EXPERIENCES OF WASTE ASSAY AND MANAGEMENT ISSUES ENCOUNTERED IN DECOMMISSIONING OLD RADIOACTIVE FACILITIES

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ABSTRACT

The history of nuclear programmes has now given rise to a legacy of radioactive contaminated facilities in the UK and elsewhere. In the UK these facilities were associated with the initial R&D programmes, both civil and military, and subsequent reprocessing programmes to reclaim ^{235}U and plutonium produced in fission reactors. The principle radioactive species produced in these systems are fission products, characterised by Cs-137 (Ba-137m), activation products, characterised by Co-60, and actinides, which are in general more difficult to assay as many of these do not have a characteristic gamma signature.

In addition, the UK was also the base for the Joint European Torus (JET), a pioneering fusion research facility. Several particle accelerator facilities were also designed and built, such as Van de Graaf machines and various cyclotron/synchrotron particle research systems. The radioactive waste types from these are activated materials from fast and slow neutron capture and more exotic radionuclides produced from particle-nucleus interactions. [1]

Of the various waste characterisation methods, gamma techniques are the simplest to use because they enable a significant volume of waste material to be interrogated, whereas alpha and beta measurements are generally of limited range and are best used in laboratory measurements. Furthermore, the detection limits for neutron measurement techniques are usually so poor that they are greater than most relevant waste sentencing criteria. Nuvia has developed a range of in-situ and ex-situ gamma based systems for monitoring wastes and these will be described in detail in the paper. These include dose rate measurements in high gamma fields (g.t. 100μSv/h), gross gamma systems for mapping and bulk waste sentencing, and gamma spectrometry systems for in-situ contamination and package (200 litre drum, 1-2 tonne bag) monitoring. These systems include the Nuvia GROUNDHOG GPS-based contaminated land mapping system, a concrete core profiling system, 2D and 3D room mapping, conveyor monitoring, excavator bucket monitoring and purpose designed gamma spectrometry systems for bulk waste monitoring.

Examples of completed projects will be described; these will cover the range of radioactive wastes produced and the appropriate measurement systems used.

1. INTRODUCTION

The paper is split into two main sections; the first on the gamma-ray based measurement techniques and the second giving details of several projects where these techniques were used to assay and characterise waste materials.

2. GAMMA-RAY MEASUREMENT TECHNIQUES

2.1 Radioactive decay properties

Radioactive decay can take place by a variety of different decay schemes, mainly gamma-ray, beta decay (often associated with gamma decay), alpha decay and spontaneous neutron decay. It is not the intention to discuss the various merits of these for assay purposes in great detail, but the main decay properties and the advantages/disadvantages of each of these techniques for quantifying radioactivity in wastes are considered in Table 1.

Table 1 Radioactive decay processes and assay capabilities

Decay process	Range	Detection capability	Detection range
Gamma-ray	~500 mm	Good	From background levels (low) to spent nuclear fuel levels (high)
Beta decay	~ 10 mm	Poor	Usually at low to medium activity levels
Alpha decay	~1 mm	Very poor	Low activity levels
Neutron	~500 mm	Poor	High activity levels

Gamma-ray measurement techniques have the advantage that they may not only be used to assay material in the laboratory, but they can also be used directly on a site to measure the activity of materials in buildings or even in the ground. The short range and measurement difficulties of the other three decay processes usually preclude their use directly on-site.

2.2 Gamma-ray measurement options

There are several options regarding gamma-ray measurements, which vary in the quality of information gathered, the activity range of operation, the complexity of the equipment, and the cost and suitability for on-site use. The various options are given in Table 2, with a detailed description on each of these options discussed below.

Table 2 Gamma-ray measurement techniques

Technique	Information quality	Activity range	Complexity	Cost	Suitability for on-site use
High resolution gamma-ray spectrometry	Excellent	Low	High	Most expensive	fair
Low resolution gamma-ray spectrometry	Very good	Low	High	Fairly expensive	fair
Window based measurements	Good	Low	Fairly high	Fairly expensive	fair
Gross gamma measurements	Good	Low to Medium	Fair	Fairly expensive	fair
Dose-rate measurements	Good	Medium to High	High	Fairly cheap	good

2.2.1 High resolution gamma-ray spectrometry

High resolution gamma-ray spectrometry systems comprise a hyperpure crystal of germanium, cooled to liquid nitrogen temperatures and connected to processing electronics. Gamma-rays de-excite in the crystal and yield electrical charge pulses, which are collected in an applied electrical field. This generates a signal, proportional to the incident gamma-ray energy, which can be processed and stored to generate a gamma-ray energy spectrum on a computer. An example of a high resolution spectrum for Cs-137 (Ba-137m; diamond symbol) and Co-60 (square symbols) is given in Figure 1.

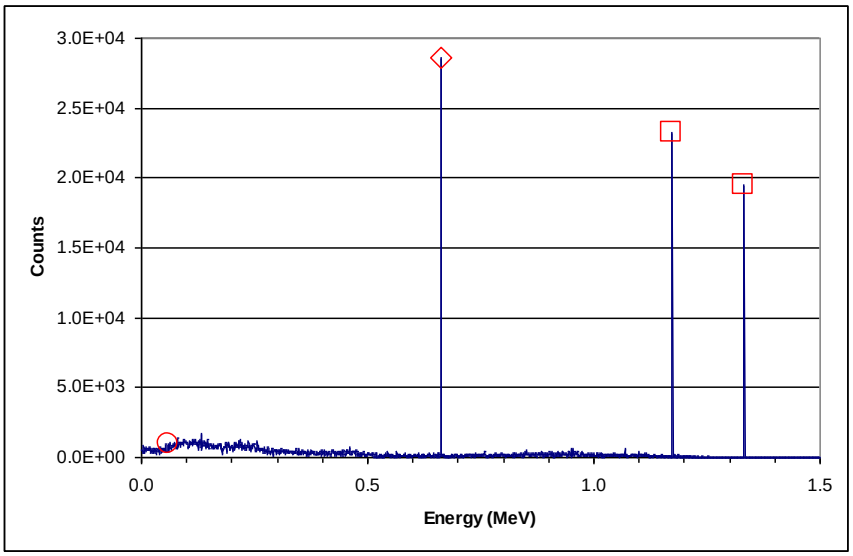

Figure 1 High resolution spectrum of Cs-137 and Co-60

2.2.2 Low resolution gamma-ray spectrometry

Low resolution gamma-ray spectrometry systems comprise a scintillation crystal connected to processing electronics. Gamma-rays de-excite in the crystal and yield a light pulse which falls onto a photocathode. Again this signal is converted to enable spectra to be obtained. A low resolution gamma-ray spectrum for Cs-137 is shown in the dotted trace in Figure 2. The resolution, or width of the peak, is as the name suggests, significantly worse for a low resolution detector than for a high resolution detector.

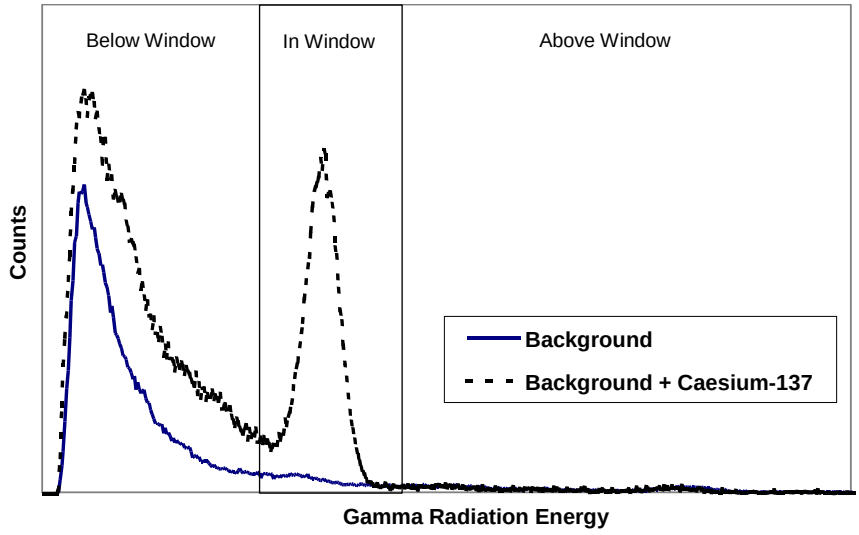

Figure 2 Low resolution spectrum of Cs-137

2.2.3 Window based measurements

This technique is most often used with low resolution systems. Instead of generating a whole spectrum, simpler electronics can be employed to reduce the measurement to a few 'regions of interest' or 'windows'. An example is given again in Figure 2, where three areas of interest are shown as Below Window, In Window and Above Window sections. Two spectra are given in the figure; the 'Background' represents the natural background spectrum from the environment, whilst the second spectrum was collected with the addition of a Cs-137 source. It can be seen that the Cs-137 only contributes to the lower two windows in this case, thus a comparison of the ratio of the accumulated counts in either of these windows, compared to the Above Window counts, can be used to indicate the presence of Cs-137. It would appear that there is little to be gained by using 'Windows' compared to accumulating and processing spectra, but this approach is beneficial when data processing is required in real time.

2.2.4 Gross gamma measurements

This technique is most often used with low resolution hardware and is really the simplest option, in that it uses all of the counts detected, with no spectral information whatsoever. Again Figure 2 can be used to explain the method in that all of the counts contained in one spectrum would be summed to give the total count-rate. The method relies on knowing the gamma-ray radioactive sources contributing to the gamma signal, but in most cases this is either known, or can be determined separately by spectrometry. The advantage of this approach is that it minimises the processing and display electronics to that of a simple ratemeter and thus is similar in use to a conventional Health Physics instrument.

2.2.5 Dose rate measurements

This approach is comparable with the gross gamma technique for low resolution scintillation crystals, but is used in higher radiation fields when spectrometer based pulse counting systems would saturate. In this case the detector types used are either ionisation chambers or Geiger-Muller (GM) tubes.

3. PROJECTS

Over a number of years, Nuvia has developed various applications of these different gamma measurement and processing techniques, and this has resulted in the design and construction of several pieces of assay/monitoring equipment to aid with decommissioning and land clearance operations. This section provides details for several projects, which have employed these techniques.

3.1 LIDO swimming pool reactor

The first example is of the LIDO swimming pool reactor at Harwell [1], which was used as a neutron source for early experimental studies of nuclear cross sections. The reactor core was exposed under water and the blue glow from Cerenkov radiation was an interesting feature that could be directly observed (Figure 3a) from above the reactor. Neutrons generated in the reactor core were channelled through the bioshield along 'beam tubes' for neutron scattering experiments and as a consequence the concrete walls in these regions became activated with gamma emitting isotopes such as Co-60, Eu-152 and Ba-133. An assessment of the activity of the activated concrete was undertaken using surface mapping and samples from cores taken from the concrete. Figure 3b shows the results from a gross gamma surface mapping, on a 10cm grid, of the inner wall of the bioshield near three of the beam tubes. Figure 3c shows the Co-60 gamma-ray activity profile into the wall of the bioshield, again on a 10 cm grid. These latter measurements were obtained by coring a section of the bioshield and then measuring the Co-60 activity profile of the core using a high resolution gamma-ray spectrometer. Separate assessments were made for the other gamma-ray emitting isotopes.

The reactor has now been fully decommissioned and demolished, and the site returned to a 'greenfield' status.

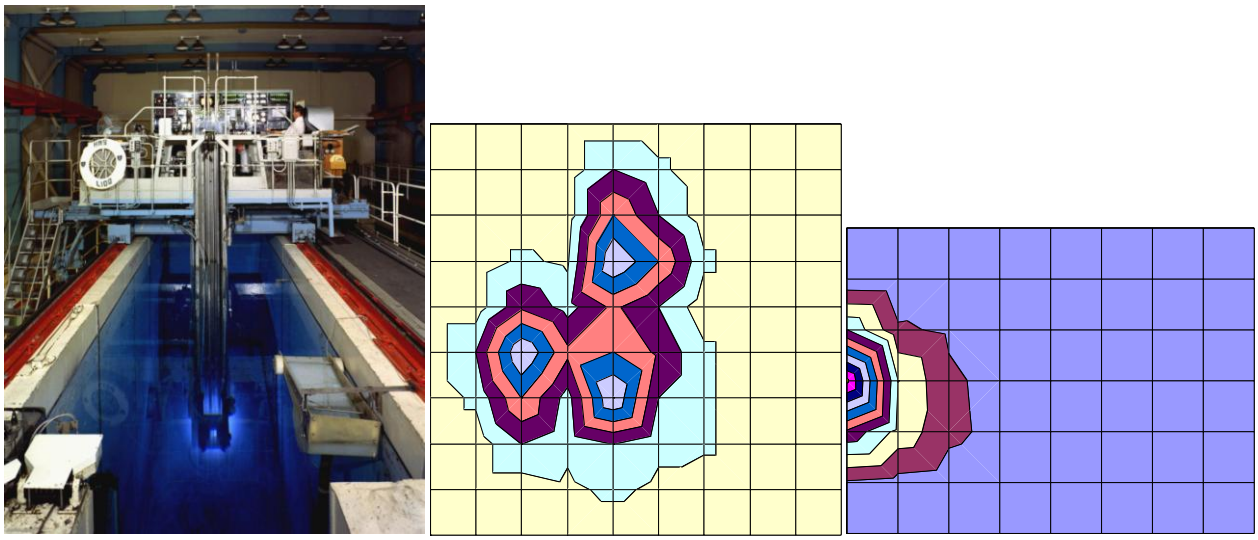

Figure 3 (a) Lido swimming pool reactor showing the Cerenkov radiation; (b) gross gamma surface mapping of the inner wall of the bioshield; and (c) Co-60 activity profile into the wall of the bioshield.

3.2 Van de Graaf accelerator and synchotron

The second example is of the radioactive wastes generated in a Van de Graaf accelerator and a synchrotron; both of these have now been completely decommissioned and returned to 'greenfield' sites. The wastes generated in these facilities were due to the presence of activation products, although there were some more exotic radionuclides generated by the various ion beams. These beams were used for many different processes; basic nuclear studies, I-123 production for nuclear medical purposes and thin layer activation (TLA) to assess engine wear in Formula 1 cars were just some examples. In general these reactions led to the production of neutrons which activated the local concrete bioshields. Information relating to the relative concentration of activation products in bioshields is given in Tables 3 and 4. Table 3 gives information on stable isotopes that can be activated by neutron capture, together with the capture cross section, the radioelement produced and its half-life. Table 4 gives the relative activity of these isotopes in a typical concrete bioshield after 35 years. Activity measurements in this application are based upon sampling and laboratory analysis, which is required in any case for beta analysis, to generate a radioisotope fingerprint of the waste material. More general on-site measurements were then conducted using high resolution gamma-ray spectrometry.

Table 3 Nuclear data on activation isotopes from elements found in concrete

Element (natural abundance)	Cross section (barns)	Radioactive element produced	Half life
Li-6 (7.5%)	941	H-3	12.3 y
C-13 (1%)	0.001	C-14	5700 y
Ca-40 (97%)	0.4	Ca-41	100300 y
Fe-54 (6%)	2.2	Fe-55	2.7 y
Co-59 (100%)	37	Co-60	5.3 y
Ni-62 (3.6%)	14	Ni-63	100 y
Eu-151 (48%)	9165	Eu-152	13.5 y
Eu-153 (52%)	312	Eu-154	8.6 y

Table 4 Activity of a bioshield, decay corrected to 35 years

Radionuclide	Activity (Bq/g)	% Activity
Co-60	4.4	0.27
Ba-133	0.2	0.01
Eu-152	168.0	10.30
Eu-154	7.1	0.43
H-3	1330.0	81.53
C-14	12.2	0.75
Ca-41	105.0	6.44
Fe-55	0.8	0.05
Ni-63	3.7	0.23
Totals	**1631.4**	**100.0**

3.3 Joint European Torus (JET)

The third example is of wastes generated at JET and again these are principally from activation products. Figure 4a shows the actual Torus and Figure 4b shows a high resolution gamma-ray spectrometry measurement of a component removed from the Torus after trials. By far the most predominant radionuclide found in this instance is Co-60.

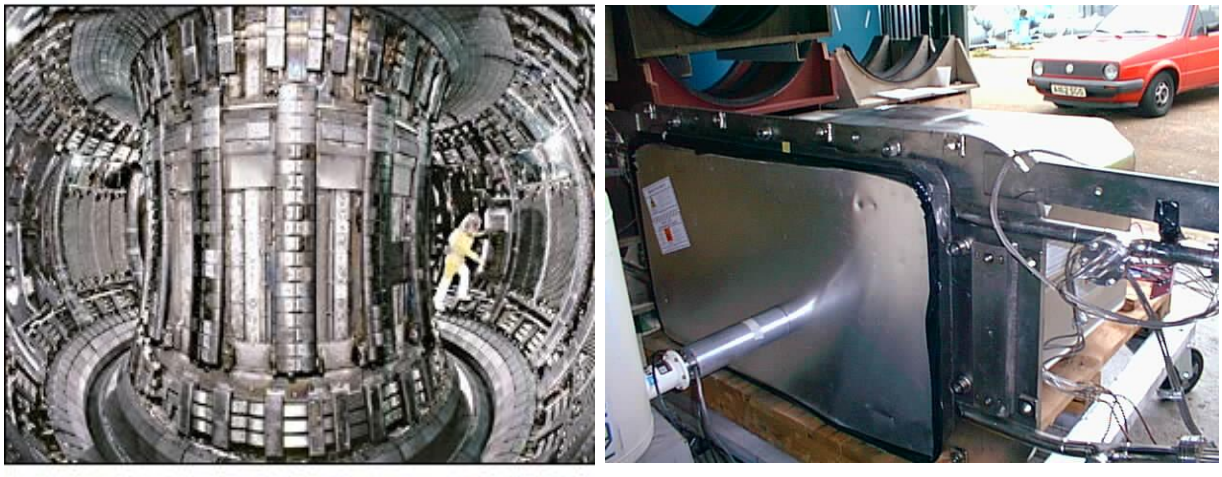

Figure 4(a) Inside the Torus and (b) Gamma spectrometry of a component removed from the Torus.

3.4 Recovery of spent fuel particles

The Dounreay nuclear facility in Scotland was built in the 1950s as part of the UK's investigations into fast reactor technology. At some point in time spent fuel particulate was accidentally discharged from the site with the result that small active particles are now found on the seabed and beaches in the vicinity of the site. Nuvia had already developed a GPS based contaminated ground surveying system called GROUNDHOG and this has been adapted to provide a particle detection capability for both the beaches and the seabed. The beach detection system comprised five large low resolution detectors mounted in a special housing on a purpose designed vehicle. This is shown in Figure 5a. The vehicle position was constantly monitored by GPS technology and all relevant data from all of the detectors and GPS system were recorded on data loggers. In this case real time measurement and processing was required so that particles could be located and recovered during the survey (during one tide), rather than after data post processing. At the time this system was initially introduced it was difficult to accumulate full spectra and then perform the spectral analysis in real time, therefore, a simpler processing approach was used. The dominant gamma contribution was from Cs-137 and the method adopted was the window approach described in Section 2.2.3 and the gross gamma approach described in Section 2.2.4. The window approach was used by taking the ratio of the In-Window (which would increase with the presence of Cs-137) and the Above-Window (which would not). This signal was compared against an alarm level to indicate the presence of a particle. In addition the total count-rate or gross gamma signal was also used. Although this may vary with surface matrix type, if one is monitoring a fairly consistent sand beach then this background variation would be small. In this context the measurement would compare directly with that of a standard Health Physics gamma contamination probe.

A seabed system was also developed in conjunction with Land and Marine Project Engineering and operates on a similar basis, again using five main sensors, but with three other sensors for detection of recovered material. A photograph of the ROV is shown in Figure 5b and the detector assembly is shown in the lower left of the picture.

Figure 5(a) Beach monitoring equipment, and
(b) ROV deployed on the seabed for particle recovery.

3.5 Decommissioning of a Post Irradiation Examination (PIE) Facility

It was a requirement of the nuclear industry to investigate the performance of fuel cladding and the fuel itself to ascertain the reliability of fuel element design. Such examination required specialised thick concrete walled 'hot cells', into which the spent fuel could be unloaded and then examined. This work required specialised facilities for such examination and one such PIE unit was located at the UKAEA Winfrith site in the UK. At the completion of the investigation phase this PIE building became redundant and a decision was made to decommission the facility. The cells in the building were severely contaminated, with dose rates of ~10Sv/h from the fuel examination processes, which had involved machining, cutting and polishing of the spent fuel. The first phase of the decommissioning process was to decontaminate these cells using remote handling technologies until the dose rate was reduced to levels of ~100μSv/h. At these levels manned entry became acceptable for short periods, however this speeded up the decommissioning process significantly as manned mapping technologies then enabled higher levels of contamination to be identified and removed preferentially. Finally, when all the gross contamination had been removed the walls were scabbled to strip off remaining surface contamination. The building was then demolished in sections, the concrete crushed and monitored for clearance purposes. Unfortunately, at this stage it was realised that there was a problem with the procedure adopted, in that small particles of fuel still appeared to be present in the crushed concrete. In the UK the sentencing level for LLW is ≥ 0.4Bq/g, which would correspond to only a 30μm fuel particle in one tonne of concrete. It was believed that as uranium is an alpha emitter, the electrostatic charge on particles of this size was sufficient for them to re-suspend in the air and then 'stick' back onto the walls after scabbling.

The problem was solved by spreading the crushed concrete from the waste into trenches and surveying them with the detectors used in the GROUNDHOG system in order to identify and remove the fuel particulate. This process is shown in Figure 6a and operates in the gross gamma mode. Two further monitoring systems, the **GEM** (**G**amma **E**xcavation **M**onitor) system, again

working on a gross gamma technology, and the **HiRAM** (**Hi**gh **R**esolution **A**ssay **M**onitor) system, using high resolution gamma technology, were also used as part of this process. These are shown in Figures 6b and 6c.

Figure 6(a) Trench monitoring using Groundhog;
(b) GEM system for measuring excavator bucket loads; and
(c) HiRAM for monitoring bagged materials.

3.6 Measurement of the inventory of nuclear material in a hot cell

There is a requirement for countries who are members of the IAEA, under the international Safeguards Nuclear Materials Accountancy Agreement, to provide information on their holdings of nuclear materials, such as uranium and plutonium. The aim of this study was to ascertain the amount of fuel that had been deposited from another PIE hot cell, again used for fuel examination, into an underground vault at the Aktau reactor in Kazakhstan 2. The approach taken was to penetrate through the thick walls of the vault and insert a dose rate monitor on the end of a manipulator arm together with a lighting system and a camera. The camera enabled a visual inspection of the vault to be made, and the dose rate system was manoeuvred within the cell to provide a three co-ordinate xyz mapping of the dose rate within the cell. The dose rate meter is shown in Figure 7a, a picture of the vault showing the drum located under the hot cell in is shown in Figure 7b, and a map of the 3D dose rate field in Figure 7c. From the measured dose rate field and knowledge of the fingerpint relating the gamma emitting radionuclides to the nuclear materials in the fuel, it was possible to calculate the amounts of plutonium and uranium in the vault.

Figure 7 (a) Dose rate meter inside the cell;
(b) Drum inside the vault; and
(c) 3D representation mapping the dose rate field.

4. SUMMARY

The purpose of this paper was to provide an insight into the history of the UK nuclear programme and describe some subsequent problems of decommissioning and remedial studies. This work required the development of specific waste assay and characterisation systems to assist with the on-site assessments of bulk waste materials. The basic assay measurement methods, using various gamma-ray measurement processes, have been described, together with a range of the practical problems encountered and the solutions that were developed.

Thanks are due to UKAEA, the UK Nuclear Decommissioning Authority, MAGNOX North and National Nuclear Centre, Republic of Khazakhstan, for permission to use technical data acquired on their sites.

5. REFERENCES

1. AERE Harwel1, Wikipedia

2. Anatoliy Ivanov, Igor Yakovlev, Sergey Pustobayev, Yevgeniy Tur, Alexander Klepikov, Andrew Herrick, David Wells , Ian Adsley and Collin Knight, Remote surveys of the BN-350 fast breeder reactor refuelling pathway (Aktau, Kazakhstan) – 11061, Paper to WM2011 Conference, February 27 - March 11, 2011, Phoenix, AZ