AN ELECTRON ACCELERATOR BASED SYSTEM FOR ASSAY OF TRANSURANIC WASTE BARRELS*

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Summary

A complete assay system for 208-liter barrels containing transuranic wastes has been developed. The system consists of an 8-MeV commercial electron accelerator, neutron moderating cavity housing the waste barrel and containing neutron detectors, high resolution germanium gamma spectrometer, and x-ray radiography camera (both film and real time). The electron linac is used to produce bremsstrahlung and high-intensity pulsed neutron flux, both of which are used to interrogate the fissionable materials. The Differential Dieaway Technique is used to assay the amounts of fissile and fertile materials. The neutron flux is also used in the Prompt Gamma Activation Assay to determine and to quantify the matrix elements present in the barrels. This information is then used to correct the assay of fissionable material. The bremsstrahlung too, is also used by x-ray radiography system to further identify the matrix.

Introduction

Current U.S. Department of Energy (DOE) guidelines for management of transuranic (TRU) waste have created a need for instrumentation to monitor TRU's at the 10 nCi/g level.[1] Because the TRU's may be contained in high density or β-γ contaminated matrices, existing passive techniques are not generally acceptable. Recent efforts, using active interrogation with either photofission by bremsstrahlung or thermal neutron fission from pulsed neutron sources, have produced methods which meet many of the basic sensitivity requirements.[2,3] However, neither of these two methods alone appears to be entirely adequate. Photofission offers good sensitivity for a large number of TRU's but, because of the similarity of photofission cross sections, identification of specific nuclides or classes of nuclides is difficult. For example, the important fissile and fertile groups cannot be readily distinguished. Thermal neutrons offer very high sensitivity for fissile elements but essentially none for fertile elements.

A combination of neutron and photon interrogation offers several distinct advantages over either applied alone, including a direct and unequivocal separation of the fissile and fertile groups.

Another important aspect of any assay, whether it be neutron or photon, is to make appropriate corrections for the matrix. Neutron-based assays are adversely affected by both low-Z materials and strong neutron absorbers such as cadmium and gadolinium. For photon interrogation and photon detection, the high-Z materials have a strong influence on the photon signature.

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Matrix constituents can be identified with thermal neutron capture gamma ray spectroscopy, which can identify low-Z materials, such as hydrogen, aluminium, calcium, and iron as well as certain strong neutron absorbers such as boron, chlorine, and cadmium. Some of the rare earth elements, for example gadolinium and samarium, have very high thermal neutron absorption cross sections and intense energetic gamma rays. High-Z materials, like lead, are difficult to detect due to small thermal neutron capture cross sections. Thus the matrix identification possible with capture gamma ray spectroscopy (also known as prompt gamma ray activation analysis or PGAA) is more helpful to neutron interrogation and detection techniques.

Recently, we reported results from the proof-of-principle experiments.[4] In this paper, we report the results from work on the engineering prototype of the practical electron-accelerator-based TRU waste assay system.

System Description

The schematic diagram of the engineering prototype of the barrel counter plus the experimental geometry are shown in fig. 1. The EG&G/EM/DOE linac was used as a source of 6-MeV electrons. In the second part of our experiments we used an 8-MeV commercial radiographic linac. The neutron moderating cavity of the barrel counter is lined with 10 cm of reactor grade graphite. The neutron detectors are placed outside the graphite layer on five sides of the cavity, and are embedded in 10 cm of polyethylene. The sixth side of the cavity contains the berylium converter target. The neutron detectors are ^{3}He-filled proportional counters. Two types of packages are used: bare, for thermal neutron detection, and cadmium wrapped for fast neutron detection. The entire assembly is shielded from the external neutrons by 10 cm layer of borated polyethylene. The system also includes Ge(Li) photon spectrometer viewing the center section of the barrel counter cavity.

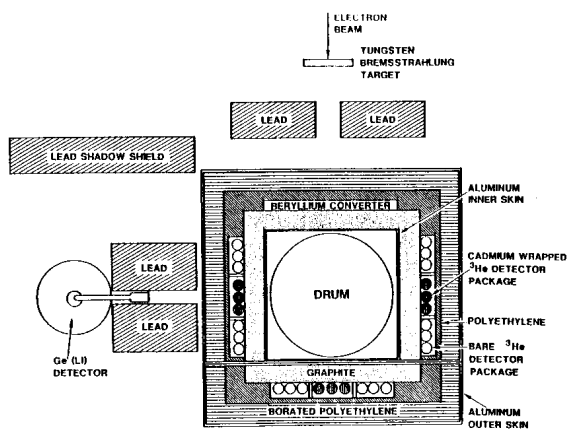

Fig. 1. Experimental arrangement showing 208-liter barrel counter and the Ge(Li) detector with its shielding.

Principle of Method and the Measurement Results

Differential Die Away Time

The pulsed beam of electrons accelerated to energies above 1.7 MeV (typically 4 or 8 MeV) strikes the tungsten target, producing bremsstrahlung. The bremsstrahlung in turn strikes the beryllium target, producing a pulse of high-energy neutrons. Within 80 µs these neutrons are slowed down to thermal energies. The resulting thermal neutron flux remains in the moderating cavity for some time, typically decaying with the half life of about 800 µs. During that time it will generate thermal neutron fissions among the fissile TRU that may be present. Fission neutrons from thermal fission are thus separated in time from the photoneutrons.

Most of the bremsstrahlung photons will pass into the volume of the waste sample where some will cause photofissions. The prompt photofission neutrons will not be distinguishable from photoneutrons that are formed in the materials of detection geometry, but the delayed neutrons from fission will be emitted continually during the whole period between the linac pulses.

Both the delayed neutron and prompt neutron emission during the thermal die away time are measured with a fast-neutron-sensitive, moderated ^{3}He proportional counter, shielded from the thermal neutron flux by cadmium. A typical signal from a cadmium-shielded detector located within the moderating assembly is shown in fig. 2.

To characterize the behavior of the barrel counter, we measured its response versus height to a point ^{252}Cf source. The results are shown in fig. 3.

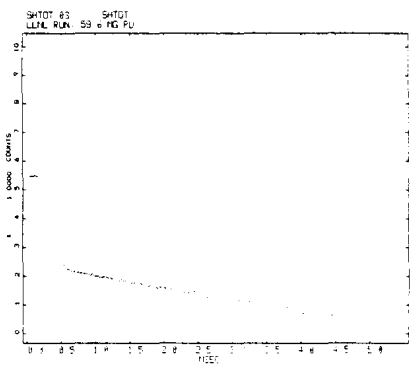

Fig. 2. Neutron count rate versus time for 60 mg ^{239}Pu.

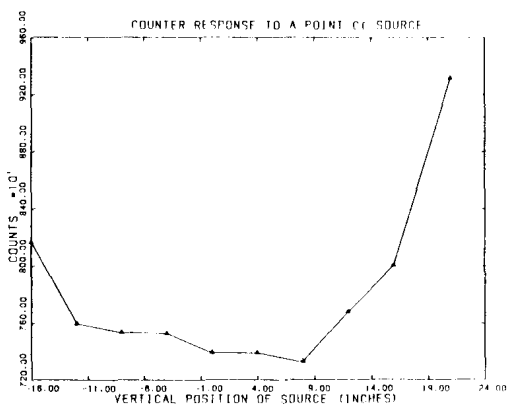

Fig. 3. Response of barrel counter to a point ^{252}Cf neutron source.

To demonstrate the relative portability of the system and that it can be used with the commercial radiographic electron accelerators, we conducted a series of measurements at the Lawrence Livermore National Laboratory (LLNL), using the Varian Linatron-2000 accelerator. The combined EG&G/EM and LLNL calibration results are shown in fig. 4.

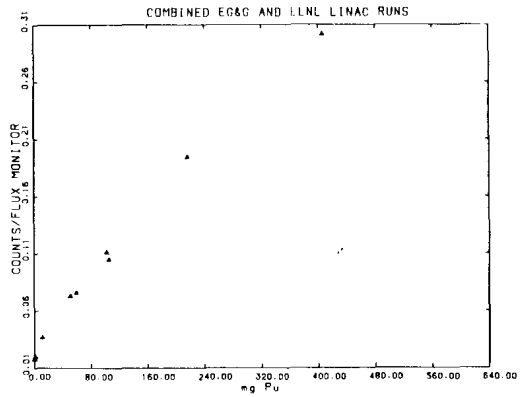

Fig. 4. Response of barrel counter versus mass of ^{239}Pu placed in center of counter. Combined EG&G/EM/DOE and LLNL linac runs.

Prompt Gamma Activation Analysis

The thermal neutrons are also captured on matrix nuclei, producing characteristic gamma radiation. By detecting this characteristic gamma radiation with a germanium detector, one may be able to determine and to some degree quantify the elements present in the matrix. Table I lists some elements that are typical matrix constituents in drums of TRU waste, their thermal neutron capture cross section, and their most intense and energetic gamma rays. Included are some elements that are particularly troublesome to either neutron or photon assay. The predominant capture gamma rays are energetic, thus minimizing the attenuation effects of the matrix. Typical gamma ray energies are in the neighborhood of 7 MeV, the neutron binding energy for many elements expected to be found in the TRU waste.

In addition to captures by the matrix nuclei contained in waste drums, the thermal neutrons will be captured on the nuclei constituting the moderating assembly. The moderating assembly contains several other elements in addition to carbon and hydrogen. These elements, which include iron, cadmium, aluminium, and boron, and which also may be found in the waste drums, will produce a nonzero or background assay when the empty drum is placed in the cavity.

For the measurements we used a Ge(Li) detector (about 6% efficiency, 1.7-keV FWHM resolution at 1332 keV) with a Princeton Gamma-Tech Model RG-11B preamplifier modified to handle high count rates.

Several matrices were placed in a 208-liter steel drum rotating in the moderating cavity. To assure that all the matrix was within viewing angle of the detector, the material was placed in a layer not exceeding 20 cm in thickness at the drum center.

A series of measurements was performed with hydrogen, cadmium, aluminium, iron, boron, chlorine, and calcium. The hydrogen was in the form of polyethylene, the boron is that of boric acid, the chlorine was in salt, and the calcium was in the form of lime. The other elements, cadmium, aluminium, and iron, were in the metallic form.

TABLE I.

TYPICAL ELEMENTS PRESENT IN BULK CONTAINS OF TRANSURANIC WASTE, THERMAL NEUTRON CAPTURE CROSS SECTION, AND CHARACTERISTIC NEUTRON CAPTURE GAMMA RAYS AND INTENSITIES

Element	Thermal Neutron Capture Cross Section	Gamma Ray Energy (keV)	Intensity (%)
Hydrogen	0.332	2223.25	100
Boron	0.103	4443.00 7005.10	75.96 47.96
Sodium	0.400	3982.00 6395.40	18.63 22.18
Aluminium	0.230	2960.01 7723.85	7.99 27.43
Silicon	0.160	3539.11 4934.39 6380.70	68.00 62.69 12.37
Chlorine	33.2	1950.93 6110.88	21.72 20.00
Calcium	0.430	1942.00 6419.90	72.55 38.89
Iron	2.55	7631.13 7645.45	28.51 24.13
Zirconium	0.185	933.70 6294.40	37.26 16.60
Cadmium	2450.0	558.60 5823.90	72.73 2.13
Gadolinium	49000.0	1186.50 6748.70	10.83 2.25
Mercury	37.6	1693.90 5966.20	14.13 13.86
Lead	0.170	6736.40 7367.70	5.04 94.06

Typical measurements of the incremental capture gamma yields accompanying the addition of matrix materials into the drum are shown in figs. 5 and 6. The gamma counts per time interval, the ordinate in these figures, are in all cases gross counts and accordingly contain counts from the constituents of the empty counter.

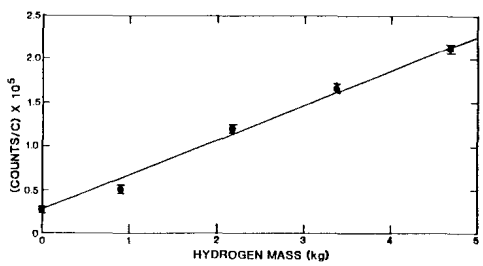

Fig. 5. Gross gamma ray counts per number of coulombs of collected charge due to added hydrogen as determined from 2223.25-keV line.

Conclusions

The feasibility of an electron-accelerator-based TRU waste assay system has been demonstrated. The system combines the neutron and photon interrogation of fissionable materials with the neutron capture identification of matrix material. Addition of the real time x-ray radiography station would make the matrix identification more complete. Use of the commercial radiographic electron accelerator with its one-button operation, makes the concept of mobile TRU waste assay system possible. We envision such a system to be based on two trucks and be able to travel to remote repository sites to perform special case assays. One of the possible concepts of this system is shown in fig. 7.

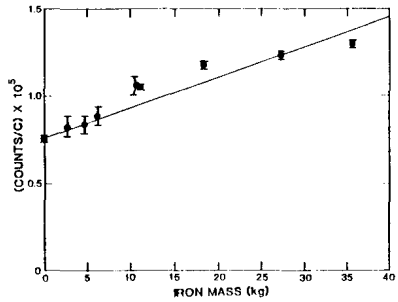

Fig. 6. Gross gamma ray counts per number of coulombs of collected charge due to added iron as determined from 7631.13- and 7645.45-keV lines.

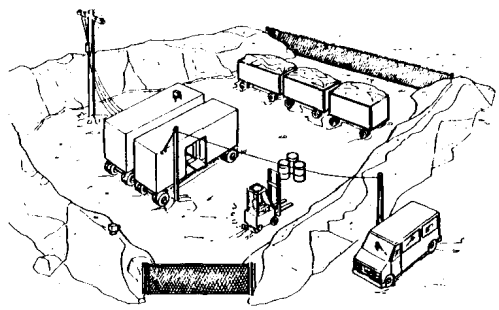

Fig. 7. Artist's conception of electron-accelerator-based mobile assay system for TRU waste. The linac, barrel counter with gamma capture spectrometer, and control and data acquisition system are housed in trucks or truck trailers. Necessary shielding is built on site from available materials.

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