NUCLEAR WASTE DRUM ASSAYER

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ABSTRACT

This paper describes a completely transportable, one-man-operable nuclear waste drum assayer capable of determining the concentrations of all radioisotopes in the drum. The principal radiation measuring devices are a segmented gamma-ray spectrometer utilizing a collimated germanium diode detector and two banks of neutron-sensitive proportional counters nearly surrounding the rotating drum. The neutron counters provide a sensitive measure of the transuranic activities present, and the germanium spectrometer provides a quantitative determination of the gamma-ray emitting radioactivities. The bremsstrahlung radiation from ^{90}Sr is also measured by the germanium detector and is used to quantify this isotope. Software algorithms determine the concentrations of the so-called difficult-to-measure radionuclides based on the directly measured nuclides and operator input regarding the source waste stream. The system can quantitatively sort waste into transuranic, low-level, or even the proposed new category of "below regulatory concern."

INTRODUCTION

The majority of radioactive waste produced in the world is packaged in 208 l drums and is generally contact handleable. These drums are further divided into categories of transuranic wastes (TRU), low level wastes (LLW), or perhaps nonradioactive wastes. Radioactive material is, after all, determined legislatively, since all materials undergo radioactive decay at some rate. Even the "stable" proton is expected to decay with some given frequency. Hence, it has been proposed to establish a category of waste called Below Regulatory Concern (BRC) which would define a concentration limit below which all waste would be considered to be nonradioactive and could be disposed of in sanitary landfills.

Large quantities of stored wastes are held at many sites and few, if any, records exist regarding the content of these drums. Since the cost for disposal of transuranic waste will be many thousand dollars per cubic meter, the disposal cost of LLW is ~$1000/m^3, and the cost for BRC waste is ~$2.00/m^3, it behooves the waste generator to qualify his waste for the cheapest possible category. This paper describes an instrument capable of making that categorization.

EQUIPMENT

The assay instrument is on rollers and can be moved anywhere within a building by one man. The system has a self-loading/unloading trailer, is easily towed, and can be readily moved anywhere. The travel trailer is designed with a fold-back roof and sides so that drums can be assayed without unloading the instrument. This design is extremely advantageous when drums must be analyzed outdoors or when only a few drums are located in several places.

The assayer uses two measuring techniques: segmented gamma-ray spectrometry and neutron counting. A drum to be assayed is placed on a rotating turntable by a self-contained electric hoist. Two banks of neutron sensitive proportional counters almost surround the rotating drum and measure the neutrons emitted by the spontaneous fission of transuranic isotopes or the reactions induced by alpha particles on light isotopes present in the matrix materials.

Simultaneously with the neutron measurement, a collimated high-resolution germanium diode gamma-ray spectrometer vertically scans the rotating drum to measure the intensity of gamma rays as a function of energy emanating from the drum. Most fission and activation products (and even some transuranics) emit measurable quantities of monochromatic photons that serve as "fingerprints" of those radioisotopes.

All data are acquired and reduced by the onboard computer, which also controls all mechanical operations except the loading and unloading of the drum.

The system does not use a transmission source to correct for matrix effects since this approach generally only leads to an appropriate correction for drums containing homogeneous waste. For nonhomogeneous drums, the transmission source leads to the average density of the drum which may or may not have any relationship to the distribution of radioisotopes within the drum. This assay system utilizes a "tomographic" approach to matrix correction which considers only the actual attenuation of radiation from within the drum. This technique will be discussed in more detail below.

DATA ACQUISITION AND REDUCTION

The gamma-ray spectrum from the drum is obtained with a lead collimated germanium diode gamma-ray spectrometer and stored in the onboard computer. The operator may choose between a continuous spiral scan or a number of horizontal segments. The latter option has the potential to provide the greatest amount of information and is the recommended mode. The number of segments is also optional and is somewhat interdependent on the aperture of the collimator to guarantee complete coverage of the drum. In practice, it is most advantageous to add the data from each segment in the computer and analyze the composite spectrum. Each individual segment can be analyzed if addi-

tional detail regarding the distribution of radioisotopes within the drum is desired. Eleven segments within a drum have been found to provide sufficient detail, and a 100 second counting time per segment provides adequate sensitivity. Hence, an entire assay, including time for repositioning the germanium spectrometer, requires on the order of 20 minutes.

The gamma-ray spectrum is deconvoluted using the computer code RAYGUN (1). Gamma-ray efficiency curves as a function of energy are obtained from calibration standards prepared in drums having different matrix materials. This method leads to a family of efficiency curves which change fairly regularly as a function of barrel weight. The "first cut" efficiency curve used to quantify the radioisotope concentrations in the drum is, therefore, based on the weight of the unknown drum. Since the RAYGUN code calculates the concentration of each radioisotope based on each gamma ray that it recognizes, the concentration of isotopes decaying by multiple gamma-ray emission, such as ^{134}Cs and ^{154}Eu, is calculated several times as a function of gamma-ray energy. If these calculated concentrations demonstrate a trend as a function of energy, that is, they do not represent a random distribution about the average, the software selects a different efficiency curve from an appropriately lighter or heavier standard until the calculated concentration based on each gamma ray is constant (within errors) as a function of gamma-ray energy. In this manner, a matrix correction is affected for the actual distribution of radioactivity within the drum and is independent of the distribution of inert heterogeneous materials in the drum.

In the absence of large quantities of gamma-emitting fission or activation products, the isotopes ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{241}Pu, ^{241}Am, ^{243}Cm, and ^{244}Cm are generally measurable at adequate sensitivity to separate TRU and non-TRU wastes based on their gamma-ray emissions. The computer utilizes these measured transuranics and the code ORIGEN (2) to predict the concentration of ^{238}Pu, ^{240}Pu, and ^{242}Pu expected to be present. The neutron yield for the assemblage of transuranic isotopes, assumed to be present as the oxide, is calculated and compared to the measured neutron emission rate obtained with the neutron sensitive proportional counters. The neutron counters have also been calibrated with standard drums containing known concentrations of appropriate transuranics in a variety of matrix materials. If the total TRU concentration based on gamma-ray counting does not agree with that based on neutron counting, within errors, the operator is flagged that an anomaly exists. One fairly common cause for an anomaly is the presence of cadmium or boron in the drum which strongly curtails the neutron emission rate. This is generally manifested in emission of recognizable prompt capture gamma rays which are identified by the computer, and an appropriate correction is applied to the neutron based TRU concentration derived from the intensity of the capture gamma-ray lines.

In many circumstances, the presence of fission or activation products masks the detection of gamma rays from transuranic isotopes. However, a positive neutron count identifies the presence of transuranics. The net neutron count rate is used to quantify the total TRU concentration of the drum based on the expected neutron yield from a mixture of transuranic isotopes of the same composition as that in the drum. The composition of transuranics in the drum can be manually entered into the default logic of the computer if known from a priori information, such as independent measurement, or can be determined by the computer itself based on measured fission product concentration ratios, such as ^{134}Cs/^{137}Cs, and the ORIGEN code for the appropriate reactor conditions and assuming no chemical separation. Fortunately, the neutron yield as a function of alpha activity for most transuranics is fairly constant with the exception of a few isotopes with high spontaneous fission branching fractions such as ^{242}Cm, ^{244}Cm, and ^{252}Cf.

Additional software built into the system contains algorithms that determine the "difficult-to-measure" radioisotopes, such as ^{63}Ni, ^{65}Ni, ^{129}I, etc., on the basis of previously measured analog ratios (3), the measured concentrations of specific fission and activation products, and operator input regarding the source of the waste.

The ^{90}Sr concentration in the waste drum can also be determined directly by measurement of the bremsstrahlung radiation in the germanium spectrum. A library of "pure" net spectra obtained from standards are stored in the computer for each radioisotope likely to be encountered and as a function of drum weight. The appropriate library spectrum is called from memory for each radioisotope observed in the unknown drum spectrum, is normalized on the basis of photopeak intensity, and is "stripped" out of the unknown spectrum leaving only a pure ^{90}Sr bremsstahlung spectrum from the unknown drum. The shape of this spectrum is verified against the appropriate library ^{90}Sr spectrum, and the ^{90}Sr is quantified on the basis of intensity.

All data acquired and all calculations performed by the patented software (4) are stored on hard disc and backed up on floppy disc for archival or subsequent manipulation. An onboard printer provides immediate hard copy of as

much or as little data as desired and can even be used to print a shipping manifest.

CONCLUSIONS

Typical sensitivities for the system are

fission/activation products = 1 pCi/g,
transuranics = 1 nCi/g, and
^{90}Sr = 100 pCi/g

which are sufficient to clearly classify a waste drum as TRU, LLW, or BRC.

A statistically significant number of radioactive waste drums from several commercial nuclear power plants have been analyzed with the system, and the majority were found to contain radioisotope levels below the proposed BRC guidelines. The savings in disposal cost to the public utilities that operate nuclear power plants would be $600,000 to $800,000 per year per reactor, and the assay system would pay for itself in a fraction of a year.

A video of the system is available.

REFERENCES

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