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NONDESTRUCTIVE MEASUREMENTS ON SPENT FUEL FOR THE NUCLEAR FUEL CYCLE

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ABSTRACT

Nondestructive measurements on spent fuel are being developed to meet safeguards and materials management requirements at nuclear facilities. Spent-fuel measurement technology and its applications are reviewed.

I. INTRODUCTION

Nondestructive measurements for spent nuclear fuels are being developed to meet more stringent requirements for safeguards and nuclear materials management at nuclear facilities. These facilities include at-reactor spent-fuel storage, away-from-reactor (AFR) storage, and spent-fuel disposal or reprocessing. The development of spent-fuel measurement technology has implications for improved international safeguards and the international management of plutonium, as well as for improved national safeguards and facility management of nuclear materials.

This paper summarizes the rapid development of spent-fuel measurement technology over the past few years, primarily in the U.S. and Europe. Applications of this technology for improved safeguards and materials management are discussed.

II. SPENT-FUEL MEASUREMENT TECHNOLOGY

Nondestructive techniques are based on the measurement of the gamma-ray and neutron signatures of the fission and activation products and the actinide inventory in the spent-fuel assembly. Nondestructive techniques that have been applied to spent-fuel assemblies are listed in Table I. All of the measurement techniques involve the direct measurement of radiations emitted by the irradiated fuel material, except for the Cerenkov-light technique that measures secondary radiation.

A. Gamma-Ray Techniques

Gamma rays originate from the fission and activation products and the actinides, with fission and activation products being the principal gamma-ray sources. The gamma-ray dose rate

TABLE I

SPENT-FUEL NONDESTRUCTIVE MEASUREMENTS

Measurement Type	Capabilities	Limitations	Reference
Cerenkov light	Radioactive material; rapid; simple instrument; above water; no fuel handling	Nonspecific; self-shielding; semi-quantitative; bare assemblies in pool	4
Gross gamma	Gamma dose rate; axial profiles; simple instruments; minimal fuel handling	Nonspecific; self-shielding; semi-quantitative	1,2,3
Gamma spectrometry	Gamma spectra; fission product specific; exposure and cooling-time correlations; well established	Self-shielding; relatively complex; relatively slow; fuel handling; geometry	5,6
Passive neutron	Neutron dose rate; penetrability; exposure correlation; relatively simple; minimal fuel handling	Nonspecific; cooling time dependence	6,7,8
Active neutron	Fissile content; penetrability; independent assay	Complex instrument; requires neutron source; fuel handling; geometry	9,10,11,12

ranges from 10 to 30 000 R/h after approximately one-year cooling time. The gross gamma-ray signature can be measured using ion chambers,[1] scintillators,[2] and thermo-luminescent detectors (TLDs),[3] or it can be measured indirectly from the Cerenkov light emission.[4]

Cerenkov light, a continuum extending into the blue region of the visible spectrum, results from the interaction of the direct radiation with the surrounding material. Measurements of Cerenkov light do not require the placement of any device in the water, because the light intensity is measured above the surface of the storage pool. This permits the rapid verification of the presence of radioactive material in fuel assemblies, and an approximate check of the declared exposure and cooling time. Measurements of Cerenkov light require that the ambient light level be reduced around the storage pool and that the fuel assembly not be stored in a canister. The other gross-gamma detectors are not limited by these restrictions. However, measurements of the gross gamma-ray signatures using ion chambers, scintillators, or TLDs require the placement of underwater fixtures in the fuel storage pool.

The use of detectors that are sensitive to gamma-energy thresholds, for example Be(γ,n) detectors, provides the capability of rapidly measuring the presence of specific fission products.[5] Gamma rays having energies greater than 1660 keV interact with beryllium to produce neutrons that can be counted using a ^{235}U fission chamber. The principal gamma ray contributing to the production of neutrons via the (γ,n) reaction is the 2186-keV gamma ray from the ^{144}Pr fission product ($t_{1/2}$ = 17 minutes), which is in secular equilibrium with its parent ^{144}Ce ($t_{1/2}$ = 285.4 days). This technique provides information about the presence of a fission product; therefore, it provides a higher level of verification for spent-fuel assemblies than that provided by gross-gamma measurements.

High-resolution gamma-ray spectrometry (HRGS) is the most widely accepted safeguards technique for examination of spent-fuel assemblies.[13,14] By using isotope activities and ratios, for example ^{137}Cs or ^{134}Cs/^{137}Cs, the exposure values of assemblies can be predicted with precisions of 5-10%, provided that the irradiation histories are known.[6] This technique can also be used to establish the consistency of cooling times for a specific set of fuel assemblies. The use of HRGS is limited by self-shielding of the fuel assembly, in that HRGS only "sees" the outer few rows of fuel rods in an assembly. Edge rods make a substantially higher contribution to the total observed gamma activity than interior rods.[15,16] Gamma rays used for HRGS usually are in the energy range 600-800 keV and are significantly attenuated. Another limitation is the complexity of the equipment required to perform these measurements. Collimating fixtures must be placed in the storage pool and the fuel scanning geometry must be controlled to obtain high-quality results.

B. Passive Neutron Techniques

Measurements of the relative neutron rates of irradiated fuel assemblies have been correlated with declared exposure values. A power-law functional relationship was used: neutron rate = $\alpha(\text{Exposure})^{\beta}$, where α and β are empirically determined constants.[6,7,8] Typical values of β are in the range 3.0 to 4.3, depending on the cooling time and type of reactor. Typical results are shown in Fig. 1 for 36 PWR fuel assemblies. The relative neutron rate is plotted as a function of the declared exposure over the range 18 813 to 38 860 MWd/MTU. The principal sources of neutrons for these exposures are the curium isotopes, ^{242}Cm and ^{244}Cm. The half-life of ^{242}Cm is 163 days versus 18.11 years for ^{244}Cm; therefore, neutrons from ^{242}Cm make a significant contribution for relatively short cooling time, but after about 2-3 years the neutron rate is dominated by ^{244}Cm.

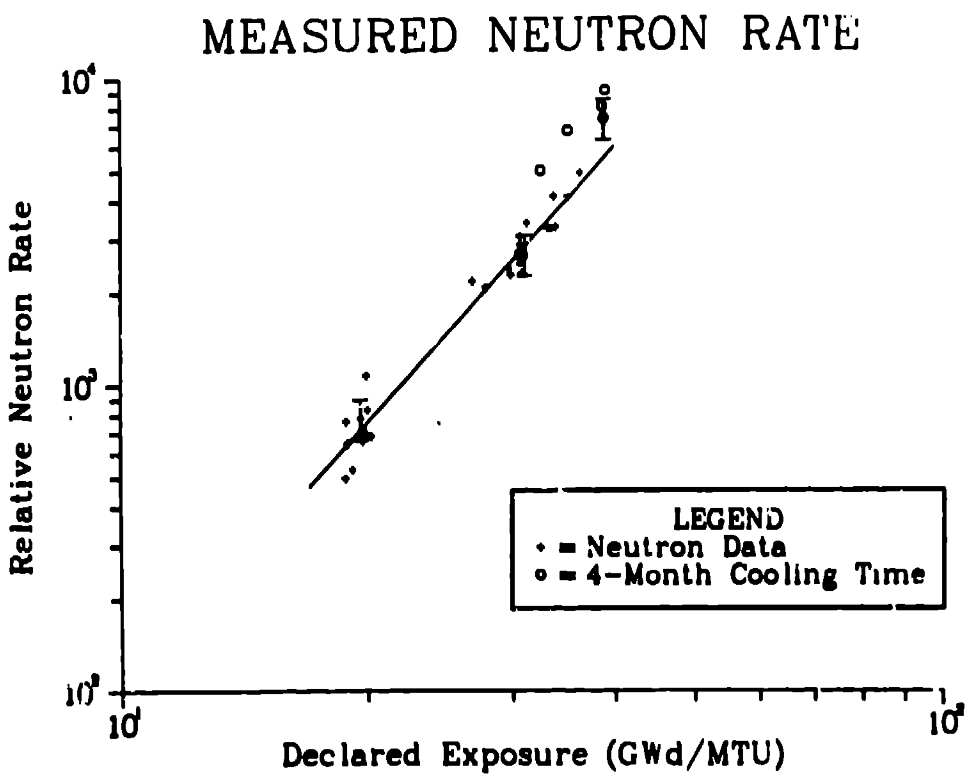

Fig. 1.
Measured neutron rates for 36 PWR fuel assemblies showing the effect of ^{242}Cm on the fuel assemblies with short cooling times.

A single-point depletion code (CINDER) has been used to calculate actinide inventories for various levels of exposure.[17] These calculations have confirmed the power-law relationship. The influence of factors, such as initial ^{235}U enrichment, power level, and other parameters, are presently under investigation.

The passive neutron technique for verification of irradiated fuels is similar to the Be(γ,n) technique, in that information is obtained about the presence of specific isotopes. It is also similar to the HRGS technique, in that the passive neutron signatures can be correlated with exposure.

An important advantage of neutron measurements is the penetrability of the neutrons. Fuel rods in the center of a PWR assembly have been shown calculationally to contribute about as much to the neutron signature as the exterior rods.[8] This is because the effects of neutron attenuation and multiplication are approximately compensating. Passive neutron signatures may also be correlated with the production of plutonium in irradiated fuel assemblies. One promising correlation between ^{244}Cm, the dominant source of neutrons after about three-year cooling time, and total plutonium has been proposed.[10] Figure 2 shows calculational results for this correlation using the detailed power history of a PWR assembly. This correlation appears to depend primarily on initial uranium enrichment.

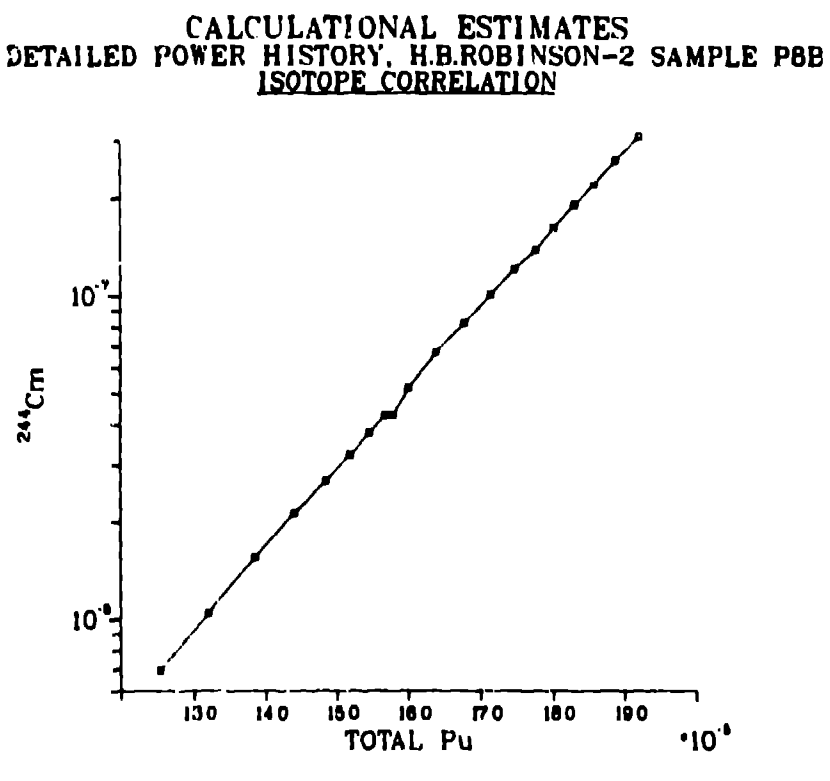

Fig. 2.
Correlation of ^{244}Cm with the total Pu for a typical PWR fuel assembly with a exposure of 31 220 MWd/MTU.

C. Active Neutron Techniques

Active neutron-interrogation techniques are being developed to determine the fissile contents of spent-fuel assemblies.[10,17,18,19] An external neutron source, either an accelerator source or an isotopic neutron source, induces fissions in the fissile isotopes, ^{235}U, ^{239}Pu, and ^{241}Pu. By measuring the prompt fission neutrons, an estimate of the fissile content can be obtained. Fast fission in ^{238}U has been estimated to be about 7% of the total count rate and is only slightly dependent on exposure.[9] If the prompt and delayed neutron signatures can be separated, the relative concentrations of the uranium and plutonium fissile isotopes may be calculated. If the fuel assembly can be removed from an aqueous environment, other assay techniques can be applied.[18,19] The active neutron technique has one distinct advantage over the techniques discussed previously: the fissile material is measured directly. The other nondestructive techniques measure signatures that may be correlated with fissile inventory.

III. APPLICATIONS

A. International Safeguards

The objective of international safeguards is "the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection."[20] To provide for timely detection, the use of materials accountancy is a safeguards measure "of fundamental importance, with containment and surveillance as important complementary measures."[20]

Nondestructive measurements can be used by international safeguards inspectors to establish the initial inventory of spent-fuel assemblies at a storage facility and to reverify the inventory if the integrity of the containment/surveillance measures fails. Nondestructive measurements can also be used during the routine inspection of a storage facility.

Two types of nondestructive measurements can be identified for inspector verification: (1) rapid qualitative measurements of spent-fuel assemblies, and (2) quantitative measurements of a limited number of the fuel assemblies. The inspection effort is limited by the available manpower and by legal constraints. An IAEA Advisory Group on the Nondestructive Measurement of Spent Power Reactor Fuels has recommended six levels of verification (Table II), ranging from verifications of the physical characteristics to measurements of the fissile contents of fuel assemblies.[14] The specific level of verification depends on the available resources and the desired level of assurance.

TABLE II

LEVELS OF SPENT-FUEL VERIFICATION AND NDA TECHNIQUES

Technique / 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	Gross Changes 1) Comparison of relative intensity of specific high-energy gamma rays 2) Comparison of relative values of measured isotope activity ratios	Gross Changes Comparison of relative neutron emission rate	1) Cerenkov 2) Mass by weighing 3) Be(γ,n) 4) TLD 5) Seals
Indication of Irradiation Exposure	Simple gross-gamma ray detection techniques	Simple passive neutron detection techniques	1) Cerenkov 2) Detection of heat 3) TLD
Presence of Fission Products or Actinides	Low- or high-resolution techniques for detection of Cs-137, Cs-134, Pr-144 and others	Verification of neutron rates expected for declared exposure	Be(γ,n)
Relative Concentrations of fission products or actinides	1) Correlations of ratios to exposure, cooling time, and initial enrichment 2) Consistency of measured and declared values	Passive: Relative exposure values $N = \alpha(\text{exposure})^{\beta}$ Active: Relative fissile contents	Be(γ,n)
Determination of Nuclear material Content	Correlations between HRGS results and destructive analyses or theoretical calculations	Passive: Cooling time dependence Active: Requires calibration standards	Not applicable

The IAEA currently relies on item counting of assemblies complemented by containment/surveillance at power reactors. The Division of Development of the IAEA has requested the development of nondestructive measurement techniques that can rapidly verify spent-fuel assemblies. Cerenkov light detectors, developed under the U.S. Technical Assistance Program,[21] have been adopted by Agency inspectors as a rapid verification technique. Other verification techniques that show promise are gross gamma-ray and passive neutron detection systems.

B. Facility Applications

Nuclear facilities that may use spent-fuel measurements to meet safeguards and materials-management requirements are those designed for interim storage or final disposition of irradiated nuclear fuels.[22] Interim storage facilities include at-reactor and AFR storages. Disposal facilities include reprocessing plants and geologic repositories designed for long-term storage of spent fuel. Table III gives a summary of potential facility applications.

Gamma-ray and passive neutron measurements currently are used for spent-fuel examinations at those reactors having examination facilities. Such measurements often are made on test fuel pins, such as mixed-oxide pins that have been irradiated in LWRs. It is possible that some reactor operators may adopt nondestructive measurements as an independent check of the fissile content of irradiated fuel prior to shipment for reprocessing.

TABLE III

FACILITY APPLICATIONS

Facility Type	Purpose	Measurement Technique
Reactor	Spent fuel examination (test pins); fissile confirmation	Gamma-ray and neutron spectrometry; passive neutron
AFR storage	S/R differences; fissile content; criticality safety; acceptance criteria; economic settlements	Active and passive neutron assay; passive neutron and gamma
Reprocessing	S/R differences; fissile content; criticality safety; efficient fissile recovery	Active neutron assay passive neutron and gamma

Additional capacity for AFR storage of spent fuel is rapidly becoming a necessity in the US, Japan, and Europe. In the US it has been proposed that the Government take title to excess domestic spent fuel (usually 5 years or longer after discharge) and that provision be made for storing a limited amount of foreign fuel.[17,23] It is currently envisioned that spent fuel will remain at an AFR storage facility for up to 30 years, pending final disposition to a geologic repository or to a reprocessing plant. If the disposition is via reprocessing, it may be necessary to return either the fissile material or an equivalent economic value to the original owner.

Nondestructive measurement systems are currently being designed to measure the total fissile and the plutonium contents of spent fuel received at AFRs.[17] System concepts include using active neutron interrogation, passive neutron, and gamma-ray techniques to meet safeguards and materials management requirements. The nuclear material content of spent fuels at power reactors is derived from reactor exposure calculations at discharge. Nondestructive measurements would be used at AFRs to close shipper-receiver (S/R) balances when the fuel is received for storage. Such measurements would also aid in the efficient utilization of available storage capacity, in economic settlements, and in answering questions that may arise concerning the fissile inventory. The measured values would provide a basis for settling S/R differences when the spent fuel is finally sent for reprocessing or permanent disposal and would assist in its efficient disposition.

Nondestructive measurement systems are being developed for reprocessing high-enriched uranium, LWR, and breeder spent fuels.[10,18,19] These measurements are intended primarily as an aid to criticality control in the dissolver tank and for efficient batch processing of the spent fuel for maximum fissile recovery. Specifications of the measurements typically include assay of the fissile contents to 5% or better with a total measurement time of 30 minutes or less. For example, the specifications for a ^{252}Cf Shuffler system to be installed at the new Fluorinel and Storage (FAST) Facility at Idaho Falls include 5% (2σ) assay precision over the range 2-10 kg ^{235}U in 30 minutes.[18] This instrument must also be capable of measuring the ^{235}U contents of high-activity solid wastes to 30 g (2σ) precision over the range 0-400 g ^{235}U in 30 minutes.

Spent-fuel measurements could also provide better safeguards at reprocessing plants. Spent fuel currently is received and stored using shipper's (reactor exposure) values. The S/R balance is not closed until the spent fuel is dissolved and the 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 head-end chop/leach operations, effectively decoupling the head-end process from the solvent-extraction process for purposes of accountability.

IV. SUMMARY

Nondestructive measurements on spent fuel will play an important role in meeting safeguards and materials management requirements in nuclear facilities. Nondestructive measurement techniques are summarized in Table I, international safeguards applications are summarized in Table II, and potential applications in various types of nuclear facilities are summarized in Table III.

ACKNOWLEDGEMENT

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