*Low-Activity Solid Waste
Measurements at Tokai Works*

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LOW-ACTIVITY SOLID WASTE MEASUREMENTS AT TOKAI WORKS

by

J. L. Parker, D. H. Beddingfield, and H. O. Menlove

ABSTRACT

There is significant interest in performing assay measurements of containerized low-activity solid waste. We have examined the cases of typical waste drum matrices containing small quantities of plutonium and fission products. We have discussed various measurement techniques and considered the advantages and disadvantages of each method. We present a new state-of-the-art passive neutron waste drum counter with minimum detectable mass limits far below those systems which we have previously fabricated.

I. INTRODUCTION

A. Purpose and Objectives

This report fills the requirement to provide a feasibility study on the possibilities of using the methods of nondestructive assay (NDA) for measuring the small plutonium content of 200-L drums of low-activity solid waste (LASW) at the Tokai Reprocessing Plant (TRP). This report has been prepared under the Department of Energy Japan Nuclear Cycle Development Institute (DOE/JNC) cooperative safeguards agreement, Action Sheet 38. Consideration will be given to the strengths and weaknesses of all reasonably applicable methods along with the relative cost effectiveness of those possible methods. Both neutron and gamma-ray methods will be evaluated for waste streams containing fission products and streams free of fission products.

B. Waste Types to Be Considered

The request for this study indicated that there are three types of waste expected from the TRP. In each case, the objective is to measure the small mass of plutonium contained in 55-gallon (208-l) drums.

First, there are some essentially clean materials from the plutonium-handling region of the main process area and the analytical laboratory. They are stored in 208-L drums with walls of 1.2-mm-thick steel. Some of these drums contain 30 to 50 kg of plastic sheet and plastic gloves, others contain about the same mass of general combustibles (paper, etc.), and yet others contain plastic-wrapped sections of metal pipe. As shown in Fig. 1, the configuration of the materials stored in the drums falls into three cases.

Second, there are drums of the same materials as in the first category, but are lightly contaminated with plutonium, perhaps as much as 10 mg per drum, but are essentially free of fission products. Most, if not all, of the external radiation dose will be caused by the 60-keV gamma ray of the ^{241}Am daughter of ^{241}Pu . At a distance of 5 cm from the drum surface, the maximum dose rate is around 1000 $\mu\text{Sv/h}$ (100 mR/h). Again, Fig. 1 shows the configuration of the materials stored in the drums that fall into the same three cases.

Third, there are waste drums from the first separation cycle that contain intense fission product activity and perhaps a maximum of 20 μg of plutonium along with ^{244}Cm with a curium ratio (mass ^{244}Cm /mass Pu) of about 0.1 after plutonium separation. Waste from the head-end dissolver area has a curium ratio that is about 2 orders of magnitude lower than after the first separation stage for the fission products. The 208-L drums have 1.5-mm-thick walls and an additional shielding inside of 50 mm of concrete. Some have, in addition, a layer of lead inside the concrete shield to further reduce dose rates from gamma rays. Again, these drums are filled with vinyl sheets and gloves, or chunks of metals. Most of the external dose rate is assumed to be from the fission product ^{137}Cs and the maximum dose value is around 30000 $\mu\text{Sv/h}$ (3 R/h) at a distance of 5 cm from the surface of the drums. Figure 2 shows the configuration of the materials packed and the interior shielding.

Table I shows the isotopic distribution of the plutonium in the waste as computed by the ORIGEN code for standard fuel composition. Apparently, the americium is removed before the plutonium gets into our second waste category so that the only americium in the waste is produced by the decay of the ^{241}Pu .

Fig. 1. Packing configurations for the fission-product-free waste generated after the first separation cycle. The external radiation dose from this waste is essentially all from the decay of plutonium and its daughter ^{241}Am .

Table I. Initial isotopic distribution in weight percent of plutonium in waste as computed by the ORIGEN code for standard fuel composition.				
^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
1.3	65.5	22.4	8.8	2.1

SHIELDED WASTE DRUM CASES (HEAVY FISSION PRODUCTS)

Size: 565 mm o.d. x 462 mm i.d. x 880 mm high

Material: SGCC

Wall Thickness: 1.5 mm

Gross Weight: 250 - 300 kg

CASE 1:

50-mm-thick concrete shield on sides, 80-mm-thick concrete shielding on bottom. Filled with bags and bundles of vinyl sheet, gloves, and other combustibles. Double wrapped in vinyl sheet.

Surface Radiation Dose: ~ 10000 micro Sv/h (~ 1 R/h)

Concrete

CASE 2:

50-mm-thick concrete shielding on sides, 80-mm-thick concrete shielding on bottom. Lead shield inside of concrete. 100-L Polypropylene pack inside lead. Filled with pieces of pipe doubly wrapped in vinyl sheet.

Surface Radiation Dose: ~ 20000 micro Sv/h (~ 2 R/h)

Concrete

Lead

Polypropylene pack

Fig. 2. Packing configurations for the waste generated in the first separation cycle. This waste contains fission products and therefore requires the extra shielding indicated. Essentially, all of the external radiation dose is from fission products and transuranic elements such as curium.

II. GENERAL DISCUSSION: ACCURACY, PRECISION, AND MINIMUM DETECTABLE ACTIVITY

Before discussing measurement systems of potential worth in the nondestructive assay of low activity solid wastes (LASW), it is useful to discuss some of the important parameters that govern the possible accuracy, precision, and sensitivity of such low-level waste screening/measurement systems. Such a discussion is also appropriate because there are varying definitions and uses for the terms accuracy, precision and sensitivity. It is useful to clearly denote the definitions and uses adopted in this report, in case it will help clarify our thinking and facilitate comparisons among various methods and systems.

A. Accuracy

In this report, accuracy will refer to the nearness of the average of a large number of replicate measurements to truth, or at least the nearness of the average to an accepted value. It will have nothing at all to do with precision, thus differing from definitions that attempt to include both high precision and low systematic error. It will, therefore, be possible under this definition to have highly accurate results of very poor precision.

This approach is preferred because of the general desire among users of nondestructive assay equipment to speak of the "accuracy" of a given method or measurement system. The precision of a measurement can be good or bad depending on the measurement time used and of the activity of the measured package, as will be carefully explained below. If the term accuracy includes precision, then the accuracy of the method or system can be good or bad depending on measurement time and the magnitude of the measured activity.

As used in this report, accuracy will be synonymous with low systematic error, which in turn is synonymous with low bias. Systematic error is the terminology used to describe the behavior of a method or measurement system when it consistently gives results which are too high or too low in varying degree. Large fractional systematic error is clearly among the least desirable properties of an NDA measurement system.

In general, NDA measurement systems depend upon the detection of neutrons or photons (gamma rays, x-rays, or both). And sometimes larger systems employ both neutron and photon detectors. Some of the reasons for systematic error are the same for both neutron- and photon-based measurements, but others are quite different. We shall discuss briefly the most frequently encountered sources of systematic error for both types of measurement.

1. Calibration Error

Calibration error is a frequently encountered source of systematic error, both in neutron- and photon-based measurements. We shall not enter into an extended discussion of instrument calibration, but will make only a few important observations. First, we assert that appropriate physical standards, or computer models of physical standards, are required to accurately calibrate almost every NDA measurement system (there are one or two notable exceptions). That is, an adequate set of appropriate standards is a necessary condition to obtain a proper calibration of such systems. We should emphasize that

“appropriate” means similar to the packages to be assayed in every important way, but that does not necessarily mean identical to the unknown packages. Often, standards can be entirely appropriate and still vary in significant ways (such as size, shape, packaging, and exact chemical composition) from the packages to be measured. Unless waste is very carefully segregated, there will be large and significant variations between many of the packages to be measured and the calibration standards. A thorough understanding of the physics involved in the measurement is required in order to determine the range of package composition and other properties for which a given set of calibration standards is appropriate. Generally, the developers of the measurement method or the designers of the measurement equipment (sometimes the same people) are best qualified to specify the fabrication of calibration standards or to decide the range of applicability for existing sets of standards.

2. Sources of Error in Neutron-Based Measurements

There are several effects that can cause error in neutron-based assay. The effects are less severe (in general) in neutron coincidence counting than in simple totals neutron counting, and correction methods developed over the past decade or so allow the effects on coincidence counting results to be further reduced. The more significant effects which might create systematic error are self multiplication (not likely to be significant in low-level waste containing small amounts of plutonium), contaminants producing extra neutrons by (α, n) reactions, variable and unknown hydrogen content, unknown uranium in what is assumed to be plutonium-only samples, the presence of neutron poisons (such as boron and cadmium), and variable density of the material to be assayed. It goes without saying that waste packages, which by their intrinsic nature are heterogeneous and of variable composition and density, will suffer more systematic errors than will the measurement of uniform, homogeneous, chemically well-characterized materials. These error sources will be discussed in somewhat greater detail later in this report. The point we emphasize here is that if the waste material is segregated into reasonable categories whose nature and composition is known, that proper calibrations can be made and corrections applied so that for coincidence counting, at least, the possible errors are greatly reduced.

3. Sources of Error in Photon-Based Measurements

Two major factors influence the accuracy of photon-based measurements of activity in large waste containers (≥ 100 L). The first is the geometry problem. In order to achieve the high efficiency required to obtain high sensitivity, the detector, or detectors, usually have to be close to the waste containers thus causing serious $1/r^2$ problems. Ordinarily, assumptions are made concerning the uniformity of the distribution of activity within the boxes or drums in order to deal with the $1/r^2$ problem. Waste — being waste — is usually highly heterogeneous and the assumption of uniformity is rarely justified.

Equally troublesome, and often worse, is the absorption of photons within the waste container. This problem is, of course, at its worst at low energies such as those of the L x-rays ($\sim 10 - 25$ keV) upon which highly sensitive screening of LASW for plutonium and uranium most often depends. Even if the problem is just that of variable density packing of paper or other low-Z materials, the problem is significant. If containers have

finite-sized particles of metal or high-density compounds (oxides for example) of uranium or plutonium, the problem is seriously exacerbated, because many of the photons will never escape from the particle in which they were emitted.

Taken together, the two problems can often cause errors of factors of 2 or more. The 2-ft³ boxes (30 cm x 30 cm x 60 cm), often used for low-density clean LASW waste in the United States, present lesser problems than many larger containers, such as the ~ 200-L drums at the TRP, but are still significant. The relatively thick steel walls (1.2 mm or 1.5 mm) of the drums at TRP will make the use of the L x-rays of plutonium impossible, though the 60-keV gamma ray emitted by ²⁴¹Am will still be very useful for high-sensitivity screening of americium-bearing waste.

In summarizing the accuracy problems with photon-based NDA measurements of LASW, it can be said that detecting the photons that do escape from a waste container is straightforward, but finding out what fraction of the emitted photons escape from the container is very difficult indeed.

B. Precision

The term "precision" has, most fortunately, retained a well-defined and commonly understood meaning. Precision is a measure of the degree of scatter among many replicate, essentially identical measurements. In essentially all measurement systems, the scatter of measurements is well described by the well-known Normal (also called the Gaussian) statistical distribution. We will not give a detailed exposition of the Normal distribution and its applications here for many exist in the literature, and we assume that most readers of this report will be familiar with both the concept and the Normal distribution. If however, any of the readers of this report are not familiar with it, one of the best introductions to the distribution and its use in describing the precision of measurements made by counting randomly emitted particles (neutrons and/or photons), and one we highly recommend, is found in Glenn F. Knoll's book, "Radiation Detection and Measurement."¹ We will give here a very brief summary of a few important general statistical concepts and of the properties of the Normal distribution which are of direct interest to this report.

Let us assume that some physical quantity x is measured N times, with results $x_1, x_2, x_3, \dots, x_N$. For example, x could be the plutonium mass of a sample measured with a neutron coincidence counter, the uranium enrichment of a sample measured by a gamma-ray scintillation detector, the mass of a plutonium oxide sample measured by a balance, or the uranium concentration of a solution measured by a titration method. In general, with few if any exceptions, the best estimate of the true value of x that can be obtained from the N measurements is the average, or mean value,

$$\bar{x} = \sum_{i=1}^n x_i / N \quad . \quad (1)$$

In general, each individual measurement deviates from the mean, thereby being a distribution of measurement values. A common and very useful indicator of the magnitude of the scatter of the distribution is the standard deviation

$$\sigma(x) = \sqrt{\frac{\sum_{i=1}^N (x_i - \bar{x})^2}{N-1}} \text{ for } (N > 1) . \quad (2)$$

Note that these two definitions are general, not depending on the Normal, or any other, statistical distribution.

The estimated standard deviation is often given as the relative standard deviation (RSD) which is defined as

$$\sigma_r(x) = \sigma(x) / \bar{x} , \quad (3)$$

if the standard deviation has been estimated from N replicate measurements, or just as

$$\sigma_r(x) = \sigma(x) / x , \quad (4)$$

if the standard deviation has been estimated by propagation from the various counts in a single measurement made by counting the randomly emitted neutrons and/or photons from the sample. We will say more about this shortly — and we will see that the ability to do such a propagation does depend upon the radioactive decay obeying a particular statistical distribution.

It is usually assumed and is almost universally true, that replicate measurements of a physical quantity are distributed about the mean according to the Normal distribution. An example is given in Fig. 3, which is a histogram of the results of 500 measurements with a Normal distribution superimposed on the histogram. The mean value of the measurements is 107.3, and the standard deviation σ estimated from Eq. (2) is 2.43. The Normal distribution allows one to estimate the fraction of the measurements that should lie within a specified interval about the mean. Table II summarizes the estimated percentages in units of the standard deviation σ . The distribution of measurements shown in Fig. 3 is very close to these estimates, as will be true for all correctly operating measurement systems. In the example plotted in Fig. 3, there is a 68.27% probability that a measurement is within ± 2.43 of the mean value (hopefully near the true value), and that similarly, there is a 95.45% probability that a measurement is within ± 4.86 of the mean value of 107.3. The application is, of course, general. For any measurement based upon the counting of radioactive particles, there is a 68.3% probability that any single measurement is within one standard deviation of the mean of a very large number of replicate measurements, a 95.5 % probability that any measurement is within two standard deviations, and so on as calculated from the properties of the Normal distribution.

Fig. 3. A histogram of 500 measurements distributed about the mean value of 107.3, with a standard deviation of ~ 2.43 . The solid line is the best fit Gaussian shape having the same mean and standard deviation.

Table II. Percentage of measurements expected to be within $\pm\sigma$ of the mean of the normal distribution.	
Width of Region $x \pm w \cdot \sigma$	Estimated Percentage of Measurements in Region
$\pm 0.6745\sigma$	50.00%
$\pm 1.0000\sigma$	68.27%
$\pm 2.0000\sigma$	95.45%
$\pm 3.0000\sigma$	99.73%

It is most useful in accountability procedures to know the probable error in measured values, and the precision — as measured by the standard deviation σ — is usually a major part of that probable error. Measuring every package multiple times in order to be able to calculate a standard deviation from Eq. (2) is just too time consuming to be practical. Furthermore, it must be strongly emphasized that, in general, there is no such thing as the “precision of a method” or “the precision of a measurement system.” The precision of the

method or system will always depend on the measurement time, varying as the inverse square root of the measurement time as will be discussed more fully below. The precision will also depend strongly on the total activity in the measured package, with more activity usually, but not always, implying better precision. It is clearly most helpful if the precision of every measurement can be estimated from the various counts involved in calculating the measured quantity.

Fortunately, it is generally possible to obtain such estimates of the standard deviation from the counts involved in calculating the measured value. The radioactive decays giving rise to the particles detected by the detectors of NDA measurement systems almost always are correctly described by the Poisson statistical distribution. The important thing about that distribution for our present purpose is that the standard deviation $\sigma(x)$ of the actual total number of counts x is given by

$$\sigma(x) \approx \sqrt{x} \text{ and that obviously, } \sigma_r(x) \approx 1/\sqrt{x} \text{ .}$$

We emphasize that x is actual counts (and therefore is an integer) and not a count rate. These two very simple expressions are the key to obtaining expressions for the standard deviations of measurements from the same actual counts used in calculating the measured quantities. They also give an immediate feel for the minimum counts required to obtain a desired precision. Clearly 100 counts cannot give better than 10% precision, 10000 counts hope for 1% precision and 1,000,000 counts must be obtained to think about 0.1% precision. As we shall see, when results are a function of several separate counts, then even higher values in all the observed counts may well be required.

If a measured quantity w is a function $w(x,y,z)$ of three measured, *independent* quantities x , y , and z whose standard deviations $\sigma(x)$, $\sigma(y)$, and $\sigma(z)$ are known, then it can be shown that

$$\sigma(w) \equiv \left\{ \left[\frac{\partial w}{\partial x} \right]^2 [\sigma(x)^2] + \left[\frac{\partial w}{\partial y} \right]^2 [\sigma(y)^2] + \left[\frac{\partial w}{\partial z} \right]^2 [\sigma(z)^2] \right\}^{1/2} \text{ .} \quad (5)$$

The extension of the formula to more or fewer variables is obvious. If any of the measured variables are not mathematically independent of one another, other terms arise and the situation is much more complicated. It is wise for instrument developers to try to design instruments so that the measured variables are independent. Now let us use the equation to find the formulas for the standard deviation of several simple functions. Let k be a constant with no uncertainty and x and y be variables with uncertainties $\sigma(x)$ and $\sigma(y)$.

$$\text{Then, if } w = kx, \sigma(w) = k\sigma(x), \quad (6)$$

$$\text{if } w = x \pm y, \quad \sigma(w) = \sqrt{[\sigma(x)]^2 + [\sigma(y)]^2}, \quad (7)$$

$$\text{and if } w = xy \text{ or if } w = x/y, \quad \sigma_r(w) = \sqrt{\sigma_r(x)^2 + \sigma_r(y)^2}, \quad (8)$$

remembering that by definition $\sigma_r(x) = \sigma(x)/x$. Thus far, the expressions are general and the variables are not necessarily the counts from detectors. Now, however, if x and y are actual counts from detection of radioactive particles so that $\sigma(x) = \sqrt{x}$ and $\sigma(y) = \sqrt{y}$, then the above expressions become respectively,

$$\sigma(w) = k\sqrt{x}, \text{ for } w = kx, \quad (9)$$

$$\sigma(w) = k\sqrt{x + y}, \text{ for } w = x \pm y, \text{ and} \quad (10)$$

$$\sigma_r(w) = k\sqrt{\frac{1}{x} + \frac{1}{y}}, \text{ for } w = xy \text{ and for } w = x/y. \quad (11)$$

If the function w becomes even moderately complicated, the resultant expressions for $\sigma(w)$ will become exceedingly complex. Frequently, judicious approximations will be required to obtain expressions that are reasonably tractable, but it can usually be done. Such expressions can be tested against the formula of Eq. (2) and have been proven adequate to estimate the precision of the measurements from many diverse NDA systems. We should comment that the precision does not need to be known with great accuracy, two significant figures usually being adequate. Just as the measured values will vary within a group of replicate measurements, so will the estimated standard deviations also vary, but with a smaller relative standard deviation, not enough, in general, to be of any importance.

We should point out that the sample standard deviation from Eq. (2) also has a distribution, the relative standard deviation (RSD) of which is given by the approximate formula

$$RSD[\sigma(w)] \approx 1/\sqrt{2(N-1)}, \quad (12)$$

where N is the number of replicate measurements of w . The expression is good within about 10% for $N > 3$. For $N=15$, $RSD[\sigma(w)] = 19\%$ ($N=15$ is often recommended by statisticians in the United States). For $N=100$, $RSD[\sigma(w)] = 7.1\%$. Clearly the inverse square root dependence on the number of measurements makes it very difficult to get a highly precise value for the precision of a measurement by replicate measurements. Nevertheless, a useful quality control measurement for an NDA system is to occasionally compare the propagated standard deviation from that calculated from replicated

measurements by Eq. (2) If the replicate measurement value is much greater than the propagated value, some problem with the NDA system is indicated, assuming of course that the equation for the propagated standard deviation is correct.

We mentioned above that the precision of quantities calculated from the detection of radioactive particles emitted from the sample varies inversely as the square root of the measurement time. For a single count, this is nearly obvious. If we count a constant rate R for a time t , then the detected counts $N \approx Rt$ and $\sigma(N) \approx \sqrt{N} \approx \sqrt{Rt}$, from which we have immediately that $\sigma_r(N) \approx \sqrt{1/N} \approx \sqrt{1/Rt}$. Now we shall assert without proof that the precision of any quantity w calculated from the counts of radioactive emissions will also vary as the inverse square root of time, no matter how complicated a function w is of the measured counts. This is true in theory and has been demonstrated experimentally through many years of experience with NDA measurements. The important conclusion is that though one can, in theory, obtain any desired precision by counting long enough, the inverse square root dependence on counting time makes it prohibitively impractical to do so when rates are low and reasonable sample throughput is required. To some extent, low rates can be made higher by employing more and more efficient detectors, but at some point there cannot be enough counts obtained in a reasonable time to obtain satisfactory precision. A point will be reached when the small activity in a package might or might not be reliably detected. This brings up the subject of the minimum detectable activity and related concepts, which we discuss next.

C. Minimum Detectable Activity and Related Concepts

When attempting to perform sensitive screening of LASW (or of any low-level material) or to go on to at least semi-quantitative measurement of the activity content, the background rates in the detectors (or in particular Regions of Interest [ROI] within the energy spectrum for energy dispersive detectors) are the most important factors governing the possible sensitivities that might be obtained. Let us first attempt to define the term “sensitivity” as it is applied to NDA systems. This is almost necessary because of the many differing definitions (values for which have varied by at least a factor of 2) and terminology which have been used among those who do low-level counting of radioactive materials.

Frequently, sensitivity has been defined as “the activity where the detector response from a source is equal to three standard deviations of the background signal.” Activity is further used as total activity, that is, it includes all decays, including alpha, beta, or gamma emitting. This definition of sensitivity is useful, and often employed, but it does not explicitly address the possibilities of false alarms when nothing is present but background, or of a package with a real, finite LASW activity failing to exceed the designated alarm level.

Instead of “sensitivity,” let us here use the designations Minimum Detectable Activity (MDA) or Minimum Detectable Mass (MDM), the MDM rather obviously being the mass of a given nuclide which gives rise to the MDA. In defining them, we will have to introduce three other concepts, those of the Critical Level (L_c), Detection Limit (L_d), and Determination Limit (L_q) as will be seen in the following discussion. These concepts

were first clearly defined and used in an important 1958 paper by Currie.² They have been elucidated and reformulated by others, notably Lochamy.³

In any measurement of radioactivity by counting discrete events, it is necessary to set a rate — which when equaled or exceeded — is declared to indicate the presence of some excess activity in addition to all that which caused the observed background rates. This rate is the Critical Level (L_c). It is usually expressed as a net rate — that is, with the background rate subtracted — though that is not necessary. As normally used, the L_c is set low enough that occasionally the fluctuations in the pure background rate will cause a measured rate greater than the L_c and thus cause a “false alarm.” On the other hand, other-than-background activity levels which cause average rates greater than L_c — again because of statistical fluctuations — can sometimes give rise to measured rates less than L_c , and thus will fail to be detected. Based on the average background rates (usually from counts much longer than will be used for routine work), one can calculate the probability for any other rate to arise from a single, relatively short measurement of background. One therefore calculates the L_c based on the fraction of false detections of activity that one is willing to accept. Further, based on the average background rate and the agreed upon L_c , one can calculate the probability that any higher average rate will give rise to a measured rate less than L_c , and thus fail to be detected. One must decide what fraction of detection failures is acceptable and then calculate the corresponding other-than-background average rate. This rate, corresponding to the agreed upon acceptable failure-to-detect probability, is often called the Detection Limit (L_d). The MDA is proportional to the L_d , being the other-than-background activity which will give rise to the total average rate equal to L_d . The minimum detectable mass (MDM) is obviously the mass of a given nuclide which gives rise to the MDA. The MDA activity is almost always much greater than the L_d rate because one almost never detects all of the radioactive decays in a contaminant source. We emphasize here that the MDA usually varies for different radioactive nuclides because of differing probabilities for emission of the particular photons or neutrons upon which a given assay is based, and because the detector efficiency varies with energy. Thus, separate MDAs will usually have to be given for all radioactive nuclides of interest. The MDMs will vary even more because of the very different half lives and atoms-per-gram values for the various nuclides of interest. The relation of MDA to MDM is given by,

$$MDA = \left(\frac{\ln(2)}{t_{1/2}} \right) \left(\frac{N_0}{A} \right) MDM ,$$

where:

$t_{1/2}$ = half life,

N_0 = Avogadro's Number, and

A = the atomic mass number of the nuclide concerned.

Note that if the MDA is given in decays per second that $T_{1/2}$ must also be given in seconds.

Thus, there is implicit in every declaration of MDA an accepted probability for false detection and an accepted probability for failure to detect. In practice, in low level measurements, both probabilities are often set at 5%, though that is by no means necessary. It should be remarked that the frequently used definition of sensitivity — the average net rate equal to three standard deviations of the average background rate — is near the value obtained by assuming 5% for both error probabilities. Both the L_c and the L_d could be set so high that the probabilities for both false detection and failure to detect will become essentially zero, but this is rarely, if ever, done. It is the near universal view of technical people that it is most efficient and beneficial to accept finite values for both probabilities.

We now give Fig. 4 in order to make as clear as possible the important concepts of L_c and L_d . On the left side of the figure is the distribution of *net counts* for pure background. In this case the *average* count has been taken to 1000. Because the distribution is of net counts, the value of 1000 is subtracted from the short counts of background. Because some counts will be greater and some smaller than the average, the distribution of net counts will have both negative and positive values and will have the symmetric form of the normal distribution as shown. The line indicating the value of L_c is positioned, in this case, so that 5% of the area under the distribution curve is on the right of the line. Thus 5% of the time a measurement of pure background will result in a value greater than L_c , which has been declared to be the value which, if equaled or exceeded, indicates the presence of radiation in excess of background. Thus we have a 5% probability of a “false alarm” on pure background. In this case the value of L_c is 52.0 net counts. Note that although a count is always an integer, average counts may be fractional and therefore the value of net counts may also be fractional.

The Normal distribution centered about the line labeled L_d is the distribution of net counts whose *average value* is L_d and for which 5% of the area under the distribution curve lies on the left of the L_c value. Thus, 5% of the time a measurement of this net activity will yield a value less than L_c and will be declared to have no activity, corresponding to a 5% failure-to-detect probability. In the example of Fig. 4, $L_d = 106.7$. The values of false-alarm probability and failure-to-detect probability may be set as desired, and the curves in Fig. 4 will shift accordingly. Expressions for calculating the values of L_c and L_d will be given shortly. The curve centered about the line labeled L_q represents the measurements of an average activity whose measured RSD = 20%. More will be said about this shortly.

Fig. 4. An illustration to make clear the concepts of Critical Level (L_c), Detection Limit (L_d), and Determination Limit (L_q). The average background count is 1000. The accepted false-alarm probability and the accepted fail-to-detect probability are both 5%. The Relative Standard Deviation (RSD) for determining L_q is 20%. The values of L_c , L_d , and L_q are respectively 52.0, 106.7, and 171.1.

For some NDA systems there might be two or three MDAs (and obviously two or three MDMs) for a single radioactive nuclide based on different ROIs in the photon spectra from energy dispersive detectors. There may be separate MDAs for different detectors or combinations of detectors. If there are several nuclides being measured, it is quite possible that as many as 50 separate L_c values would be in use in screening operations. One would probably not want to set the L_c probability at 5% for all of them, for that would result in frequent false alarms. In such a situation it might be wise to set the L_c probability much lower, possibly near 0.1%, so that, overall, there might be only around a 5% probability of having a false alarm on any given box of "thought-to-be-clean" LASW.

Strictly speaking, radioactive decay is best described by the Poisson statistical distribution, but if the numbers of events detected are great enough (roughly 10 or greater), the Normal statistical distribution provides a sufficiently accurate description and the expressions are easier computationally. For most NDA systems (but not all) the rates will be high enough that the Normal distribution may be used. The required expressions are derived by Currie in absolute number form and by Lochamy in rate form, though Lochamy gives only expressions for equal false-alarm and failure-to-detect probabilities.^{2,3}

We will give some figures to provide some feeling for the functional behavior of L_c and L_d . These particular figures were originally generated as part of a feasibility study for an NDA system to screen thought-to-be-clean waste paper in cardboard boxes of 30 cm by 30 cm by 60 cm dimensions. It was assumed that the detectors would be energy dispersive and that many ROIs might be employed, so the L_c probability (the false alarm probability) has been set at 0.1%. Figures 5 and 6 give L_c and L_d as functions of the average background rate, which is assumed to have been measured in a count very much longer than the two minutes (120 seconds) assumed available for the measurements. Finally, the probability for failure-to-detect was taken as 5.0%. Figure 5 gives L_c and L_d for background rates from 0.1 to 50 counts/sec, while Fig. 6 shows values for background rates from 0.1 to only 10 counts/sec. Examination of the figures indicates how much improvement in L_d one gets for improvements in background rate. It is seen that in general the improvement is near, but not quite as good, as the inverse square root of rate behavior that one might intuitively expect. We might also intuitively expect that multiplying the count time by a factor F would have the same effect on L_c and L_d as dividing the background rate by the same factor. This expectation is true for L_c but not exactly so for L_d . It will be well to give the explicit, rate-based expressions for both L_c and L_d — as derived from the Normal statistical distribution — so that the behavior of L_c and L_d can be clearly understood.

$$L_c = K_\alpha \sqrt{\frac{R}{t}} \quad (13)$$

$$L_d = L_c + \frac{K_\beta^2}{2} \left(1 + \sqrt{\frac{1}{t^2} + \frac{4L_c}{K_\beta^2 t} + \frac{4L_c^2}{K_\alpha^2 K_\beta^2}} \right), \quad (14)$$

where

K_α = a constant (a multiple of the standard deviation of the background distribution) corresponding to the accepted probability for false alarm,

K_β = a constant (a multiple of the standard deviation of the net count distribution of average rate L_d) corresponding to the accepted probability for failure to detect,

R = the background rate measured in a long count, and

t = the count time for the measurements being done.

Fig. 5. The Critical Level (L_c) and the Detection Limit (L_d) as functions of the average background rate (s^{-1}) for rates up to $50 s^{-1}$. They are given as net (background subtracted) rates. Probability for false alarm is 0.001; failure-to-detect probability is 0.05; count time is 120 s.

Fig. 6. The Critical Level (L_c) and the Detection Limit (L_d) as functions of the average background rate (s^{-1}) for rates up to $10 s^{-1}$. They are given as net (background subtracted) rates. Probability for false alarm is 0.001; for failure-to-detect probability is 0.05; count time is 120 s.

We give in Table III values for K_α and K_β for some commonly used probabilities for either false alarm or failure to detect. Other values may be found in standard tables of the Normal distribution.

Table III. Commonly used values of K_α and K_β corresponding to selected probability values.	
Probability	K-Value
0.05	1.645
0.01	2.327
0.005	2.575
0.001	3.090
0.0005	3.290
0.0001	3.715

These expressions were used to compute the values in Figs. 5 and 6, as well as Figs. 7, 8, and 9 that explicitly show the behavior of L_c and L_d with counting time at fixed background rate. Figure 7, which gives L_c and L_d versus the reciprocal of the square root of time, shows that indeed L_c is proportional to the reciprocal of the square root of time, while L_d is nearly, but not quite so. Figures 8 and 9 make explicit the difficulties in keeping L_c and L_d low in the face of short count times. Really, of course, the difficulty has to do with small total count numbers independent of the time required to obtain them.

Fig. 7. The Critical Level (L_c) and the Detection Limit (L_d) versus $(1/T^{1/2})$ to illustrate behavior of L_c and L_d with time. Probability of false alarm = 0.001; failure-to-detect probability = 0.05. Average background rate = 10 s^{-1} . Count times from 1 to 100 s.

Fig. 8. Detection Limit (L_d), as a function of background rate for six different total count times. False alarm probability = 0.001; failure-to-detect probability = 0.05.

Fig. 9. Critical Level, (L_c), and Detection Limit (L_d) as a function of count time, average background rate constant at $20 s^{-1}$. Probability of false alarm = 0.001; probability of failure to detect = 0.05.

We see that at the lowest rates the concept of precision is not very useful. We speak instead of the MDA, with its associated probability for failure to detect as well as the probability for false alarm on pure background. As the average other-than-background rates increase, the probability for failure to detect becomes smaller and the concept of precision becomes more useful, the assays become at least semiquantitative, and then truly quantitative as the failure-to-detect probability becomes essentially zero.

The precision of measurements based on a single measured net rate depends upon both the background rates and the measured rate — as well as the measurement time. A useful concept defined in the literature is the Determination Limit (L_q), which is that average net count rate which will give rise to a specified relative standard deviation. The expression for L_q is

$$L_q = \frac{1}{2t(RSD)^2} \left(1 + \sqrt{1 + 4tR(RSD)^2} \right), \quad (15)$$

where

RSD = the desired relative standard deviation (or precision),

R = the background rate measured in a long count, and

t = the measurement count time.

In the example of Fig. 4, the value of L_q for RSD = 20% is shown along with the resulting distribution of net counts. We note that for this value of RSD the probability of measuring a value less than L_c is very small, the actual value being 0.025%. The value of L_d has been chosen so that the failure-to-detect probability is 5%. It is interesting to note that this corresponds to an RSD value of 31.2%.

In order to get some feeling for the relationship of L_c , L_d , and L_q for various choices of RSD, we present Fig. 10. It gives (as a function of average background rate) L_c and L_d (as before, false-alarm probability is 0.001 and failure-to-detect probability is 0.05), and L_q for RSDs of 20%, 10%, 5%, and 2%. We note that L_q for RSD=10% is only about twice L_d . Indeed, L_d corresponds to an RSD of about 23% for background rates about 1/sec. This means that if a given package yields a measured value with an estimated precision of 23%, in about 5% of replicate measurements the measured value will be less than L_c and will therefore be declared to be without measurable activity. The value of RSD to which L_d corresponds is different here than in the example of Fig. 4 because the false-alarm probabilities are different.

With accuracy and precision defined as they will be used in this report, and with the concepts of Critical Level (L_c), Detection Limit (L_d), and Determination Limit (L_q) defined as a means of understanding what is really meant by “sensitivity,” we are prepared to go on to the discussion of various possible measurement approaches.

Fig. 10. Critical Level, (L_c), Detection Limit (L_d), and Determination Limit (L_q) for RSDs of 10%, 5%, 3%, and 2% as function of average background rate. Probability for false alarm = 0.001; probability for failure to detect = 0.05. Count time is 120 s.

III. POSSIBLE MEASUREMENT APPROACHES

In about 1965, it was realized that a number of the standard methods used in experimental nuclear physics, along with the vast body of knowledge concerning the elements and all their isotopes, could be exploited to nondestructively measure a number of properties (mass, isotopic distribution, concentration) of special nuclear materials in a variety of forms and packaging. Serious, competent development efforts began about 1967. Since then, a number of groups of researchers around the world have invested thousands of man years of effort in finding, developing, and optimizing NDA methods for literally hundreds of applications. Several fundamental methods have been identified and vigorous development efforts have brought them to the status of mature technologies. Those fundamental methods have been, and still are being applied to a multitude of NDA measurement problems. The search for fundamentally different and new methods also continues.

After all the effort expended there are still two basic method categories, though each has a multitude of varied applications. Those two broad categories are, of course, the neutron-based methods and the photon-based methods. The two broad classes of methods depend respectively on the detection of neutrons and the detection of photons emitted from the materials being assayed. The neutrons and photons may be emitted spontaneously during the decay of radioactive nuclides or be induced by external radiation, usually neutrons but sometimes photons. Methods based on the detection of spontaneously emitted particles are usually referred to as passive methods, and those in which the particle emission is induced by external radiation are referred to as active methods. Possibly useful methods for the NDA of LASW from the Tokai reprocessing facility, which is the subject of this report, also fall into the two broad categories of neutron-based or photon-based methods.

The research and development of NDA methods has been well documented during the past thirty years. In the United States the Nuclear Regulatory Commission has commissioned a number of books on various aspects of NDA methods.^{4,5,6} Other books summarize the vast body of knowledge concerning the detectors that could be used.^{1,7} We will not try to reproduce here all that has been learned but only to summarize some basic data and to generally describe those methods which seem best adapted to measurements of plutonium in LASW. The high fission-product activity in a significant fraction of the expected waste is sufficient to exclude photon-based methods for that fraction. As such, we shall discuss neutron-based approaches first and then photon-based approaches. Finally, the possibility of systems that combine neutron and photon detection will be briefly discussed.

A. Neutron-Based Approaches

1. *Neutrons from Plutonium*

The neutrons useful in the NDA measurement of plutonium arise from spontaneous fission of the even-A isotopes of plutonium, from neutron-induced fission of plutonium isotopes, and from alpha-particle or proton reactions with a number of isotopes of low-Z elements. There is a great deal of information about all of these processes that is of use to

the designers of NDA methods and systems. We will give only enough information to enable the reader to follow the logic which makes some methods and procedures useful, and some of the useful ones preferable to others.

The spontaneous fission of plutonium, uranium, thorium, and some other heavy elements is an important source of neutrons that are useful in NDA. Without attempting to discuss the physics of spontaneous (or induced) fission we give Table IV which lists the spontaneous fission neutron yields (and other useful information) for fifteen nuclides of importance in the NDA of special nuclear materials. Note that this information only concerns the *prompt* neutrons from spontaneous fission, those emitted as part of the fission process in a very small fraction of a second ($\ll 10^{-6}$ s).

Perhaps a few comments about the information in the table are in order for those not familiar with the subject. All the listed nuclides decay both by alpha emission and by spontaneous fission. The total half-life is, of course, the observed half-life, the period in which half of all the nuclei decay, either by alpha emission or by fission (or in one or two cases, by beta decay). The spontaneous fission half-life is the time it would take for half of the nuclei to decay if spontaneous fission were the only decay mode. Because the decay constant of any decay mode (the probability of decay by that mode per unit time), $\lambda = (\ln 2)/(\text{half-life for that mode})$, it is easily seen that the ratio of fission decays to total decays is the ratio of *total* half-life to *fission* half-life. A quick examination of the ratios indicates that the fission decay fraction is $\leq 10^{-6}$ for all the listed nuclides except ^{252}Cf , for which it is 0.031.

The spontaneous fission multiplicity columns give the average number of neutrons per fission. We note that the multiplicity is different for induced fission than for spontaneous fission. It is also true that the multiplicity for induced fission depends somewhat on the energy of the inducing neutron, an example of the complexity of neutron interactions. The multiplicity distribution is roughly a Normal distribution, meaning that it is nearly symmetric. For all the listed nuclides except ^{252}Cf , there are rarely more than five or six neutrons emitted. The average energy of all the spontaneous fission neutrons is about 2 MeV, far greater than the nominal 0.025 eV of thermal neutrons (those in thermal equilibrium with their surroundings at an ambient temperature of 20 degrees Celsius). The energy distribution is the asymmetric Maxwellian distribution that has a high-energy tail allowing a small fraction of neutrons in the 8 to 10 MeV range. The multiplicity of the prompt fission neutrons is important because it allows the separation of the fission events from other neutrons producing reactions that result in only the emission of one neutron at a time. Generally, we refer to the detection of the fission events by the time correlation of the multiple neutrons as *coincidence counting*. This much used and very useful method will be discussed below as probably the most useful basic method to be applied to the NDA of LASW at the TRP.

Table IV. Spontaneous fission half-lives, multiplicities, and yields for 15 high-Z nuclides. ^a							
Isotope A	Number of Protons Z	Number of Neutrons N	Total Half-Life	Spontaneous Fission Half-Life (yr)	Spontaneous Fission Yield (n/s-g)	Spontaneous Fission Multiplicity $\bar{\nu}$	Induced Thermal Fission Multiplicity $\bar{\nu}$
²³² Th	90	142	1.41×10^{10} yr	$>1 \times 10^{21}$	$>6 \times 10^{-8}$	2.14	1.9
²³² U	93	149	71.7 yr	8×10^{13}	1.3	1.71	3.13
²³³ U	92	141	1.59×10^5 yr	1.2×10^{17}	8.6×10^{-4}	1.76	2.4
²³⁴ U	92	143	2.45×10^5 yr	2.1×10^{16}	5.02×10^{-3}	1.81	2.4
²³⁵ U	92	143	7.04×10^8 yr	3.5×10^{17}	2.99×10^{-4}	1.86	2.41
²³⁶ U	92	144	2.34×10^7 yr	1.95×10^{16}	5.49×10^{-3}	1.91	2.2
²³⁸ U	92	146	4.47×10^9 yr	8.20×10^{15}	1.36×10^{-2}	2.01	2.3
²³⁷ Np	93	144	2.14×10^6 yr	1.0×10^{18}	1.14×10^{-4}	2.05	2.70
²³⁸ Pu	94	144	87.74 yr	4.77×10^{10}	2.59×10^3	2.21	2.9
²³⁹ Pu	94	145	2.41×10^4 yr	5.48×10^{15}	2.18×10^{-2}	2.16	2.88
²⁴⁰ Pu	94	146	6.56×10^3 yr	1.16×10^{11}	1.02×10^3	2.16	2.8
²⁴¹ Pu	94	147	14.35 yr	2.5×10^{15}	5×10^{-2}	2.25	2.8
²⁴² Pu	94	148	3.76×10^5 yr	6.84×10^{10}	1.72×10^3	2.15	2.81
²⁴¹ Am	95	146	433.6 yr	1.05×10^{14}	1.18	3.22	3.09
²⁴² Cm	96	146	163 days	6.56×10^6	2.10×10^7	2.54	3.44
²⁴⁴ Cm	96	148	18.1 yr	1.35×10^7	1.08×10^7	2.72	3.46
²⁴⁹ Bk	97	152	320 days	1.90×10^9	1.0×10^5	3.40	3.7
²⁵² Cf	98	154	2.646 yr	85.5	2.34×10^{12}	3.757	4.06

^aAll information in Table IV taken from Table 11-1 of Ref 4.

Without discussing the nuclear physics involved, we note that the even isotopes of plutonium (those with even numbers of neutrons and of protons) have spontaneous fission yields thousands of times greater than any of the isotopes of uranium and thorium or of the odd isotopes of plutonium. These high fission yields are the basis for the numerous passive neutron NDA measurement systems. They will almost surely be the basis for possible systems of use at the Tokai facility. The two isotopes of curium, ²⁴²Cm and ²⁴⁴Cm, are seen to have neutron yields per gram about 10^4 times greater than the plutonium isotopes. This implies that microgram quantities of curium, if they are proportional (as they are thought to be) to the mass of plutonium in the high activity waste from the first separation cycle, would enable an indirect but very sensitive measurement of the very small masses of plutonium in that waste. Finally, we note the exceedingly high neutron yield of ²⁵²Cf, which has made it a very useful source of neutrons in active (those in which fission is induced) NDA measurement systems.

As mentioned above, there are delayed neutrons emitted in connection with the fission process. They are emitted by highly excited nuclides formed during the beta decay of the fission fragments. The excited nuclides decay instantly upon formation so that the delayed neutrons are emitted with the half-lives of their precursors. Though there are many fission fragment nuclides, all delayed neutrons can be categorized into six groups with decay half-lives in the range from 0.2 s to 55 s. Importantly, the delayed neutrons only constitute about 1% of the neutrons emitted so they are not a large perturbation on passive counting methods. We note also that the average energy of the delayed neutrons is only 300 to 600 keV, rather than the ~2 MeV of the prompt neutrons.

Delayed neutrons are exploited in active NDA systems in which the fission-inducing neutrons cannot be separated from the prompt fission neutrons, but the delayed neutrons can be detected after the stimulating source is turned off. The lower yield of delayed neutrons is made up by using a more intense inducing source. We note that the induced fission cross sections of the fissile plutonium isotopes (^{239}Pu and ^{241}Pu) are factors of two to three times as great as for the fertile isotopes (^{238}Pu , ^{240}Pu , and ^{242}Pu) which provide nearly all the prompt neutrons in spontaneous fission. This circumstance means that active NDA measurements are more nearly a measure of fissile content, and therefore their accuracy does not depend as strongly on accurate knowledge of the isotopic composition of the plutonium being measured as does the passive measurement of the less abundant even-even isotopes.

A final remark now about emissions from spontaneous fission is that both prompt and delayed gamma rays also occur. Their uses and the difficulties caused by them will be described later.

One other class of neutrons deserves a short discussion. Early in this century it was discovered that alpha particles emitted by some high-Z elements impinging on certain low-Z elements caused reactions in which neutrons were emitted. Two such examples are:

One suspects immediately that the fluorides and oxides of uranium and plutonium will emit some measurable fluxes of neutrons produced by this mechanism, and indeed they do. One would also expect that plutonium and uranium mixed with other low-Z elements might produce measurable neutron fluxes, and that is also true. The physics describing these so called (α, n) reactions is quite complicated. We shall qualitatively describe only a few aspects of it. The range of alpha particles in solid materials is very short. For the 4- to 6-MeV alpha particles emitted in the decay of uranium and plutonium isotopes, the range in full density oxide is ~0.005 cm. Thus, particle sizes must be small and the mixing intimate in order to get significant yields of (α, n) neutrons. The molecularly bonded compounds obviously fully meet the requirements for maximum production.

Table V gives information about (α ,n) yields in the fluorides and oxides of 18 high-Z nuclides including those of uranium and plutonium. A few comments about the table may be helpful. Note first that the yields from the fluorides are about 100 times as great as for the oxides, reflecting the greater cross sections for (α ,n) reactions in ^{19}F compared to ^{18}O . Secondly, note that the yields are inversely proportional to the half-lives of the nuclides. This is expected because the decay rate of any nuclide is given by $R = \lambda N$, where N is the number of atoms of the nuclide and the decay constant, $\lambda = (\ln 2)/(\text{half-life of the nuclide})$. As in Table IV, the fraction of alpha decay is given by the ratio of the total half-life to the alpha decay half-life. It is clear then that in all cases, save those of ^{241}Pu (where the alpha decays are only 0.0024% of the total), of ^{249}Bk (where the alpha decays are only 0.0014% of the total), and of ^{252}Cf (where the alpha decays are 97 % of the total) the alpha-decay fraction is very close to 100%.

Isotope A	Total Half-Life	Alpha Decay Half-Life	Alpha Yield (α /s-g)	Average Alpha Energy (MeV)	(α ,n) Yield in Oxide (n/s-g)	(α ,n) Yield in UF ₆ /PuF ₄ (n/s-g)
^{232}Th	1.41×10^{10} yr	1.41×10^{10} yr	4.1×10^3	4.00	2.2×10^{-5}	
^{232}U	71.7 yr	71.7 yr	8.0×10^{11}	5.30	4.49×10^4	2.6×10^6
^{233}U	1.59×10^5 yr	1.59×10^5 yr	3.5×10^8	4.82	4.8	7×10^2
^{234}U	2.45×10^5 yr	2.45×10^5 yr	2.3×10^8	4.76	3.0	5.8×10^2
^{235}U	7.04×10^8 yr	7.04×10^8 yr	7.9×10^4	4.40	7.1×10^{-4}	0.08
^{236}U	2.34×10^7 yr	2.34×10^7 yr	2.3×10^6	4.48	2.4×10^{-2}	2.9
^{238}U	4.47×10^9 yr	4.47×10^9 yr	1.2×10^4	4.18	8.3×10^{-5}	0.028
^{237}Np	2.14×10^6 yr	2.14×10^6 yr	2.6×10^7	4.77	3.4×10^{-1}	
^{238}Pu	87.74 yr	87.74 yr	6.4×10^{11}	5.49	1.34×10^4	2.2×10^6
^{239}Pu	2.41×10^4 yr	2.41×10^4 yr	2.3×10^9	5.15	3.81×10^1	5.6×10^3
^{240}Pu	6.56×10^3 yr	6.56×10^3 yr	8.4×10^9	5.15	1.41×10^2	2.1×10^4
^{241}Pu	14.35 yr	5.90×10^5 yr	9.4×10^7	4.89	1.3	1.7×10^2
^{242}Pu	3.76×10^5 yr	3.76×10^5 yr	1.4×10^8	4.90	2.0	2.7×10^2
^{241}Am	433.6 yr	433.6 yr	1.3×10^{11}	5.48	2.69×10^3	
^{242}Cm	163 days	163 days	1.2×10^{14}	6.10	3.76×10^6	
^{244}Cm	18.1 yr	18.1 yr	3.0×10^{12}	5.80	7.73×10^4	
^{249}Bk	320 days	6.1×10^4 yr	8.8×10^8	5.40	1.8×10^1	
^{252}Cf	2.646 yr	2.731 yr	1.9×10^{13}	6.11	6.0×10^5	

^a All information in Table V taken from Table 11-3 of Ref. 4.

One other aspect of (α ,n) neutrons has important consequences: they are randomly emitted with a multiplicity of one. This means that although they constitute an annoying

background for neutron coincidence counting, they are separated very effectively from the prompt fission neutrons whose multiplicities are almost all ≥ 2 for important nuclides. Because of this property, mixtures of various high-Z alpha emitters and low-Z elements such as oxygen, fluorine, beryllium, lithium, and boron make sources of single, random neutrons with varying intensities and energies which are useful in many ways, including some NDA applications.

2. *Passive Total Neutron Counting*

In total neutron counting all pulses from neutron interactions in the detectors are counted. No attempt is made to separate the high-multiplicity fission neutrons from the multiplicity-one neutrons from (α, n) or other sources. Such systems are simple and less costly than systems employing coincidence counting, but for many applications are not as accurate as the coincidence systems. This is particularly so for waste where uncontrolled composition can give rise to variable and unpredictable intensities of (α, n) neutrons and also variable and unpredictable intensities of neutron induced fissions. For well-characterized materials, the accuracy may be adequate and for some things such as samples of plutonium fluorides which have high (α, n) yields, the minimum detectable quantities can be low. Chapters 14 and 15 of reference 4 describe in extensive detail the principles of total neutron counting (including much about the interactions of neutrons in the many varied materials encountered) and also describe many useful detectors and systems which have made use of those principles in spite of the known limitations.

3. *Passive Neutron Coincidence Counting*

A few comments about the type of detector systems most often used in neutron coincidence counters are appropriate as an introduction to our discussion of coincidence counting. Most detection systems designed for the coincidence counting of prompt fission neutrons consist of a polyethylene sample cavity in which an appropriate number of cylindrical ^3He counter tubes are embedded. The detection probability for ^3He tubes is a monotonically decreasing function of energy and they have maximum sensitivity to thermal neutrons. Helium-3 detectors have a much higher sensitivity to neutrons than do other neutron detectors such as fission chambers or BF_3 counters. This higher efficiency is the key to the success of ^3He counters in coincidence counting applications. The polyethylene effectively moderates the ~ 2 MeV fission neutrons down to thermal energy. The detection is by the reaction

Because there is always only 765 keV of energy available to ionize the gas in the detector tube, the pulse-height spectrum has just one broad peak. No energy information about the original neutrons is available, but that limitation becomes an asset in high count rate environments. Gamma rays interacting in the gases give much smaller pulses than do neutrons so that at low rates virtually no gamma-ray-induced events are recorded at all. As the gamma-ray event-rate goes higher, however, the pulses start to pileup and eventually significantly perturb the neutron count. A great deal of effort has been

expended to increase the ability of ^3He tubes to operate in high gamma-ray environments. Current tube designs can operate in coincidence counters while exposed to gamma-ray doses as high as 100 R/h per active foot in extreme cases.¹¹ In typical systems, gamma-ray dose rates up to 20 R/h per foot of active detector length can be tolerated without significant degradation of the system performance. Higher dose rates require special design consideration. Inasmuch as the maximum expected gamma-ray dose for the LASW drums at TRP is about 3 R/h, a system based on ~1m active length ^3He detector tubes would perform optimally without requiring shielding to reduce the gamma-ray dose to the detector tubes. The addition of shielding, if required, would increase the rate of cosmic-ray induced background events that can be almost completely eliminated from the signal. Detectors explicitly designed for passive total neutron counting usually have one or more ^3He tubes in appropriate moderator assemblies but are not in a well (usually annular) configuration. There are also fast scintillator materials useful for neutron detectors but not frequently used in NDA systems. Much more information about neutron detectors is available in references 1 and 4.

As explained above, the prompt neutrons from spontaneous fission are emitted essentially simultaneously ($\ll 1 \times 10^{-6}$ s). For the plutonium isotopes, the average number of prompt neutrons emitted (the *multiplicity*) is ~2.2, the minimum number is zero for a fraction of a percent of the decays, and as many as six neutrons are emitted in a fraction of a percent of the decays. In order to be efficiently detected in the ^3He counters which have proven of most use, the ~2 MeV energy of the neutrons must be reduced to near thermal energies. This is accomplished as the neutrons undergo multiple scattering in carefully designed assemblies of moderating material (which is often polyethylene). Again, we will not take the space to attempt a detailed explanation of the rather complicated physical processes involved, but will refer those interested to chapters 12 and 13 of *Passive Nondestructive Assay of Nuclear Materials*,⁴ or similar sources. An important aspect of the process is that it takes rather long times. The simultaneously emitted neutrons become separated in time and their detections may be many microseconds apart. The average neutron population in the detector assembly decays in an approximately exponential fashion. Depending on the purpose of the detector, the time constant (usually called the “die-away time”) of the neutrons may be as long as 100 μs or as short as 20 μs . Thus the “coincidence” of the detection is not nearly as “tight” as we are accustomed to in many nuclear physics experiments. However, over 25 years of research into the detailed physics of the process and equally intense development of equipment to detect and extract the maximum amount of information available from the pulse train from the detectors have resulted in an impressive ability to detect the correlated bursts of neutrons and separate them from singly-emitted neutrons. The accuracy of coincidence instruments is thus little influenced by the chemical form of the plutonium, which can result in the emission of variable numbers of multiplicity-one neutrons from (α, n) reactions, or by variable background rates of multiplicity-one neutrons.

As seen in the data of Table IV, almost all of the contributions of prompt fission neutrons from plutonium come from the even isotopes ^{238}Pu , ^{240}Pu , and ^{242}Pu . Even though ^{238}Pu and ^{242}Pu have higher neutron yields per gram per unit time, the higher

isotopic fraction of ^{240}Pu in most materials to be measured usually causes the ^{240}Pu to dominate the detected neutron signal. Because of this, it has become convenient and customary to define an effective ^{240}Pu mass for coincidence counting by

$$^{240}\text{Pu}_{\text{eff}} = 2.52 \text{ }^{238}\text{Pu} + ^{240}\text{Pu} + 1.68 \text{ }^{242}\text{Pu}. \quad (16)$$

This quantity is the mass of ^{240}Pu that would give the same coincidence response as that obtained from the sum of the ^{238}Pu , ^{240}Pu , and ^{242}Pu isotopic fractions in the actual item being measured. The odd isotopes of plutonium are not considered in $^{240}\text{Pu}_{\text{eff}}$ because their spontaneous fission rates are negligible. The coefficients in the expression are determined primarily by the relative neutron yield, and in a minor way, by the differences in multiplicity and by details of detection circuitry. The coefficients given are those for the modern shift register circuits developed at Los Alamos National Laboratory, but would change only slightly for other commonly used circuits. Depending on the isotopic composition of the plutonium material being measured, the effective mass is 2% to 20% higher than the actual mass of ^{240}Pu . And, of course, the ^{240}Pu mass is usually $\leq 25\%$ of the plutonium total mass, with the ^{238}Pu and ^{242}Pu fractions being much smaller. These facts emphasize the necessity of accurately knowing the isotopic distribution of the plutonium being measured in order to obtain results for the total plutonium mass.

Now although coincidence counting by itself is usually more accurate than just total neutron counting, the total count can often provide information that complements the coincidence count. The circuitry of modern coincidence systems automatically acquires the total rate along with two-fold and often three-fold coincidences. For a wide range of material types the analysis algorithms make use of both the coincidence and the total neutron data.

As carefully explained in Section II-C, one of the most important parameters governing the minimum detectable activity (MDA) of most NDA systems is the background rates. Passive coincidence counting is no exception. Background coincidence rates are greatly reduced by adequate shielding and proper location. Usually there remains a component of background caused by cosmic ray spallation in or near the detector. As an example of this, it was once observed that 77 kg of lead in a coincidence counter produced the same coincidence rate as 0.7 g of plutonium (20% ^{240}Pu) at the high elevation of Los Alamos.¹² Because cosmic-ray-induced neutron production is much more likely in high-Z than in lower-Z elements, one method of reducing background is to minimize the use of heavier metals (such as cadmium and iron) in the detector body. Modern coincidence systems for drums will probably use more than 100 ^3He tubes in an annular or square array surrounding the drums. By considering the very high multiplicity of the spallation neutron events, the rate of cosmic-ray coincidences in the background may be still further reduced. This is the basis of the truncated multiplicity (TM) approach of background reduction.

In recent years there have been significant advances in low-activity waste counters. These advances have, for the most part, been fueled by the economics of waste disposal. For passive neutron detector systems, the results of this R&D effort have been to

significantly reduce the MDM of plutonium in containerized wastes. The advances that have been realized extend to optimized physics design, software improvements, and innovative electronic configurations^{13,14,15}. The result of these advances had been to reduce MDM values for passive neutron systems to remarkably low values.

The coincidence detection efficiency is proportional to the square of the single-neutron detection efficiency. Recent coincidence counter designs in sizes that can accept 200-L drums have efficiencies up to 40% for single neutrons. The high-efficiency, optimized electronics and the various background reduction efforts combine to give quantitative measurements down to about 1 mg of ²⁴⁰Pu. The add-a-source matrix correction procedure and multiplicity analysis greatly reduce measurement bias in the expected heterogeneous LASW drums. We finally note that there is a small component of cosmic-ray-induced background coincidence events which is non-Poisson in its nature and which places a difficult-to-remove limit on the possible sensitivity that can be achieved.

In the waste from the first separation cycle at TRP, the mass of ²⁴⁴Cm is about 1%-10% of the mass of plutonium. It is important to notice that the spontaneous fission yield of ²⁴⁴Cm is 10,000 times as great as that of ²⁴⁰Pu. This means that the neutron output is dominated by the ²⁴⁴Cm. Even though the multiplicity of the curium decay is somewhat higher than that for the decay of the plutonium isotopes, the decays from the two sources cannot be differentiated. The sensitivity for ²⁴⁴Cm, however, is very high, being less than 1 ng in a 200-L drum. As long as the curium/plutonium ratio is near constant (or can be determined with adequate frequency on a batch basis), the curium measurement provides an indirect but very sensitive measure of plutonium. Because the curium/plutonium ratio is near 0.1, the sensitivity for plutonium would be near 1 ng with only uncertainty in the ratio contributing significant uncertainty in the value. Because the high level of fission product gamma rays prevents any gamma-ray-based assay of this waste, this indirect measure of plutonium by the passive neutron counting of the curium spontaneous fission neutron emission seems to be the best possibility for determining the plutonium content in this waste from the first separation cycle.

Neutron-based assay systems have the advantage of maintaining a uniform systems response over an extremely wide range of sample matrix types and densities. This fact is related to the high penetrability of neutrons when compared to any other radiation signature that might be exploited in an NDA system.

Table VI shows the change in the efficiency of a waste drum assay system (WDAS) system from the matrix materials in the drums at Plutonium Fuel Production Facility (PFPPF). The wide variety of matrix types and the insensitivity of the system are impressive. Current drum counter designs have a decreased matrix sensitivity. The totals ratio to the empty case corresponds to the efficiency change. We see that most of the materials have less than a 10% change in efficiencies. Figure 11 shows a plot of the totals data where the rates have been normalized to unity for an empty drum.

Table VI. Totals Neutron Counting for 200-L Drums.

Matrix	Mass (kg)	Waste Totals/Empty Totals
Empty	0	1.00
Paper	54	1.01
Steel Pipes, Valves	66	0.99
PVC & Rubber Gloves	26	0.94
Borated Glass	173	0.96
Wood	51	1.00
Concrete Rubble	220	1.04
Iron Pieces	196	1.05
Polyethylene Tubes	33	0.94

Fig. 11. Totals neutron count rate normalized to an empty drum for various waste matrices.

In a recent adaptation of an old idea, the “Add-a-Source” (AAS) scheme is used to make corrections for matrix-induced variability in detector response. As we use the term here, “matrix” means everything in a package except the plutonium being measured. In this scheme, after the purely passive measurement, a small ^{252}Cf source (the source intensity is about 10^5 n/s) is introduced near the drum. The perturbation of the ^{252}Cf coincidence rate is used to determine a matrix correction for the sample. The measurement errors introduced from matrix materials in 200-L drums have been reduced by an order of magnitude by the use of this technique. It should also be noted that the technique can also detect the presence of neutron shielding materials inside of a drum.

4. Active Neutron-Based Methods

There are two active neutron-based methods that should be discussed briefly. They are the shuffler-type instruments and the differential die-away-type instruments. Both are much more costly and complex than the passive neutron methods discussed above and any advantages will probably not be worth the extra cost. There is a third active-type system, namely the active-well coincidence counter, but its sensitivity can never be good enough to be a candidate for the measurement of the Tokai LASW. Detailed information on each of these systems is provided in Ref. 4.

Shuffler-type instruments generally use a ^{252}Cf neutron source to induce fissions and then detect delayed neutrons from the fission. The source is rapidly moved by a flexible cable from a shield where the detectors are protected from the source neutrons to positions near the package to be assayed. After a few seconds of irradiation, the source is rapidly moved back to a shielded location. The delayed neutrons are detected for another period of several seconds. Remember that the delayed neutron groups have half-lives of from about 6 s to 55 s. This irradiate-measure cycle is repeated until the precision is satisfactory or until the maximum allowable measurement time is reached. A measurement cycle typically occupies about a total of 10 s. This procedure is required, of course, because the prompt fission neutrons from the sample are usually only a small fraction of the neutrons from the ^{252}Cf source, especially for small masses of fissile material ($\ll 1$ g). Also, the ^3He tubes of the detector may be actually rendered useless during the irradiation because of exceedingly high count rates.

We note that, as mentioned above, if high sensitivity is not required, a relatively small AmLi source may be placed in a fixed position to irradiate the sample. The coincidence events from the induced fission can be separated successfully from the increased singles background caused by the (α, n) source, thus allowing active measurements of fissile quantities greater than about a gram. This is called the active-well coincidence method. The active-well coincidence method is not useful for sample volumes greater than about 30 liters.

Because the fission cross sections are much higher for the fissile isotopes (^{239}Pu , ^{241}Pu , ^{235}U , ^{233}U), shufflers and active-well coincidence counters measure approximately the sum of fissile mass in the packages. There are, of course, many situations where that is just what is desired. The shuffler method is now well developed and there are many systems in the field, often specifically customized to the specific problems. We note that any system that can do the shuffler-type measurement can also do a passive measurement, though the system is optimized for the active shuffler measurement of fissile material. Such hybrid systems are often used. The passive neutron coincidence method described above, in which the AAS source is used in a shuffler mode to give an active assay of fissile material, is another hybrid procedure, though in that case the system is optimized for the passive measurement.

Shuffler systems for many assay requirements are the optimum solution. However, if not actually required, they would not be used because of complexity and cost. The ^{252}Cf source is, of itself, not complex, but it is expensive, and because it cannot be turned off, there is a requirement for permanent and substantial shielding, both to protect personnel

from excessive radiation doses and to protect the system detectors from the intense neutron flux. The flexible cable mechanisms used to "shuffle" the source between the shield and the irradiation locations are also somewhat complex and must be of very high reliability. Thus the shuffler is not likely to be the best choice for measuring the plutonium in the TRP LASW drums.

The differential die-away systems generally use a sealed-tube-type pulsed accelerator to generate bursts of 14-MeV neutrons by the reaction ${}^2_1D + {}^3_1T \rightarrow {}^4_2He + {}^1_0n$. In these accelerators, the deuterons are accelerated onto a tritium target. The high-energy neutrons slow down to near thermal energies in the package to be assayed and the lining of the assay chamber, during which time the detectors recover from the paralysis induced by the intense burst of interrogating neutrons. The thermal or near-thermal neutrons then induce fission in the fissile content and the fast fission neutrons are detected as the assay signal. The moderated interrogating neutrons are kept from the detectors by layers of material (often cadmium) with very high cross sections for absorbing thermal neutrons. The actual detectors are usually the same 3He tubes used in passive neutron coincidence systems.

Differential die-away time systems have good sensitivity because the interrogation is by means of thermal neutrons, the cross sections for absorption followed by fission being very high in ${}^{239}Pu$ and ${}^{235}U$. On the other hand, accuracy is a difficult problem because of the rapidly changing value of the absorption probabilities at near-thermal energies. Additionally, the use of the accelerator for producing the neutrons increases the cost and complexity of the system greatly. Because of the complexity, cost, and accuracy problems, the differential die-away-type systems are probably not the most advantageous for the Tokai reprocessing facility unless there was a requirement for measurements (though not of high accuracy) of quantities of ${}^{235}U$. However, if the sample contains ${}^{235}U$, the active measurement does not provide the ${}^{239}Pu$ content directly.

B. Photon-Based Approaches

1. Photons from Plutonium

Both gamma rays and x-rays accompany the decay of all the plutonium isotopes of interest (${}^{238}Pu$, ${}^{239}Pu$, ${}^{240}Pu$, ${}^{241}Pu$, ${}^{242}Pu$). We note in passing that x-rays and gamma rays are identical in nature and respond in identical ways in detectors. The distinction between them is based only on their mode of production. Gamma rays are emitted from excited nuclei while x-rays are emitted from excited atomic states. For many purposes, this is a distinction without a significant difference and we shall often prefer the generic term photons. Among the plutonium isotopes, there are wide differences in the number of unique-energy gamma rays emitted and in the energies and yields of those that are emitted. Frequently, those who have worked to exploit the gamma rays of plutonium (and uranium too, for that matter) have wished that they had been better endowed with useful photon emissions. Nevertheless, there are a number of very useful photon-based NDA methods in use by which the isotopic ratios of isotopes are measured, by which element concentrations are measured, and by which masses of various isotopes may be measured quantitatively.

As with neutrons, there is a large body of knowledge concerning the photons from plutonium and their interactions in matter. And again, we will not attempt to give an overview of all of it, but will discuss only a few basic matters which will make the advantages and disadvantages of photon-based assays more understandable.

First, we shall draw a few distinctions between neutrons and photons and their detectors. Spontaneous fission neutrons are emitted with a wide spectrum of energies, and the detectors usually employed do not preserve any energy information. It is therefore difficult to definitively conclude from measurements what nuclides are emitting the observed neutrons. Photons, on the other hand, have unique energies and if high-resolution detectors are used, the emitting nuclides can usually be identified, even if quantitative measurement is difficult or impossible. We give Table VII that gives the energies and yields for some of the more useful gamma rays of plutonium and uranium isotopes, along with their mean free paths in representative high-Z and low-Z materials. We just remark that ^{239}Pu emits hundreds of unique gamma rays over the range from about 30 keV to about 800 keV though many are, for a variety of reasons, not very useful. Plutonium-241 and its daughters emit several tens of gamma rays, while ^{238}Pu and ^{240}Pu emit only a few gamma rays. It is seen that the yields (gamma rays/s-g) are in some cases greater than for the prompt neutrons from spontaneous fission. This does not mean, however, that photons will usually give a more sensitive assay. Gamma-ray detectors are usually relatively small. In many cases less than 1% of the emitted photons will enter the detector and be registered at full energy, while the neutron-based systems usually have "near-four-pi" geometry with efficiencies up to 40%. The result is that neutron-based instruments will often have lower (that is, better) MDAs. There are some photon detectors (usually low-Z plastic scintillators) that do not have adequate energy resolution to identify the emitting nuclides. They are nevertheless useful in some situations.

The decay of the plutonium isotopes is also accompanied by the emission of x-rays, which can also for some problems be used to advantage. Table VIII gives the energies and yields of both K-x-rays and L-x-rays for the plutonium isotopes. We note that the average yields (photons/g-s) of L-x-rays for the plutonium isotopes are high, being emitted in about 10% of the decays. For some circumstances, this relatively high yield makes possible highly sensitive screening operations.

Table VII. Energies and yields for some of the more useful gamma rays from plutonium and uranium isotopes; also some mean free path data.^f

Nuclide	Energy (keV)	Activity (γ/g-s)	Mean Free Path ^a (mm) (High-Z, High-ρ) ^b	Mean Free Path ^a (mm) (Low-Z, High-ρ) ^c
²³⁴ U	120.9	9.35×10^4	0.23	69
²³⁵ U	143.8	8.40×10^3	0.36	73
	185.7	4.32×10^4	0.69	80
²³⁸ U	766.4 ^d	2.57×10^1	10.0	139
	1001.0 ^d	7.34×10^1	13.3	159
²³⁸ Pu	152.7	5.90×10^6	0.40	75
	766.4	1.387×10^5	9.5	139
²³⁹ Pu	129.3	1.436×10^5	0.27	71
	413.7	3.416×10^4	3.7	106
²⁴⁰ Pu	45.2	3.8×10^6	0.07	25
	160.3	3.37×10^4	0.45	76
	642.5	1.044×10^3	7.4	127
²⁴¹ Pu	148.6	7.15×10^6	0.37	74
	208.0 ^e	2.041×10^7	0.86	83
²⁴¹ Am	59.5	4.54×10^{10}	0.14	38
	125.3	5.16×10^6	0.26	70

^aMean Free Path is thickness of absorber which reduces gamma-ray intensity to 1/e.

^bUranium (Z=92) oxide with $\rho = 10 \text{ g/cm}^3$.

^cAluminum (Z=13) oxide with $\rho = 1 \text{ g/cm}^3$.

^dFrom the ²³⁸U daughter ^{234m}Pa. Equilibrium assumed (which requires ~ 100 days).

^eFrom the ²⁴¹Pu daughter ²³⁷U. Equilibrium assumed (which requires ~25 days).

^fInformation in Table VII is from Table 1-2 of Ref. 4. This reference itself references many other useful sources of data.

Table VIII. Some energies, yields, and mean free paths for some low energy photons from ^{239}Pu , ^{240}Pu , & ^{241}Am .

Nuclide	Photon ^a	Energy ^b (keV)	Yield (photons/g*s)	Mean Free Path in Cellulose ^c (g/cm ²)
^{239}Pu	U-K _{α1}	98.44	1.57×10^5	6.1
^{239}Pu	U-K _{α2}	94.66	0.97×10^5	6.0
^{239}Pu	U-L _γ	20.3	1.33×10^7	~1.7
^{239}Pu	U-L _β	17.1	5.24×10^7	~1.0
^{239}Pu	U-L _α	13.6	3.79×10^7	~0.7
^{240}Pu	U-K _{α1}	98.44	8.4×10^3	6.1
^{240}Pu	U-K _{α2}	94.66	5.7×10^3	6.0
^{240}Pu	U-L _γ	20.3	1.1×10^8	~1.7
^{240}Pu	U-L _β	17.1	4.2×10^8	~1.0
^{240}Pu	U-L _α	13.6	3.1×10^8	~0.7
^{241}Am	Np-K _{α1}	101.1	2.50×10^6	6.2
^{241}Am	Np-K _{α2}	97.1	1.55×10^6	6.1
^{241}Am	Gamma	59.536	4.56×10^{10}	5.2
^{241}Am	Np-L _γ	22.0	0.60×10^{10}	~1.7
^{241}Am	Np-L _β	17.5	2.34×10^{10}	~1.0
^{241}Am	Np-L _α	13.9	1.65×10^{10}	~0.7

^a All designations such as U-L_α imply x-rays, this example indicating the L-alpha x-ray of uranium. Remember that the alpha decay of plutonium isotopes results in uranium x-rays, and the alpha decay of americium results in neptunium x-rays.

^b All K-x-ray energies are the weighted average of two components with near equal energies. All L-x-ray energies are the weighted average of six to ten components in an approximately 2-keV energy range.

^c Cellulose is the basic component of paper. Multiplying these values by the actual spatial density of waste paper in a container will give the average mean free path in centimeters.

Neutrons more easily penetrate high-Z, high-density materials such as iron, lead, and tungsten, while being more highly attenuated by lower-Z materials of greater moderating power. Conversely, photons are much more strongly attenuated by higher-Z materials than by lower-Z materials. We give Fig. 12 that plots the mass attenuation coefficients of a range of elements versus energy for energies in the range from 10 keV to 1000 keV. We see that from about 100 keV to 400 keV, the attenuation coefficients for higher-Z elements are much greater than for lower-Z elements, which explains the differences in mean free paths noted in Table VI.

The basic law of photon attenuation is exponential and is given by

$$F = e^{-\mu \rho x},$$

where F is the fraction of photons that penetrate a layer of material of thickness x, of density ρ and mass attenuation coefficient μ without absorption and without inelastic

scattering. This fraction is often referred to as the gamma-ray transmission. The mass attenuation coefficients for all the elements are known with accuracies of ~1% over the very great range from ~1 eV to 100,000,000 eV. The coefficients for compounds and composite materials are easily found by combining the coefficients for the elements according to simple rules.

Two important things are evident from a study of Fig. 12. The first is that plutonium (and other high-Z elements) is very good at absorbing its own emitted photons. That means that self shielding will be a problem if there are finite-sized lumps of high-Z, high-density material in the waste. The second important consideration is that in packages as large as 200-L drums the attenuation of emitted photons by the matrix can be very significant, especially if assays are based on photons of ≤ 400 keV which includes most of the useful gamma rays from plutonium. The consequence is that sensitivity is reduced and large corrections must be made for the attenuation of the emitted photons within the packages. For relatively clean waste (without significant activity from fission products), there are procedures for making the required corrections. For over twenty years, approximate corrections have been made by measuring the transmission (fraction of gamma rays passing through the container as given above) through the container with an external gamma-ray source of appropriate energy from which is computed the fraction of gamma rays escaping from the container. This procedure was carried out in a segmented way, as the assay of a set of somewhat overlapping segments roughly defined by a collimator. The operator was required to assume uniformity and homogeneity within each segment, an assumption that was rarely fully justified in heterogeneous waste, and accuracy suffered accordingly. More modern systems essentially do much more detailed active/passive tomographic scans of barrels of clean waste and obtain much more accurate results.

There is a final, very important consideration that will preclude the use of photon-based NDA systems for much, if not all of the fission-product-containing waste from the first separation cycle at the TRP. This is the fact that the photon detectors see the photons from fission products just as well as those from plutonium. It will be very difficult, if not impossible, even when using the highest resolution detectors, to do a quantitative assay of small amounts of plutonium (let ~1 g of plutonium be regarded as small) if the fission-product, gamma-ray dose at the surface of the drums is above, perhaps, 10 or 20 mR/h. At higher dose rates, the presence of the plutonium is completely concealed by the overwhelming radiation from the fission products.

The information upon which this feasibility report is based indicates that the fission-product-free, low-activity drums generated after the first separation cycle probably have dose rates of ≤ 100 mR/h at the surface. This radiation dose is, however, primarily from the decay of the ^{241}Am daughter of ^{241}Pu , and does not prevent photon-based measurements of the plutonium. Unfortunately, as seen from Table VII, the specific yields for the higher energy gamma rays from all of the isotopes are so low that the possible sensitivities are on the order of one gram of plutonium in containers as large as the 208-L drums. However, the emission rate for the 59.5-keV gamma ray of ^{241}Am is 4.5×10^{10} $\gamma/\text{g/s}$, more than 10^5 times greater than for any of the higher-energy gamma

rays of the plutonium isotopes. If the americium has been separated from the plutonium, and the time since separation is known, then the plutonium/americium ratio can easily be calculated and measurement of the ^{241}Am by means of the 59.5 keV gamma ray becomes a sensitive, indirect way to measure the plutonium. For plutonium (with the isotopic distribution given in Table I) that has been stored for ten years, extrapolation from the performance of a photon-based system built at LANL shows that the sensitivity of this indirect measurement for plutonium would be about 20 ng for low density, low-Z materials. After twenty years in storage, the sensitivity would be about 10 ng of plutonium. This scheme to obtain an indirect measurement of the plutonium in the waste generated after the first separation cycle is obviously very similar to the curium-based method suggested above to obtain a sensitive, although indirect, measurement of the plutonium in the waste from the first separation cycle. However, because the penetrability of the 59.5-keV γ -ray is low, the signal source will be limited to only the surface layer.

Fig. 11. The mass attenuation coefficients for a variety of elements — from hydrogen to plutonium — as a function of energy, given as the usual log-log plot. The potential difficulties with photon-based measurements of variable density, low-Z waste at low energies are evident.

If there is waste that is known to be clean (paper from the facility, for example) that, has only an exceedingly small probability of being contaminated, but that needs to be

confirmed clean, then the same photon-based method used for the indirect measurement of plutonium may be used effectively to verify its cleanliness at levels that may well permit inexpensive disposal. This will be discussed at greater length below.

2. *Passive Low Resolution Systems*

There is quite a large class of NDA systems designed to screen waste for low levels of fission product activities. They generally consist of measurement chambers, often large enough for 200-L drums, surrounded (or nearly surrounded) by sheets of plastic scintillation material. Because of the "near-four-pi" geometry, the sensitivity is quite good but the plastic scintillation detectors have essentially no energy resolution and there is no way to separate the plutonium photon events from fission-product photon events. This whole class of systems is therefore not at all suitable for measuring the plutonium in the LASW drums at Tokai. As mentioned, they are useful for screening waste for low levels of fission product activities, and perhaps doing semi-quantitative measurements of it. Poor efficiency for the 59.5-keV gamma ray along with the lack of resolution eliminate this type of system for consideration for use in the photon-based, indirect method of measuring plutonium discussed above. We will therefore give no further consideration to systems of this type.

3. *Passive Medium-to-High Resolution, Exploiting Higher-Energy Photons*

For many years, good quality, reasonably accurate, photon-based assays of ^{239}Pu have been done using medium-resolution sodium-iodide scintillation detectors as well as high-resolution germanium gamma-ray detectors. These have been based primarily upon the 413.7-keV gamma ray of ^{239}Pu which is emitted at a rate of $\sim 3.4 \times 10^4 \text{ s}^{-1}\text{-g}^{-1}$. The penetrability of the photons at $\sim 400 \text{ keV}$ is reasonably good and even with heterogeneously packed 200-L drums, accuracies within 10% of "truth" have generally been possible. Corrections for the attenuation within drums are based on the fraction of gamma rays from an external source that pass through the drum along a diameter (see Chapter 6 of reference 4). The corrections required an assumption of uniformity and homogeneity — at least within reasonably well-defined, horizontal segments — that is not really justified in heterogeneous waste. The assays using the high-resolution germanium detectors are generally more accurate and less influenced by small intensities of interfering gamma rays. Successful use of the sodium iodide scintillation detectors generally requires a constant and known isotopic composition of the plutonium involved.

In recent years much more complete and sophisticated scanning has been done employing extensions of the analysis methods used with standard industrial x-ray tomographic procedures. These modern methods have much improved the accuracy of passive gamma-ray assays. Unfortunately, the sensitivity is still not very good, being around one gram of ^{239}Pu , and the material must be at least "almost clean" with respect to fission-product gamma rays. As a result, such systems will not be appropriate for the specified Tokai waste, impressive as their performance is on heterogeneous but fission-product-free waste at levels of one gram or more.

4. *Passive Medium-to-High Resolution, Exploiting Lower-Energy Photons*

As has been briefly described above, an NDA procedure has been developed during the past few years at the Los Alamos National Laboratory (LANL) that may be of use at the TRP in dealing with the fission-product-free waste generated after the first separation cycle. The method was developed when it was realized that LANL was burying — at very high expense — in the Radioactive Waste Landfill, many cubic meters of clean but “suspect” waste. This clean waste was mostly just paper or plastics from the clean areas of facilities which handled and processed radioactive materials and was “suspect” only because it originated in those facilities. It was realized that if that waste could be verified to be “clean” with sufficient sensitivity then it could be disposed of in the public landfill for about \$10 /m³ rather than for approximately \$1500 /m³ at the Radioactive Waste Landfill. The procedure is used in two modes at LANL. In the first mode, the scrap paper is spread on a moving belt without packaging and is examined by an array of six phoswich detectors. In this mode the system is sensitive to all radioactive nuclides except low energy beta emitters.⁸ In the second mode the waste is packaged in 115-L plastic drums which are viewed by an array of three well-shielded phoswich detectors.^{9,10} In this second mode, the system is sensitive to essentially all photon emitters but not to beta emitters because the beta particles are almost all absorbed by the box. As examples of the MDAs achievable with the procedure (in 1000 s assay times), we cite the following: 20 nCi (0.3 μg) for ²³⁹Pu, 1 nCi (0.3 ng) for ²⁴¹Am, 1 nCi (0.5 mg) for ²³⁵U, and 3 nCi for ¹³⁷Cs.

Because of potential usefulness, we will briefly describe this procedure in modest detail, so that the basis for its performance can be understood. The key to the sensitivity of this procedure is the use of phoswich detectors to detect the more intensely emitted low-energy (~15- to 25-keV) L-x-rays of plutonium and uranium (along with the very intense 59.5 keV-gamma ray of ²⁴¹Am). Phoswich detectors are sandwiches of two different kinds of scintillation phosphors viewed by a single photomultiplier tube. Those used in the LANL systems consist of 3 mm of NaI, backed by 50 mm of CsI. The open face of the NaI crystal is covered with a thin window of beryllium or aluminum in order to admit photons down to ~ 10 keV with little loss. There is a 1.25-cm-thick light pipe — usually of quartz — between the CsI crystal and the face of the photomultiplier tube. All the crystals and the light pipe are 12.7 cm in diameter, as is the photomultiplier.

The usefulness of this combination of scintillators depends upon the fact that the light generated in the NaI crystal decays with a time constant of ~ 0.23 μs while the light generated in the CsI decays with a time constant of ~ 1.0 μs. Because of this difference, pulses that are generated solely in the NaI can be separated quite cleanly from those generated solely in the CsI or that have contributions from both crystals. Events in which a photon Compton scatters from the CsI back into the NaI are a major source of the low-energy background spectrum of the NaI crystal. Cosmic ray muons also cause events with light in both crystals. By doing an electronic pulse shape analysis, almost all of the events with light from both crystals can be eliminated from the NaI spectrum. This results in a decrease in background rates of between a factor of 4 to 5 compared to an equivalent NaI crystal with an inert light pipe behind it — a combination which has been much used in the past. As explained earlier, reduction of background rates is most important to

achieving low MDAs. The phoswich detectors used in the LANL systems are not experimental devices but have seen considerable use for many years for in vivo detection of lung burdens of plutonium, americium or uranium of workers in nuclear facilities.

The phoswich detectors offer a second advantage if it is desired to detect photons over a wide energy range, for example from the ~10-keV L-x-rays of plutonium to the 1332.5-keV gamma rays emitted by ^{60}Co . The problem arises because of the difficulty in having sufficient data storage channels in the multichannel analyzers to accurately show the low-energy structure of the photon spectra. In the LANL systems, the thin NaI detectors that detect the low-energy part of the spectrum record only up to about 400 keV. The CsI crystals give only about half as much light output per unit energy input as does NaI. Combined with some electronic shaping, the CsI detectors view an energy range about 2.5 times that of the NaI detector — up to ~1800 keV — in the same number of data channels used for the low-energy spectra. The output of such detectors is separated into sodium iodide and cesium spectra that can be separately analyzed. The two spectra from each of the phoswich detectors (for however many are used) are easily stored separately in modern analyzer systems.

A system similar to that mentioned above for screening 115-L plastic drums of combustible waste would be suitable for performing the ^{241}Am -based indirect measurement of plutonium discussed briefly in Section III-B-3. The 1.2-mm-thick walls of the 208-L drums would completely eliminate the 10-25 keV L-x-rays of the plutonium and americium isotopes but about 20% of the 50.5-keV gamma rays from the americium would penetrate. This escape fraction would suffice to give a Minimum Detectable Mass (MDM) of about 1 ng for ^{241}Am in measurements of about 1000 s. After 10 years decay, the plutonium/americium ratio is about 20:1, therefore the MDM for plutonium would be about 20 ng, as stated above. Because the ^{241}Pu decays into ^{241}Am with a half-life of about 14.4 years, after about 20 years, the plutonium/americium ratio would be approaching 10:1 and the MDM for plutonium would be approaching 10 ng. We comment that the sensitivity of the ^{241}Am -based measurement for plutonium becomes more sensitive with time because americium builds up with little loss because of the 432-year, half-life of the ^{241}Am . On the other hand the ^{244}Cm -based indirect measurement for plutonium in the first-separation cycle wastes becomes somewhat less sensitive with time because of the 18.1-year, half-life of the ^{244}Cm , though not enough to be of any practical importance.

We finally remark that the “open belt” version of the system we have been describing is thought to provide the best available verification that clean paper and other thin combustibles really are free of radioactive contamination. This, combined with a reasonable throughput, may make the procedure useful in a number of facilities in reducing waste disposal costs.

C. Combination Neutron and Photon-Based Approaches

Some few NDA systems have been constructed and used which combine the use of neutrons and photons in interesting ways. These systems have been too complex, too expensive, or not appropriate for the problem of measuring small amounts of plutonium

in the waste at TRP. We include a few comments on the subject mainly to assure the staff at TRP that essentially every reasonable possible combination of methods has been considered and tested during the past 35 years.

Quite early (in the late 1960s), small linear accelerators were used to produce intense beams of Bremstrahlung photons by which photofission was induced in fissile materials inside waste containers. The resultant fission neutrons, either prompt, delayed, or both, were detected as a measure of the amount of fissile material. Reversing this procedure several years later, successful fuel-rod scanning devices have been built which used ^{252}Cf -neutron sources to induce fission in the fuel as it was drawn quite rapidly through the irradiation assembly. The uniformity of the loading of the fuel pellets was checked by detecting the delayed-fission gamma rays as the irradiated fuel rods passed through the detector assembly several seconds later.

Other major NDA systems have had (and still do have) both photon-based and neutron-based components. The separate components are, however, just parts of an integrated system and could operate independently. As an example of such schemes, a neutron-based method measures the fissile content of the material and a gamma-ray spectrometer device provides the isotopic distribution of the plutonium as well as, quite frequently, the plutonium/uranium ratio.

IV. RECOMMENDED BEST APPROACHES

As the approach best suited and most cost effective for measuring the plutonium in both the waste from the first separation cycle and from the waste generated after the first separation cycle, we recommend a *state-of-the-art passive neutron coincidence counting system*. This system is similar to the WDAS design, but employs many recent advances in system design. We call this design the *Super-WDAS*. By the indirect ^{244}Cm -ratio based measurement, such a system can attain an MDM of about 1 ng of plutonium in the waste from the first separation cycle, and a directly measured MDM of about 5 mg of plutonium in the waste generated following the first separation cycle. If a lower MDM is required for the waste generated following the first separation cycle, we recommend a medium-resolution photon-based system detecting the 59.5-keV gamma ray of ^{241}Am to do an indirect measurement of the plutonium. Such a system should have an MDM of about 10 ng of plutonium for low density, low-Z waste if it has been 20 years or more since the americium was separated from the plutonium. However, the performance of a photon-based system would be very poor for high density and/or high-Z matrices. We will now discuss each of the two recommended approaches in detail.

A. State-of-the-Art Passive Neutron Coincidence System

1. MDM and Applicability

For the indirect measurement of plutonium in the waste from the first separation cycle by the ^{244}Cm -based method, the Determination Limit for a Relative Standard Deviation (RSD) of 20% is about 1 ng of plutonium in measurement times of 1000 s. The exact value of the MDM will depend strongly upon the background rate in the system, which in turn, will depend upon how well it is shielded (an economic as well as a technical decision), upon the use of "low background" materials in the counter, and on the use of the best software for elimination of cosmic-ray-generated coincidences. In Fig. 13 we give as nanograms of ^{244}Cm , the Critical Level, the Detection Limit — which is the MDM in this case because it is given as nuclide mass — and the Determination Limit for RSD = 20% versus the total coincidence background rate. The conversion from curium mass to plutonium mass will depend upon the plutonium-to-curium ratio in the waste, which, in the information provided as the basis for this report, was assumed to be 10:1.

For the direct measurement of plutonium in the waste generated after the first separation cycle, the MDM is about 1 mg of ^{240}Pu equivalent, which for the ^{240}Pu isotopic fraction of about 24%, translates into about 3 mg of plutonium, again in measurement times of about 1000 s. The exact same considerations on the influence of the background rates apply here as for the waste from the first separation cycle. In Fig. 14, the Critical Level, the Detection Limit (again equal to the MDM), and the Determination Limit for RSD = 20% are given in milligrams for ^{240}Pu .

Fig. 13. For a state-of-the-art passive neutron coincidence counter, we give the Critical Level (L_c), Minimum Detectable Mass, (MDM), and the Determination Limit for $RSD=20\%$, all expressed as ng of ^{244}Cm . The false-alarm probability and the failure-to-detect probability are both 5%. The count time is 1000 s. The plutonium-to-curium ratio is assumed to be constant and must be determined by chemical means. In the preliminary estimates of plutonium masses in waste drums the ratio was assumed to be $\sim 10:1$.

Fig. 14. For a state-of-the-art passive neutron coincidence counter, we give the Critical Level (L_c), Minimum Detectable Mass (MDM), and the Determination Limit for $RSD = 20\%$ (L_q), all expressed as mg of ^{240}Pu . The count time is 1000 s.

In order to understand the actual coincidence rates corresponding to the values given in Figs. 13 and 14 in mass units, we give Fig. 15, which again presents the Critical Level, the Detection Limit, and the $RSD = 20\%$ Determination Limit, but as net real coincidence count rates. These net rates were calculated using the Normal statistical distribution and consideration of the effects caused by the time correlation of the fission neutrons. The

time correlations require a correction to the expressions that arise from the assumption that all the neutron events are randomly distributed. Based on much experimental work, a multiplicative correction factor of 1.3 has been applied to the values obtained by assuming a random distribution of neutrons.

Fig. 15. For neutron coincidence counters, we give the Critical Level (L_c), Detection Limit (L_d), and Determination Limit for RSD = 20% (L_q), all as net rates. The count time is 1000 s. Note that the values depend only mildly on the detector efficiency, the variation in the correction for the effects of time correlation in the detection of the spontaneous fission neutrons causing the only difference.

In converting the calculated net rates to equivalent masses of ^{244}Cm and ^{240}Pu , one must know the specific fission rates and the multiplicity distributions for the two nuclides as well as the detector efficiency and the necessary parameters of the shift-register circuitry. The specific fission rates and the multiplicity distributions are tabulated in standard references. For the results presented here, the detector is assumed to have a singles efficiency of 40%, a die-away time of 60×10^{-6} s, a pre-delay time of 1.5 μs , and a gate length of 128 μs . In both cases, the assay count time has been taken as 1000 s. Because of uncertainties in the correlation correction factor and because the final detector parameters may vary somewhat from those assumed, the overall difference between the values presented here and those achieved by the final system may be as much as 25%. However, the value of the background coincidence rate will still have the greatest influence on the performance achieved, and this is accurately portrayed in Figs. 13, 14, and 15. Coincidence systems similar to the size required at TRC have been constructed with doubles background rates in the vicinity of 0.1 s^{-1} . It is thought that a well designed system for use at TRC will not have background rates higher than that.

For the case of a Super-WDAS, we can predict the MDM on the basis of the anticipated performance parameters. The sensitivity for measuring plutonium is a function of the detection efficiency, the neutron die-away time, the room background, and the cosmic-ray spallation neutron background. For the Super-WDAS, the use of 10 atm

^3He tubes and the shortened neutron die-away time significantly improve the performance compared with the WDAS. The MDM of plutonium can be estimated if we define MDM as being the mass of plutonium that gives a signal equal to 3σ above the background. The MDM can be calculated from

$$MDM = \frac{3(1.3)}{a} \sqrt{\frac{B + A + a(MDM)}{t}}, \quad (17)$$

where

a = the response constant = 108 cps/g (see Table VIII),

B = doubles background $\cong 0.009$ cps,

A = accidental rate = $T^2G \cong 0.0104$ cps,

T = totals rate $\cong 9$ cps,

G = coincidence gate = 128 μs ,

t = measurement time = 1000 s, and

(1.3) = correlation error factor (unitless).

The above equation is simplified by the assumption that the background B is measured for a time interval that is much longer than t .

For the Super-WDAS (shielded building) and a measurement time of 1000 s, we obtain

$$MDM = \frac{3(1.3)}{108} \sqrt{\frac{0.009 + 0.0104 + 108(MDM)}{1000}}$$

$$\begin{aligned} MDM &= 0.00025 \text{ g } ^{240}\text{Pu} \\ &= 0.00076 \text{ g Pu (33\% } ^{240}\text{Pu}_{\text{eff}}) \end{aligned}$$

Thus, the MDM equals approximately 0.76 mg plutonium for a Super-WDAS-type detector located in a concrete-shielded building. Figure 16 shows the MDM as a function of cosmic-ray spallation doubles background rate for Super-WDAS. The doubles rate has been reduced by using recently developed advanced-background reduction methods that reject both large and small cosmic-ray spallation neutron counts.

If the drum is measured for 1 h, the MDM reduces to ~ 0.40 mg Pu. However, if the drum contains combustibles with a high hydrogen content, the counting efficiency will be reduced and the MDM will increase in proportion to the efficiency change of the system.

We have developed advanced methods for cosmic-ray neutron background reduction. Table IX gives a summary of the effects of detector design advances and the application of new background reduction techniques. The new methods reduce the neutron background by an order of magnitude.

Fig. 16. Trend in LLD resulting from the cosmic-ray reduction in moving from Los Alamos (elev. 2200m) to a concrete building at Tokai (sea level). The detector is assumed to be 40% efficient and the coincidence gate is set at 128 μ s.

TABLE IX. Performance and Background Estimates for WDAS and Super-WDAS.

Parameter	WDAS ^a	Super-WDAS ^b
Efficiency	18%	43%
Number of tubes	64	114
Calibration constant (counts/s g ²⁴⁰ Pu)	18	108
Totals bkg. (sea level) (count/s)	4	9
Doubles bkg. (sea level) (counts/s)	0.015	0.009
Totals bkg. (sea level basement) (counts/s)	2	4
Doubles bkg. (sea level basement) (counts/s)	0.006	0.003
MDM for 1000-s count (g ²⁴⁰ Pu)	8.3×10^{-4}	2.5×10^{-4}
MDM for 1000-s count (g Pu)	25×10^{-4}	7.6×10^{-4}

^aRates from WDAS units at PFPF and PWTF.

^bRates extrapolated from WDAS including reductions from TM and LCV methods.

2. Hardware

The Super-WDAS is a coincidence counter with a measurement cavity large enough to accommodate a 208-L drum, having a singles efficiency of about 43%. Background coincidence events will be reduced as much as feasible by eliminating heavy metals such as iron and cadmium from the body of the detector and by optimized shielding. The system makes use of add-a-source functionality when there is a need for significant matrix correction. The electronics will be of the most advanced shift register type that

allows for minimal effective die-away time and for long effective determinations of the background rates. Appropriate provision for placing the drums — which will have masses up to 500 kg — into and removing them from the counter will be provided.

The conceptual design of the Super-WDAS is shown in Fig. 17. The new design greatly improves the sensitivity of WDAS type systems for the measurement of plutonium in drums and boxes. The advanced passive neutron system uses more ^3He tubes with 10 atm gas pressure compared with the original 4 atm tubes in the WDAS. The new tubes give a higher efficiency and a shorter neutron die-away time. The tubes are arranged to minimize matrix sensitivity to a wide variety of sample compositions. The conceptual system has an efficiency of 43% and an MDM below 1 mg of plutonium for a shielded installation at sea level.

Fig 17. Diagram of conceptual passive neutron drum counter.

3. Software

Software routines are available that control calibration routines, measurement quality control routines, and the sample measurement routines. The analysis software INCC (NCC WIN) code provides this capability.

4. Calibration Standards and Measurement Control Standards

Appropriate standards, both for calibration and for regular measurement quality control, are necessary adjuncts of the system. It is probably best that these be fabricated at TRP in consultation with the development staff at LANL.

The standards will probably comprise a two-component system. The first component will be one or more 208-L drums packed with appropriate matrix materials and with provision for placing small capsules containing plutonium or ^{252}Cf at the desired locations within the volume of the drum. The second component is obviously the actual capsules containing the plutonium and/or ^{252}Cf . Again, the range of plutonium or ^{252}Cf masses to be used, the number of standards required, and their active material masses are best determined in consultation with the system developers.

The MDMs are computed from the parameters of the counting system and the observed background rates. If it is desired to demonstrate that the calculated values are correct, attempting to make very small value standards to demonstrate MDM should wait until after the system is in place and the actual MDM has been computed. One can fabricate a standard with the appropriate mass of material, either plutonium or ^{252}Cf , and then see if the system fails to detect it the appropriate fraction of the time. An alternate demonstration is to make standards at the Determination Limit for a selected RSD value (perhaps 20%) and then experimentally demonstrate with replicate measurements that the RSD of the measurements is nearly equal to the selected value. If this were true, then there would be high confidence that the MDMs were as calculated.

5. *Sample Matrix Effects in Neutron Drum Counters*

Neutron-based assay systems are highly regarded for their intrinsic insensitivity to matrix composition. A wide variety of matrix materials and densities can be assayed without the need for special calibration. The types of matrices that can be accurately assayed without special standards can be increased by employing add-a-source functionality to an assay system. It is desirable to optimize a neutron counter to reduce the matrix sensitivity of the system.

In the present case, the types of matrices that are typically encountered are reasonably well defined. The matrices at Tokai will be either hydrocarbons or iron/hydrocarbon mixes as well as the concrete and lead shielding. The Super-WDAS neutron assay system has been optimized to minimize the sensitivity to these matrices over as broad a range of loadings as possible.

JNC provided information regarding six typical storage drum and matrix configurations. This information was shown in Figs. 1 and 2. Each of these typical cases is summarized in Table X. We examined the effects of the drum and matrix on the optimized Super-WDAS passive neutron counting system described above. The matrix types can be generalized into two types: hydrocarbon and iron/hydrogen mix. Each of the matrix types in the appropriate drum types was simulated using the MCNP program. The results of these simulations will be presented in this section.

Table X. Summary of drum and matrix types of interest at Tokai Works.				
Case Number	Drum Type	Matrix Type	Matrix Mass (kg)	Matrix β - γ activity (μ Sv/hr)
1	Normal 208-L SGCC with 8, 15-L poly packs	Vinyl Sheet and Gloves	~50-70	~200-1,000
2	Normal 208-L SGCC	Vinyl Sheet and Flammable Wastes	~50-70	~200-1,000
3	Normal 208-L SGCC with 200-L poly pack	Vinyl Wrapped Metal Pipes	~50-70	~200-1,000
4	Normal 208-L SGCC	Vinyl Wrapped Metal Pipes	~50-70	~200-1,000
5	Normal 208-L SGCC with concrete shield	Vinyl Sheet, Gloves	~80-130	~10,000
6	Normal 208-L SGCC with concrete shield, Pb shield, and 100-L poly pack.	Vinyl Wrapped Metal Pipes	~80-130	~20,000

Before considering the performance of the Super-WDAS design for the various drum/matrix cases of Table X, it is instructive to examine the effect of the various drum types with no waste matrix. The system performance for the various empty drum types for totals and doubles counting is shown in Fig. 18.

Fig. 18. Effect of empty drum types on Super-WDAS performance.

From Fig. 18, it is clear that the type of drum does not significantly affect the performance of the assay system. The small effect of the concrete-lined drums is due to the neutron moderation in the concrete. The use of the add-a-source method would remove any possible bias introduced by the drum itself.

It is useful to note the two shielded drum cases in Fig. 4 would prove nearly impossible to assay using a gamma-ray system. The concrete-liner would attenuate the gamma rays so severely that a reliable assay measurement would become impossible in a reasonable time period. The addition of a lead-liner to the drum, while having no effect on the neutron counting, would further aggravate the gamma-ray attenuation problem.

Case 1. Normal 208-L drum with eight 15-L poly packs filled with vinyl sheet and gloves.

This case can be simulated by modeling a standard 208-L drum and a polyethylene matrix at various densities. A graph of the behavior of the Super-WDAS with increasing polyethylene loading is shown in Figs. 19 and 20. The effect of the poly packs is shown in these figures. The hydrogenous content of the waste adds to this effect. Accommodating the effects of increasing moderation is most difficult aspect of design optimization of neutron drum counters. The need to minimize the effect of hydrogenous loading and the effects of metallic matrices work in opposition to one another. For the Super-WDAS, the system response to increasing polyethylene loading was minimized while constrained by the other matrix types.

The matrix effect on the neutron measurement is dominated by the hydrogen density in the matrix. The figures from the MCNP calculations are presented in units of kg of polyethylene. However, the hydrogen density for hydrogenous waste is typically a factor of ~2 less than for polyethylene. For example, 70 kg of paper gives about the same neutron perturbation as 30 kg of polyethylene.

Fig. 19. Behavior of Super-WDAS system efficiency with hydrocarbon loading.

Fig. 20. Trend in the doubles count rate per gram $^{240}\text{Pu}_{\text{eff}}$ for a 64 μs coincidence gate.

Case 2. Normal 208-L drum with vinyl sheet and flammable waste.

This case can be simulated by modeling a standard 208-L drum and a polyethylene matrix at various densities just as in Case 1. Figures 19 and 20 show the system behavior, but the poly pack mass offset shown in the plots does not apply in this case.

Case 3. Normal 208-L drum with a 200-L poly pack containing vinyl wrapped metal pipes.

This case can be simulated by modeling a standard 208-L drum with a mixed-iron and hydrogen matrix. The iron-to-hydrogen mass ratio for this case is 300g iron per gram hydrogen. The system behavior for this case is shown in Figs. 21 and 22. In this case the effects of the iron and the hydrogen nearly offset one another.

Fig. 21. Behavior of Super-WDAS system efficiency with case 3 iron/hydrogen loading.

Fig. 22. Trend in the doubles count rate per gram $^{240}\text{Pu}_{\text{eff}}$ for a 64 μs coincidence gate for case 3 iron/hydrogen loading.

Case 4. Normal 208-L drum containing vinyl wrapped metal pipes.

This case can be simulated by modeling a standard 208-L drum with a mixed iron and hydrogen matrix. The iron-to-hydrogen mass ratio for this case is 100g iron per gram hydrogen (less hydrogen content than Case 3). The system behavior for this case is shown in Figs. 23 and 24.

Fig 23. Behavior of Super-WDAS system efficiency with case 4 iron/hydrogen loading.

Fig. 24. Trend in the doubles count rate per gram $^{240}\text{Pu}_{\text{eff}}$ for a 64- μs coincidence gate for case 4 iron/hydrogen loading.

Case 5. Normal 208-L drum with a concrete shield, containing a vinyl sheet and gloves.

This case can be simulated by modeling a standard 208-L drum with a concrete liner, as described by JNC, and a polyethylene matrix at various densities. A graph of the behavior of the Super-WDAS with increasing polyethylene loading is shown in Figs. 25 and 26.

Fig. 25. Behavior of Super-WDAS system efficiency with case 5 polyethylene loading in a concrete-lined drum.

Fig. 26. Trend in the doubles count rate per gram $^{240}\text{Pu}_{\text{eff}}$ for a $64 \mu\text{s}$ coincidence gate for case 5 polyethylene loading in a concrete-lined drum.

Case 6. Normal 208-L drum with a concrete shield, a Pb shield, and 100-L poly pack containing vinyl wrapped metal pipes.

This case can be simulated by modeling a standard 208-L drum with concrete and lead liners, as described by JNC, and an iron/polyethylene matrix at various densities. A graph of the behavior of the Super-WDAS with increasing polyethylene loading is shown in Figs. 27 and 28. The iron-to-hydrogen mass ratio for this case is 100 g iron per gram hydrogen.

Fig. 27. Behavior of Super-WDAS system efficiency with case 6 iron/hydrogen loading in a concrete-lined drum.

Fig. 28. Trend in the doubles count rate per gram $^{240}\text{Pu}_{\text{eff}}$ for a $64\ \mu\text{s}$ coincidence gate for case 6 iron/hydrogen loading in a concrete- and lead-lined drum.

From the system behavior shown in the preceding figures, it is clear that the Super-WDAS is remarkably tolerant of a wide variety of matrices and loadings. By using add-a-source functionality, the system can be made nearly invariant over the entire range of matrix types and possible loadings. Figure 29 shows the extent of matrix perturbation both with and without add-a-source correction in the High-Efficiency Neutron Counter system.¹⁶ After the add-a-source matrix correction, the average assay error for the entire set of matrix loadings is reduced to 3%.

Fig. 29. Effectiveness of add-a-source correction in an optimized drum counter.

B. Medium Resolution, Low-Energy-Photon-Based System

1. MDM and Applicability

For the case of waste generated following the first separation cycle, a system employing medium-resolution phoswich detectors can be used to detect the 59.5-keV gamma ray of ^{241}Am . The americium/plutonium ratio calculated from the known time interval since the original separation of plutonium from americium allows conversion to plutonium mass. Based on a system (conveyor belt sample handling) in operation at LANL, the MDM for ^{241}Am was at the nanogram level (assuming false-alarm and failure-to-detect probabilities of 5%) in measurement periods of 1000 s. If the period since americium/plutonium separation has been 10 years, the plutonium MDM will be about 20 ng. If the period since americium/plutonium separation has been 20 years, the MDM will be down near 10 ng. As noted here, the MDM for plutonium improves with time, a consequence of the fact that the ^{241}Am grows in from the decay of the 14.3-year half-life ^{241}Pu . It is also clear that in the first few years when the americium/plutonium ratio is very small, this method would not provide a very sensitive measure of plutonium mass. Determination Limits for RSD = 20 % are approximately 1.5 times the MDM values.

It is worth noting again that a system like this could screen clean, low-Z, low-density waste at levels sufficiently low to possibly permit cheap disposal of the waste. If such waste were measured in thin-walled plastic drums with diameters no greater than about 45 cm, then the L-x-rays of the various isotopes could be usefully detected. For ^{239}Pu , the directly measured MDM would be about $0.15 \mu\text{g}$ (~20 nCi), for ^{235}U , the directly

measured MDM would be about 0.5 mg (~1 nCi), and for ^{241}Am , the directly measured MDM would be about 0.3 ng (~1 nCi).

As with the neutron coincidence system, the MDMs are strong functions of the background rates and therefore of the effectiveness of the shielding. The calculations of the net rates corresponding to Critical Level, Detection Limit, and the Determination Limits are simpler than for the coincidence counters because the emission of the detected gamma rays is truly random. Converting from the net rate values to masses of ^{241}Am requires the specific gamma-ray emission rates (gamma rays/s-g) and the efficiency of the detector array. The various values given are for detector efficiencies and background rates that are thought to be reasonably attainable. Figure 30 gives the Critical Level, the Detection Limit — equal here to the MDM because the quantities are given as nuclide mass — and Determination Limit for $\text{RSD} = 20\%$, all as nanograms of ^{241}Am , as a function of background rate. Figure 31 gives the same quantities as net rates. In both figures, the count time is taken to be 1000 s. In a system at LANL employing three phoswich detectors, the background for the 60-keV gamma rays was about 0.6 s^{-1} . Any possible system for use at TRP would employ at least that many phoswich detectors, and perhaps as many as six. The estimates of the various limits have been based on the observed performance of the three-detector LANL system.

Fig. 30. For a state-of-the art medium-resolution, low-energy-photon NDA system, we give the Critical Level (L_c), Minimum Detectable Mass, (MDM), and the Determination Limit (L_d) for $\text{RSD} = 20\%$ (L_q); all expressed as ng of ^{241}Am . The false alarm probability and the failure-to-detect probability are both 5%. The count time is 1000 s and the false alarm and failure-to-detect probabilities have both been assumed to be 0.05. If the beginning plutonium isotopic distribution is known, the plutonium-to-amerium ratio can be calculated from the known time since americium was last removed.

Fig. 31. For gamma-ray systems using a simple background subtraction to obtain the measured net rate of a particular gamma ray, we give the Critical Level (L_c), Detection Limit (L_d), and the Determination Limit (L_q) for $RSD = 20\%$, all as net rates. The count time is 1000 s, and the false alarm and the failure-to-detect probabilities have both been assumed to be 0.05.

2. Hardware

The system requires a shielded measurement cavity large enough to enclose the 208-L drums and the detectors. There would be a minimum of three 12.5-cm-diameter phoswich detectors and a maximum of six such detectors. The shielding would consist of at least 5 cm of lead lined with 1-mm-thick layers of cadmium (or tin) and copper. The layers of cadmium (or tin) and copper reduce the intensity of lead x-rays which are fluoresced from the lead shielding and interfere with the low-energy peaks of importance in assays. Cadmium has been habitually used for the purpose, but if the neutron background is significant, neutrons can be captured in the cadmium and create gamma rays that add to the background. Some systems have been built with much thicker layers of iron instead of lead but that is not likely to be cost effective. Actual shielding would depend on the intensity of local backgrounds in the location of the system and on economic and health and safety factors. The last component of the mechanical system is adequate provision for placing the drums to be measured into and removing them from the system.

The electronics required to process the pulse-height information from the detectors would occupy perhaps two NIM bins. Most of it would be standard, commercial modules with perhaps a single custom LANL-designed module to make the processing of the information from multiple detectors in a single multi-channel analyzer more efficient. A single, small computer having adequate computational power and storage capacity containing the required multichannel analyzer cards would be required. It would control data acquisition, data analysis, calibration routines, measurement quality control functions, and storage and archiving of results.

3. *Software*

The necessary software routines to control data acquisition; to perform data analysis, calibration routines, and measurement quality control functions; and to store and archive results would be provided.

4. *Calibration Standards and Measurement Control Standards*

As with the passive neutron coincidence system, appropriate standards, both for calibration and for regular measurement quality control will be necessary adjuncts of the system. And again, it is probably best that these be fabricated at TRP in consultation with the development staff at LANL. We note that the calibration standards may be satisfactory for use in measurement quality control.

The standards will probably consist of a two component system. The first component will be one or more 208-L drums packed with appropriate matrix materials and with provision for placing americium-bearing standards at desired locations within the drum. The second component is obviously the actual packages containing known masses of americium. They will be fundamentally different from the metal capsules containing calibration material for the passive neutron coincidence counter system. The americium standards will probably consist of pads of absorbent material sandwiched between plastic sheets. They will, out of necessity, be more fragile than metal-clad capsules, but with due care can be used successfully and safely.

The same comments about standards appropriate to demonstrate MDM values that were given in Section IV-A-4 in connection with the passive neutron coincidence counter are equally valid here.

SUMMARY

For the special case of waste generated following the fission product removal, gamma-ray assay using phoswich detectors can provide excellent sensitivity for measuring the 59.5-keV gamma ray from ^{241}Am . If the time interval from the chemical separation of the plutonium from the fission product and actinides is known, the ^{241}Am measurement can be used to calculate the plutonium with good sensitivity. This method only applies to wastes without fission product contamination and with a homogeneous distribution, and this low-energy gamma ray does not penetrate the walls of shielded drums or emerge from the interior of heterogeneous drums.

The passive neutron detector systems can be used to measure the plutonium mass in shielded drums and in all of the matrix categories considered in this study. However, for the drums containing waste prior to the first separation stage, the neutron signal is dominated by the neutrons from ^{244}Cm . The approximated ratio of $^{244}\text{Cm}/\text{Pu}$ can be estimated from the average burnup of the input fuel assemblies. Alternatively, measurements of the curium and plutonium in the dissolver and waste solutions can be used. This stream-averaged $^{244}\text{Cm}/\text{Pu}$ ratio can be used to convert the measured ^{244}Cm mass to a plutonium mass.

The MDM sensitivity is very good for the waste containing curium because the neutron yield per gram from ^{244}Cm is 10,000 times higher than for ^{240}Pu . The actual mass of ^{244}Cm is about 1% that of plutonium, so the total neutron coincidence signal is about two orders of magnitude higher than that for plutonium alone. Thus, the sensitivity for measuring plutonium by its association with curium improves by two orders of magnitude. For an optimized neutron detector system, we calculated the direct plutonium measured MDM at 0.78 mg (see Section IV-A-D). The MDM improvement for the ^{244}Cm association method is ~8 nanograms of plutonium.

After the first separation cycle, the neutron measurement directly provides the ^{240}Pu -effective mass. The resulting MDM levels are about 0.8 mg and 0.4 mg of plutonium for measurement times of 1000 s and 4000 s, respectively. When totals counting, rather than coincidence counting, is used, the MDM is reduced by a factor of 2. However, the impurity levels can obscure the assay result based on totals counting. The totals assay result based on an oxide assumption will be correct or if there are impurities in the MOX, the assay will be high. Thus, the totals neutron count rate provides a sensitive flag (or upper limit) for the presence of plutonium or ^{241}Am in the waste.

The matrix corrections for neutron counting are modest and, after the add-a-source correction, the matrix errors are reduced to 3–5%. We expect that the dominant errors will come from the plutonium isotopic uncertainties and from the uncertainty in the $^{244}\text{Cm}/\text{Pu}$ ratio for the waste containing fission products. The measured $^{244}\text{Cm}/\text{Pu}$ ratios in the high-activity liquid waste going to Tokai Vitrification Facility can be used to predict the ratios in the LASW.

The WDAS-type neutron systems have been authenticated for use by the International Atomic Energy Agency for establishing the plutonium content in waste drums. The signal is analyzed using the INCC software and an independent calibration is performed by the Agency.

The inhomogeneous distribution of the plutonium in the LASW is a problem that is a major source of error in the measurements - especially for gamma-ray measurement systems. For the super-WDAS neutron system, we have introduced new technology to greatly reduce this source of error. The add-a-source matrix correction makes the assumption that the plutonium is well distributed throughout the matrix, and this is often not the case. The new technology uses multiplicity counting to measure the three observables (singles, doubles, and triples). The three parameters let us solve for the three unknowns (^{240}Pu mass, alpha, and efficiency). Thus, the change in efficiency caused by the inhomogeneous distribution of the matrix and the plutonium is directly measured in the three-parameter analysis. This advanced analysis, using both add-a-source and multiplicity, is already incorporated in the INCC software that has been authenticated and approved for use by the IAEA.

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