Characterization of Decommissioned Reactor Internals: Direct-Assay Method Assessment

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ABSTRACT

This study describes the direct-assay technique for measuring activation levels of irradiated reactor component hardware. It also compares the direct-assay technique with calculational analysis methods that predict activation levels. Direct assay is performed in four steps: (a) planning and component selection, (b) onsite measurements, (c) radiochemical analysis, and (d) data analysis and classification. Uncertainties are estimated for each step of this process, and an overall uncertainty in the classification accuracy is calculated as about $\pm 35\%$. Numerous research ideas are identified to help reduce the uncertainty level; many of these ideas would improve activation determinations performed by either direct assay or by calculational analysis methods.

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1. INTRODUCTION

1.1 Background

Nonfuel-bearing metal components in operating power reactors activate to relatively high levels of radioactivity through absorption of neutrons during power operation. Typical components are of stainless steel; the principal activation products are ^{55}Fe , ^{60}Co , and ^{63}Ni , the first two having relatively short half-lives. The list of components includes boiling water reactor (BWR) control rod blades and in-core instrument tubes, and some pressurized water reactor (PWR) in-core guide tubes, rod-control cluster assemblies, and burnable poison control assemblies. Routine reactor refueling operations discharge many "used" components as waste into the plant fuel-pool storage area, where the water acts as a radiation shield for the intense ^{60}Co radiation and as a cooling medium. These discharged waste components are periodically classified in accordance with the requirements of 10 CFR 61,¹ with subsequent packaging and shipment to low-level waste disposal sites. Because of the high radiation levels from the pieces, often exceeding 20,000 R per hour at contact, component characterization, classification, and packaging occur in the fuel pool. Irradiated hardware usually has a Class C waste classification. The shorter-lived ^{60}Co and ^{55}Fe activities have no concentration limits, and the concentrations of the lower activity radioactive constituents, ^{59}Ni , ^{63}Ni , and ^{94}Nb , that generally control the waste classification, fall below their Class C limits when averaged over the package.

Reactor internal structural components that remain in the pressure vessel during the life of the reactor, particularly BWR and PWR core shrouds and PWR core barrels, present a more difficult classification problem, since they are large components and can be accessed only in situ rather than in the spent fuel pool. They are not amenable to measurement during the reactor life, and considerable uncertainty exists regarding their eventual waste classification, which may be greater than Class C (GTCC). This latter classification would make the Department of Energy (DOE) responsible for their disposal. In fact, some of these components may be marginally either Class C or GTCC, which emphasizes the need for accuracy in their method of classification. Although the bases for the Nuclear Regulatory Commission (NRC) waste classification rules contain considerable conservatism, waste categories are distinct and contain no leeway for interpretation.

1.2 Activity in Reactor Internal Hardware

Core shrouds and core barrels are cylinders surrounding the reactor core. A typical BWR shroud has a volume of metal of about 3.3 m³. A PWR shroud has typically 1.6 m³ of metal, and a core barrel has about 3.3 m³.² Stainless steel (SS) 304 is the structural material of core shrouds and barrels and also of most other reactor internal hardware. This alloy consists of iron (~90%), nickel (8.9 ± 0.5%), and molybdenum (0.4 ± 0.2%). The nickel contains trace amounts (usually "uncontrolled" contaminants) of cobalt (0.10 ± 0.06%) and niobium (0.014 ± 0.011%) (Reference 2). Stainless steel also has a minor constituent of nitrogen that activates to ^{14}C . Cobalt activates to produce ^{60}Co , giving high radiation levels from the component; and the niobium activates to ^{94}Nb ,

which has low permitted concentrations in low-level waste. Resulting concentrations of ^{94}Nb , and the ^{59}Ni and ^{63}Ni , produced from activation of the nominal 9% nickel content in the alloy, usually dominate the waste classification. Activation cross sections for these elements are complex, having some resonance structure for thermal, epithermal, and fast neutron energies. Thus, knowledge of neutron fluences in these energy regions are necessary for accurate estimates of the production of those radionuclides in activation of the hardware. Other activation products produced in irradiated hardware, ^{54}Mn , ^{55}Fe , ^{58}Co , ^{65}Zn , ^{99}Tc , $^{110\text{m}}\text{Ag}$, and ^{125}Sb either have no Class C limits or show concentrations that are well below the limits for these nuclides. Additionally, these other nuclides contribute little to the radiation dose rate from the component, except in freshly discharged components. However, the high variability in the concentrations of all of the small percentage constituents of SS 304 can contribute large inaccuracies in estimating the activities of nuclides important for classification and, thus, in the classification itself. This is particularly true for cobalt whose activations usually serve in characterization as the normalizing activity for other radionuclides.

Components in contact with reactor coolant are coated with surface deposits transported by the coolant (commonly called "crud" deposits). The surface film contains activation and fission products, and transmuted fuel (uranium and transuranic nuclides). The activation is of structural corrosion material suspended in the coolant. The activation transports through the system as resuspended and sloughed ionic and colloidal material. Activities include ^{55}Fe , ^{60}Co , ^{63}Ni and ^{94}Nb radionuclides, but deposited activities are in concentrations only a small fraction of that in the activated component itself. Fission products, e.g., ^{90}Sr , ^{129}I , and ^{137}Cs , and U and TRU, result both from tramp fuel outside the fuel bundles and from leaking fuel rods and are the source of all of these activities deposited on activated hardware. The activity concentrations in the surface deposit are not predictable and depend upon the history of reactor fuel integrity and the coolant water chemistry during component use. The activity concentrations of many of the nuclides in the surface deposit are required for waste classification and may be significant to the overall classification of a component.

1.3 Activation of Decommissioning Hardware

The bulk of reactor hardware requiring disposal during decommissioning is outside the reactor core area during its lifetime and receives a lower neutron fluence than the in-core components such as BWR control rods and in-core instruments, for which there are considerable data on activation levels and waste classification. However, hardware outside the reactor core receives irradiation for a longer period than most of the in-core components. Few data are available on typical neutron flux levels outside the core, particularly for those portions of the shrouds and barrels that extend above and below the core midplane. Difficulties in accurate activation estimates are exacerbated by the nature of the neutron activation cross sections for all the elemental constituents that contain resonance structure in the epithermal and high-energy regions as well as significant thermal neutron capture cross sections. Uncertainty in the neutron energy distribution as well as in the average value of the thermal flux seen by the component constitutes a significant source of uncertainty in estimating the activation level and classification of hardware, when such activation levels are calculated.

1.4 Classification of Activated Hardware

Prior to its disposal, low-level radioactive waste is subject to waste classification according to the requirements of 10 CFR 61. The regulations provide concentration limits for certain radionuclides

and categorize wastes into classifications of A, B, C, or GTCC based upon these limits. Most of the key characterization nuclides are activation or fission products having relatively long half-lives. Many of the activities do not emit measurable gamma radiation in their decay modes, and measurement occurs only through complex radiochemical and radiation counting procedures. NRC positions recognize that direct measurements for these nuclides in every waste component is impractical and allow licensees to estimate concentrations of these "hard-to-detect" radionuclides through scaling from measurable activities. Scaling factors are obtained from analyses of representative samples of each waste stream or component "type" (waste characterization). Waste classification proceeds through measurement of the normalizing (key) nuclide concentration (usually ^{60}Co or ^{137}Cs) and application of the appropriate scaling factors to estimate the other activities in the waste. Estimates of the concentrations of the key nuclide activities are usually through a form of direct measurement, either sampling, direct gamma-ray scanning, or dose-to-curie analyses.³ Accuracy in the classification is usually controlled by the representativeness of the sample used for waste characterization (scaling factor determination), accuracy of the scaling-factor analyses, and accuracy of the key nuclide analyses.

Conventional low-level waste sampling and classification methods are not practically applied to irradiated hardware because of their high levels of activity and the inhomogeneity of the activities within any component. The NRC has also recognized this impracticality and allows the use of two techniques for classification of irradiated hardware, "Activation Analysis" and "Direct Assay." Both techniques use radiation dose rates to determine the differential and average ^{60}Co concentrations in a component, but differ in methods to estimate the concentrations of the other nuclides. Application of the methods thus far has been primarily to components discharged in normal refueling operations. These components include control rod blades (CRBs) and local power range monitors (LPRMs) from BWRs and thimble plugs. The PWR components include tubes and burnable-poison rod assemblies (BPRAs). Most of these components are long and relatively thin items that have very inhomogeneous activity concentrations along the length (some ranging from $>20,000$ R/hr at one end to <100 R/hr at the other).

1.4.1 Activation Analysis

Activation Analyses (AA) are computational approaches that use neutron activation computer programs (usually ORIGEN2,⁴ MICROX,⁵ DTF,⁶ RADCOR,⁷ and ANISN⁸) to determine absolute or relative concentrations of radionuclides. Nearly all applications use a combination of dose-rate measurements and dose-to-curie computations (MICROSHIELD⁹ or PATH¹⁰) to estimate the average ^{60}Co concentrations in a component. Vendors of fuel-pool cleanup services use variations of activation analysis techniques. In all cases, application of the method requires information on the neutron exposure history of each component (i.e., neutron flux and energy distribution and exposure periods) as data input to the computations.

One vendor (General Atomics) uses a procedure for determining the ^{60}Co concentrations in each component using measured dose rates at 6 in. from the center line of each component at 24- to 36-in. increments along its length. They then convert the dose rates to ^{60}Co concentration using the three-dimensional gamma-ray shielding code, PATH.

General Atomics (GA) computes the concentrations of other important activation products, ^3H , ^{14}C , ^{54}Mn , ^{55}Fe , ^{59}Ni , ^{63}Ni , ^{65}Zn , ^{94}Nb , and ^{99}Tc by direct computation using the material composition and neutron exposure data for each component. GA does not correlate these activation products to the ^{60}Co content because the company believes that the variability of the cobalt constituent in stainless steel is too large for accurate correlations. Material composition for the component for these computations is either supplied by the utility or the manufacturer or is taken from structural material handbooks or reference documents.¹¹ The MICROX code computes neutron spectrum-dependent capture cross sections and the DTF code calculates the relative neutron fluences for the three energy groups: thermal, epithermal, and fast.

General Atomics estimates concentrations of corrosion and fission products present as surface contamination by measuring the radiation dose from a cold (unirradiated) metal stored in the fuel pool, characterized by analyzing silt taken from the pool, and assuming a uniform coating of all this material on the surfaces of all components.

Another important vendor, Waste Management Group (WMG), performs similar computations of component activation using RADCOR with the neutron irradiation history and elemental constituents for each component as input to the code. However, WMG uses only the computed relative concentrations of the nuclides in its analyses, using their concentration ratios to ^{60}Co as scaling factors for the other nuclides. They determine ^{60}Co concentrations through dose-rate measurements (at 6-in. separation from the component center line) at 12- to 18-in. intervals along the length of the component and dose-to-curie conversion factors using calculations with MICROSIELD. WMG also characterizes surface deposited activity using a sample of silt collected from the fuel pool and the assumption that the silt deposits on the entire surface of each component to a specific thickness (~ 0.001 in.).

1.4.2 Direct Assay

In 1985, Science Applications International Corporation (SAIC) began developing a technique of direct assay of irradiated hardware and made the initial application to CRBs and LPRMs at the Peach Bottom Power Station. The technique uses a combination of dose-rate measurements and gamma-ray scanning of the component to measure directly the ^{60}Co and other gamma-ray emitting radionuclide concentrations and the factor to convert measured radiation dose to activity concentration (dose-to-curie factor). The technique takes samples of material from the component for radiochemical analyses to obtain measured activity concentrations relative to that of ^{60}Co (scaling factors). Later modifications of the method also acquire samples of surface activities so that their scaling factors are also measured. Values of radiation dose rate along the component are converted to ^{60}Co concentration using the measured dose-to-curie factor. Concentrations of the other nuclides result from application of the measured scaling factors. The direct-assay technique has since been applied to classify irradiated hardware from more than 25 operating power reactors using the combination of sampling, direct gamma-ray measurements, radiochemical analyses, and radiation dose measurements to obtain the individual radionuclide concentrations and the total activity content for irradiated components.

The direct-assay method currently used to classify irradiated hardware has six principal steps: (a) identify the number of structural material batches ("vintages") for each type of component (i.e., identify number of samplings required); (b) identify the scan and sampling locations for each sampling, perform a quantitative gamma-ray scan and a contact radiation dose rate, and obtain a bulk and surface sample at each; (c) measure a contact radiation dose rate profile along the length of every component, taking readings at 6- to 18-in. intervals along the length; (d) radiochemically analyze both the bulk and surface samples for specific nuclides; (e) reduce the data from all the measurements to derive scaling factors for all of the relevant nuclides, scaled to ^{60}Co , and dose-to-curie factors; and (f) classify the component using the dose-rate profile data, dose-to-curie factors, and scaling factors. Application of direct assay provides a direct measurement of the factors required to classify the components at designated locations on specific components and assumes their representativeness for application to the entire component and to other like components.

1.4.3 Comparison of Characterization Approaches

Differences between the activation analysis and direct-assay approaches are significant. Activation analysis relies on representative average constituent composition of the component alloys and on knowledge of the neutron fluence seen by the component in the reactor, whereas direct assay relies on the representativeness of the sampling and scanning locations and on the accuracy of their analyses. Differences, less significant to most component classification, but significant in terms of accuracy, lie in surface-activity analyses. Activation analysis assumes fuel-pool silt as quantitative representation of the activity, whereas direct assay measures surface deposition directly. If surface deposits occur during residence in the pressure vessel, as is believed, fuel-pool silt may not be representative of the material character or concentration. In cases where a component, while in service, was close to a leaking fuel rod, surface deposits of TRU have influenced the component classification (Reference 3).

Table 1 presents a comparison of two activation analyses and the direct-assay techniques in six aspects of classification of an irradiated component: (a) assumptions of structural material composition, (b) determination of dose-to-curie factors, (c) dose-rate partitioning, (d) scaling factors, (e) surface activities, and (f) use of component irradiation history. From the classification standpoint, the largest differences in classification of components results from assumptions regarding elemental constituents and the neutron cross sections, flux values, and energy distributions. A recent comparison of classification results^a for CRBs from two BWRs showed higher concentrations of key nuclides estimated by activation analysis, 30–400 percent higher for some activities, rather than by direct assay. Estimated concentrations of surface activities also differed greatly in the comparisons.

a. M. Hauf, R. Hollis, and J. E. Cline, *Evaluation of Waste Characterization Methodologies for Irradiated Waste Components*, EPRI Research Project 2813-31, Draft Report, January 1992.

Table 1. Comparison of activation analyses and direct-assay techniques.

Classification aspect	Activation analysis		
	General Atomics (GA)	WMG	Direct assay
Material composition	General Atomics relies on typical or average material compositions obtained from supplier data or reference handbooks or publications. These data are used to compute concentrations of all activities but ^{60}Co .	WMG relies on typical or average material compositions obtained from suppliers data or reference handbooks or publications. These data are used to compute activities of all activities relative to the ^{60}Co activity.	Direct assay does not depend upon assumptions of material composition except to assume that the component that was measured was representative of all other like components purchased at the same time.
Dose to curie factor	GA uses PATH, a point-kernel shielding code, to compute the conversion factor from measured dose rate to ^{60}Co concentration.	WMG uses MICROSHIELD, a point-kernel shielding code, to compute the factor to convert dose rate to ^{60}Co concentration.	Direct assay measures the dose-to-curie conversion factor for ^{60}Co in the geometry configuration of the dose-rate profile scan.
Dose rate from ^{60}Co (partitioning)	GA uses the MICROX and DTF codes to compute the relative radiation dose from the gamma-ray emitters and the fraction from ^{60}Co .	WMG uses RADCOR to compute the partitioning of the total dose to that from ^{60}Co .	Direct assay uses the gamma-ray scan and MICROSHIELD to partition the total dose to that from ^{60}Co .
Scaling factors	GA does not use scaling factors but relies on computations to give absolute concentrations of each nuclide.	WMG computes scaling factors with RADCOR using typical material compositions and the irradiation history.	Direct assay measures scaling factors for bulk and surface activities and assumes they are representative of those like components purchased at the same time.
Surface activity	GA characterizes surface activities with fuel-pool silt and assumes uniform coatings equal to that of "cold" items in the pool.	WMG characterizes surface activities from fuel-pool silt and assumes a uniform coating of this material over all the components.	Direct assay measures the surface activity at the sampling locations and assumes that they are proportional to bulk activities.
Irradiation history	GA relies on irradiation history records for computations of absolute concentrations of nuclides.	WMG relies on plant records of irradiation history for computations of scaling factors.	Direct assay uses irradiation histories only for radioactive decay corrections (time since removal).

2. APPLICATION OF DIRECT ASSAY TO IRRADIATED HARDWARE

The direct-assay technique uses a combination of material sampling, gamma-ray scanning, dose-rate measurements, and radiochemical analyses to classify irradiated hardware. Figure 1 shows the four stages of progression in classifying irradiated hardware: (a) planning and component selections, (b) onsite measurements, (c) radiochemical analyses, and (d) data analysis and classification.

2.1 Planning and Component Selection

It is not practical to sample and scan all components in a characterization campaign. The cost and required time would be prohibitive. Thus, a selection process identifies a smaller number of components that are considered as representative of the remainder. The selection process identifies these representative samples.

Although concentrations of the elemental constituents can vary considerably among different batches of an alloy such as SS 304, variations within the same batch should show a much smaller variation. Therefore, direct-assay waste characterization attempts to identify components that were made from the same batch of the base alloy. Characterization planning assumes that like components (e.g., CRBs or LPRMs) that were purchased from a vendor at the same time were fabricated from the same batch of alloy and should have similar elemental compositions. Plant purchase records and core modification records identify those components that were purchased in one order, and these components are designated as being of one batch or "vintage." The first stage in the direct-assay process then randomly selects one specie from each vintage and component type for detailed measurements by sampling and scanning. Surface-activity assumptions require their use in the same fuel cycle. To apply the direct-assay results to components other than those selected as representative, the data must reference to the time of discharge from the core. Thus, component irradiation history is necessary for radioactive decay adjustments.

2.2 Onsite Measurements

Onsite measurements are gamma-ray scans, radiation dose-rate measurements, and sample procurement for each of the identified representative components. Measurements also include radiation dose-rate profiles for all components in the characterization campaign.

2.2.1 Gamma-Ray Scanning

Description of Method. The gamma-ray scanner consists of a calibrated high-purity germanium gamma-ray detector housed in a cart-mounted tungsten-collimated shield. The detector is a portion of a high-resolution spectrometer having a computer-based data acquisition, analysis, and operational management system. The cart sits at the edge of the fuel pool, and the instrumentation remains in a clean, low-radiation-level location. Figure 2 shows a sketch of the operation of the gamma-ray scanner. A 14-ft, 1-in. diameter air collimator, attached to the end of the shield, extends the collimation through the water in the fuel pool to the component itself. The collimator is strengthened during operation in the pool by a support cable to prevent sagging that would affect alignment. Testing and alignment adjustment precede any pool measurements. The component usually suspends from the spent fuel-pool platform bridge for the scanning.

Figure 1. Schematic flow diagram of stages in component waste classification by direct assay.

Figure 2. Sketch of gamma-ray scanner operation for measuring a CRB.

The scanner spectrometer uses a calibration technique developed for use with extended sources and applied to onsite assay work by the International Atomic Energy Agency.¹² The technique extends a point-source detector calibration and spatial response mapping to extended geometries such as the fuel-pool application. Detector calibration and response measurements occur at several distances along the symmetry axis of the detector at many angles with respect to this axis. Data from the calibration measurements are fitted to a calibration model to obtain the values of the parameters describing the detector response. Calculations of detection efficiency for any given geometry then use point-kernel numerical integration computations that include the detector gamma-ray response as a function of energy and spatial location of the source together with self and external shielding considerations. The technique has obtained accuracies as low as 10% for several complex geometries.^{3,13} The geometry of the collimated irradiated component is relatively simple, nearly approximating a point source because of the long collimator length (~14 ft) and small diameter (1 in.), and should yield relatively high accuracy.

Example of Use. Table 2 presents the results from a gamma-ray scan of a typical BWR control rod blade. Results are net concentrations, having the background contribution already removed. Background data were taken in the same geometry but with the collimator not pointed at a component in the pool. The data in the table show the strong presence of ^{60}Co and only weak indications of other nuclides. The summary analysis includes only the ^{65}Zn activity (from trace zinc in the alloy) in addition to ^{60}Co since the other entries had uncertainties (one-sigma net counting statistics) greater than 50%. A MICROSIELD calculation showed that this ratio of ^{65}Zn to ^{60}Co activities would result in 99.5% of the radiation dose coming from the ^{60}Co . Note that scans of "fresh" components at other pools have shown relative contributions for ^{60}Co as low as 45%, the remainder being ^{54}Mn (from trace manganese), ^{58}Co , and ^{65}Zn .

Table 2. Results from a gamma-ray scan of a typical BWR CRB.

Spectrum filename: C:CRB1.SPC Counting time: 1,000 seconds Detection efficiency name: CRB				
Nuclide analysis				
Nuclide	Gamma-ray energy (keV)	Disintegration rate (d/s)	Percent uncertainty	Weighted average concentration ($\mu\text{Ci/cc}$)
^{95}Zr (fission product)	724.2	2.24E+07	85.6	— ^a
	756.7	—	—	—
^{95}Nb (fission product)	765.8	3.72E+06	236.9	— ^a
		—	—	—
^{134}Cs (fission product)	563.3	—	—	—
	569.3	—	—	—
	604.7	—	—	—
	795.8	5.44E+06	193.8	—
	801.8	—	—	—
	1365.1	—	—	— ^a
^{54}Mn	834.8	2.10E+07	46.8	— ^a
^{59}Fe	1099.2	2.69E+07	76.5	—
	1291.6	1.59E+07	68.2	(3.2 $\pm$ 1.1)E+02
^{65}Zn	1115.5	6.58E+07	33.8	— ^a
^{60}Co	1173.2	3.30E+09	0.70	(1.68 $\pm$ 0.16)E+04
	1332.5	3.63E+09	0.67	—

a. Values not computed when uncertainties are larger than 50%.

2.2.2 Radiation Dose-Rate Measurement

Description of Method. The dose-rate measuring instrument is an underwater model of an ionization chamber system that has three probes covering the radiation range from 1 mR/hr to 20,000 R/hr. The chamber is energy compensated to give a response uniform to within 20% for gamma radiation from 100–3,000 keV. The chambers are of plastic-lined aluminum and filled with air at one standard atmospheric pressure. The high-range chamber is 2.5-cm in diameter by 1.5-cm long with an active volume of 7 cm³.

Radiation measurements are made in contact with the component at measured intervals, usually every 15 cm (6 in.), along the length. Since it is often difficult to see clearly through the 10–15 ft of water, contact measurements are assured by recording the highest reading obtained during small adjustments in the positioning of the dose measuring probe. For horizontally suspended objects such as the LPRM hot sections, positioning occurs along a vertical line until a maximum dose rate is seen. For vertically suspended components, such as the control rod blades, the reading is recorded when the probe rests against the component, annunciated by back pressure against the probe and a stability in the dose-rate reading. Profiling begins at one end, and increment positions are noted through graduations in a scale marked on the bridge platform for horizontally scanned objects and through graduations on the probe cable for vertically scanned objects. Dose-rate measurements of small components are measured at several equally spaced locations around the perimeter to obtain an average dose rate for the component. Measurements include dose-rate determinations at the position of the gamma-ray scan.

Example of Use. Figure 3 shows results of the dose-rate profiling of the CRB example (Footnote a). Also shown on the figure are locations of the gamma-ray scans, discussed above, and samplings, discussed below. The data show a profile of a typical blade depleted of boron and discharged at the end of its useful life. Table 3 summarizes the values of the dose rates measured at the five scanning locations of Figure 3. Included in the table are the ⁶⁰Co concentrations measured by the gamma-ray scanner at those locations. The bottom row gives the values of the measured dose-to-curie factors that will be discussed in the analysis section. The last column of the table lists the average of the dose rate and the ⁶⁰Co concentration in the component. The average dose rate for the piece is an average of eight equally spaced readings over the CRB, and the average ⁶⁰Co concentration of the same item is the average dose rate multiplied by the average dose-to-curie factor for that component. Typical classification of irradiated hardware by direct assay measures the dose-to-curie factor at only one location.

2.2.3 Bulk and Surface Material Sampling

Description of Method. Collecting material samples from the components stored in the fuel pools requires the fuel-pool sampler developed for this purpose. The device consists of a sampler head, segmented manipulating rods, connecting hoses, a detachable in-line filter cartridge, and a pneumatic-driven grinder and water pump. The sampler head contains the pneumatic drill, fitted either with a grinding burr or a wire brush as sample removers. The rotating head removes material from the component into the plenum shroud surrounding the head where water carrying the material is drawn through the in-line filter. Discharged filtered water returns into the pool. The sample holder has quick-disconnect fittings to assist in its removal without loss of filter integrity. Radiation monitoring of the filter housing during collection determines when an adequate sample is collected.

Table 3. Contact radiation dose rate and ^{60}Co concentrations measured for a typical control-rod blade.

Control rod dose-rate and gamma-ray scan						
Parameter	Scan position					Average
	No. 1	No. 2	No. 3	No. 4	No. 5	
Measured ^{60}Co concentration [from scan] (Ci/m^3)	16,840	87,950	77,260	40,760	112,100	55,900 ^a
Measured dose rate (R/hr)	6,000	16,000	18,000	20,000	20,000	15,350 ^b
Measured dose-to-curie factor (Ci/m^3)/(R/hr)	2.81	5.50	4.29	2.04	5.61	4.1 ± 1.4^c

a. Product of average dose rate and average dose-to-curie factor.

b. Average of eight equally spaced readings over the CRB.

c. Average and standard deviation around average for measured factors in the five locations.

Figure 4 shows a sketch of the use of the fuel-pool sampler in sampling a control rod blade. Operation requires two individuals: one at the edge of the pool controlling the air supply to the sampler head and the water pump and monitoring the radiation level of the sampler filter, and the other, who is located over the component on the platform bridge, positioning the sampler head in the desired position for sampling. The bulk and surface samples are obtained sequentially, after changing the sampling head between the burr and the brush. Between samplings, the system is flushed through a clean filter to remove any residual material in the sampler or lines.

Dose limits have been set for the bulk sample at a maximum of 100 mr/hr in contact with the sample filter to reduce personnel exposure during transport and analysis, and this level also provides an adequate sample for analysis. Accumulation of a sample can occur quickly when sampling a component having dose rates of several thousand R/hr as shown in Figure 3. Acquiring a surface sample requires several seconds of brushing over an area typically 50–100 cm^2 . An increase of 2–5 mR/hr above background usually denotes acquisition of an adequate sample.

Example of Use. Use of the sampler acquires bulk and surface samples of each component selected for sampling. After acquisition, the sample cartridges are capped on each end, labeled, sealed in plastic bags, analyzed for shipping by the plant laboratory, and loaded into DOT Type 6M shipping containers for transport to the analytical laboratory.

DOSE-RATE PROFILE

Figure 3. Plot of radiation dose-rate data from a typical BWR control rod blade.

Figure 4. Sketch of typical operation collecting a sample of a control rod blade.

2.3 Radiochemical Analyses

Description of Method. Radiochemistry is performed on the samples by a qualified laboratory whose Quality Assurance Program is based on the 18 criteria of 10 CFR 50, Appendix B.¹⁴ The laboratory is periodically audited by numerous utilities and is qualified as a supplier of analytical services.

Bulk samples of irradiated metals receive radiochemical analyses for the nuclides, ^{14}C , ^{59}Ni , ^{63}Ni , ^{94}Nb , ^{99}Tc , and gross alpha activity, and gamma-ray analysis for ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$, ^{125}Sb , and ^{137}Cs . Radiochemical separations are made for ^{137}Cs , if it is not adequately found in the gamma-ray analysis of the entire sample. The activity concentrations are referenced to activity per entire sample, since the sample is contained on a wet filter and an accurate mass is impractical for the milligram-sized sample. Since only activity ratios are used in the analysis for scaling factors, this referencing is adequate.

Radiochemical analyses of the surface samples are for the activities, ^{90}Sr , ^{129}I , ^{238}Pu , ^{239}Pu , ^{241}Pu , ^{241}Am , ^{242}Cm , and ^{244}Cm . Gamma-ray (or radiochemical) analyses specifically look for ^{137}Cs , the reference nuclide in the surface sample. Since the activity of ^{129}I is usually several orders of magnitude below the detection limits for this very small sample (the analyses have only reported lower limits), the expensive radiochemical analysis for this activity is not always done.

Example of Radiochemical Analysis. Table 4 shows the results of radiochemical analyses of a bulk and a surface sample from Position 5 of the control rod blade indicated in Figure 3. The data from the analyses are shown in the second two columns (the section on analysis that follows will discuss the remainder of the data in the table). As indicated, the surface sample was analyzed for fission products and TRU, whereas the bulk sample was analyzed for activation products in the metal and for the reference activities, ^{137}Cs and gross alpha. These latter two analyses of the bulk sample allow normalization of the surface-sample data to the bulk-sample results. Uncertainties listed with the data are the statistical and chemical yield one-sigma uncertainties propagated in computing the final concentrations.

2.4 Data Analysis

2.4.1 Derivation of Scaling Factors

Description of Method. Scaling factors for irradiated components are factors for each radionuclide that are ratios of concentrations of that nuclide to the concentration of ^{60}Co in the bulk material. Their use gives the concentration of the nuclide through multiplication of the scaling factor with the ^{60}Co concentration, obtained through direct measurement or through dose-to-curie conversion. For the bulk activation activities, ^{54}Mn , ^{55}Fe , ^{58}Co , ^{59}Ni , ^{63}Ni , ^{65}Zn , ^{94}Nb , ^{99}Tc , $^{110\text{m}}\text{Ag}$, and ^{125}Sb , scaling factors are the ratios of activity to ^{60}Co measured in the bulk sample. Activity ratios for surface deposits are adjusted for the volume-to-surface ratio of the sample, and, when applied to the irradiated component, the surface-to-volume ratio of the component. Laboratory tests of the sampler grinding apparatus measured an average grinding depth of 0.040 cm (16 mil) during sample collection with the burring tool. This volume-to-surface ratio of $0.04\text{ cm}^3/\text{cm}^2$, when multiplied by the surface-to-volume ratio for the component, gives the dimensionless factor used to adjust measured

Table 4. Measured sample activity concentrations, scaling factors, and average concentrations in a classified control-rod blade.

Nuclide	Activity concentration ($\mu\text{Ci/sample}$)		Scaling Factors (to ^{60}Co)	Average concentration (curies/ m^3)
	Surface sample	Bulk sample		
^{14}C	—	$(3.8 \pm 0.4)\text{E-04}$	$(2.1 \pm 0.3)\text{E-6}$	0.13 ± 0.02
^{54}Mn	0.5 ± 0.2	4.0 ± 0.5	$(2.2 \pm 0.3)\text{E-2}$	$(1.4 \pm 0.2)\text{E+3}$
^{55}Fe	—	120 ± 12	0.67 ± 0.08	$(4.2 \pm 0.6)\text{E+4}$
^{60}Co	16.0 ± 1.6	180 ± 18	—	6.4E+4
^{59}Ni	—	$(5.3 \pm 2.5)\text{E-02}$	$(2.9 \pm 1.5)\text{E-4}$	18 ± 9
^{63}Ni	$(3.8 \pm 1.3)\text{E-05}$	7.6 ± 0.8	$(4.2 \pm 0.5)\text{E-2}$	$(2.6 \pm 0.3)\text{E+3}$
^{90}Sr	—	$<1.3\text{E-04}$	$(2.2 \pm 1.3)\text{E-8}$	$(1.4 \pm 0.8)\text{E-3}$
^{94}Nb	—	$<1.2\text{E-05}$	$<7\text{E-7}$	<0.05
^{99}Tc	Not measured	—	$<7\text{E-8}$	<0.004
^{129}I	$(3.9 \pm 0.4)\text{E-03}$	$(5.6 \pm 0.6)\text{E-03}$	1.2E-11	7.6E-7
^{137}Cs	$(1.3 \pm 0.1)\text{E-04}$	—	$(2.3 \pm 0.4)\text{E-6}$	$(1.4 \pm 0.2)\text{E-1}$
^{238}Pu	$(6.7 \pm 0.7)\text{E-05}$	—	—	—
^{239}Pu	$(1.0 \pm 0.1)\text{E-02}$	—	—	—
^{241}Pu	$(9.8 \pm 1.0)\text{E-05}$	—	$(1.2 \pm 0.2)\text{E-6}$	9.4 ± 1.4^a
^{241}Am	$(1.8 \pm 0.2)\text{E-05}$	—	—	—
^{242}Cm	$(5.7 \pm 0.6)\text{E-05}$	—	$(2.2 \pm 0.5)\text{E-9}$	$(1.7 \pm 0.2)\text{E-2}^a$
^{244}Cm	—	$(1.1 \pm 0.1)\text{E-04}$	—	—
Gross α	—	—	—	—
TRU>5yr	—	—	$(4.2 \pm 0.8)\text{E-8}$	$(3.3 \pm 0.5)\text{E-1}^a$
Act.<5yr	—	—	0.69 ± 0.08	$(4.3 \pm 0.7)\text{E+4}$
Intermediate and auxiliary values				
Average dose rate (R/hr)			15,350	
Dose-to-curie factor			4.1 ± 1.4	
Surface-to-volume factor ^b			0.0728	
Gross $\alpha/^{60}\text{Co}^c$			$(6.1 \pm 0.8)\text{E-7}$	
$^{137}\text{Cs}/^{60}\text{Co}^c$			$(3.1 \pm 0.5)\text{E-5}$	
$^{90}\text{Sr}/^{137}\text{Cs}^d$			$(9.7 \pm 3.5)\text{E-3}$	
$^{129}\text{I}/^{137}\text{Cs}^d$			5.0E-5^e	
$^{241}\text{Pu}/\text{Gross } \alpha^d$			27 ± 4	
$^{242}\text{Cm}/\text{Gross } \alpha^d$			0.049 ± 0.006	
TRU>5yr/ gross α^d			0.95 ± 0.03	
a. Activities expressed in nanocuries per gram (required by 10 CFR part 61).				
b. Includes surface-to-volume ratios of the CRB and of the bulk sample.				
c. From the bulk sample.				
d. From the surface sample.				
e. Generic Scaling Factor from Reference 14.				

ratios of surface activities in the bulk sample, i.e., ^{137}Cs and gross alpha, to obtain scaling factors applicable to the component.

The surface sample is used to determine the ratios of ^{90}Sr and ^{129}I to ^{137}Cs and to determine the ratios of the individual TRU activities to gross alpha. The ratios for the first two are the ratios of activities measured in the sample. The TRU factors result from ratios of the individual measured activities to the sum of all the alpha-emitting activities seen in the sample (i.e., ^{238}Pu , ^{239}Pu , ^{241}Am , ^{242}Cm , and ^{244}Cm). The NRC and DOT regulations treat alpha activities with greater-than-5-year half-life as a single item denoted as TRU (or α) >5-yr. Thus, scaling factor analyses determine the factor for this category as a ratio of the sum of the ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm activities to the gross alpha activity. These values determined from the surface samples are then multiplied by the adjusted ratios of ^{137}Cs or gross alpha, as appropriate, to the ^{60}Co determined from the bulk sample, to obtain final scaling factors.

Example of Use. Table 4 includes scaling factors derived from the radiochemical data. The fourth column contains the resulting factors, and the bottom portion of the table shows some intermediate ratios used in the determinations. The surface-to-volume factor in the table includes the volume-to-surface ratio for the bulk sample, 0.04, and the surface-to-volume ratio of the CRB, 1.82. Intermediate values for the activity ratios listed in the bottom portion are from the results of the appropriate sample analyses, as indicated, and have not been adjusted by a surface-to-volume factor. However, scaling factors for ^{137}Cs , ^{90}Sr , and the TRU activities, listed in the top portion of the table have been adjusted by this factor. Since no detectable activity was found for ^{94}Nb and ^{99}Tc in the analyses, the table lists factors derived only from the detection limits. The ^{129}I activity was not measured and the value for the scaling factor in the table is a "generic" factor.¹⁵ The scaling factor for "act. <5 yr" is the total value for all the beta- and gamma-emitting activities with half-lives less than 5 years. The principal contributor to this category in irradiated hardware is ^{55}Fe ; lesser contributors are ^{54}Mn , ^{58}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$, and ^{125}Sb . Only ^{54}Mn was identified in the sample of Table 4.

2.4.2 Derivation of Dose-to-Curie Factors

Description of Method. The dose-to-curie factor is the ratio of the ^{60}Co concentration measured by the gamma-ray scanner to the contact radiation dose rate measured at that same location on the component (apportioned into that fraction of the dose rate resulting from ^{60}Co). In most irradiated components, the fraction of the dose rate resulting from the Co isotope is greater than 95% and apportioning is not required. The gamma-ray scan gives concentrations of the gamma-ray emitting radionuclides that contribute to the dose rate. Calculation of the ^{60}Co contribution requires computing the relative dose equivalents of the other contributors, using a shielding code such as MICROSHIELD. As an example of such calculations, for a CRB, the relative dose rate contributions from equal concentrations of ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn , and ^{125}Sb are, respectively, 0.34, 0.40, 1.0, 0.23, and 0.17.

Example of Use. Table 3 includes dose-to-curie factors computed for the five scan locations of the CRB (Figure 3). The data from scans, typified by the data in Table 2 that gives the data from one location, showed that 98% of the gamma-ray activity was from ^{60}Co , and, thus, the measured dose rate required no adjustment for the other nuclides. Had this not been the case, the factors given above for the other nuclides would have been used, e.g., a scan of a fresher CRB showed relative

concentrations of ^{54}Mn and ^{58}Co of 35% and 15%, respectively. The apportionment calculation was as follows:

	<u>Fraction</u>	<u>Factor</u>	<u>Product</u>	<u>Dose Fraction</u>
^{54}Mn	0.35	0.34	0.12	$0.12/0.68=18\%$
^{58}Co	0.15	0.40	0.06	$0.06/0.68=9\%$
^{60}Co	<u>0.50</u>	<u>1.00</u>	<u>0.50</u>	<u>$0.50/0.68=74\%$</u>
Total	1.00		0.68	100%

The average value and uncertainty for the dose-to-curie factor, listed in the last column of Table 3, is the average of the five measured values.

2.4.3 Component Classification

Description of Method. Figure 5 is a schematic diagram showing the flow paths of the component measurements and analyses, and their inter-relationships in the classification of an irradiated component by direct assay. The box at the top represents all the items identified for classification and that portion on the right indicates those components selected as representative samples for gamma-ray scanning and material sampling. All components receive dose-rate profiles.

Boxes on the right indicate the bulk and surface sampling of the representative components, the radiochemical analyses for each and the inter-relation to derive scaling factors for each of the relevant nuclides, relative to the dominant ^{60}Co concentration. The gamma-ray scan and the dose-rate measurements at the same locations on the same representative components give the dose-to-curie conversion factor for each type and vintage of component, represented by that selected sample. The classification of each component then completes by applying the appropriate dose-to-curie factor to the average dose rate to obtain the average ^{60}Co concentration in that component, and applying the appropriate scaling factors to compute the average concentrations of the other activities required in the classification. The remainder of the classification process is to compare the resultant concentrations to the Table 1 and Table 2 concentration limits in 10 CFR 61 and to establish the waste classification through individual nuclide concentration ratios to the limits and through summing the individual fractions for the nuclides in each table.

Example of Use. The last column in Table 4 lists the average concentrations of the nuclides in the control rod-blade example required for 10 CFR 61 classification. These values resulted from the process shown in Figure 5, using the average dose-rate and dose-to-curie factor for the component, shown in the list of auxiliary values, and the scaling factors in Table 4. Table 5 reproduces these concentrations and compares them to the Class C limits in 10 CFR 61. As is usual with stainless-steel components, the two nickel isotopes control the classification. For this component, no nuclide has concentrations above the Class C limit and the sum of the fractions for both Table 1 and Table 2 nuclides is less than half the limit, showing the component to be a Class C waste.

Figure 5. Schematic flow diagram of direct-assay classification of irradiated hardware.

Table 5. Classification of example control-rod blade.

Control-rod blade waste classification				
Nuclide	Class "C" limits (Ci/m ³)	Average concentration (Ci/m ³)	Fraction of class "C" limit	
			"Table 1 Nuclides"	"Table 2 Nuclides"
¹⁴ C	80	0.13	0.02	
⁶⁰ Co	No limit	64,000		No class
⁵⁹ Ni	220	18	0.08	"C" limit
⁶³ Ni	7,000	2,600		
⁹⁰ Sr	7,000	0.0014		0.37
⁹⁴ Nb	0.2	<0.05	<0.25	2.0E-07
⁹⁹ Tc	3	<0.004	<0.0013	
¹²⁹ I	0.08	7.6E-7	9.5E-06	
¹³⁷ Cs	4,600	0.14		
²⁴¹ Pu	3,500 ^a	9.4 ^a	0.0027	3.0E-05
²⁴² Cm	20,000 ^a	0.017 ^a	1.7E-7	
TRU > 5 yr	100 ^a	0.33 ^a	0.0033	
Nucl. < 5 yr (includes ⁵⁴ Mn, ⁵⁵ Fe, ⁶⁵ Zn, etc.)	No limit	43,000		No class "C" limit
Sum of fractions			<0.36	0.37
a. Activities in nanocuries per gram.				

3. ASSESSMENT OF METHOD ASSUMPTIONS AND UNCERTAINTIES

This section assesses the accuracy of the direct-assay technique of waste classification when applied to irradiated hardware discharged in normal operation and to larger components that would be removed in decommissioning, particularly core barrels and shrouds. The assessment considers the analytical accuracies and their effects on the accuracy of estimating radionuclide concentrations in the components and, therefore, the accuracy of its classification. Some uncertainties in the results of individual stages in direct assay arise from true random analytical variances (e.g., random radioactivity counting statistics), symmetrical about the reported value, while others are bias errors, either one-sided or symmetric about the measured value (e.g., alignment errors). Each measurement in the process of direct assay is independent, and an uncertainty in the results from one of the steps has no relation to an uncertainty in the results from another step. The technique for combining uncertainties in quantities computed from several measured values assumes this independence and uses "standard" uncertainty techniques with no co-variant terms. Appendix A presents the propagation methods used in the discussion of direct-assay uncertainty estimates. The discussion also includes assessments of the dependence of classification accuracy on the validity of the assumptions made in the direct-assay process. These assessments primarily arise from the results of experimental measurements and tests of the assumptions.

3.1 Determination of Dose-to-Curie Factor

3.1.1 Analytical Accuracy

Gamma-Ray Scanning. Spectrometer calibration uses a technique developed over the past several years that allows the extension of an accurate "point-source" calibration, using National Institute of Standards and Technology (NIST) traceable sources, to objects of extended geometry. The method is an alternative to the use of phantom standards that attempt to duplicate the geometry of the object with a uniform, homogeneous, and known concentration of standard source material. Producing uniform, homogeneous, and known concentrations in solid extended-geometry sources has always been difficult both to make and to verify. For activated hardware, where the counting geometry is a 15-ft collimated beam, the required high activity in the phantom adds another extreme level of impracticality.

The calibration procedure uses a semi-empirical model of detector response (Reference 12) that treats the detector as a point along its axis of cylindrical symmetry whose position becomes a function of energy, so that sources counted along the symmetry axis give a response proportional to $1/D^2$, D being the distance along the axis from the detector front surface. Detection response to point sources located off axis is a well-behaved function of the angle with respect to the axis and of gamma-ray energy. The calibration considers each detector and collimator configuration as an entity, requiring its own complete calibration. The procedure is to determine, empirically, the parameters of the model through a mapping of the detector response to a point-source standard for any location in the half-infinite space in front of the detector.

Detection efficiencies for the defined counting geometry of an extended source result from an integration of the response function over the volume of the source. Calculations are similar to shielding-code point-kernel integrations, taking into account the self-attenuation in the source and

attenuation in containers, absorbers, and air columns between the source and the detector. Calculations assume that the distribution of activity within the scanned portion of the component is known, either uniform or as a known function within the volume element. For the data base of components discharged in normal operation, the assumption of uniform activation has a basis of isotropic neutron irradiation fluence and should be accurate to less than 1-2%. For core barrels and shrouds, where the neutron fluence varies through the thicker material, there would be a variation in activity along the radial (thickness) dimension of the component. Uncertainties in the scan analyses resulting from uncertainties in knowledge of this variation should not contribute significantly to the overall uncertainty of the scan.

Calibrations using the model were used extensively at TMI-2 during the recovery efforts, at the International Atomic Energy Agency (IAEA, Reference 13), and in power plants (Reference 3) and have shown accuracies better than $\pm 20\%$ for large tanks of homogeneous liquids or resins, when compared with sampling results. Most comparisons are difficult (Reference 3), since accurate knowledge of the actual source contents is not usually precise. For application to a source located at 15 ft from the detector and collimated to a 1-in. diameter, a $1/D^2$ on-axis response is a good approximation, and accuracies are estimated at $\pm 5\%$. Counting statistics add to this uncertainty but are negligible for the usually strong ^{60}Co gamma rays.

Determination of the relative contributions from all the gamma-ray emitters in the component, for apportionment of a measured dose rate to that from ^{60}Co , is easily and accurately done with the collimated germanium detector spectrometer. Gamma rays from the principal contributors to the dose, ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$, and ^{125}Sb are easily resolved; and the analyses of relative abundances are usually accurate to $\pm 5\%$.

Uncertainties in ^{60}Co concentrations from gamma-ray scanning result principally from uncertainties in the distance between the end of the collimator and the component being scanned and on the alignment of the 15-ft collimator. One centimeter of water will attenuate ^{60}Co gamma rays by approximately 6%. Since scans are made at contact with the component, observable from above the pool, an error in positioning greater than one centimeter is unlikely, giving a possible underestimation bias of as much as -6% to the measurement.

Misalignment of the collimator probably contributes the largest uncertainty in the measurement, again as an underestimation bias, since misalignment will always yield a concentration smaller than actual. Three forms of misalignment can occur: (a) scanning the component surface at an unknown angle, (b) not having the object fully enclosed within the 1-in. diameter field of view, and (c) an arc in the 15-ft long collimator itself. For components larger than 1-in. diameter, the first two forms are not serious contributors. However, for in-core instrument tubes such as LPRMs that have a diameter of this magnitude, alignment is a significant source of uncertainty. This will be discussed further in the summary on uncertainties in dose-to-curie factor determinations. Bowing in the collimator can also be a serious problem. Hence, early modifications in the collimator went to thick-walled box channel and additional support struts. The present method of collimator alignment using support on each end and maximizing the counting rate reduces the accuracy uncertainty from this source to about -10%. Overall accuracy of gamma-ray scanning, including the asymmetrical bias, is estimated, using the method in Appendix A, at about +5%, -13%. The geometry of an in-situ scan of a large cylindrical core barrel or shroud should not affect this estimate of accuracy. If scans could be made

of both sides of a core barrel at a given location, accuracies in assumptions of the distribution of activity in the component thickness could be verified.

Dose-Rate Measurements. The vendor of the Model RO-7 dose-rate monitor, Eberline Instrument Division, calibrates the dose-rate instrument using a set of NIST-certified ^{137}Cs sources. Each chamber is calibrated at five dose-rate values in an air media at standard temperature and pressure (STP). Quoted accuracies are better than $\pm 5\%$ over 0.16–320 R/hr. The manufacturer believes the accuracy in the high range to 20,000 R/hr is also better than $\pm 5\%$. Recalibration occurs annually. The underwater housing used in measurements in the fuel pool attenuates the readings about 5%, reflected in the calibration factor for underwater use. However, application of the instrument to dose-rate measurements and profiles on irradiated hardware uses only relative measurements, since readings are referenced to a measured ^{60}Co concentration, and all measurements use the same instrument. Therefore, only reproducibility of measurements is the limiting factor in dose-rate measurement uncertainties. Reproducibility is estimated, from repeated measurements, at $\pm 5\%$. For thick-walled components, such as core barrels, radiation self-attenuation is important and must either be accounted for or assumed constant throughout the component. Uncertainties in this assumption should have only a small effect on the relative dose-rate accuracy.

Positioning accuracy is a more important aspect of the dose-rate measurements. Measurement geometries when the probe (or a jig positioning the probe at a fixed distance) is in contact are the easiest to reproduce. Applications of direct assay have used contact dose-rate measurements where contact is ensured by the back resistance of the piece. As was mentioned above, one centimeter of water attenuates the radiation from ^{60}Co by about 6%. Because of the awkward geometry of some scans, positioning can result in measurement errors of about 12%. Uncertainties in results caused by errors in positioning are one-directional biases in the dose-to-curie factors in the opposite direction from those of the gamma-scan positioning errors and approximately of the same magnitude, except for small-diameter pieces, giving an asymmetric overall uncertainty for relative dose-rate measurements of about +13%, -5%.

Overall Accuracy in Dose-to-Curie Factors. The asymmetric uncertainties in the gamma scanning and in the dose-rate measurements are opposite and approximately equal. Propagation of the estimated uncertainties from the two measurements used to compute the dose-to-curie factor give an estimated bi-directional uncertainty of $\pm 14\%$.

An indication of the true accuracy of dose-to-curie factor determinations may result from examination of the reproducibility in values determined for a given type of component. Since 1985, direct assay has measured dose-to-curie factors in 17 BWR control rod blades and 22 BWR LPRMs. These two component types, both of standard and uniform design, provide the best available examples for evaluations of reproducibility. A CRB has a cross section in the shape of a cruciform with the sides of each quadrant about 5 in. wide and 1/3-in. thick. Scan and dose-rate measurements are of the blade area near the center of the cross. An LPRM is a thin-wall hollow tube 0.75 in. in diameter. Both components are primarily of SS 304. Table 6 presents the average value and standard deviation in measurements of dose-to-curie factors for those components. The average and standard deviation calculations exclude one measurement of a CRB factor and five measurements of LPRM factors, rejected as extreme outliers. The rejected CRB value resulted from a gamma-ray scan result identified as having a spectrometer failure. The rejected LPRM values are greater than a factor of three low and are believed to have resulted from a severe misalignment of the gamma-ray scan collimator. The

standard deviations shown in the table indicate reproducibilities of $\pm 28\%$ for dose-to-curie factors from CRBs and $\pm 24\%$ from LPRMs (with outlier rejection). These uncertainties are approximately double the estimated uncertainties and indicate either an underestimated uncertainty in a measurement or an unidentified source of uncertainty. The source is currently unknown.

3.1.2 Validity of Assumptions

Application of a measured dose-to-curie factor to the average dose rate for an entire component assumes that the factor is a constant over the component, i.e., that the dose rate per unit of concentration of ^{60}Co is constant and that the proportion of the dose resulting from ^{60}Co is also constant. The first assumption is that the cross-sectional dimensions of the component are constant, and the second is that the elemental constituents and the relative activation of these elements are constant over the length.

Validity of the first assumption needs to be examined for each component, but has been demonstrated, as discussed below, within the measurement capability for common components such as CRBs and LPRMs and should be valid for core barrels and shrouds. The second assumption is valid by definition for any component in which the ^{60}Co contributes the bulk ($>90\%$) of the dose, which is the case for most SS 304 hardware that has cooled for times greater than a year. The uniformity of the alloy composition in a single-alloy component depends upon the quality control of the fabricators and their suppliers but is assumed by most parties to be uniform to within a small percentage. Relative activation of the different elements contributing to the dose relies on the dependence of the activation cross sections on neutron energy and the variation in the relative neutron energies along the length of the component. All important activations are by single neutron capture in common materials and none have a large epithermal neutron resonance structure in their cross sections. Thus, there is no reason to suspect significant variations in relative activation along the component length.

Components important in decommissioning have long irradiation histories in which the ^{60}Co activity has reached a near equilibrium concentration that will completely dominate that from shorter-lived components. Longer-lived gamma-ray emitting activities, ^{94}Nb or fission products in the crud deposits, should still have concentrations well below that of ^{60}Co , even considering the variation in neutron flux and energy over the large component volume. Thus, ^{60}Co should contribute over 95%

Table 6. Reproducibility of dose-to-curie factor measurements in irradiated components.

Component	Number of measurements	Average value and standard deviation
Control-rod blade	16 ^a	2.9 ± 0.8 (28%)
Local power range monitor	17 ^b	56 ± 13 (24%)

a. One additional measurement rejected as extreme outlier.

b. Five additional measurements rejected as extreme outliers.

of the radiation dose rate from these components and the dose-to-curie factor should be constant over the volume of these components.

Measurements were made to assess the uniformity assumptions. Table 7 shows relative gamma-ray intensities of ^{54}Mn , ^{65}Zn , and ^{60}Co at five positions along the length of a CRB (Reference 11). The data are of the CRB whose dose-rate profile and scanning and sampling positions were shown in Figure 3.

An external variance [root mean square (RMS) uncertainty] significantly larger than the internal variance (propagated uncertainties of nuclide analyses) would suggest the presence of a systematic uncertainty, unaccounted for in the measurement uncertainties. In the values in Table 7, there is no indication of a significant difference in the two variances and no statistically significant indication of variation in relative activation along the length of the rod.

The assumption of applicability of a measured dose-to-curie factor to the same component type as that measured depends upon the representativeness of the alloy in the sample for the entire batch and on knowledge of the decay history of each component in the pool. The assumption in selecting a representative sample from a collection of components having the same "vintage" is that all components of the same type and purchased at the same time were fabricated from the same batch of alloy material. The accuracy of this assumption is not known and is difficult to ascertain. Decay knowledge is necessary to correct the dose-to-curie factor for the relative contribution of ^{60}Co to the measured dose if the adjustment factor is important. Accuracy is directly related to the size of the adjustment and the radioactive decay.

Table 7. Relative concentrations of gamma-ray-emitting nuclides along a control rod blade length.

	Position along control-rod blade					Average
	18 in.	48 in.	84 in.	108 in.	132 in.	
	7,000 R/hr	12,500 R/hr	18,500 R/hr	20,000 R/hr	21,000 R/hr	
Nuclide	Relative concentration at each location					
^{54}Mn	0.006 ± 0.003	0.015 ± 0.002	0.012 ± 0.001	0.013 ± 0.003	0.016 ± 0.002	0.012 $\pm$ 0.002 (0.004)
^{65}Zn	0.019 ± 0.006	Not identified	0.013 ± 0.004	0.020 ± 0.005	0.015 ± 0.004	0.017 $\pm$ 0.005 (0.003)
^{60}Co	1.0	1.0	1.0	1.0	1.0	

(The first value of the uncertainty is the internal uncertainty, i.e., from propagation of measurement uncertainties. The second value is the external uncertainty, i.e., RMS about the average.)

3.2 Determination of Scaling Factors

3.2.1 Analytical Accuracy

Sampling. Sampling of bulk activity gives an accurate representation of the material at the position of sampling unless there is cross contamination from a previous sample. Between samplings of bulk material, a flushing occurs of the sampler head, lines, and filter housings. Furthermore, since a surface sample is generally acquired between bulk material samplings, the sampling head is removed and cleaned before re-use. Inaccuracies from cross contamination between surface sample and bulk material sample activities are also unlikely and would cause minimal problems, since sampling is in the same general area on the piece and analyses of the two samples are for different radionuclides. Hence, inaccuracies in direct-assay classification from sampling should be negligible.

Radiochemical Analyses. Reported uncertainties in scaling factors determined through radiochemical analyses result from propagation of uncertainties for individual measurements of nuclide concentrations. The uncertainties in the individual measurements include statistics of radioactivity counting, background determinations, chemical yield determinations, and calibration uncertainties. The magnitude of any uncertainty depends upon the relative concentration of the nuclide in the sample, the reliability of the chemical separation (particularly the chemical yield), and the type of radioactive decay emission. For easily analyzed gamma-ray emitting nuclides with relatively high concentration (e.g., ^{60}Co), the uncertainty is dominated by spectrometer calibration uncertainties and has a minimum uncertainty of $\pm 10\%$. For less abundant activities, such as ^{137}Cs , uncertainties are usually controlled by chemical yield and counting statistics. Uncertainties of $\pm 50\%$ are not uncommon for some of these low-concentration nuclides.

Radioactive deposits on component surfaces are highly variable and depend upon the condition of the reactor fuel while the component was in service. If the ^{90}Sr and TRU activities of these deposits are sufficient for importance in classification, accuracies in their analyses are typically about $\pm 10\%$. A larger source of uncertainty is the surface-to-volume factor applied to surface activities. This has two parts: (a) the surface-to-volume ratio of the component, and (b) the volume-to-surface ratio of the bulk sample taken from the component (i.e., the average depth of the grinding). That portion regarding the surface-to-volume ratio of the component depends upon knowledge of the component details and is usually accurate to within $\pm 1\text{--}2\%$. However, the portion for the bulk sample is a function of each sampling. The value currently used is based upon laboratory measurements that showed an average grinding depth of about 0.4 mm in a typical grinding operation. The accuracy of this value applied to all bulk samplings is estimated at $\pm 30\%$.

Particular difficulties occur in factors for the fission product, ^{129}I , and the activation products, ^{59}Ni , ^{94}Nb , and ^{99}Tc . Except for ^{59}Ni , these activities are present only in small concentrations and are seldom identified in radiochemical analyses; only minimum detectable activities are reported and those are relatively high because of the low analytical sensitivities and the small sample sizes. The ^{59}Ni activity cannot be separated from the more dominant ^{63}Ni , and since the nuclide emits only a difficult-to-analyze x-ray, analytical uncertainties on its measurement are often high. Since classification requires values for all five activities, any reported minimum detection limits, translated into upper limits on scaling factors, can give unrealistically high activity concentration estimates that may be close to the Class C limits for these nuclides. As applied in cases where real values for the activities are

not observed, direct-assay characterization reports scaling factors that result from the smaller of: (a) the minimum detection limit in the sample, (b) a scaling factor from an historical data base of positive results from other similar components, and, (c) in the case of ^{129}I where positive values have never been measured in hardware samples, an historical data base from samples of other primary waste streams, scaled to ^{137}Cs . Use of the alternate scaling factors is believed to yield more realistic results. In some few instances, the estimated concentration of ^{94}Nb exceeds the Class C limit, even though the activity is not found in the sample.

Overall Accuracy in Scaling Factors. A statistical assessment of analytical scaling factor determinations is difficult. Actual values depend not only on the analytical accuracy but also on the alloy constituents and, to a lesser degree, on the activating neutron energy distribution. Table 4 contains the uncertainties on the listing of scaling factors for the example CRB of this document. These are estimates of the total analytic uncertainty, propagated from uncertainties in measured individual nuclide values. Uncertainty estimates of scaling factors for the radionuclides controlling waste classification range from $\pm 10\%$ to $\pm 50\%$ (excluding those from minimum-detection limits).

However, only measurements of decay-corrected activity ratios known to be constant would indicate true analytical accuracies for comparison to the estimated values. The ratio of the two nickel radioactive isotopes fit that criteria, but the distribution of measured values of the ^{59}Ni to ^{63}Ni ratio will provide an indication of actual analytical accuracy only for those two nuclides and not for the elusive surface-to-volume factor or for sampling errors. The only other indication of scaling factor accuracy is through reported uncertainties and the quality control in the analytical laboratory (e.g., duplicate and split sample analyses).

3.2.2 Validity of Assumptions Involving Scaling Factors

Application of scaling factors of bulk activities measured at a single location to an entire component makes assumptions of representativeness similar to those for application of the dose-to-curie factors. In particular, a required assumption is that the constituents of the alloy and their relative activation are constant over the volume of the component. Through the same arguments discussed earlier, there is no reason to expect variations in either the constituents or relative activation over the volume of a single component.

Assumptions regarding surface activities are: (a) fission products and TRU activities are only in the surface deposits, (b) these deposits occurred while the component was in service within the reactor vessel, (c) relative concentrations of the surface activities are constant over the volume of the component, and (d) surface concentrations are proportional to the concentration of activation products in the component bulk at that location. Nonfuel-bearing hardware contains no special nuclear material and would produce neither fission products nor TRU. Therefore, these activities should appear only in the surface deposits. Deposition of surface activities while the component is stored in the fuel pool should consist only of loose material that settles on horizontal surfaces and is readily removed during manipulation prior to sampling. The surface material is more likely "crud" that deposited during in-core service and would be characteristic of the activities in the reactor coolant during that time. The nuclide distributions within this crud deposit should be relatively constant. The last assumption postulates that deposition rates for the crud are greater in the higher-temperature regions of the more active reactor core areas, a thesis supported by measurements of deposits on fuel assemblies.

Measurements were made in attempts to verify or disprove the assumption of proportionality. Table 8 presents the scaling factors from five sets of samplings along the length of the example control rod blade. The total activation at the five locations was substantially different, as indicated by the ^{60}Co concentrations at each location. The last column gives the weighted average of the individual measured factors and their internal and external uncertainties. That the external variances for the activation products, ^{14}C , ^{55}Fe , ^{59}Ni , and ^{63}Ni , are significantly larger than the internal variances suggests additional sources of uncertainties in the analytical process beyond those accounted for in the propagation of measurement uncertainties. Their source is also unknown.

The results of the analyses show several other interesting features:

1. *Variation in Alloy Constituents along the Length of the CRB.* There are no statistically observable trends in the scaling factors of bulk activation products at four of the five positions along the length of the blade (all except the sampling at the location with the lowest concentration). The variation in the carbon, iron, and nickel activities relative to cobalt are random with respect to position and are not statistically significant. However, results for the other sampling, while lying only slightly outside the variance of the other four measurements, indicate the possibility of a lower nickel activity content, relative to iron, than in the other samples. This anomalous ratio indicates either a variation in the nickel content, the relative activation, or, more likely, an anomalous ^{63}Ni sample analysis in this lower-activity sample.
2. *Variations of Relative Surface Activities along the Length of the CRB.* Scaling factors show no statistically observable variation in the surface activities, ^{90}Sr , ^{137}Cs , and TRU, along the CRB length. This suggests that the assumption of surface activities being proportional to bulk activities is at least approximately correct and that surface deposition is proportional to the axial neutron fluence encountered by each component in the active core region.

This constancy of scaling factors of surface deposits is partially validated for in-core components, but may not be valid for those away from the core, particularly if there are decontamination activities. Sampling will be necessary on core barrels and shrouds to establish correlations of surface activity on those pieces.

The assumption of applicability of scaling factors measured for one component to like components of the same "vintage" batch of structural material depends, as with application of dose-to-curie factors, upon the representativeness of the sample for the alloys of the other components. Representativeness assumes that all components of that type, purchased at the same time from a single vendor, were fabricated from the same batch of alloy. The accuracy of this assumption has not been established, and its use depends upon adequate records and identifications for each component. Accuracy in classification of these other components depends directly on the validity of this assumption.

Table 8. Comparison of scaling factors measured at five positions along a control rod blade.

Nuclide	Scaling factor				
	Position No. 1 $^{60}\text{Co}(\text{Ci}/\text{m}^3)$ 16,800	Position No. 2 $^{60}\text{Co}(\text{Ci}/\text{m}^3)$ 87,950	Position No. 3 $^{60}\text{Co}(\text{Ci}/\text{m}^3)$ 77,260	Position No. 4 $^{60}\text{Co}(\text{Ci}/\text{m}^3)$ 40,760	Position No. 5 $^{60}\text{Co}(\text{Ci}/\text{m}^3)$ 112,100 Average
^{14}C	1.1E-6± 14%	3.0E-6± 14%	8.7E-7± 14%	1.5E-6± 14%	2.0E-6± 14% 1.7E-6± 6.3% ^a [45%] ^b
^{55}Fe	0.72± 14%	1.00± 14%	0.91± 14%	0.56± 14%	0.77± 6.3% [21%]
^{59}Ni	5.6E-4± 41%	5.2E-4± 17%	—	2.3E-4± 40%	4.7E-4± 19% [30%]
^{63}Ni	0.022± 14%	0.043± 14%	0.052± 14%	0.032± 14%	0.039± 6.3% [26%]
^{90}Sr	4.8E-9± 30%	—	3.2E-9± 62%	—	4.5E-9± 34% [18%]
^{137}Cs	4.8E-7± 14%	1.4E-7± 14%	6.9E-7± 14%	—	3.9E-7± 7.3% [55%]
	8.5E-8± 30%	—	—	—	1.1E-7± 17% [17%]
^{241}Pu	3.1E-11± 30%	—	—	—	17% [17%]
	2.4E-9± 16%	—	—	—	8.8E-10± 17% [65%]
^{242}Cm	—	—	—	—	3.8E-9± 17% [23%]
TRU>5yr	—	—	—	—	—

a. Internal uncertainty, from propagation of measurement uncertainties into the average.

b. External uncertainty, standard deviation about the average.

3.3 Summary of Accuracy Assessment

Figure 6 repeats the schematic flow diagram of Figure 5 and is annotated to show estimates of analytical uncertainties for each step in the direct-assay process. The values for dose-to-curie factor and scaling-factor analyses are standard deviations on repeated values that include uncertainties in the assumption of representativeness of these values over an entire component. The analyses indicate that the accuracy for analysis of the nuclides in waste classification is $\pm 20\text{--}52\%$, using internal uncertainties, and $\pm 33\text{--}56\%$, using external uncertainties. Accuracy of nuclides important to classification, and, therefore, the "classification accuracy" itself is estimated at about $\pm 35\%$.

There is no reasonable way to quantify the validity in application of the scaling factors to other components or to place true limits on their classification uncertainties. However, this general uncertainty is common to all methods of irradiated hardware waste classification.

Accuracies of applying direct-assay techniques to core barrels and shrouds should be comparable to accuracies of applying direct-assay techniques to in-core components. Surface activities, although having larger uncertainties in their estimation, should contribute a minor role in the waste classification. The unknown validity of assumptions regarding uniformity of the alloy over large samplings is of concern for the question of uniformity over the entire bulk of these large components, and some verification is required.

$\sim \pm(20-52)\%$, $\sim [33-56]\%$ on controlling nuclides and sum of fractions

Figure 6. Schematic flow diagram of direct-assay classification of irradiated hardware, showing uncertainties for each process step and for the final classification.

4. POTENTIAL FOR REDUCING UNCERTAINTIES

The estimated accuracy of about $\pm 35\%$ in a waste classification of an irradiated component by direct assay is within the desired NRC goals of a factor of 10.¹⁶ Extension of the measured scaling factors to other components of the same type is less certain but probably still falls within the NRC guidelines. However, a classification that yields either ratios of individual nuclides to the Class C limits or sum-of-the-fractions greater than unity abruptly classifies a waste as GTCC waste with no "gray area" of definition. Since many irradiated components fall on the borderline between Class C and GTCC wastes, accuracy in classification becomes important to reduce the conservatism built into most estimations. Hence, it is beneficial to investigate the potential to enhance the accuracy of direct-assay characterization and its use in determining the classification of reactor internal hardware generated from decommissioning activities.

The discussion of activities that have potential for improving the accuracy of waste classification by direct assay is in three distinct areas: (a) searches for existing and more accurate data that could improve the accuracy of classification estimates, (b) measurements that would supply data necessary for more accurate analyses, and (c) technique developments that could improve the accuracy of the measurement process. Many of the items discussed would also improve the accuracy of classifications from activation-analysis. Some of the proposed enhancements, such as concentration averaging, are not directly applicable to improving the accuracy of a specific characterization technique, but apply more to applications within the characterization process that would affect whether components would be Class C or GTCC wastes.

4.1 Information Collection

4.1.1 Improved Characterization of Cobalt, Nickel, and Niobium in Alloys

Purpose: Concentrations of the nuclides ⁵⁹Ni, ⁶³Ni, and ⁹⁴Nb control waste classification both individually and as principal contributors to the "sum-of-the fractions" for 10 CFR 61 Table 1 and Table 2 nuclides. Since direct assay depends upon measuring the ⁶⁰Co concentration and using scaling factors for the other nuclides related to the cobalt activity, accurate cobalt concentrations are also important. Hence, improved knowledge about variances in cobalt, nickel, and niobium concentrations in alloys used in reactor internals, SS 304 and 316, zircaloy-4, and Inconel 718 would help bound the activation levels and waste classification of those materials. There is particular interest in the character of steels used in BWR core shrouds and in PWR core shrouds and barrels. For example, the measured range of nickel in the SS 304 of these components has been reported as $(8.9 \pm 0.5)\%$, of cobalt as $(0.10 \pm 0.06)\%$, and of niobium as $(0.14 \pm 0.11)\%$ (Reference 2). Better knowledge of the actual compositions of particular components could reduce the uncertainties in applying measured scaling factors for these nuclides to other components not directly measured.

- Procedure:** Use plant and nuclear supplier records to identify the material fabricator. Contact the steel supplier for assay records of alloys used in fabrication of reactor internal components. Identify other components or material supplies that would provide unirradiated samples of that alloy for instrumental assay for nickel, cobalt, and niobium.
- Practicality:** The process is feasible if records exist for desired components adequate to identify the alloy manufacturer and for the particular batch by the manufacturer. Many of these records may be available from utility or reactor steam system suppliers, studies of pressure-vessel embrittlement, or other sources.
- Cost:** The program cost would be the labor of the investigator to track the records through the utility, reactor supplier, and fabricator. Feasibility of ferreting out these data and their usefulness should be established quickly (100–200 hours) to assess the desire to continue the study.

4.1.2 Improved Data Base for Neutron Flux at Decommissioning Hardware

- Purpose:** Neutron fluxes at all energies, thermal, epithermal and fast, are uncertain in regions beyond the reactor core in both BWR and PWR power plants (Reference 2). Few measurements of these levels are reported in the open literature, and computations of these levels are used in many estimates of radioactivity induced in decommissioning hardware. Differences of as much as a factor of 100 are reported between some measured and calculated flux values. The differences have substantive consequences in estimating the waste classification of these components, which may be GTCC. Additional unpublished data may exist from utilities and nuclear steam suppliers that would be useful in establishing these levels.
- Procedure:** Perform additional searches for industry data of measured or estimated neutron fluences in the regions of core shrouds and barrels for both BWR and PWR plants. This would require extensive use of contacts with the utilities and with nuclear steam supply system vendors. Evaluate the acquired data and prepare the best available estimates of these neutron fluence values for thermal, epithermal, and fast energy ranges.
- Practicality:** The study is feasible using utility and industry contacts and library reference literature searches.
- Cost:** The cost for this study is estimated at 150–200 person hours plus a literature search cost estimated at \$1,000.

4.1.3 Improved Data Base of Niobium and Nickel Cross Sections

- Purpose:** Activation cross sections for the three important activities, ^{59}Ni , ^{63}Ni , and ^{94}Nb , are not well known¹⁷ either in the thermal or epithermal regions. No data are reported for neutron activation cross sections of the individual isotopes of nickel where data is given only for capture by the natural element having five stable isotopes. Estimates of individual cross sections are inferred by the calculational routines with considerable

uncertainty. Even for niobium, although having a single stable isotope, the thermal and epithermal capture cross sections are not reported; and scatter in reported values of total cross sections gives considerable uncertainty in the accuracy of the values used in the codes.

- Procedure:** Evaluate available cross section data for thermal, epithermal, and fast neutron capture for nickel and niobium isotopes to produce ^{59}Ni , ^{63}Ni , and ^{94}Nb . The search would be for reactor activation cross section measurements and for activations measurements that used selective-energy filters to determine absorption in the different energy regions. The search would utilize reference data bases and the DOE National Data Centers (which include foreign data) for basic and evaluated data. The study would prepare an evaluation and summary of the retrieved data.
- Practicality:** This data search is feasible using reference data base searches and contacts with nuclear data centers. The INEL has personnel involved in this area who could be of significant help in this effort.
- Cost:** The cost for this data search is estimated at 100–150 person hours, plus a reference material search cost estimated at \$1,000.

4.1.4 Consequences of Concentration Averaging Requirements

- Purpose:** Regulators recognize that a significant degree of inhomogeneity exists in nuclide concentrations within low-level waste. Due to this recognition, they generally allow averaging of nuclide concentrations over the volume of waste in a package in determining classification. Recent positions taken by the NRC^b propose more restrictive requirements. The proposed Branch Technical Position on concentration averaging is currently out for comment. If accepted by waste disposal sites, a larger fraction of irradiated components would become GTCC waste. Consequences of different averaging scenarios should be examined.
- Procedure:** Investigate the consequences to GTCC waste volumes of various concentration averaging scenarios for irradiated hardware. Packaging and storage volume requirements would be included in the investigation as well as waste volumes. The study should consider the disposal-site intrusion scenarios that have formed the basis of averaging considerations.
- Practicality:** Electric Power Research Institute (EPRI) has a study of concentration averaging currently under way. The study could be easily extended to include decommissioning hardware.
- Cost:** Costs for including decommissioning hardware in the EPRI study are estimated at 100–150 person hours.

b. U.S. Nuclear Regulatory Commission, *Technical Position on Concentration Averaging and Encapsulation*, Draft for Comment, March 1992.

4.2 Measurement Program

4.2.1 Activation Measurements at Reactors

- Purpose:** Uncertainties in levels of neutron fluence, alloy composition, and activation cross sections result in large uncertainties in waste classification of large-volume reactor internals that lie beyond the reactor core. These uncertainties could be mitigated through direct-assay analyses at reactors that have retired from service but have not been decommissioned or decontaminated. Several of these units exist in the U.S. that could provide the data base for a measurement program of waste characterization to reduce uncertainties in classification of all core shrouds and barrels.
- Procedure:** Characterize core shrouds or barrels at retired BWR and PWR plants using direct assay with underwater dose-rate profiles and samplings. Candidate plants for such measurements are Humbolt Bay, Dresden Unit 1, LaCrosse, Rancho Seco, and Yankee Rowe. These plants have fuel removed from the pressure vessel, and access is available from the top of the vessel after removing the head. The average dose rate of the components would be combined with dose-to-curie factors, either computed or measured, measured scaling factors, and decay and irradiation histories of the unit to estimate the activities and the average neutron fluxes seen by the components. These results would be used to predict activities expected in components in service at operating plants. Experimental results and interpretations would be documented in the report on the project.
- Practicality:** The program would require complete cooperation by the operator utilities of the plants selected, requiring operational support from plant personnel, perhaps even including assistance in measurements. Since the data from the measurements would be necessary to the utility for decommissioning, this degree of cooperation might easily be obtained. With this cooperation, the program should require only easily made modifications to previous fuel-pool classification campaigns.
- Cost:** The cost of this program for a single reactor unit is estimated at \$60,000 to \$90,000 exclusive of utility support. A program involving more than one plant should cost somewhat less per plant. Utility costs would hopefully be borne by them as a cost-sharing contribution.

4.2.2 Definition of ^{59}Ni -to- ^{63}Ni Activity Ratio

- Purpose:** The two activation products of nickel important in waste classification, ^{59}Ni and ^{63}Ni , usually dominate classification of irradiated components. Although ^{63}Ni concentrations are usually a higher fraction of the allowed 10 CFP 61 limit than those of ^{59}Ni and usually make up over 95% of the "Table 2" activities, the latter isotope can combine with ^{94}Nb and TRU concentrations to give high sum-of-the-fractions for Table 1 activities. ^{59}Ni is relatively difficult to measure because of the higher concentration (~100 times) of ^{63}Ni in the sample, and uncertainties in ^{59}Ni measurements are often high (>30%). The ratio of the two activities should be nearly a constant for all irradiated hardware and the accuracy of

⁵⁹Ni determinations would be improved if only the more abundant isotope were measured and the ⁵⁹Ni determined through a precisely determined ratio. This ratio is not known at present with enough precision to allow its use for accurate estimations.

- Procedure:** Procure samples of an irradiated high-nickel-containing alloy (e.g., inconel) and perform repeated high-precision and accurate measurements of the activity ratios of ⁵⁹Ni to ⁶³Ni, using either mass spectrometry or radiochemical techniques with calibrations from ⁵⁹Ni and ⁶³Ni standards and multiple laboratories. The program should establish the ratio with a precision of 5%.
- Practicality:** Samples could be collected as a portion of other commercial hardware classification programs. Sample splitting would be performed by one laboratory, and the samples would be shipped to all the participating facilities. Such programs have operated successfully in the past.
- Cost:** Analytical costs are estimated at \$1,000–1,500 per sample for mass spectrometry, less for radiochemistry. Both are necessary. Add-on costs to a classification campaign for collecting and distributing the additional samples are estimated at \$10,000.

4.2.3 Sampling Representativeness

- Purpose:** Although a few measurements have been made of the representativeness of scaling factors from a single sample when applied to the entire component, additional confirmation is desirable. Verification of representativeness should be for both bulk and surface activities. In the thick-wall core barrel and shroud components, measurements should also be of activity on both sides of the component along a radial direction to establish the radial activity variation. The activities, ⁶⁰Co, ⁶³Ni, ⁹⁴Nb, ⁹⁰Sr, ¹³⁷Cs, and gross α , should be the representative nuclides sought in these studies to reduce the uncertainty of scaling factor applications.
- Procedure:** Procure and radiochemically analyze samples taken along the length of a component to determine the variation in scaling factors taken from differing regions of a large component and along the radial dimension of the thick-wall components. These samples should be analyzed with high precision for the radionuclides: ⁶⁰Co, ⁶³Ni, ⁹⁴Nb, ⁹⁰Sr, ¹³⁷Cs, and gross α . The measurement program would best be a part of 4.2, Item 1, Activation Measurements at Reactors.
- Practicality:** Collection of samples for these measurements would be coordinated with any other measurement programs or with another commercial hardware classification campaign. The program is feasible in that framework.
- Cost:** Estimated costs for the analysis of a pair of samples for these six activities are estimated at \$1,500–2,000. Analysis costs would be shared with any other cooperative measurement program.

4.3 Development Program

4.3.1 Improved Sensitivity for ^{94}Nb Analyses

- Purpose:** Class C limits for the concentration of ^{94}Nb in irradiated hardware are very low (0.2 Ci/m^3). Although the radionuclide emits a gamma ray with an energy in a range that usually has a high sensitivity, the very high relative concentration of ^{60}Co in the samples, 10^5 - 10^7 times that of the niobium activity, results in low sensitivities for ^{94}Nb , even after enhancement by radiochemical separations. These detection sensitivities are often well above the actual activity in the sample and sometimes result in estimations of component concentrations above the 10 CFR 61 limit. This often results in a component being classified as GTCC partially on a minimum detection limit (MDL) for niobium and adds to the uncertainties in waste classification where the fraction of the limit for ^{94}Nb , based on MDL values, can exceed that from measured values of ^{59}Ni .
- Procedure:** Develop a practical radiochemical procedure for ^{94}Nb that can detect the nuclide in small samples containing ^{60}Co at a concentration 10^7 higher than that of the niobium (i.e., capable of yielding real scaling factors of $\leq 10^{-7}$).
- Practicality:** The procedure development is a practical solution to a problem that has presented difficulties for both the utilities and regulators in classifying irradiated hardware. The same inconsistencies would affect the DOE GTCC program if left unresolved.
- Cost:** Cost estimates for this development program should be 100–200 person hours.

4.3.2 Improved Precision in Radiochemical Procedures

- Purpose:** Because of potential loss of ^{14}C and other volatile products in radiochemical dissolutions, the bulk and surface samples acquired from irradiated components are not homogenized before aliquoting for individual analyses. A result is that often there is an assumption made of homogeneous activity on the sampling filter. The validity of this assumption is not known and its use may be a significant contributor to the differences between the internal and external variances discussed previously and shown in Table 8. Elimination of this uncertainty should reduce the uncertainties in resulting controlling scaling factors shown in Figure 6 to $\pm(10\text{--}15)\%$.
- Procedure:** Develop radiochemical procedures that would either ensure homogeneity of the entire sample before aliquots are taken or reduce the requirements for the homogeneity (e.g., take large enough aliquots to give accurate analyses of the normalizing ^{60}Co activity for each aliquot). One practical approach might be: since the radionuclides controlling waste classification of irradiated hardware are particularly volatile and their loss in dissolution should not be large, homogenize the larger fraction of the sample used in their analyses before aliquoting.
- Practicality:** The development would be a practical solution to reducing external variances in scaling factor measurements and should not compromise analysis procedures.

Cost: The cost of development should be small, since it should involve only a change in procedures by the contractor laboratories.

4.3.3 Improved Control of Gamma-Scanning Geometries

Purpose: A source of uncertainty in dose-to-curie factor measurements is the geometry of measurements of gamma-ray concentrations at specific locations on the component (gamma-ray scanning). This uncertainty is largest in components such as in-core instruments where the diameter of the component is comparable to that of the collimator diameter. A device that would position the scanner precisely on the component would improve the accuracy of results from these scans and significantly reduce the number of anomalous outlier measurements that result from improper alignments.

Procedure: Develop a jig to position the gamma-ray scanning collimator end in contact with and at a defined geometry with the component centered in the collimator, particularly for small-diameter components.

Practicality: The collimator could have a plastic sleeve fitted over the end that conforms to the geometry of the piece. Installation of such a jig and its use should be feasible.

Cost: Development costs for the positioning jig should be under \$5,000.

4.3.4 Improved Control of Dose-Rate Geometries

Purpose: Another source of uncertainty in dose-to-curie factor measurements is the geometry of dose-rate measurements. This uncertainty is also largest in components such as in-core instruments where the diameter of the component is comparable to that radiation probe. A device that would position the scanner precisely on the component would improve the accuracy of results from these dose-rate profiles. With an accurate positioning jig, the uncertainty in the dose-rate portion of the dose-to-curie factor and determining the average dose rate could be reduced to the order of 5-7%.

Procedure: Develop a jig to position the dose-rate monitoring probe in contact with or at a fixed but small distance from the profiled component, particularly for small-diameter and long components.

Practicality: The dose-rate probe could have a plastic sleeve fitted over the end that conforms to the geometry of the piece. Installation of such a jig and its use should be feasible, although its use might slow processing of multiple components during pool cleanup operations.

Cost: Development costs for the positioning jig should be under \$5,000.

4.3.5 Improved Control of Bulk Sampling

- Purpose:** Uncertainty in knowledge of the surface-to-volume ratio in the bulk sampling of a component adds some uncertainty to the estimates of surface activity determinations. Although these activities are not often influential in the waste classification process, measured concentrations of TRU on the surfaces can contribute a substantive portion of the Table 1 sum of the fractions. Reduction of the uncertainty in this factor should reduce actual uncertainties in TRU scaling factor measurements by about half.
- Procedure:** Measure the variance of the surface-to-volume factor in samplings of bulk material from nonradioactive stainless pieces in simulated pool sampling environments. Measurement of average grinding depths could use micrometer "feeler gauges." Develop procedures or modifications in the sampling head to reduce this variance.
- Practicality:** Practical development could use a local swimming pool for the testing.
- Cost:** Costs of measurements and development are estimated at one man-month.

4.4 Summary

This section has proposed 12 studies, measurements, and developments that can improve the accuracy of waste classification of irradiated hardware by direct assay and that would reduce the uncertainties in estimating GTCC wastes from decommissioning commercial power reactors. Suggested studies within the three areas of study were ranked in order of decreasing priority even though most are believed important. The measurement programs are the most important. For these programs, efficiency and economy could be gained in a combined measurement program performed at power reactors that have been retired.

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Appendix A

Propagation of Measurement Uncertainties

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Propagation of Measurement Uncertainties

The process of uncertainty propagation used in the assessment assumes that each individual uncertainty is independent and random, having a one- or two-sided Gaussian distribution about the measured value. Propagation of individual uncertainties to achieve estimates of the uncertainty in a component classification then uses the conventional propagation rules where the uncertainty in a result is the square root of the sum of variances of the components that make up the result. In a mathematical form, this is expressed as:

$$\sigma_v = \sqrt{\sigma_x^2 \left(\frac{\partial V}{\partial x}\right)^2 + \sigma_y^2 \left(\frac{\partial V}{\partial y}\right)^2 + \sigma_z^2 \left(\frac{\partial V}{\partial z}\right)^2 \dots etc.} \quad (1)$$

where

σ_i = the uncertainty (standard deviation) in the i^{th} variable

V = the product variable

$x, y, z, \text{ etc.}$ = the component variables .

The equation reduces to the square root of the use of the squares of the individual percentage uncertainties for the multiplication and division operations in characterizing and classifying waste by direct assay.

The propagation procedure treats each side of an asymmetric variable independently to compute the total uncertainty.

An example of an uncertainty propagation is as follows:

In Figure 6, the uncertainty for the average dose rate, A , is +5%, -12%, that for the average dose-to-curie factor, D , is $\pm 14\%$, and that for the scaling factor for ^{63}Ni , S_{63} , is $\pm 10\%$. The equation for computing the average concentration of ^{63}Ni in the component, C_{63} , is:

$$C_{63} = A \text{ (R/hr)} \times D \text{ (}\{Ci_{Co-60}/m^3\}/\{R/hr\}\text{)} \times S_{63}(Ci_{Ni-63}/Ci_{Co-60}) \quad (2)$$

Using Equation (1) to propagate the uncertainties associated with Equation (2) results in the equation:

$$\sigma(\%)_{C_{63}} = \sqrt{\sigma(\%)_A^2 + \sigma(\%)_D^2 + \sigma(\%)_{S_{63}}^2} \quad (3)$$

where the terms are the percent uncertainties.

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Thus, the upper uncertainty is:

$$\sigma_{C_{63}}(+)=\sqrt{5^2+14^2+10^2}=18\% \quad (4)$$

and the lower uncertainty is:

$$\sigma_{C_{63}}(-)=\sqrt{12^2+14^2+10^2}=21\% \quad (5)$$

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