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## **Nondestructive Verification and Assay Systems for Spent Fuels**

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## EXECUTIVE SUMMARY

This interim report reviews the potential role of nondestructive measurements on irradiated light-water reactor (LWR) fuels. The study is being performed for the Savannah River Interim Spent-Fuel Storage Project Office, with technical review by Savannah River Laboratory. Until recently, the scope of this study was to develop the conceptual design of a spent-fuel nondestructive verification and assay system (VAS) for future away-from-reactor (AFR) spent-fuel storage facilities and to fabricate, test, and demonstrate a prototype VAS system. The design of a prototype VAS is ~80% complete, and some of the hardware already has been procured.

Recent changes in Government policy regarding the acquisition and storage of excess spent fuel in federally operated AFR storage facilities may terminate the present study. At the same time, the Department of Energy (DOE) is pursuing a new policy for implementing new technologies to enable the domestic nuclear power industry to store its own spent fuel at reactor sites as much as possible, pending the startup of spent-fuel reprocessing plants. In the meantime, the nuclear power industry, under the joint sponsorship of DOE and the Electric Power Research Institute (EPRI), is investigating new fuel management and storage techniques aimed at using high-burnup fuels. Techniques in an advanced stage of development include high-density fuel storage, fuel-bundle disassembly with repackaging of new assemblies, and compaction and storage of high-burnup rods.

Nuclear materials accounting and control for safeguards and nonproliferation and for new spent-fuel management technologies require nondestructive measurements that span a wide range of fuel characteristics and fuel types (Table S-I). In addition to satisfying safeguards requirements, these measurements can satisfy facility requirements, such as in-plant materials management, process control, and criticality control. Several nondestructive techniques are already well advanced, primarily because of this project and the US program of technical assistance to the International Atomic Energy Agency (IAEA). The applicability of these nondestructive techniques is not limited to LWR spent-fuel assemblies. The basic technology can be applied to any spent-fuel material (material test reactor and liquid-metal fast breeder reactor fuel materials). This new spent-fuel nondestructive measurement technology should be transferred to the nuclear industry.

Specific recommendations of this interim study include

- The Cerenkov viewing device and the neutron ring detector, both already developed for the IAEA, should be transferred to the domestic nuclear industry. These devices are portable, relatively easy to use, and provide confirmation of the integrity and the burnup of spent-fuel assemblies.
- The spent-fuel nondestructive VAS should be completed, and a program of field testing and demonstration should be carried out. This system would be the prototype for future in-plant, nondestructive measurement systems at spent-fuel storage and reprocessing facilities. As such, an on-site program of field testing and demonstration is needed at an actual facility before a final system can be considered ready for in-plant use.
- Application of nondestructive measurements for spent fuel should be studied in relation to the new techniques for fuel management and storage currently under development by DOE, EPRI, and the nuclear industry.

Volume II includes a comprehensive survey of the integrated capability developed at Los Alamos for the design, fabrication, and testing of prototype spent-fuel measurement systems. This capability includes state-of-the-art computer programs for the calculation of radiation source terms and emissions from spent fuel; a hot-cell experimental facility for handling unirradiated fuel materials and a variety of neutron and gamma-ray sources; a wide range of instrumentation for experimentation, data collection, and analysis; and an integrated capability for fielding all types of nondestructive measurement systems at spent-fuel storage facilities.

TABLE S-I  
LEVELS OF SPENT-FUEL VERIFICATION AND NONDESTRUCTIVE TECHNIQUES

Level of Verification	Gamma-Ray	Neutron	Other
Physical characteristics	Not applicable	Not applicable	(1) Item counting (2) Coloration (3) Mass by weighing (4) Serial number
Physical integrity of fuel assemblies	<u>Gross Changes</u> (1) Comparison of relative intensity of specific high-energy gamma rays (2) Comparison of relative values of measured isotope activity ratios	<u>Gross Changes</u> Comparison of relative neutron emission rate	(1) Cerenkov (2) Mass by weighing (3) $Be(\gamma, n)^a$ (4) TLD <sup>b</sup> (5) Seals
Indication of irradiation	Simple gross gamma-ray detection techniques	Simple passive neutron detection techniques	(1) Cerenkov (2) Detection of heat (3) TLD <sup>b</sup>
Presence of fission products or actinides	Low- or high-resolution techniques for detection of $^{137}Cs$ , $^{134}Cs$ , $^{144}Pr$ , and others	Verification of neutron rates expected for declared burnup	$Be(\gamma, n)^a$
Relative concentrations of fission products or actinides	(1) Correlations of ratios to burnup, cooling time, and initial enrichment (2) Consistency of measured and declared values	<u>Passive:</u> Relative burnup values $N = \alpha(\text{burnup})^{\beta}$ <u>Active:</u> Relative fissile content	$Be(\gamma, n)^a$
Determination of nuclear material content	Correlations between HRGS results and destructive analyses or theoretical calculations	<u>Passive:</u> Correlations with plutonium content <u>Active:</u> Absolute fissile content	Not applicable

<sup>a</sup>Detector sensitive to high-energy gamma rays.

<sup>b</sup>Thermoluminescence detector.

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## NONDESTRUCTIVE VERIFICATION AND ASSAY SYSTEMS FOR SPENT FUELS

by

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### ABSTRACT

This is an interim report of a study concerning the potential application of nondestructive measurements on irradiated light-water-reactor (LWR) fuels at spent-fuel storage facilities. It describes nondestructive measurement techniques and instruments that can provide useful data for more effective in-plant nuclear materials management, better safeguards and criticality safety, and more efficient storage of spent LWR fuel. In particular, several nondestructive measurement devices are already available so that utilities can implement new fuel-management and storage technologies for better use of existing spent-fuel storage capacity. The design of an engineered prototype in-plant spent-fuel measurement system is 180% complete. This system would support improved spent-fuel storage and also efficient fissile recovery if spent-fuel reprocessing becomes a reality.

## I. INTRODUCTION

### A. Synopsis of US Spent-Fuel Storage Policy

In April 1977, President Carter announced the indefinite deferral of spent-fuel reprocessing. In October 1977, the US Department of Energy (DOE) announced a companion policy to accept and take title to spent nuclear fuel accumulating at domestic reactor sites and a limited amount of spent fuel from overseas. To implement this policy an Interim Spent-Fuel Storage Project Office was established at DOE Savannah River Operations Office. Research begun then has produced a generic environmental impact statement for the new policy,<sup>1</sup> an on-going study of the need for away-from-reactor (AFR) spent-fuel storage,<sup>2</sup> and a scoping study of the AFR interim spent-fuel storage concept.<sup>3</sup>

These studies indicate that the projected need for AFR storage capacity is highly uncertain. It depends strongly on assumptions concerning the

installation of high-density storage racks by utility operators, the maintenance of full-core reserve storage capacity, and the possible transshipment of spent fuel between reactor sites. Thus, the need for AFR storage was questioned, notably by the General Accounting Office (GAO).<sup>4</sup> For planning purposes DOE selected a base case of 755 t of excess spent fuel by 1985.<sup>3</sup> However, even this assumption was questioned, and on March 9, 1981, DOE announced that it was discontinuing Federal AFR storage but would support the development of technology for more efficient storage of spent fuel at the reactor sites and would encourage commercial spent-fuel reprocessing.

The DOE announcement means that the utilities will continue to bear the primary responsibility for storage and maintenance of irradiated fuel. Increased fuel costs and the uncertainty associated with the so-called "back end" of the fuel cycle

will force the nuclear power industry to implement several rather costly measures to improve fuel utilization, to maximize the use of existing fuel-storage capacity, and to modify some current practices.

1. High-Density Racks. High-density storage racks contain a neutron absorber (boron) so that the packing density of spent-fuel assemblies can be increased, perhaps by 50-70%. Most utilities have now applied for license amendments to permit installation of high-density racks in their existing storage pools.

2. Transshipment of Spent Fuel. As the name implies, this involves transporting spent fuel among several storage sites for optimum utilization of storage capacity. The Nuclear Regulatory Commission (NRC) is currently permitting this on a case by case basis, where there is a severe shortage of storage space, but it is doubtful that this practice will become routine because of its political sensitivity.

3. Relinquishing Full-Core Reserve Capacity. This reserve capacity is for removal and storage of the entire core in the cooling pond in case of emergency. This is not required by NRC, but no utility will willingly operate without it; thus, it is imprudent to rely on this "extra" capacity in planning for spent-fuel storage requirements.

4. Rod Storage. When rods are stored, they are removed from assemblies and packed in storage canisters. The increase in effective storage capacity over normal assembly storage may be ~50-70%. (Close-packed rods are less reactive under water than full assemblies.) This technique appears promising and is in the development stage.<sup>5-8</sup> However, the safety and safeguards implications of disassembly and rod storage must be investigated further before the rod storage technique can be put into routine use.

5. High-Burnup Fuels. Modern power plants are designed to use high-burnup fuels, up to 40 000 MWD/tU\* or more for boiling-water-reactor (BWR)

\*Megawatt-days (thermal) per metric ton of initial uranium. Note that this is properly the unit of exposure; however, this quantity is commonly referred to as burnup. The term burnup is used consistently throughout this report.

fuel and 50 000 MWD/tU or more for pressurized-water-reactor (PWR) fuel. This means that more reactive, higher initial enrichment (>4% in <sup>235</sup>U) fuels are being fabricated and tested under the uranium utilization program supported by DOE and the Electric Power Research Institute (EPRI). Some of these fuels cannot be stored in high-density racks unless it can be verified that they have undergone at least one cycle of irradiation in the reactor. The NRC currently licenses fuel storage in terms of criticality limits under the conservative assumption that the spent fuel is as reactive as fresh fuel. A burnup calculation currently is not acceptable as an indicator of reduced reactivity, but a measured value of burnup might eventually be accepted, thereby increasing the effective storage capacity of existing pools for all types of spent fuel.

6. Reconstitution of Assemblies. Reconstituting an assembly involves disassembling one or more irradiated fuel assemblies, repackaging new assemblies using irradiated rods plus possibly some unirradiated fresh rods, and storing the "used-up" rods, either in partially filled assemblies or in rod-storage canisters. The technique of reconstituting assemblies has been demonstrated for both BWR and PWR fuels and is in an advanced stage of development. The basis for sorting the rods in irradiated assemblies, in lieu of a direct measurement, presumably depends on the average burnup calculated for the assembly. However, some type of direct nondestructive examination of the irradiated assemblies would be useful for sorting the rods, and some type of quality assurance might be required for reconstituted assemblies before they could be returned to the reactor.

#### B. Los Alamos Participation in the Interim Spent-Fuel Storage Program

Since June 1979, the Los Alamos safeguards research and development groups, supported through the Interim Spent-Fuel Storage Project Office at Savannah River and with technical review by Savannah River Laboratory, have been developing nondestructive measurement technology for spent-fuel verification and assay. The principal objective is to design, fabricate, and demonstrate a prototype verification and assay system at a

selected spent-fuel storage facility in the FY82-83 period. Such systems would provide storage-facility operators with base-line data on burnup and plutonium content of irradiated light-water-reactor (LWR) fuels, which could be used to satisfy safeguards and nonproliferation requirements and operating requirements for safe and efficient storage.

Los Alamos has successfully demonstrated, both by calculation using the EPRI-CINDER code and by experimentation at various spent-fuel storage sites, that certain gamma-ray and neutron emissions correlate directly with burnup and fissile content. Laboratory demonstrations show that the measured neutron response to the interrogation of unirradiated fuel assemblies by an external neutron source is sensitive to changes as small as 1% of the fissile content. That is, if one or at most a few low-enriched uranium fuel rods are replaced with depleted uranium rods, neutron response is affected.

During the Fall of 1980 a series of experiments was initiated at the GE-Morris Operation Spent-Fuel Storage Facility to gather basic physics and facility operating data for designing a spent-fuel verification and assay system. The experimental results are especially important in specifying engineering design requirements for operationally acceptable in-plant measurement systems. Companion experiments also were fielded at GE-Morris to demonstrate portable nondestructive assay (NDA) equipment, including the Cerenkov viewing device and the neutron ring, which were developed under the auspices of the US technical assistance program to the International Atomic Energy Agency (IAEA). These portable NDA instruments were developed to help safeguards inspectors verify spent fuel in situ with minimum disruption of the operations at a facility.

This effort was initiated before the Los Alamos involvement in the interim spent-fuel storage program.<sup>9</sup> Thus, our effort under the spent-fuel storage program could build on experience gained previously under the international safeguards program. We prepared a report for Savannah River summarizing the current experience in applying the Cerenkov technique at a variety of spent-

fuel storage facilities<sup>10</sup> and delivered a Cerenkov viewing device to Savannah River personnel for evaluation. We provided Savannah River with a second report that reviews passive neutron and gamma-ray techniques developed for spent-fuel verification by IAEA inspectors.<sup>11</sup>

An earlier concept study of safeguards for spent nuclear fuels was carried out for the DOE Office of Safeguards and Security (DOE/OSS).<sup>12</sup> This study indicated that, in safeguards, spent fuel is primarily of concern for international safeguards and nonproliferation. Efforts to develop portable nondestructive instruments for verification of spent fuel were endorsed, and, at the same time, it was recommended that NDA instruments be considered for in-plant use by operators of spent-fuel storage facilities. A safeguards and nonproliferation study by Savannah River Laboratory<sup>13</sup> later reinforced and expanded these conclusions.

We have documented Los Alamos progress under the interim spent-fuel storage program in a series of informal monthly letter reports and in a progress report.<sup>14</sup> The present report, supplemented by Refs. 10, 11, and 14, encompasses all Los Alamos activities under the interim spent-fuel storage program and for the most part replaces all previous reports.

### C. Scope and Organization of the Report

The report consists of two volumes. The first comprises work on the conceptual design of a non-destructive verification and assay system (VAS) for spent-fuel storage facilities, including sections on the role of nondestructive measurements in spent-fuel storage facilities, the description of a reference spent-fuel storage facility, the conceptual VAS system, VAS measurement subsystems, and conclusions and recommendations. The second volume comprises six appendixes consisting of essential supporting technical information for the study.

## II. THE POTENTIAL ROLE OF NONDESTRUCTIVE MEASUREMENTS AT SPENT-FUEL STORAGE FACILITIES

Until recently, nondestructive measurements on spent fuel were used almost exclusively for

postirradiation examination of irradiated fuel materials at specially equipped facilities. Several new NDA techniques and instruments now are either available or in an advanced stage of development for various uses in spent-fuel storage facilities.

#### A. In-Plant Materials Management

Operators of spent-fuel storage facilities are concerned with criticality safety and radiological protection. These functions are closely related to materials control and accounting. The storage of high-burnup fuel assemblies or close-packed rods in high-density racks requires nondestructive verification of the burnup. Under the enhanced uranium utilization program, DOE, EPRI, and the nuclear industry are investigating the application of nondestructive techniques as burnup monitors. Nondestructive measurements that can determine the distribution of the burnup in irradiated fuel assemblies would be especially useful for fuel-bundle disassembly and repackaging of fuel assemblies. Such measurements could be used to separate burned-up rods for storage from those that are reusable in new assemblies. Accurate measurements of the fissile content would provide useful materials management data when the fuel is stored initially and when it is shipped finally for reprocessing or long-term storage.

Nondestructive measurement systems are currently being developed for reprocessing facilities.<sup>15</sup> These measurements are intended primarily as aids to criticality control in the dissolver tank and for efficient batch processing of the spent fuel for maximum fissile recovery. Spent-fuel measurements also could provide better safeguards at fuel reprocessing plants.

Spent fuel currently is received and stored using shipper's (reactor exposure) values. The shipper/receiver (S/R) contents of the input accountability tank are measured. The closure of this balance is complicated because the integrity of the input batch may be compromised by back-cycle streams or heels in the dissolver and accountability tanks, and because losses in the high-activity solid wastes are difficult to measure. A better procedure may be to close the S/R balances using

nondestructive measurements on the spent-fuel assemblies while they are still integral items. Such measurements could then be used to obtain materials balances across the headend chop/leach operation, effectively decoupling the headend process from the solvent-extraction process for purposes of accountability.

#### B. Materials Control and Accountability

Materials control and accountability (MC&A) provides information to the operator and the licensing authorities on the locations and amounts of all special nuclear materials (SNM) in the facility (App. A). Domestic MC&A is based on a system of facility records and reports covering all receipts and shipments of SNM and all SNM on inventory.<sup>16</sup> The operator must verify inventory records by periodic physical inventories, and his MC&A system is subject to verification checks by NRC inspectors at any time.

Physical inventories at power reactor storage basins are usually performed annually and normally involve visual identification of the fuel assemblies in storage. The SNM inventory records maintained by the facility are based on predicted values from burnup codes.

Similar licensing rules and procedures have been established for AFR storage facilities.<sup>17,18</sup> In addition, essentially all licensed nuclear facilities must conform to new reporting requirements established under the US/IAEA safeguards agreement.<sup>19</sup>

The policy of storing spent fuel based on predicted SNM values was established when it was envisioned that the interim storage time of irradiated power reactor fuel before reprocessing would be, at most, a few years. Utility operators are now anticipating storage of irradiated fuel containing significant quantities of plutonium for 10 yr or more. Because of the current uncertainty with regard to the back end of the fuel cycle, sound safeguards and nuclear materials management practice call for a direct verification of the identity, integrity, and SNM content of spent-fuel assemblies. The same comment is reinforced if fuel bundles are disassembled for rod compaction and storage or for repackaging into new assemblies.

The integrities of the original items (assemblies) are destroyed, and new items (rod storage containers or new assemblies) are created. Each such action must be reported to the safeguards authorities (NRC and IAEA) as an inventory change, including data on the SNM contents of the old and new items, which are checked by on-site inspection and verification. Nondestructive measurement techniques can provide the necessary capabilities.

### C. International Safeguards

Since 1978, Los Alamos has been developing portable NDA equipment for use by IAEA inspectors to verify spent-fuel assemblies stored at power reactors. The Cerenkov technique currently is applied by inspectors using a portable, hand-held viewing device,<sup>10</sup> which provides an image of the top of the fuel assembly. Usually the inspector stands on the bridge of the fuel-handling crane, which is positioned over each fuel assembly to be measured. The main advantage of the technique is that the fuel assemblies are measured in situ in the storage racks without introducing any equipment into the storage basin.

Passive gamma-ray and neutron techniques for IAEA uses are also in an advanced stage of development.<sup>11</sup> A prototype device called the neutron ring is being evaluated by the IAEA. As the name implies, this is a closed loop of detectors, usually consisting of a square array of ion chambers and <sup>235</sup>U fission chambers, that is placed over the top of a single storage location in the spent-fuel storage rack. The fuel assembly is partially withdrawn from the storage rack through the ring of detectors using the fuel-handling crane, and gamma-ray and neutron emissions are measured simultaneously on all four sides of the assembly. The measurements can be repeated at various axial positions by lifting the fuel assembly partially out of the storage rack.

The objectives of IAEA safeguards under the Nuclear Nonproliferation Treaty are to detect and deter the diversion of nuclear materials.<sup>20,21</sup> The safeguards goal for irradiated power reactor fuel is to detect a diversion of 8 kg of plutonium in 1-3 months or 75 kg of contained <sup>235</sup>U in 1 yr.<sup>22-24</sup>

For high-burnup fuels, the 8 kg of plutonium approximately corresponds to two PWR or four BWR fuel assemblies. To achieve the safeguards goal, inspectors verify the inventory at spent-fuel storage facilities every 2-4 months by verifying the identity, location, and integrity of the irradiated fuel assemblies. A subset of the fuel assemblies is selected for measurement using a statistical sampling plan. This plan is based on the verification of selected attributes of the fuel assemblies, ranging from visual observations of the assemblies up to the application of nondestructive measurements. The number of stored assemblies that must be examined to meet the safeguards goal with the desired level of assurance can be large, requiring approximately three-quarters of the PWR assemblies and half the BWR assemblies to achieve the 95% confidence level. Thus, the methods used for routine verification must be rapid and must not interfere unreasonably with the normal operation of the facility. This usually dictates the application of semiquantitative, confirmatory NDA techniques. If discrepancies are found during a routine check of the inventory, then a detailed inventory verification may be performed using more quantitative NDA techniques.

### D. Levels of Spent-Fuel Verification and Available NDA Techniques

NDA techniques at spent-fuel storage facilities verify various attributes of spent fuel contained in integral items, either fuel assemblies or storage containers. This verification ranges from detection of gross radiation emissions to the direct measurement of fissile content. No single NDA technique can satisfy all the requirements for nondestructive measurements, that is, cover all types of LWR fuel, all the different spent-fuel parameters and their possible ranges, and all the levels of desired measurement accuracy and precision. Thus, a variety of NDA techniques and instruments is being developed. Table I emphasizes this point and Table II elaborates the capabilities and limitations of currently available NDA techniques.<sup>15</sup>

TABLE I

## LEVELS OF SPENT-FUEL VERIFICATION AND NDA TECHNIQUES

Level of Verification	Gamma-Ray	Neutron	Other
Physical characteristics	Not applicable	Not applicable	(1) Item counting (2) Coloration (3) Mass by weighing (4) Serial number
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Indication of irradiation	Simple gross gamma-ray detection techniques	Simple passive neutron detection techniques	(1) Cerenkov (2) Detection of heat (3) TLD <sup>b</sup>
Presence of fission products or actinides	Low- or high-resolution techniques for detection of <sup>137</sup> Cs, <sup>134</sup> Cs, <sup>144</sup> Pu, and others	Verification of neutron rates expected for declared burnup	Be( $\gamma$ ,n) <sup>a</sup>
Relative concentrations of fission products or actinides	(1) Correlations of ratios to burnup, cooling time, and initial enrichment (2) Consistency of measured and declared values	<u>Passive:</u> Relative burnup values $N = \alpha(\text{burnup})^{\beta}$ <u>Active:</u> Relative fissile contents	Be( $\gamma$ ,n) <sup>a</sup>
Determination of nuclear material content	Correlations between HRGS results and destructive analyses or theoretical calculations	<u>Passive:</u> Correlations with plutonium content <u>Active:</u> Absolute fissile content	Not applicable

<sup>a</sup>Detector sensitive to high-energy gamma rays.

<sup>b</sup>Thermoluminescence detector.

## III. REFERENCE SPENT-FUEL STORAGE FACILITY

Two generic classes of spent-fuel storage facilities are being considered: water-basin storage and dry-surface storage. Because much more operational experience is available for water-basin storage, we have selected it as our reference facility. In particular, we have selected the GE-Morris Operation Spent-Fuel Storage Facility as our model.<sup>25-27</sup> The Spent-Fuel Storage Facility was originally designed as the interim storage pool at the headend of the reprocessing plant; however, now its principal use is the storage of spent nuclear fuel under conditions that maintain the fuel integrity and assure the health and safety of the operating personnel and the general public.

The facility has been licensed and used as a spent-fuel storage facility since January 1972. It is licensed to store 750 t of spent nuclear fuel from PWR and BWR reactors. The Morris facility is described in detail in Refs. 25-27. The following is an abbreviated description, emphasizing the spent-fuel unloading facilities that are of special interest for spent-fuel nondestructive measurements.

The configuration of the unloading and storage basins is shown in Fig. 1. The basins are constructed of reinforced concrete poured against bedrock and lined with stainless steel. Channels in the concrete behind the stainless-steel liner collect any water that might leak through the liner

TABLE II  
SPENT-FUEL NONDESTRUCTIVE MEASUREMENTS

Measurement Type	Capabilities	Limitations
Cerenkov light	Imaging device; radioactive material; rapid; simple instrument above water; no fuel handling; well developed	Nonspecific; self-shielding; semiquantitative
Gross gamma	Gamma dose rate; burnup profiles; rapid; simple instruments; minimal fuel handling; well developed	Nonspecific; self-shielding; semiquantitative
Gamma spectrometry	Gamma spectra; fission-product specific; correlations with burnup and cooling time; well developed	Self-shielding; relatively complex; relatively slow; fuel handling; affected by changes in geometry
Passive neutron	Neutron dose rate; interior rods; correlations with burnup and plutonium content; relatively simple; relatively fast; minimal fuel handling; well developed	Nonspecific; cooling time dependence
Active neutron	Fissile content; interior rods; independent assay; steady-state technique using $^{252}\text{Cf}$ ready for field testing	Relatively complex; relatively slow; requires external neutron source; fuel handling; affected by changes in geometry; pulsed-neutron technique still in R&D stage

or in through the concrete. This water is collected in a sump with a level-detection alarm system. The three basins contain 675 000 gal of water. No neutron poison materials are used. Minimum water depth above the spent-fuel assemblies in storage is 4-7 m. This minimum depth is decreased to 3 m during fuel-transfer operations. At the present time, ~310 t of fuel are stored in the two storage basins. Individual fuel assemblies are stored in specially designed storage baskets--four PWR fuel assemblies or nine BWR fuel assemblies per basket.

Fuel assemblies are shipped to the GE-Morris Operation either in a railcar-mounted cask, usually capable of carrying 7 PWR or 18 BWR fuel assemblies, or in a highway-trailer-mounted cask capable of carrying 1 PWR or 2 BWR fuel assemblies. The shipping cask is placed on the unloading pit shelf, and the cask-head fasteners are removed. Then the

cask is lowered into the bottom of the unloading pit where the cask head is removed. The appropriate fuel-storage basket is moved into the unloading pit to receive the fuel assemblies from the cask. Each fuel assembly is removed individually from the shipping cask and is placed in the storage basket. At this time in the fuel receiving operation, the fuel assembly could be examined nondestructively with minimum impact upon the facility operation. A nondestructive examination at this point in the unloading operation would increase the unloading time by the measurement time; however, any other measurement procedure would require additional fuel handling. If the examination were not completed at this time, the fuel assemblies would have to be retrieved at a later time from the storage basins for examination. The cask unloading basin is the only area of the facility equipped for handling individual fuel assemblies.

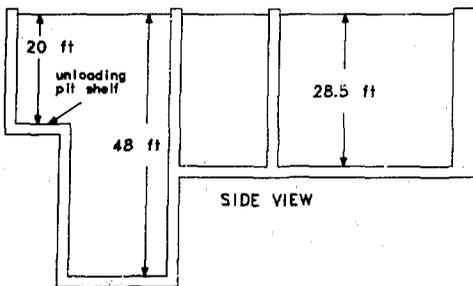
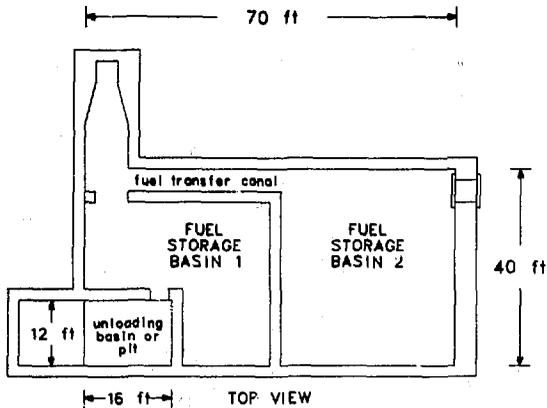


Fig. 1.

GE-Morris spent-fuel storage facility.

Once the fuel assemblies are placed in the fuel-storage baskets, the baskets are moved into storage basin Nos. 1 or 2 for storage, the shipping cask head is replaced, and the cask is moved to a decontamination pad for cleanup before return shipment to the reactor facility. This entire fuel unloading process requires 20-48 h for the casks presently in use.

Figures 2 and 3 show a BWR railway cask and a PWR highway cask in the bottom of the unloading pit. It is evident from the two photographs that there is very little space available for a nondestructive measurement system. The system must be designed to be placed in the unloading pit so that it does not interfere with the normal fuel unloading operations. Appendix B describes an experimental measurement apparatus currently installed in the unloading basin at GE-Morris. Section IV describes a conceptual nondestructive VAS for in-plant use.

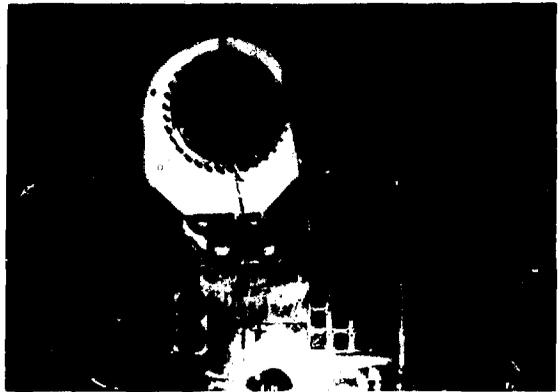


Fig. 2.

BWR railway cask in the unloading pit at GE-Morris Operation.

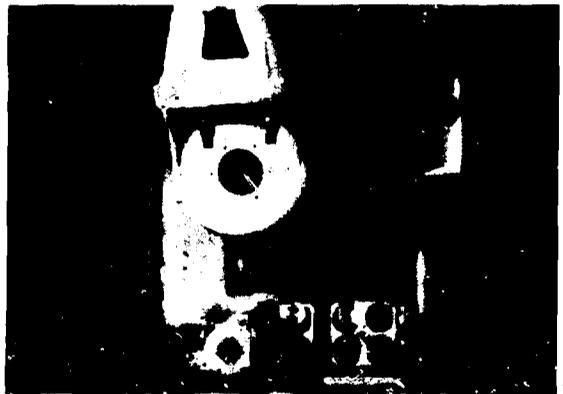


Fig. 3.

PWR highway cask in the unloading pit at GE-Morris Operation.

#### IV. VERIFICATION AND ASSAY SYSTEM

##### A. Objective

The overall objective of the VAS is to provide timely nondestructive data on irradiated fuel assemblies to satisfy the MC&A and operational requirements described in Sec. II. No single NDA technique can meet this objective for the different types of reactor fuel and the ranges of fuel parameters that may be encountered (Sec. II, Table I). Therefore, a combination of nondestructive techniques that are mutually complementary and that together are comprehensive is necessary. In addition, prototype systems must be tested for

operational compatibility and acceptability in actual spent-fuel storage facilities.

### B. Functional Description

Basically, the VAS will provide two kinds of data. The first kind includes burnup and cooling-time values to verify the identity of each fuel assembly and to compare with reactor-operator-declared values. These data and comparisons would be available to the storage facility operator essentially in real time to identify problems and gross discrepancies before placing the fuel in the storage basin.

The second kind of data includes estimates of the fissile content, the plutonium content, and the remaining  $^{235}\text{U}$  content. These values also are compared with reactor-operator-declared values obtained from burnup calculations. Statistically significant differences would be investigated, and a set of "best values" would be determined and entered into the inventory records maintained at the storage facility. Because this second kind of assay data need not be as timely as the first kind, there is time for investigation of differences and analysis of the data to determine best values. Normally, best values would consist of an appropriately weighted linear combination of the calculated and measured values.

Figure 4 shows a functional layout of the VAS system. Four types of nondestructive measurements are obtained to cover the anticipated fuel types and fuel-parameter ranges. Gross gamma-ray and passive neutron data provide axial and angular profiles of the burnup. These burnup profiles are normalized by absolute burnup measurements obtained from high-resolution gamma-ray and active neutron measurements. Assay data are obtained by combining and analyzing the gamma-ray and neutron data using a small computer (the VAS computer), which is also used to make preliminary comparisons with the declared attributes of the fuel and to alert the operator immediately if there are gross discrepancies. The final assay values for each fuel assembly are entered into the facility records. We assume that these records are maintained at the facility on a central computer that also maintains data relevant to other facility functions, such as fuel handling, storage, criticality safety, and radiological protection.

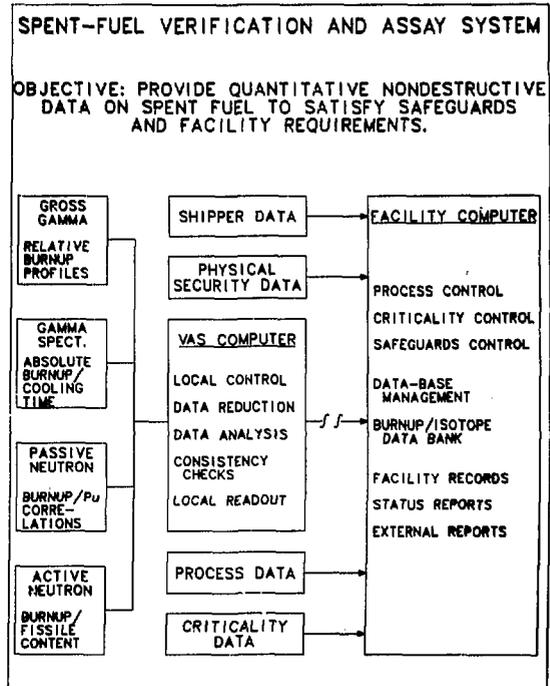


Fig. 4.  
Functional description of the VAS system.

### C. Design Requirements

1. Design Considerations. The proposed VAS system is the first spent-fuel measurement system that uses four types of radiation measurements: gross gamma-ray, high-resolution gamma-ray spectroscopy, passive neutron, and active neutron. The design of each of the measurement subsystems is considered in Sec. V. General design considerations for the entire VAS system can be summarized as follows.

- (1) The system will measure entire fuel assemblies underwater.
- (2) The measurements will be completed in ~0.5 h.
- (3) The system will occupy minimal space.
- (4) The system will meet applicable safety and licensing requirements.
- (5) The system will be highly reliable and simple-to-operate and maintain.
- (6) The system will be capable of measuring both PWR and BWR fuel assemblies that might be encountered at the spent-fuel storage facilities.

(7) The system will have flexibility of design so that changes in measurement requirements can be accommodated.

(8) The system will be thoroughly tested in actual spent-fuel storage facilities.

Each type of measurement requires precise positioning of the radiation detectors relative to the fuel assembly. To achieve a measurement accuracy of 1%, it is desirable to maintain detector-to-assembly distances constant to within a few millimeters.

For both passive and active neutron measurements, access to at least two opposite sides of the assembly is required. For measuring flux tilts using gross gamma-ray methods, access to a minimum of three sides of the assembly is required, and access to the fourth side is desirable. Routine high-resolution gamma-ray measurements are planned for only one axial position on the assembly because of measurement time constraints (500- to 1000-s measurement times).

The  $^{252}\text{Cf}$  source used for active neutron measurements is moved vertically away from the fuel assembly during the passive neutron measurements. It must then be repositioned precisely (relative to the detectors) for active neutron measurements.

For a number of reasons, it appears desirable not to move the fuel assembly during standard measurement procedures. If acceptable to the facility operator, the simplest approach would be to position the assembly with the fuel handling crane and to leave the assembly attached to the crane during the measurement.

Some thought has been given to the question of whether passive neutron and gross gamma burn-up-profile systems should consist of a single detector that is stepped along the assembly or should consist of a number of detectors accumulating data in parallel. At present neither option has been eliminated, but there is a leaning toward the use of multiple detectors placed at specified axial positions and operated in parallel.

2. Design Approach. Based on the considerations outlined above, the best design approach is to place components for gross gamma-ray, passive neutron, and active neutron measurement systems in

vertical pipes (750-100 mm in diameter by 10 m long) that extend from the top of the pool to the assembly, as shown schematically in Fig. 5. The measurement components (detectors, shielding,  $^{252}\text{Cf}$  sources, and cables) would then be mounted in rigid canisters that fit inside the vertical pipes. This arrangement should provide the desired ease of maintenance and design flexibility. The high-resolution gamma-ray measurement uses an air-filled slant collimator pipe, with a germanium detector mounted at poolside, also shown in Fig. 5.

Several arrangements were considered for the horizontal layout of vertical pipes and slant collimator relative to the fuel assembly. Figure 6 shows an arrangement in which three vertical pipes are rigidly mounted to a U-shaped mechanical frame, sized for PWR assemblies. Hollow aluminum inserts are used as a spacer when BWR assemblies are to be measured. Although this arrangement has the significant advantage of simplicity, it also has certain drawbacks. Because not all BWR and PWR assemblies have the same cross-sectional dimensions, it may be necessary to use several different inserts. It might be difficult to maintain (with this

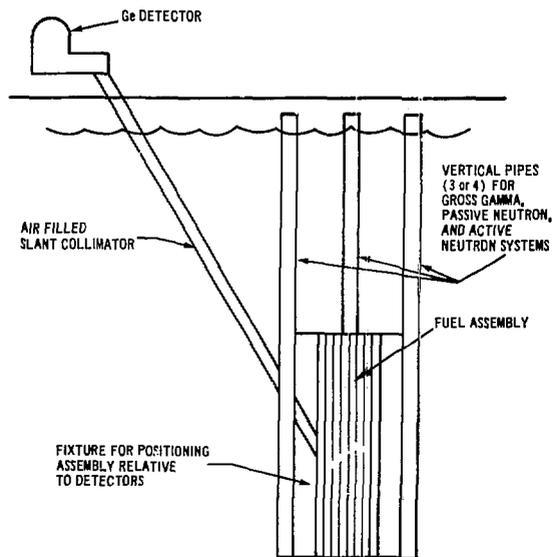


Fig. 5.  
General features of the VAS measurement system.

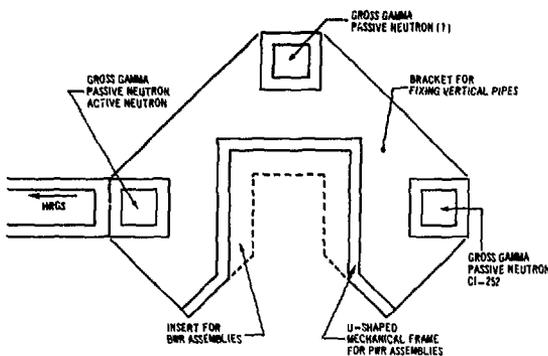


Fig. 6.  
Cross section of measurement system option that uses mechanical inserts to position BWR and PWR assemblies relative to detectors and  $^{252}\text{Cf}$  source.

arrangement) a detector-to-assembly tolerance of a few millimeters, and there may be fuel handling problems in placing an assembly in a U-shaped fixture. Furthermore, access to the fourth side of the assembly is denied.

Figure 7 shows two horizontal layouts, both of which employ two L-shaped "clam shells" to position detectors on four sides of the assembly. In this arrangement, one of the "L's" is fixed and the other one moves. The assembly is placed adjacent to the fixed L and the movable L is brought into position for either a BWR or PWR assembly. Although this arrangement can accommodate a variety of fuel sizes, it is a complex design that will require careful engineering. Space requirements for the two approaches shown in Figs. 6 and 7 are slightly different. The clam-shell layout (Fig. 7) requires slightly more floor space in the pool, and the U-shaped layout requires space to store the inserts when they are not in use. Another possible arrangement would be to design two separate measurement systems, one optimized for PWR assemblies and the other optimized for BWR assemblies.

A decision regarding the layout of the measurement system must be based on the actual characteristics of the specific facility where the instrument is to be installed. The availability of space in the pool, fuel-handling procedures, and safety requirements are overriding factors. Fortunately, it appears that a layout of the measure-

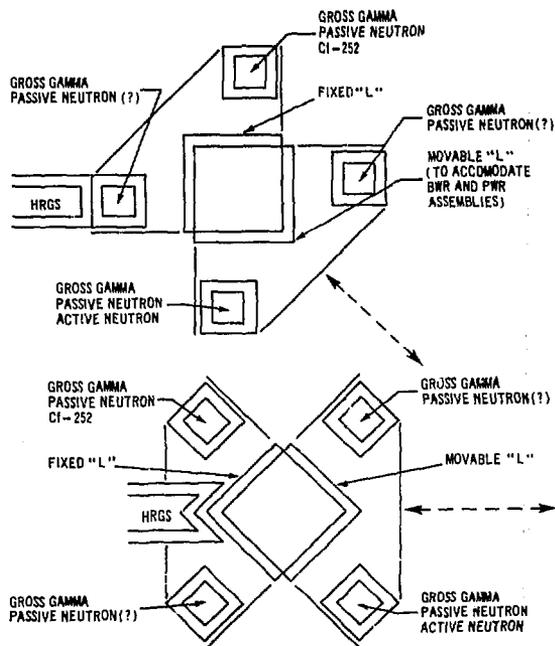


Fig. 7.  
Cross sections of measurement systems that use clam-shell arrangements to position BWR and PWR assemblies relative to detectors and  $^{252}\text{Cf}$  source.

ment system can be selected that is compatible with the practical constraints of existing facilities. The present design is for power reactor fuel assemblies. The design consideration would be similar for a system to measure other types of fuel assemblies, for example, materials test reactor (MTR) fuel elements and liquid-metal fast breeder reactor (LMFBR) fuel assemblies.

#### D. Status of the Conceptual VAS Design

At the time when this study was terminated because of a shift in spent-fuel storage policy, the design of a prototype VAS system was ~80% complete. This prototype system is not intended to be the final design of an engineered system for in-plant use, but it is an R&D tool designed for field testing and evaluation, leading to a final engineered VAS design. As such, it represents the next logical step beyond the experimental apparatus currently installed at the GE-Morris storage facility. (The current system at GE-Morris is described in App. B.)

The new prototype experimental system is illustrated in Fig. 8. It consists of a rigid, vertical array of U-shaped fuel-assembly-positioning and detector-locating brackets. (A single bracket is shown in Fig. 8.) The U-shaped brackets are sized for PWR assemblies, and vertical pipes are used to locate gross gamma and passive neutron detectors symmetrically at the four assembly corners. The gross gamma and passive neutron detectors are placed at various axial positions (five to nine total) in each of the vertical pipes. BWR assemblies could also be measured using this arrangement, but with some loss of measurement symmetry. The opening of the U is oriented parallel to the storage-basin wall so that the gamma-ray slant-collimator pipe connected to the detector above the surface of the water can be located parallel to the basin wall and, at the same time, not interfere with any of the vertical detector pipes. A  $^{252}\text{Cf}$  source is placed in a vertical pipe located at the center of one side of a standard-sized PWR assembly, along with a so-called neutron-tailoring region designed to interrogate the cross section of the fuel assembly more or less uniformly from a single neutron source. Another vertical detector pipe is placed at a point opposite the  $^{252}\text{Cf}$  source for uniform counting of the induced fission neutrons. The  $^{252}\text{Cf}$  source can be moved to a vertical position away from the fuel assembly for passive neutron and gamma-ray counting, or it can be positioned at various axial positions along the fuel assembly for active neutron measurements.

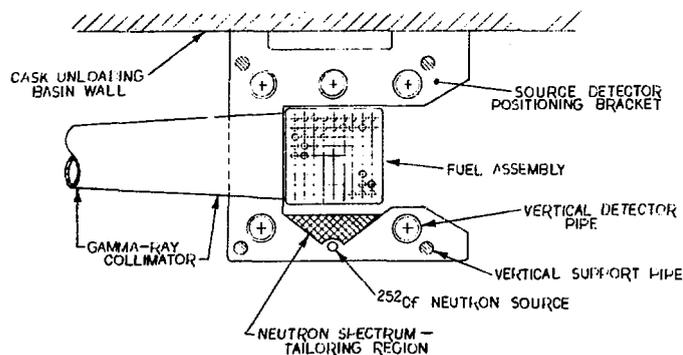


Fig. 8. Conceptual design of a prototype VAS instrument. The spent-fuel positioning and detector bracket is shown. Profile measurements are made at several axial positions. The data are collected, analyzed, and displayed under micro-processor control.

## E. Prediction of Spent-Fuel Parameters from Combined Nondestructive Measurements

As shown in Fig. 9, four different measurement methods are proposed: high-resolution gamma-ray, gross gamma-ray, passive neutron, and active neutron. The desired assay results are burnup, cooling time, fissile content, and plutonium content. Thus, four types of measurements are available, and the desired assay results are obtained from suitable combinations of the measurements.

### 1. Parameters Determined.

a. High-Resolution Gamma-Ray Spectrometry (HRGS). Gamma radiation emitted by spent fuel arises mainly from the fission products. With a high-resolution gamma-ray spectrometer, certain fission products can be identified from their characteristic gamma rays. A HRGS system can also be used to determine burnup, either from absolute fission-product activity ( $^{137}\text{Cs}$ ) or from activity ratios ( $^{134}\text{Cs}/^{137}\text{Cs}$ ,  $^{154}\text{Eu}/^{137}\text{Cs}$ ). Using fission products with substantially different half-lives (such as  $^{95}\text{Zr}$ ,  $^{144}\text{Pr}$ , and  $^{137}\text{Cs}$ ), cooling time also can be estimated.

There are two major drawbacks to HRGS: it requires a relatively long measurement time (about 15 min/measurement), and it only assays the first few outer rows of fuel pins in the assembly because of source self-attenuation. Because of the assay time, only one HRGS measurement would be made routinely per assembly. Other more rapid (but less accurate) axial burnup measurements can be combined with the single HRGS measurement to determine average burnup for the fuel assembly.

# VAS DATA INTERPRETATION

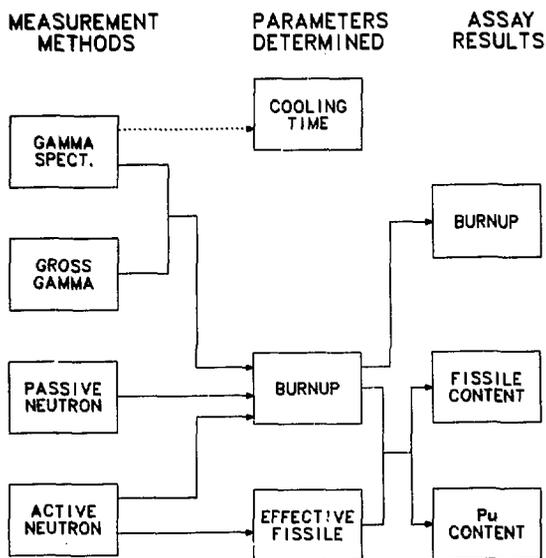


Fig. 9. Spent-fuel parameters obtained from combined nondestructive measurements.

**b. Gross Gamma Ray.** For rapid determination of the axial burnup distribution, it is possible to use the gross gamma signature measured with one or more detectors, such as ion chambers. The assay time is relatively short (5-10 s/position). However, there are some disadvantages of gross-gamma measurements; for example, not all of the fission products ( $^{134}\text{Cs}$ ,  $^{154}\text{Eu}$ ) are linearly proportional to burnup, and, in the case of high-burnup assemblies, these nuclides may account for a substantial portion of the gross gamma radiation. In this situation, a detector sensitive to high-energy gamma rays (>1.6 MeV), such as the  $\text{Be}(\gamma, n)$  detector, provides a more accurate burnup profile, but the assay time is somewhat longer ( $\sim 2$  min/position).

Because in some reactors the burnup may not be uniform across the assembly, it is also advisable to measure the angular burnup distribution. The same detectors [ion chambers,  $\text{Be}(\gamma, n)$  detectors] used for axial profiles can also be used for this application.

**c. Passive Neutron.** The intrinsic drawback of any gamma-ray assay is that it is sensitive predominantly to the outer few rows of fuel rods of the assembly. The passive neutron assay, however, is different; it is more sensitive to the interior rods of the assembly because of the higher penetrability of the neutrons. Passive neutron emission from spent fuel arises mainly from the  $^{242}\text{Cm}$  and  $^{244}\text{Cm}$  nuclides for burnups above 10 GWd/tU. Because of the relatively short half-life of  $^{242}\text{Cm}$  ( $t_{1/2} = 163$  days), passive neutron emission is sensitive to cooling time during the first 2 yr of cooling. For longer cooling times the neutron emission rate depends primarily upon the 18.11-yr half-life of  $^{244}\text{Cm}$ . For cooling longer than 2 yr, passive neutron emission is approximately proportional to burnup raised to the fourth power. Because of this relationship, passive neutron count rate can be used to measure the axial burnup profile of a single fuel assembly or the relative burnup values of a set of assemblies.

The major disadvantages of passive neutron assay are (1) for cooling times shorter than 2 yr, the method is difficult to interpret because of the  $^{242}\text{Cm}$  contribution and (2) correlations of passive neutron emission with burnup may be difficult to determine accurately for all types of FWR and BWR fuels.

**d. Active Neutron.** The three methods discussed above measure burnup indirectly by monitoring the buildup of certain fission products or by determining the concentration of specific actinides. The active neutron technique, however, measures directly the remaining fissile content of the spent fuel. Active-neutron-interrogation methods can be designed such that they provide relatively uniform response across the assembly. With active interrogation, a linear combination of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  and, to a lesser degree,  $^{241}\text{Pu}$  is measured. The contribution of  $^{241}\text{Pu}$  is significant only at high burnup. Because the active neutron measurement requires an assay time of 10-20 min, only one such measurement along the assembly can be performed routinely.

**2. Verification.** The division between assay and verification is somewhat subjective. Roughly

speaking, nondestructive spent-fuel verification can be divided into the following levels:

- (1) dose rate,
- (2) cooling time and fission products,
- (3) burnup profiles, and
- (4) burnup.

a. Dose Rate. Both gamma dose rate and neutron count rate depend to varying degrees on the cooling time, burnup, and irradiation history. The measured gamma and neutron dose rates can be checked to determine whether they are consistent with the declared burnup and cooling time.

b. Cooling Time and Fission Products. For the next level of verification, cooling time estimated from HRGS results can be compared with the declared value, and, in addition, the gamma-ray spectra can be checked to see that they are indeed typical of the expected fission products, and are not due to other radiation sources.

c. Burnup Profiles. The burnup profiles measured by two different methods, gross gamma and passive neutron, can be checked to see if they are in agreement and consistent with expected profiles. The burnup profiles measured by gamma and neutron methods may be different for legitimate reasons, and these differences should be explainable in terms of irradiation history.

d. Burnup. The burnup can be arrived at by three different methods: spectral gamma-ray measurement, passive neutron measurement, and active neutron measurement. Burnup can be measured using high-resolution gamma spectrometry to determine the amount of specific fission products present in the fuel assembly. The absolute intensity of  $^{137}\text{Cs}$  or the isotopic ratios  $^{134}\text{Cs}/^{137}\text{Cs}$  or  $^{154}\text{Eu}/^{137}\text{Cs}$  can be correlated with burnup using destructive analysis for confirmation. Similarly, passive neutron emission rates for fuel assemblies can be used to estimate burnup. Active neutron measurements involve the determination of the remaining fissile content in a fuel assembly, which can be directly related to burnup. For the most advanced verification, the burnup values determined from the three measurements would be examined, the differences explained, and a best value determined in conjunction with the operator-declared value.

3. Assay. Some of the quantities measured for verification, such as burnup and cooling time, can also be used as assay results. In addition to the burnup and cooling time, the fissile content and total plutonium content can be estimated from the passive measurements using isotope correlations and can be determined directly from the active neutron measurements. The procedure is as follows. From the average burnup for an assembly, total plutonium content can be calculated by the Pu/U vs burnup correlation, provided the initial uranium inventory is known. In addition, calculations indicate that passive neutron and gamma-ray signatures correlate directly with the fissile and plutonium contents. These correlations would be compared with the effective fissile content measured by active neutron interrogation to obtain estimates of the separate fissile contents ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$  plus  $^{241}\text{Pu}$ ) and the plutonium content.

Thus, it appears possible to start from the four types of nondestructive measurement methods and arrive at the desired verification and assay data for spent fuel, that is, to start with HRGS, gross gamma, passive neutron, and active neutron measurements, and arrive at estimates of burnup, cooling time, fissile content, and plutonium content. The detailed data-interpretation procedures must still be developed and tested experimentally. A program to develop these procedures is in progress using data, both destructive and nondestructive, obtained for well-characterized burnup samples.

## V. MEASUREMENT SUBSYSTEMS

Each of the four VAS measurement subsystems is described in this section. Additional supporting technical information is provided in the appendixes (Vol. II). Appendix C is a reasonably comprehensive bibliography of reports and symposia papers pertaining to the nondestructive measurement of spent fuel.

### A. Gross Gamma-Ray Profile

1. Objective. To obtain a gross gamma-ray profile that is representative of the relative axial burnup of a BWR or a PWR fuel assembly. This

axial profile is used as an integrating function for relating spectral gamma-ray, passive neutron, and active neutron measurements at a specific axial location to the entire fuel assembly.

**2. Equipment.** The basic hardware for measuring the gross gamma activity profile of irradiated fuel assemblies was developed and tested during previous spent-fuel exercises (App. B). The unit consists of a microprocessor with an RS-232-compatible communications port that will support any defined ASCII protocol, digital electronics (with the exception of preamplifiers) for measuring fission-chamber output at count rates up to 500 000 counts/s and analog electronics that will support up to 48 ion-chamber input channels (Fig. 10). The current system is expandable in units of 16 analog channels and 4 digital channels. The analog electronics circuitry is shown in Fig. 11. A negative bias is applied, and all the detectors are operated from the same power supply. This mode of operation reduces the adverse effect of current leakage. Because of the extremely low current levels ( $5 \times 10^{-12}$  to  $10^{-8}$  A), the signals cannot be multiplexed before the conversion to voltage. Isolation resistances of  $10^{12} \Omega$  are maintained.

The microprocessor increases the versatility of the measurement system, for example, by allowing multiple measurements to be performed with the ion chamber to obtain an average measurement. Design of the chassis is well documented as is the software package.<sup>28</sup> The serial data link is accomplished using standard RS-232 hardware and can

support any ASCII protocol. The interface between the microprocessor and any other processor, such as the VAS central computer, is straightforward requiring minimal software support once the protocol is defined.

**3. Data Analysis.** The use of gross gamma-ray profiles as burnup integrating functions was investigated for both BWR and PWR fuel assemblies.<sup>11,29</sup> Typical results for a BWR and a PWR fuel assembly are shown in Fig. 12, including ion-chamber results and spectral gamma-ray ( $^{137}\text{Cs}$ ) results. Cesium-137 is widely accepted as being the best single isotopic-burnup monitor. It has a long half-life (30.17 yr) and has approximately the same fission yield for  $^{235}\text{U}$  (6.22%) and  $^{239}\text{Pu}$  (6.69%). The gamma emission from the  $^{137}\text{Cs}$  isotope correlates linearly with burnup over the range from 0-50 GWd/tU (Sec. V.B).

Figure 12 also shows the results of axial  $\text{Be}(\gamma, n)$  measurements for the BWR fuel assembly. The  $\text{Be}(\gamma, n)$  measurements are only sensitive to gamma rays with energies  $>1660$  keV or, in these examples, the 2186-keV gamma ray of  $^{144}\text{Pr}$ . The gross gamma-ray profile compares favorably both with the  $^{137}\text{Cs}$  and the  $\text{Be}(\gamma, n)$  profiles.

All of the gamma rays emitted by a fuel assembly contribute to the dose measured by the ion chamber. The percentage of the total dose caused by each of the nine most prominent gamma rays was calculated for four BWR fuel assemblies and four PWR fuel assemblies. Declared burnup values and cooling times for each of the fuel assemblies are

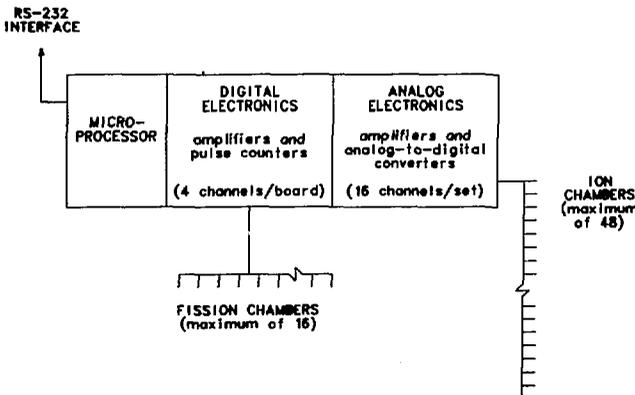


Fig. 10. Microprocessor with digital electronics for processing fission-chamber data and analog electronics for ion-chamber data.

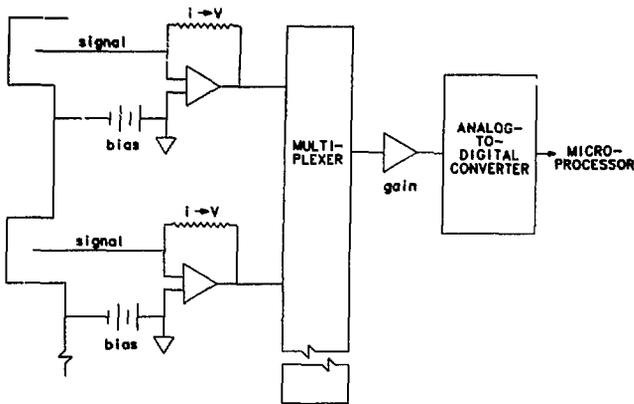


Fig. 11.  
Current-to-voltage converter  
for multiple ion chambers.

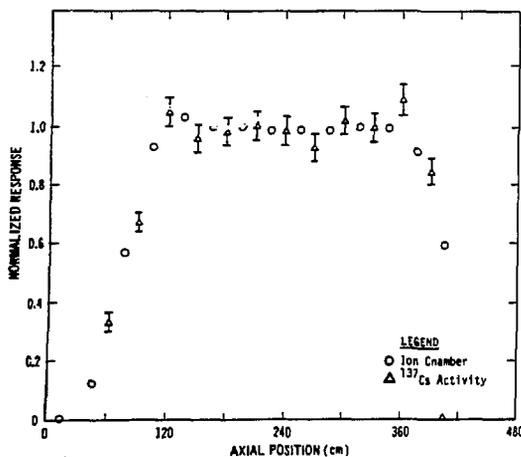
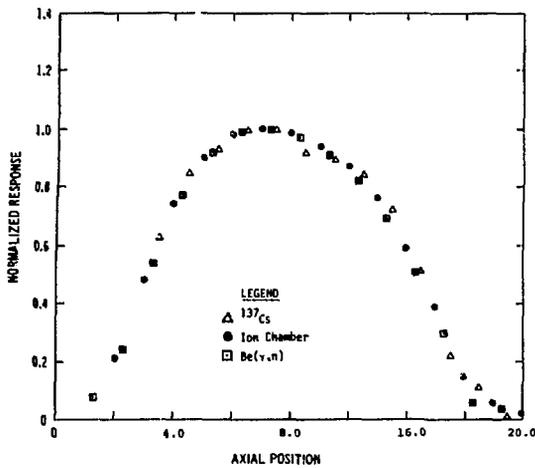


Fig. 12.  
Comparison of gross gamma-ray profiles of a  
BWR (upper plot) and a PWR (lower plot) fuel  
assembly with  $^{137}\text{Cs}$  axial profiles.

given in Table III. The gamma rays used in the calculation are given in Table IV. Figure 13 shows the contributions from each of the nine most prominent gamma rays as a function of source-to-detector distance. In every case, the relative contribution of the  $^{144}\text{Pr}$  (2186-keV) gamma ray becomes dominant as the distance increases. At a spent-fuel storage facility, the fuel assemblies may have longer cooling times than considered here. The precursor of  $^{144}\text{Pr}$  is  $^{144}\text{Ce}$  with a half-life of only 285 days, so that the contribution from the  $^{144}\text{Pr}$  may be significantly reduced from that indicated by Fig. 13.<sup>30</sup>

Figure 14 shows the percentage contributions to the total dose for the nine gamma rays as a function of cooling time.<sup>30</sup> Superimposed upon these plots is a line that divides the two sets of isotopes into those that originate directly from fission ( $\phi$ ) and those that are formed by fission followed by one or more nuclide captures ( $\phi^2$ ). The latter so-called shielded isotopes are  $^{134}\text{Cs}$  and  $^{154}\text{Eu}$ . The different production mechanisms for these two isotopes are particularly important because the neutron energy spectrum may change significantly over a fuel assembly, such as in the upper part of BWR fuel assemblies where the void fraction can change appreciably. The absorption cross sections of these particular isotopes have resonances in the epithermal energy range; therefore, any change in the neutron energy spectrum or flux can significantly change their production

TABLE III

BURNUP AND COOLING TIMES  
FOR FUEL ASSEMBLIES

Assembly	Declared Exposure (Mwd/tU)	Cooling Time (days)
BWR-1	4 356	873
BWR-3	12 459	1451
BWR-11	18 804	324
PWR-3	17 776	832
PWR-6	19 826	524
PWR-12	31 851	279
PWR-14	32 185	279

TABLE IV

ISOTOPES AND GAMMA-RAY ENERGIES  
USED IN DOSE CALCULATIONS

Isotope	Gamma-Ray Energy (keV)
$^{134}\text{Cs}$	605
$^{137}\text{Cs}$	662
$^{95}\text{Zr}$	724
$^{95}\text{Zr}$	757
$^{95}\text{Nb}$	766
$^{134}\text{Cs}$	796
$^{154}\text{Eu}$	1275
$^{144}\text{Pr}$	1489
$^{144}\text{Pr}$	2186

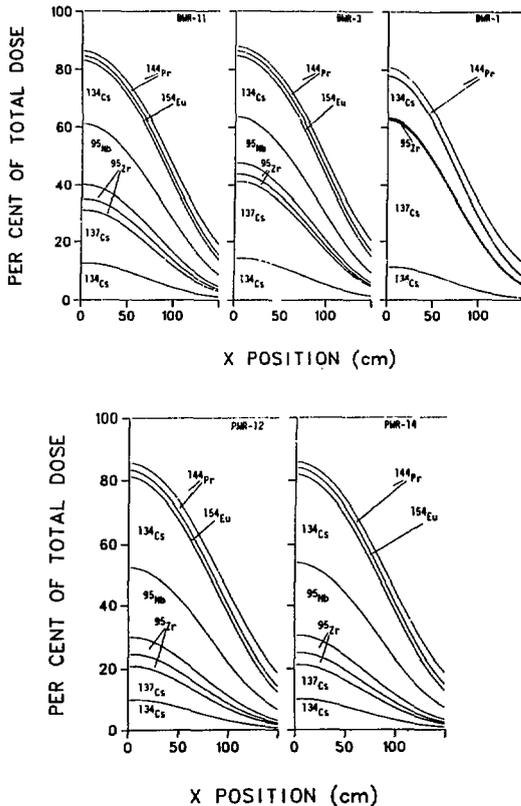


Fig. 13.

The percentage contribution to the total dose rate as a function of source-to-detector distance.

rates. This effect is minimal for the fuel assemblies examined to date, but it could be important for future measurements.

The most representative gross gamma-ray axial profile of a fuel assembly is a weighted average of the profiles measured on two or more corners or sides of the assembly. The ion chamber is an uncollimated detector; therefore, it measures (averages) the gross gamma-ray activity over axial regions of the fuel assembly. If the gross gamma-ray profile were to be measured at four angular orientations (corners or sides), it would probably be necessary to have at least one chamber about every 30 cm to obtain a representative profile. The time required to obtain an ion-chamber measurement is only about 10 s, so that the time required for these measurements is insignificant when compared to the time required for the other measurement techniques.

#### B. Spectral Gamma Ray

1. Objective. To obtain a gamma-ray spectrum (200-2200 keV) that is representative of the irradiated fuel assembly at a single axial position. To analyze the gamma-ray spectrum and identify all prominent peaks, including gamma-ray energy, net area, and their respective uncertainties. The data are transmitted to the VAS computer for more complete analysis and for correlation with data from the other three measurement subsystems.

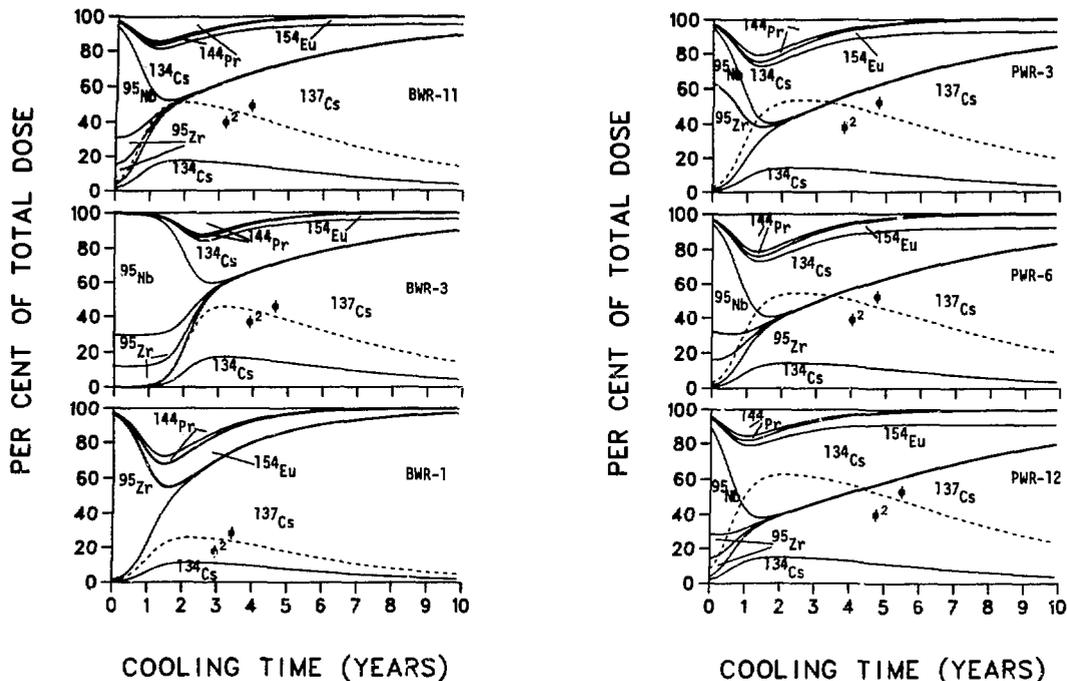


Fig. 14.  
The percentage contribution of the nine gamma rays to the total dose as a function of cooling time.

**2. Equipment.** The spectral gamma-ray subsystem consists of essentially three components: (1) collimating assembly that precisely defines the volume segment of the fuel assembly from which the gamma-ray spectra are obtained, (2) high-resolution detector, and (3) data acquisition unit for recording the spectra and calculating the net areas and uncertainties of all the prominent gamma-ray peaks. Each of these three components will be discussed briefly to identify potential problem areas that may have to be addressed to obtain a satisfactory spectral gamma-ray subsystem.

Of the three components, the collimating assembly is probably the most critical because, if it is designed incorrectly, the spectral results will be erroneous no matter how sophisticated the detector and data acquisition unit. The collimator may consist of either a hollow pipe of circular cross section or a hollow channel of rectangular cross section (that is, a rectangular slit) that extends from the detector above the surface of the

water to the axial midpoint (or any specified axial position) of the irradiated fuel assembly under-water. The circular pipe geometry has the advantage of being easier to fabricate and more rigid than the rectangular channel. However, a pipe collimator does not provide a representative sample of the fuel assembly because there is more contribution from the central region of the fuel assembly than from the edges. This is illustrated by Fig. 15 for three different sizes of pipe collimators. This effect is particularly important if BWR fuel assemblies are being examined from the side rather than from the corner.

Figure 16 shows the cross section of a BWR fuel assembly indicating the relative positions of the different regions of enrichment and burnable poisons. The production of specific fission products depends upon the initial fuel enrichment (App. D). By using a rectangular channel as the collimator, each of the enrichment regions of the BWR fuel assembly would contribute approximately

OUTER SURFACE RODS

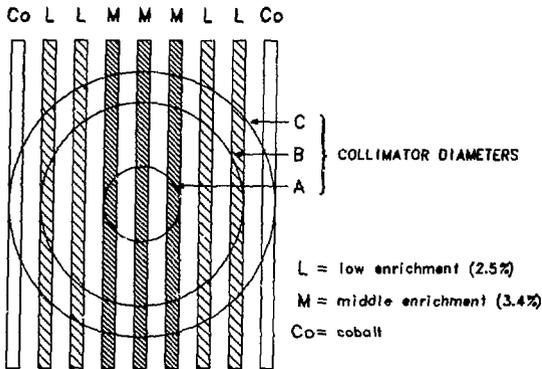


Fig. 15.

Fields of view of circular collimators with different diameters. The central fuel region contributes more than the edges in all cases.

equally to the gamma-ray spectra, resulting in a representative sample at the specified axial position.

Because of self absorption, most of the gamma rays emitted by a fuel assembly originate in the outer few rows of rods. The outer row of a BWR fuel assembly can attenuate the 661.6-keV (<sup>137</sup>Cs) gamma rays from the interior fuel rods by nearly 80% if the gamma rays pass through the middle of the rod. This self attenuation can limit the ability of any gamma-ray system to obtain a representative spectrum of the fuel assembly. Asymmetric burnup profiles across BWR fuel assemblies are common because of the use of control rod blades that can be adjacent to two of the sides of the assembly. It may be necessary to rotate BWR assemblies, taking a gamma-ray spectrum on all four sides, to obtain a representative measurement.

Similar asymmetric burnup profiles are observed in PWR fuel assemblies because of core location and the possible presence of burnable poison clusters (App. B). During the startup of PWR cores, a large number of fuel assemblies contain burnable poison clusters. For a Westinghouse PWR fuel assembly, there are 20 available positions within the 15 by 15 array. Various configurations of poison rods are used; each can produce a distinctly different burnup pattern across the assembly (Fig. 17). Significant variations in the gamma

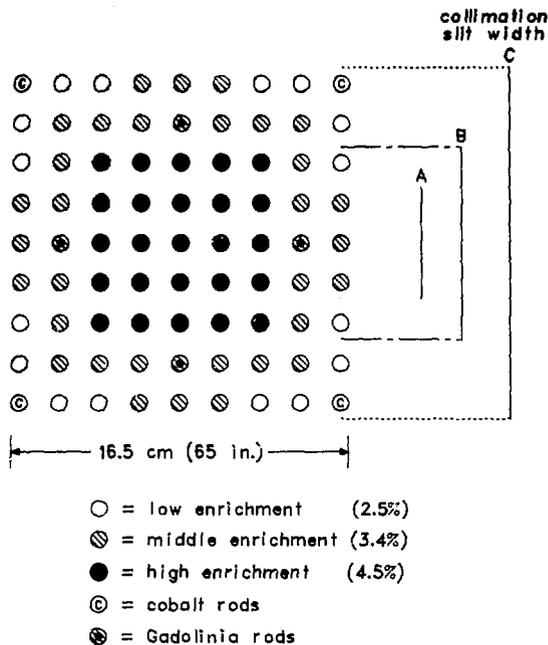


Fig. 16.

Fields of view for rectangular slit collimators of different sizes. Contributions from different regions of the fuel are equally weighted for collimator C.

activities (20-40%) have been observed at the corners of PWR fuel assemblies (App. B). Thus, to obtain representative spectra of irradiated fuel assemblies, a slit-type collimator that views the entire assembly cross section is preferred. It may be necessary to rotate the fuel assembly as well. Figure 18 shows three possible orientations for the collimator. Orientation A views the fuel assemblies from the corner; however, each of the fuel rods at the outer surface would not contribute equally to the gamma-ray spectra because of differences in path length through the water. This problem is reduced by using orientations B or C.

Additional collimation and spectral shaping is required at the end of the collimator adjacent to the detector to reduce the count rate to an acceptable level for the data acquisition unit. Low-energy gamma rays and x rays are effectively eliminated by inserting absorbers (Pb, Cd, and Cu) in the beam path. Low-energy radiation must be eliminated to permit more efficient collection of

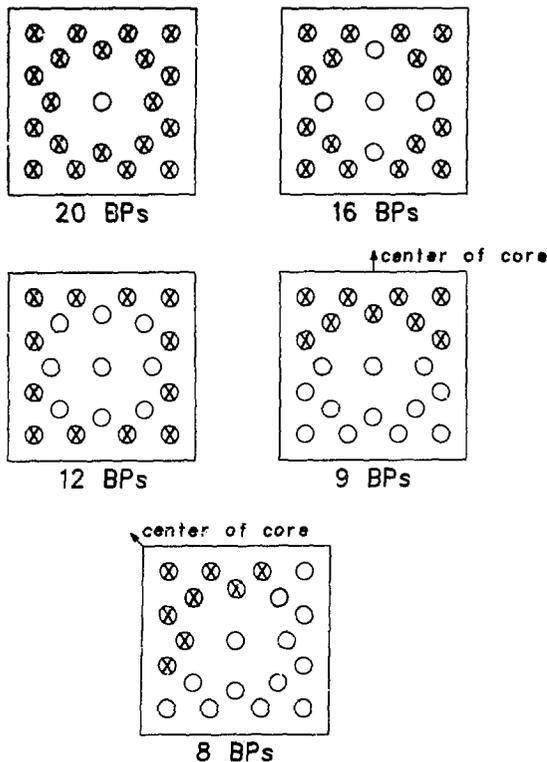


Fig. 17.  
Distributions of burnable poison rods  
in PWR assemblies.

the higher energy gamma rays from the fission and activation products in the fuel assembly.

The detector assembly is a high-resolution germanium detector with a resolution of at least 2.5 keV full width at half maximum (FWHM) at the  $^{60}\text{Co}$  gamma peak. For an irradiated fuel assembly, there are only a limited number of fission and activation products that produce detectable gamma rays. The more prominent gamma-ray emitting isotopes are listed in Table V, along with the half-lives, fission yields from  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , and the gamma-ray energies and branching ratios. The gamma rays present after a 1- to 2-yr cooling time can be easily resolved using a germanium detector with a resolution of 2.5 keV. None of the gamma rays emitted directly by the transuranics ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ) are listed in the table because their gamma rays (<450 keV) are obscured by the Compton

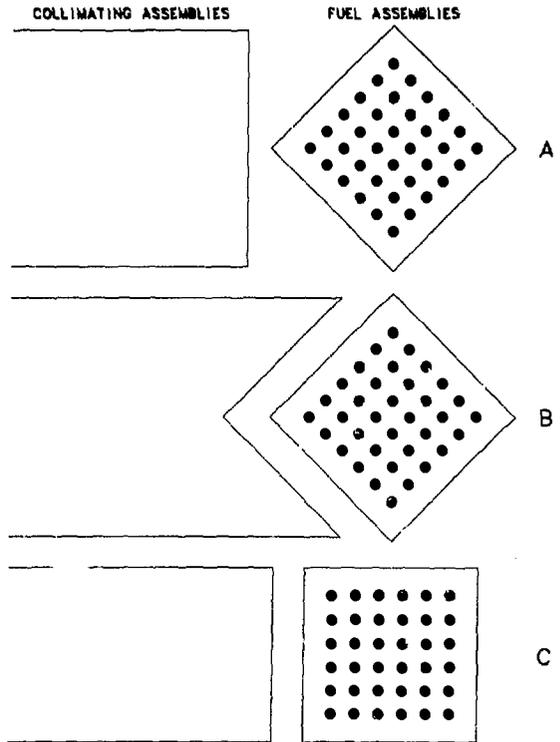


Fig. 18.  
Possible collimator orientations for  
the spectral gamma-ray subsystem.

scattering of the higher energy gamma rays from the fission products.

The data acquisition unit consists of a multi-channel analyzer with at least 4096 channels, peak stabilization, pile-up rejection, and the capability to determine accurately the energy, net area, and uncertainty of each of the prominent gamma-ray peaks in the spectra. This information is then transmitted to the VAS computer for further analysis.

**3. Data Analysis.** We have demonstrated, both experimentally and computationally, that gamma rays from the  $^{137}\text{Cs}$  isotope and the two isotopic ratios,  $^{134}\text{Cs}/^{137}\text{Cs}$  and  $^{154}\text{Eu}/^{137}\text{Cs}$ , correlate with burnup (Ref. 11 and App. D). Figure 19 shows calculational results for PWR fuel with burnups up to 50 GWd/tU with different constant power levels, and up to 31 GWd/tU with an actual irradiation history for the H. B. Robinson-2 reactor. The

TABLE V  
MEASURABLE ISOTOPES IN A TYPICAL LWR FUEL ASSEMBLY

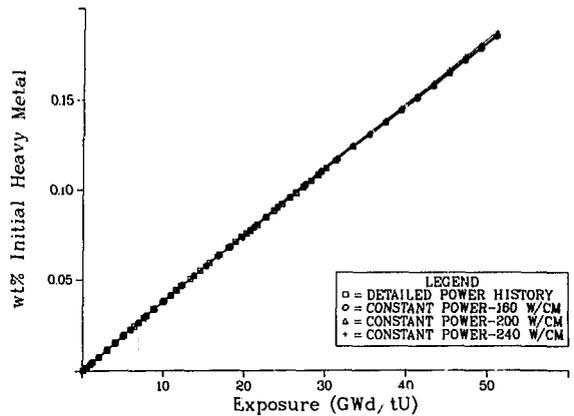
Fission Products		Fission Yield (%)		Gamma-Ray Energy in keV (Branching Ratio)
Isotope	Half-Life	<sup>235</sup> U	<sup>239</sup> Pu	
<sup>137</sup> Cs	30.17 yr	6.222	6.689	661.6 (0.851)
<sup>154</sup> Eu <sup>a</sup>	8.5 yr	2.69-6	9.22-5 <sup>b</sup>	1274.4 (0.355), 1004.8 (0.174), 996.3 (0.203)
<sup>125</sup> Sb	2.71 yr	0.0294	0.1110	427.9 (0.30), 600.8 (0.18), 686.2 (0.12), 463.5 (0.11)
<sup>134</sup> Cs <sup>a</sup>	2.062 yr	1.27-5	9.89-4 <sup>b</sup>	604.7 (0.976), 795.8 (0.854), 801.8 (0.087), 1365.1 (0.0304), 1167.9 (0.018)
<sup>106</sup> Ru-Rh	366.4 days	0.1018	4.280	622.2 (0.098), 1050.5 (0.016)
<sup>144</sup> Ce-Pr	284.5 days	5.475	3.736	696.5 (0.0134), 2185.6 (0.0066), 1489.2 (0.0026)
<sup>95</sup> Zr	63.98 days	6.496	4.892	756.7 (0.546), 724.2 (0.431)
<sup>103</sup> Ru	39.35 days	3.043	6.948	497.1 (0.864), 610.3 (0.054)
<sup>95</sup> Nb	34.97 days	6.496	4.893	765.8 (0.9982)
Activation Products				
<sup>60</sup> Co	5.27 yr			1173.2 (1.00), 1332.5 (1.00)
<sup>58</sup> Co	70.3 days			811.1 (0.99), 511.0 (B <sup>+</sup> annihilation)
<sup>54</sup> Mn	312.2 days			834.8 (1.00)

<sup>154</sup>Eu and <sup>134</sup>Cs yields are given only for direct production of the isotope from the fission process. Each isotope is produced primarily through neutron absorption. For PWR fuel material irradiated to 25 GWd/tU, the "accumulated fission yields" of <sup>154</sup>Eu and <sup>134</sup>Cs were calculated as 0.15 and 0.46% for the total fissions, respectively.

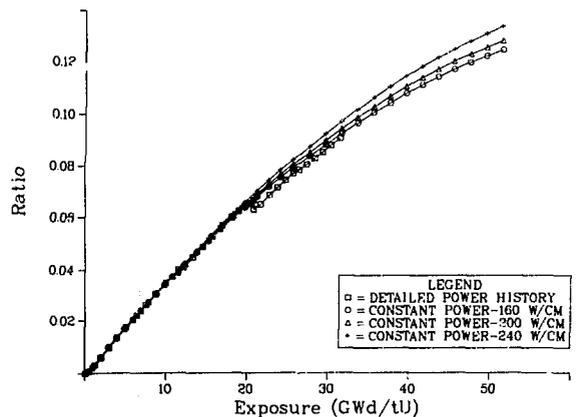
<sup>b2.69-6</sup> should be read as 2.69 × 10<sup>-6</sup>.

<sup>137</sup>Cs isotope production correlates very well with burnup because the fission yields from <sup>235</sup>U and <sup>239</sup>Pu are nearly identical. This isotope would be the preferred signature for burnup if the system could be calibrated for each type of fuel assembly to be examined. Slight changes in the scanning geometry, source self-attenuation, and attenuating material in the beam can significantly alter the measured results; therefore, isotopic ratios, in particular <sup>134</sup>Cs/<sup>137</sup>Cs and <sup>154</sup>Eu/<sup>137</sup>Cs, are often used instead of <sup>137</sup>Cs because the ratios are less sensitive to these experimental effects. The relative detection efficiency over the gamma-ray energy range can be determined by analyzing the gamma rays from fission products that have more than one gamma ray in the measured spectra (for example, <sup>134</sup>Cs, <sup>106</sup>Ru-Rh, and <sup>144</sup>Ce-Pr). This normalization process removes some of the influence of self attenuation and gamma absorption, so that measured gamma-ray ratios for one fuel assembly can be directly compared with ratios for a different assembly. Ratios of gamma rays for a single isotope can also be used as a consistency check for a set of identical fuel assemblies.

Cs-137



Cs-134/Cs-137



Eu-154/Cs-137

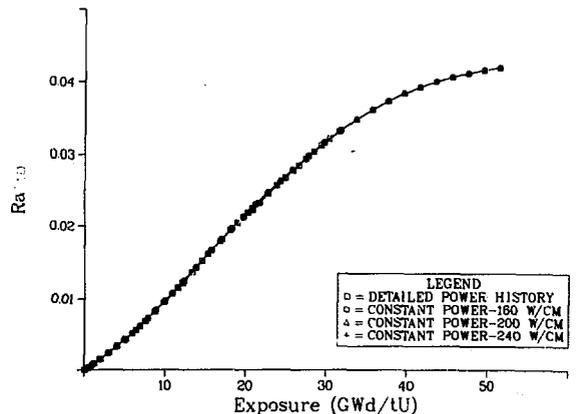


Fig. 19.

Calculational results for four irradiation histories: three constant power levels up to a burnup of 50 GWd/tU and a detailed history up to a burnup of 31 GWd/tU.

Cesium-134 has a relatively short half-life of 2.062 yr. Therefore, the  $^{134}\text{Cs}/^{137}\text{Cs}$  isotopic ratio can be significantly influenced by changes in the irradiation history. In Fig. 19 the effect on this ratio of a 61-day downtime for refueling is shown. Europium-154, with an 8.5-yr half-life, is not influenced nearly as much as is  $^{134}\text{Cs}$ . However,  $^{154}\text{Eu}$  has a complicated neutron-capture production sequence, starting with  $^{148}\text{Nd}$ ,  $^{150}\text{Nd}$ , and  $^{145}\text{Pr}$ , that may be affected by changes in the neutron energy spectrum. Results obtained using the  $^{134}\text{Cs}/^{137}\text{Cs}$  isotopic ratio may also be affected by changes in the neutron energy spectrum because the  $^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$  reaction depends strongly upon the resonance neutron cross section.

Initial fuel enrichment can also affect these two isotopic ratios, as is shown in Fig. 20. The isotopic ratios were calculated for a typical PWR fuel assembly as a function of burnup for a range of initial  $^{235}\text{U}$  enrichments of 2.2-4.5%. In BWR fuel assemblies the enrichment can also vary from 2.5-4.5%, with similar effect on the isotopic ratios.

Both the  $^{134}\text{Cs}/^{137}\text{Cs}$  and  $^{154}\text{Eu}/^{137}\text{Cs}$  isotopic ratios show a reasonably linear correlation with burnup up to 30 GWd/tU. Above this burnup range, they are no longer linear. As utilities attempt to improve uranium utilization, fuel assemblies will be exposed routinely to the 50-GWd/tU level or more. Figure 21 shows the correlations of these two isotopic ratios with the total fissile inventory remaining in the PWR fuel assemblies. The total fissile inventory has been defined as the summation of  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  isotopes. These total fissile correlations appear to be reasonably linear over the entire burnup range, that is, up to 50 GWd/tU.

### C. Passive Neutron

**1. Objective.** To obtain a passive neutron signature that is representative of the burnup of the irradiated fuel assembly at one or more axial positions.

**2. Equipment.** Uranium-235 fission chambers appear to be the best choice for measuring the neutron signatures of irradiated fuel assemblies.

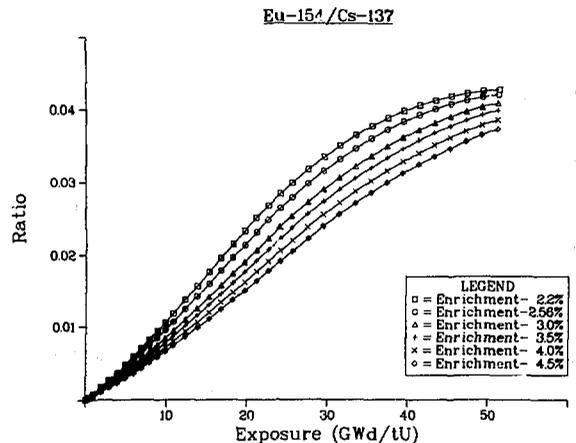
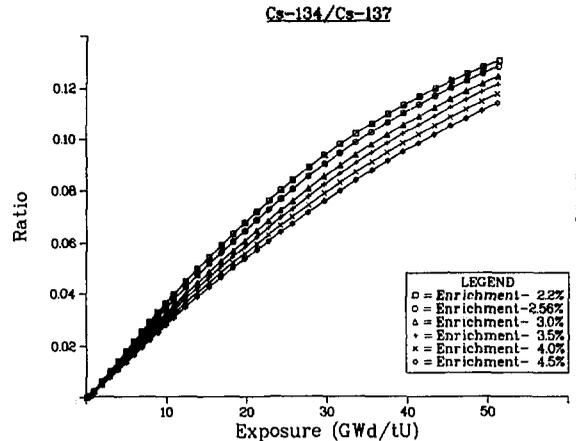


Fig. 20.  
Effect of initial fuel enrichment upon the isotopic ratios  $^{134}\text{Cs}/^{137}\text{Cs}$  and  $^{154}\text{Eu}/^{137}\text{Cs}$ .

They have been operated successfully in high gamma-ray environments (50 000 R/h).<sup>11</sup> The data collection module was described in Sec. V.A. Counting rates up to 250 000 counts/s are handled easily. Based upon the neutron measurements performed at GE-Morris (App. B), a count rate of 1000-10 000 counts/s is the expected range using fission chambers containing 130 mg of  $^{235}\text{U}$ . These rates do not cause significant problems for the electronics.

There are several possible detector arrangements that could be used to measure the passive neutron signature at one or more axial positions along the fuel assembly. Two possible arrangements are shown in Fig. 22 in which fission chambers are

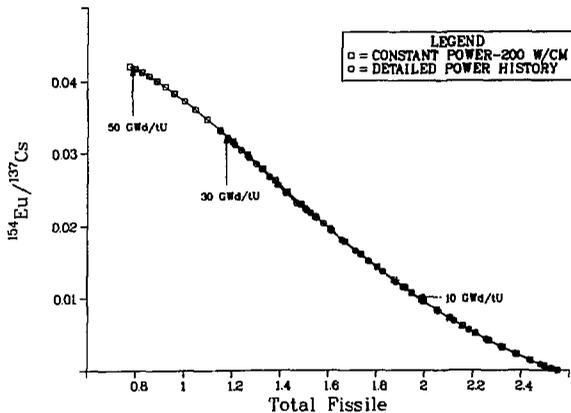
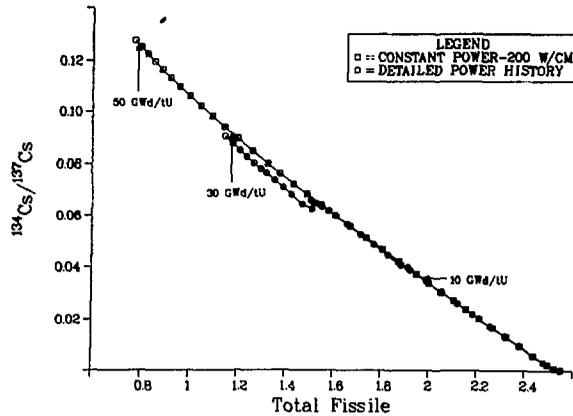


Fig. 21. Correlations of  $^{134}\text{Cs}/^{137}\text{Cs}$  and  $^{154}\text{Eu}/^{137}\text{Cs}$  with total fissile content of PWR fuel assemblies.

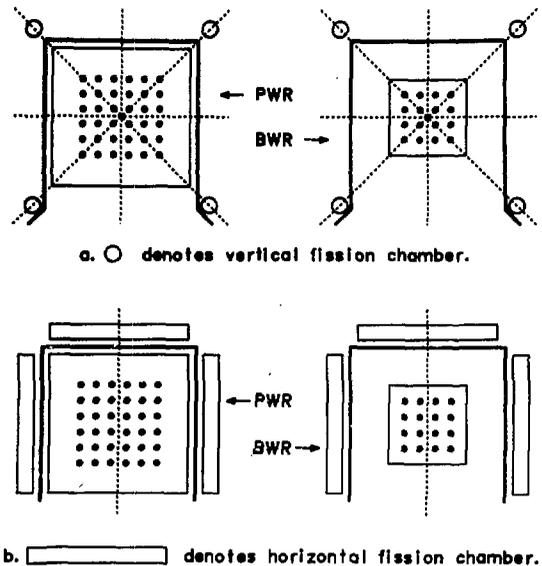


Fig. 22.

Possible detector geometries measuring the passive neutron emission of spent-fuel assemblies. Planes of symmetry are indicated by the dashed lines.

placed on the corners (Fig. 22a) or on the sides (Fig. 22b) of PWR and BWR fuel assemblies. In either case, one side of the geometry is left open to allow insertion of the fuel assembly. In Fig. 22, planes of symmetry for the neutron measurements are indicated by dashed lines. The arrangement of detectors at the four corners has more planes of symmetry than at three sides. This implies that the positioning of the fuel assembly may not be as critical for the corner arrangement as for the side arrangement. Because fuel assemblies vary slightly in size and configuration, it is necessary to make the counting geometry as independent of source position as possible. A symmetric positioning of neutron detectors makes the measurement

system relatively insensitive to the exact assembly position. A positioning requirement of  $\pm 0.5$  cm should be satisfactory, based upon laboratory and field measurements (App. B). The corner arrangement could result in too much importance being assigned to the fuel rods located in the corners, whereas if the side arrangement were used, all of the rods on a surface would contribute more equally to the measured response. Additional laboratory and calculational investigations are in progress to define which detector arrangement is preferred.

Passive neutron measurements should be performed at nearly the same axial location as the spectral gamma-ray and active neutron measurements to permit comparisons of the results. If possible, the same detectors should be used for both passive and active neutron measurements. Because of the presence of the gamma-ray collimator, the neutron detectors may have to be displaced by  $\sim 20$  cm to ensure a uniform water environment around the detector. The neutron measurements also should be performed on all four sides or corners of the assembly, if possible. If this is not possible,

then measurements on three corners or sides may be used to extrapolate to the fourth side.

During the experimental program at the GE-Morris facility (App. B), neutron measurements were obtained at different corners of the same fuel assembly and were found to vary by 30-40%. This variation in neutron counts corresponds to a difference in burnup of about 15-20%, using the power relationship shown in Fig. 23.<sup>11</sup> The sources of these burnup differences include the core position of the fuel assembly and the effect of burnable poison clusters. As was discussed in the spectral gamma-ray section, these clusters are often not symmetrical (Fig. 17).

Passive neutron measurements are performed at selected axial positions for comparison with the gross gamma-ray profile. This comparison can be used to confirm the gamma-ray profile as the axial burnup profile. Approximately five to seven axial positions on four corners or three sides should be satisfactory for this purpose.

**3. Data Analysis.** Experimentally and calculationally we have shown a power-law functional relationship between the measured neutron emission rate of PWR fuel assemblies and the declared burnup (Figs. 23 and 24). The principal sources of neutrons in irradiated LWR fuel assemblies with burn-

ups >10 GWd/tU are the two curium isotopes,  $^{242}\text{Cm}$  ( $t_{1/2} = 162.8$  days) and  $^{244}\text{Cm}$  (18.11 yr). (See App. D where a detailed discussion of the buildup of transuranics and their influence on the passive neutron emission rate is presented.) The influence of  $^{242}\text{Cm}$  is clearly indicated in Fig. 24, which shows the data from calculations for a PWR fuel assembly. At the time of discharge from the reactor (0 months), the influence of  $^{242}\text{Cm}$  on the neutron source term is a maximum; however, after 2 yr it becomes insignificant compared with  $^{244}\text{Cm}$  ( $t_{1/2} = 18.11$  yr). Fuel assemblies in spent-fuel storage facilities often have been cooled 4 yr or more. Note that the calculated results in Fig. 24 do not include the effects of neutron absorption or multiplication within the fuel assembly. These effects can be included by using Monte Carlo methods, and such investigations are in progress.

Correlations of the passive neutron rate with total fissile, total plutonium, fissile plutonium, and total plutonium minus  $^{239}\text{Pu}$  are plotted in Fig. 25. For cooling times >12 months, correlations with fissile plutonium may be useful for irradiated fuel assemblies; however, the dependence of these correlations on initial  $^{235}\text{U}$  enrichment must also be considered (App. D).

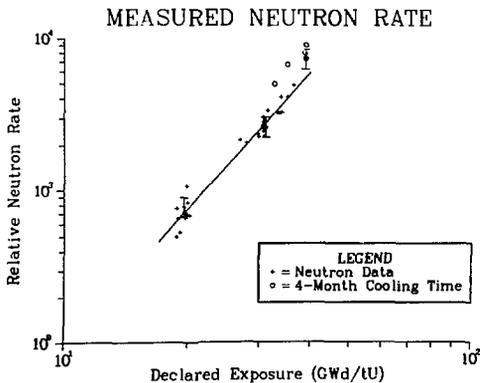


Fig. 23.

Experimentally measured data for PWR fuel assemblies with 4-40 months cooling time. The principal sources of neutrons are  $^{242}\text{Cm}$  ( $t_{1/2} = 162$  days) and  $^{244}\text{Cm}$  (18.11 yr). The effect of  $^{242}\text{Cm}$  is most significant for the shortest cooling time.

CALCULATIONAL ESTIMATES  
NEUTRON RATE VERSUS EXPOSURE

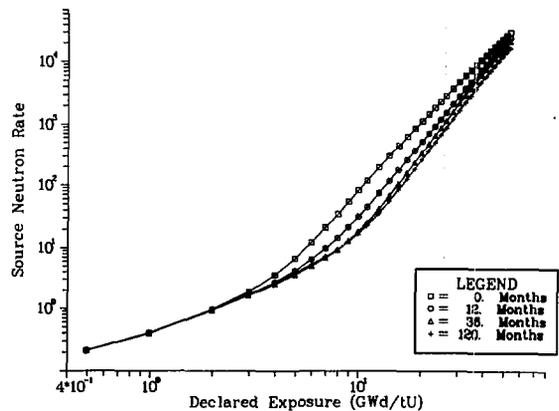
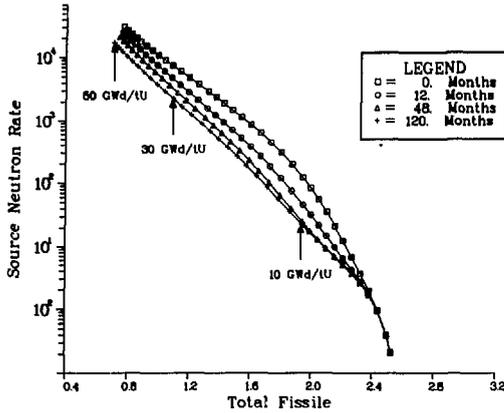


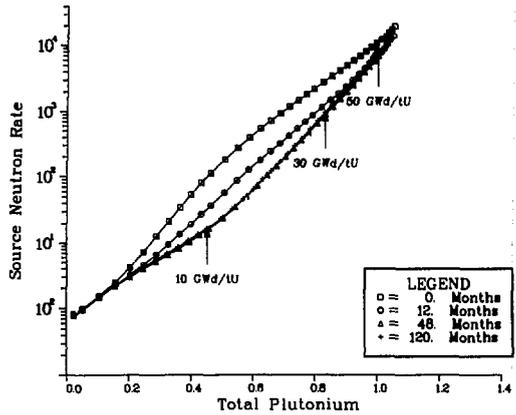
Fig. 24.

Calculated neutron source rate as a function of declared burnup for various cooling times.

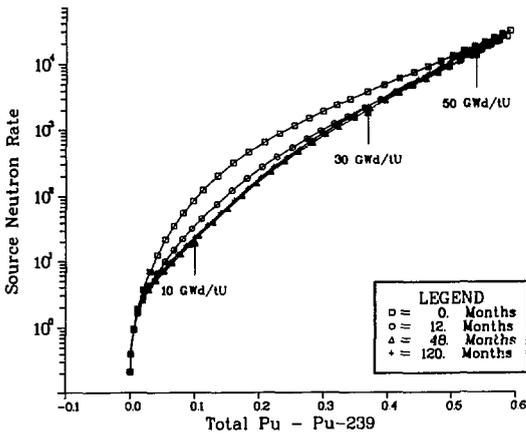
**CALCULATIONAL ESTIMATES**  
**NEUTRON RATE VERSUS ISOTOPIC CONCENTRATION**



**CALCULATIONAL ESTIMATES**  
**NEUTRON RATE VERSUS ISOTOPIC CONCENTRATION**



**CALCULATIONAL ESTIMATES**  
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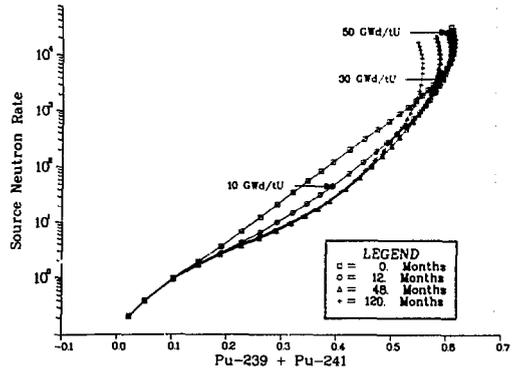


Fig. 25.  
 Correlations of the source neutron rate with total fissile, total plutonium, fissile plutonium, and total plutonium minus  $^{239}\text{Pu}$  for various cooling times.

**D. Active Neutron**

**1. Objective.** To develop a system for direct nondestructive assay of the remaining fissile content of spent LWR fuel assemblies by active neutron interrogation. The system will operate underwater in the cooling pond of spent-fuel storage facilities. Measurements will be performed on individual fuel assemblies. The data will be used to

- (1) verify that assemblies contain fissile isotopes,

- (2) quantify an assembly's effective fissile content,
- (3) verify assembly burnup, and
- (4) estimate the separate uranium and plutonium fissile contents.

**2. Technique.** Direct nondestructive measurements of the fissile contents of irradiated fuel assemblies require active neutron interrogation techniques. The intense neutron background from high-burnup assemblies, mainly caused by  $^{242}\text{Cm}$

and  $^{244}\text{Cm}$ , will require the use of reasonably strong sources for the active assay measurement to override the background. The detector response for an active measurement depends on the quantities of fissile isotopes in an assembly. The quantities of fissile isotopes, which are functions of initial enrichment and burnup, produce a measured response representative of an effective fissile loading. Appropriate calibration curves enable the effective fissile measurement to be used to determine fuel-assembly burnup. Specification of the fissile quantities of uranium and plutonium in an assembly require at least two independent measurements. Methods to correlate data collected from passive and active measurements are being developed to permit the separate uranium and plutonium fissile quantities to be calculated. Direct methods that measure the prompt and delayed fission neutron signatures and use these two measurements to separate the uranium and plutonium fissile quantities are also being investigated (App. E).

Prompt- and delayed-neutron emissions for active measurements on fuel assemblies depend on the quantities of the fissile isotopes  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$ . To calculate the prompt and delayed emission sensitivities with respect to burnup, representative spent-fuel compositions as a function of burnup are required.

Atom densities for uranium and plutonium isotopes have been obtained from CINDER calculations for the H. B. Robinson PWR fuel. The calculated atom densities compare closely with data obtained from destructive analysis of this fuel that had been irradiated up to 30 000 MWd/tU (App. D). A plot of the calculated concentrations of uranium and plutonium isotopes as a function of burnup is shown in Fig. 26. A list of the atom densities for each isotope at intervals of 5 000 MWd/tU, as calculated by the CINDER code, is given in Table VI.

Based on the above spent-fuel composition, a simple analysis of the effect of changing fissile densities on prompt- and delayed-neutron responses from thermal-neutron fission was completed. This analysis assumes that a unit volume of fuel is subjected to low-energy neutron irradiation. The neutron production resulting from fission of the

## CALCULATIONALLY ESTIMATED QUANTITIES PWR SENSITIVITY SERIES -- CASE A-2

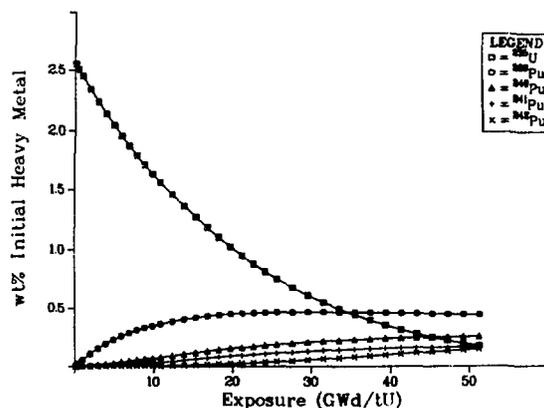


Fig. 26.  
Uranium and plutonium isotope concentrations  
for H. B. Robinson fuel.

fissile isotopes is calculated. The yield of prompt neutrons from a thermal irradiation flux of strength  $\phi$  ( $\text{n}/\text{cm}^2 - \text{s}$ ) is described by

$$Y = (\bar{\nu}_{fN})_{235} \phi + (\bar{\nu}_{fN})_{239} \phi + (\bar{\nu}_{fN})_{241} \phi \quad (1)$$

The response equation Y can be rewritten in the form

$$Y = a N_{235} + b N_{239} + c N_{241} \quad (2)$$

where Y represents an effective neutron yield from thermal fission of the fissile isotopes in a unit cell. The nuclear data corresponding to thermal energies for the fissile isotopes in Table VI are listed in Table VII.

The coefficients [a and b in Eq. (2)] for low-energy fission (0.005-0.14 eV) were computed by Gozani using a self-shielded group cross-section set.<sup>31,32</sup> These coefficients are listed in Table VIII along with the coefficients corresponding to thermal fission. The group cross-section set provides a more accurate value for the relative fission rate of the various fissile isotopes. The results indicate that the coefficients for  $^{239}\text{Pu}$

TABLE VI

CINDER CALCULATED FISSILE MATERIAL DENSITIES  
AS A FUNCTION OF BURNUP

Burnup (GWd/tU)	Atom Densities Atoms/b-cm			
	<sup>238</sup> U	<sup>235</sup> U	<sup>239</sup> Pu	<sup>241</sup> Pu
0	2.13-2 <sup>a</sup>	5.67-4	0	0
5	2.12-2	4.53-4	4.78-5	1.19-6
10	2.11-2	3.63-4	7.44-5	5.15-6
15	2.10-2	2.83-4	9.02-5	1.19-5
20	2.09-2	2.21-4	9.76-5	1.78-5
25	2.08-2	1.71-4	1.01-4	2.29-5
30	2.07-2	1.32-4	1.02-4	2.68-5
35	2.07-2	1.01-4	1.01-4	2.97-5
40	2.06-2	7.20-5	9.95-5	3.21-5
45	2.05-2	5.37-5	9.78-5	3.34-5
50	2.03-2	3.46-5	9.30-5	3.37-5

<sup>a</sup>Read 2.13 × 10<sup>-2</sup>.

TABLE VII

THERMAL NEUTRON PARAMETERS FOR THE FISSILE ISOTOPES  
<sup>235</sup>U, <sup>239</sup>Pu, AND <sup>241</sup>Pu

Isotope	$\sigma_f$ (b)	$\bar{\nu}$ (n/fission)	$\beta$ (delayed n/fission)
<sup>235</sup> U	577.1	2.43	0.0158
<sup>239</sup> Pu	740.6	2.87	0.0061
<sup>241</sup> Pu	950.0	3.14	0.0156

TABLE VIII

PROMPT NEUTRON YIELD COEFFICIENTS  
FOR LOW-ENERGY FISSION

Energy	Coefficients	
	a	b
Thermal (0.025 eV)	1.52	2.13
Group Set (0.005-0.14 eV)	1.7	2.3

and <sup>241</sup>Pu are underestimated by 12 and 8%, respectively, if only the thermal cross-section values are used.

The prompt-neutron yield, corresponding to the data of Table VI and the group set coefficients (a = 1.7, b = 2.3) of Table VIII, is plotted as a function of burnup in Fig. 27. The yield (Y) decreases from a maximum of 5.67 × 10<sup>-4</sup> atoms/b-cm,

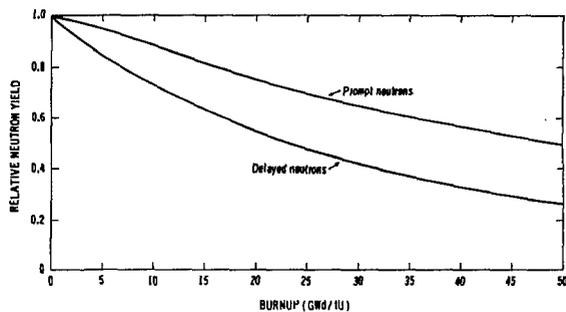


Fig. 27.  
Relative yield of prompt and delayed  
neutrons from a PWR fuel assembly as  
a function of burnup.

corresponding to a PWR assembly with no burnup, to 3.67 × 10<sup>-4</sup> atoms/b-cm at a burnup of 30 000 MWd/tU. This represents a 35% decrease in the prompt-neutron yield.

The fraction of the prompt yield produced by each of the fissile isotopes as a function of burnup is given in Table IX. A fresh assembly with no burnup produces all of its fissile yield from <sup>235</sup>U. Increasing burnup produces higher relative quantities of the <sup>239</sup>Pu and <sup>241</sup>Pu isotopes. At 30 000 MWd/tU, 47.2 and 16.8% of the prompt-neutron yield is produced from <sup>239</sup>Pu and <sup>241</sup>Pu, respectively, whereas the <sup>235</sup>U produces only 36% of the yield. It is important to remember that these neutron yields do not include effects caused by multiplication and absorption in a fuel assembly. They represent the yield caused only by thermal fission of a unit cell of fuel. The multiplication and absorption effects are being investigated, both in the laboratory and calculationally using Monte Carlo techniques.

The delayed-neutron yield from a unit cell as a function of burnup for the low-energy group structure (0.005-0.14 eV), was determined by Gozani<sup>31,32</sup> to be

$$Y_d \propto N_{235} + 0.6 N_{239} + 1.65 N_{241} \quad (3)$$

The delayed-neutron yield for the atom densities of Table VI is listed in Table X, along with the fraction of the signal produced by each of the

TABLE IX

FRACTION OF THE PROMPT-NEUTRON RESPONSE  
PRODUCED FROM EACH FISSILE ISOTOPE

Burnup (Gwd/tU)	Effective Fissile Density (atoms/b-cm)	Prompt Response Fraction		
		<sup>235</sup> U	<sup>239</sup> Pu	<sup>241</sup> Pu
0	5.67-4 <sup>a</sup>	1.0	0	0
5	5.37-4	0.844	0.151	0.005
10	5.02-4	0.723	0.252	0.025
15	4.64-4	0.610	0.339	0.059
20	4.28-4	0.517	0.388	0.096
25	3.95-4	0.433	0.434	0.133
30	3.67-4	0.360	0.472	0.168
35	3.41-4	0.296	0.504	0.200
40	3.15-4	0.229	0.537	0.234
45	2.97-4	0.181	0.560	0.259
50	2.70-4	0.128	0.585	0.287

<sup>a</sup>Read 5.67 x 10<sup>-4</sup>.

fissile isotopes. The relative delayed yield is plotted as a function of burnup in Fig. 27 for comparison with the relative prompt yield. The delayed-neutron yield is a stronger relative function of burnup than is the prompt yield. However, the much lower absolute yield of delayed neutrons compared to prompt neutrons and the high neutron background make a delayed-neutron measurement more difficult (see App. E).

The effective delayed-neutron fraction ( $\beta_e$ ) decreases as a function of burnup because the plutonium isotopes increase and the <sup>235</sup>U decreases. The value of  $\beta_e$  is calculated for thermal fission parameters using the relation

$$\beta_e = \frac{(\beta \bar{\nu}_{f,N})_{235} + (\beta \bar{\nu}_{f,N})_{239} + (\beta \bar{\nu}_{f,N})_{241}}{(\bar{\nu}_{f,N})_{235} + (\bar{\nu}_{f,N})_{239} + (\bar{\nu}_{f,N})_{241}} \quad (4)$$

Two curves are plotted in Fig. 28. The lower curve corresponds to the value of  $\beta_e$  when the contribution from the <sup>241</sup>Pu isotope is ignored, whereas the upper curve includes the contribution of <sup>241</sup>Pu. At higher burnup values, the <sup>241</sup>Pu isotope makes a significant contribution to the delayed-neutron fraction.

An active neutron measurement of an irradiated fuel assembly underwater will produce a response

TABLE X

FRACTION OF THE DELAYED-NEUTRON RESPONSE  
PRODUCED FROM EACH OF THE FISSILE ISOTOPES

Burnup (Gwd/tU)	Effective Fissile Density (atoms/b-cm)	Delayed Response Fraction		
		<sup>235</sup> U	<sup>239</sup> Pu	<sup>241</sup> Pu
0	5.67-4 <sup>a</sup>	1.0	0	0
5	4.84-4	0.937	0.059	0.004
10	4.17-4	0.871	0.107	0.022
15	3.57-4	0.793	0.152	0.055
20	3.09-4	0.715	0.190	0.095
25	2.69-4	0.635	0.225	0.140
30	2.37-4	0.556	0.258	0.186
35	2.11-4	0.480	0.288	0.233
40	1.85-4	0.390	0.323	0.287
45	1.67-4	0.321	0.350	0.329
50	1.46-4	0.237	0.382	0.381

<sup>a</sup>Read 5.67 x 10<sup>-4</sup>.

caused primarily by thermal-neutron-induced fissions in the fuel. The relative yield of neutrons from each of the fissile isotopes changes significantly with burnup, as shown in Tables IX and X. The combined yield appears as an effective fissile component that is related to assembly burnup, as shown by Figs. 27 and 28.

An active neutron measurement is, in fact, much more complicated than indicated by these unit-cell calculations. The actual measurements depend strongly on the distribution of fissile material in the fuel assembly, combined with neutron multiplication and neutron transport effects.<sup>33</sup> In addition, the detector response also complicates the interpretation of these measurements.

3. Equipment. The reference case for nondestructive measurements requires that the fuel assembly be positioned underwater in a test stand and not moved during the duration of the assay. The test stand must provide accurate positioning between the assembly and the measuring apparatus. The length of fuel assemblies and the time allotted for data acquisition prevent making an active assay measurement along the entire length of the fuel assembly. Therefore, data using the neutron-interrogation source will be obtained at a few axial positions along the fuel assembly and then normalized to the full length using burnup profiles obtained with ion-chamber or passive neutron measurements.

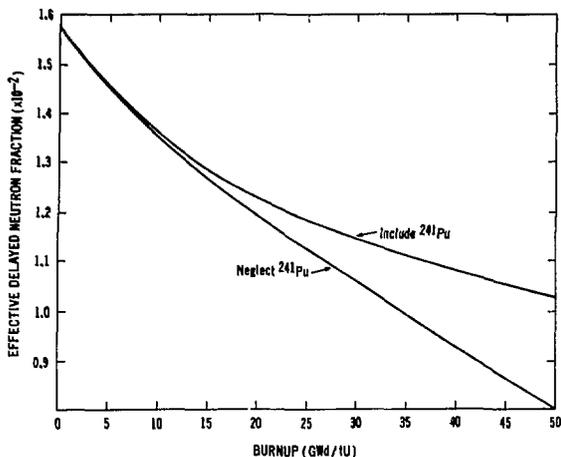


Fig. 28.

Effective delayed-neutron fraction for thermal fission of a PWR fuel assembly as a function of burnup.

Items that must be considered in the detailed design of an active neutron system include

- (1) detectors (number, type, and location);
- (2) sources (neutron emission rate, energy spectrum, number, and location); and
- (3) materials (type, shape, placement around the source, and detectors).

Spent-fuel assemblies produce large gamma-ray (>50 000 R/h) and neutron backgrounds ( $\sim 10^8$  n/s). Neutron detectors must be able to operate without producing bias effects, such as pulse pileup, that could be caused by the intense gamma field. In addition, the large neutron fields require low-efficiency detectors to reduce count rates and limit problems associated with excessive downtime losses. Low-efficiency counters also enable close geometric coupling between the detectors and the fuel assembly. Neutron current chambers do not exhibit the problems of downtime and pileup that are characteristic of pulse counters; however, the chambers are not capable of detecting neutron-only events in the presence of a large gamma-ray field. They also do not provide sufficient signal for low-burnup, long-cooling-time assemblies. For these reasons, pulse-type detectors must be used for the neutron measurements.

A few types of neutron detectors are capable of meeting these requirements, and among them fission chambers offer some distinct advantages. They

require essentially no gamma-ray shielding and are capable of detecting individual neutrons in the presence of an incident gamma-ray flux of  $10^6$  R/h. Their low detection efficiency permits neutron counting with minimal downtime and pileup effects when exposed to large neutron and gamma-ray fields. Boron-10-lined chambers have a higher detection efficiency than fission chambers and are able to operate in large gamma fields if adequately shielded. Spent-fuel measurements would require that these counters be shielded with lead and be placed further from the assembly to reduce the neutron count rate. Development of the active and passive neutron assay portion of a spent-fuel measurement system currently is based on using fission chambers.

The types of materials, number of detectors, and their locations relative to the neutron-interrogation source and fuel assembly are currently being studied. Several detectors may be required to ensure that all pins in an assembly produce a measured response characteristic of their loading and independent of their location. The detectors may be placed in polyethylene and wrapped in cadmium to measure the response to epithermal neutrons. Decisions concerning detector location, placement, and shielding and source-tailoring materials are based on producing a measured response that is independent of fuel-pin position and that enhances the signal-to-noise ratio of the measurements. Design calculations and laboratory experiments are underway using unirradiated fuel materials (App. F).

A number of neutron-interrogation sources are possible for measurements underwater. The water serves to reduce neutron energies below the fission threshold of  $^{238}\text{U}$ , causing most of the induced-fission signal to be from the desired fissile isotopes (>90%). To date, we have primarily investigated designs based on using a  $^{252}\text{Cf}$  spontaneous fission source. The half-life (2.65 yr), the wide range of possible neutron yields ( $<10^{10}$  n/s), the high reliability, and the small size make this a good choice. To perform these measurements a  $^{252}\text{Cf}$  source of  $10^8$ - $10^9$  n/s ( $\sim 0.1$ - $1$  mg) would be required. The  $^{252}\text{Cf}$  neutron source is placed on one side of the fuel assembly with the detectors on the other side. The source may be moved to

several positions for active measurements along the length of the assembly. Before each measurement using the neutron source, measurements of the neutron background must be collected. If the passive neutron detectors are also used for the active measurement, their data will provide the necessary background measurements.

The neutron source is placed in a block of material at each measurement position to distribute the neutrons across the edge of the assembly. The desired effect is to obtain uniform irradiation of the fuel-assembly cross section using a single "point" neutron source. The proper source distribution and tailoring material arrangement produces a response that is reasonably independent of pin position and that limits fissions in  $^{238}\text{U}$  caused by source neutrons. Multiplication in an assembly will produce a  $^{238}\text{U}$  fast-fission response (~7%) that is independent of the interrogation source. Initial calculations and experiments to design an optimum source-tailoring configuration are described in App. F.

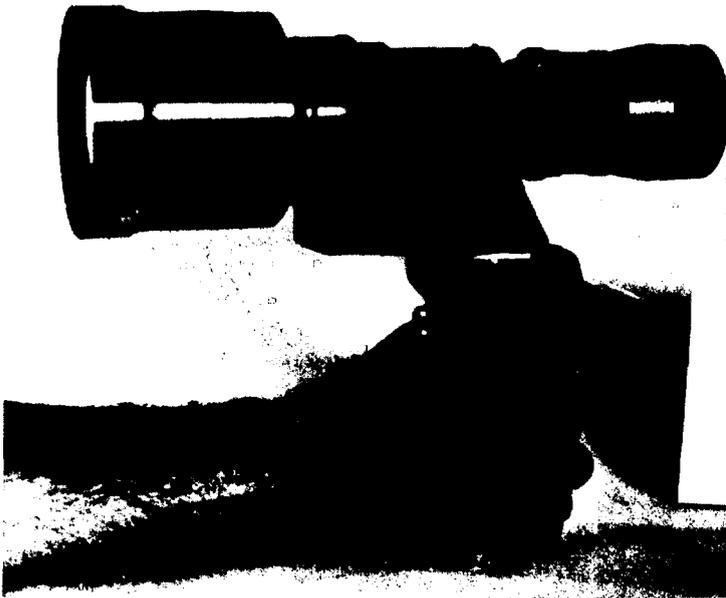
## VI. CONCLUSIONS AND RECOMMENDATIONS

The following conclusions and recommendations are based on the current interim study of nondestructive measurement techniques for irradiated LWR fuel assemblies stored underwater. The spent-fuel measurement technology developed under this program should be transferred to the nuclear industry. The basic technology is applicable to the measurement of MTR or LMFBR fuel materials and should not be limited only to the measurement of LWR fuel assemblies. The study is incomplete in that some of the proposed measurement systems must still be tested in the laboratory and in the actual environment of an operating spent-fuel storage pond. Also the scope of the current study does not include new spent-fuel storage technologies, such as bundle disassembly, rod compaction, and underwater storage, or dry storage in air of irradiated rods or assemblies.

### A. Conclusions

- Nondestructive measurements of the following spent-fuel parameters would help operators of spent-fuel storage facilities satisfy various regulatory and in-plant materials management requirements:

- (1) fuel imaging by measurement of Cerenkov radiation,
  - (2) gross gamma and passive neutron dose rates,
  - (3) gamma-ray and neutron emissions from specific fission products and actinides,
  - (4) average burnup and the distribution of burnup over the fuel material,
  - (5) total fissile content and the separate uranium and plutonium fissile contents, and
  - (6) total plutonium content.
- The most useful nondestructive measurement systems for spent-fuel storage facilities are those capable of measuring a combination of spent-fuel parameters covering a wide range of irradiation histories for different types of fuel.
  - In general, gross gamma-ray and passive neutron emissions from irradiated LWR fuels correlate well with angular and axial burnup profiles. The spectral gamma-ray and passive neutron emissions also appear to correlate with the total plutonium and fissile plutonium contents, but these correlations must still be verified experimentally. The passive neutron correlations are most useful for irradiated fuels with burnups >10 GWD/tU and cooling times >2 yr.
  - Hardware and software subsystems for Cerenkov, gross gamma, and passive neutron measurements of irradiated LWR fuel have been brought to an advanced stage of development. Portable devices, such as the Cerenkov viewing device (Fig. 29) and the neutron ring (Fig. 30), have been tested in the field and are available for routine use in spent-fuel storage facilities. These devices provide confirmatory data regarding the integrity, average burnup, and burnup distribution of spent-fuel assemblies.
  - The HRGS technique is widely recognized as a useful burnup monitor. This technique is expected to provide reasonably accurate nondestructive measurements of burnup and cooling time. However, the HRGS technique



JAVELIN MODEL 222 NIGHT VISION DEVICE

THIS DEVICE CAN BE USED BY THE IAEA INSPECTOR TO OBTAIN QUALITATIVE INFORMATION FROM THE CERENKOV GLOW FROM SPENT FUEL ASSEMBLIES. SUCH INFORMATION AS THE UNIFORMITY OF THE GLOW, THE SPATIAL DISTRIBUTION, THE OVERALL INTENSITY AND THE GENERAL APPEARANCE OF THE ASSEMBLY CAN BE USEFUL TO HIM IN ESTABLISHING THE AUTHENTICITY OF THE ASSEMBLIES INSPECTED. THIS METHOD IS RAPID AND THE INSTRUMENT IS LIGHTWEIGHT AND EASY TO USE.

Fig. 29. The Cerenkov spent-fuel viewing device.

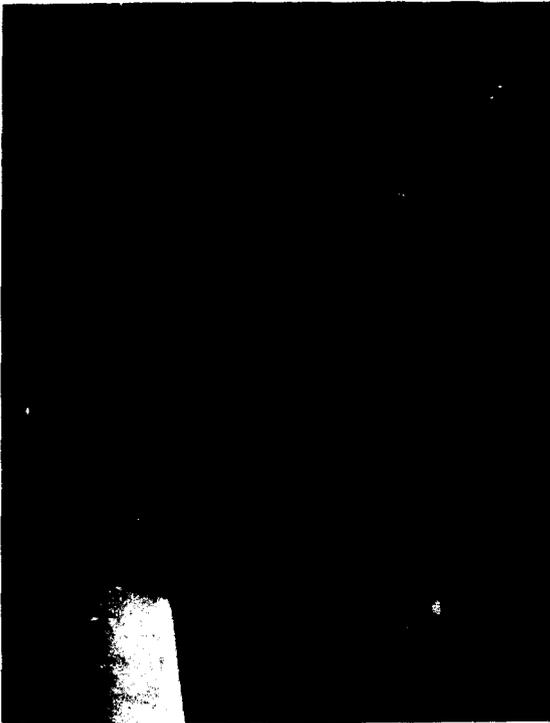


Fig. 30.  
The neutron ring device. This device measures both gross gamma and passive neutron emissions.

requires relatively long measurement times and should be used in conjunction with gross gamma and passive neutron techniques to obtain the average burnup and burnup profile in a reasonable measurement time, that is, ~30 min or less per fuel assembly.

- Hardware and software for the HRGS subsystem are in an advanced stage of development and are essentially available for in-plant use (Fig. 31).
- Active neutron measurements of the fission-neutron yield induced by an external isotopic neutron source ( $^{252}\text{Cf}$ ) are sensitive to small changes (a few per cent) in the fissile content or the fissile distribution of fuel assemblies. This technique still must be demonstrated for irradiated fuel assemblies. As with HRGS, it should be used in conjunction with gross gamma and passive neutron systems because the measurement time is ~15-30 min.
- The active neutron technique using steady-state, isotopic neutron sources is essentially ready for field testing. The pulsed-neutron technique, which potentially could be used to separate the uranium and plutonium fissile components by separating the induced prompt and delayed fission



Fig. 31.

Experimental setup for high-resolution gamma-ray measurements at Big Rock Point Nuclear Plant.

neutrons, must await further development in the laboratory before it can be considered ready for field testing.

- The conceptual design of a prototype VAS is ~80% complete (Fig. 8 in Sec. IV.D). The prototype system includes gross gamma, passive neutron, high-resolution gamma, and active neutron measurement subsystems. If completed, this system would be valuable for gathering field-test data and experience at operating spent-fuel storage facilities.

#### B. Recommendations

- The Cerenkov viewing device and the neutron ring detector, both already developed for the IAEA, should be transferred to the domestic nuclear industry.
- The prototype VAS should be completed and a program of field testing and evaluation should be carried out in an actual spent-fuel storage facility.
- Correlations of gamma-ray and passive neutron emissions with plutonium content, both total and fissile contents, should be verified experimentally for irradiated LWR fuels. Work is in progress and should continue to obtain and analyze suitable nondestructive and destructive data for well-characterized burnup samples covering the burnup range up to 50 Gwd/tU.
- Work at the R&D level should continue on the potential applications of active neutron techniques to spent-fuel measurements. In particular, investigations of pulsed-neutron techniques for this application should continue, including calculations, laboratory experiments, and some field evaluations. The results of these investigations should be reviewed before a commitment is made to build a full-scale prototype system.
- The study of the potential application of nondestructive measurements for spent fuel should be extended to the new techniques for fuel management and storage currently under study by the DOE, EPRI, and the nuclear industry. The potential role of nondestructive measurements in supporting these new technologies should be investigated.

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