10CFR61 IRRADIATED COMPONENT CHARACTERIZATION AT CRYSTAL RIVER UNIT 3

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ABSTRACT

Since the enactment of 10CFR61, nuclear utilities have experienced difficulty in obtaining representative samples of irradiated hardware. Problems stem from the very high dose rates of irradiated hardware (1,000 - 50,000 R/hr), which necessitates the execution of a sampling procedure underwater in fuel storage pools. FPC Crystal River unit 3 approached the problem by utilization of direct assay technology, which quantifies the 10CFR61 radionuclides that are either gamma-emitting or neutron-emitting (TRU) radionuclides.

The direct underwater assay of in-core instrumentation and SSHT bolts was performed by using direct assay equipment which was submerged in the fuel pool. This work was done in conjunction with an EPRI technology demonstration project which evaluated the current technologies and techniques used for direct measurement of radionuclides in waste packages. Additionally, the direct assay equipment characterized 10CFR61 radionuclides in waste packages containing CVCS filters, primary resins, radwaste resin, and DAW. This paper presents the results of the direct assay techniques demonstrated at Crytal River unit 3.

BACKGROUND

The disposal of low level radioactive wastes has become a major issue for nuclear power plant operators. The enactment of 10CFR61 and 49CFR170-178 has increased analysis and packaging requirements for radwaste. Waste disposal rate structures now include charges for radioactivity content in addition to the usual volume and radiation level of the waste. 10CFR61 further requires the identification and quantification of radionuclides which cannot be measured with radiation detection equipment currently available on-site at nuclear stations. Consequently, nuclear stations have had to rely on commercial radioanalyticial laboratories for "representative" waste sampling and analysis of radioisotopes specified in the 10CFR61. Unfortunately, not all waste streams are conducive to "representative" sampling because of nonuniformity and/or high dose rates. Reactor components which have been irradiated certainly fall into this category.

In response, Florida Power Corporation and the Electric Power Research Institute engaged in a research project specifically aimed at evaluating the technology and methods currently available for performing direct measurements of the radioactivity emitted from irradiated components at Crystal River unit 3. This paper presents a summary description of the direct assay technologies and methods demonstrated; the results of the assays; and the cost savings realized.

Project Description

Crystal River unit 3 (CR-3) was confronted with the difficult task of radionuclide characterization of irradiated in-core instruments (ICIs), core barrel bolts (CBB), and control rod retainer clips. The total volume of this waste was approximately 14 cu. ft, including the waste canister. Dose rates from the storage canisters ranged from 8,000 R/hr to 21,000 R/hr.

CR-3 has available two calculation methods for determination of the curie content of irradiated components: ORIGEN-S Code and the dose to curie content conversion calculation reported in the Health Physics Journal, Nov. 1976 referred to as WP-103. Conservative application of the ORIGEN code assumes that the irradiated component saw the maximum in-core neutron flux and that the component was irradiated uniformly over the volume. During Refuel II, the irradiated ICIs were cut and evenly distributed by high and low dose rate into four storage canisters. During Refuel IV, the high dose rate ICIs ends were put into one canister and the lower dose rate parts into three other canisters. This produced a problem in calculating the curie content using the ORIGEN code since the curie content for a canister was assumed to include the entire ICI. The WP-103 calculation proved to be a problem because of the high waste density and the energy correction factor. After considering the problems, inaccuracies and conservatism with the application of the two calculational methods, CR-3 decided to investigate the use of direct assay technology.

Under cosponsorship with EPRI, researchers from Battelle Pacific Northwest Laboratories (BNW) and Science Applications International Corporation (SAIC) were brought on-site in November 1985 to demonstrate three direct assay techniques, one for transuranics and two methods for gamma-emitting radionuclides. Surface scrape samples, as well as metal samples, were taken of irradiated components and sent to SAIC for radiochemistry analysis to allow a basis for comparision with the direct assay results.

Direct Assay Technology

EPRI had implemented a research project to evaluate and demonstrate direct assaying techniques. The advantages of utilizing radiation detection technology for direct identification and quantification of radionuclides in the waste package are: (1) reduces the errors introduced by sampling, (2) reduces the associated personnel radiation exposure involved with sampling, in particular high dose rate non-uniform waste streams, (3) reduces the need to sample waste

streams, (4) and yields timely results so that packaging and transportation decisions can be made. Direct assay techniques were to be evaluated on the accuracy, practicality, cost and conformance with the ALARA principle.

Transuranics by Direct Assay

BNW uses technology and methods which allow for direct measurement of gamma- and neutron-emitting radionuclides. The TRU assay technique is based upon the measurement of the neutron emissions from the transuranics present in the various radwaste streams. These neutrons are produced from either spontaneous fission of the TRU radionculides or via their alpha particle emissions by (α,n) interactions with light isotopes present in the waste. The neutrons emitted from a waste package are detected by a special array of $^{10}BF_3$ tubes surrounding, or adjacent to, the waste package (see Fig. 1). The TRU assay system is calibrated to a known ^{244}Cm standard in the exact geometry the waste is to be assayed in. A computer code, which incorporates input from the ORIGIN code, converts the total neutron counts measured by the detector system into individual transuranic concentrations, including ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm and ^{246}Cm. The technology has been demonstrated on a wide variety of waste package geometries and radwaste materials, including activated components (ICI, LPRM, CRB), resins, filters, evaporator bottoms and DAW.

A submersible direct assay system was used for TRU characterization of irradiated components at CR-3. The system is composed of 29 $^{10}BF_3$ neutron detection tubes (66-cm long x 5-cm dia.) encased between lucite moderator material and housed in a retangular, water-tight steel box with 20-cm thick lead gamma shield, Fig. 2. This assay system has a detection sensitivity of approximately 10 nanocuries/gm of total TRU (half-lives greater than 5 years, not including ^{244}Cm and ^{241}Pu) for a 10-minute count of the 48-cm long x 20-cm dia. storage canisters. This sensitivity could be lowered to about 1-2 nanocuries/gm for a 1,000-minute count.

Gamma-Emitters by TLD Dose Profiling Method

In order to obtain a complete 10CFR61 characterization the CR-3 irradiated components, BNW uses a second direct assay technique for identification and quantification of gamma-emitting radionuclides. The method uses a 1-inch ionization chamber, a PGT intrinsic germanium (IG) detector and calibrated thermolumenescent dosimeters (TLDs). The ion chamber and the TLDs provide dose and dose rate profiles, while the IG detector measures the gamma energies present, see Fig. 3. All instruments are calibrated to NBS traceable sources prior to use. The 1-inch chamber used only as backup to the TLDs is calibrated at the BNW calibration facility. Several dose points are taken ranging from exposures of 10 R to 1000 R. A calibration in R/minute was established for direct reading of the ion chamber electronics. TLDs were also calibrated to Co-60 at the BNW calibration facility. TLDs have a near one to one energy response from 0.1 to 5.0 MeV which makes them an excellent dosimeter for measuring mixed source fields. The gamma scan provides the relative ratio of the radionuclides contributing to the dose rate measured by a series of TLDs placed every four inches along the length of the storage canisters. This infomation becomes input to the ISOSHLD computer code along with the geometry of the canisters. The radionuclide concentrations are then varied in the same measured relative ratios until the dose rate measured by the TLDs are matched.

Through the use of correlation data and the elemental composition of the ICI material present in the canisters, the curie contents of the remaining activation and fission products radionuclides specified in 10CFR61 are calculated. Correlation data were taken from NUREG/CR-3474, "Long Lived Activation Products in Reactor Materials," August 1984.

Gamma-Emitters by Spectroscopy

SAIC uses collimated high-resolution spectroscopy to measure quantitatively the gamma-emitting radioactivity in bulk waste. The system is tradenamed QuantiScan™. The system consists of a high-purity germanium gamma-ray detector installed in a shielded collimator and a portable microcomputer that controls the acquistion, analysis and reporting of data. The technique that makes this system of direct scanning usable for quantitative measurements is the calibration method. Without the ability to measure quantitatively the different geometries found in all plant radwaste, a gamma-ray scan can give only qualitative results that are of minimal use in radwaste assay. Quantiscan™ and an RO-7 teleprobe were used for direct assay of the CR3 irradiated component storage canisters. The Quantiscan™ provides the relative ratio of the gamma emitters. The RO-7 provides an average dose rate. This information serves as input to the computer code which calculates the radionuclide concentrations of the gamma-emitters.

TRU Results

Table I presents the results of the direct underwater assay of irradiated component canisters as measured by BNW TRU assay system. The total TRU concentrations ranged from 2 to 16 nanocuries/gm. There is some uncertainty associated with the results from canister 1-4, in that the high dose rate may have interfered with the neutron counting due the the gamma flux. The Class C limit for total TRU is 100 nanocuries/gm of waste material. All canisters contained TRU concentrations well below this limit.

TABLE I

Direct Underwater Assay of TRU Contents
of In-Core Radwaste Components
Crystal River, November 1985

Sample	Maximum Dose Rate (R/hr)	Total TRU† (nCi/gm)	^{242}Cm (nCi/gm)	^{241}Pu (nCi/gm)
1-3	11.4	1.5 ± 7.6	0.024 ± 0.2	40 ± 200
1-4	21,700	16 ± 3*	0.25 ± 0.05*	430 ± 93*6
2-2	13,470	8.2 ± 4.3	1.8 ± 0.9	150 ± 80
2-3	13,128	5.6 ± 2.6	1.2 ± 0.6	103 ± 48
2-4	12,510	7.6 ± 3.2	1.7 ± 0.7	141 ± 60
10CFR61				
Class "C" Limit		100	20,000	3,500
"B" Limit		---	---	---
"A" Limit		10	2,000	350

*Possibly some interference with the neutron counting due to extremely high gamma-ray flux.

†Includes all significant TRU isotopes with half-lives greater than 5 years.

Scrape samples and one metal fragment sample were taken from a low dose rate piece of ICI. SAIC

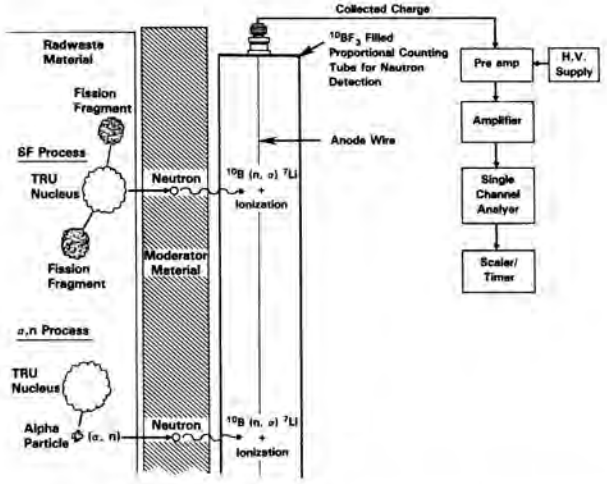

Fig. 1. Schematic Diagram of Neutron Detection from TRU Radionuclides in Radwastes.

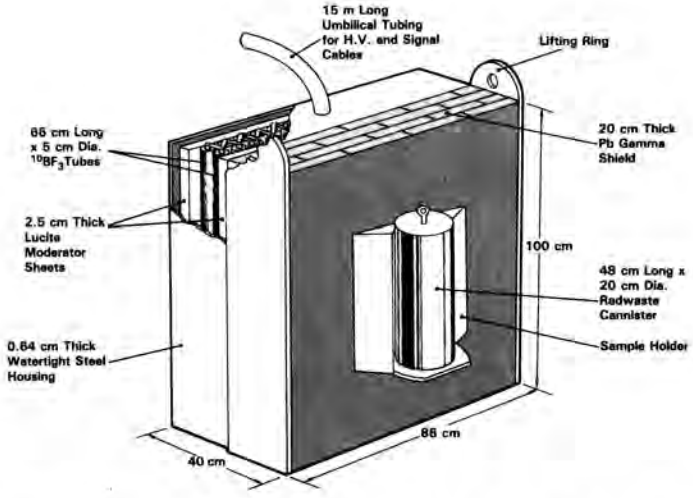

Fig. 2. Underwater Neutron Detection System for Direct Assay of TRU Contents of Highly Radioactive Waste.

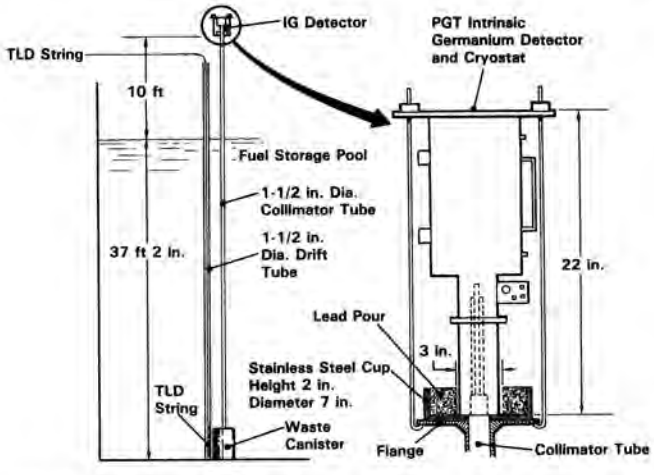

Fig. 3. Collimator Tube and Drift Tube Positions fro Gamma Spectrum and TLD Measurements on Canisters of In-Core Waste Components.

performed the chemical analysis of these samples. The results for the TRU are shown in Table II.

TABLE II

Scrape Sample TRU Radiochemistry Results

Sample ID	Total TRU (nCi/TRU)	^{242}Cm	^{241}Pu
1-4	0.72	0.0089	22.0
2-3	0.72	0.0089	22.0
2-4	0.72	0.0089	22.0

These quantities are about a factor of ten below the BWN TRU direct assay results. It is difficult to compare these results, since the radiochemistry is based on a very small sample, it is not known whether the deposit of TRU on the surface of these samples is uniform in distribution across all the ICIs in the canisters. It is clear from both the BNW direct TRU assay and the SAIC radiochemistry results that this waste would not be above class C due to TRU.

Gamma Assay and Scaling Factor Results

Figures 4 and 5 show the BNW result on dose profiles of the ICI storage canisters. These results graphically show that the dose profile was relatively constant over the height in which the ICI pieces reached inside the canister. Table III presents the ^{60}Co results from the BNW and SAIC gamma assays and the in-house methods CR3 initially considered using.

TABLE III

Curies of ^{60}Co in Canisters

ID#	BNW	SAIC	WP-103	ORIGIN
1-4	707	690	1,970	5,250,000
2-2	439	N.R.	1,300	785,000
2-3	427	540	1,260	763,000
2-4	407	600	1,200	727,000

These results show that both the BNW TLD dose profile method and the SAIC Quantiscan/RO-7 method were in reasonable agreement for the total curies of ^{60}Co in each canister. It is apparent that the WP-103 and the ORIGEN methods, with the very conservative assumptions used, would have resulted in a total curie content greater than a factor of two high.

Table IV and V show the ^{63}Ni curie content and the ^{55}Fe content when the direct assay results were scaled. BNW using scaling factors from NUREG/CR-3474, which defines the elemental content of Inconel and stainless materials. SAIC ratioed their radiochemistry results to the direct assay results to obtain total curie quantities for ^{63}NI and ^{55}Fe.

TABLE IV

^{63}Ni Curie Content (Scaled #s)

Sample ID	BNW	SAIC	ORIGEN	WO-103
1-4	1,070	84	439	2,990
2-3	531	63	70	1,570
2-4	507	72	67	1,500

TABLE V

^{55}Fe Curie Content (Scaled #s)

Sample ID	BNW	SAIC	ORIGEN	WP-103
1-4	358	192	324	1,000
2-3	263	192	37	770
2-4	251	216	35	736

Benefits

Along with the increased confidence in the curie characterization using direct assay method, there is the obvious benefit of cost savings. The conservative calculational methods FPC had considered using would have greatly penalized CR3 in cost for disposal. The curie surcharges for each method are shown in Table VI below. The packaging, transport and other burial charges are equal.

TABLE III

Curie Surcharge Costs for Disposal

FPC Computer Code	$95,000
WP-103 Dose to Curie	$60,000
BNW	$40,000
SAIC	$45,000

SUMMARY

The results show that BNW direct assay method for TRU is sensitive enough to detect transuranic levels sufficient to allow for waste classification. Both BNW method and SAIC method for gamma emitting radionuclide quantification demonstrate a significant improvement over the FPC calculatinal methods for curie content determination. These methods could be adopted by radwaste operations as a means for more accurately defining the curie content of high dose rate waste and consequently result in a cost savings. The cost savings for low activity waste would diminish as the total curies in a waste package diminish. All the results from the direct assay project will be published by EPRI.

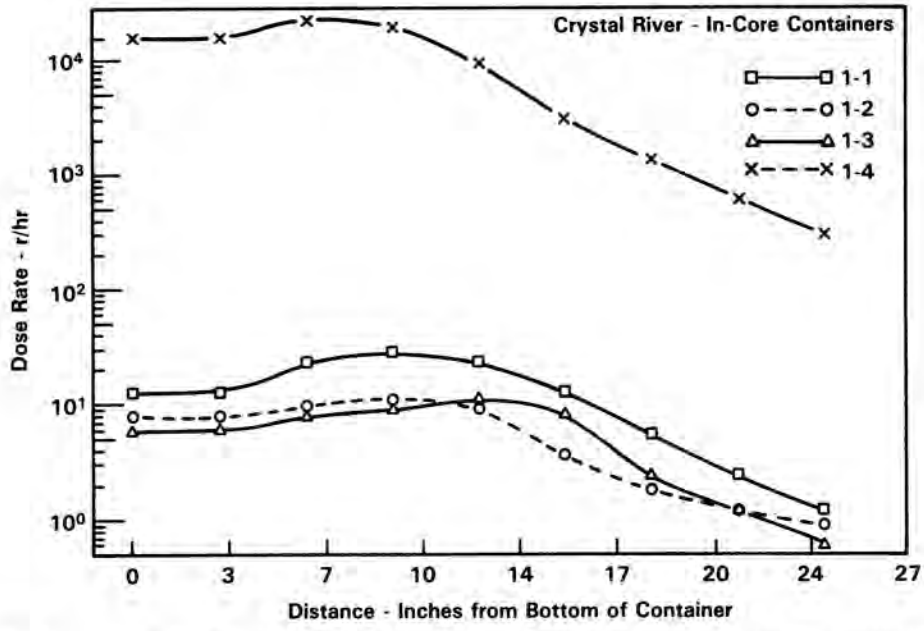

Fig. 4. TLD Dose Rate Profiles of Cannisters of In-Core Monitor Waste Components.

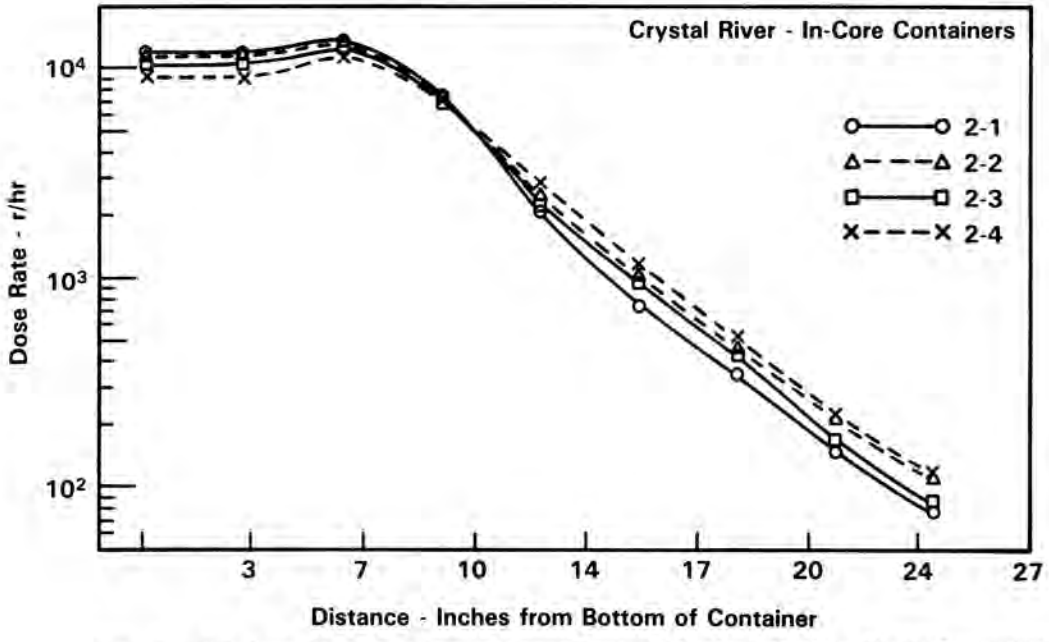

Fig. 5. TLD Dose Rate Profiles of Cannisters of In-Core Monitor Waste Components.