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# PERFORMANCE OF TRANSMISSION-CORRECTED HIGH-RESOLUTION GAMMA ASSAYS IN A NUCLEAR FUEL FABRICATION FACILITY

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## ABSTRACT

The usefulness of a portable high resolution gamma assay unit in a nuclear fuel fabrication facility quickly becomes apparent when improved characterization of intermediate products, wastes, and other materials within the operating facility is desired. The performance of this high-resolution gamma assay method was examined and the method was qualified for a wide variety of intermediate products and wastes. The method proved to be versatile and accurate and was quickly integrated into an operating facility for fuel processes, criticality safety, nuclear material accountability, material storage, and waste disposal.

Nuclear fuel fabrication at AREVA's Richland, WA facility uses many different kinds of processes, including conversion of uranium hexafluoride to uranium oxide, powder pelletizing, rod fabrication and bundle assembly, solvent extraction, solid and liquid uranium extraction and recovery, and incineration. The facility works with enriched uranium-bearing materials and other isotopes which must be characterized for processing. Qualification results are reported for uranyl nitrate (UNH) enrichment, uranium-bearing materials, dewatered waste sludge, uranium ash, waste liquids, contaminated soils, and combustible wastes.

High-purity germanium (HPGe) detector-based spectrum data were transmission-corrected for container and matrix attenuation, geometry, collimator effects, and shifting facility background radiation levels. The assay results were compared against standards for validation and functional testing. The assay method was then qualified for each different type of uranium-bearing material by comparison with traditional methods of characterization involving grab and composite sampling, chemical extraction, and inductively coupled plasma/mass spectroscopy (ICP/MS) analysis methods. ICP/MS requires that solid samples have uranium extracted using a combination of nitric acid, microwave digestion, before analysis.

Results for the HPGe transmission-corrected assays compared favorably with ICP/MS under a wide range of physical characteristics such as specific gravity (0.3 -1.9), container thickness (0.05 – 0.1 inch), %U-235 enrichment (0.7 – 5%), uranium concentration (0.01 – 70 gU/l), and configurations such as metal and plastic drums, B-25 boxes, in-situ soil and sands, buckets and pails, and a variety of sample bottles. An overall correlation coefficient $R^2 > 0.998$ is reported for a variety of containers and materials containing between 2 to 15,000 grams of enriched U under in-plant operating conditions. As expected, assay accuracy decreased for lower uranium materials such as slightly-contaminated soil, where background effects from stored and in-transit uranium in the plant become more significant.

## INTRODUCTION

Fabricating nuclear fuel at AREVA's Richland, WA facility requires many different processes such as conversion of uranium (U) hexafluoride to U oxide, powder pelletizing, rod fabrication and bundle assembly, solvent extraction, solid and liquid U extraction and recovery, and incineration. The company has also designed and built many different waste treatment processes for sludge retrieval and dewatering, heavy metals removal, ion exchange, filtration with filter aids, soils washing, and selective uranium leaching and crystallization. Low-level and radioactive mixed (hazardous) wastes are assayed, packaged and disposed offsite. Enriched U-bearing materials containing other isotopes are characterized for processing and for disposal. The usefulness of a portable high-resolution gamma assay unit in a nuclear fuel fabrication facility quickly becomes apparent when improved characterization of intermediate products and wastes, and other materials within the operating facility is desired for productivity and analytical improvements.

An Ortec ISOCART High-Purity Germanium (80% HPGe) system equipped for field assay and office transmission-correction was used in this work. The field HPGe assay unit was used to confirm or improve the U assay of old drums containing wet wastes, filters, and mop heads, that were packaged that were assayed years ago. There are many applications where HPGe assays can be used to evaluate existing criticality safety control assays as well. Production rates increased by receiving results via HPGe rather than lab analysis.

Another obvious advantage of in-situ gamma assay over laboratory methods of grab sampling, and U extraction from small (1-2g) samples, is the ability to measure the overall gamma flux from an object, sample, waste, or other material. Laboratory sampling relies on homogeneity and efficiency of extraction of U from the solids, but has very accurate isotope analysis of the liquid phase using Inductively Coupled Plasma/Mass Spectroscopy (ICP/MS). Since performance is currently measured lab analyses terms, HPGe results were compared against them, with care was taken to ensure both lab sampling is representative and transmission-correction modeling parameter accuracy.

The performance of this HPGe field assay unit was examined and the method was qualified for a wide variety of intermediate products and wastes. The method proved to be versatile and accurate and was quickly integrated into an operating facility for fuel processes, criticality safety, nuclear material accountability, material storage, and waste disposal.

HPGe spectrum data were then transmission-corrected for container and matrix attenuation, geometry, collimator effects, and shifting facility background radiation levels. The assay results were compared against laboratory results and standards, for validation and functional testing. The assay method was then qualified for each different type of U-bearing material by comparison with traditional methods of characterization involving grab and composite sampling, chemical extraction, and inductively coupled plasma/mass spectroscopy (ICP/MS) analysis methods. ICP/MS requires liquid phase for analysis, and solid samples must have U extracted using a combination of nitric acid, heat, and microwave digestion, before analysis.

Qualification results are reported here for uranium-bearing materials such as dewatered waste sludge, combustible wastes, incinerator ash, contaminated soils, and uranyl nitrate (UNH).

## RESULTS

Results for the HPGe transmission-corrected assays compared favorably with ICP/MS under a wide range of physical characteristics such as specific gravity (0.3 -1.9), container thickness (0.05 – 0.1 inch), %U-235 enrichment (0.7 – 5%), uranium concentration (0.01 – 70 gU/l), and configurations such as metal and plastic drums, B-25 boxes, in-situ soil and sands, buckets and pails, and a variety of sample bottles. An overall correlation coefficient $R^2 > 0.998$ is reported for a variety of containers and materials containing between 2 to 15,000 grams of enriched U under in-plant operating conditions. As expected, assay accuracy decreased for lower uranium materials such as slightly-contaminated soil, where background effects from stored and in-transit uranium in the plant become more significant.

### Uranium-Bearing Materials

Results shown in Figure 1 below are HPGe field analyses of U-bearing materials having a wide range of U concentrations, with a variety of container shapes, weights. Dewatered waste sludge, debris, soils, and incinerator ash data are represented in the data below. The results compare well over 3 orders of magnitude of concentration, and give a correlation coefficient of 0.998. Soil results from 21 soil samples are discussed in greater detail later (see Figure 1).

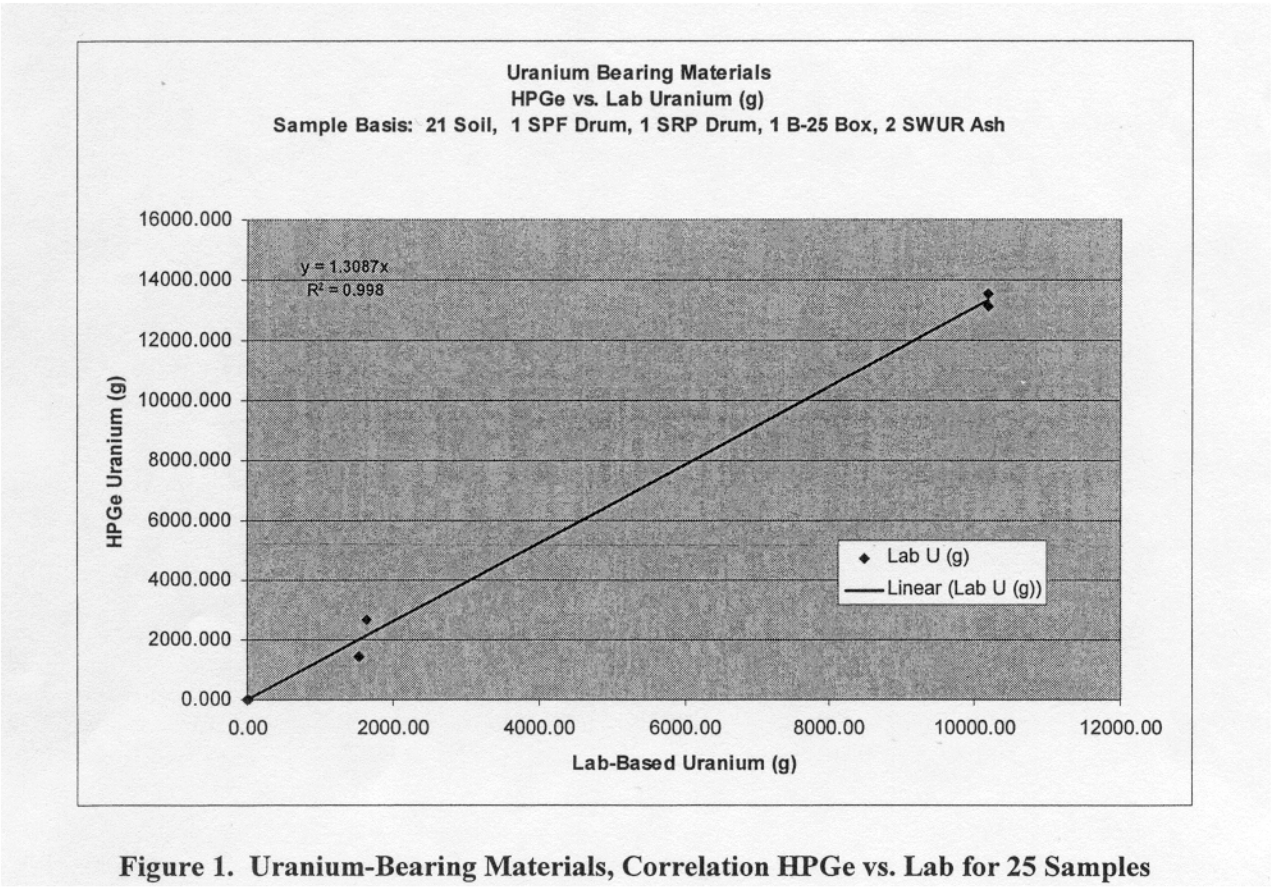


**Figure 1. Uranium-Bearing Materials, Correlation HPGe vs. Lab for 25 Samples**

Enhancements to Criticality Safety

Criticality safety methods employed at this site rely upon 3 independent controls, including U concentration, %U-235 enrichment, and mass controls. Data provided by the HPGe unit can provide all three, and this is especially important in verifying lab results independently. Laboratory response time is typically 1 – 3 days, while the HPGe unit gives results in under an hour, and this has improved plant productivity and reduced lab analysis costs in some cases. This savings is higher where composite sampling, and U chemical extraction, and both ICP/MS or KPA analyses are all required.

Wastes which are not homogenous can make representative sampling and characterization difficult. The HPGe unit was used to assess and enhance existing criticality safety by confirm the analyzed safety margins and comparing results with existing sampling and analysis methods. Typical waste containers are shown in Figure 2.

Ash from incinerators contain silica powders, metals, glass and aluminum balls, klinkers, and other in-homogenous materials. Samples of each of these materials are retrieved, pulverized, and homogenized to obtain a representative sample. The sample is sent to the lab, and a 1 - 2 g aliquot is retrieved from the sample for analysis. Methods such as this which rely on human performance to ensure that the sample taken in the field is representative. The U-235 enrichment level of the ash must be monitored because feedstocks and enrichments are always changing and this affects batch size and safety margins.

Thirteen (13) pails of ash from a randomly selected campaign were analyzed for enrichment. The HPGe and lab methods compared favorably, with average U-235 enrichment of 3.69% vs. 3.59% respectively, with standard deviations of 0.083 and 0.073 respectively. Some differences in individual samples were seen, as expected, due sampling and analysis error, and differences in methodology.

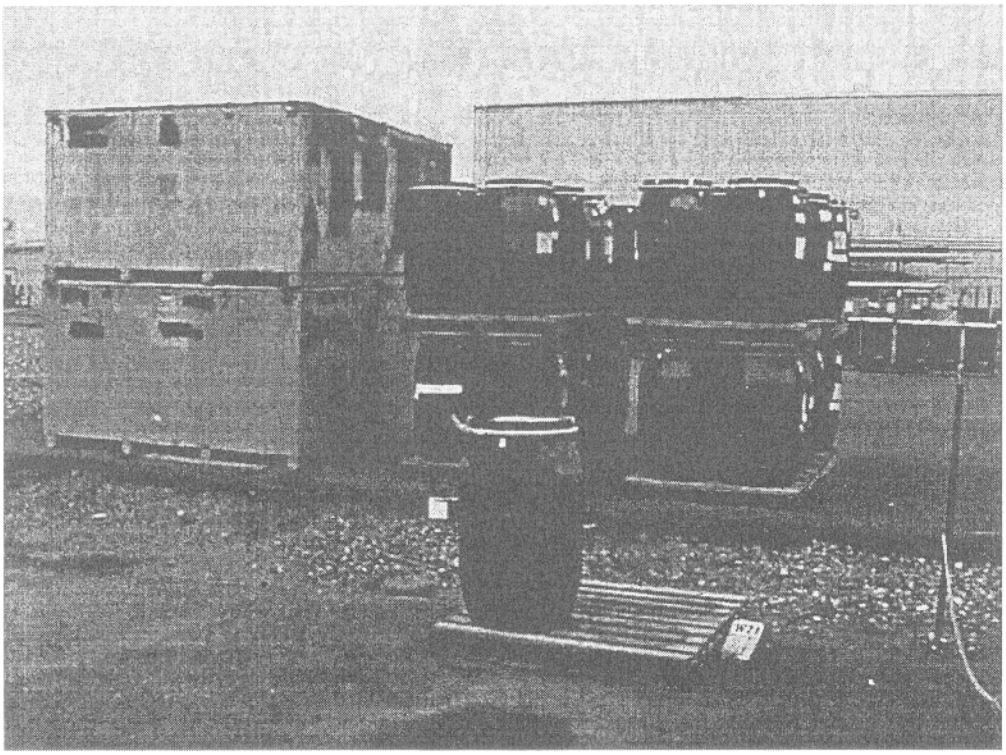

Figure 2. Miscellaneous U-Bearing Materials

In another facility, sludges are retrieved by dredge and quarantined in a large feed tank, where mixers suspend the sludges in a slurry. HPGe analysis were used to confirm the accuracy of lab analyses on U mass and enrichment. The variability of enrichment and concentration within the sludge was studied, and the results for a one lab grab sample and analysis vs. 11 HPGe analyses in the same batch of sludge gave U concentrations of 3.5 g/l and 3.5 g/l respectively, and the 11 HPGe analyses had a standard deviation of 0.286. Some variability within the sludge was confirmed but was minor, and mixing efficiency was confirmed. For the enrichment data, 11 samples in the batch were analyzed via HPGe and averaged 2.69% U-235 with a standard deviation of 0.234, and 95% confidence level of 0.157, while the single grab sample analyzed by the lab yielded 2.88% U-235. HPGe unit was able to discern the variability within the slurry and also confirmed the safety margin for the slurry, which had a limit of <4% U-235.

Uranyl Nitrate is an intermediate product resulting from the dissolution of U powders, U pellets, U-bearing solids and wastes, and solvent extraction. Drums of UNH are stored in warehouses pending processing and the conversion $UO_2$ or $U_3O_8$. A need arose for in-situ enrichment data for a project involving UNH conversion. The analyses would provide an over-check of lab results, mainly to ensure the UNH was <5% U-325 enrichment for criticality safety purposes. The HPGe assay unit was moved into the warehouse, with its diverse background radiations, and an attempt was made to determined the impact of using only 1 background correction in a complex environment where hundreds of drums are stored in close proximity. The detector was placed within 1" of the drum.

Results show that all drums were below 5% U-235 as expected, and there was a good correlation in the range tested (2.5 – 3.9% U-235). The method yielded a correlation coefficient of 0.9419 for 7 different drums this range of enrichments. Sources of error include varying background radiation from other stored drums in close proximity to the rear of the lead-collimated detector.

Contaminated Soils and Demolition Debris

Contaminated areas in the plant were representatively sampled by taking numerous grab samples, compositing and homogenizing, and submitting a sample of the composite to the lab. These results were compared against an HPGe assay for a disk of soil 4" deep and about 7 feet diameter, and the total U present in that disk are shown in Figure 3 below. Lab sampling and analysis error, grab sample variation, and changing background errors are the likely sources of variation between the two.

All in-situ soil areas were assayed for 10 minutes, with a separate background analysis except for points 17-21. The effect of background radiation from stored fuel onsite can be seen in points 17 – 21 where samples were taken near a fuel storage area. Without no background correction, the apparent detector U results increased as the HPGe unit approached the fuel, even with 5/8" lead shield. A 2" lead shield is better, and in a fuel fabrication facility, or any operating facility, the dynamic background changes from ongoing operations will make it more difficult to get accurate results. This is especially challenging on very low U samples, and samples which are near the natural background background. Therefore, frequent background readings must be taken and any unusual fuel or product movements in the vicinity must be taken into account on low U assays. A low-traffic, low-background remote area is recommended for assaying low U materials.

Buildings and structures are frequently modified or demolished to make room for new processes, process enhancements, or closures (Figure 4). Debris is typically hand surveyed. In-situ HPGe assays can confirmed that the object is clean or prove it exceeds cleanup levels. Field assays of the cement, insulation, ductwork, sheetrock, and other materials can be done quickly in an area of low-background radiation and low traffic. In an operating fuel fabrication facility this is more difficult as U-bearing materials are being moved frequently and may impact the background levels used in the analysis, but a selecting a low-traffic area can minimize this effect.

Demolition work, which relies upon hand-held survey meters and laboratory sampling to dispose of waste, can proceed more quickly when the HPGe unit is used in conjunction with, or replaces, traditional methods of characterization. The HPGe unit is superior to lab analysis because results can be deliver within a few hours rather than days. And wastes can be packaged for disposal the same day they are generated. This reduces the amount of debris that can accumulate, awaiting lab results, and make for a more cost-effective operation.

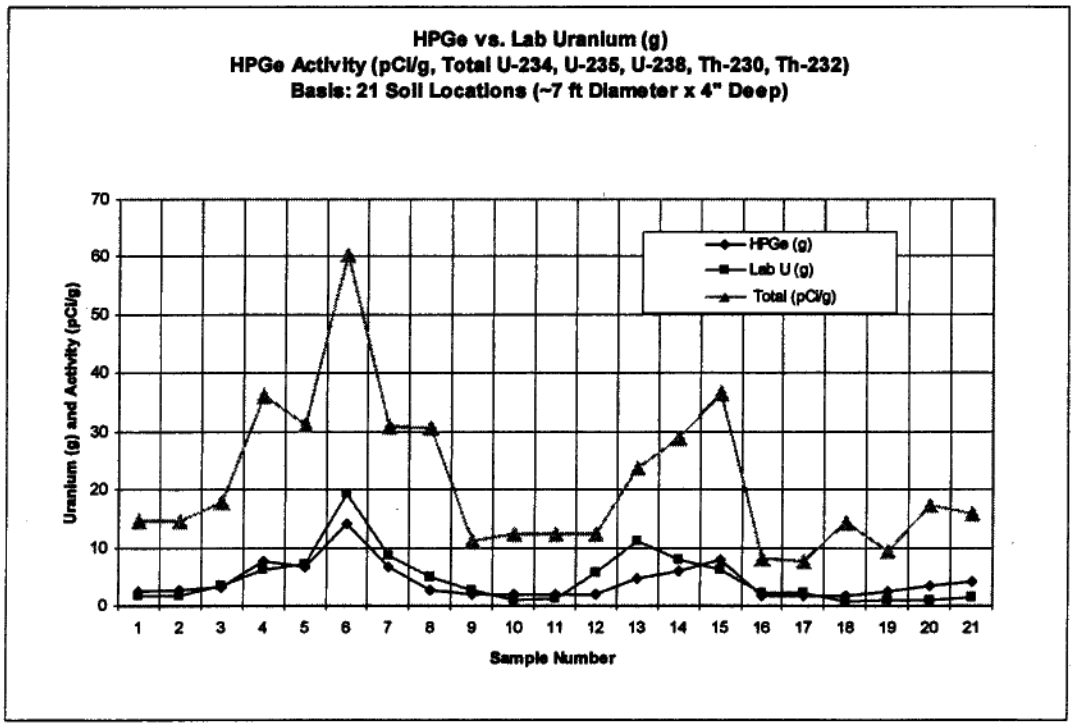


**Figure 3. Comparison of HPGe vs. Lab Uranium Results**

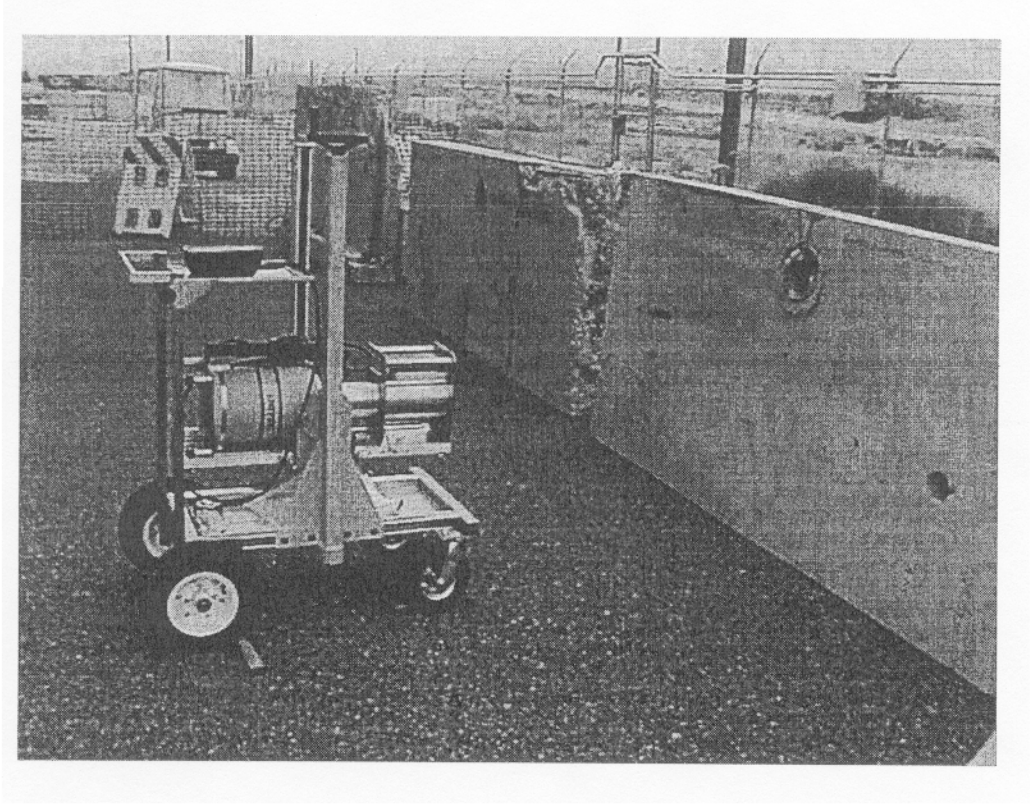

**Figure 4. Building Debris Being Verified as Clean with the field HPGe Assay Unit**

## Conclusions

The usefulness of a portable high resolution gamma assay unit in a nuclear fuel fabrication facility quickly becomes apparent when improved characterization of intermediate products, wastes, and other materials within the operating facility is desired. The performance of this high-resolution gamma assay method was examined and the method was qualified for a wide variety of intermediate products and wastes. The method proved to be versatile and accurate and was quickly integrated into an operating facility for fuel processes, criticality safety, nuclear material accountability, material storage, and waste disposal.