DEVELOPMENT OF AN INTEGRATED ASSAY FACILITY

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ABSTRACT

Initial results of active neutron and active gamma-ray interrogation of a 500 litre cemented simulated CAGR intermediate level radioactive waste drum are described. The basis of the interrogation systems was the Harwell electron linear accelerator HELIOS, which was used to produce the interrogating neutrons and gamma-rays. Several sets of neutron detectors were located around the drum to count signature neutrons. The responses of the system were measured by placing known samples at many different locations within the drum. In general, measured responses confirmed calculated responses. Good agreement was obtained for the azimuthal angle dependences. The absolute responses agreed well for gamma-ray interrogation, but the calculations were apparently over-estimates for neutron interrogation. Those aspects requiring consideration in the practical application of assay techniques are identified.

INTRODUCTION

Regulatory authorities and waste managers may require waste packages to be examined before disposal in a repository to ensure that total or specific radioactivity limits are being respected. For the particular case of cement encapsulated Intermediate Level Waste (ILW), a non-destructive method of comprehensive assay has been proposed by a consortium of Taylor Woodrow, Rolls Royce, Plessey Controls and Ray Technologies. To assist further development, the UK Department of the Environment and the Commission of the European Communities awarded a research contract to Taylor Woodrow to investigate appropriate examination techniques and their application to a practical assay facility. The work is also supported by the UK Nirex Ltd., the Inspectorate of Nuclear Installations and the industrial consortium, and forms part of the European Atomic Energy Community's cost sharing research programme on "Development of Test Methods for Quality Assurance" (Contract Nos. FIIW 0102, 0244 UK(H)). This paper describes work carried out, mainly at Harwell, forming a major part of the overall program.

The quality checking system envisaged in the present work would incorporate gamma-ray radiography, passive gamma-ray scanning, passive neutron counting, and active neutron and active gamma-ray interrogation. The data provided by these measurements would be integrated to produce a best estimate of the radioactive contents of drums. Of the five techniques, gamma-ray radiography, active neutron interrogation (for assessing the fissile content (eg ^{235}U) of drums) and active gamma-ray interrogation (primarily for assessing the total actinide content (in practice mainly ^{238}U) of drums) would be electron linac based. For active neutron interrogation, the use of an electron linac driven fast neutron source, with its much greater output than the alternative compact D/T neutron generator tube, is a significant advantage when interrogating drums with a strongly neutron absorbing cement matrix. Previous work on neutron and gamma-ray interrogation systems using electron linacs has been carried out at Los Alamos (1). However the present systems, which draw on experience obtained from the work performed under the UK QCTF programme (2, 3), are different in several aspects, in particular those of package size (500 litre instead of ~200 litre) and density (cement encapsulated instead of non-encapsulated), both of which make interrogation for actinide content more difficult.

Because in general the design and performances of active neutron and gamma-ray interrogation systems (especially electron linac based active neutron systems) are less advanced than gamma-ray radiography, passive gamma-ray scanning and passive neutron counting systems, most effort within the present programme has been directed at the active neutron and gamma-ray systems. Previous descriptions of earlier stages of the present work have appeared elsewhere (4, 5, 6).

EXPERIMENTAL FACILITY

An experimental active interrogation facility has been assembled in the Low Energy (LE) Cell of the Harwell electron linear accelerator HELIOS. The facility can operate in both neutron and gamma-ray modes, using energy analyzed ~5, ~7 and ~15 MeV electron beams. A schematic diagram of the assembly is shown in Fig. 1.

In the neutron mode, fast neutrons from a composite tantalum/beryllium (3 g/cm^2 Ta, 200 x 200 x 50 mm Be) target driven by a ~15 MeV electron beam penetrate the cement drum. After thermalization, these neutrons produce a thermal flux throughout the drum which decays with time. This thermal flux induces fission of fissile actinide isotopes within the drum, and a small fraction of the resultant fast fission neutrons are counted in fast neutron detectors outside the drum. In principle the net counts from the detectors are proportional to the fissile actinide density within the drum.

In the gamma mode, bremsstrahlung with a ~7 MeV endpoint from a tantalum radiator (1 g/cm^2 Ta) penetrates the drum and induces photofission and photoneutron reac-

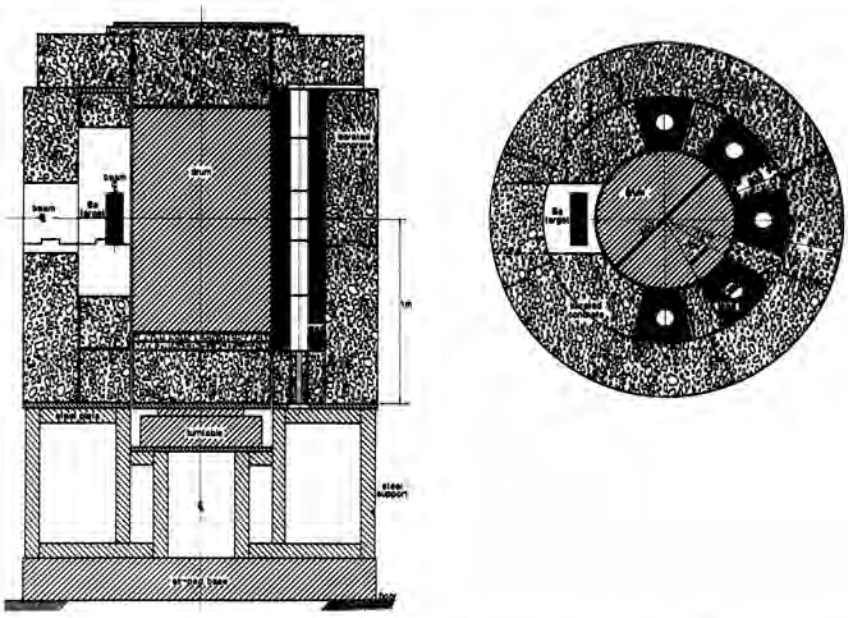

Fig. 1. Schematic representation of the experimental composite interrogation system. The system is shown configured in its neutron mode. Because only one electron beam line is available for the present work, the whole assembly has to be translated by 500 mm and rotated through 90° to change to gamma mode. To facilitate this the assembly has been constructed with a hovercraft-type base.

tions in all actinide isotopes within the drum. Again, a small fraction of the resultant neutrons is counted, in this case by thermal neutron detectors outside the drum. The neutrons photoproduced by the bremsstrahlung from the deuterium naturally present in the hydrogen in the water in the cement can be measured separately and then subtracted by reducing the bremsstrahlung endpoint energy to ~5 MeV. This energy is well below the characteristic fission barrier and neutron separation energies of ~6 MeV in actinide nuclei. The net counts are in principle proportional to the total actinide density within the drum. There is a useful synergy between the neutron and gamma modes, in that measurement of the water content of the drum from photoproduction from deuterium helps to establish the neutronics of the drum for both neutron and gamma-ray interrogation.

In both neutron and gamma modes, the source of interrogating radiation and the neutron detectors are located essentially on opposite sides of the drum, thereby producing a rough balance between the effects of variation of attenuation of the interrogating radiation and the variation in neutron detection efficiency. It is therefore important that neutrons produced in or near the source do not leak around the drum to the detectors. Hence the drum and detectors are enclosed in borated concrete shielding lined on the inside with cadmium sheet everywhere except over the 135 and 225° detector channels. In addition, because of the intense gamma-flash from the linac and because of possible radiation dose rates of ~1000 rads. hour -1 which are to be expected near the surfaces of 500 litre ILW drums (7), the counters have to be protected by lead shielding. The thickness of lead surrounding the counters in the present arrangements is ~100 mm.

The assembly has been designed to accommodate a wide range of counters. The detector channels at 90, 180 and 270° are intended primarily for fast neutron detector packages for neutron interrogation, and the channels at 135 and 225° for thermal neutron detectors for gamma interrogation. Several different detector packages have been used, incorporating 50 mm diameter ^{3}He counters, "4-packs" of 25 mm ^{3}He counters (each of which should recover from overload more quickly than a single 50 mm counter), and 50, 25 and 13 mm $^{10}BF_3$ counters.

The data recorded from the experimental assembly are the time spectra (relative to the electron beam pulses) of neutrons counted in the detector packages, and the electron beam charge delivered to the targets and associated collimators. The heart of the data acquisition system is a multi-input, multi-shot time digitiser as used routinely by neutron time-of-flight experiments for nuclear physics and nuclear data measurements on HELIOS. For the present purpose, the digitiser has been configured with eight separate inputs each with 1 μs time channels extending out to 2 ms, and is connected to a DEC LSI-11/73 computer. For each linac pulse, the digitiser clock is started with the electron gun trigger pulse, and the digitiser records the times of occurrence of neutron detector signals. The signals from the counters are also recorded in blind CAMAC scalers and, (so that the operation of the counters may be continually

checked visually) in Harwell 2130 scalers. Further details of the experimental system are given in (6).

EXPERIMENTAL MEASUREMENTS AND RESULTS

Measurements have been made on a 1100 kg 500 litre drum containing cemented simulated CAGR waste with the experimental system in both its active neutron and active gamma-ray interrogation modes. A system of holes and plugs in the drum allowed actinide samples and calibration sources to be placed at known positions within the drum. The neutron detection efficiencies for the various counters used were measured using a strong calibrated ^{252}Cf neutron source.

In the neutron mode, measurements were made on the drum with no fissile samples present, and also with a series of ^{235}U samples (in the form of foils 0. 05 mm thick to avoid self-shielding) at many different positions within the drum. Seventeen counters were used as follows: three fast neutron detectors configured as three 4-packs of 25 mm ^{3}He counters 300 mm long in the 180° detector channel one above the other (the middle detector centred in the drum mid-plane); two fast neutron detectors based on 50 mm ^{3}He counters 1 m long in the 90 and 225° channels; one bare 25 mm 10BF3 counter 120 mm long in the 135° channel in the mid-plane; one 13mm $^{10}BF_3$ counter 50 mm long to measure the thermal flux within the drum; and one 25 mm $^{10}BF_3$ counter 90 mm long in lead and cadmium shielding, outside the assembly, to monitor the epi-cadmium neutron flux directly from the target.

Measurements of responses were made for the following drum and fissile sample configurations: drum with no fissile samples; as a function of fissile sample mass at the center of the drum; for a fixed mass fissile sample (55 g ^{235}U), as a function of vertical position on the central axis, azimuthal angle at 240 and 300 mm radial positions in the mid-plane, and azimuthal angle at 240 and 300 mm radial positions in a plane 300 mm above the mid-plane. In addition, activation measurements of the absolute output of the neutron target were made using the ^{41}K (n,p) ^{41}Ar reaction, and subsequently counting the 1294 KeV gamma line. The ^{41}K (n,p) ^{41}Ar reaction was chosen so that there would be no spurious activation by gammas in the highly gamma-contaminated neutron radiation field.

In the gamma mode, measurements were made on the drum with no actinide samples present, with a set of 275 g natU cylinders 19 mm in diameter, and with 64 g of heavy water, at many different positions within the drum. Four counters were used as follows: two bare 50 mm $^{10}BF_3$ counters 400 mm long in the 180 and 225° detector channels; a bare 25 mm $^{10}BF_3$ counter 120 mm long in the 135° channel; one fast neutron detector based on a 50 mm diameter ^{3}He counter 1 m long in the 90° channel.

Measurements of responses were made for the following drum and actinide sample configurations: drum with no samples at 7 MeV; as a function of ^{nat}U mass at the center of the drum at 7 MeV; for the D_20 sample at the center of the drum at 5 and 7 MeV; as a function of azimuthal angle for one ^{nat}U cylinder and for the D_20 sample at a 240 mm radial position at 7 MeV; for the D_20 sample as a function of vertical position on the central axis at 7 MeV; for a drum with no samples, as a function of energy between 5 and 7 MeV; as a function of azimuthal angle for the D_20 sample at 240 mm at 5 MeV. The measurements with the D_20 sample were made because of the important subtraction of the deuterium photoproduction response from the response measured at 7 MeV due to both actinide and deterium components. Azimuthal responses were measured only for samples in the mid-plane.

In Fig. 2 are shown typical time spectra from counters in both the neutron and gamma modes. For operation in the neutron mode, the component in the time spectrum produced by introducing fissile material into the drum is obvious; the 135 μs i/e die-away time of this component agrees very well with the characteristic die-away time of the thermal flux within the drum, as measured by the 13 mm diameter $^{10}BF_3$ counter inserted into a 20 mm diameter hole running through the drum. For the gamma mode, time spectra from both thermal and fast neutron detectors are shown. The thermal detector time spectrum dies away relatively slowly (mostly because of the slow die-away of neutrons within the pure lead shielding around the counter), whereas the fast detector spectrum dies away with the ~30 μs characteristic decay time of the detector package.

In Fig. 3 is shown the response function for the system in its neutron interrogation mode as a function of ^{235}U mass at the center of the drum. The quantity plotted is the ratio of counts at later times in the time spectra (mostly due to the thermal neutron induced fissions) to counts at earlier times (mostly due to the interrogating neutrons themselves) corrected for the background in the absence of ^{235}U samples.

In Fig. 4 is shown the dependence of the response on azimuthal position of the fissile material. Two sets of measurements are shown (for the mid-plane and bottom 4-packs in the 180° detector channel). Also shown are values calculated previously (8).

In Fig. 5 is shown the response function for the system in its gamma-ray interrogation mode as a function of actinide mass at the center of the drum. The quantity plotted is the total count in the 225° detector normalized to the electron beam charge delivered to the bremsstrahlung radiator

and corrected for the background in the absence of actinide samples. In Fig. 6 is shown the dependence of the response on azimuthal angle for actinide material. Also shown are calculated predictions.

DISCUSSION

In its neutron mode, the response of the system as a function of fissile material mass is satisfactorily linear. The small deviations from absolute linearity can be explained on the basis of the differing configurations of the thin foil ^{235}U samples and the inherent small spatial variations in response over distances of ~100 mm corresponding to the extended sizes of the configurations. The (fast) neutron output from the neutron target deduced from the 41K activation measurements was 1×10^{9} neutrons/μC. The absolute response at the center of the drum is 0.2×10^{-10} counts at times greater than 200 μs per gram of ^{235}U per interrogating neutron. This is lower than the value of $\sim 1 \times 10^{-10}$ obtained by correcting previous predictions (4,5) for neglected neutron absorption effects in the matrix (2). Preliminary indications, from neutron detection efficiency, thermal neutron flux and target fast neutron ouput measurements, are that it is the thermal neutron flux in the drum which is lower than predicted. The measured and calculated azimuthal angle dependences are in satisfactory agreement. The decreases towards -180° and $+180^{\circ}$ in the response for the 4-pack out of the mid-plane can be explained by the greater sample-to-counter distances for samples in the mid-plane and counters out of the mid-plane, than for counters in the mid-plane when the samples are nearer the counters.

In its gamma mode, the response of the system as a function of actinide mass is also satisfactorily linear. Responses, as in (8), were calculated by folding together the spectrum of bremsstrahlung as a function of photon energy and angle from the radiator and the ^{238}U photofission and photoneutron cross-sections, and assuming exponential attenuation of the incoming bremsstrahlung photons and the outgoing photoproduced neutrons. The absolute measured and calculated responses for actinide material at the center of the drum are 2×10^{-4} and 4×10^{-4} counts per gram of ^{nat}U per microcoulomb of charge delivered to the bremmsstrahlung radiator respectively, for 7 MeV electrons. The discrepancy can be explained by the failure to take account of the soft, and therefore rapidly attenuated, photoneutron component of the photoproduced neutron energy spectrum in the neutron detection efficiency prescription assumed in the calculation. There is good agreement between the measured azimuthal angle distributions and the distribution calculated as described above.

The azimuthal angle distributions for the D_20 sample (not shown in the present paper) are similar in shape to the actinide distribution shown in Fig. 6. One of the reasons for making the D_20 measurements is to understand the differences in the azimuthal angle distributions at 5 and 7 MeV. If water (in the cement encapsulation) is distributed homogeneously throughout the drum, then differences in the shapes of the response functions at 5 and 7 MeV are relatively unimportant. However, if the water is not distributed homogeneously, then the differences in the responses have to be understandable and predictable.

CONCLUSIONS AND FUTURE WORK

The results of the present experimental work show that there are no particularly severe unexpected difficulties in operating the system as previously envisaged at the design stage. Apart from the overestimate of the absolute neutron interrogation response, which is currently being investigated, the results of the measurements give confidence that the calculations developed earlier within the present programme can be used to understand the behavior of the active neutron and gamma-ray interrogation systems. It is intended to repeat the present measurements on simulated cemented Magnox and PCM 500 litre waste drums in which the neutron and gamma-ray transport properties are quite different.

PRACTICAL ASPECTS OF AN ASSAY FACILITY

Practical application of the techniques mentioned in Section 1 involves consideration of:-

- the relationship(s) between radioactive material content and measured output signal(s)
- the usage pattern of the facility
- data processing and information presentation
- the engineering design of the facility

The first of these points is of course basic to the whole concept of assay. Although there are some further aspects to be resolved, a clear correlation has been established between actinide content and neutron count for both active neutron and active gamma-ray interrogation. It is expected that calibration will be required, possibly involving the use of standard calibration drums on a regular basis. It is obviously essential to have a good understanding of the factors which can upset the correlations, and this may require further research.

The pattern of use of a facility will depend on the number of waste drums and the proportion of these to be assayed. The latter has not so far been resolved. The possibility of 100 per cent assay cannot be excluded from consideration, if only because of public opinion. In the UK, this would necessitate a short cycle time (about 20 minutes) and virtually continuous operation.

For routine assay, operators need to be presented with reliable information about each individual drum, in an easily

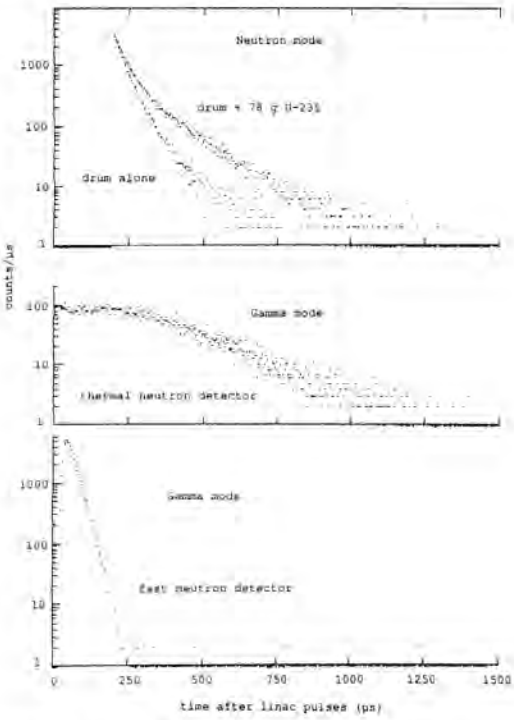

Fig. 2. Time dependences of signals from detectors in neutron and gamma modes. The digital signals from the counters are obtained from single channel analyzers on the outputs of the (pulse shaping) main amplifiers following the charge sensitive preamplifiers. Top: in neutron mode, from fast neutron detector, with (upper) and without (lower) 78 g ^{235}U at the center of the drum. Center and bottom: in gamma mode, from thermal neutron and fast neutron detectors respectively as described in the text.

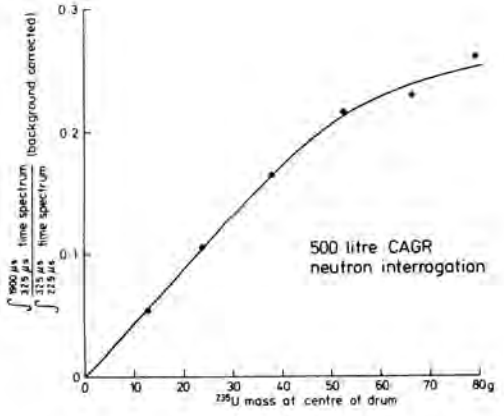

Fig. 3. Variation of the response of the system in neutron mode with fissile mass at the center of the drum. The quantity plotted is the ratio of the 325-1900 μs region of the time spectrum from the mid-plane 4-pack to the 225-325 μs component as described in the text. The line is a fit to the data points.

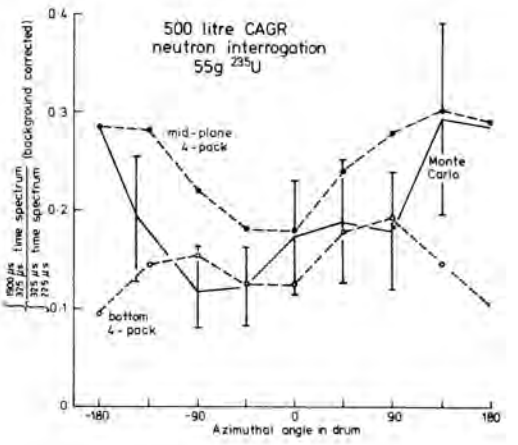

Fig. 4. Variation of the response of the system in neutron mode with azimuthal angle for fissile materials at 240 mm radius in the mid-plane for the mid-plane 4-pack in the 180° detector channel. The Monte Carlo calculations for the mid-plane line, shown with corresponding uncertaintiers, have been normalized to the measured mid-plane values (solid circles) at -180° (180°). Also shown in a measured distribution (open circles) for the bottom 4-pack. The lines simply serve to guide the eye.

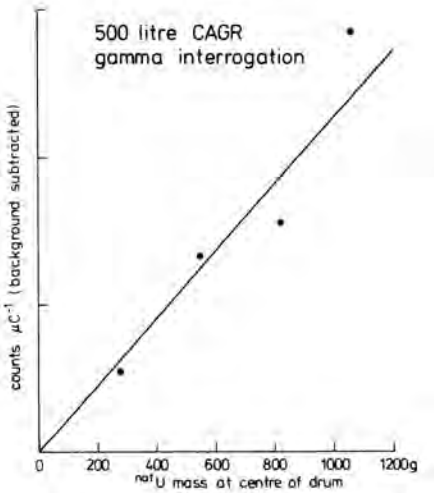

Fig. 5. Variation of the response of the system in gamma mode with actinide mass at the center of the drum. The quantity plotted is the total count from the 225° thermal neutron detector per microcoulomb of charge delivered to the radiator. The line is a fit through the data points.

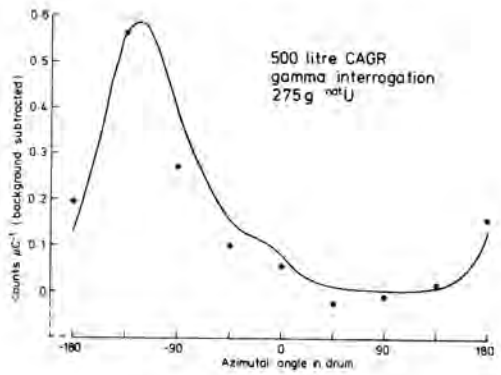

Fig. 6. Variation of the response of the system in gamma mode with azimuthal angle for actinide material at 240 mm radius in the mid-plane. The predicted distribution line, calculated as described in the text, has been normalized to the measured values (solid circles) at -135° (225°).

understandable form. The ideal is probably a graphical and digital screen display with video recording and hardcopy printout. Raw data eg neutron counts etc. need to be available as a secondary display or simply as hardcopy. It is desirable that the complex mathematical processes by which the raw data are converted into meaningful information are carried out by the data processing system, and not by the operator. Estimates of radioactive material quantities will have to be corrected for the effects of distribution within the package and any other factors which may affect the correlations. A time based 3D visual simulation of the package would be useful if it can be developed.

To engineer a facility, its functional requirements have to be defined. Many of the specific engineering problems have been considered elsewhere (6, 8).

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