

## **In-Situ Characterization of Underwater Radioactive Sludge - 8031**

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### **ABSTRACT**

A fundamental requirement underpinning safe clean-up technologies for legacy spent nuclear fuel (SNF) ponds, pools and wet silos is the ability to characterize the radioactive waste form prior to retrieval. The corrosion products resulting from the long term underwater storage of spent nuclear fuel, reactor components and reprocessing debris present a major hazard to facility decontamination and decommissioning in terms of their radioactive content and physical / chemical reactivity. The ability to perform in-situ underwater non-destructive characterization of sludge and debris in a safe and cost-effective manner offers significant benefits over traditional destructive sampling methods.

Several techniques are available for underwater measurements including (i) Gross gamma counting, (ii) Low-, Medium- and High- Resolution Gamma Spectroscopy, (iii) Passive neutron counting and (iv) Active Neutron Interrogation. The optimum technique depends on (i) the radioactive inventory (ii) mechanical access restrictions for deployment of the detection equipment, interrogation sources etc. (iii) the integrity of plant records and (iv) the extent to which Acceptable Knowledge which may be used for “fingerprinting” the radioactive contents to a marker nuclide.

Prior deployments of underwater SNF characterization equipment around the world have been reviewed with respect to recent developments in gamma and neutron detection technologies, digital electronics advancements, data transfer techniques, remote operation capabilities and improved field ruggedization.

Modeling and experimental work has been performed to determine the capabilities, performance envelope and operational limitations of the future generation of non-destructive underwater sludge characterization techniques. Recommendations are given on the optimal design of systems and procedures to provide an acceptable level of confidence in the characterization of residual sludge content of legacy wet storage facilities such that retrieval and repackaging of SNF sludges may proceed safely and efficiently with support of the regulators and the public.

## INTRODUCTION

A commonly encountered problem in legacy ponds and silo is the accelerated corrosion of fuel elements and their casings underwater. As a consequence of this corrosion, large volumes of sludge and debris accumulates. This complicates the decommissioning and decontamination (D&D) efforts as the sludge is often difficult to remove. The fine particulate nature of the sludge causes major technical obstacles as cleanup activities often result in agitation which leads to reduced visibility and heightened airborne contamination. Removal of sludge is therefore one of the highest priority activities in waste cleanup activities for both historic facilities and operational plants.

Very often the long and diverse operational history of these facilities results in poor records of the sludge contents and uncertainties in its distribution. In addition the chemical and radiological activity of the sludge (key parameters required for building a defensible safety case for clean-up) are often based on very conservative estimates. More accurate knowledge of the radionuclide activity of sludge in various parts of the pond or silo is an important first step in the D&D process.

An overview is provided of applicability, advantages and disadvantages of the following methods suitable for underwater surveying of sludge:

- High Resolution Gamma Spectrometry (HRGS)
- Low Resolution Gamma Spectrometry (LRGS)
- Intermediate Resolution Gamma Spectrometry (IRGS)
- Dose Rate Measurements
- Neutron Measurements
- Monte Carlo N-Particle (MCNP) Modeling

Recommendations are provided regarding design of a ruggedized in-situ sludge survey technique based on state of the art technology that can be used as a generic method for underwater characterization of sludge and spent fuel debris

## GAMMA TECHNIQUES

Gamma-emitting waste can be measured either by counting the total number of gamma-rays produced by a sample (*gross gamma counting*) or by counting the number of gamma-rays produced as a function of gamma-ray energy (*spectrometry*).

The choice of technique depends on the application. As a rule of thumb, the less that is known (or can be assumed) about the material that is to be measured, the more complex and sophisticated the measurement system that is required. Depending upon the detector technology, the degree to which the energies of different gamma-rays can be resolved determines the application for which a particular technique can be used. In some applications the techniques may be used in combination (e.g. with an instrument using one technique being used to pre-sort material for measurement by an instrument that uses a different technique). The following sections describe the different instrument types that are in common use.

## **Gross Gamma Counting**

Gross gamma methods include use of plastic scintillators, Geiger-Muller tubes and ionization chambers. With these detectors, useful spectroscopic information is not provided. The main advantages of these methods are low cost, high efficiency, simple electronics, low maintenance and room temperature operation. However, the sensitivity of the technique is limited by its lack of energy resolution such that different isotopes cannot be resolved and events cannot be separated from the background.

The lack of radionuclide specificity means that if the sample differs significantly from what is expected, the wrong radionuclide inventory will be produced and furthermore any unexpected radionuclides that are present will not be reported.

Gross gamma surveys have been carried out on a number of occasions at spent fuel processing facilities which have provided important information for plant operators [1]. More complex analyses can be carried out using computer codes and / or MCNP modeling to interpret measured dose rates in terms of activity. Examples of application of this technology are:

- Determination of activity held up in evaporators at highly active process plants,
- Channel measurements in Spent Fuel Canisters at reprocessing plants,
- Dose rate logging to provide information on reprocessing efficiency for special fuels,
- Contamination mapping to locate the source of high dose rate levels in process caves.

## **Low Resolution Gamma Spectrometry (LRGS)**

The gamma-rays produced by different radionuclides have characteristic energies. The measurement of the energy of each gamma-ray as it is counted by the detection system allows the activity of different radionuclides to be determined separately. The interaction of gamma-rays with the surrounding material will modify the energy spectrum of the gamma-rays that reach the detector and this must be taken into account in calibration.

A common example of a LRGS system is a thallium-activated sodium iodide, NaI(Tl), scintillator crystal coupled to a photomultiplier in order to convert the light produced by each incident gamma-ray into an electronic pulse whose amplitude depends upon the energy deposited in the crystal. Sodium iodide detectors typically have an energy resolution (full width at half maximum, FWHM) of 6-7% at the Cs-137 gamma-ray energy of 662 keV, and are suitable for use when the gamma-spectrum is relatively simple, e.g. by virtue of it containing one or a few well-spaced photo-peaks or by its having a broad energy region dominated by one isotope.

NaI detectors are cheaper and require less maintenance than their high-resolution counterparts and they do not require special operating conditions (e.g. liquid nitrogen or electro-mechanical cooling). LRGS detectors can also generally be made to be larger and as such they can provide a highly cost-effective and high efficiency counting system with respect to HRGS.

The choice of detector size and scintillator material is a function of the measurement application; larger detectors are used when a high counting efficiency is required and/or when the gamma-rays of interest are of high energy and unlikely to deposit all of their energy in a

smaller crystal.

Cesium iodide and bismuth germanate are alternative materials that, because of the higher atomic number of their constituent materials, offer a better stopping power than sodium iodide for a given crystal size. These materials have the drawback of poorer light transmission and other factors, such as their robustness to mechanical damage and sensitivity to temperature may also affect the choice of materials used.

Bismuth germanate is particularly useful for gamma-ray measurements in high ambient neutron backgrounds as the material is relatively insensitive to neutron capture, compared with sodium iodide.

The output from scintillator detectors, especially those using photomultiplier (PM) tubes, is often temperature dependent. Consequently, spectrum stabilization is usually employed such that the detection system gain (usually by controlling the high voltage to the PM tube) compensates for changes in the location of a known photo-peak.

Improvements can be made to LRGS measurements using spectral de-convolution techniques such as that developed by Southampton University [1] which provides more detailed information from complex NaI spectra. This technique improves the energy resolution and peak to Compton ratio, enabling more peaks to be resolved and in some cases allowing previously unseen photo-peaks to emerge from the gamma continuum.

However, the chief drawback is that unexpected radionuclides are not always discernable and may be missed or misinterpreted. Also the scintillator material may be fragile and as a result is easily damaged by mis-handling or when exposed to severe temperature changes.

A number of portable NaI detectors and handheld systems have now been developed by a variety of manufacturers which can be used for an initial, quick, qualitative assessment of simple combinations of commonly encountered radionuclides such as Cs-137 and Co-60.

### **High Resolution Gamma Spectrometry (HRGS)**

As is the case for LRGS, high-resolution gamma spectrometry based systems work by measuring the energies of the gamma-rays reaching the detector. The techniques employed for radionuclide identification and quantification, system calibration and correction for matrix attenuation are broadly similar to those used in LRGS systems. The improved energy resolutions brings two main benefits:

- it allows more complex mixtures of radionuclides to be measured
- it improves the signal to noise ratio of the counts in the photo-peak relative to the ambient Compton continuum.

The most common form of detector technology used for HRGS is based on crystals of the high purity germanium (HPGe) semiconductor material. These detectors typically have energy

resolution (FWHM) of 0.1 – 0.2 % at 662 keV. They are essential when the gamma-ray spectrum to be measured is complex, containing many gamma-ray lines from several radionuclides.

All HPGe detectors have to be cooled to a temperature below 100 K in order for them to operate. (Intrinsic germanium has a small semiconductor *band gap* which means that at higher temperatures electrons are promoted into the conduction band and the material stops behaving as a semiconductor.)

Traditionally HPGe detectors are cooled using liquid nitrogen. Although various Dewar and cryostat types and configurations are available that give liquid nitrogen hold up times from 1 hour up to 3 weeks, the requirement for regular refilling means that such detectors have to be used in applications where they are in accessible locations and close to a reliable supply of liquid nitrogen.

The use of electrical refrigeration as a means of cooling for HPGe is an obvious attraction for applications where regular liquid nitrogen filling is problematic. In recent years the technology has matured sufficiently to provide a reliable and cost-effective alternative to liquid nitrogen.

HRGS allows measurement of complex mixtures of gamma-emitters and may be used to detect unexpected radionuclides or to interpret unknown radionuclide mixtures. Energy dependent matrix attenuation correction can be applied to the measurements.

HRGS systems are easily the most expensive gamma counting platform. The electronics tend to be complex and thus may be more susceptible to problems such as electrical interference and component malfunction.

Another drawback is the long warmup and cool-down times required for the detectors, with up to 48 hours required to perform a full thermal cycle on a detector that loses coolant. Both the hardware and the data interpretation are considerably more complex for HRGS systems further increasing operational burden and cost of the measurement.

## **Intermediate Resolution Gamma Spectrometry**

Many recent advances in gamma detection technology have occurred in the area of Intermediate Resolution Gamma Spectrometry i.e. with resolution between that of HPGe and NaI. The holy grail of such research is to produce a detector material with near-HPGe resolution that can be operated at room temperature and manufactured in large physical dimensions at a price level near that of NaI.

Cadmium Zinc Telluride (CdZnTe) is one of the most widely used Intermediate Resolution Gamma Spectrometry detectors and has undergone significant development effort in recent years. The main benefits of CdZnTe are:

- Good detection efficiency for a given detector size
- A high band gap energy enabling it to at (or close to) room temperature,
- Improved temperature stability over NaI(Tl),

- Relatively low cost,

The disadvantage of the high band gap is that the energy needed to create an ion pair is high resulting in a low number of ion pairs. The resulting resolution, although worse than HPGe, is several times better than NaI(Tl) detectors, conferring sufficient advantage to make it attractive for the measurement of reasonably complex spectra that NaI(Tl) detectors would be unable to resolve effectively.

However, the present technology precludes growth of large CdZnTe crystals with acceptable charge transport characteristics. Coplanar grid technology yields CdZnTe detectors of typically  $1 \text{ cm}^3$  with good peak shape and resolution at a wide range of energies. These detectors have good pulse-height resolution at high energy, typically better than 2-3% FWHM for a gamma-ray energy of 662 keV.

The small size of the detectors limits the measurements to applications where there is a lot of gamma activity, so that they are ideal for high activity environments such as spent fuel debris. However, one major drawback is the incomplete charge collection effect which results in low energy tailing that can complicate the spectral analysis.

This problem is addressed by use of larger crystals, surface contouring, passivation techniques, use of Peltier cooling at the detector head to reduce the crystal temperature by 30K below ambient and improvements in electrode designs.

Recent advances in scintillator material have resulted in the development of cerium activated lanthanum bromide ( $\text{LaBr}_3$ ) detectors. These detectors offer room temperature intermediate energy resolution, fast emission and good temperature and linearity with fast decay times. At 662 keV the FWHM is around 3-4% [2].

Although this scintillator is more brittle than other popular materials,  $\text{LaBr}_3$  has the advantage of having a nearly flat photo-emission over a wide temperature range. For a more rugged detector,  $\text{LaCl}_3$  is a good alternative although the disadvantage is its reduced detection efficiency and slightly poorer resolution.

Unlike CdZnTe, lanthanum bromide can be produced in large volume crystals including the standard 3" x 3" configuration. This gives a significant performance advantage for measurement of high energy photons without the peak shape problem that CdZnTe suffers.

The only major drawback with lanthanum halides is these materials are currently relatively expensive to produce with crystal prices approaching that of HPGe. As a result this technology is usually only favoured over NaI where there is a specific need for the improved level of resolution and where operational difficulties preclude deployment of HPGe with its associated cooling requirements.

## NEUTRON METHODS

Neutron emissions tend to be more penetrating than gamma rays. However under water their range is limited to a few meters by the moderating and absorbing properties of intervening hydrogen nuclei.

Several methods have been developed for assay of fissile materials and neutron emitting isotopes using active and passive neutron based methods:

- Active methods are not usually favoured for underwater in-situ measurements due to the complexity of presenting an interrogation source to the target materials and the problem of thermal neutron absorption in the water.
- Passive methods are widely deployed in verification of spent fuel. Total Neutron Counting (TNC) is often used taking advantage of the strong specific neutron emission from Cm-244.

Fission chambers have been extensively used for neutron measurements in areas of high gamma dose rate, such as in the presence of spent fuel assemblies or fission products in reprocessing plants. Recent advances [4] in He-3 tube design for high gamma environments also makes these detectors (which offer low cost and high efficiency) an attractive option.

Passive neutron methods however are generally less favourable for in-situ sludge measurements than gamma methods as the concentration of the primary neutron emitter Cm-244 is a strong function of fuel burnup. As a result it is difficult to correlate the neutron measurement to the sludge radionuclide activity at facilities where a wide range of materials were processed with variable burnup. The mixing of the corroded fuel in the resultant sludge compounds this problem.

## MCNP MODELING

MCNP modeling is an extremely useful tool for estimating radionuclide activities from a set of raw measurement data. This method takes into account the measurement situation and models the effects of geometry, collimation, attenuation, scattering and detector type. MCNP techniques can be applied in the interpretation of both gamma and neutron data in spent fuel storage facilities.

## SLUDGE SURVEY METHODS

Initial surveying of underwater sludge can be performed with NaI focusing on mapping out the relative distributions of fission products (by measuring Cs-137) and activation products from (Co-60). The simplicity and robustness of NaI makes this the ideal detector for this job.

A probe comprising an uncollimated 3" x 3" NaI detector can be used to survey the location and activity distribution of sludge, spent fuel and other reactor hardware in the pond or silo. The probe requires a multichannel analyzer housed within the assembly and control from a laptop compute to enable acquisition and storage of spectra.

Measuring the gamma activity of Co-60 and Cs-137 enables the activities of other nuclides can be estimated by the 'fingerprint' or 'Acceptable Knowledge' method. Known isotopic fractions are determined with respect to Co-60 (for activation products) or Cs-137 (for fission products and actinides). Fissile content for criticality control is achieved by use of conservative scaling factors to the Cs-137 activity.

The underwater NaI probe may be positioned at a grid of pre-determined survey points underwater. The acquired spectra can then be analyzed with a mathematical model to determine the activity inventory and approximate spatial distribution of key radionuclides. Results can be combined with radiochemistry analysis where available to further improve isotope specific data on the sludge inventory of the structure under study.

These measurements may than be augmented with more detailed assessments on individual containers or debris at specific locations in the pond. An underwater gamma probe using an appropriate detector (either Geiger Muller, HPGe, NaI, LaBr<sub>3</sub> or CdZnTe) will be housed in a shield block/collimator assembly. A key part of the sub-surface assay system design is the collimator. This device restricts the field of view of the detector to a specific assay region to ensure that background activity (especially of concern in highly contaminated legacy facilities) is acceptably low. The collimator together with the detector's shield housing also plays an important role in ensuring that the detector does not 'saturate' in the presence of high count rates

Where necessary, additional filters or attenuators may be placed in front of the detector to further reduce the input count rate to a level that is within the specification of the detector electronics. The shield block should be designed to enable the detector to be presented to the container or zone of interest at known, fixed and reproducible geometries.

The sensitivity to gamma rays that originate outside of the target zone will be minimized by:

- careful design of the shield block / collimator assembly,
- intervening water; which can provide very effective shielding
- close proximity of the detector to the target; which significantly enhances efficiency to the target tank due to the inverse square law.
- use of multiple measurement locations to enable "cross-talk" effects to be corrected for during off-line analysis.



A suitable pole or rig can be used to facilitate deployment from a fixed operator platform. The gamma probe would be wired via an integral umbilical cable to portable electronics and computer station situated on or near to the operator's platform.

The probe needs to include a jig/standoff to ensure that the operator can easily present the detector at a known and reproducible distance / location to the target regions. High count rates from spent fuel will ensure that each survey would take no more than 5 minutes per target.

A live visual output is essential at the operator platform for the operator to verify data acquisition and determine the time required for each measurement. For each target a report can be generated off-line showing the radionuclide inventory. A graphical activity profile showing the location of the activity at various target zones can also be produced.

In addition to providing an estimate of the activity present in each target zone the proposed technique enables a dose profile to be generated. The survey can be easily repeated during D&D to determine the effectiveness of the subsequent clean-out process. This would enable operators to target areas where significant quantities of residual sludge remain in the pond.

A post clean-out gamma survey of all tanks could be used to confirm that there is no significant sludge hold-up and the dose rates and quantities of fissile materials are sufficiently low for subsequent safe removal and disposal.

This method offers several advantages over sampling.

- The total activity determination is based on a survey of the entire contents rather than from a small number of samples assumed to be representative of the contents as a whole,
- There is no requirement to remove, handle, transport, and dispose of potentially highly active samples,
- Careful design of in-pond monitor equipment and survey methodology can result in rapid surveys potentially minimizing operator time and dose uptake,
- The approach reduces the need for visual / boroscope inspection of in-pond hardware and containers to obtain verification that sludge has been removed,
- The generic approach is widely applicable across different facilities,
- Use of relatively inexpensive off-the-shelf equipment allows for ease of maintenance and component replacement.

## **SUMMARY**

A variety of underwater radiometric characterization methods have been identified that provide plant operators the ability to determine the fissile content and radionuclide composition of diverse sludge compositions.

Materials with a wide range of gamma activity can be characterized in-situ in legacy ponds and silos without the need to procure complex neutron based detection equipment in downstream processing facilities.

These methods also offers technical advantages over the neutron drum assay approach in that matrix related “dead zones” (due to the neutron moderating / absorbing effects of hydrogen present in sludge) are avoided thus eliminating reliance on uniform source distribution assumption that often underpin neutron based drum assay.

In addition to assay of fully corroded sludge, the method allows measurement on underwater pieces of spent fuel ranging in size from small particles through to whole fuel elements.

An optimal system design has been identified that, together with appropriate operational procedures, has been specified to provide an acceptable level of confidence in the characterization of residual sludge content of legacy wet storage facilities. This technology can allow D&D teams to safely and efficiently retrieve and repackage spent nuclear fuel sludges and other debris waste.

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