THE USE OF *IN SITU* GAMMA SPECTROSCOPY TO SAVE TIME, DOSE, AND MONEY IN OPERATING NUCLEAR FACILITIES

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ABSTRACT

In situ gamma spectroscopy has been successfully used for decommissioning surveys and environmental remediation surveys for many years. With the recent advancements in technology of very portable and reliable Ge detectors, high quality digital MCAs, and mathematical efficiency calibration algorithms that are fast and accurate, this technology is now available to operating nuclear facilities.

In situ gamma spectroscopy offers advantages over the traditional method of extracting a representative sample, transporting it to a laboratory, and then preparing the sample for counting.

Getting the sample is the major problem. Some samples are physically difficult to obtain (subsurface soil, concrete, activated materials, material inside pipes or tanks, etc.). Some samples are physically dangerous to take (very radioactive items like resin beads, concrete cores or steel coupons, pressurized gases, hot water, corrosive or toxic items). Since *in situ* spectroscopy is a non-contact process, and since the sample doesn't need to be physically extracted, these problems are greatly minimized. And, for high activity samples, the measurement time will be very short, much less than the sample extraction time, therefore the dose should be lower.

Some contamination situations are difficult because it is hard to find a truly representative sample for lab analysis. Because *in situ* spectroscopy measures the entire item, or a large portion of it, small local non-homogeneities are less of a problem. Since transporting the sample is not needed, and since preparing the sample in the lab is not needed, the costs, doses, risks of injury, and time associated with these tasks are eliminated.

In situ spectroscopy can give near-instantaneous results, and therefore allow prompt decisions to be made while the equipment is in the field. Frequently, new or unusual tasks have too much personnel protection required based upon a lack of knowledge of what might be encountered. This may cause the work to take longer, and increase the dose, rather than reduce it. The availability of nuclide-specific quantitative activity, rather than just gross count or dose-rate information can allow better decisions to be made by the plant HP to define the optimum amount of personnel protection for the job.

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INTRODUCTION

When planning the radiological protection portion of a project, the HP typically relies on various dose-rate measurements from the area, and his experience about the plant conditions and past similar operations. It is not very common to have nuclide-specific data available for job planning, other than from historical knowledge or expectations. But, in most cases, reliable knowledge of exactly what radionuclides are present, where they are located, and the activity or concentration will allow the job to be planned better. This better knowledge should lead to a safer operation, lower dose, lower risk of things going wrong, lower cost, and a quicker finish.

Historically, if the HP needed nuclide-specific information, samples must be extracted, packaged, transported to the laboratory, prepared for analysis, and counted. Even for "rush" samples with an on-site laboratory, this process takes hours or days, especially if the sample is "different" than normal. The sample extraction process is often dangerous, and difficult. Finding a small sample that is easy to extract and that is representative of the entire object is normally very difficult. As a result, few nuclide-specific analyses are performed for each new job, relying instead on dose rate measurements, and a history of previous jobs performed that were similar. Today, with the advancement in technology, practical, portable, quantitative, Ge gamma spectroscopic tools are available to the HP for even better job planning.

IN SITU GAMMA SPECTROSCOPY BACKGROUND

In situ gamma spectroscopy isn't very new, having been used extensively for fallout monitoring staring in the 60's. But, it was rather difficult then, with large MCAs, and low resolution NaI detectors. Today, the availability of Ge detectors with high resolution and low background makes the analysis of the spectrum much easier. High purity Ge, instead of Ge(Li) detectors now can be stored at room temperature, although they must be operated at low temperature. Reliable cyrostats can hold two or five days of liquid nitrogen (LN), and can be aimed in all directions without spilling. Small book-sized powerful Multichannel Analyzers (MCA) can run all day from a single battery, and have full Digital Signal Processing (DSP) for high count-rate and good temperature stability. Equally small and powerful PCs control the MCA, and run the spectral analysis software. Today, instead of taking the sample to the gamma spectroscopy counting lab, the counting lab can be carried by one person to the sample. For use in high background areas, or where there are multiple sources to be measured, shielding or collimation might be required. This is also available, as shown in Figure 1. This shield set has both 25 mm and 50 mm lead shielding in the back, at the sides, and with various aperture openings in the front of the detector. For close quarters, as is common in Nuclear Power Plants (NPPs), the shield assembly is easily broken down into parts.



Figure 1 The ISOCS shield and Ge detector

The final element to complete the package is the availability of mathematical efficiency calibration software, such as the Canberra ISOCS (In Situ Object Calibration Software). With this software, the user can generate accurate (5%) efficiency calibrations with no calibration sources. All that is needed is a few dimensions describing the source, any absorbers, material composition, and the location of the detector. The results are available in a few minutes. This allows the user to generate accurate results in the field, immediately after the acquisition. The software is very flexible, allowing calibrations at any energy between 45 and 7000 keV, at any distance from contact with the detector out to 500 meters, and in any location surrounding the detector. A wide variety of sample shapes may be calibrated. Figure 2 is intended to convey the versatility of the ISOCS (In Situ Object Counting System) device.



Figure 2 The ISOCS System can be used to assay many different things

HOW IN SITU GAMMA SPECTROSCOPY CAN HELP IN NPPS

NPPs are good places for gamma spectroscopy. Most nuclides of concern are either good gamma emitters, or correlated with other nuclides that are good gamma emitters. US NPPs all take a wide variety of sample for 10CFR61 compliance, and therefore have a rich database of correlation factors between nuclides for various waste streams. These waste streams are also typical of conditions encountered in routine or emergency plant operations. Remarkably, there seems to be little difference in nuclide ratios from plant to plant, so if plant-specific data isn't available, this generic data can be used to convert from easily measured nuclides to total inventory. The proper application of *in situ* spectroscopy adds more information to the database that can be used by the HP to properly plan and assess a new job. Now, in addition to dose rate and count rate information, the HP has nuclide information and the activity of each nuclide at that particular job site, not just historical information from previous jobs. This allows the HP to optimize the amount of protection for each job. Too little protection is obviously bad, but too much protection can also be bad, as it can also increase the worker's dose. Wearing respirators where there is little chance of internal deposition but where there is external exposure, will increase the stay-time and therefore the dose. Face masks and eye shields reduce the vision and work efficiency; unless there is a potential for exposure to the lens of the eye, then these may also increase total body dose. Protective clothing, especially when applied in multiple layers, also reduces work efficiency and adds extra stress to the worker which may cause more harm than the amount of skin contamination it prevents. And, these "safety" devices do increase the risk of falls, heat stress, or other occupational injuries in the industrial conditions encountered in NPP maintenance operations. So, unless they provide a commensurate reduction in radiation dose and risk, they are not ALARA. Isotopic composition knowledge of the proposed operation can help better plan the optimal protection program.

Are respirators really necessary? If the *in situ* measurement of the workplace indicates the presence of certain nuclides like Cs-134, Cs-137, Zr/Nb-95, Ru/ Rh-106, then perhaps they are, as these indicate, failed fuel with the possible presence of Ce/Pr-144, fuel "fleas" and fragments, and other alpha emitters. But, if only Co-58, Co-60, Mn-54, Zn-65, and other common activation products are present, then external exposure is very likely to far exceed the internal exposure and respirators may not be warranted.

Is a high beta dose rate field expected, which may require heavy gloves or eye protection? If only Co-58, Mn-54, Zn-65, and Cr-51 are present, then not likely, as these emit few betas. Co-60, Fe-59, and Sb-125 only emit low energy betas which are easily shielded. But if Cs-137, Ce/Pr-144, I-131 are dominant, then one should be more protective as they have many high energy betas. Does the dose rate as measured outside a pipe or valve or tank tell much about the conditions inside the object? Only if you know the nuclides causing the dose rate. Low energy nuclides like Cr-51, Sb-125, Cs/Pr-144, Am-241, are easily shielded, and there could be a lot more inside. But, if only high energy photons like Co-60, Co-58, Mn-54, Zn-64 are present, then the internal contents are likely to be pretty well estimated.

How much temporary external shielding is necessary? This is frequently added to reduce the dose rate but since the installation process causes dose, adding too much is not ALARA. If the nuclides and activities causing the dose rate are known, then the optimum thickness of shielding can be planned and added.

Many times, when pipes, tanks, or valves are opened, they release radioactive gasses (e.g. Xe-133/135). While not usually harmful, this release may set off alarms, and cause much additional paperwork and a reduction in confidence in the safety operation. *In situ* gamma spectroscopy would be helpful in determining the presence of these gases in advance of the procedure so that the necessary actions may be taken.

TYPICAL APPLICATIONS

In the first three years since the introduction of the ISOCS system, the commercial acceptance by users has been quite good. There are about 130 units in use now world-wide, with about 20 of them in NPPs, about half in the US. The following examples of the use of *in situ* gamma spectroscopy are from a polling of those users, and Canberra's experiences as an *in situ* measurement service provider.

At a Midwest NPP, several drums of waste were placed in a shielded shipping container many years ago, without adequate documentation. Even though the container was 5 cm of lead, it was an easy task to identify and quantify the contents as Cs-137 (1 GBq) and Co-60 (7 GBq), without opening the container. At the same facility, a spent fuel storage pool had been drained, and the amount of contamination on the wall was needed. Unfortunately, the waste container was near to the pool, and could influence the readings. But, the use of shielding and collimation around the detector minimized this problem. The wall contamination was found to be Co-60 (\sim 30,000 dpm/100 cm²) and Cs-137 (\sim 2,000 dpm/100 cm²). This was done without the expense of scaffolding or special supports or even entry into the pool area. Figure 3 shows the detector aimed at the pool walls, with the shipping container in the background.



Figure 3. ISOCS aimed at spent fuel pool walls

A NPP in Europe had some spent ion exchange resin for waste disposal. The disposal regulations define a maximum activity of each nuclide per container. Since the disposal costs are per container and quite expensive, it is desired to place the maximum amount of activity in each container as legally permissible. Because the resin was from various operations the Co-60 and Cs-137 (the dominant nuclides) fractions varied significantly, so that traditional dose-rate to activity conversion factors can only be used with much conservatism. Taking samples of a few mg for laboratory analysis is both costly and causes worker dose. ISOCS was used to count each 5-10 kg batch of resin before it was added to the drum. A running total of activity for each nuclide was kept, and the operation was stopped when the next batch would have exceeded the drum's legal capacity. This process saved the company about \$2M in waste disposal costs over the conventional method of dose rate to activity conversions.

Every NPP uses ion exchange resins for water purification. These must all be classified for disposal as radioactive waste. The typical process involves extracting a very small sample (few beads or flakes) to limit the amount of radioactivity, and then counting in the laboratory. This takes time and incurs dose, and leaves open the question of representativeness of the small sample to the full batch. A much better way would be to use *in situ* spectrometry to assay a large fraction or the entire container. This has lower dose, eliminates the sampling error, and gives near-instantaneous results.

Floor, wall, and ceiling contamination assessments are commonly done in potentially contaminated areas. For floors and accessible flat wall surfaces, large surface area beta count rate probes are more practical as they are lower in cost and easier to use. But, what is the correct calibration factor? The conventional method of using a thin beta source that generally is not the nuclide being measured is not correct. The source on the floor or wall is most likely not on the surface, but has penetrated in the pores of the concrete and/or covered by paint or grime. Since gammas aren't affected as much by this, a good application of ISOCS would be to determine the concentration in a few areas, and then establish site-specific correlation factors for the beta probes. For walls that are high, or for ceilings, or any surface with complex features, then the cost of erecting/removing scaffolding and the extra time to survey will make the ISOCS method much more attractive, as it can be done all from the floor area. Collimation can be used to define the field of view and to remove the influence of adjacent hot sources.

Pipe measurements are quite common in NPPs to track changes in contamination as a function of water chemistry. Taking samples here is very difficult, so *in situ* measurement programs are normally done. Historically these are not quantitative, and sometimes not even nuclide-specific, just trending measurements. They must be repeated with the same detector at the same location each time. But with ISOCS, these are nuclide specific and quantitative, and the same pipe area can be measured from many different angles, if the plant configuration changes. Nuclide-specific activities are most important today to define the effect of the many new water chemistries that are being refined to reduce occupational dose. The ISOCS shield is modular allowing to be broken down and carried into confined spaces. The collimation allows only the pipe of interest to be measured, as shown in Figure 4. The collimation and spectroscopy also has allowed the main steam line to be measured during power operations at a Southern US NPP. Normally, these are done during an outage as access is not allowed because of the steam hazard potential. But, since the measurements were done from another room, and the wall shielding accounted for in the calibration, these could be done during operation, thus shortening the subsequent outage.



Figure 4. Pipe measurements in contaminated area with ISOCS

NPPs have many tanks of liquids or gasses. Some of these are designed for representative sampling, but most are not. ISOCS can be effectively used to measure the entire tank. This is especially good where a representative sample of the contents of the tank is not easily obtained (plated to walls, precipitated, nonliquids, no sampling ports, no mixing methods), and when the contents are dangerous (high temperature, corrosive or toxic, high radioactivity, high pressure). With *in situ* spectroscopy, the process of taking the sample, transporting it to the lab, then further processing for counting, can be bypassed, and near-instantaneous results can be obtained. Figure 5 shows an example.



Figure 5. In situ measurements of an "empty" tank

Historically when something is too hot to deal with, it is put into the spent fuel pool. When cleaning campaigns are conducted, this material must be assayed for proper disposal. In the US, if certain nuclides have too high a concentration (greater than Class C) there is no waste disposal pathway. Underwater *in situ* spectroscopy with ISOCS has been used at several NPPs to classify this waste for proper shipment. The mathematical efficiency algorithms properly handle these geometries. Figure 6 shows a special detector housing with an external pin-hole collimator being placed in a spent fuel pool.



Figure 6. Underwater in situ measurements in spent fuel pool

Chemical or abrasive cleaning of the primary system is frequently done to reduce operational dose. ISOCS has been used at a Canadian NPP to monitor the progress of this effort to determine when that process is no longer effective. Figure 7 shows a steam generator with the various *in situ* measurement points.



Figure 7. In situ measurements to monitor the progress of steam generator cleaning

Concrete is frequently activated from neutron flux or contaminated from spills. To determine the proper remediation, one needs to know the activity vs. depth profile. Traditionally, this meant removing a core, slicing it in thin layers, followed by laboratory assay. But, *in situ* spectroscopy can do this without coring in many cases. As long as there are various gamma lines (e.g. Eu-152 in activated concrete) it can be done with a single measurement, and fitted to a series of distribution profiles to find the best match. For other gamma emitters (Cs-137, Co-60) this can be done with several collimated measurements from different angles. ISOCS was also successfully used during an outage to quickly quantify nuclides and activity in the wall to show that demolishing it would not cause an airborne hazard; therefore respirators or special tents were not required.

N-16 can be measured directly and quantified. Because of the short half-life, sampling isn't normally an option. ISOCS was used at a European NPP to quantify the N-16 content of an entire turbine building in a few hours, with results agreeing acceptably with those from many months of calculations.

ISOCS has been used successfully at a Southeastern US NPP as a temporary Whole Body Counter at the contamination control area. If the total body contamination monitors show activity, this is frequently from Xenon or Radon adsorption. The Ge spectrum can document this, and prove the absence of other more significant radionuclide depositions. Using ISOCS as a portable WBC was very cost effective compared to the labor cost of taking the "contaminated" workers to the plant WBC nearly a mile away.

Free-release measurements are very common at any facility handling radioactive material. In situ gamma spectroscopy is very effective to assay complex items that are difficult or impossible or expensive to fully assay with traditional hand-held beta instruments. At a Northeast US NPP ISOCS has been used to free release heat exchangers without destructive assay, proving that the internal contamination did not exceed regulatory limits, as shown in Figure 8. It has been used to prove that various other large and complex items are not contaminated at various other NPPs, as shown in Figure 9 from a European NPP project. It can even be used to survey entire rooms in a single measurement using an unshielded detector in the center of the room, as shown in Figure 10. Nuclides that might be hidden from conventional surveys would be easily detected here with minimal effort.



Figure 10. An unshielded detector can assay an entire room for hidden contamination

Figure 8. ISOCS used to assay heat exchangers for release

CONCLUSION



Figure 9. In situ spectroscopy can assay complex shapes that are difficult to survey

In situ gamma spectroscopy is now a practical measurement tool for the operational health physicist. It is reasonably robust and easy to use. It requires minimal additional skill beyond traditional gamma spectroscopy and an understanding basic physics of radiation interaction. When conventional measurements are combined with nuclide-specific quantitative measurements, the NPP health physicist can now make better and quicker evaluations of the radiation status of the working environment and the appropriate action to be taken to best accomplish the job at the lowest cost and dose.