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**An Evaluation of NDA Techniques and
Instruments for Assay of Nuclear Waste
at a Waste Terminal Storage Facility**

E. D. Blakeman
E. J. Allen
J. D. Jenkins

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AN EVALUATION OF NDA TECHNIQUES AND INSTRUMENTS FOR ASSAY OF
NUCLEAR WASTE AT A WASTE TERMINAL STORAGE FACILITY

E. D. Blakeman
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AN EVALUATION OF NDA TECHNIQUES AND INSTRUMENTS FOR ASSAY OF
NUCLEAR WASTE AT A WASTE TERMINAL STORAGE FACILITY

E. D. Blakeman E. J. Allen J. D. Jenkins

ABSTRACT

The use of Nondestructive Assay (NDA) instrumentation at a nuclear waste terminal storage facility for purposes of Special Nuclear Material (SNM) accountability is evaluated. Background information is given concerning general NDA techniques and the relative advantages and disadvantages of active and passive NDA methods are discussed.

The projected characteristics and amounts of nuclear wastes that will be delivered to a waste terminal storage facility are presented. Wastes are divided into four categories: High Level Waste, Cladding Waste, Intermediate Level Waste, and Low Level Waste. Applications of NDA methods to the assay of these waste types is discussed. Several existing active and passive NDA instruments are described and, where applicable, results of assays performed on wastes in large containers (e.g., 55-gal drums) are given.

It is concluded that it will be difficult to routinely achieve accuracies better than ~10-30% with "simple" NDA devices or 5-20% with more sophisticated NDA instruments for compacted wastes. It is recommended that NDA instruments not be used for safeguards accountability at a waste storage facility. It is concluded that item accountability methods be implemented. These conclusions and recommendations are detailed in a concurrent report entitled "Recommendations on the Safeguards Requirements Related to the Accountability of Special Nuclear Material at Waste Terminal Storage Facilities" by J. D. Jenkins, E. J. Allen and E. D. Blakeman.

1. INTRODUCTION

This report presents the results of a study to investigate the potential use of nondestructive assay (NDA) instrumentation at a nuclear waste terminal storage facility for purposes of special nuclear material* (SNM)

*Special Nuclear Material is defined in 10 CFR 70.4 paragraph (m) to be (1) plutonium, uranium 233, uranium enriched in the isotope 233 or in the isotope 235, and any other material which the commission pursuant to the Atomic Energy Act of 1954, determines to be special nuclear material, or (2) any material artificially enriched by the foregoing but does not include source material.

accountability. This study was performed for the Office of Waste Isolation (OWI) operated by Union Carbide Corporation — Nuclear Division (UCC-ND) for management of the National Waste Terminal Storage (NWTs) program. The objective of the NWTs program is to provide facilities in various deep geological formations at various locations in the United States for safe disposal of commercial radioactive waste. An assessment of the applicability of NDA methods at such a facility is representative of one of the numerous technical support activities required to thoroughly demonstrate the feasibility of the NWTs program.

A variety of NDA techniques and instruments have been developed and are currently in use to assay the fissile nuclide contents in a wide assortment of materials in containers ranging in size from a few cubic centimeters to large containers containing several 55-gal drums (barrels). The use of NDA techniques enables nuclear materials to be assayed without opening the container, or in other ways affecting the material. For waste containing nuclear materials which are highly radioactive or radiotoxic, and which are sealed in a container designed for permanent storage, this type of measurement if technically feasible provides clear advantages. For this reason NDA instrumentation should be evaluated for applications in a terminal waste storage facility. Incoming nuclear waste containers could be nondestructively assayed for SNM contents, thereby providing information for safeguards accountability.

This report is divided into six sections. Following the Introduction, Section 2 discusses the different NDA methods and provides comparative advantages and disadvantages for each. Section 3 describes the expected quantities, types, and characteristics of the wastes which will be received at a waste repository. In the fourth section, the problems associated with the assay of each waste type are discussed. Instrumentation in use or currently being developed for assay of wastes similar to those which will be received at a waste repository, and the results of assays performed with these instruments are described in the fifth section.

Finally, based on this information, conclusions and recommendations are presented.

2. REVIEW OF NONDESTRUCTIVE ASSAY (NDA) TECHNIQUES

2.1 Introduction

Nondestructive Assay (NDA) techniques can be grouped into two general categories. One category contains the "passive" methods in which radiation resulting from spontaneous fission or radioactive decay is detected and measured. Calorimetric methods, where the heat generated by α decay of fissile and fertile isotopes is used as a measurement of nuclear material content, are included in the passive NDA category.

The other category contains the "active" methods in which samples are "interrogated" with radiation and a material response is measured and related to the amount of special nuclear material present. Either neutrons or gamma rays may be used for interrogation. Induced radiation which can be related to SNM content include prompt fission neutrons, prompt fission gammas, and delayed neutrons. In general, passive NDA devices are inherently simpler than active devices, but are more susceptible to interference when the material to be assayed is in a very dense or heterogeneous matrix.

Figure 2.1 shows the various available NDA methods. These methods will be discussed in the following sections of this report. An attempt has been made to keep this section general in nature in order to give the reader an overall view of the extremely broad scope of nondestructive assay techniques. However, because the ultimate goal of this report is to evaluate application of NDA techniques to nuclear waste, techniques that do not have potential for this particular application are discussed only briefly. References are provided for the reader who desires further detail.

2.2 Passive NDA Techniques

2.2.1 Passive gamma methods

These techniques involve the measurement of gamma rays of discrete energies emitted from materials in a sample during radioactive decay. The most commonly measured spectral lines in fissile material assay work

Fig. 2.1. Categorization of NDA methods.

are the 186 keV line from ^{235}U and the 414 keV line from ^{239}Pu . These are moderately intense signatures and do not have severe interferences. Other lines such as the 129 keV and 375 keV lines of ^{239}Pu are also useful. Table 2.1^{1,2} provides a convenient listing of major gamma-ray signatures for fissionable isotopes, along with additional remarks concerning the usefulness of gamma signatures for assay purposes.

Two types of detectors are commonly used for gamma assay work. These are the NaI scintillation detector and the Ge(Li) solid state detector. Ge(Li) detectors have much better energy resolution (about 1% as compared to about 10% for NaI detectors).^{3,4} With the Ge(Li) detector, discrete spectral lines can be resolved and measured, whereas, with the NaI detector many spectral lines become "smeared" together making accurate assays more difficult, particularly if the isotopics of the sample are not well known. Ge(Li) detectors are limited in volume to about 80 cm³,* whereas, NaI detectors can be made up to 100 times this volume.³ NaI detectors can be made with a much higher efficiency than Ge(Li) detectors and therefore, require shorter counting times to achieve an equivalent number of counts. Ge(Li) detectors must be operated at liquid nitrogen temperatures (an operational complication) while NaI detectors may be used at room temperatures. The choice of detector depends on the required application. For detailed isotopic analysis, Ge(Li) detectors, because of their resolution capability, are preferred. For assay of samples containing small amounts of nuclear material or when short counting times are desirable, NaI detectors may be the better choices because of their higher efficiencies.

The primary difficulty encountered in employing gamma ray spectroscopy for material assay results from gamma absorption within the sample. Over 90% of the gammas detected come from within two mean-free-paths (mfp) of the surface of the sample. This means for example, in a $\text{PuO}_2\text{-UO}_2$ mixture with a density of 10 g/cm³, one mfp is about 0.3 cm and 0.07 cm for gammas of 385 keV and 186 keV respectively. Therefore, in a gamma

*Detector sizes are limited by the unavailability of large, sufficiently pure germanium crystals, and the difficulty of drifting lithium ions to greater depths than 1.5 cm.²

Table 2.1. Major gamma-ray signatures for the fissionable isotopes^a

Isotope	Energy (keV)	Intensity (g-s) ⁻¹	Comments
²³⁵ U	185.72	4.3×10^4	Only intense gamma ray. Resolved with NaI as well as Ge(Li). Useful for enrichment and quantitative measurements. Several much weaker peaks are seldom useful.
²³⁸ U	1001.10 766.40	1.0×10^2 3.9×10^1	These actually arise from the ^{234m} Pa daughter of ²³⁸ U. After chemical separation about 100 days are required for the activity to come into equilibrium at the levels stated. Plutonium-238 gives rise to the same 766.40-keV gamma and would produce interference in U-Pu mixtures. Useful for work with Ge(Li) or NaI.
²³⁸ Pu	766.40 152.77	1.5×10^5 6.5×10^6	Most useful for quantitative assay. Ge(Li) or NaI. Useful for isotopic determinations with Ge(Li).
²³⁹ Pu	413.69	3.4×10^4	The 413.69 usually provides the basis for Ge(Li) assays. The 413.69 plus the 375.03 and its weak neighbors form a complex upon which NaI assays are based.
²³⁹ Pu	129.28	1.4×10^5	Useful for isotopic determinations with Ge(Li). Plutonium-239 has over 100 gamma rays, some of which are useful for careful work with Ge(Li).
²⁴⁰ Pu	—	—	Several weak gamma rays but all suffer bad interference from gammas of other isotopes. Requires very careful work with high-resolution detector to make use of any of them.
²⁴¹ Pu	207.98	2.0×10^7	Actually from ²³⁷ U daughter and requires about 25 days after chemical separation to come into equilibrium at stated value. May also have a few percent interference from ²⁴¹ Am which emits same gamma. Nevertheless a good clean strong gamma useful with both NaI and Ge(Li).
	164.59	1.8×10^6	Useful with Ge(Li). Also from ²³⁷ U.
	148.60	7.5×10^6	Useful with Ge(Li). Direct from ²⁴¹ Pu.
²⁴¹ Am	59.54	4.6×10^{10}	Very strong gamma but attenuation problems. Useful with Ge(Li) or NaI. Has several other much less intense gammas sometimes useful for Ge(Li) work.
²⁴² Pu	—	—	No useful gamma rays at all. Nature failed us at this point.

^aFrom: LA-5651-M, September 1974.

assay of this density material, 90% of the detected ^{235}U gamma rays originate in the outer 0.14 cm of the sample. For ^{239}Pu 90% of the detectable gammas come from the outer 0.6 cm.^{3,4} Clearly, for dense or large samples, passive gamma assay is not feasible unless it can be assumed that the sample material is a homogeneous mixture in which gamma rays originating close to the surface are characteristic of the entire sample.

Another problem associated with passive gamma techniques arises from high background interference with the desired spectral line measurements. Much of this background is the result of Compton scattering within the detector. LASL has performed extensive assay work using Ge(Li) detectors and resolving background problems using computational methods.^{1,2} For small uranium and plutonium sample assay work, accuracies of ~1% have been readily achieved with careful assays.

The equation $M = CR \cdot CF / K^*$ illustrates the correction factors required to calculate the mass of a material from a measured count rate.^{1,2}

The attenuation correction factor, CF, can be determined by placing the sample between the detector and a known gamma source and measuring the gamma transmission through the sample (for large samples in circumstances where less accuracy is required, samples can be weighed and the gamma attenuation factor estimated from the calculated sample density). The factor K is a calibration constant which incorporates a measure of the detector efficiency, the specific activity of the gamma signature of interest, and the effects of sample geometry. This factor is deduced by first counting a known standard. Knowing CF for the standard and the mass M, the factor K can then be calculated.

In summary, passive gamma assays require the use of calibration standards and of some method (usually transmission measurements) to determine the matrix attenuation effects. To reduce effects due to geometrical inhomogeneities, samples are usually rotated during assay. In general, passive gamma detection methods cannot be relied on if the sample does not

*

M = mass of isotope

CR = measured gamma count rate from signature of isotope

CF = correction factor for sample attenuation

K = calibration factor (corrected counts 1 gram)

permit a source transmission measurement. Large, opaque and heterogeneous samples are not amenable to passive gamma assay techniques nor are samples containing high level extraneous sources of gamma radiation. Thus, passive gamma techniques are ruled out for several classes of OWI waste (see Sects. 4.2 and 4.4).

2.2.2 Passive neutron methods

Passive neutron methods like passive gamma measurements depend on the inherent radioactivity of the material. They enable assays to be performed by measurement of the neutrons emitted from a material of interest. Neutrons are produced by two processes: spontaneous fission and (α, n) reactions. Spontaneous fission occurs in all fissionable isotopes, but is about 10^4 times more frequent in the even Pu isotopes than the odd, i.e., in ^{238}Pu , ^{240}Pu , ^{242}Pu , and greater by an additional factor of $\sim 10^4$ in the higher even numbered actinides such as ^{242}Cm and ^{244}Cm . Therefore, its utility as an indicator is limited to the measure of fertile species, particularly ^{240}Pu in materials uncontaminated by higher isotopes.

The (α, n) neutrons are produced primarily by alpha particle reactions with ^{18}O in oxide fuels. Other low Z elements, especially fluorine and boron, which may be present in nuclear waste, have high (α, n) yields. Unless the chemical composition of a sample to be assayed is known and the sample is homogeneous, (α, n) neutron counting cannot be relied on to provide accurate quantitative assays because different isotopic and chemical combinations can give the same neutron output. Thus (α, n) neutrons are more of a nuisance than a benefit and should be considered as a useful indicator only for verification or comparison checks.

It is difficult to measure the SNM in a sample from the total neutron count since this value includes both spontaneous fission neutrons and (α, n) neutrons. It is possible however, to distinguish between spontaneous fission and (α, n) yields by means of neutron coincidence counting. The coincidence technique registers a count only when two neutrons are detected simultaneously. Because two or more neutrons are produced by a spontaneous fission, this technique enables spontaneous fission events to be counted while (α, n) neutrons are discriminated against.

Two types of neutron detectors commonly used are: BF_3 and ^3He detectors. BF_3 detectors use the reaction $^{10}\text{B}(n,\alpha)$ for thermal-neutron detection and can be used in the presence of comparatively large gamma fluxes because of high specific ionization and the large energy of the charged alpha particles which are released.⁵ Because the cross-section for the $^{10}\text{B}(n,\alpha)$ reaction drops rapidly with an increase in neutron energy, the reaction is insensitive to fast neutrons. For fast neutron detection, the counter must be placed in a neutron moderator material to slow fast neutrons down to thermal energies.

^3He detectors use the $^3\text{He}(n,p)^3\text{H}$ reaction. The cross-section for this reaction is slightly higher at thermal energies than that of the $^{10}\text{B}(n,\alpha)$ reaction, but is more than twice the $^{10}\text{B}(n,\alpha)$ cross-section at 1 MeV. ^3He detectors can be operated at higher gas pressures than BF_3 detectors; therefore they are usually more efficient.

A number of other neutron detectors (gas chambers, scintillators, semiconductors, nuclear emulsions, etc.) are available for specific applications. The reader is referred to Ref. 5 for details.

As in the case of passive gamma counting, matrix materials and inhomogeneities can cause variable detector responses. The presence of moderating materials tend to increase the absolute detector efficiency (as compared to an empty container with only a source present) and in the case of hydrogenous materials, the detector response can be changed by as much as 20%. Techniques do exist (the source addition technique⁶) by which the matrix material influence can be "normalized out." But all such stratagems result in additional system complexity and introduce their own uncertainties in the data analysis.

In summary, passive neutron techniques suffer in the presence of matrix inhomogeneities, unknown matrix compositions and matrix contamination by the higher actinides. For commercial reprocessing wastes, application is limited to assay of low level wastes and then, only when the plutonium isotopics are well characterized.

2.2.3 Calorimetric methods

The principle behind NDA by calorimetric techniques is to measure the isotopic decay power generation in a sample and correlate this value to

the isotopes present. To do so, the relative isotopics of the sample must be known.

Calorimetry is the most precise method for assay of plutonium in small samples. Extensive work has been performed by Mound Laboratories in this area.^{7,8} It is however, not useful for direct assay of ^{235}U or ^{238}U since the power output of these two isotopes is about 10^{-4} that for ^{239}Pu . The method is not suitable for waste that is contaminated with radioactive fission products nor is it suitable for assay of large samples because of the long equilibrium times required. Assay by calorimetry cannot therefore be considered as a useful method for assay of nuclear wastes in barrels or canisters.

2.3 Active NDA Techniques

2.3.1 Neutron interrogation methods

Neutron interrogation methods comprise one of the two categories of active NDA techniques. Depending on the limiting energy of the neutron source, interrogation may be made by subthreshold (<1 MeV) neutrons which induce fission in fissile nuclides or superthreshold (>1 MeV) neutrons which induce fission in fissile and fertile nuclides. Some active neutron devices permit the determination of both the fissile and fertile content of a material by making separate measurements using neutrons with energies above and below the fertile fission threshold. This is achieved by appropriately moderating or "tailoring" the source neutrons to the desired energy.

Because of the good penetrating qualities of high energy neutrons, active neutron methods may be successfully used for the assay of highly compacted materials in large containers (e.g., 55-gal drums). All neutron moderating and absorbing materials. The presence of varying amounts of hydrogen in nuclear wastes can cause a wide range of fission neutron yields for samples with similar quantities of SNM, thereby making an accurate assay of the material impossible unless the hydrogen content is known or can be corrected for.

Neutron interrogation techniques are also sensitive to nonhomogeneous matrix effects and therefore, rotation and translation of samples to

average out these effects is usually required. As in the case of passive gamma and neutron methods, self-attenuation effects also occur if the nuclear material is present in lumps. For active NDA techniques self-attenuation affects both the interrogating radiation and the detected radiation.

For purposes of this report, neutron interrogation techniques are grouped according to the method by which the interrogating source neutrons are produced. Four such categorizations are described based on (d,n), (p,n), spontaneous fission, and (α ,n) or (γ ,n) sources. We also categorize these techniques according to the radiation detected. For purposes of this report, two such categories are considered:

1. Detection of prompt fission neutrons and/or prompt gammas.
2. Detection of delayed fission neutrons.

Prompt neutron yields are more than two orders of magnitude greater than delayed neutron yields.⁹ Thus, better counting statistics are available when it is possible to count prompt radiation without source interference. However, there is a compensating effect due to the lower emission energies of delayed neutrons. Neutron detectors are inherently more sensitive to slow neutrons than fast neutrons and are therefore, more efficient for the detection of delayed neutrons than for prompt neutrons.⁵

Of the methods discussed in subsequent sections, delayed neutron counting is employed with (d,n) or (p,n) sources and prompt radiation is counted using (γ ,n) or (α ,n) sources. Systems for detection of either prompt or delayed radiation have been devised using spontaneous fission ^{252}Cf sources.

2.3.2 Interrogation by (d,n) sources

This assay method uses the exothermic (d,n) reaction:

The deuterons producing this reaction must have an energy of 100 keV or greater and are produced by either a Cockroft-Walton generator or a sealed gas discharge tube. The deuterons impinge upon a tritium target and produce neutrons of 14 MeV. Neutron yields are in the range of

10^{11} n/sec for the Cockroft-Walton generator and 10^{10} n/sec for the gas discharge tube.^{3,4} The most promising assay procedure is to pulse the source with pulse widths up to 1 second, allowing as long as 1 second between pulses to measure the delayed neutrons. Detectors used are BF_3 or ^3He imbedded in a polyethylene moderator and covered with Cd to reduce thermal neutron background. A reasonably high efficiency for detection of fast neutrons can be obtained.

The delayed neutron yields for fissionable isotopes decreases rapidly¹⁰ for interrogating neutron with energies above 5 MeV. It is necessary therefore, to moderate the 14 MeV source neutrons to energies below 5 MeV by placing moderating materials between the neutron beam and the assay sample. This procedure also enables interrogation at energies above and below the fission threshold for fertile isotopes (~ 1 MeV), thereby allowing determination of fissile and fertile contents.

Neutron interrogation techniques employing (d,n) neutron sources have potential for applications to the assay of nuclear wastes. They produce neutron fluxes of sufficient intensity to overcome background problems present in contaminated samples and of sufficient energy to allow penetration of large samples. Drawbacks to their application in industrial environments include source reliability, maintenance, and cost considerations.

2.3.3 Interrogation by (p,n) sources

The production of neutrons for this method is based on the endoergic reaction

High energy protons are directed on a Li target producing the interrogating neutron beam. The proton source, which must produce protons of 1.63 MeV or greater, is generally a Van de Graff generator. Unlike the (d,n) method which produces monenergetic neutrons of 14 MeV, this method allows for variation of the maximum energy of the neutron spectrum by controlling the energy of the proton source. Therefore, moderating the neutron beam to "tailor" the neutron energies becomes unnecessary. The

method allows for accurate discrimination between the fissile isotopes by measuring delayed neutron yields at different interrogating energies.

These advantages however, are offset by the expense and complication of the Van de Graff generator system. It is believed that this factor prohibits consideration of this technique for the practical assay of nuclear waste at a waste repository location.

2.3.4 Interrogation by spontaneous fission sources

The use of a ^{252}Cf spontaneous fission source to produce neutrons for active neutron interrogation has become feasible in the past few years due to a reduction in the cost of ^{252}Cf to about \$10/ μg . ^{252}Cf decays with a half-life of 2.6 years by alpha-decay. (The spontaneous fission half-life is 88 years.) Neutrons are produced at the rate of 2.34×10^6 n/sec/ μg . Therefore, for practical assay work where high intensity neutron sources of $\sim 10^8 - 10^{10}$ n/sec are required, quantities of the isotope in the range of ~ 100 μg to milligram amounts are needed.

^{252}Cf offers an inherent advantage over the use of accelerators for NDA because a complex machine is replaced with a reliable neutron source. However, the ^{252}Cf source cannot be turned off and on like an accelerator, and therefore, measurement techniques other than pulsing the source and measuring prompt or delayed neutron yields must be employed. Two such methods are described below.

- a. Detectors sensitive to fast neutrons can be used to count prompt fission neutrons if the source neutrons are moderated to energies below the detector sensitivity to prevent source interference.
- b. The source can be "shuffled" back and forth from a shielded area to a position where the sample is irradiated. During the time the source is away from the sample in the shielded area, the delayed neutron yield can be counted without source background interference.

A ^{252}Cf source could be used for superthreshold as well as subthreshold interrogation for assay of fertile as well as fissile materials in containers. However, as can be seen from Table 2.2,¹¹ the neutron spectrum begins to drop rapidly in intensity above energies of about 1-2 MeV, indicating that such an application is not very viable.

Table 2.2. Neutron spectrum from spontaneous fission of 1 gram of $^{252}\text{Cf}^a$

Energy (MeV)	Neutrons/sec · gram of nuclide
0 — 0.5	2.8×10^{11}
0.5 — 0.5	3.7×10^{11}
1.0 — 2.0	7.6×10^{11}
2.0 — 3.0	4.6×10^{11}
3.0 — 4.0	2.8×10^{11}
4.5 — 5.0	1.6×10^{11}
5.0 — 6.0	5.6×10^{10}
6.0 — 7.0	4.0×10^{10}
7.0 — 8.0	1.3×10^{10}
8.0 — 10.0	9.9×10^9
10.0 — 13.0	2.2×10^9
Total	2.4×10^{12}

^aRef. 11.

2.3.5 Interrogation by (α ,n) and (γ ,n) sources

(α ,n) reactions such as $^{239}\text{Pu-Be}$, $^{239}\text{Pu-Li}$, $^{210}\text{Po-Li}$, and $^{241}\text{Am-Li}$ or (γ ,n) reactions as $^{124}\text{Sb-Be}$ and $^{88}\text{Y-Be}$ may also be used as neutron sources for active assay purposes. [Reference 12 provides an extensive discussion of (α ,n) and (γ ,n) neutron sources including values for neutron yields, energy spectra, half-lives, costs, etc.] An advantage of a (γ ,n) source over a ^{252}Cf source is that the spectrum is softer, ensuring that all source neutrons are below the fertile fission threshold. This ensures that only fissions from fissile materials are detected and also reduces the interference of source neutrons with the detection of fission neutrons. Conversely, the softer spectrum makes this method less suitable for assay of large samples, particularly if the samples contain large amounts of neutron absorbing materials. A disadvantage of the two (γ ,n) sources mentioned are the short half-lives of the source materials. Half-lives of ^{124}Sb and ^{88}Y are 60.9 days and 107 days respectively.

Most (α, n) neutron sources give a harder spectrum than ^{252}Cf and therefore offer possibilities for the assay of fertile materials by superthreshold interrogation. However, it is difficult to produce, shield, and handle (α, n) sources above 10^7 – 10^8 n/sec. These are not intense enough for most waste assay applications.

Two devices which use (α, n) and (γ, n) reactions are the Random Source Interrogation System (Random Driver)¹³ and a photoneutron ^{124}Sb -Be system,¹⁴ both developed at LASL. The Random Driver uses a ^{241}Am -Li source of about 5×10^5 n/sec and is employed primarily to determine the ^{235}U content in containers up to 5 gal in size. The photoneutron ^{124}Sb -Be system has been used primarily to assay small samples such as PWR and BWR fuel rods and HTGR fuels to an accuracy of up to 1%. Both systems utilize coincidence neutron counting techniques. Gulf General Atomic has developed the Sub-MeV System using a ^{124}Sb -Be source for the assay of HTGR fuel and achieved accuracies in the range of 1%.¹⁵

Assay systems using (γ, n) or (α, n) sources appear suitable for assay of small samples such as fuel rods or fuel assemblies which are characterized by a homogeneous matrix and contain appreciable amounts of fissionable materials. For larger, heterogeneous samples, the soft spectrum of (γ, n) sources and the low intensity of (α, n) sources make these unattractive for accurate assay of large heterogeneous samples.

2.3.6 Interrogation by accelerator generated gammas

This technique uses high energy gamma rays from 5–10 MeV to produce photofission in samples. The resulting prompt neutrons emitted are then detected and the SNM content determined. Although delayed neutrons or gammas can be detected, the higher prompt neutron yield provides a better measurement.

The gamma source is produced by directing high energy electrons on a high Z target (converter). Bremsstrahlung is emitted. The electrons are usually produced by a linear accelerator (LINAC), although an electron microtron can also be used. At gamma energies of about 6 MeV, photofission is possible in all fissionable isotopes.

The fission neutrons are detected by BF_3 detectors imbedded in a hydrogenous matrix. For the first millisecond following the gamma burst,

prompt neutrons are detected. From 1-5 ms after the gamma burst, the bulk of the counts are due to delayed neutrons.

Figure 2.2³ provides a plot of neutron yield vs electron energy for several nuclides. As in the case for neutron induced fission the photo-fission delayed neutron yield for each nuclide is about 2 orders of magnitude below the prompt neutron yield for the same electron energy. Below electron energies of about 7 MeV, near the (γ ,f) threshold, neutron yields vary rapidly with energy. Above 7 MeV the change in yield is more gradual, roughly a factor of 2 for a 20% change in electron energy. It is possible to distinguish between various isotopes by making two or more measurements at different energies and measuring changes in neutron yields.

Active gamma assays can be greatly affected by the presence of ^2H , ^9Be , and ^{13}C in the matrix of the sample being interrogated. These three elements, all of which may occur in nuclear waste, have a high (γ ,n) cross section and may cause increased background problems. By taking two measurements, one above and one below the (γ ,f) threshold of the isotope of interest, these (γ ,n) effects can be corrected for.

Photofission stimulation techniques are applicable to assay of waste materials and, in fact, considerable experience has been obtained to date in this application (see Sect. 5.7). Primary drawbacks for application in industrial environments include the complexity of the source with the attendant cost and maintenance problems.

2.3.7 Interrogation by accelerator generated neutrons

A linear electron accelerator may also be used to produce neutrons for active interrogation.^{15,16} This is done by placing a secondary target in the bremsstrahlung beam to convert the gamma radiation to neutrons by (γ ,n) or (γ ,f) reactions. The resulting neutron beam is then directed on the assay sample and the resulting fission neutrons detected as in active neutron techniques. Figure 2.3 shows the LINAC configurations for both gamma and neutron interrogations.

Figure 2.4¹⁷ shows the neutron intensity from the secondary target as a function of the electron energy. In the figure several materials are considered for the secondary target and an optimal high-Z converter such as tungsten is assumed for the primary target. For an electron

Fig. 2.2. Prompt and delayed neutron yields in fissile and fertile materials produced by electrons of energy E_e .

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C CONVERTER - PRODUCES GAMMA RAYS FROM ELECTRON BEAM
 S SAMPLE
 D DETECTOR - DETECTS FISSION NEUTRONS
 T SECONDARY TARGET - PRODUCES NEUTRONS FROM GAMMA RAYS BY (γ, n) OR (γ, f) REACTION

--- ELECTRON BEAM (5-8 MeV)
 ~~~ GAMMA BEAM
 ——— NEUTRON BEAM OR FISSION NEUTRONS

Fig. 2.3. (a) Active gamma and (b) active neutron interrogation by use of an electron accelerator.

Fig. 2.4. Neutron yields as a function of electron energy.

energy of 8 MeV with a 50 μ A average electron current, the average neutron intensity is about 10^{10} n/sec using a 1-cm thick D₂O or beryllium target. This yield is roughly the same as that achieved by a 14 MeV neutron generator. A yield of about 6 times greater can be obtained using a ²³⁸U target. By using large volumes of D₂O, Be, or a combination of these materials and ²³⁸U, the average neutron intensities produced can exceed those of a 14 MeV neutron generator. These high neutron yields are one advantage of an electron accelerator as an active neutron assay device. The second major advantage is the capability of rapidly pulsing a LINAC and measuring the prompt neutron yields between pulses.

Figure 2.5¹⁷ shows the neutron spectrum obtained from a D₂O target for different electron energies. For an electron energy of 4 MeV, the maximum neutron energy is about 0.9 MeV. For an 8 MeV beam, the maximum neutron energy is about 2.9 MeV. Depending on the selected electron energy, neutrons can be produced for either sub- or superthreshold interrogations.

2.3.8 Electron microtron sources^{17,18}

Another type of electron accelerator under consideration for assay work is the electron microtron. This instrument, analogous to a cyclotron, electromagnetically accelerates electrons in a circular orbit to an escape energy. A microtron has several advantages over a LINAC electron accelerator. It inherently has much better energy resolution of the electron beam (0.25–1%). To obtain resolution widths of 100 keV on a LINAC, a bending magnet must be used with a defining slit arrangement. Stray radiation results and shielding problems arise. The microtron is more stable, safer, and easier to operate than a LINAC. On the other hand, an electron LINAC can produce a larger beam current than a microtron. It can also produce high instantaneous current in short bursts in the 10^{-9} second range.

2.3.9 Isotopic gamma source interrogation

Additional active NDA methods that use an isotopic gamma interrogation source rather than an electron accelerator, should be mentioned. These are: (1) X-Ray Fluorescence, (2) Absorptivity and (3) Absorption

Fig. 2.5. Neutron spectrum from the photodisintegration of the deuteron induced by bremsstrahlung.

Edge Densitometry.¹⁹ In the first of these techniques, X-ray emission lines are excited by a radioisotopic source. The other two techniques require the use of gamma transmission measurements to calculate the nuclear material in a sample. These techniques have been widely used to assay solutions and small samples. They are not applicable to nuclear waste in large containers and will therefore not be further discussed in this report.

2.4 Summary

Table 2.3 is presented as a summary of the various NDA methods and their possible applications. Specific applications of these methods to wastes that will be delivered to a nuclear waste storage facility will be discussed in Sect. 3 and examples of NDA instruments that are in use or are being currently developed will be presented in Sect. 4.

Table 2.3. General summary of NDA methods

NDA method	Description	Advantages	Disadvantages	Possible applications
<u>Passive</u>				
I. Passive gamma	Detection of γ signatures using Ge(Li) or NaI detector	1. Simple 2. Direct isotopic determination by unique signatures	1. Absorption in large or dense samples 2. Attenuation in lumps 3. Cannot be used if extraneous gamma sources (i.e., fission products) are present	Low density or homogeneous plutonium waste. Particularly useful for accurate assay of small containers with unknown isotopic content.
II. Passive neutron	Detection of spontaneous fission or (α, n) neutrons	1. Simple 2. Can be made insensitive to gamma radiation 3. Good for dense materials	1. Spontaneous fission and (α, n) neutrons must be distinguished from each other 2. Only measures fertile Pu directly 3. Sensitive to hydrogenous matrix	1. Measurement of ^{240}Pu in all waste types 2. May be useful as backup for passive gamma to determine if lumps are present or for assay when sample is too dense for passive gamma assay
III. Calorimetry	Nuclide decay heat measurement with calorimeter	Most accurate method if relative isooptics are known	1. Affected by extraneous heat sources (fission products) 2. Long measuring time	Only useful for small container assay. Measuring times are too long for assay of large containers
<u>Active</u>				
I. Neutron interrogation	Deuterons from sealed tube of Cockcroft-Walton Generator impinge on ^3H target to produce 14 MeV neutrons	1. Useful on dense or large samples 2. Can be used with gamma sources present 3. Sub- and superthreshold interrogation possible	1. Requires shielding 2. Sensitive to hydrogenous matrix 3. Expensive/complicated	Assay of all waste types. Fissile and fertile isotope determination
B. (p, n) source	Van de Graft Generator impinges protons on deuterium target	1. Useful on dense or large samples 2. Can be used with gamma sources present 3. Sub- and superthreshold interrogation possible 4. Control of neutron energy without use of spectrum tailoring targets	1. Requires shielding 2. Sensitive to hydrogenous matrix 3. Expensive/complicated	Useful at laboratory. Too expensive/complicated for plant use.

Table 2.3 (continued)

NDA method	Description	Advantages	Disadvantages	Possible applications
<u>Active (continued)</u>				
C. ^{252}Cf source	Spontaneous fission of ^{252}Cf produces interrogating neutrons	1. Useful on dense or large samples 2. Can be used with gamma sources present 3. Reliable source 4. Simpler than (d,n) or (p,n)	1. Requires shielding 2. Sensitive to hydrogenous matrix 3. Only accurate for sub-threshold interrogation	Assay of all waste types. Determination of fissile isotopes.
D. (α, n) source	α particles from α -emitting source impinge on target	1. Useful on dense or large samples 2. Can be used with gamma sources present 3. Sub- and superthreshold interrogation possible 4. Hard spectrum for super-threshold analysis	1. Requires shielding 2. Sensitive to hydrogenous matrix 3. Difficult to produce and shield large sources	Possible assay all wastes, but best for fuel rods/assemblies. Fissile and Fertile isotope determination.
E. (γ, n) source	γ particles from source impinge on target	1. Useful on dense or large samples 2. Can be used with gamma sources present 3. Sub- and superthreshold interrogation possible 4. Softer spectrum allows better tailoring and less interference of source with detector than for ^{252}Cf	1. Requires shielding 2. Sensitive to hydrogenous matrix 3. Softer spectrum only good for subthreshold interrogation 4. Short half-life of source	Same as above, but only for fissile determination
II. Active gamma interrogation	γ source produced by impinging electrons on high-Z target	1. Useful on dense or large samples 2. Can be used with gamma sources present 3. Sub- and superthreshold interrogation possible 4. Versatile - can also be used for active neutron interrogation with (γ, n) target	1. Requires shielding 2. Has problems associated with both gamma and neutron NDA 3. Expensive/complicated	Assay of all wastes. Possible fissile and fertile determination.

3. NUCLEAR WASTE CHARACTERISTICS

It is currently thought that nuclear wastes accepted for storage at a waste repository will consist of four basic types:²⁰ (1) High Level Wastes (HLW); (2) Cladding Wastes (CW); (3) Intermediate Level Waste (ILW); and (4) Low Level Waste (LLW). These wastes are defined and described below:

1. High Level Wastes (HLW)

Those aqueous wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuels. HLW contains:

- a. All nonvolatile fission products.
- b. Several tenths of 1% uranium and plutonium in spent fuels.
- c. All other actinides formed by transmutation of uranium and plutonium in the reactor.

(Note: HLW does not include ^3H , noble gas fission products and 99.9% of I, Br fission products.

2. Cladding Wastes (CW)

Contaminated fuel element materials from reprocessed spent fuel assemblies. CW contains:

- a. Fuel element structural materials: Zircaloy, stainless steel, Inconel, other miscellaneous materials.
- b. Up to 0.1% of plutonium originally in the spent fuel.
- c. 0.05% of actinides and fission products in spent fuel.

3. Intermediate-Level TRU Wastes

Those solid materials, other than high level and cladding wastes, which contain long-lived alpha activities greater than 10 nCi/g and have gamma radiation levels sufficient to require biological shielding and remote handling techniques. ILW contains:

- a. A wide assortment of items: paper, cloth, wood, plastic, rubber, glass, ceramic, metal, and immobilized salts and sludges that arise in the treatment of liquid waste streams and filters from clean-up of offgas.

b. Contains average of 0.025 g of Pu or U/ft³ (uncompacted)
(0.883 g/m³)

c. Contains assumed 0.025% fission products of spent fuels.

4. Low-Level TRU Wastes

Same as ILW except that external radiation levels are low enough to allow handling without supplementary shielding. LLW arises principally at fuel preparation/fabrication plants and to a lesser extent at fuel reprocessing plants. LLW contains:

a. Similar to ILW

b. Contains average 0.25 g of Pu or U/ft³ (uncompacted)
(8.83 g/m³) (Fuel losses of 0.25% in preparation, 0.5% fabrication, 0.1% reprocessing)

Characteristics of each waste type including densities, chemical and radionuclide composition, and surface dose rates are presented in Table 3.1.^{20,21} These waste characteristics are consistent with assumptions in Ref. 20 that spent fuel is reprocessed and plutonium recycled.

Table 3.1. Characteristics of nuclear waste types accepted at the reference design Waste Terminal Storage Facility

	Approximate density	Approximate composition	Approximate actinide content	Approximate surface dose rate
HLW	3.3 g/cc	SiO ₂ 25-40 wt % B ₂ O ₃ 10-15 wt % Waste oxides 20-35 wt % ZnO 5-10 wt % Alkali metal oxides 5-10 wt %	70 kg/m ^{3a}	10 ⁵ -10 ⁶ rem/hr canister surface
CW	4.5 g/cc	Zircaloy 88 wt % ^b Stainless steel 9 wt % Inconel 2.5 wt %	6.65 kg/m ^{3a}	10 ³ rem/hr at canister surface
ILW	2 g/cc (compacted)	Metals, ceramics, ash Fission products, actinides	10 g/m ³ (Pu)	10-1000 mrem/hr at canister surface
LLW	2 g/cc (compacted)	Same as for ILW	50-100 g/m ³ (>95% Pu)	<10 mrem/hr at container surface

^aThese are total actinide contents. For HLW, CW, and ILW, plutonium amounts are only a few percent of total actinide contents.

^b~100% stainless steel for LMFBRS.

It has been proposed that standard containers be used for storage of each waste type. High Level Waste, Cladding Waste, and Intermediate Level Waste require external shielding and will probably be delivered in canisters of identical design, each having a waste volume of .18 cubic meters. The proposed design is shown in Fig. 3.1.²² Low Level Waste does not require external shielding and will probably be delivered in 55-gal drums, each with a waste volume of .21 cubic meters.

The four waste types considered above comprise very general categorizations of nuclear wastes and may vary considerably in composition. At Rocky Flats, for example, plutonium wastes are segregated into 52 separate categories, each with unique compositional characteristics.²³

The High Level Wastes, for purposes of this report are assumed to be solidified in borosilicate glass. This process seems to be the most feasible at this time, although other solidification processes are under consideration.²⁰ Compared to ILW and LLW, HLW will not vary greatly in chemical or isotopic composition. The relative isotopics of the actinides present will be in approximately the following proportions: 77% U, 4% Np, 2.3% Pu, 13% Am and 3.5% Cm.²⁰ These values will change with the age of the waste. About .03-.08 m³ of HLW will be generated per ton of fuel reprocessed. It is anticipated that by the year 2010, a volume of 9.98×10^3 m³ of HLW will be accumulated at a federal waste repository. The total actinide content of this waste will be about 700 MT with a total radioactivity of about 72,000 Mci.²⁰

Cladding wastes from LWR fuels are not expected to vary substantially in composition or isotopics except to the extent to which they are compacted. These wastes can be compacted to within 70% of their theoretical density. This will increase their density from ~ 1 g/cm³ to 4.5 g/cm³. Assuming a factor of 4.5 compaction, about .065 m³ per metric ton of PWR fuel, .082 m³ per metric ton BWR fuel, and .25 m³ per metric ton of LMFBR fuel will be generated. It is estimated that 8.51×10^3 m³ of compacted cladding wastes will be accumulated by 2005 in a federal waste repository with a total actinide mass of 56.62 MT and a total radioactivity of 406 Mci.²⁰ The relative actinide isotopics of the waste will be about the same as that of the spent fuel prior to reprocessing.

Fig. 3.1. Assumed standard canister for high-level, cladding, and intermediate-level wastes (generic repository description).

Intermediate Level Wastes and Low Level Wastes, unlike HLW and CW, will vary considerably in chemical composition, depending on the origin of the wastes in the fuel fabrication or reprocessing process. A large fraction (1/2-1/3) of LLW, particularly lower density wastes such as cloth and paper, are combustible and may be reduced by incineration by factors of about 50 and 20 in volume and weight respectively. Also, about 1/2 to 3/4 of the wastes may be reduced in volume by factors of 2 to 10 by compaction. More development work must be done, however, before wastes can be routinely compacted by a factor of 10. Mound Laboratory has performed research in this area and has compacted drums of LLW containing >10 nci/g <100 nci/g by a factor of 4.²⁴ Compaction factors of 10 should be achieved for these wastes in the late 1980's.

About 280 m^3 of Intermediate Level Wastes are generated per MT of plutonium (or ^{233}U) processed. An expected $37.9 \times 10^3 \text{ m}^3$ volume of waste will be accumulated in a repository by 2005 with a total plutonium mass of 300 kg. (Isotopics are similar to those of spent LWR fuel.) About 280 m^3 , 560 m^3 , and 113 m^3 of Low Level Waste are produced by fuel preparation, fabrication, and reprocessing respectively. It is anticipated that a total volume of $0.111 \times 10^3 \text{ m}^3$ of LLW will be accumulated in a repository by 2005 with a total actinide mass of $9.18 \times 10^3 \text{ kg}$.

Figures 3.2 and 3.3 summarize the estimated waste volumes and actinide masses for each of the four waste types that will accumulate in a federal repository from the present to the year 2010 for HLW and 2005 for CW, ILW, and LLW. It can be seen that the projected volume accumulation of LLW will be greater than twice that of the other wastes combined by the year 2005, and greater than the volume of HLW by more than an order of magnitude. However, because of its concentrated nature, the accumulated actinide content of HLW will be more than 5 times that of the other wastes combined and more than 40 times that of LLW. Since the actinide content of HLW from LWR's contains about 2.3% plutonium, whereas actinide content in LLW is $>95\%$ Pu, the accumulated plutonium content of both waste types will be about the same.

Table 3.2 presents estimated quantities (kg/MT fuel) of fission products, structural materials, and heavy metals found in each of the four waste types. The composition of a typical PWR spent fuel assembly

Fig. 3.2. Estimated accumulation of nuclear wastes at a federal repository.

Fig. 3.3. Estimated accumulation of nuclear wastes at a federal repository.

Table 3.2. Nuclear waste contents

kg of fission products, structural materials,
and heavy metals per MT PWR fuel reprocessed

Waste type	Fission products	Structural materials	Heavy metals
	(kg)	(kg)	(kg)
HLW	29.1	0	5.48
CW ^a	.0174	271	.483
ILW ^b	.0087	0	~.25 (1% Pu)
LLW ^c	0	0	~.1 (95% Pu)
Fuel assembly	34.8	271	965

^a Assumes .05% of fission products and heavy metals from spent fuel.

^b Assumes .025% of fission products from spent fuel and .025 g-Pu/ft³ waste (uncompacted).

^c This value is assumed to be the same for fabrication and preparation.

is also given since these may be regarded as waste if fuel reprocessing is not implemented. These values were determined by the ORIGEN²⁵ computer code and are consistent with assumptions regarding losses in fuel preparation, fabrication, and reprocessing as stated in Ref. 20. From Table 3.2, it is observed that Cladding Wastes contain only about .06% as much fission products and heavy metals per MT of fuel reprocessed as High Level Wastes. However, the heavy metal/fission product ratio is about the same and indicates similar assay problems will be experienced. Additionally, Cladding Wastes contain activated structural materials which will contribute to the gamma background. Although more dilute than HLW or CW, Intermediate Level Wastes have substantial fission product contamination which presents background interference with assay measurements.

Table 3.3 presents a breakdown of the heavy metal isotopes for typical LWR spent fuel and for the concentrated HLW effluent from a reprocessing plant as obtained using the ORIGIN computer code. Typically,

Table 3.3. Grams of heavy metals in spent fuel and reprocessed nuclear waste for one MT of PWR fuel^a

Nuclide	Spent fuel		Reprocessed waste (HLW)	
	Initial	10-year decay	Initial	10-year decay
Pb-208	4.59 E-7	4.15 E-5	7.88 E-7	3.47 E-6
Th-288	1.82 E-6	1.94 E-5	2.76 E-6	1.72 E-7
Th-230	9.13 E-4	4.44 E-3	1.06 E-3	1.08 E-3
Th-232	2.34 E-4	1.53 E-3	2.90 E-4	2.97 E-4
Th-234	1.36 E-5	1.36 E-5	1.36 E-5	6.79 E-8
Pa-231	5.13 E-4	5.91 E-4	5.17 E-4	5.17 E-4
Pa-233	1.58 E-5	1.17 E-5	1.66 E-5	1.67 E-5
U-232	2.83 E-4	8.13 E-4	1.74 E-6	4.08 E-6
U-233	4.80 E-3	6.34 E-3	2.45 E-5	1.73 E-3
U-234	121	134	.610	1.02
U-235	7980	7980	39.9	39.9
U-236	4550	4550	22.7	22.7
U-237	10.6	1.92 E-5	1.55 E-7	9.41 E-8
U-238	9.43 E+5	9.43 E+5	4710	4710
Np-237	472	486	482	483
Np-239	79.7	7.81 E-5	7.82 E-5	7.82 E-5
Pu-236	6.57 E-4	5.81 E-5	2.97 E-6	2.60 E-7
Pu-238	161	160	.836	5.50
Pu-239	5190	5270	26.3	26.4
Pu-240	2170	2170	10.8	20.1
Pu-241	1030	643	5.06	3.15
Pu-242	354	354	1.77	1.78
Am-241	25.1	412	46.3	47.5
Am-242m	.942	.900	.940	.899
Am-242	.0783	1.08 E-5		
Am-243	94.3	94.4	94.4	94.4
Cm-242	10.1	2.17 E-3	5.14	2.17 E-3
Cm-243	.0807	.0650	.0799	.0643
Cm-244	30.2	20.6	29.7	20.2
Cm-245	1.93	1.93	1.93	1.93
Cm-246	.222	.221	.222	.221
Cm-247	2.86 E-3	2.86 E-3	2.86 E-3	2.86 E-3
Cm-248	1.93 E-4	1.93 E-4	1.93 E-4	1.93 E-4

^a 3.3% enriched U-235; 33,000 MWD burnup.

HLW isotopics will approximate those presented in the Reprocessed Waste column of Table 3.3 while heavy metal isotopics in cladding wastes will be similar to those shown in column 1. Plutonium isotopics for LLW will also approximate those in column 1, although other heavy metals will be present only in small amounts. Typically, the actinide content of LLW will be about 95% plutonium.

ILW wastes will have variable isotopics depending on their source, but the majority will be bounded by the values in Table 3.3.

Table 3.4 presents estimates of average neutron production rates from the four waste categories and Fig. 3.4 shows pictorially, the estimated span of densities and radiation levels anticipated. These data have significant impact on the applicability of NDA devices for assaying the four types of materials as discussed in the following section.

Table 3.4. Estimated (α, n) and spontaneous fission neutron production rates in nuclear wastes (per m^3 of waste)

Waste type	Neutron source	Initial (n/s - m^3)	One year (n/s - m^3)	Ten years (n/s - m^3)
HLW	S. F.	5.65×10^9	4.46×10^9	2.98×10^9
	(α, n)	7.13×10^8	2.12×10^8	5.82×10^7
	Total	6.36×10^9	4.67×10^9	3.04×10^9
CW (compacted)	S.F.	2.96×10^6	2.52×10^6	1.60×10^6
	(α, n)	4.16×10^5	2.25×10^5	9.04×10^4
	Total	3.38×10^6	2.75×10^6	1.69×10^6
ILW ^a	S.F.	4.99×10^5	4.25×10^5	2.70×10^5
	(α, n)	7.02×10^4	3.81×10^4	1.52×10^4
	Total	5.64×10^5	4.63×10^5	2.85×10^5
LLW ^a	S.F.	$\sim 2.9 \times 10^4$	$\sim 2.9 \times 10^4$	$\sim 2.9 \times 10^4$
	(α, n)	$\sim 6.4 \times 10^4$	$\sim 6.7 \times 10^4$	$\sim 8.5 \times 10^4$
	Total	$\sim 9.3 \times 10^4$	$\sim 9.6 \times 10^4$	$\sim 1.1 \times 10^5$

^a Both wastes are compacted with densities of about 2 g/cc.

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Fig. 3.4. Estimated ranges of density and surface dose rate for nuclear wastes.

4. ASSAY OF NUCLEAR WASTES

4.1 Introduction

Selection of the most suitable NDA technique for a particular type of nuclear waste requires consideration of the following three inter-related factors. These will be discussed in detail in later sections for each waste type:

1. Waste Characteristics — These include the chemical, physical, and nuclear properties, size of containers, and homogeneity of waste mixtures.
2. Type/accuracy of Assay Required — This depends on the isotopes that must be measured, the assumptions one can make about the waste, and the amount of time available to perform assays. It may be feasible to measure only the fissile isotope contents for one waste type, whereas for another it may be required to directly measure both fissile and fertile isotope contents. Some assays may require accuracies of a few percent whereas for others only a "crude" estimate may be acceptable. All of these factors influence the selection of a practical assay system.
3. Waste Standards — The availability of representative standards influences the calibration and therefore the achievable accuracy of NDA instrumentation.

4.2 Passive Assay of Fission Product Contaminated Waste

High Level, Cladding, and Intermediate Level Waste all produce high radiation backgrounds due to the fission products present. Because of this high gamma radiation background, passive gamma methods cannot be used to assay these wastes. Furthermore, these wastes contain appreciable amounts of ^{242}Cm and $^{244}\text{Cm}^*$ (see Table 3.3), both of which have a high spontaneous fission activity producing a high neutron background. The

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*Curium is not a fission product, but is produced during irradiation in the reactor.

spontaneous fission half-lives of ^{242}Cm and ^{244}Cm are 7.2×10^6 years and 1.4×10^7 years respectively.²⁶ This corresponds to respective spontaneous fission rates of 7.59×10^6 spontaneous fission/s/g for ^{242}Cm and 3.87×10^6 spontaneous fission/s/g for ^{244}Cm . These values are about 4 orders of magnitude greater than the spontaneous fission rate of ^{240}Pu . ^{244}Cm is the longer lived of these two nuclides ($T_{1/2} = 18$ years as compared with 163 days for ^{242}Cm) and is expected to be present in Cladding Wastes, and Intermediate Level Wastes in quantities of about a hundredth of that of ^{240}Pu . Much higher relative amounts are estimated for High Level Wastes, because actinides other than plutonium and uranium are concentrated in these wastes. Thus, neutrons from curium in these wastes will mask the spontaneous fission neutrons from even-numbered plutonium isotopes and preclude use of passive neutron methods. Only active interrogation methods appear to be applicable to these three classes of waste.

4.3 Active Assay of High Level and Cladding Wastes

Neither High Level nor Cladding Wastes are amenable to passive assay techniques because of their high neutron and gamma backgrounds. However, it may be technically feasible to utilize active interrogation methods. The total neutron cross-section of zirconium, which is about 88% of the chemical composition of LWR Cladding Waste, is about 7 barns for 1 MeV neutrons. If it is assumed that these wastes are compacted to a density of about 4.5 g/cc, the mean-free-path of 1 MeV neutrons in the material is about 5 cm. Since the waste can be assumed homogeneous, it is reasonable to expect that, even in the most compacted situation, enough neutron penetration can be achieved to assay them by active neutron interrogation methods. Similarly, for gamma rays of 3 MeV or greater, the mass attenuation coefficient of zirconium is $\sim 0.04 \text{ cm}^2/\text{g}$. This corresponds to a mfp of about 5.5 cm for the most compacted case. Again, it is reasonable that, even if the waste is highly compacted, effective penetration of Cladding Wastes may be achievable using high energy gamma interrogation methods, and assays may be possible. High Level Wastes, which have lower densities than highly compacted Cladding

Wastes, can similarly be penetrated by 1 MeV neutrons or high energy gammas.

The presence of high neutron backgrounds in these wastes complicate the successful application of NDA techniques. For Cladding Wastes, the ratio of curium to the fissile isotopes is about the same as for spent fuel assemblies. Problems associated with overcoming these background effects will be similar to those encountered in spent fuel assay (Sect. 4.8). Ratios of curium to fissile uranium and plutonium in High Level Waste is expected to be greater than 100 times the relative amounts in LWR spent fuel assemblies.* The neutron background effects from these wastes will be awesome. Extremely high intensity interrogation sources ($\sim 10^{11}$ – 10^{12} n/sec) only achievable with an electron accelerator or neutron generator may have to be employed to successfully assay these wastes. No attempts have been made to assay materials of this type and the utility of such an undertaking is questionable.

The authors believe that no consideration should be given to the assay of High Level or Cladding Wastes at a waste storage facility for safeguards purposes. HLW contains an average actinide content of about 70 kg/m³ of which about 2% is plutonium. Thus about 250 grams of plutonium will be present in each HLW canister. While this may be a considerable amount and is, in fact about an order of magnitude greater than the anticipated plutonium content of a drum of LLW, one must recognize that first, sophisticated technology will have already been applied to extract all of the plutonium from the material prior to its becoming waste and second, additional processing will have been performed to convert the residue to as tightly bound and insoluble a form as possible. Thus, the ability of a diverter to extract usable amounts of plutonium from solidified HLW assuming he were intrepid enough to steal the material, is negligible. These considerations, coupled with the associated radiological hazards, make the probability of diversion of this waste remote.

The utility of assaying HLW in its solidified and encapsulated form is also of little value from the material balance point of view. Typically the material will originate from large liquid holding tanks

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*Determined by the previously mentioned ORIGEN code.

where it will have resided and been homogenized for a considerable (nominally 5 years) period of time prior to solidification. Its assay will therefore have little value to the material balance records of the shipper who presumably will sample and assay incremental additions to his waste tanks, and none to the receiver. If a final assay is required, the logical point is just prior to or following calcination when the material is still sampleable and amenable to chemical assay.

A canister of CW will contain on the average only slightly more plutonium than a drum of LLW. For this reason, plus the arguments advanced above for HLW, no serious consideration should be given to the nondestructive assay of this waste.

4.4 Passive Assay of Low Level Waste

Because of the wide variation of matrix composition within this category, Low Level Waste presents problems for assay by passive non-destructive methods. LLW can be assayed by passive gamma methods, but the presence of lumps or matrix inhomogeneities presents limitations. If it is assumed for example that a barrel of LLW contains a homogeneous matrix consisting of carbon with a density of 1.5 g/cm^3 , then the mean free path for 414 keV gammas emitted from ^{239}Pu is about 6.9 cm (disregarding self-shielding from nuclear material present).² Since the radius of a 55-gal barrel is $\sim 28 \text{ cm}$, this means that less than 2% of the 414 keV gammas emitted from the center of the barrel would arrive at the surface without an interaction. A matrix of higher density or of higher Z material would allow even less penetration of the signature gammas. Nuclear material close to the center of the waste drum may therefore be particularly difficult to detect.

Table 4.1 shows the results from computer calculations run to investigate the magnitude of the self shielding effects caused by several idealized inhomogeneities of plutonium distribution within a waste drum. Details on the calculations are given in Appendix A.

The results indicate that the combined effects of matrix density and SNM distribution in a drum of waste can cause drastic variations in the intensity of the emitted radiation. Since the location of the SNM

Table 4.1. LLW computer simulation
 Relative transmission of ^{239}Pu 414 keV gamma radiation in
 LLW for carbon matrices with densities of 1 g/cc and 2 g/cc

Plutonium distribution in waste matrix	Transmission of 414 keV gamma line (normalized to homogeneous case for matrix density of 1 g/cc)			
	Cylindrical geometry		Spherical geometry	
	1 g/cc	2 g/cc	1 g/cc	2 g/cc
Homogeneous plutonium distribution	1	.529	1	.546
Plutonium located in outer radial interval ($\Delta R = .5$ cm)	3.26	2.96	2.43	2.11
Plutonium located in center interval ($R = 3$ cm)	.111	4.27×10^{-3}	.120	3.13×10^{-3}
Plutonium located in lump in center of container	NA	NA	1.64×10^{-2}	8.77×10^{-4}

in a waste container and the waste density are expected to vary considerably for LLW, it is apparent that a wide range of values could be obtained for the same SNM contents in a LLW drum. Corrections for these factors can be partially obtained by performing transmission measurements on a sample to determine matrix attenuation effects, and by rotating the sample to average assymmetric SNM distribution. Rotation does not, however, account for radial variations in the SNM distribution and cannot differentiate between a lump of SNM near the center of a drum and a homogeneous SNM distribution. Similarly, transmission measurements only provide average matrix attenuation corrections, and do not account for radial variation of the matrix within a waste drum. It is doubtful therefore, if passive gamma measurements could be effectively used for precise quantitative assay at a waste repository unless they could be used in conjunction with passive or active neutron measurements to detect lumps of heterogeneous matrix/SNM distributions.

4.5 Active Assay of Low Level and Intermediate Level Wastes

LLW that has been highly compacted or ILW that contains fission product activity cannot be effectively assayed by passive gamma methods. The most effective way to assay these wastes is by use of the active neutron or gamma interrogation methods described in Sect. 2.3. Waste densities of ~ 2 g/cc do not prevent penetration of moderated neutrons for subthreshold interrogation unless neutron absorptive material such as boron is present. Also, the small amounts of SNM present in these wastes (e.g., 20 g) are not likely to be present in "lumps" large enough for appreciable neutron self-attenuation effects.

Active assay of ILW will present some problems due to the presence of fission product gamma background radiation and neutron background due to the presence of ^{242}Cm and ^{244}Cm . Surface doses as high as 1000 mrem/hr (see Table 3.1) will have to be attenuated to acceptable detector bias levels (e.g., 100 mrem/hr) by lead shielding.²⁷ Also, greater neutron source intensities than used for LLW assay will be necessary to override the neutron background present in ILW.

The bulk of the effort expended to date on assay of low and intermediate level waste containers has been directed towards active interrogation techniques. Section 5 presents examples of active instruments. Much of the experience with these instruments is applicable to the assay of wastes anticipated at a waste terminal storage facility.

4.6 Assay of Waste Near the 10 nci/g Level

The discussion thus far has centered on the applicability of specific NDA techniques to generic classes of waste assuming "average" properties for each class. The LLW and ILW materials present a peculiar problem for NDA techniques, however, at the lower end of their concentration limits — that is, can they be assayed at all?

Estimated average plutonium quantities in ILW and LLW are about 2 g for a canister of ILW and about 10–20 g for a 55-gal drum of LLW. Alpha activities of wastes containing these amounts of plutonium are considerably greater than 10 nci/g, the lower limit for which ILW and LLW will be

considered for nonretrievable storage at a waste repository. Table 4.2² presents the alpha activity for the principal isotopes found in transuranic waste, and the amount of each isotope needed to produce 10 nci/g in a 55-gal drum with a matrix density of 1 gm/cm³.

Table 4.2. Alpha activities for plutonium, uranium, and americium

Isotope	Alpha half-life (year)	Alpha specific activity (α /s-g)	Alpha ci/g	Grams in 55-gal waste drum for 10 nci/g equivalent ^a
²³³ U	1.6×10^5	3.5×10^8	9.5×10^{-3}	0.22
²³⁸ Pu	86	6.4×10^{11}	17	1.2×10^{-4}
²³⁹ Pu	2.4×10^4	2.3×10^9	6.2×10^{-2}	3.4×10^{-2}
²⁴⁰ Pu	6.6×10^3	8.4×10^9	.23	9.1×10^{-3}
²⁴² Pu	3.8×10^5	1.4×10^8	3.8×10^{-3}	.55
²⁴¹ Am	460	1.2×10^{11}	3.2	6.5×10^{-4}

^a Assume for waste in drum, $\rho = 1$ g/cm and volume = 2.08×10^5 cm³
10 nci/g is equivalent to 2.08×10^{-3} ci/drum.

A reasonable assumption for the plutonium isotopics in LLW 5 years after generation and just prior to delivery to a waste repository is 1% ²³⁸Pu, 59% ²³⁹Pu, 24% ²⁴⁰Pu, 12% ²⁴¹Pu, and 4% ²⁴²Pu.²⁰ The ²⁴¹Pu does not have appreciable alpha activity, but beta decays to ²⁴¹Am which has a specific-alpha activity of 1.2×10^{11} α /sec-g. The ²⁴¹Am content after 5 years is about 3.5% of the plutonium content. From the values in Table 4.2, the specific alpha activity of this typical material is calculated to be about 1.35×10^{10} α /sec-g or about .36 α -ci/g (due primarily to the ²³⁸Pu and ²⁴¹Am content). For a 55-gal drum of LLW with a matrix density of 2 g/cc, 10 nci/g is equivalent to 4.16×10^{-3} ci/drum. Therefore, only 1.15×10^{-3} g of plutonium (plus ²⁴¹Am) is required to reach the 10 nci/g alpha activity in a drum of LLW.

It is reasonable to expect that waste drums containing activities as low as 10 nci/g will be received for burial at a waste repository even though the average activity/l per drum will be much greater than

this amount. Activities this low in containers as large as 55-gal drums are not amenable to assay by passive methods, and will be difficult to accurately assay by any method.

As an example of the lower sensitivity limits for current active techniques, LASL has estimated the sensitivity of their ^{252}Cf "Shuffler"²⁸ to be about 80 mg ^{239}Pu for a carbon matrix of 1.0 g/cm^3 in a large container (see Sect. 5.5). This value is about an order of magnitude greater than the total plutonium (containing ^{238}Pu and ^{241}Am) content at the 10 nci/g level. If it is assumed that ^{239}Pu is the only isotope present, this sensitivity would still be about twice the plutonium amount equivalent to 10 nci/g. This indicates that while it may be possible to assay wastes with very low alpha activity levels, wastes in the 10 nci/g range will probably remain below the level of achievable sensitivity for some time.

4.7 Assay of ^{233}U Waste²⁹

One eventuality which the waste repository must plan for is that the use of High Temperature Gas Cooled Reactors will comprise as much as 2% of installed nuclear capacity during the 1990's. This will lead to nuclear wastes containing ^{232}Th and ^{233}U . If this materializes, it is projected that by the year 2005 approximately 280 m^3 of low level ^{233}U waste with a total actinide content of 19 kg will be accumulated at a waste terminal storage facility. Although this amount is only 0.25% of the expected plutonium low level wastes, NDA problems peculiar to this type of waste should be addressed.

Assay of ^{233}U waste is complicated considerably by the high gamma activity and associated background interference from small and variable amounts of ^{232}U . This isotope is produced in the following ways. (See Figs. 4.1 and 4.2.)

1. (n, 2n) reactions in ^{233}U
2. (n, 2n) reactions in ^{232}Th followed by (n, γ) reactions in ^{231}Pa
3. Two sequential (n, γ) reactions on ^{230}Th and its daughter, ^{231}Pa

For passive gamma assay, ^{235}U has no prominent gamma lines equivalent to the 186 keV line from ^{235}U or the 414 keV line from ^{239}Pu . The two

Fig. 4.1. Decay chains of ^{232}Th and ^{232}U .

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Fig. 4.2. Uranium-233 decay chain.

most prominent lines at 42 and 97 keV are too weak and obscured by ^{232}U background activity to be useful for practical assay work. One particular daughter product in the ^{232}U decay chain does have high-energy gamma lines which may be useful for assay purposes. ^{208}Tl , which has a 76 year half-life, emits an intense 2.61 MeV gamma-ray during beta decay to ^{208}Pb . The intensity of this line at equilibrium is 35.93 gammas per 100 decays of ^{232}U or 2.9×10^{11} γ 's/sec/g- ^{232}U . Thallium also results from the decay of ^{232}Th but, because of this isotope's long half life, the thallium activity from this source is small. If it is assumed that 100 ppm of the total uranium is ^{232}U , then the intensity of this line is about 2.9×10^7 γ 's/sec/g-uranium. By comparison with Table 2.1 it is noted that the 2.61 MeV ^{208}Tl gamma line will be more intense than the plutonium or uranium lines when the ^{208}Tl has reached equilibrium with the ^{232}U (about 10 years). Measurement of this line by passive gamma methods is reasonable. Section 5.9 discusses a system that uses this technique for passive assay of ^{233}U waste in 55 gal drums. This technique to determine ^{233}U content can only be used if a detailed history of the waste is known. If the ^{208}Tl is in equilibrium with the ^{232}U , then the amount of ^{232}U can be determined and the $^{233}\text{U}/^{232}\text{U}$ ratio is defined. In general, $^{232}\text{U}/^{233}\text{U}$ ratios will vary with the thorium-uranium isotopics and with reactor burnup. ^{232}U amounts may vary from 100 ppm to 1500 ppm of the total uranium content.

^{233}U waste may also be assayed by active methods similar to those used for plutonium or ^{235}U waste. Passive neutron methods cannot be used since the spontaneous fission rates of ^{233}U and its associated isotopes are small.

4.8 Assay of Spent Fuel Assemblies

If reprocessing of spent nuclear fuel is not implemented, spent fuel assemblies will be considered as nuclear waste to be disposed of at a waste terminal storage facility. It is possible, in this event, that some requirements may be imposed to measure the fissile contents of these assemblies for safeguards purposes. Table 3.2 gives estimated quantities of fission products, structural materials, and heavy metal contents in

nuclear wastes obtained from the reprocessing of 1 MT of PWR fuel. All of these constituents will be present in spent fuel assemblies. Because of the fission products and activated structural materials present, assay of spent fuel assemblies poses problems similar to the assay of HLW or CW. The use of passive radiation instruments is not feasible and any instrument using active interrogation techniques will require the use of shielding material (Pb) between the detector and fuel assembly to reduce the high gamma flux to appropriate detector limits.

Science Applications, Inc. under subcontract from IRT Corporation has investigated the feasibility of direct nondestructive measurement of fissile material contents of spent LWR fuel assemblies.¹¹ The use of both isotopic sources and accelerator based techniques were studied, though emphasis was put on the use of isotopic sources because of their relative simplicity. Two such isotopic sources considered practical for spent fuel assembly assay are ^{252}Cf spontaneous fission sources and photoneutron (primarily Sb-Be) sources. Sb-Be sources, though limited by their 60.2 day half-life, produce a soft (keV) interrogating neutron spectrum which does not interfere with fission neutron detection. Sb-Be and ^{252}Cf sources are both available with the large neutron intensities ($\sim 10^9$ n/sec) necessary to overcome the neutron background from the ^{242}Cm and ^{244}Cm present in spent fuel. Current (α, n) sources are not readily available with high enough intensities and are therefore, not recommended for this application.

Figure 4.3¹¹ depicts a "generic measuring system" which forms the basis for the SAI/IRT study. This system is designed to operate under water in a pool where the spent fuel is located and cooled. As indicated in Fig. 4.3, lead shielding is employed to protect detectors from gamma background. The lead shield is surrounded by a thin polyethylene moderator to further slow down prompt and delayed neutrons. Source neutron spectrum tailoring is accomplished by the H_2O coolant. If a ^{252}Cf source is used, fast neutron detectors can be located on the far side of the source and the induced prompt fission neutrons counted. Alternatively, the source can be cycled to and from the irradiation position and delayed neutrons counted. An Sb-Be source can be used in the same manner. One

1 PWR FUEL ASSEMBLY CONSISTING OF ABOUT 200 LOW-ENRICHED URANIUM FUEL PINS IMMERSSED IN WATER	4 POLYETHYLENE (CH_2) MODERATOR
2 LEAD SEGMENTS	5 BF_3 TUBES AND POLYETHYLENE SPACERS
3 LEAD SHIELDING	6 POLYETHYLENE (CH_2) REFLECTOR
	7 POSSIBLE SOURCE LOCATION

Fig. 4.3. Cross-section of the generic measuring system.

method of "turning-off" a Sb-Be source for delayed neutron measurements, is to rapidly remove the Sb-gamma source from the Be converter.³⁰

An important observation in the SAI/IRT study is that neutron self-shielding effects are partially cancelled out by fast neutron multiplication within the fuel assemblies. It is also argued that the use of suitable standards will allow corrections to be made for the residual self-shielding effect.

Similar studies have been performed at Oak Ridge National Laboratory on the conceptual design of a system for the nondestructive total assay of a spent LMFBR fuel subassembly.³¹ This system is shown in Fig. 4.4. The original design utilized 12 ^{106}Ru - ^{106}Rh sources containing about 50 g of ^{106}Ru . Gamma rays from these sources produce in the D_2O photo-neutrons of about 0.02 MeV. An updated design incorporates Sb-Be photo-neutron sources intense enough to produce fission neutrons exceeding the neutron background due to spontaneous fission neutrons. Other background sources are (α, n) neutrons and neutrons produced in the D_2O by gamma rays from the spent fuel itself. The inner lead shield attenuates these gamma rays and the B_4C shield attenuates low energy neutrons that would not penetrate the fuel.

Analyses of this system were performed using the ORIGEN and ANISN codes. It is estimated that assays of accuracies of 1-5% can be achieved for a throughput of 5 tons/day (2 to 4 spent fuel subassemblies per hour).

As is the case of High Level and Cladding Wastes, the authors consider any attempt to assay spent fuel at a waste repository for material accountability purposes totally unnecessary. While spent elements will contain substantial (~ 5 kg/element) amounts of plutonium, the possibility of diversion of this material by surreptitious extraction without destruction of the element is inconceivable. Accordingly item accountability and identification is all that is required for material control purposes.

4.9 Waste Standards

The accuracy of any assay instrument depends upon proper calibration prior to assay of a particular waste type. In order to obtain accurate

Fig. 4.4. Spent-fuel-element nondestructive assay system.

calibration factors applicable to the waste assayed, waste standards used must match waste types as closely as possible in matrix material and SNM quantity and distribution. Because of the wide variation in waste types expected in the low level and intermediate level categories delivered to a repository, a large number of standards with variable compositions and distributions of nuclear material would have to be employed. For LLW, for example, as many as ~30 different standards would be necessary.

In addition, since all 55-gal drums look alike, some drum classification and identification system would be required to insure that the proper calibration curves were used in interpreting the results for each drum assay. The associated record keeping and verification system (to insure the shipper was properly identifying each drum's contents) and handling problems (since different types of waste might have to pass through different instruments) will be complicated and expensive.

4.10 NDA Instruments as Attribute Indicators

Nondestructive assay methods can be loosely grouped into two general categories^{32, 33} in their application to SNM accountability. These are (1) variable methods and (2) attribute methods. Variable methods, by application of one or more of the NDA techniques previously described, are used to make absolute measurements of the amount of nuclear material in an item, assigning to that value some reasonable measurement error. Up to this point, our discussion of NDA instruments has assumed their use as variable NDA instruments. Such instruments for safeguards applications must be capable of detecting diversion attempts in which small amounts of SNM have been removed or otherwise diverted from the system.

Attribute measurements on the other hand, are "go/no-go" type measurements, sensitive to diversion efforts in which a large fraction (e.g., 30-100%) of the SNM in an item has been removed and replaced by degraded material. Attribute measurements provide useful safeguards information and may be a reasonably alternative to the performance of accurate NDA measurements at a waste repository. They in general rely

upon relatively rapid and crude techniques to indicate if the SNM contents of a waste container deviate substantially from expected values. Passive neutron methods or a combination of passive neutron and passive gamma methods (NaI detectors) are feasible for this kind of application. Rocky Flats is currently developing an instrument that uses multiple passive neutron detectors to assay large crates of plutonium contaminated wastes. Errors on the order of ± 50 –100% are expected, but it will be possible to determine if grossly different than expected amounts of SNM are present.

Another "go/no-go" instrument in the conceptual stage at Rocky Flats is a device that reads radiation signatures from waste containers (crates or drums). This instrument would be used to measure the radiation signature of a container prior to shipment to the waste repository and again upon arrival. The two signatures would be compared to determine if the container shipped was the container received or if tampering occurred during shipment. This instrument would utilize both gamma (NaI) and neutron detectors. Although an approximate assay could be performed, the instrument's primary purpose would be to obtain and compare radiation signatures. Some development work will be necessary before the feasibility of this instrument can be determined. One disadvantage is the necessity of maintaining and cross calibrating identical equipment at the shipper and the repository stations.

4.11 Summary

The preceding sections have pointed out the effects that the various waste characteristics have on attempts to implement the different non-destructive assay techniques. We have seen that passive techniques, which are preferable from an operational point of view and which are best able to give detailed information on isotopic distributions, are ruled out for high level, intermediate level, cladding wastes and spent fuel assemblies. We have concluded that assay of the high level and cladding wastes (and spent fuel elements, should they eventually be discarded) will be extremely difficult by any technique.

Intermediate and low level wastes in 55-gal drums are assayable by active interrogation techniques but, because of the wide variations in

composition and density expected at the repository in these categories, accurate measurements of SNM content will be difficult and will require maintenance of multiple standards for instrument calibration.

There is, however, an alternative application of NDA techniques for material control at a waste repository. That is, in the role of attribute indicators where their principal function will be to provide gross verification of the contents of waste containers and shipper-receiver verification. Implementation of such a system presents no significant technical problems.

The following section presents examples of NDA instruments applicable to the assay of nuclear waste in large containers such as 55-gal barrels. It will be seen that certain of the instruments show promise of potential use at a waste repository within the limitations as summarized in the above remarks.

5. REVIEW OF EXISTING WASTE ASSAY INSTRUMENTS

5.1 Introduction

This section describes NDA instruments that have been developed and used or are currently being developed to assay nuclear waste in waste drums or other large containers. In subsections 5.2–5.9, eight instruments are described. Five employ passive means (either passive gamma, passive neutron, or a combination of both). Three use active neutron or gamma methods. Where applicable, assay results obtained or expected with these instruments are discussed, with an assessment of their potential application at a waste repository.

5.2 Eight Channel Drum Scanner and Segmented Gamma Scanner

This instrument³⁴ was developed by LASL to assay large waste containers up to 55-gal barrels using passive gamma techniques. The unit was designed to supplement/complement the active interrogation techniques tested in MONAL (Mobile Nondestructive Assay Laboratory), particularly for barrels containing plutonium — bearing hydrogenous waste.

The instrument uses a vertical assay of eight 2 in. × 2 in. NaI detectors to measure sample activity in the 385 keV complex from ^{239}Pu , the 766 or 153 keV gamma rays from ^{238}Pu , or the 186 keV gamma rays from ^{235}U . Detectors are spaced over the entire barrel height about 4–6 in. apart and about 5 in. from the barrel surface. The output of each detector passes through a single-channel analyzer which brackets the 385-keV gamma complex. Eight 100 μCi ^{22}Na sources, placed 180° opposite the gamma detectors, are utilized for an external transmission scan. Barrels are rotated during counting to reduce the dependence of the response on the azimuthal plutonium distribution.

The detectors are collimated both vertically and horizontally. The effect of the horizontal collimation is to selectively reduce the response of material located near the periphery of the barrel. Also, by positioning the detector close to the barrel and using horizontal collimation, the radial distribution of the plutonium can be determined from a

measurement of the counting rate vs the angle of rotation. This data can be collected, displayed, and analyzed using the multiscaling unit of a multichannel analyzer.

The 8-channel drum scanner was used to assay the ^{239}Pu content in scrap and waste in 55-gal drums at Rocky Flats. For these measurements, average accuracies were about 11% (1σ) for assay times of about 10 min.

Utilizing the same concepts, a segmented gamma scanner has been developed and marketed by Camberra Industries, Inc.³⁵ This instrument utilizes a single high resolution Ge (Li) detector for assay of uranium or plutonium contaminated wastes in 55-gal drums. Assays are performed by horizontally collimating the detector and moving the drum past the collimator such that the drum is effectively segmented into horizontal slices approximately 3 in. thick. Transmission correction measurements are performed for each segment. Drums are rotated several revolutions during each counting interval to compensate for azimuthal matrix inhomogeneities.

For assay of ^{239}Pu or ^{235}U , the 414 keV or 186 keV lines respectively are measured. A multichannel analyzer is used to define regions of interest and enable background subtraction. For determination of correction factors, a ^{75}Se (400.1 keV) transmission source is used for ^{239}Pu waste and a ^{169}Yb (177.2, 198.0 keV) transmission source for ^{235}U waste. Sources with gamma lines very close to the plutonium or uranium lines of interest were chosen in order to determine the most accurate transmission correction factors. This, however, necessitates the use of a high resolution Ge(Li) detector in order to ensure that the transmission source lines can be separated from the uranium or plutonium lines. The Ge(Li) detector used is 45–50 mm in diameter by 2.3 cm thick and has a resolution <1.2 keV (FWHM) at 122 keV.

While assay data is being acquired, a correction source is also counted to compensate for instrument dead-time and pulse pile up. Apparent intensity changes in this source are measured and a correction factor determined. Correction sources used are ^{133}Ba (356.3 keV) for ^{239}Pu and ^{57}Co (122.0 keV) for ^{235}U .

Operation of the system is flexible and automated. With pushbutton control, the following functions are available: initialization, calibration, assay, abort, and source shutter operational check. Control of the system is accomplished with a Digital Equipment PDP-11 mini-computer.

Tests have shown that this instrument is capable of determining the presence of 5 or more grams of ^{239}Pu in 55-gal drums with matrix densities up to 0.5 g/cc with an accuracy better than $\pm 20\%$ in a scanning time of about 10 min. For assay of ^{235}U , similar sensitivity and accuracy is obtained for matrix densities up to 0.2 g/cc. Complete assay of a 55-gal drum can be performed in about 15 min. Beyond densities of about 0.5 g/cc transmission measurements cannot be accurately made and use of this instrument is not feasible.

This instrument reflects the current state-of-the-art in gamma assay for measurements of wastes in large containers. Its use is recommended for applications to lower density materials (< 0.5 g/cc). However, due to the dense, heterogeneous nature of LLW expected to be received at a waste terminal storage facility, its use at such a facility would be extremely limited for accurate NDA work.

5.3 4π Neutron Coincidence Counter

This instrument^{28, 34, 36} was developed by LASL for the assay of large samples such as 55-gal drums using passive neutron detection. The coincidence counting technique is used to distinguish between spontaneous fission neutrons and (α, n) neutrons using BF_3 neutron detectors located in the walls of the sample chamber. Forty-eight detectors are used. Each is 2 in. in diameter and positioned between 1 in. thick polyethylene slabs. An additional 15 BF_3 detectors are contained in both the top and the bottom of the chamber. The chamber is shielded by a 2 ft thick water shield. A door in the side of the counter allows samples to be admitted and an entrance ramp to the chamber facilitates drum handling.

For preliminary research with the 4π neutron coincidence counter, assays were performed on mocked-up 55-gal barrels containing hydrogenous and nonhydrogenous matrices. Matrix materials included iron, fire brick, graphite, CH_2 , and rags. Responses were normalized to an empty barrel.

Spontaneous fission neutrons were produced by a ^{252}Cf source placed at various radial and axial locations. The source strength was about 10^4 n/sec. (This is equivalent to about 10 grams of ^{240}Pu , which produces about 1.0×10^3 spontaneous fission neutrons per second per gram.) For nonhydrogenous materials, spatial responses corresponding to different axial and radial source locations did not vary appreciably. It is estimated that about $\sim 10\%$ uncertainty in assays will arise due to spatial variation in the detector response. For barrels with hydrogenous matrix materials, large spatial variations are encountered and an estimated $\sim 30\%$ measurement uncertainty is expected due to these variations. Figure 5.1³⁶ compares the spatial effects observed in a nonhydrogenous matrix (iron) and a hydrogenous matrix (CH_2).

The 4π neutron coincidence counter was used to assay mocked up 55-gal drums containing a vacuum filter sludge matrix consisting of approximately 40% solids and 60% water.⁶ A 20 g sample of plutonium (5% ^{240}Pu) was located at various positions in the drum to determine spatial response. The detector responses varied by as much as an order of magnitude from center to edge, indicating a very strong spatial effect. Also, volume averaged detector response indicates that coincidence counting only samples approximately one third of the waste volume. Wastes such as these are obviously very difficult to assay by passive means, using either neutron or gamma methods. It is believed at LASL that assay accuracies of $\sim 25\%$ may be achieved if the drum is reasonably homogeneous and suitable calibration factors can be obtained.

An instrument of this type could be successfully employed at a Waste Terminal Storage Facility to obtain measurement accuracies on the range predicted above on LLW drums. However, due to the previously discussed limitations in all passive neutron devices, this instrument could only be recommended to provide "back-up" NDA measurements.

5.4 Gamma Counter II

This instrument,³⁴ developed at Rocky Flats, uses NaI scintillation detectors to measure gamma rays in the plutonium 385 keV complex. BF_3 detectors are also used to count (α, n) and spontaneous fission neutrons.

Fig. 5.1. Spatial detector response of neutron well counter to (a) non-hydrogenous and (b) hydrogenous matrices in 55-gal waste containers.

Gamma counts are used for quantitative assay, while the ratio of neutron counts to gamma counts is used for diagnostic purposes. Corrections for gamma ray attenuation by the sample matrix are based on the matrix weight. The instrument has been calibrated for several matrices, including combustibles, resin, graphite, lead gloves, Raschig rings, CWS filters and insulation.

Three 1 in. $\times$ 2 in. NaI detectors are used. These are placed in a vertical array at a distance of about three drum diameters from the sample. Sixteen polyethylene moderated BF_3 detectors are used for the neutron counting. The instrument is located in a shielded underground cavity.

Using counting times of 100 seconds, accuracies of $\pm 10\%$ have been achieved on samples consisting of waste in miscellaneous categories and containing 100 g of plutonium. Barrels containing lead gloves were assayed to $\pm 30\%$ and expected assay of CWS filters is $\pm 25\%$.

An instrument of this design offers potential for possible use at a Waste Terminal Storage Facility. Although the gamma measurements above are unreliable for dense barrels, a combination of gamma and neutron measurements can still be used for most LLW types to provide rough estimates (within say 50-100%) of a waste drum contents, or simply (as a signature device) to compare shipper and receiver measurements.

5.5 ^{252}Cf "Shuffler"

The "Shuffler,"²⁸ developed by LASL, is an active neutron interrogation device utilizing a ^{252}Cf neutron source. To ensure that the ^{252}Cf source does not interfere with measurements, the instrument is operated by transferring or "shuffling" the source from the interrogation region to a shielded position prior to measuring the induced delayed neutrons.

A schematic diagram of the "Shuffler" is shown in Fig. 5.2.²⁸ The shielding tanks and fast-neutron irradiation chamber are shown as well as the ^3He detectors used to measure delayed neutrons while the ^{252}Cf source is cycled from the irradiation position to the dwell position. Twenty-five ^3He tubes in a CH_2 moderating matrix are used. They are connected in parallel to a single preamplifier and amplifier. The irradiation tank is on wheels and can be positioned either closer to the

Fig. 5.2. Modulated ^{252}Cf assay system, "shuffler," showing the shielding tanks and fast-neutron irradiation chamber. The ^{252}Cf source cycles between the irradiation position and the dwell tank; during the latter time intervals (i.e., ^{252}Cf in dwell tank) the delayed neutrons induced in the sample are counted using the ^3He detectors.

source for better small sample assay or it can be removed entirely for assay of large assemblies as barrels or fuel assemblies. Cycling of the ^{252}Cf source is accomplished with a Teleflex Helix-wound cable driven by a stepping motor. Transfer times as low as 0.5 sec are possible.

The ^{252}Cf "Shuffler" can be used for both fast- or thermal neutron interrogation, depending on the sample to be assayed. For fast neutron interrogation, the sample chamber is lined with cadmium to absorb low energy (<0.3 eV) neutrons from the interrogation, and the neutron tailoring assembly consists of tungsten, beryllium, and nickel. For interrogation with thermal neutrons, the cadmium liner is removed and neutrons are tailored with lead and CH_2 .

Published results of measurements of large Pu containing samples with the "Shuffler" system are not currently available. Estimates, however, have been made of the instrument sensitivity. If a carbon matrix is assumed ($\rho = 1.0 \text{ g/cm}^3$) with $10 \text{ g } ^{239}\text{Pu}$ uniformly distributed, it was estimated by LASL that the instrument sensitivity for a 1000 seconds count is $\sim 80 \text{ mg } ^{239}\text{Pu}$ at the 3σ level.³⁷ Experimental measurements since have verified a sensitivity of 42 mg of ^{235}U which corresponds to a sensitivity of about 63 mg of ^{239}Pu .³⁸

Although it has not yet been commercially developed and marketed, the "Shuffler" design could be used at a waste repository to assay most low level wastes with sensitivities as indicated above and with probable accuracies within the 10-25% range. The instrument also has potential applications to assay of ILW, but the higher neutron background levels expected from this material will degrade performance and/or require a larger interrogating source. More extensive research however, will be necessary to fully determine the capacity of this instrument.

5.6 "Jumbo" Random Driver

This instrument^{39,40} was built by National Nuclear Corporation for the assay of materials in large containers, including 55-gal waste drums. Both passive and active modes can be used to assay for uranium or plutonium. The basic detector unit consists of four plastic scintillator detectors. These are mounted with associated photomultipliers in a

steel box large enough to accommodate samples up to 64 cm × 64 cm × 100 cm. For use in the passive mode, the unit is capable of counting fast fission neutrons and fission gammas detected simultaneously by any 1, 2, 3, or 4 of the detectors. For active neutron interrogation, up to 4 Pu-Li sources, each with a strength of approximately 10^6 n/s, can be inserted at opposite sides of the device. The use of multiple sources in this manner eliminates the need for a turntable for rotating the assay sample. Also, a uniform response is obtained throughout the detection volume.

Measurement sensitivities of 0.2 grams for ^{235}U and 0.15 grams ^{239}Pu have been obtained with this instrument in the active mode. Sensitivities are based on 2σ statistics and 10 minute counts.

This instrument represents current state-of-the art development in a Random Driver for assay of waste drums. Measurement accuracies for most low level waste types are in a range comparable to the ^{252}Cf "Shuffler" as described in Sect. 5.5. However, it does not have as low a sensitivity as the "Shuffler," nor can it be used for assay of fission product contaminated waste due to the sensitivity of scintillation detectors to gamma activity. It could be used at a waste repository for assay of compacted LLW. However, as discussed in Sect. 2.3.5, the development potential of an (α, n) or (γ, n) NDA instrument is considered secondary to an instrument using a spontaneous fission ^{252}Cf source.

5.7 GAMAS (LINAC)

A mobile assay system (GAMAS),^{34, 41} was designed and built by Intelcom Rad Tech. GAMAS is a hybrid assay system utilizing active and passive neutron and gamma assay devices, all of which are located in an 8 ft by 36 ft trailer. The system includes instruments as listed below and is shown in Fig. 5.3.⁴¹

1. An electron linear accelerator (LINAC) for x-ray beam interrogation.
  2. A shielded high efficiency neutron detector used to measure spontaneous fission neutrons or x-ray induced neutrons.
  3. NaI and Ge(Li) detectors for passive gamma measurements.
  4. An isotopic source assay system (ISAS) using a ^{252}Cf source.
- This device is only for assay of small samples.

Fig. 5.3. Cutaway drawing of GAMAS.

An on-line computer is included in the system for collecting and processing data and to interface electronics equipment with associated detectors.

The LINAC, used for active gamma assays, is used to produce a pulsed beam of electrons with energies ranging from 5.0 to 8.9 MeV. A tantalum converter is used to produce a continuous bremsstrahlung x-ray spectrum with energies up to that of the electron beam. The x-ray beam is then collimated to a 12 in. wide by 6 in. high spot on the sample at a distance of 5 ft from the accelerator output. Neutrons produced by photofission (γ, f) and by (γ, n) reactions in the sample are detected by modulated banks of BF_3 tubes.

A commonly used method for assaying a sample involves two separate irradiations, one at 7.5 MeV and another at 5.5 MeV. During the 7.5 MeV irradiation both (γ, n) and (γ, f) prompt neutrons are measured. During the 5.5 MeV irradiation, only (γ, n) neutrons are produced and measurements can be used to correct the 7.5 MeV measurement for (γ, n) background. Also, for some samples, fissile and fertile isotopes can be calculated by measuring prompt to delayed neutron ratios at two different energies.

The GAMAS System was used to measure a wide variety of nuclear materials at several locations. At NRTS, Idaho, 14 waste barrels were actively assayed by the LINAC and also passively assayed by the GAMMAS barrel scanner (see Sect. 5.4). Comparative results are shown on Table 5.1.⁴¹ Three of the four barrels containing greater than 5 grams of plutonium were relatively dense and had a significant hydrogen content. A representative sample barrel was not available, so the available data were used to calculate a gamma-beam and neutron attenuation correction. For barrel 771-7828, containing some very dense material in the bottom half of the barrel, active assay results were more accurate than passive gamma results (within 3%). For the other barrels, errors were large and results were not significantly different for the two methods of assay.

Table 5.2³⁴ is also presented as a summary of results of assays performed on 55-gal drums and fuel assemblies. Because of the complexity and expense involved, an electron accelerator, such as the one in the

Table 5.1. Comparison of active and passive measurements of SNM in 55-gal drums

Barrel	Shipper's value Pu (g)	Gulf Rad Tech	
		Passive Pu (g)	Active Pu (g)
771-19720	0	0	—
771-20042	0	0.6	—
771-7816	37	20.5	18.7
771-7817	34	12.3	11.4
771-7949	2	1.7	—
771-19977	0	0	0
771-19705	0	0	—
771-7895	18	22.7	30.2
771-19721	0	0.1	—
771-19706	0	0	—
771-19719	0	0	—
771-7828 ^a	40	19.9	38.9
771-19715	0	0.4	—
771-19700	0	2.7	—

^aVery dense barrel.

Table 5.2. GAMAS (LINAC) assay results

SNM	Sample description	(P) Precision (A) Accuracy	Assay time per sample	User
Pu	Waste material in 55-gal drums (20-200 g Pu/drums).	(A) $\pm 15\%$ (1 σ) (poor standards; no segregation) (A) $\pm 7\%$ (1 σ) (good standards and segregation).	10 min	Rad. Tech. at Rocky Flats
U	Waste material in 55-gal drums.	(P) $\pm 2\%$ (1 σ) (dry waste) (P) $\pm 9\%$ (1 σ) (wet waste).	10 min	Rad. Tech. at UNC, Hermatite
U	Fuel assemblies ATR & ETR fuel assemblies.	(P) $\pm 1\%$ (1 σ)	3 min	Rad. Tech. at NRTS

GAMAS system, is not at this time considered advisable for installation at a waste repository. The development of such a device for this purpose should be considered only if it becomes necessary to achieve assay accuracies that cannot be achieved with simpler isotopic source instruments. However, future development work may produce simpler, effective accelerators, making their potential use in the applied NDA field a more attractive alternative. This trend is indicated by a portable accelerator constructed by Varian Associates and tested by JRB Associates for potential NDA applications.^{42,43} This device is comparatively simple, compact (29 in. x 28 in. x 66 in., 2000 lb), and usable without a magnetic analysis system usually required for system stability with a LINAC. Although this instrument was constructed for radiographic work at Stanford University, an instrument of this or a similar design definitely offers potential for practical NDA work.

5.8 Neutron Generator Assay System (NGAS)

A joint contract between the AEC and United Nuclear Corporation resulted in the design and operation of a 14 MeV sealed beam neutron generator⁴⁴ for active assay of highly enriched uranium at UNC's Rhode Island Scrap Recovery Plant. The system was installed in a bunker-like structure located in the drum storage yard in the Scrap Recovery Plant.

Figure 5.4⁴⁴ shows a floor plan of the structure. Drums to be assayed are brought into the structure through a door on the south side. During assay, the drums are moved vertically by a 2-ton capacity elevator at about 1 fpm and rotated by a turntable at approximately 3 rpm. A low energy neutron shield consisting of natural boron carbide powder packaged in 16-gauge sheet metal is mounted on the elevator platform to prevent thermal neutron room return from reaching the assay sample.

A 14 MeV sealed beam neutron generator with neutron yields of $\sim 10^{10}$ n/sec was used for the active assay work. The generator produces neutron pulses approximately every 2 sec of about 1 sec duration, allowing for delayed neutron counting between pulses. Eighteen ³He proportional counters imbedded in polyethylene are used to detect delayed neutrons.

Fig. 5.4. Active assay shield structure - floor plan.

The counters are 1 in. in diameter and 20 in. in length. An electronic gating circuit is used to permit counting only between generator pulses.

Two fission chambers containing ^{235}U are used to monitor the drums for hydrogen. One is placed about 8 in. below the middle of the drum and the other at about the same distance above. Weighted responses from these chambers are averaged to produce a fission chamber response factor to establish calibration curves for the system.

For calibration of the instrument, 55-gal barrels were mocked up by preparing uranium samples containing 50, 500, 1500 and 2000 grams of ^{235}U (in uranium enriched to 97.0 w/o in ^{235}U). The samples were placed in plastic bags which were inserted in barrels. Water or a hydrogenous material was put into the barrels in various quantities.

As in the use of electron accelerators, the installation of a 14 MeV generator at a waste repository is not considered desirable due to the comparative complexity and expense involved. Future development work however, may make such a design a more attractive alternative for such applications.

5.9 Device for Passive ^{233}U Assay

A system for the passive NDA of ^{233}U waste in 55-gal drums by measurement of the ^{208}Tl 2.6 MeV gamma ray was constructed at Oak Ridge National Laboratory.⁴⁵ This system employs two 3 x 3 in. NaI scintillation detectors mounted in 2 in. thick cylindrical lead shields. The detector centers are located 1 in. above and below the center of the upper and lower half of the assay drum respectively. Drums are rotated at 1 rpm during assay. The detector housing is mounted on tracks to permit variation of the distance between the waste drum and the detectors. Counting is achieved with a multichannel analyzer. The technique depends on knowing the time since separation of the ^{232}U daughters from the ^{233}U . This time is used to establish a relationship between ^{233}U content and the ^{208}Tl gamma ray intensity.

The instrument was used to assay simulated waste drums containing from 8.42–31.39 grams of UO_2 in a foam glass matrix. Sample positions were varied. Accuracies from $\sim$ 1–5% were achieved for these

well-characterized samples. Results of measurements of 5 unknown drums are reported in Ref. 42. Precisions obtained in these measurements were all in the $\pm 6\%$ range (95% confidence interval).

This instrument represents a prototype design. It demonstrates the possibility of assaying ^{233}U waste by passive gamma NDA means. Its potential use in a waste repository is very limited, however, because of the requirement for knowing the process history of the contained ^{233}U .

6. CONCLUSIONS AND RECOMMENDATIONS

Based on the considerations discussed in the preceding Sects. 2 through 5, we conclude:

1. Calorimetric techniques are not applicable to the types of material and containers expected at a waste repository.
2. Assay of High Level and Cladding Waste canisters by non-destructive techniques to determine residual fissile material content is almost impossible using available technology. The probability of success for developing such techniques for routine application at a waste repository is very small.
3. Assay of intermediate level waste canisters for fissile content using passive gamma or neutron techniques is not feasible because of the high fission product gamma activity and neutron activity from the higher actinides.
4. Assay of Low Level Waste by passive gamma methods is possible but severely complicated by the high density and variability of the waste material and by the need to calibrate measured results against representative standards. For well characterized low level wastes, passive gamma techniques can be employed, but the possibility of unknown matrix inhomogeneities reduces confidence in such measurements.
5. Assay of Low Level Waste by passive neutron methods is possible, but complications due to the presence of (α, n) neutrons and uncertainties in plutonium isotopics relegates these methods to the "consistency check" level.
6. Active interrogation methods, using either gammas or neutrons, are the most promising techniques for quantitative determination of the fissile content of intermediate and low level waste containers. Highest accuracies (5-20%) may be achieved on LLW in 55-gal drums using particle accelerators to generate the interrogating radiation but these bring with them the attendant problems of high cost and operating and maintenance problems. Isotopic [^{252}Cf or (γ, n)] sources can be successfully employed to assay times to achieve these accuracies will be of the order of 10 min/drum.

Several different devices, tailored to specific waste types within each category, will probably be required to cover the spectrum of materials and activities in the low and intermediate waste categories. Development work will be required to implement such a system.

7. Attribute measurements, go-no go measurements and radiation signature measurements are simpler to make and are well within the range of existing technology. While specific equipment would have to be designed for these applications, we see no severe technical problems. These measurements would be made by passive or by a combination of active-passive techniques using isotopic sources.

In view of the above technical considerations, we recommend:

1. No consideration should be given or effort expended on attempts to assay the fissile content of high level and cladding waste containers. In the event that spent fuel elements are accepted for internment, these too should be accounted for by item control methods. If knowledge of the fissile contents of these waste categories is needed (for criticality control purposes, etc.), it should be obtained by sampling chemical analysis techniques prior to encapsulation or, in the case of spent fuel, from exposure history calculations.
2. If the decision is made to assay intermediate and low level waste, techniques should be selected from the active neutron method ranks and preference given to isotopic source driven devices. We do not recommend such a course of action, however. The difficulties anticipated plus the rather low level of accuracy which can be expected lead us to conclude that alternate methods for material control and accountability be instituted; specifically we recommend that item identification and control measures and tamper indicating procedures be relied on.* Responsibility for closing the material balance loop should rest with the shipper where the material is more accessible,

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* This topic is addressed in a comparison document.⁴⁶

better characterized, and amenable to sampling. Appropriate controls and procedures should also be instituted at that point to insure that the repository criteria for material form, content, etc., are complied with.

Appendix A

COMPUTER ANALYSIS OF GAMMA RADIATION FROM LLW FOR PASSIVE NDA

A computer simulation of the radiation emitted from a drum of LLW was performed to investigate the effect of SNM distribution and matrix density variation on the nondestructive assay of Low Level Waste by passive gamma methods. The XSDRNPM⁴⁷ one dimensional transport code was used. A container of LLW was mocked up in both cylindrical and spherical geometries. For the cylindrical case, the radius of the cylinder corresponded approximately to that of a 55-gal waste drum. For the spherical geometry, the radius of the container was about the same as that of a 55-gal drum. 17.2 grams of plutonium (assumed average actinide content of a drum of LLW) were distributed in the container. The gamma source from the plutonium was input as discrete energy lines using only the most prominent spectral lines as shown in Table 2.1. The waste matrix was assumed to be carbon for which two matrix densities were considered: 1 g/cc and 2 g/cc.

Three different distributions of the plutonium source were considered.

1. The source was homogeneously distributed over the container volume. (Subsequent results were normalized to results from this case for the 1 g/cc matrix density).
2. The source was located in the outer 0.5 cm of the spherical or cylindrical matrix.
3. The source was located in a center interval of 3 cm radius in the matrix. Also, in the spherical geometry, a case was considered in which the plutonium was placed in a small lump of pure PuO₂ in the center of the sphere. Only for this last case was the plutonium attenuation effect considered. For other cases the plutonium concentration was considered dilute enough to have a negligible effect on the matrix transmission properties.

Table 4.1 shows the relative transmission of the 414 keV gamma radiation from ²³⁹Pu as calculated close to the surface of the waste container. Results obtained for the two geometries are approximately equivalent. For both geometries it is seen that if the plutonium is

located near the center of the container, the transmission of the 414 keV ^{239}Pu line is reduced by a factor of about 10 for the 1 g/cc matrix and by ~200-300 for the 2 g/cc case. For the extreme case in which the plutonium is in a small lump in the center of the container (spherical), the relative transmission is reduced by a factor of ~1000.

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