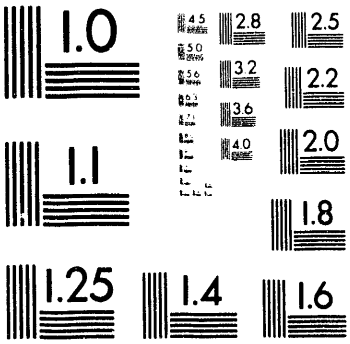
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COMPOSITE WASTE ANALYSIS SYSTEM

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ABSTRACT

Nondestructive analysis (NDA) of radioactive waste forms an integral component of nuclear materials accountability programs and waste characterization acceptance criterion. However, waste measurements are often complicated by unknown isotopic compositions and the potential for concealment of special nuclear materials in a manner that is transparent to gamma-ray measurement instruments. To overcome these complications, a new NDA measurement system has been developed to assay special nuclear material in both transuranic and low level waste from the same measurement platform. The system incorporates a NaI detector and customized commercial software routines to measure small quantities of radioactive material in low level waste. Transuranic waste analysis is performed with a coaxial HPGe detector and uses upgraded PC-based segmented gamma scanner software to assay containers up to 55 gal. in volume. Gamma-Ray isotopics analysis of both waste forms is also performed with this detector. Finally, a small neutron counter using specialized software is attached to the measurement platform to satisfy safeguards concerns related to nuclear materials that are not sensed by the gamma-ray instruments. This report describes important features and capabilities of the system and presents a series of test measurements that are to be performed to define system parameters.

INTRODUCTION

The Plutonium Facility at Los Alamos National Laboratory (LANL) is a U.S. national defense facility involved in the recovery and processing of plutonium and other transuranic elements. Wastes are routinely generated here from many stages of plutonium metal fabrication, including pyrochemical and aqueous processing and uranium and plutonium fuel fabrication. The processing steps generate a wide variety of leaner scrap and waste forms such as plutonium oxide from burned residues, Pu-bearing salts from production/reduction and metal purification processes, impure plutonium metal, metal reduction slags, ash, ceramics, cleaning rags, plastics, HEPA filters, and other remnants and apparatus generated from processing and cleanup tasks. Solid wastes created from this processing are divided into two categories: transuranic waste (TRU) containing greater than 100 nCi/g and Low Level Waste (LLW) containing less than this amount. TRU waste is obtained from various processing and cleanup steps within the processing gloveboxes and may be packaged in containers ranging from small cans up to 55 gal. drums. LLW is derived from locations outside the gloveboxes where contamination is expected to be light. Both solid waste categories receive nondestructive analysis (NDA) to determine radioactive content. For low bulk density materials ($\leq$ 1.5 g/cc) such as paper, plastics, ash, powders, etc, the analyses are performed using x-ray or gamma-ray analytical methods. Denser materials, such as metals, leaded gloves, and some plutonium salts are measured with passive or active neutron counter analysis.

In general, measurements of low density LLW utilize a combined x-ray and gamma-ray analysis routine in which the contributions from ^{235}U, ^{238}Pu, ^{239}Pu, ^{241}Am, and fission products are collected, then added together to determine the total activity in the waste[1,2]. In this scheme, the L x-ray region of the spectrum is first monitored for low radioactivity levels in the waste. As the activity level increases, the higher energy gamma rays from the isotopes of interest are examined to assay the sample. Because this technique does not correct for matrix attenuation or self absorption effects, it is suitable only for low bulk density materials with diffuse contaminations of plutonium or uranium. The sample is rotated and vertically repositioned before one or more NaI or HPGe detectors to smooth the spatial response from the sample. Correlation factors for non-gamma ray emitting species may be incorporated into the software to account for radioactive species not directly detected by the instrument[1]. This analytical method provides both high sensitivity, although relatively poor accuracy, using x-ray detection at low activity levels (~ 10 nCi/g) with poorer sensitivity, but improved accuracy, using gamma-ray detection at higher activity levels. However, despite the use of the two spectral regimes, there are several limitations to this method of LLW analysis. First, the method is generally insensitive to unknown radioactive contaminants or non-standard isotopic distributions. That is, contaminants such as ^{237}Np, ^{243}Am, or ^{244}Cm which are often associated with plutonium processing may not be revealed with this method. Failure to detect their presence can lead to significant underestimation of the total activity in the waste. In addition, more than one plutonium isotopic distribution is typically encountered at plutonium processing facilities. Because the principal isotopes in the distributions (^{239}Pu and ^{240}Pu) have substantially different specific activities, failure to determine the correct distribution can lead to inaccuracies in calculating the total activity in the waste. A second limitation in LLW analyses concerns their susceptibility to density variations in the waste. In the past, matrix attenuation of the gamma-ray and x-ray signals in low density LLW has been discounted as a relatively minor constituent of the overall measurement uncertainty. Typically a standard with the same density as the average LLW density was used to calibrate the instrument. However, as the variability in waste matrix types has grown and under increasingly stringent regulatory requirements for more accurate measurements of LLW, corrections for matrix attenuation effects have become necessary. A third limitation on existing x-ray and gamma-ray analysis of LLW relates to the inability of these instruments to detect plutonium concealed within photon shielding materials. Here, the concern is not that the radioactive content in the waste will be underestimated as discussed above; but rather that comparatively large quantities of plutonium (100s of grams) may be concealed within gamma-ray shielding materials such that they are invisible to the detector. For example, significant amounts of plutonium could be contained within relatively thin lead shielding; thereby presenting a diversion scenario of significant proportions to safeguards analysts.

Low density TRU waste is typically assayed with Segmented Gamma Scanners (SGS)[3,4]. Here the sample is rotated during the assay and a vertical sequence of collimator-defined horizontal segments are measured with a high resolution (generally) gamma-ray detector to minimize the effect of sample inhomogeneities. An attached external transmission source, emitting gamma radiation near the energy of the isotope being assayed, is used to correct for matrix attenuation effects. Rate-related losses (deadtime and pulse pileup) to the measured count rate are usually corrected by means of a separate, external gamma-ray source affixed to the detector head. SGS assays are also limited by certain deficiencies. As with LLW measurements, unexpected isotopic distributions or contaminant radioactive species cannot be distinguished in the measurements. This can result in increased measurement inaccuracy, underestimation of the radioactive material content, and failure to meet waste disposal criterion. Also, segmented gamma-ray analysis is insensitive to plutonium concealed within gamma-ray shielding material. The shielding may be due either to external shielding surrounding the plutonium or to self shielding in larger clumps of plutonium. Again, serious underestimations of the total radionuclide content in TRU waste containers can result.

To surmount these limitations on x-ray and gamma-ray measurements of waste, a new instrument has been developed jointly by personnel at the LANL Plutonium Facility and Canberra Industries, Inc. From the same measurement platform, the instrument can operate in either an SGS mode to analyze TRU waste and leaner residues or can be used in a multienergy analysis mode to measure low level waste. In the SGS mode, the instrument performs both conventional segmented gamma scan assays; and, in addition, is capable of performing high resolution gamma-ray isotopics analyses to search for unforeseen plutonium isotopic distributions or unexpected radioactive contaminants. In addition, during the SGS measurement, the passive neutron counter inspects the waste for neutron-emitting materials that are shielded from the gamma-ray assay. The collimator assembly and transmission source holder have also been redesigned for the new instrument.

In the multienergy analysis mode for LLW, the software inspects the ^{235}U, ^{238}Pu, ^{239}Pu, ^{241}Am, and fission product gamma-ray and x-ray spectral regions with the NaI detector typical of conventional LLW counters, and additionally performs gamma-ray isotopics analysis using a high resolution detector. Algorithms have also been installed in the measurement software to correct for bulk density variations in the waste and provide realistic assessments of the measurement uncertainty. Finally, in the LLW measurement mode, the neutron counter measurement again examines the waste for the presence of radioactive materials that are shielded from the gamma-ray analyses.

This paper will first describe the hardware and software components of the composite waste analysis system in more detail. Included in the description are all major revisions to current commercially available TRU or LLW measurement systems. Then a series of tests to be performed to determine performance parameters of the system will be discussed. In addition to hardware and software checkout, these include a series of measurements to determine instrument stability, sensitivity, susceptibility to geometric effects, efficacy of the LLW attenuation correction, uncertainty analysis, and measurement control parameters.

SYSTEM DESCRIPTION

The measurement frame for the new instrument was taken from an old SGS unit that was decommissioned and retrofitted to accommodate both low level and transuranic waste containers. Its footprint is approximately 5' x 5' although the dewar for the HPGe detector protrudes an additional 18" at one side. All power supplies, electronic modules, oscilloscope, counter/timer and motion controllers are housed in one standard 180 cm high x 56 cm wide x 76 cm deep NIM cabinet. The computer, printer, and keyboard are separately mounted on an adjacent 24" x 24" desk.

At this facility, low level waste is normally packaged in 1' x 1' x 2' cardboard boxes. The composite instrument is capable of measuring waste boxes in these containers; and, in addition, can measure LLW in 30 gal and 55 gal drums. Measurement time for the cardboard boxes is approximately 5 minutes. The instrument is programmed to detect ^{235}U, ^{238}Pu, ^{239}Pu, ^{241}Am, and fission products using a single 5" diam x 2" long NaI detector optically coupled to a photomultiplier tube. Shielding around the detector consists of a specially fitted 1.75" thick lead cover surrounding the sides, head, and back of the NaI crystal, except where the 2.0" wide vertical collimator allows x-ray and gamma-ray access from the sample. The collimator permits the entire length of the LLW container to be viewed throughout the measurement. This collimator arrangement, plus rotational and vertical scanning, permit smoothing of the detector spatial response to the waste package. The LLW measurement electronics, consisting of a preamplifier/discriminator, amplifier, high voltage power supply, analog to digital converter, and Canberra System 100 MCA board, form an independent, stand-alone, unit that controls only the LLW measurements. Periodic measurements of a shuttered ^{133}Ba source mounted to the platform provide gain and intensity control for the detector. Table I contains a more detailed listing of the components for each of the measurement systems in the composite counter. In addition, there will be simultaneous gamma-ray isotopic analysis using an HPGe detector performed during LLW measurements. Detection is via a single, coaxial, pure germanium detector. For low level waste measurements, this detector has a specially fitted, 2.0" wide vertical collimator which allows the entire length and width of the container to be viewed by the detector. The purpose of the high resolution measurement is to ascertain the isotopic distribution of the waste and to detect unexpected contaminants. An analytical balance, calibrated from 0 - 30 kg, is also connected to the system computer in order to calculate the activity per unit gram of waste. Finally, a small 1' x 1' x 4" wide neutron slab counter, containing four 8" long ^{3}He detectors surrounded by polyethylene, operates during LLW measurements. The high voltage supply and amplifier for this counter are internally mounted. The amplifiers convert the output of the ^{3}He tubes to a TTL pulse train directed to the counter/timer which, in turn, is interfaced to the computer. The neutron counter is attached to the side of the measurement platform and its purpose is to detect the presence of relatively large amounts of plutonium that may be transparent to the NaI detector. Because only totals neutron counts, not coincidences, are registered by these detectors, it is not intended to accurately assay the plutonium content in the waste. Rather, it is meant only to detect the presence of relatively large quantities of fissioning nuclei. Motor controllers for rotational and vertical positioning of the sample and shutter regulation for the gain source support the detection apparatus.

The modified SGS unit for analysis of TRU waste uses a high purity Ge detector to measure containers up to 55 gal drums in size. A specially-constructed collimator assembly allows the vertical LLW collimator arrangement to be quickly reconfigured to a horizontal position for segmented analysis in this mode of operation. The position of the collimator is automatically detected by a Hall-effect sensor connected to the computer.

Table I. The table lists the equipment used with each of the major components of the composite waste analysis system. Each component is a stand-alone unit in the sense that measurements can be performed with that unit without invoking the electronic control modules from other components.

LLW Waste Measurements
5" diam x 2" long NaI(Tl) Scintillation Detector with thin Be window and ~ 8 % resolution (at 662 keV).
Circular grid type, 10 stage Photomultiplier Tube with overall gain of ~10^5 and charge sensitive preamplifier.
Amplifier.
High Voltage Power Supply.
Analog-to-Digital Converter.
Gain control using shuttered ^{133}Ba source.
4096 data channel Multichannel Analyzer integrated within computer.
Analytical balance.
Stepping motor controllers for vertical and rotational scanning.
TRU Waste and Gamma-Ray Isotopics Measurement
20 % efficient pure Ge coaxial detector with thin Be window.
Amplifier.
High Voltage Power Supply.
Analog to Digital Converter.
4096 data channel Multichannel Analyzer integrated to computer.
Digital Stabilizer.
Six position Transmission Source Holder.
Stepping motor controller for rotational and segmented scanning.
Neutron Meas
1'x 1'x 4" wide Polyethylene Slab housing four 8" long ^{3}He detector tubes.
Internal Amplifier/Discriminator.
High Voltage Power Supply.
100 MHz Counter/Timer.

Adjustable collimation widths of ½" and 2" can be specified with the assembly. Shielding for the detector consists of a 2.0" thick lead cover surrounding the front, sides, and back of the crystal and preamplifier cylinder except where the collimator opens to the sample to allow gamma-ray access. An independent electronics package and motor controllers for rotational and segmented sample scanning and transmission source shutter management provide electronic and mechanical support for the measurements. Matrix attenuation correction is furnished by a shuttered, transmission source holder capable of housing up to six different gamma-ray emitting radioisotopes. This unit is controlled by the computer which permits automated transfer from one type of transmission source to another. The multiple holder allows quick realignment to a different transmission source in order to perform more accurate matrix attenuation corrections at gamma-ray energies closer to those of the assayed radionuclide. Rate loss corrections are effected with a ^{109}Cd source mounted next to the HPGe detector crystal. Simultaneous gamma-ray isotopics analysis of TRU waste containers is performed with the same high resolution detector and electronics modules, but using an independent software analysis routine. For this measurement, the individual segment spectra are summed to a single composite spectrum which is then analyzed for plutonium isotopic composition and unanticipated radioactive contaminants. The small neutron counter, described above, also operates during TRU waste assays to detect the presence of large amounts of plutonium that are shielded from the gamma-ray detector. Measurement time for 55 gal. drums is approximately 30 minutes. A diagram of the composite system is shown in Figure 1.

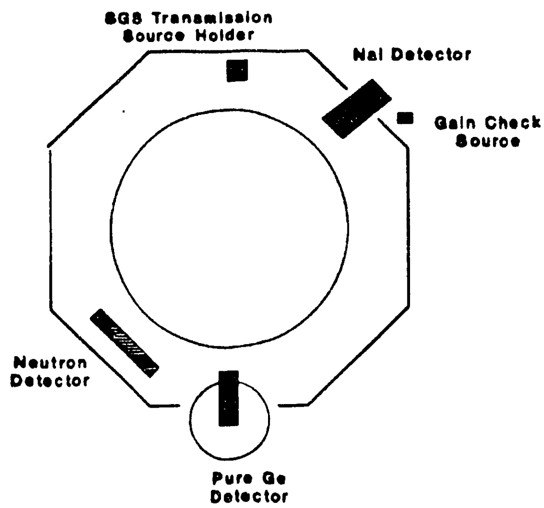

Figure 1. Schematic of the Composite Waste Measurement System showing the measurement platform and the three detectors.

The software for the composite system is accessed through the Canberra System 100 data acquisition and analysis structure which features a PC-based multichannel analyzer system and operates through the Microsoft Windows operating environment for program execution and multitasking control. Data analysis is based on three existing applications programs for the gamma-ray measurements and a new program tailored for the neutron counting application. The LLW and SGS measurement programs are a combination of FORTRAN and C with I/O predominantly in C and analysis algorithms predominantly in FORTRAN. Use of Microsoft Windows and the S 100 board allow the menu-driven operator options and the x-ray/gamma-ray spectrum to be simultaneously accessible. The interface between the operator and the overall system is through the video terminal; that is, instrument output and operator options are displayed and controlled via this terminal. A main access menu has been added to the system to allow the operator to select LLW measurement, TRU waste measurement, gamma-ray isotopics analysis, container weight input, S 100, or Microsoft Excel. Each of these programs can be run independently of the others. In addition, the operator can exit to DOS through this menu.

Conventional LLW analysis permits measurement control analyses, calibrations, and assays using fixed window spectral acquisition at operator determined x-ray and gamma-ray energies. In addition to these types of analyses, the new instrument includes supplemental software to perform the following functions in the LLW measurement mode. To allow gamma-ray isotopics analysis to be performed simultaneously, the software first checks that the collimator for the Ge detector is in the vertical position. If it is and if the LLW assay performed by the NaI detector also indicates that the plutonium content in a waste container exceeds a preset value, a gamma-ray isotopic analysis and peak search is automatically triggered on the high resolution detector. The isotopic analysis is performed with a commercially available software package (Canberra's SPECTRAN) which features peak search, multiplet deconvolution, radionuclide identification, decay corrected activity, operator controlled default settings, and library editing features. The results of the isotopics measurement are saved on a file in the computer whose identifier is also listed on the assay report. In addition, computer-controlled zero and gain checks over the linear range, and resolution and efficiency checks based on low and high energy peaks from a ^{133}Ba source mounted on the instrument platform, have been added to the NaI measurement software. These system checks are performed during periodic background measurements. If the supervisor-controlled limits for these checks are exceeded, error messages, warnings, and corrective action statements are displayed on the terminal, printed on the printer, and written to the permanent file for that measurement. Next, an analysis file for matrix corrections has also been incorporated into the measurement algorithm. Here, a series of container standards with known densities will first be used to determine attenuation correction factors[5] for the gamma-ray energies employed in the low level waste measurement. These factors and their range of applicability will then be entered into the software to provide corrections to the assays based on the density of the waste packages. The software also includes an analysis of the minimum detection limit for the measurement. Periodic evaluation of the minimum detection limit will be incorporated into the measurement control sequence for the system. The next software addition to the LLW algorithm is an estimate of the error in the measurement. Because all the sources of error and their ranges have not yet been determined for the instrument, these cannot yet be installed into the software. Therefore, the software provides for inclusion of 5 pairs of data points of percent error as a function of plutonium value. A second order fit will be made to these data points and the resulting equation used to provide the error for individual waste containers. Finally, if the count rate from the concurrent neutron measurement exceeds a preset value above the background level, then this information is also reported on the terminal display, printer output, and permanent file. A list of changes to conventional measurement software for the composite system is presented in Table II for both LLW and TRU waste measurement.

The low level waste measurement file and printer report will contain the following information: waste type, date, time, operator identification, container identification and room of origin, sample weight, isotopic and summed activity of the waste, measurement error estimates, lower limit of detection, neutron count rate warning message (if the preset has been exceeded), gamma-ray isotopics identification and warning message (if preset limits have been exceeded), and matrix attenuation correction factors used in the activity calculation. All report data are written into a formatted ASCII file read into a Microsoft Excel spreadsheet.

Several changes have also been added to the segmented gamma scanner software. Conventional SGS software includes algorithms for background analysis, system gain, measurement control,

Table II. Summary of software changes in the Composite Waste Analysis System.

Description of Change	LLW	TRU
Main Access Menu	x	x
Preset to trigger SPECTRAN Analysis	x	
Peak Energy for Zero Check	x	
Peak Energy for Gain Check	x	
Count Time for Neutron Background	x	x
Preset Background Neutron Count Rate	x	x
Matrix Correction	x	
Historical Error Analysis	x	
Minimum Detection Level	x	
Simultaneous Gamma Isotopic Analysis	x	x

calibrations, and assays based on high resolution, fixed window analysis of preset gamma-ray peaks. All assays include corrections for container, matrix, and rate loss effects. In addition to these types of analyses, the SGS software for the new instrument includes several additional features. First, because the collimator for the pure Ge detector can be positioned either horizontally or vertically, a sensor adjacent to the collimator communicates to the software whether it is correctly positioned for SGS measurements. If it is not, a message appears on the terminal that the collimator is out of position. Next, the spectral data gathered by the detector is directed to both the SGS software and to the gamma-ray isotopics package, thereby enabling a peak search for contaminants and confirming the plutonium isotopic distribution. Simultaneously, the neutron detector is activated and data from this instrument is sent to neutron counting software to determine whether the count rate is within preset limits. The SGS software also contains additions to the measurement diagnostics. During the daily background measurement to check SGS instrument stability and background effects, a supplemental check of the background neutron count rate and ^{3}He detector efficiency is also performed. If this rate is above acceptable safeguards limits, then a warning message appears on the terminal, printer output, and permanent computer file for the measurement. Also, corrective action for the operator is displayed on the terminal when diagnostic limits have been exceeded. In addition to the results of the segmented gamma scanner assay, generic information on the measurement (date, time, operator ID, etc), and diagnostic messages; the TRU waste printer and computer file also contain data on the weight of the sample, identification of the isotopic constituents, and the neutron count rate (if the preset limits are exceeded).

PROJECTED STUDIES

After fabrication and instrument checkout, a series of evaluations are planned for the system to determine its operating envelope for both LLW and TRU waste. In the LLW measurement mode, a special set of NIST-traceable standards will be constructed for use with the evaluations. The standards will span the allowable nuclear material ranges and LLW box weight loadings encountered at this facility. One evaluation will study matrix and attenuation effects in these measurements. Because part of the LLW measurement algorithm involves analysis of the spectral region from 13 kev to 60 kev, x-ray and gamma-ray attenuation effects can be substantial if left uncorrected. The evaluation will study photon attenuation in each of the algorithm's spectral regions of interest using standards of known activity content and representative weight loadings. Based on this study, appropriate correction factors for matrix attenuation effects will be determined. A second evaluation to be performed in the LLW measurement mode involves assessment of the effect of source position on detector response. Again, sources of known plutonium content will be placed in different locations within the low level waste container and their count rates will be registered. The sources will be measured both alone and mixed with matrix materials commonly encountered at this facility so that combined matrix and geometric effects can be studied. In addition, short term (several hours) and long term (several months) stability investigations will be performed as part of the instrument's evaluation. Here, repeated measurements of standards will be carried out in both the LLW and TRU waste measurement modes to establish valid measurement control criterion for the instrument's operation. Another evaluation will determine the sensitivity of the gamma-ray and neutron instruments in both measurement nodes. The Plutonium Facility has relatively high neutron and gamma-ray backgrounds and their effect on instrument performance must be assessed prior to placing the system into regular service. Finally, LLW measurement error analyses based on the evaluations discussed above will be calculated and incorporated into the software so that realistic uncertainty statements can accompany the system's assays.

SUMMARY

A new NDA system has been developed to assay low level and transuranic waste generated from plutonium and uranium processing operations. The system uses a single platform to perform both types of measurements on containers as large as 55 gal drums. Conventional SGS and multienergy x-ray/gamma-ray low activity analysis algorithms are supplemented in the system to include gamma-ray isotopics capability to assess plutonium isotopic distribution and the presence of radioactive contaminants. In addition, a small neutron slab counter has been attached to the instrument for the purpose of sensing the existence of relatively large amounts of nuclear material that cannot be detected by the gamma-ray instruments. Algorithms have also been added to the LLW mode to correct for matrix effects and determine minimum detection levels.

Fabrication and delivery of the instrument to this facility was completed in July, 1993. Checkout and evaluation is expected to take approximately six months at which time the system will be put into service. An adjacent facility at LANL with similar waste generation as that encountered at this facility has ordered a duplicate of this system to meet their measurement needs.

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