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GROSS GAMMA DOSE RATE MEASUREMENTS FOR TRIGA SPENT NUCLEAR FUEL BURNUP VALIDATION

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ABSTRACT

Gross gamma-ray dose rates from six spent TRIGA fuel elements were measured and compared to calculated values as a means to validate the reported element burnups. A newly installed and functional gamma-ray detection subsystem of the In-Cell Examination System was used to perform the measurements and is described in some detail. The analytical methodology used to calculate the corresponding dose rates is presented along with the calculated values. Comparison of the measured and calculated dose rates for the TRIGA fuel elements indicates good agreement (less than a factor of 2 difference). The intent of the subsystem is to measure the gross gamma dose rate and correlate the measurement to a calculated dose rate based on the element s known burnup and other pertinent spent fuel information. Although validation of the TRIGA elements' burnup is of primary concern in this paper, the measurement and calculational techniques can be used to either validate an element's reported burnup or provide a burnup estimate for an element with an unknown burnup.

INTRODUCTION

The United States Department of Energy (DOE) is currently responsible for over 200 types of spent nuclear reactor fuels [1]. Most of these fuels stem from 40 years of DOE-operated and sponsored research, test, and other experimental reactors, but many commercial fuels are part of the inventory as well. These fuels have been stored at various domestic and foreign sites around the world and are now being consolidated for interim storage at the Idaho National Engineering and Environmental Laboratory (INEEL), the Hanford site, and the Savannah River Site (SRS).

Spent fuel from many of these reactors, now decommissioned, had unique design features such as core configuration, fuel and assembly geometry, reflector and coolant materials, operational characteristics, and neutron spatial and spectral properties. Over the past 40 years, important reactor data were not always recorded and maintained in accordance with a modern quality assurance program compatible with current Office of Civilian Radioactive Waste Management requirements. As a result, much of the spent fuel inventory today has inadequate (or non-existent) burnup and fuel specific information for spent fuel characterization purposes.

In order to characterize a spent fuel element in terms of its radionuclide inventory, calculations are relied on to provide the necessary activity estimates. Usually anywhere from 50-150 important radionuclides

(activation, actinides, and fission products) are required to fully characterize a fuel element for handling, shipping, and storage requirements. Measurement of these radionuclides for each element would be prohibitive, hence the need for calculational characterization. An accurate calculational estimate of the radionuclide inventory in turn, relies on characterization information as input data that includes the following data:

(1) Burnup (MWd, MWd/MTU, %U235, etc),
(2) Exposure or power history,
(3) Beginning-of-life (BOL) heavy metal loading,
(4) Date element loaded into the core,
(5) Date element removed from the core,
(6) Cooling or decay time, and
(7) Applicable neutron cross sections.

The availability and accuracy of the above characterization information determines the accuracy of the characterization calculation. The burnup, by far, is the most important piece of information needed to characterize a spent nuclear fuel element. Most of the other information is available or can be conservatively estimated based on circumstantial information in order to fill in data gaps. With the burnup and the other pieces of characterization information, one can calculate the spent fuel element's radionuclide inventory, activity levels, decay heat, and gamma-ray emission spectrum. From the gamma emission spectrum, one can further calculate gross gamma dose rates at a distance from the element. Dose rate correlates to element burnup and it is this correlation that is used here to validate a reported element burnup.

The gross gamma dose rate detection system was conceived and developed specifically for the purpose of experimentally measuring spent fuel gross gamma dose rates as a function of distance from the element. The measured dose rate can then be compared to a calculated dose rate based on an estimated burnup. If the dose rate comparison is good, the reported burnup and the estimated burnup used in the calculation should be close. If the burnups are close, then the reported burnup is assumed to be validated. Alternatively, if a spent fuel element does not have a reported burnup, the burnup used in the calculation can be iterated until the calculated and measured dose rates agree. The burnup in the final iteration can then be used as an estimated burnup for the element and the corresponding calculated radionuclide inventory can be used to characterize the element. Therefore, the technique can be used to either validate a reported fuel element burnup, or provide a burnup estimate for a fuel element with an unknown burnup.

SYSTEM DESCRIPTION

The In-Cell Examination System is crane-portable and it consists of multiple subsystems, which include a shielded, collimated, gross gamma dose rate measurement and gamma-ray spectroscopy systems with integral shielded laser distance measurement and video camera components (Figure 1). The In-Cell Examination System is physically located in the hot cell of the Irradiated Fuel Storage Facility (IFSF) building at the INEEL and is mounted on a moveable platform and integrated with other remote hot cell handling equipment for versatility and operation in the IFSF building. The major In-Cell Examination System components will be described in some detail below.

Gross Gamma System

The Gross Gamma Detection System is composed of two Eberline RO-7 ion chamber detector heads (fill gas is air), digital readout instrument, and various connecting devices. The ion chambers provide for remote monitoring of high range beta and gamma radiation fields. For gamma radiation, the ion chambers' detection range can extend from 1.0 mR/hr to 20,000 R/hr. The whole system is mounted to an INEEL-designed and fabricated partial collimator/shield.

The cylindrical collimator shield is a carbon steel shell with lead shielding and an open front face that makes an otherwise omni-directional head primarily sensitive to the gamma radiation entering the head from the front. The outer carbon steel wall thickness is 5.5-mm, the lead thickness 17.9-mm, and the inner carbon steel liner 1.2-mm thick. The collimator front face opening is 3.81-cm in diameter with the detector heads recessed 10.16-cm from the opening. The recessed detector position mitigates back-scatter radiation from the walls, ceiling, and floor but was designed to allow sufficient viewing angle for the detector to receive direct gamma radiation from the entire fuel element length. For standard TRIGA elements with active fuel lengths of 38-cm, the entire element can be viewed at distances as close as 51-cm (20-in).

The detector response to the gamma radiation field is then converted into a calibrated dose rate signal. This dose rate signal (mR/hr) is reported as a function of distance and fuel element identification number and is the measured dose rate.

The two ion chamber detectors, one a mid-range head (<200 R/hr) and the other a high-range head

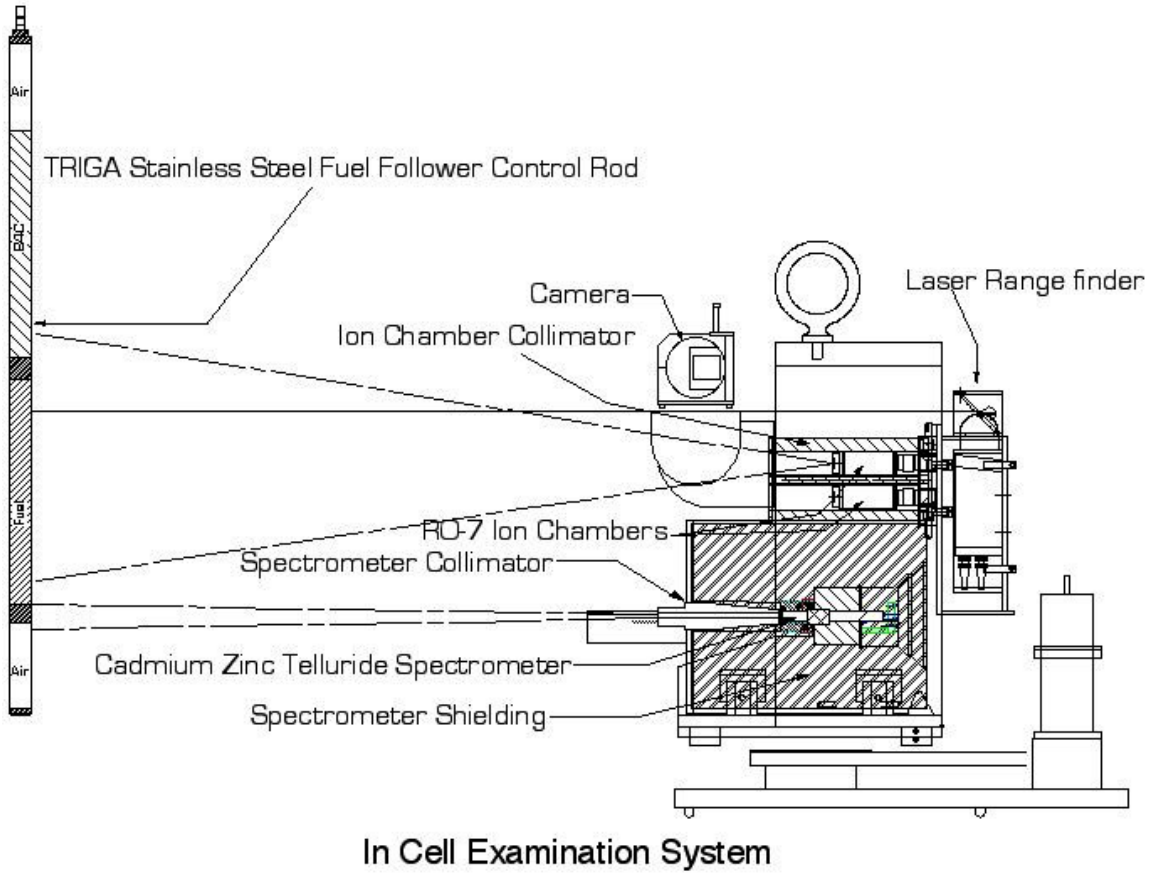

Figure 1. Sketch showing the In-Cell Examination System with the gross gamma RO-7 ion chamber detectors, gamma-ray spectrometer, shielding, platform, laser, and FFCR TRIGA fuel element.

(<20kR/hr) were calibrated to a 450 Ci (1982) Cs-137 source both with and without the collimator. The detectors were mounted on the High Level Calibration Well slide and positioned so that the detector head was parallel to the collimated beam. The Cs-137 source is mechanically raised and lowered to change the distance between the source and detectors. The mid-range head was calibrated to its maximum value of 200 R/hr and the high range head to 300 R/hr. Because the ion chamber function is linear, it is believed that the high-range head will be correct at all higher values. The total estimated uncertainty for the setup and calibration is ±11.8% at the 95% confidence interval.

Gamma Spectrometer System

The Gamma Spectrometer System consists of an eV Products SPEAR cadmium-zinc-telluride (Cd-Zn-Te) gamma spectrum detector, a tungsten collimator, bismuth shielding, and cabling. It is situated on the moveable platform at an elevation of approximately 30-cm. The detector is a miniature semiconductor unit that is approximately the size of a writing pen, including the preamplifier. The Cd-Zn-Te crystal is a cube, 5mm on a side, and is positioned 1.3 mm back from the window. The crystal operates at room temperature.

The Cd-Zn-Te detector is very efficient for low energy gammas (<100keV) and is useful up to approximately the Cs-137 line at 662 keV. The intrinsic efficiency (counts per gamma-ray) is 1.05E-3, 5.1E-6, and 9.2E-7 for 121.8, 778, and 1408 keV gamma energies, respectively. The Full Width Half Maximum is measured to be approximately 12 keV for the Cs-137 662 keV line, but has a strongly tailed pulse shape at lower energies.

The bismuth shield is approximately 33.02-cm OD and 15.24-cm thick with a stepped opening at the front end for insertion of a collimator. The shield can be fitted with one of three INEEL-designed and fabricated tungsten collimators units (1.0, 3.0, and 10.0 mm apertures) to control saturation of the detector. Collimator lengths are approximately 20-cm. The collimators are discrete units that are remotely changed by using an electro-mechanical manipulator located in the hot cell dry handling cave.

The spectroscopy system was not used in the gross gamma measurements, but it should be useful in future measurements for estimating spent fuel cooling times as well as burnup.

Laser Range Finder

An Acuity Research Accurange Class IIIa laser rangefinder is mounted in an INEEL-designed and fabricated shielded box for the purpose of measuring the distance (up to 55 feet) from the source (fuel element). The rangefinder axis is parallel to the axis of the ion chambers and is shielded to minimize damage to the electronics by high radiation fields. It is also mounted behind the spectrometry detector's bismuth shield unit with the beam directed from a 90-degree mirror to eliminate direct exposure of the return sensor to direct gamma photon sources.

Video Camera

The RJ Electronics RCS-510 shielded, radiation-hardened video camera provides a view of the fuel element being evaluated. A closeup view allows confirmation of item serial numbers and cladding integrity.

The whole set of sensors are all mounted as a unit that can be rotated by operating an electrically-driven rotary table to point at a fuel element suspended on a handling tool. Being able to aim the unit allows a high degree of flexibility with respect to where measurements can be taken during the fuel element and canister handling process.

SPENT FUEL ELEMENTS

In order to test the gross gamma detection system, spent fuel elements were required to perform test measurements. Upon completion of the system setup, a shipment of Foreign Research Reactor (FRR) fuel elements were received at the IFSF in June 2001. The shipment included TRIGA (Training, Research, and Isotope General Atomics) spent nuclear fuel elements from the Heidelberg II TRIGA reactor in Heidelberg, Germany. The fuel elements required repackaging in the IFSF hot cell and were destined for semi-permanent storage in the IFSF dry storage area. Permission to remove several elements from the canisters provided the opportunity to take gross gamma measurements.

While in the hot cell, the six elements were individually withdrawn from their storage canisters and suspended over the IFSF hot cell floor for the dose rate measurements. Gross gamma dose rates at the element mid-plane (maximum dose rate) were measured along with the distance between the detector and the element using the laser sensing system.

TRIGA Element Description

The six Heidelberg TRIGA elements consisted of 2 aluminum-clad standard elements and 4 stainless steel-clad fuel follower control rods (FFCR).

The fueled or active part of each aluminum-clad element was 3.56-cm (1.4-in) in diameter and 35.56-cm (14-in) in length. The fuel meat was uranium-zirconium hydride (U-$ZrH_{1.0}$) with an 8.0 wt% uranium loading enriched to 19.9% U-235. BOL uranium loadings were 36.0 grams of U-235 and 144.0 grams of U-238. Above and below the fuel are two cylindrical graphite reflectors. Each element is clad in 0.076-cm (0.03-in) thick aluminum with aluminum end fixtures at both end of the element. These elements experienced relatively low burnups in the range of 1-4% U-235 fissioned.

The FFCR element consisted of an active region with a 3.016-cm (1.19-in) diameter and 38.1-cm (15-in) length. The fuel meat was U-$ZrH_{1.6}$ with 8.5 wt% uranium loading enriched to 19.9% U-235. BOL uranium loadings were 32.5 grams of U-235 and 128.5 grams of U-238. Above the top graphite reflector, the FFCR also has a 38.1-cm long boron carbide neutron absorber section with the boron enriched to 18.5% B-10. The FFCR element is shown in Figure 1. These 4 elements experienced similar mid-range burnups of 12.3% U-235 fissioned.

A more detailed physical description of these two types of TRIGA elements is given in reference [2].

The Appendix A Data

Part of the INEEL's Foreign Research Reactor (FRR) acceptance criteria is the disclosure of the spent fuel characterization information on an Appendix A form. The Appendix A contains a full physical description of the spent fuel elements along with the element s identification number, fuel type, burnup estimate (%U235), BOL uranium mass loading, EOL uranium and plutonium isotopic masses, decay heat, core residency time, and decay time. Any subsequent verification checks by INEEL analysts of the EOL isotopics or decay heat must always be based on the burnup data given in the Appendix A. The reported burnups for the 6 Heidelberg elements are from the operator-supplied Appendix A data [3].

METHODOLOGY

The calculational methodology used to generate the TRIGA parametric study results [4] that in turn is used to estimate the Heidelberg fuel element dose rates as a function of burnup is based on three standard and well-known reactor physics computer codes. These codes collectively calculate burnup-dependent neutron cross sections, perform the depletion calculation, generate the gamma source term, and transport the gamma radiation for the dose rate estimates. The calculational methodology is described in detail in Reference [5] and will not be repeated here other than to mention the codes and their specific application to the parametric study of TRIGA fuels.

Computer Codes

The MCNP4A (Monte Carlo N-Particle code version 4A) code is a general purpose, continuous energy, generalized geometry, coupled neutron-photon-electron Monte Carlo transport computer code [6]. The MCNP code performs the neutron transport calculation to determine the flux and reaction rates in the geometry model cells that translate into neutron cross sections. A wide variety of nuclide cross sections and reactions are available from the ENDF-4, ENDF-5, and ENDF-6 data sets. MCNP was also used to perform the gamma radiation transport calculation to estimate the fuel element dose rate as a function of burnup and distance from the element.

The ORIGEN2 (Oak Ridge Isotope Generation) computer code [7] is used for calculating the buildup, destruction, and decay of stable and radioactive isotopes. Coupled ordinary differential equations describing the complex burnup and decay mechanisms between the nuclides are solved using the matrix exponential method. Several standard neutron cross section, decay, and production data libraries are available with the code for a narrow range of applications. A special feature of the code is a user option to modify the standard cross section libraries with user calculated cross sections, thereby allowing a burnup calculation to be reactor-specific. ORIGEN2 performs the burnup calculation with effective one-group cross sections with depletion controlled either by power or irradiation flux level.

The MOCUP (MCNP-ORIGEN2 Coupled Utility Program) code [8] is a utility code composed of a system of external processors that links input and output files from the MCNP4A and ORIGEN2 codes in order to perform a time-dependent burnup of a fuel composition. MOCUP is composed of three processing modules, namely, mcnpPRO, origenPRO, and compPRO. Each module performs specific, sequential tasks during each time step or burnup iteration.

Calculational Methodology

The calculational process begins with the development of an MCNP material and geometry model of the reactor core. For the TRIGA parametric study, a 1/12-

core model of a MARK I TRIGA reactor was developed which included 12 full and partial fuel elements, light water coolant, a graphite radial reflector, and an outer light water reflector. The MCNP model is then used to calculate average fluxes and reaction rates in the fuel elements for 37 different fuel actinides and 77 fission products. Calculated neutron reaction cross sections include radiative capture and fission, and four threshold particle reactions: (n,2n), (n,3n), (n,alpha), and (n,p).

Four separate models were developed for each of the 4 different TRIGA fuel elements considered in the parametric study. Each model had a full core loading of each element type. Cross sections were developed only for BOL core conditions for the parametric study depletion calculations.

A separate MCNP4A TRIGA fuel element model was developed specifically for the parametric study dose rate predictions. The ORIGEN2 generated gamma emission rates as a function of burnup were used as the fuel element source in the model. MCNP4A performed the gamma radiation transport through the fuel element and clad into the surrounding air. Dose rates were calculated at clad contact, 3-feet, and 3-meter distances from the element at midplane.

TRIGA Parametric Study

The calculated dose rates used in the dose rate comparison are based on interpolated values from a TRIGA spent fuel parametric study [5]. The parametric study is a comprehensive analysis which presents complete radionuclide inventories (145 radioisotopes), decay heat, and gamma dose rates as a function of the TRIGA element type, burnup, and cooling time. The parametric data are presented in the form of curves and tables for easy interpolation with the intent to quickly characterize a given spent TRIGA fuel element with limited characterization information.

The following 4 TRIGA fuel element types were considered in the parametric study:

(1) Aluminum-clad standard element,
(2) Stainless steel-clad standard element,
(3) High enrichment (70%) FLIP (Fuel Lifetime Improvement Program) element, and the
(4) FLIP-LEU-I (Low Enrichment Uranium) element.

The parametric study dose rate curves are presented as a function of burnup, decay time, and distance from the source. It is these dose rate curves that can be interpolated to compare with the measured dose rates.

The parametric data can be used to characterize a TRIGA spent nuclear fuel element without a time-consuming depletion calculation for each individual spent fuel element. It can save a considerable amount of time and allows one to characterize the TRIGA spent fuels with reasonable accuracy. Given only the limited spent fuel element information in a fuel element's Appendix A, one can still characterize the spent fuel element using the parametric study data.

Parametric Study Application

The calculated gross gamma dose rates for the six TRIGA elements were estimated based on the Heidelberg Appendix A data and the parametric study. The relatively straightforward procedure used to estimate the calculated dose rates is given below.

First, the Appendix A burnup data (% U-235 burnup) is used to determine the mass (grams) of U-235 fissioned and the corresponding MWd (Megawatt-days). The dose rates in air are plotted parametrically as a function of fuel element burnup (MWd) and as a function of distance from the element in the study s dose rate versus distance plots. The burnup (MWd) determines which parametric curve will be used. Once the dose rate figure and curve are established, the decay time (from the Appendix A) is used to determine the dose rate (mR/hr). The burnup may lie between two parametric values and the determination of the dose rate will necessitate an interpolation between the two curves.

Once the dose rate is determined, it needs to be adjusted through multiplication by two additional factors. These two multiplicative factors are needed in order to take into account the effect of the element s in-core residency time and the difference in the actual distance from the element to the detector relative to the parametric study distance of 3 meters. These two multiplicative factors are then multiplied by the interpolated dose rate to obtain the final calculated dose rate estimate for the spent fuel element.

RESULTS

The table below presents the calculated dose rate estimates as determined from the Section 4 methodology above and the actual gross gamma dose rates as measured by the RO-7 gross gamma detector system.

Comparison between Measured and Calculated Gross Gamma Dose Rates.

Element ID	Element[a] Type	Burnup[a,b] (%)	Decay[c] Time (days)	Time[a] in Reactor (years)	Distance (cm)	Meas. Dose Rate (mR/hr)	Calc. Dose Rate (mR/hr)	Ratio[d]
4240E	Al-clad	3.9	8,765	10.4	311.76	100	77	1.30
3428E	Al-clad	1.9	10,957	5.0	104.95	600	342	1.75
7171	FFCR	12.3	725	21.5	110.92	2,200	2,343	0.94
7172	FFCR	12.3	725	21.5	105.54	2,200	2,588	0.85
7173	FFCR	12.3	725	21.5	117.40	2,100	2,092	1.00
7170	FFCR	12.3	725	21.5	99.09	3,300	2,932	1.13

a. Heidelberg Appendix A data [4].
b. Burnup is number of grams of U-235 fissioned.
c. Decay time is the time provided in the Appendix A plus 1-year (gross gamma measurements were taken approximately 1-yr later).
d. Ratio=Measured/Calculated

CONCLUSIONS

The agreement between the measured and calculated dose rates presented in the table is very good. The far right column (Ratio) indicates that the measured and calculated dose rates are within a factor of two of each other, and all except one are within ±30% or better. The agreement appears to be slightly poorer for the 2 aluminum-clad standard elements relative to the FFCRs. This may be due to greater uncertainty in the reported Appendix A burnup for the two aluminum-clad elements. These elements would have had a greater potential for in-core movement over their lifetimes versus the fixed-core position FFCRs. The good agreement between the measured and calculated dose rate values should provide some level of confidence and validation for the burnups reported in the Heidelberg II Appendix A.

Several variables can contribute to the uncertainty in both the calculated and measured dose rate values. These include:

(1) the accuracy of the Appendix A burnup data,
(2) use of the parametric study interpolation procedure versus a detailed depletion calculation for each individual element,
(3) error in the distance measurement, and
(4) the detector dose rate measurement error.

The In-Cell Examination System with its various subsystems has the potential to provide a valuable service for spent fuel characterization. It can validate Appendix A reported burnups for TRIGA as well as other spent nuclear fuel received by the INEEL and has the potential, combined with the depletion calculations, to provide a burnup estimate for spent fuel elements with unknown burnups.

ACKNOWLEDGMENTS

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