MATHEMATICAL EFFICIENCY CALIBRATION OF GE DETECTORS FOR LABORATORY SAMPLE GAMMA SPECTROSCOPY

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The current practice in most radioanalytical laboratories is to perform efficiency calibrations with radioactive sources. From the technical standpoint, and from an economical standpoint, this is an excellent solution when water-based calibration sources are used to prepare calibrations for water-based samples. But, there are problems with this common technique, both technical and economic.

- Samples that are not water [e.g. soil, gases, vegetation, metals, …] have different density and Z, and therefore will have different efficiencies than water.
- Chemicals can plate or precipitate out of the solution, or volatilize, and therefore have a different efficiency than intended.
- It is difficult to find calibration-quality long half-life water-soluble standards at medium energies, and at low energies and near K/L edges where many datapoints are needed.
- Many nuclides have coincidence summing effects and therefore are not suitable for close geometries.
- Radionuclides decay, and eventually must be replaced.
- Liquid radioactive sources require special handling and have potential contamination risks to the laboratory, but surrogates for the liquids rarely have the same response function.
- Source-based calibrations are expensive, when the cost of materials and labor are considered. These costs include source purchase, container costs, labor for calibration sample preparation, labor to perform the calibration and documentation, labor for source inventory maintenance, and source disposal costs.

Mathematical calibrations have been successfully performed in the past by several methods. Some use radioactive sources as a reference point, or as an angular response correction factor [e.g. HASL-258 for *in-situ* measurement of soils], but these are not useful for close-in laboratory geometries. Some [e.g. the K-zero technique for neutron activation analysis] use mathematical formulas to compute the intrinsic response of the detector, but only work for simple geometries, and not complex shapes like Marinelli beakers. Many Monte Carlo techniques exist [e.g. MCNP, GEANT, EGS] but these are complex to program and take significant computer time. But, when properly applied, they do give excellent results. [1] While these can work fine for reference and special applications, they are not generally suitable for the ordinary radioassay laboratory.

Canberra has recently introduced a new computer program, ISOCS [*In-Situ* Object Calibration Software] that allows the user to perform complete mathematical efficiency calibrations of Ge detectors. No radioactive sources are necessary, except for efficiency/FWHM calibrations, and this can be a single low-cost un-calibrated multi-emergy point source. The original application for this calibration software has been for *in-situ* counting, where the large sample size leaves the users with few calibration options, however, ISOCS is also useful for laboratory calibrations.

The ISOCS mathematical efficiency calibration software uses a combination of Monte Carlo calculations and discrete ordinate attenuation computations. At the factory, the complete dimensions of the customer's Ge detector and its mounting and housing hardware are placed into an MCNP model. Then a large number of point computations are run covering the 50-7000 keV energy range, the 0-50 meter distance range, and the 0-360 degree angular range. This large set of data is combined into a series of mathematical equations which are supplied to the user. The parameters for the equations are specific to the customer's detector, not just generic parameters. The user then selects the detector via. the ISOCS software, and enters the physical dimensions and parameters describing the source and it's relationship to the detector. Air temperature, pressure, and humidity are also used for attenuation corrections for spaces not occupied by the source. The ISOCS software then uses this information to compute the efficiency. This is done by a series of quasi-random source volume subdivisions. Attenuation corrections are then made for intervening source matrix, non-source attenuators like containers and air, and sample support shelves. This process is done in an iterative manner for each source volume and for each energy, increasing the number of source voxels each iteration, until the specified convergence is achieved. Although there are a great many computations, today's fast computers can do typical laboratory geometries in a few seconds.

Calibrations can be performed for many different sample shapes:
> Cylindrical objects viewed from end
> Cylindrical objects viewed from the side
> Box-shaped objects
> Marinelli beakers

Each of these objects can be located on the detector axis, or offset from it. The detector axis can be aligned with the sample axis, or at a different angle. The sample container can be of any elemental composition and density. The sample matrix can be of any elemental composition and density. The efficiency can be computed for 20 different energies from 50 keV to 7000 keV.

Validation testing of the ISOCS efficiency calibration software has been accomplished by comparing the ISOCS efficiency results with those from 109 reference sources. These comparisons covered *in-situ* geometries [47 tests], collimated geometries [15 tests], and laboratory geometries [47 tests]. Laboratory geometries were defined as small sources closer than 1 meter to the detector, and included points, liquid scintillation vials from the end and side, larger beakers, and Marinelli beakers. The conclusion of the validation testing for laboratory geometries was that the ISOCS computation method is accurate to within 4.5% sd at high energies and 7% sd at low energies. [2]

These uncertainties are believed to be consistent with the errors inherent in calibrations with radioactive sources, when the calibration variables described at the beginning of this document are considered. Improvements already completed in the factory detector characterization algorithm, and are underway for a more versatile sample container algorithm to better reflect the complex shapes of the bottoms and sides of laboratory containers. These should further reduce the sd of the ISOCS calibration.

As an example of the potential errors the authors have observed in typical laboratory calibrations, consider these 2 cases.

A: use of epoxy calibration standard with density 1.15 the for the assay of samples of water at density 1.0

B: use of epoxy soil calibration standard with a density of 1.6 for the assay of typical laboratory samples of dried soil at a density of 1.3.

Figure 1 shows the ratio of the efficiency of the calibration standard to the efficinecy of the sample. The sample was a cylinder 15cm diameter and 10 cm high, in a 2mm thick PVC container, on the endcap of a BEGe5025 Ge detector. ISOCS was used for these calculations. These errors of 3-8 % for water and 7-28% for soil can be avoided by the use of exact calibration standards, which is what ISOCS allows.
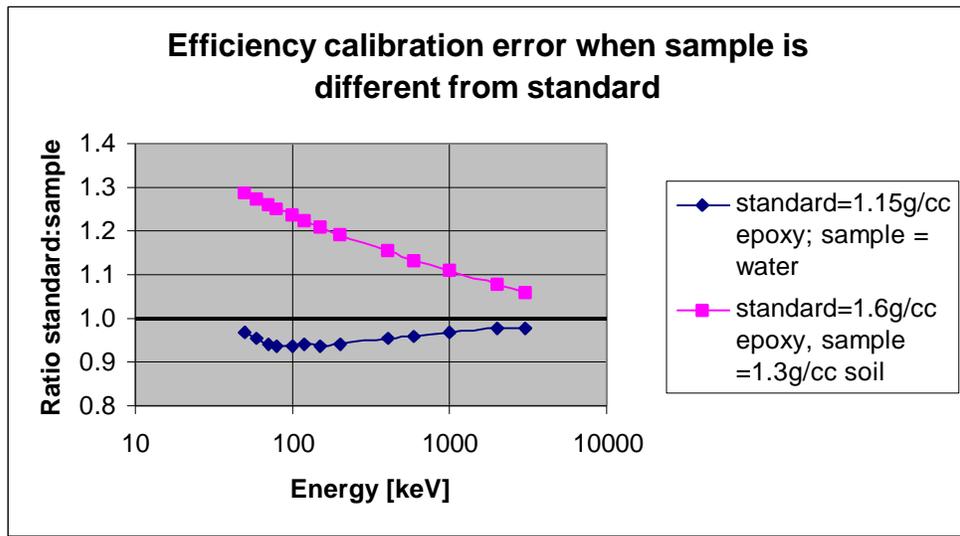
Also, consider what happens if there are small amounts of high Z materials in a sample that were not present during the calibration. Figure 2 shows the effect of the addition of a few percent of Fe in a soil sample that was not present in the calibration standard. The values are typical of Fe content in various types of soils from certain regions. Higher Z constituents have a proportionally larger effect.

Fig 2 Efficiency calibration error vs Fe soil content
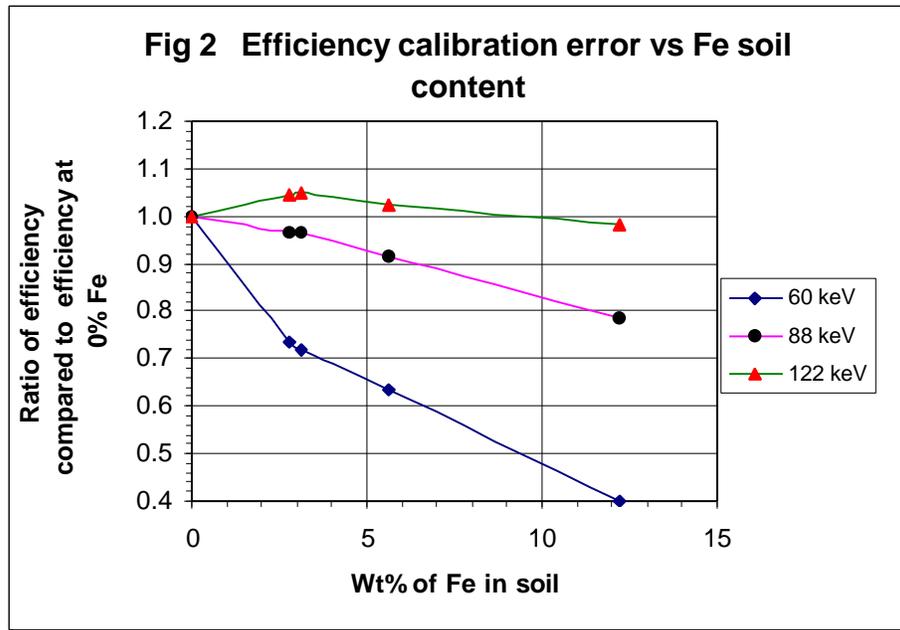
Mathematical efficiency calibrations also allow the reduction of the labor involved in sample preparation. No longer is it necessary to spend time forcing all samples to fit the few available calibration geometries. No need to try to make the samples match water in density. Place as much sample as available in the counting container to maximize the efficiency. Use whatever density and composition exists in the real sample. Calibrations can be performed in a few seconds for any sample type and sample height in the container.

Of course, a good quality assurance program is still necessary to prove to the laboratory operator and regulator that the calibrations are accurate and adequate. This includes a well designed and executed quality control program to prove that the characteristics of the detector have not changed since the characterization was done, and also includes participation in a blind proficiency testing program.

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[1] Validation of the MCNP Monte Carlo Code for Germanium Detector Gamma Efficiency Calibrations, F. L. Bronson and L. Wang, Canberra Industries, Inc., Waste Management '96 proceedings, Tucson AZ, Copies available from Canberra.

[2] ISOCS Efficiency Calibration Validation and Internal Consistency Document, ISOX-VIC 4/98, Canberra Industries, Inc. Copies available from Canberra.