DEVELOPMENT OF NON-DESTRUCTIVE ACTINIDE ASSAY METHODS FOR RADIOACTIVE WASTE

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ABSTRACT

The need to measure the actinide content of radioactive waste has led to the development at Harwell of a range of radioactive waste assay equipment. This paper describes the current status of a number of Harwell standard assay systems, giving the performance and limitations of passive gamma, passive neutron and active neuron interrogation systems. Methods have been developed to correct for the effects of the waste matrix material on the measurements. Other techniques are being developed to tackle the problem of the self-shielding in lumps of fissile material.

INTRODUCTION

Radioactive waste arises from a wide range of operations both within the nuclear industry and elsewhere. Between being produced and final disposal, the waste may undergo transport and storage by various operators in different sites. It also is likely to be involved in various processing operations such as sorting and compaction. It is important that certain information about the waste should always be available so that sensible decisions can be taken about the way the waste is treated and handled. Information on the external dose-rate is necessary for handling operations; the special nuclear material content is necessary for both accountancy and criticality control; the content of a range of radionuclides relevant to national waste disposal limits may be required before the waste can be placed in a repository.

In certain cases, for particular types of waste, it may be possible to obtain sufficient information on the waste as it arises, and to keep sufficiently detailed records in order to satisfy all future requirements. However, in many cases such information is not available when the waste is produced, or the information becomes irrelevant, for example when waste is sorted into shreddable and non-shreddable streams with the transfer of activity between the streams being unknown. In such circumstances it is necessary to make measurements on the waste. Destructive analysis can produce very precise estimates of radioactive material, but it has major shortcomings: Only a fraction of drums can be measured, the sampling within a single drum is likely to be non-representative in the case of solid heterogeneous waste, and handling of active material and further waste processing is necessary. Non-destructive assay too has shortcomings, and it is the purpose of this paper to outline what can be achieved within these shortcomings and how they can be minimized.

This paper considers the following techniques used to determine the actinide content of the drum: Passive gamma assay, passive neutron assay and active neutron interrogation. Passive gamma-ray analysis gives information on those nuclides which are important in determining the external dose-rate from the drum. Passive neutron measurements mainly give information on the plutonium, curium and californium content of the waste. Active neutron interrogation mainly gives information on the fissile content of the waste (^{239}Pu and ^{235}U). A knowledge of these nuclides satisfies the requirements of dose-rate, nuclear materials accountancy and criticality safety. These are sufficient for the short term handling and storage of the material. Some information which may be required for admission of the waste to a repository (such as total alpha activity) may be available from a combination of this information (e.g., Pu-239 content plus the plutonium isotopic composition). Other information, in particular the total beta activity, may be available in certain cases from isotopic correlation techniques.

PASSIVE GAMMA

Characteristic gamma-rays are emitted by many common radioactive materials including plutonium and uranium. It is possible by using a high resolution germanium detector to obtain unique signatures for many radionuclides. The ability of an external detector to make a quantitative measurement of the amount of radioactive material in the waste depends on a number of factors. The main difficulty arises due to non-uniform distribution of the gamma-emitting material in an absorbing matrix. The absorption of the gamma-rays depends on their energy and also on the density and composition of the matrix. Table I shows the attenuation of various gamma rays through a variety of matrix materials.

TABLE I
Gamma Ray Transmission Under Various Conditions

Gamma Energy (MeV)	Transmission Through 10 cm of Solid Material: Polyethylene 0.9 g/cm^3	Concrete 2.3 g/cm^3	Iron 7.87 g/cm^3
1.3 (^{60}Co)	0.77	0.30	0.15
0.67 (^{137}Cs)	0.74	0.16	0.10
0.41 (^{239}Pu)	0.74	0.11	0.09
0.19 (^{235}U)	0.76	0.06	0.02

Corrections can be made for the attenuation of the matrix in the drum if the transmission of gamma-rays of similar energy can be measured through the drum. In order

to allow for variation in composition of the drum, the attenuation measurement is best carried out in a number of small slices.

Another problem is the self-attenuation of gamma-rays by the source material itself (e.g., a lump of plutonium metal). Approximate values for the escape probability from a number of spheres of different materials is given in Table II.

This self-attenuation problem can be partly solved by considering the ratios of gamma-rays of different energies which escape from the source, but can represent an important source of uncertainty on the measurement.

Figure 1 shows a Segmented Gamma Scanner for use with drums up to 200 liters. The system consists of a turntable onto which the drum can be loaded. This rotates at 10 rpm and can accept containers up to 1,250 kg maximum weight. To the side of the turntable a high resolution intrinsic germanium detector views the drum through a lead collimator. A hydraulically-operated platform is used to raise the detector enabling the drum measurement to be made in fixed vertical increments of 50 mm; 18 increments are used to measure a 200 liter drum. An external radiation source located on the far side of the drum from the detector is used to measure the density of the waste matrix. Gamma spectra are accumulated by a multichannel analyzer, with complete system control and data processing being performed by a dedicated personal computer.

Fig. 1. Segmented Gamma Scanner for 200 liter drums

The system is capable of operation with matrix absorption up to 90% with gamma radiation levels up to 0.4 mGy/hr. The typical ^{239}Pu measurement range is from 0.2 g in a single segment to 10 g in a single segment. Typically, the 1σ spread on plutonium mass for ten 0.1 g plutonium oxide spheres uniformly distributed in a segment for a segment counting time of 5 minutes is 8% (10%) for wastes of density 0.3 (0.5) g/cm^3 respectively. The accuracy of measurement is dependent on geometrical variations in the position of the plutonium in the drum, this is minimized to typically ±8% by drum rotation and horizontal collimation. The major sources of error are sample self absorption and system calibration. When the result of the transmission measurement indicates the presence of a high density matrix, the drum is normally remeasured using one of the neutron measurement systems described below.

PASSIVE NEUTRON

Passive neutron emission is caused by spontaneous fission (e.g., ^{240} Pu, Cm and Cf) and (α, n) reactions on light elements. These two types of neutron are generally distinguished using a coincidence technique, making use of the fact that spontaneous fission produces several neutrons per event whereas (α, n) neutrons are produced singly. The effect of lumps of source material are much less for the typical fission neutrons from plutonium than for gamma-rays. In addition, the effect of the matrix on neutrons is different from that on gamma-rays and thus the use of both techniques is complementary and covers a wider range of waste types. Generally, the soft waste (tissues, etc.) can be measured well with the SGS system, whereas harder (non-shreddable) material can be measured well with a passive neutron system.

Figure 2 shows a passive neutron monitor for drums up to 200 liters. The assay chamber consists of a rectangular steel frame with a hinged door for loading and unloading the waste drum. All six sides of the structure are clad

TABLE II
Escape Probabilities From Spheres For Gamma-rays

Mass of Sphere g	Fraction of Gamma Rays Escaping Pu metal (414 KeV)	PuO_2 (414 KeV)	U metal (186 KeV)	U_3O_8 (186 KeV)
0.1	0.70	0.79	0.24	0.37
1.0	0.49	0.62	0.16	0.20
10.0	0.28	0.40	0.05	0.09
100.0	0.14	0.21	0.025	0.04

Fig. 2. Passive Neutron Monitor for 200 liter drums

internally and externally with 25 mm and 38 mm thick polyethylene sheet respectively, to form 75 mm air gaps. The air gaps house a total of 36 ^{3}He proportional counters (two atmospheres pressure), mounted horizontally, with the intercounter distance adjusted to give uniformity of coincidence counting efficiency within ±5%. The system is provided with microprocessor-controlled coincidence counter electronics, with operational control and data analysis performed by personal computer. This system has been calibrated for a variety of waste types. The limit of detection for suitable waste streams, in coincidence mode is 10 mg of ^{240}Pu. When used in totals counting mode, the chamber can give an upper level indication of the presence of ^{240}Pu of 6 milligrams. The total plutonium content may be obtained from a plutonium isotopic measurement on the passive gamma system.

Figure 3 shows a passive neutron monitor for packages up to 12 liters. This transportable system is designed for both the measurement of small waste packages for waste management purposes and also cans of plutonium product for nuclear materials accountancy. The assay chamber consists of an annular polyethylene well counter fitted with 18 ^{3}He proportional counters 25 mm diameter and four atmospheres pressure. As the polyethylene annulus forms and undermoderated assembly, the efficiency at the top and bottom of the chamber is increased preferentially with respect to the center by addition of outer polyethylene rings. In conjunction with polyethylene and plugs this result in a coincidence counting efficiency uniform to within ±1.5% over 450 mm of the total 460 mm internal height. Coinci-

Fig. 3. Transportable Passive Neutron Monitor

dence electronics and a personal computer form part of the total system package.

Figure 4 shows a modular passive neutron monitor for irregular shaped packages. The requirement for a transportable passive neutron assay system which could be offered to the waste package and configured to surround it, lead to the development of this instrument. The main components are a series of 8 polyethylene slabs 1,050 mm x 270 mm x 100 mm each housing three ^{3}He proportional counters 50 mm diameter and two atmospheres pressure. The slabs can be configured in a variety of ways to surround, for example, a 200 liter drum in circular format or a rectangular passage in two parallel rows of four slabs. Again, the system is provided with coincidence electronics and a personal computer.

ACTIVE NEUTRON INTERROGATION

Passive neutron detection systems have two particular limitations; they cannot measure the uranium content of the waste and, in general, they cannot distinguish between neutrons from plutonium, curium and californium. This latter limitation can cause difficulties, as the number of neutrons emitted from one milligram of ^{252}Cf is of the same order as is emitted by 2,000 kg of ^{240}Pu. Active neutron interrogation systems can provide a direct measure of ^{239}Pu and ^{235}U

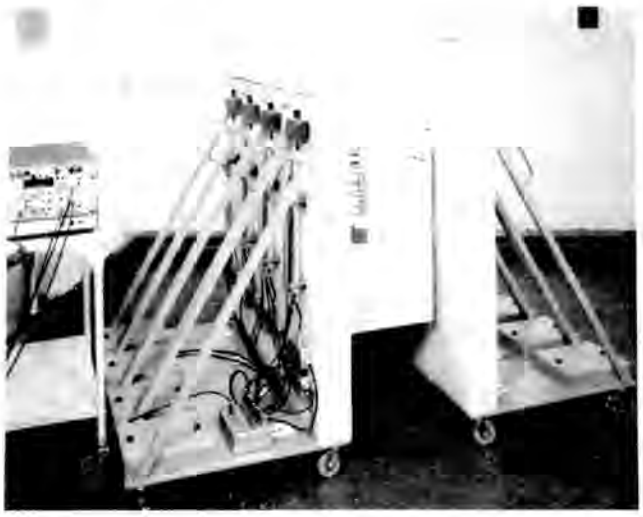

Fig. 4. Transportable Modular Passive Neutron Monitor.

which are of most significance for criticality control and nuclear materials accountancy.

Active neutron interrogation refers to a technique in which external neutrons are used to cause emission of secondary neutrons from any fissile material in the waste. The source neutrons need to penetrate the bulk material of the waste (the "matrix") and then penetrate the fissile material itself. The secondary neutrons need to escape from the sample, pass through the bulk material and be counted in the detection system.

The ability of the interrogation neutrons to penetrate the matrix depends both on the properties of the matrix and the energy of the interrogating neutrons. If the penetration is poor, the response to fissile material depends on its distribution within the waste drum. For thermal neutrons, the most significant matrix property is the absorption cross-section, which is very high for materials like cadmium and boron and high for material like hydrogen and iron. The neutron penetration depends on the product of the absorption cross-section, the density of the bulk material and the path length through which the neutrons have to travel (i.e., the size of the waste package). Higher energy neutrons can penetrate cadmium, boron and iron relatively easily, but they are moderated by hydrogen-containing matrices to give thermal neutrons which have the problems of absorption mentioned above. Generally, the matrix has a large effect on the response of particular systems to fissile material. For specific waste types, it is possible to produce calibrations relating to the measured counting rate to the amount of fissile material. In operating conditions where it is necessary to measure mixed or unknown types of waste, it is important to have an arrangement to monitor the type of waste and select an appropriate calibration. One particularly difficult matrix is cement encapsulated waste because of its high density and high hydrogen content. A special technique has been developed to deal with this waste type and this will be described in a later section.

The neutron flux in the vicinity of the fissile material is determined by the composition of the matrix, but the reaction rate per gram of fissile material depends on its configuration. In thermal rate per gram of fissile material depends on its configuration. In thermal neutron systems, neutron self-shielding becomes important for very small amounts of plutonium or highly enriched uranium. For example, in the case of metallic uranium (95% ^{235}U), the reaction rate in a 0.1 g sphere is only about 25% of that of dilute material. For a 10 g sphere, the rate is about 6% of the rate of dilute material. When the fissile material is more dilute, because the density is reduced (e.g., plutonium oxide), or because non-fissile material is present (low-enriched uranium), the effects of self-shielding are smaller. For example, the reaction rate in a 100 g sphere of uranium oxide (2.5% ^{235}U) with a density of 5 g/cm^3 is about 80% of the dilute reaction rate.

The secondary neutrons from fission are fast and have little difficulty in escaping from most fissile material samples which are expected to be encountered in waste, in contrast to some of the passive gamma emissions.

The escape of the secondary neutrons from the matrix is affected by the same processes as affect the matrix penetration of the interrogating neutrons. The probability of escape depends on the energy of the neutrons and, generally, higher energy neutrons escape more easily.

The efficiency of the detection system depends on the type of neutrons it is designed to detect. In differential dieaway systems, it is necessary to discriminate against thermal neutrons. Therefore, the thermal neutron detectors are wrapped in polyethylene and then cadmium. This gives a relatively low absolute detection efficiency and also means that any matrix which moderates the secondary neutrons to a significant degree reduces the detection efficiency. Total neutron counting systems such as those used for Cf shufflers are less sensitive to the emitted neutron spectrum as they count thermal as well as higher energy neutrons. They also have a higher absolute detection efficiency. The detection system may need to operate in the presence of high gamma dose-rates from certain samples and may also need to withstand (but not necessarily operate in) high neutron doses from the primary neutron source.

Several active interrogation instruments for the measurement of fissile material have been developed at Harwell. These instruments have been designed to minimize the problems which have been outlined above. The development relies on a combination of modelling with neutron transport codes, and measurements using experimental prototypes. The differential dieaway system and Cf

shuffler are general instruments designed to cope with a variety of waste streams and package sizes. The most appropriate choice for a particular application depends on the waste matrix type, the fissile material content of the waste and the required measurement limits. The asymmetric dieaway system which originated at Harwell was designed specifically to deal with the difficult problems posed by cement encapsulated waste drums.

Differential Dieaway System

The differential dieaway system uses a primary pulsed neutron source of 14 MeV neutrons. These fast neutrons slow down to thermal energies in about 10 μseconds and the resulting thermal neutrons have a lifetime of 100 s of μseconds. These thermal neutrons can produce fast neutrons by causing fission in any fissile material which is present. The detection system is sensitive to fast neutrons but insensitive to thermal neutrons and so gives a signal proportional to the amount of fissile material present. Differential dieaway systems have been designed and operated at Harwell for a range of waste from sizes from 50 (13 U.S. gal) to 500 liters (130 U.S. gal). Figure 5 shows the 500 liter version.

One important feature of the Harwell series of dieaway chambers is the monitoring of the waste type. As has been described above, the type of waste in the drum can have a large effect on the calibration of the device. The Harwell system uses two additional measurements to characterize the waste type. The first involves the measurement of the thermal flux distribution in the chamber. This flux is modified by the presence of the matrix and its absolute value and time behavior allows a correction to be made for the effect of the matrix on the thermal neutrons. The second measurement is made using a californium source which is placed in the center of the chamber beneath the drum. The waste matrix affects the neutrons from the source and hence, the count-rate of the main detectors gives information on the matrix type. These two measurements enable measurements on unknown waste types to be related to a suitable calibration. This has been demonstrated experimentally for a wide range of matrices in 200 liter waste drums (1). This procedure allows drums containing unknown bulk matrices to be assayed to an accuracy of 25% provided that the matrix falls within the calibrated range.

Fig. 5. Differential Dieaway 500 litre Assay Chamber.

For 200 liter waste drums of combustible or non-combustible waste with a bulk density of between 0 and 1 g/cm^3, the limit of detection is a few milligrams of Pu or ^{235}U in a 15 minute measurement using a pulsed neutron tube with an output of 10^8 neutrons/second. Similar performance can be obtained from the 500 liter assay system.

In addition to the calibration of the standard systems for a wide range of waste types, work is currently in progress on new techniques to solve the problem of self-shielding in lumps of fissile material. One possible approach is the DANI (Dual Active Neutron Interrogation) technique in which both the prompt neutrons from the thermal dieaway induced fission and delayed neutrons from fission are recorded in the intervals between the primary source pulses. The delayed neutrons are produced not only from the thermal fission but also from fission caused by higher energy neutrons from the primary pulse. This delayed neutron signal is therefore less affected by self-shielding in the sample than the prompt signal from thermal fission. The ratio of the two results can be used to give information on the presence of lumps of fissile material in the waste (2).

Californium Shuffler

In a californium shuffler system, an intense moveable californium source is used to irradiate a waste drum for a few seconds. The source is then withdrawn and a count is made of the number of delayed neutrons which are emitted. This process is repeated a number of times until sufficient statistical accuracy has been achieved. The detection chamber is designed to measure neutrons of all energies which are emitted from the sample during the counting phase. This property makes the shuffler chamber well suited to passive neutron measurements in addition to the active assay. The Harwell waste assay shuffler, shown in Fig. 6, was designed to accommodate 200 liter (55 U.S. gal) waste drums, but has also been calibrated for 50 liter (13 U.S. gal) drums.

In this system, also, it is important to monitor the type of waste matrix. This is done by monitoring the thermal and epicadmium neutron flux which is present beneath the waste drum when the primary californium source is irradiating the drum. These measurements show to what extent

Fig. 6. Californium Shuffler Assay System for 200 liter drums

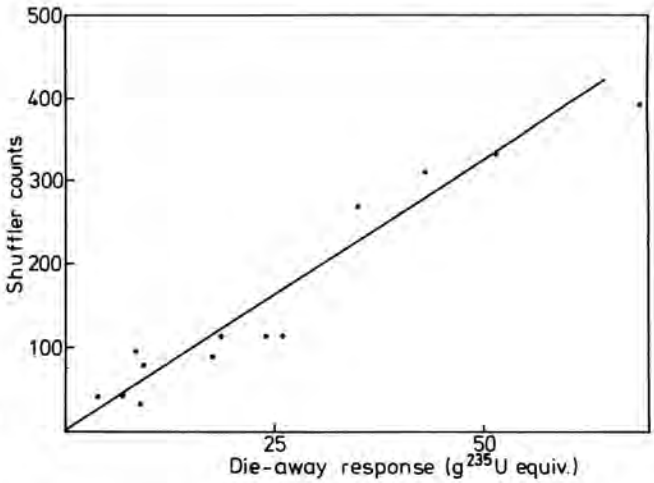

Fig. 7. Comparison Between Results from Cf Shuffler and Dieaway Assay

the thermal neutrons are affected by the matrix and allow a correction to be made to the response of the system for each waste type. This has been demonstrated for a range of matrix types (3).

The source movement mechanism is controlled by a personal computer. The same computer controls the data acquisition system (using Harwell electronic modules) and performs data analysis.

The limit of detection of a waste assay system depends on the size of the primary neutron source. For 200 liter drums with bulk waste density in the range 0 - 1 g/cm^3, the limit of detection is a few tens of milligrams of plutonium or ^{235}U in a ten-minute measurement using a 10^9 n/s ^{252}Cf source. The system also measures the passive neutron output form the waste drum with total and coincidence counting electronics and this gives a detectable limit of 10 mg of ^{240}Pu.

Other shuffler systems have been designed for special applications; e.g., for high gamma activity waste and for measuring hold-up in a spent fuel dissolver. An investigation into the use of epicadmium interrogation neutrons has shown that for some waste types, the effect of self-shielding can be considerably reduced. For example, in a 10 g sphere of 95% ^{235}U, the thermal self-shielding reduces the response to about 6% of the dilute value but in an epicadmium flux, the reduction is to about 35% of the dilute value.

Comparison of Differential Dieaway and Californium Shuffler

A series of actual waste drums containing fissile material were selected in order to compare the performance of the differential dieaway system and the californium shuffler. Figure 7 shows a plot of the counts recorded in the shuffler assay system for each of the drums as a function of the fissile mass measured using the differential dieaway technique. There is a good measure of agreement. Measurement and analysis are continuing to complete the study of this set of drums. At a later stage, it is intended to unpack the present drums in order to more clearly relate the measured values to the actual contents.

Cement Encapsulated Waste Measurement

Work is in progress on the use of a linear accelerator (linac) to provide the neutron source for this interrogation technique (5). The neutron source intensity is much greater than a neutron tube and the limit of detection is expected to be reduced to 0.1 g of fissile material. The linac can also be used as a gamma radiation source in order to check the total actinide content of the waste package using a photofission method. In an integrated radioactive waste inspection facility a linac may be used to check the package integrity using radiography and therefore the additional use of the linac as a neutron source may be cost-effective.

SUMMARY

The performance of three types of actinide assay systems has been given: Passive gamma, passive neutron and active neutron assay. Each system gives information on the actinide content of particular waste types under certain conditions.

Passive gamma measurements give the plutonium content (and possibly ^{235}U) content of 200 liter drums of low density waste. The limit of detection is a few grams.

If the drum is shown by the gamma measurement to

absorb gamma rays strongly, or if lower levels of detection are required, a passive neutron chamber can be used to give estimates of ^{240}Pu at the 10 milligram level.

If the drum contains other neutron emitters (Cm or Cf) or the ^{235}U is required, then active neutron interrogation is needed to give the ^{239}Pu and ^{235}U content. The lower limit of detection for suitable waste forms is a few milligrams.

If the fissile material is in the form of large lumps, errors are caused in all of the above methods. In some waste types (non-moderating), it is possible to overcome this problem by Dual Active Neutron Interrogation (for the differential dieaway technique) or epicadmium interrogation (for the californium shuffler).

Because the performance of each system depends on the type of waste, it is extremely useful from the assay point of view to segregate (as far as possible) different types of waste into different streams.

REFERENCES

1. ARMITAGE, B. H. and SHERWOOD, A. C., "Matrix Effects in the Neutron Dieaway Assay of Fissile Material in 200 Liter Drums," 9th ESARDA Symposium on Safeguards and Nuclear Materials Management, London, 1987.
2. ARMITAGE, B. H. and COGBILL, M. J., "Dual Active Neutron Interrogation of Fissile Material in Waste Drums," Proc. 11th ESARDA Symposium on Safeguards and Nuclear Materials Management, Luxembourg, May 1989.
3. PACKER, T. W., SWINHOE, M. T. and SYME, D. B., "Non-Destructive Assay of the Fissile Content of Radioactive Waste Using a Californium Shuffler," Proc. 11th ESARDA Symposium on Safeguards and Nuclear Materials Management, Luxembourg, May 1989.
4. PACKER, T. W. and SWINHOE, M. T., "The Differential Dieaway Technique Applied to the Measurement of the Fissile Content of Drums of Cement Encapsulated Waste," Harwell Laboratory Report AERE R13137, 1988.
5. MOLESWORTH, T. V., FINDLAY, D. J. S., SENE, M. R. and SWINHOE, M. T., "Development of an Integrated Assay Facility for Immobilized Intermediate-Level Waste," Proc. BNES Conf. on Radioactive Waste Management, Brighton, UK, May 1989.