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**Application of PINS and GNAT to the Assay
of 55-gal Containers of Radioactive Waste**

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ABSTRACT

The Portable Isotropic Neutron Spectroscopy (PINS) and Gamma Neutron Assay Technique (GNAT) assay systems that were developed with funding from the Office of Research and Development (NN20), were taken to the Stored Waste Examination Pilot Plant (SWEPP) facility at the Radioactive Waste Management Complex (RWMC) and applied to the assay of surrogate and Rocky Flats Plant waste contained in 55-gal drums. PINS, a portable prompt γ , neutron activation analysis technique, was able to identify key elements in both the surrogate and real waste so that three main waste categories: metal, combustible material, and cemented chlorinated sludge wastes could be identified. GNAT, a γ , neutron assay technique for the identification and quantification of fissioning isotopes, was able to identify ^{240}Pu in surrogate waste in which nine 1-g nuclear accident dosimeters were inserted. GNAT was also able to identify ^{240}Pu in real 55-gal waste drums containing 15- and 40-g of plutonium even in the presence of high activity concentrations of ^{241}Am .

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Application of PINS and GNAT to the Assay of 55-gal Waste Containers of Radioactive Waste

1. INTRODUCTION

The Portable Isotopic Neutron Spectrometer (PINS)¹ and the Gamma Neutron Assay Technique (GNAT)² systems were developed with funding from the Office of Research and Development (NN20) for the in-field, nonintrusive identification of chemical munitions and nuclear weapons, respectively. Last September these systems performed analyses at the Stored Waste Examination Pilot Plant (SWEPP) to determine their applicability for the assay of key elements, or fissile materials, present in low-level waste (LLW), transuranic waste (TRU), and mixed waste in 55 gal. (2086) drum containers from Rocky Flats. Low-level and Transuranic waste was shipped in 55 gal. drums to the INEL from the Rocky Flats Plant (RFP) for temporary storage at the Radioactive Waste Management Complex (RWMC). The waste is categorized by the radionuclide activity concentration (plutonium for TRU) and the material matrix for each drum. The coarse matrix groupings consist of cemented chlorinated sludges, combustible materials, and metals.

Measurements were made on surrogate TRU waste and on real waste to determine if the PINS and GNAT could provide meaningful information on the key elements and on the plutonium content present in this type of waste. The results and conclusions drawn from these measurements follow.

2. PINS MEASUREMENTS

The objective of PINS, a portable prompt γ neutron activation analysis (PGNAA) technique, was to determine if it could identify the waste matrix of each drum from the assay of key elements in the matrix to permit the sorting of these drums into three coarse categories (i.e., cemented chlorinated sludges, combustibles, and metals).

The PINS system consists of a $\sim 5\mu\text{g}$ ^{252}Cf neutron source, reflector/moderator block, shadow shield, and a portable Ge spectrometer interfaced to a notebook PC. Eighty-one-ninety-two-channel spectra were acquired from the ^{252}Cf neutron irradiation of three surrogate waste drums and from three actual waste drums. These data were transferred to a VAX computer for convenience and analyzed with the gamma-ray spectral analysis program VAXGAP³. The surrogate waste drums consisted of one with metals (various types of steel), one with combustible material (paper, wood, plastic, etc.), and one with cemented chlorinated sludge (e.g., lubricating and cutting oils). Spectra from three different types of waste drums containing surrogate waste were acquired and are shown in Fig. 1, 2, and 3. Spectra were also acquired from three different types of actual waste. These spectra are shown in Figs. 4, 5, and 6. The γ -ray peaks in these spectra are labeled by energy, associated element, reaction that produced it (i.e., n,γ ; n,n' ; or decay) and interaction in the occurring detector. From these spectral plots the following conclusions are made:

1. The PINS system was able to identify the following elements present in the drums: hydrogen, iron, chlorine, nickel, chromium, calcium, silicon, and aluminum.
2. The PINS system was able to identify aluminum, nickel, chromium, and iron from close lying prompt γ -ray peaks between 7500 and 9800 keV in the surrogate waste containing walrath cans made of stainless steel and, aluminum and iron scrap, and in real waste containing metal pump parts primarily of iron.
3. The PINS system was able to identify hydrogen, calcium, chlorine, silicon, and iron present in the surrogate and real waste drums of cemented chlorinated sludge. Because several different kinds of cement were used in the disposal of sludges at the RFP, lack of a strong calcium peak from Portland cement in the PGNAA spectrum does not necessarily ensure the waste is not a cemented sludge.

4. For the drums containing combustible materials the PINS system identified a strong hydrogen prompt γ -ray peak at 2223.25 keV, and less intense peaks (relative to sludge) associated with chlorine, silicon and aluminum in the surrogate and real waste drums as shown in Figs. 2, 3, 5 and 6.
5. Spectra were acquired from two of the actual waste drums (cemented chlorinated sludge and pump parts) at an expanded gain with the ^{252}Cf source removed. These γ -ray spectra clearly show the presence of ^{241}Am and ^{239}Pu as shown in Fig. 7.
6. Examination of the spectra in the energy range from 7000 to 9000 keV indicates that capture γ -ray peaks with good signal-to-continuum ratios from iron, nickel, and chromium are present as seen in Fig. 1. Examination of the spectra in the energy range from 1500 to 2500 keV indicates that inelastic scatter and thermal neutron capture peaks with good signal-to-continuum ratios from iron, calcium, silicon, chlorine, and hydrogen are present as shown in Fig. 6. Together, the data from these energy regions may inform the operator of which elements and hence waste category is in the drum being assayed.
7. From an examination of the PINS acquired spectra it is possible to sort the waste into three categories: combustible material, cemented chlorinated sludge, and metal from an identification of the key elements present in each waste and from the relative peak intensities of these key elements.

3. GNAT MEASUREMENTS

The new portable Gamma Neutron Assay Technique (GNAT), that consists of eight liquid scintillation BC-501 neutron detectors and three 25% duet style Ge detectors, was used for waste assay measurements for Plutonium at the RWMC from September 27 through October 7, 1993. The ^{239}Pu content would be derived from the measured ^{240}Pu and the $^{239}\text{Pu}/^{240}\text{Pu}$ ratio. A schematic of the experimental set up is shown in Figure 8. The tests were carried out in the TRUPACT building at the RWMC.

The first set of measurements was performed by placing nine 1-g NAD's (94% ^{239}Pu and 6% ^{240}Pu) in different positions inside the steel surrogate 55-gal (calibration) drum. The coincidence data for both γ - γ and neutron- γ pairs were collected. Also, in order to provide an efficiency calibration for the Ge detectors, γ -ray singles were accumulated at the same time. The spontaneous fission half-lives for ^{239}Pu and ^{240}Pu are 5.5×10^{15} y and 1.34×10^{11} y, respectively. This means that only ^{240}Pu has a short enough half-life to measure the associated neutrons produced by spontaneous fission.

Pulse-shape discrimination techniques were used to separate γ -ray pulses from neutron pulses in the neutron detectors. A typical spectrum in which pulse-shape discrimination was applied for the above measurements is shown in Figure 9. If the NAD sample contained a pure ^{239}Pu source (i.e., no ^{240}Pu present), only the γ band (lower band) would be observed. The existence of the neutron band (upper band) in this spectrum indicates that the ^{240}Pu isotope is present in the NAD samples located in the steel calibration drum.

After the calibration measurements, three waste drums with different amounts of ^{239}Pu were transported to the TRUPACT building for our investigation. They contained 15 g, 40 g and 130 g of plutonium, respectively. The γ -ray singles and the total γ -ray coincidence spectra for the waste drum containing 15 g of plutonium are shown in Figures 10 and 11, respectively. The peaks in these spectra are labeled according to their energies in units of keV. Even though both spectra show the very strong presence of ^{241}Am , which has a half-life of 432.7y the γ -n and n-n coincidences positively identify the presence of a spontaneously fissioning radionuclide in this case ^{240}Pu . Each spectrum was dominated by the presence of ^{241}Am . This radionuclide, can be troublesome in normal passive/active assay systems that cannot easily discriminate between fission neutrons and (α , n) reaction produced neutrons. Transitions from ^{241}Am are observed in the spectra with energies ranging from 60 keV to 768 keV. In the case of the drum with 130 g

of Pu, the count rate from the ^{241}Am was so high that it was necessary to move the Ge detectors away from the drum in order to prevent the Ge preamplifier from saturating. Even with this drum 20 feet from the detection system the 60-keV γ ray of ^{241}Am was easily observed. Use of a suitable attenuator around the detector housing to reduce the intensity of the 60-keV ^{241}Am radiation is necessary for GNAT to be an effective tool in measuring the amount of plutonium in a waste drum in the presence of high ^{241}Am radiation.

The presence of γ -rays at 129, 375 and 413 keV indicates the presence of ^{239}Pu in the waste drum. Also, $K\alpha$ X-rays from the decay of plutonium are observed in these spectra at 104 keV. The intensities of ^{239}Pu gamma-rays could be used by themselves, if the plutonium distribution were homogeneous, to determine the amount of this isotope. This is not typically the case with waste. The 295-keV and 1280-keV transitions are from spontaneous fission of the ^{240}Pu isotope and are associated with the decay of the fission fragment ^{134}Te which consists of 2% of the total fission yield. These peaks show up weakly in the total γ -ray coincidence spectrum (Fig. 11) as compared with the γ -ray singles spectrum (Fig. 10). Quantification of the amount of ^{240}Pu isotope in the drums containing 13 g and 40 g of plutonium was not possible, but the comparison of these spectra with the spectrum taken with NAD's implied that the amount of ^{240}Pu is much less than 1 g. From the ratio of the γ -ray intensity of ^{239}Pu to the γ -ray intensity of ^{134}Te in the γ -ray singles spectrum, one should be able to determine the percentage of ^{240}Pu in the Pu mixture but this requires precise efficiency calibrations that may not be possible on waste drums in which the radionuclide content is not homogeneously distributed. Several γ -ray analysis programs have been developed to measure isotopic or radionuclide ratios from each of the isotopes (or radionuclides) of interest by analyzing a close-lying γ -ray doublet in which each component is from one of the isotopes (or radionuclides) being assayed. The applicability of these programs to the present assay needs to be determined.

Other radio-isotopes that were identified in spectra from the waste drum are labeled in the spectra in Figs. 10 & 11.

4. CONCLUSION

The PINS system has been successfully used to identify key elements in three different matrix categories of surrogate and real waste drums stored at SWEPP. From the presence and relative intensities of the peaks associated with these key elements, the three waste categories: metals, combustible material, and cemented chlorinated sludge can be sorted. Because the range of relative intensities for hydrogen, chlorine, and calcium may overlap for different types of combustible materials and cemented chlorinated sludge wastes, additional measurements on all types of cemented chlorinated sludge are recommended before all waste subcategories can be confidently sorted with high reliability. Additional measurements of several drums from each subcategory of cemented chlorinated sludge and of combustible materials would be required to verify that subcategories of cemented chlorinated sludge and combustible materials can be sorted by PINS with confidence.

The coincidence technique used by the GNAT system reduces the background substantially and increases the peak/background ratio by a factor of 10. This is very important in assaying very high-level wastes as the background problem causes misidentification of some radioisotopes and sometimes even makes it impossible to identify other radioisotopes. A large amount of ^{239}Pu , ^{241}Am and other isotopes was identified in the three different waste drums that were assayed. The ^{240}Pu content of the 15- and 40-g drums were found to be less than 1 g. In the case of the drum containing 130-g of plutonium, however, the americium activity was so high that the detectors had to be moved away from the drum. Therefore, the measurement of ^{240}Pu content was not possible. Based on these tests we learned that future measurements of real waste will need to incorporate an attenuator to absorb the 60-keV γ ray from ^{241}Am (2-mm thick Sn reduces transmission to <0.1%); interference from higher-energy γ rays on a successful assay will then be evaluated.

5. SUMMARY

The use of a PINS system incorporating a $\sim 5\mu\text{g}$ ^{252}Cf neutron source reflector/moderator block, shadow shield, and a portable Ge spectrometer has been successfully tested for the identification of key elements in transuranic (TRU), low-level (LLW), and mixed waste by prompt gamma neutron activation analysis (PGNAA). The specific waste forms assayed were cemented chlorinated sludge, combustible material and metal wastes in 55-gal drums from the Rocky Flats Plant and stored at SWEPP. These measurements identified the following elements in drums of surrogate or real waste: chlorine, hydrogen, silicon, calcium, aluminum, iron, nickel, and chromium. The presence of a subset of these elements and their apparent relative concentrations can be used to identify and sort the three main matrix categories of waste measured: cemented chlorinated sludges, combustible material, and metal wastes.

GNAT, a Ge spectrometer and neutron counter assembly that measures singles and coincidence counts following fission, demonstrated its ability to detect nine 1-g NAD (nuclear accident dosimeters) placed in a surrogate 55-gal waste drum. These sources contain $\sim 6\%$ ^{240}Pu the source of spontaneous fission upon which the GNAT method is based. The total plutonium content is determined from the ^{240}Pu content and the plutonium isotopic ratio. Following this success, real waste drums containing 15-, 40-, and 130-g quantities of plutonium were counted with the GNAT system. The drums investigated were found to contain a large amount of ^{239}Pu , ^{241}Am and other γ -ray emitting radioisotopes. The radionuclide ^{240}Pu was specifically identified in the drums containing 15- and 40-g of plutonium from γ - γ and γ -n coincidences. Because of the high level of the ^{241}Am activity in the drum containing 130-g of plutonium causing preamplifier saturation, the GNAT detector assembly had to be located about a meter from the drum. This poor geometry resulted in a severe reduction in coincidence counting efficiency. As a result, this waste drum containing 130-g plutonium could not be assayed with confidence using the GNAT γ - γ , γ -n and γ - and n- singles spectra. Future GNAT measurements of high-level waste will incorporate a suitable attenuator (e.g., 2- or 3-mm

thick tin sheet surrounding the detectors) to allow measurements in close proximity to the waste drum even when high activity concentrations of ^{241}Am are present.

6. REFERENCES

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FIGURES

1. PGNAAs spectrum of a 55-gal surrogate waste drum containing walrath cans, aluminum, and iron. Each labeled peak consists of an energy in keV, the activated element symbol, whether it is a full-energy (no prime), single-escape peak (single prime), or double-escape peak, and whether the peak is from thermal neutron capture (c), or from inelastic scatter (s).

55-gal. surrogate waste drum
 Contents: walrath cans, aluminum,
 and iron

55-gal. surrogate waste drum
 Contents: walrath cans, aluminum,
 and iron

2. PGNAAs spectrum of a 55-gal surrogate waste drum containing combustible material. Each labeled peak consists of an energy in keV, the activated element symbol, whether it is a full-energy (no prime), single-escape peak (single prime), or double-escape peak, and whether the peak is from thermal neutron capture (c), or from inelastic scatter (s).

55-gal. surrogate waste drum
Contents: combustible material

55-gal. surrogate waste drum
Contents: combustible material

3. PGNAAs spectrum of a 55-gal surrogate waste drum containing cemented chlorinated sludge. Each labeled peak consists of an energy in keV, the activated element symbol, whether it is a full-energy (no prime), single-escape peak (single prime), or double-escape peak, and whether the peak is from thermal neutron capture (c), or from inelastic scatter (s).

55-gal. surrogate waste drum
Contents: cemented chlorinated sludge

55-gal. surrogate waste drum
 Contents: cemented chlorinated sludge

4. PGNAAs spectrum of a 55-gal real waste drum containing pump parts. Each labeled peak consists of an energy in keV, the activated element symbol, whether it is a full-energy (no prime), single-escape peak (single prime), or double-escape peak, and whether the peak is from thermal neutron capture (c), or from inelastic scatter (s).

55-gal. real waste drum
Contents: pump parts

55-gal. real waste drum
Contents: pump parts

5. PGNAAs spectrum of a 55-gal real waste drum containing combustible material. Each labeled peak consists of an energy in keV, the activated element symbol, whether it is a full-energy (no prime), single-escape peak (single prime), or double-escape peak, and whether the peak is from thermal neutron capture (c), or from inelastic scatter (s).

55-gal. real waste drum
Contents: combustible waste

55-gal. real waste drum
 Contents: combustible material

6. PGNAAs spectrum of a 55-gal real waste drum containing cemented chlorinated sludge. Each labeled peak consists of an energy in keV, the activated element symbol, whether it is a full-energy (no prime), single-escape peak (single prime), or double-escape peak, and whether the peak is from thermal neutron capture (c), or from inelastic scatter (s).

real
 55-gal. surrogate waste drum
 Contents: cemented chlorinated sludge

55-gal. real waste drum
 Contents: cemented chlorinated sludge

7. Spectrum of γ radiation emitted from real waste drum with energies of key peaks labeled in keV.

Passive γ -ray spectrum of real waste drum

8. Detector assembly of GNAT including three duet style Ge spectrometers and eight neutron detectors.

9. Neutron, γ coincidence distribution. Pulse shape discrimination techniques were applied to the liquid scintillation BC 501 neutron detectors.

Figure 4. Neutron-gamma discrimination for $^{241-242}\text{Pu}$ source.

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10. Singles spectrum taken with one of the Ge diode spectrometers of TRU waste in a 55-gal drum containing 15-g of plutonium from the Rocky Flats Plant. Peak energies are labeled in keV.

11. Coincidence spectrum taken with Ge duet spectrometers of TRU waste in a 55-gal drum containing 15-g of plutonium from the Rocky Flats Plant.

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