neutrons emitted: α, n ; spontaneous fission; and induced fission. Coincidence methods only measure fission neutrons or gamma rays. Both α, n and fission neutrons are emitted with high-energy spectra. These neutrons, and also gamma rays, can be directly detected using scintillation detectors. Other detectors using BF_3 or ^3He have useful sensitivity only to thermal neutrons and must be surrounded by moderator material. Coincidence neutron techniques can be either active or passive. Active techniques measure fission-neutron flux induced by an external neutron source, while passive techniques measure fluxes of spontaneous fission neutrons. The use of additional neutron sources is difficult in an in situ experiment, and only passive techniques will be considered here.

2. Applications to Holdup Measurements

The major subjects which must be considered when applying neutron techniques to holdup problems are shown schematically in Fig. 6. In general, holdup assay techniques utilizing neutrons have the advantage of being relatively insensitive to the container attenuation and self-absorption problems encountered with gamma-ray techniques. However, self-multiplication due to induced fission can be a problem for large amounts of material. Perhaps the most serious problem with neutron methods is their sensitivity to the background neutron flux which is certain to be present in holdup assay situations. This is due mainly to the nondirectional character of most neutron detectors. Background contributions are most serious when counting total (α, n plus fission) Neutrons.

A typical thermal neutron detector assembly is shown schematically in Fig. 7. Two equal size slabs of polyethylene contain BF_3 neutron proportional counters. The detector slabs are placed on opposite sides of the suspected holdup container. The container can be almost any component in the fuel

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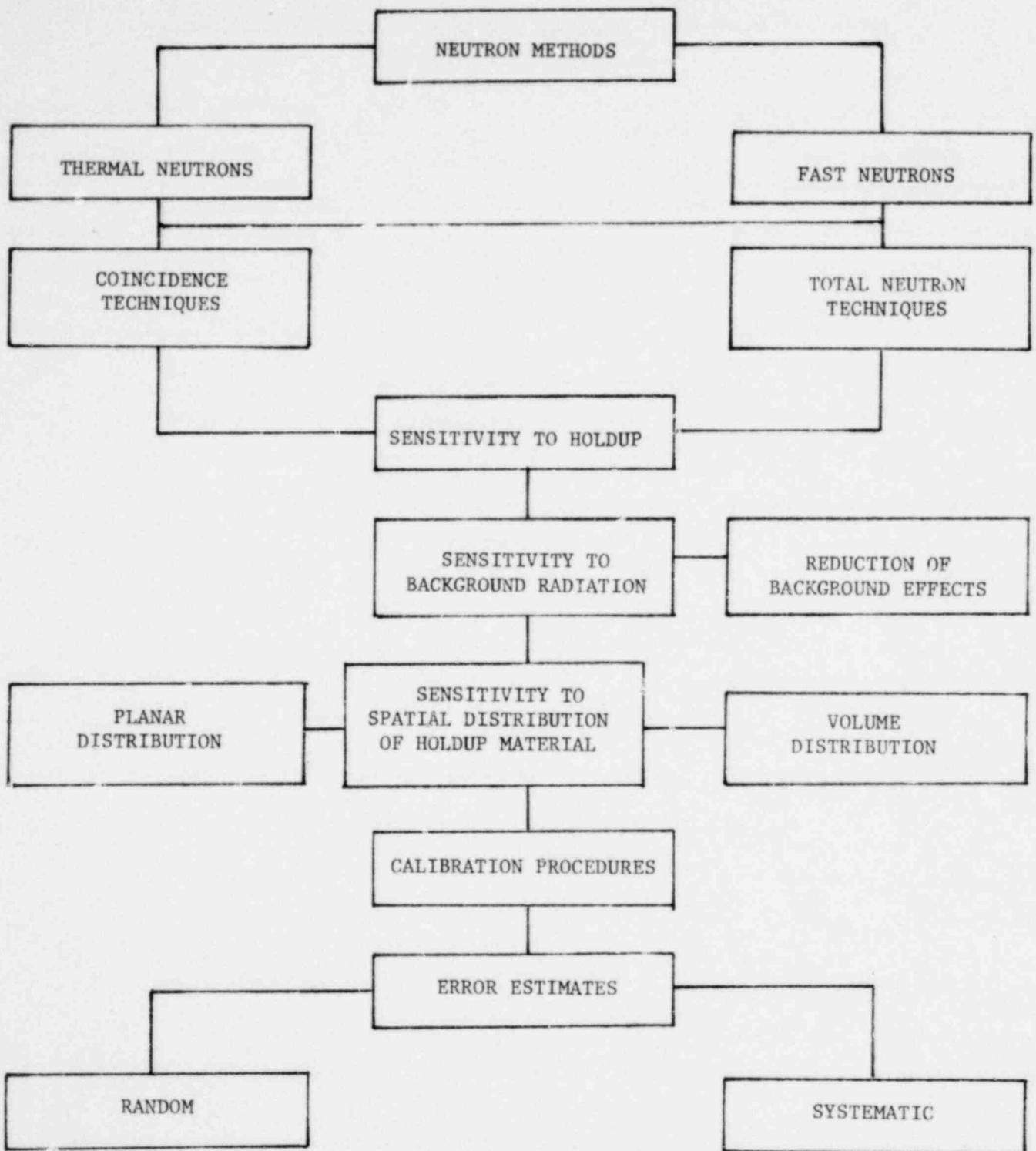


Fig. 6. Steps in development and evaluation of neutron measurement techniques.

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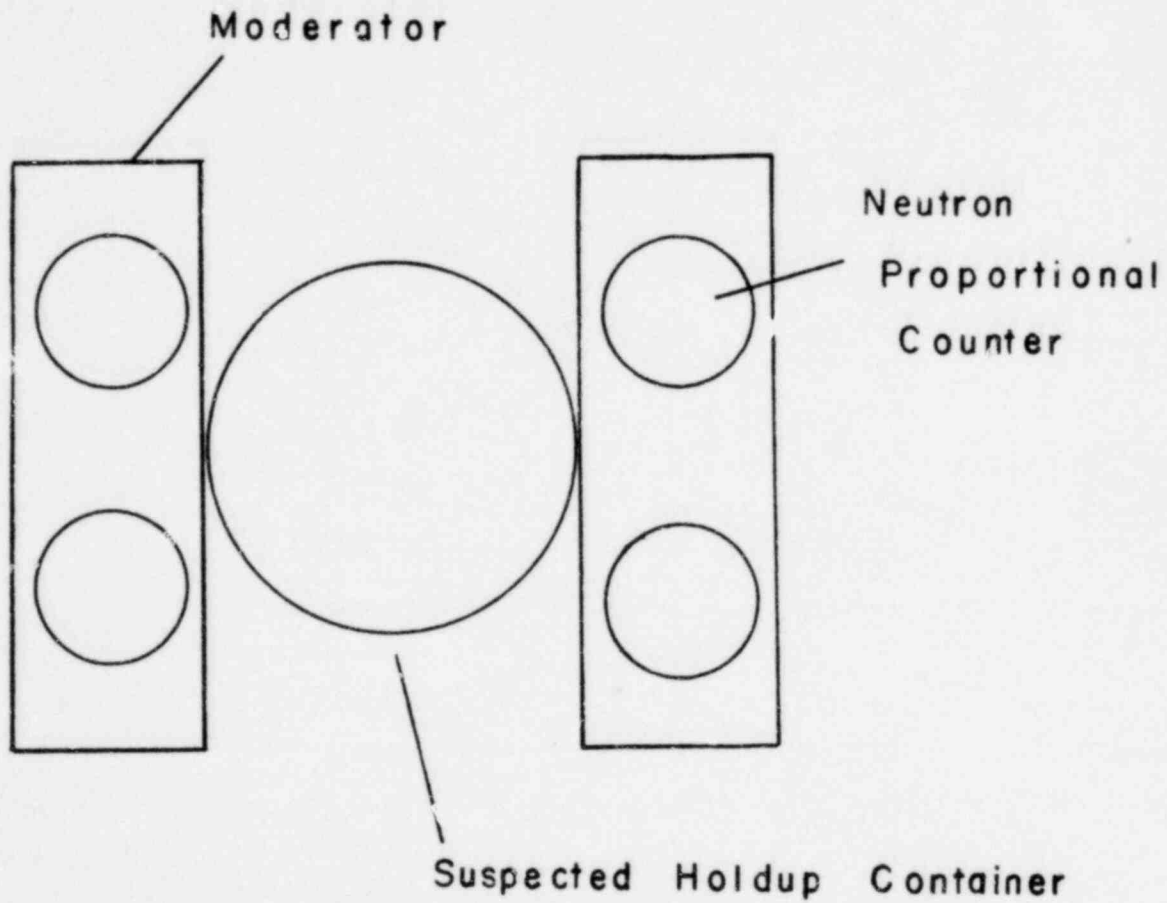


Fig. 7. Schematic diagram of apparatus for neutron measurement of holdup.

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fabrication facility. If fast neutrons were being detected, the slabs might be plastic scintillator material, and photomultiplier tubes would replace the BF_3 counters. Both thermal neutron detectors, with their moderator material, and scintillation detectors constitute rather large and cumbersome assemblies, and it is sometimes difficult to place these assemblies close to the material measured. The use of moderator/absorber material to reduce background neutron and gamma-ray effects contributes to this problem. It is important to keep the source-detector separation, R , small, since, for a point neutron source, the detector response to total neutrons varies as $1/R^2$. If coincidence methods are used, this response has a $1/R^4$ dependence, since the coincidence efficiency varies as the square of the total neutron efficiency. The introduction of large amounts of moderator material into a plutonium processing area poses another problem from criticality safety considerations.

Future experiments will study the response of such detector assemblies, as shown in Fig. 7, to plutonium in potential holdup configurations and to plutonium in other locations near the detector which can contribute to background. Some preliminary results have been obtained,¹⁶ but more is needed. Further development work will be undertaken into ways of minimizing the effect of background neutrons. Moderator/absorber systems will be evaluated for their effectiveness at background flux reduction. These experiments are needed for both fast- and thermal-neutron detection methods.

As in the case of gamma-ray counting techniques, uncertainties in sample location with respect to detector location will result in uncertainties in plutonium determination. Even for constant source plane-detector plane distance, the response of a neutron detector is not uniform for a point source of neutrons at various points along the detector plane. More serious is the uncertainty arising from variations in source-detector distance, R , especially in large containers. Since the coincidence neutron count rate varies as $1/R^4$, uncertainties in the value of R lead to large uncertainties in the amount of material determined by a neutron-assay technique. Further experiments will be performed to determine the sensitivity of neutron measurements to variation of sample position within various containers.

Also, as in the case of gamma-ray techniques, evaluation and calibration experiments using realistic holdup simulation are needed.

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These experiments will allow optimum procedures and configurations to be selected and both random and systematic errors to be estimated.

In summary, even though neutron assay techniques have many characteristics which make them difficult to use for in situ holdup measurements, they may prove useful in some applications. Their insensitivity to self-attenuation or attenuation by containers may make neutron methods attractive where large concentrations of holdup are suspected, or where container attenuation is particularly severe. For example, if a gamma-ray assay using intensity ratio indicates a very thick plutonium deposit, the gamma-ray result might be verified using a neutron-assay method. The work of Tape, Close, and Walton²¹ has shown that estimates of total room holdup can be made using total neutron counting if the neutron flux from adjoining rooms is not large.

C. Thermal Techniques

1. Introduction

The primary mode of radiative decay of plutonium isotopes 238, 239, and 240 is by α -particle emission. These α -particles are stopped in the plutonium after a short mean free path. The energy transfer to the surrounding plutonium matrix results in self-heating of the matrix. The measurement of this heat has been utilized in the calorimetric-assay technique.^{7,22} Calorimetry, however, is not a practical technique for holdup measurements.

As a result of self-heating, a plutonium source will be at a slightly higher temperature than its environment. This temperature difference will depend on the amount and isotopic composition of the plutonium and the various heat-loss mechanisms operative. Containers of plutonium holdup might be expected to be as much as 1°C warmer than ambient. Temperature differences as small as 0.1°C can be detected by measuring the intensity of long-wavelength infrared radiation. Compact infrared equipment is available with 0.1°C temperature resolution.

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2. Applications to Holdup Measurements

A recent experiment has indicated that small local power sources in a stainless steel pipe can cause temperature differences detectable by infrared techniques.¹⁶ An example is shown in Fig. 8.¹⁶ The heat source is an 0.4-inch long electrical resistor in contact with the inner wall of the pipe. The plutonium isotopic mixture likely to be found in recycle fuel material will have a specific power of about 4 mW gm^{-1} . Thus Fig. 8a represents the case of an approximately 25-gram lump of plutonium on the wall of a pipe.

It is not likely that the infrared method will become a quantitative assay technique, nor is the method likely to be useful for locating or measuring uniformly distributed material since this would not result in a temperature difference. However, the technique could become quite useful as a rapid scanning method for locating large concentrations of holdup. More experiments are planned to define the limits of sensitivity for plutonium in various configurations and containers. Measurements are also needed to determine the effect of material distribution within a given container. This will involve realistic holdup simulation experiments, important for evaluation of all techniques.

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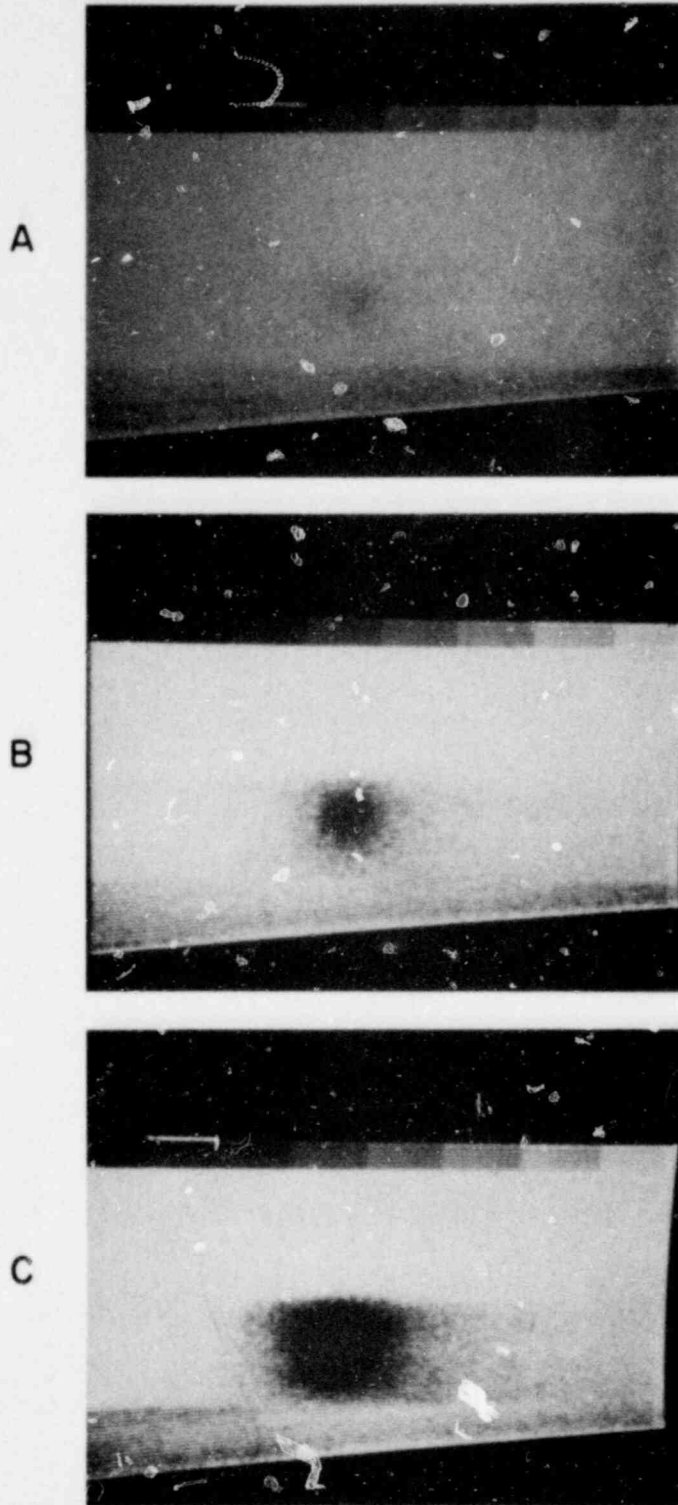


Fig. 8. Infrared scans of stainless steel pipe with various power input to electrical resistor attached to inner pipe wall. (a) 104 mW, (b) 213 mW, (c) 360 mW.

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