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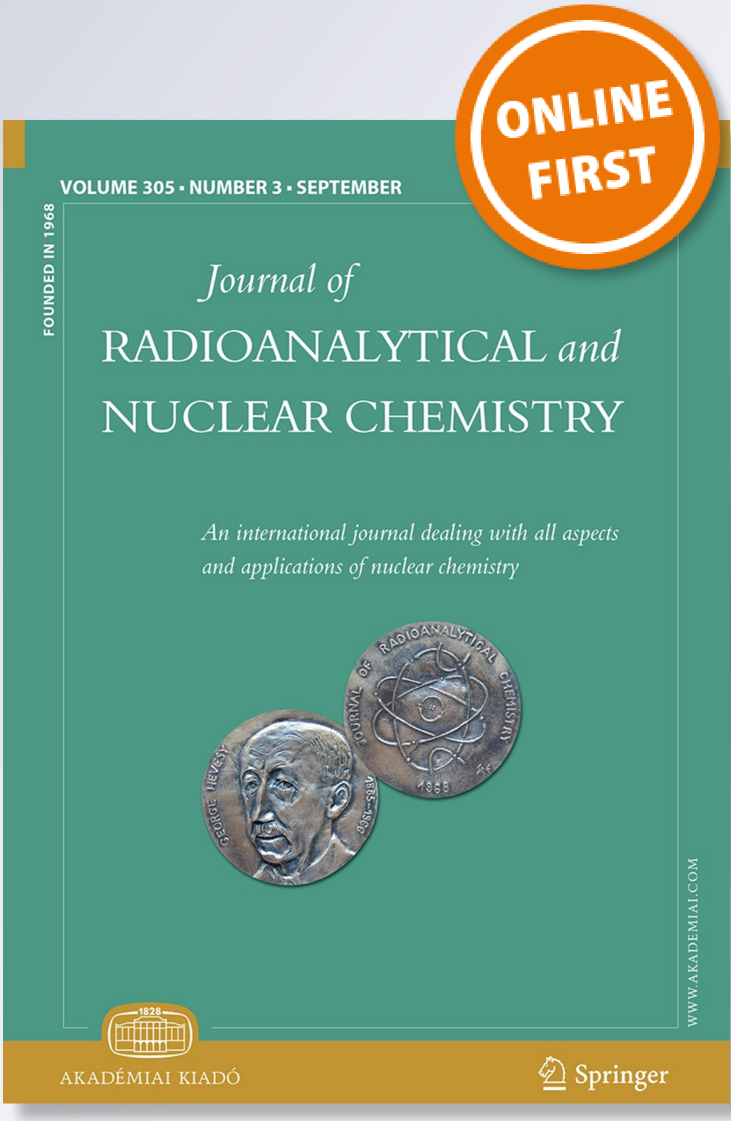

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A Monte Carlo approach to food density corrections in gamma spectroscopy

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Abstract Evaluation of food products by gamma spectroscopy requires a correction for food density for many counting geometries and isotopes. An inexpensive method to develop these corrections has been developed by creating a detailed model of the HPGe crystal and counting geometry for the Monte Carlo transport code MCNP. The Monte Carlo code was then used to generate a series of efficiency curves for a wide range of sample densities. The method was validated by comparing the MCNP generated efficiency curves against those obtained from measurements of NIST traceable standards, and spiked food samples across a range of food densities.

Keywords Gamma spectroscopy · Monte Carlo · MCNP

Introduction

Correction of the efficiency curve for food density is needed for gamma spectroscopy samples where the photon energies of the isotopes in the sample are low, and when the sample geometry is unfavorable (e.g. bulk samples placed in bins atop the detector). Traditionally this correction has been performed using a series of multiline standards at different densities in the geometry being used for the testing. This approach is both expensive and time consuming. Further, the list of isotopes necessary in the reference and proficiency test standards has been outlined by Jerome et al. [1]. and few of them are commercially available.

Alternative methods of performing the density correction using computer models of the high purity germanium (HPGe) crystal and counting geometry are also available. Jackman and Biegalski [2] have provided a summary of the codes that are available for this purpose, and Jonsson et al. [3] have provided an outline of errors associated with use of the codes to correct for geometry and density differences. One of the more popular codes is provided by Canberra Industries. Their package, LABSOCS/ISOCS [4], will develop and provide efficiency curves for many geometries. The HPGe crystal is characterized at the factory and the dead layer is measured. The tool then uses the Monte Carlo N-Particle Transport Code (MCNP) [5] and other methodologies to generate efficiency curves for numerous geometries and sample densities.

Ametek/Ortec also offers a package, ANGLE [6], which uses extended attenuation curves to generate photon efficiency curves for particular geometries and sample densities. This methodology requires one measured efficiency curve and detailed inputs on the crystal and sample geometry in use.

In this work, we use MCNP as a primary tool to generate food density efficiency curves for a characterized HPGe crystal and sample geometry. If the dead layer of the crystal is not known, an iterative approach is used to find it by generating MCNP efficiency curves that match a measured curve produced by a multiline standard at one density. By contrast, Novotny and To [7] used a non-linear least squares estimation to determine the unknown crystal characteristics, to include the dead layer.

Once agreement between the measured efficiency curve and the MCNP generated curve was achieved, the model was used to generate efficiency curves across the range of

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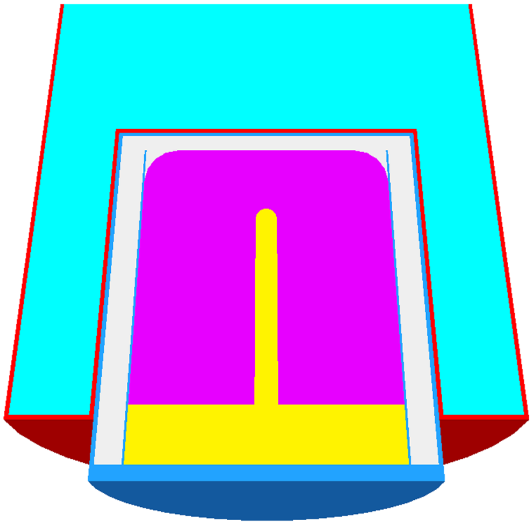

Fig. 1 MCNP model of the P type HPGe crystal and sample geometry used in this work

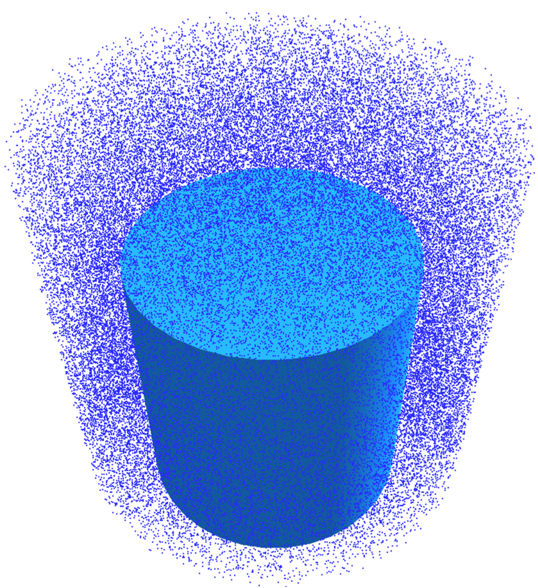

Fig. 2 Verification of the source region in the MCNP model. The radioactive material is uniformly distributed throughout the sample in the Marinelli beaker

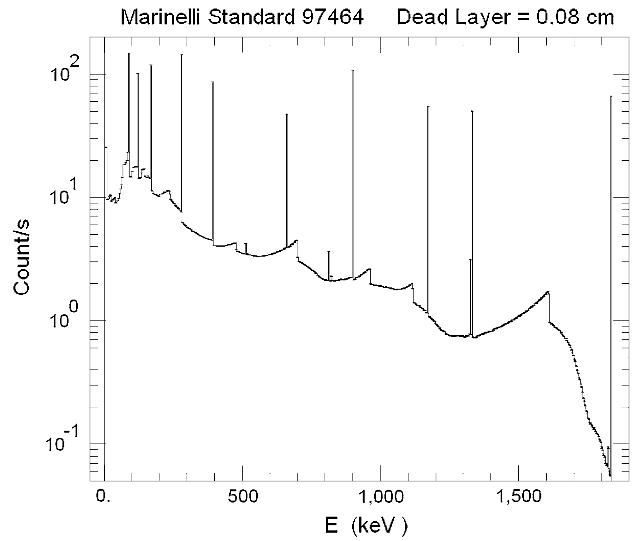

Fig. 3 MCNP generated spectrum of a multiline standard. The F8 tally was used

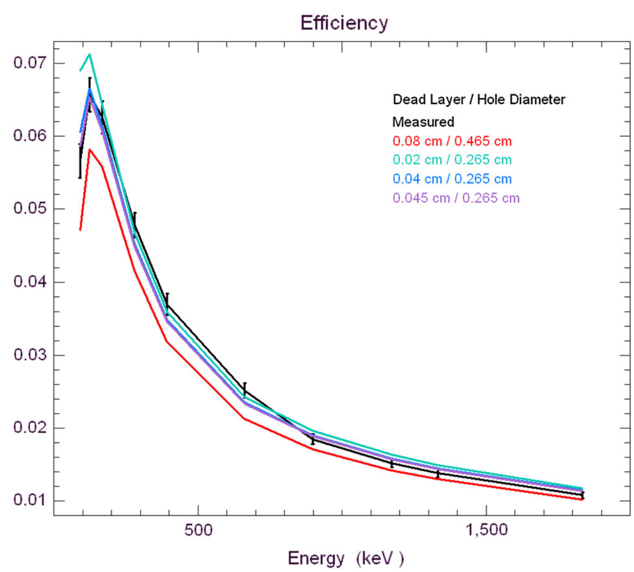

Fig. 4 MCNP generated photopeak efficiency curves at different dead layers on the HPGe crystal plotted with the measured efficiency curve from a multiline standard

food densities expected and was tested with food samples of various types spiked with ^{241}Am and ^{137}Cs.

The MCNP model

The primary MCNP model was developed using a geometry composer, Moritz [8], to speed the development of the input file for the transport code. Development and verification of the crystal model and counting geometry required 2 days, with one additional day to determine the unknown dead layer by iteration. The model, once complete, should not change significantly over the life of the crystal, except for minor adjustments of the dead layer. The crystal chosen was a traditional P type crystal, 62 mm in diameter and 60 mm in length with a 40 % relative efficiency. The thickness of the dead layer was unknown. For the food samples, a 500 mL Marinelli beaker was chosen. The HPGe crystal, the cold finger, aluminum cap, and polyethylene Marinelli beaker were modeled (Fig. 1), and

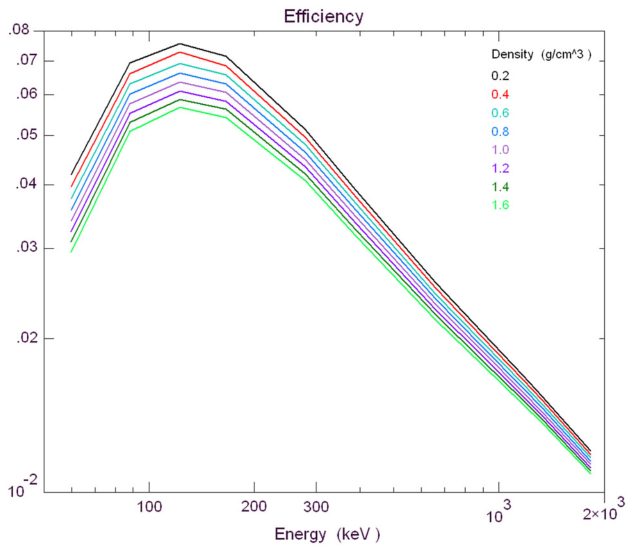

Fig. 5 MCNP generated efficiency curves over the range of expected food densities

the germanium dead layer was estimated. Once the model and source regions were verified (Fig. 2), a series of runs were performed with different dead layer thicknesses. Run times of 15 min with a multi-core Xenon processor were sufficient to generate 10^8 histories and adequate convergence of the statistical criteria. The MCNP F8 tally was used to generate a spectrum (Fig. 3) and the efficiency was determined for energies from 60 keV (^{241}Am) to 1.836 MeV (^{88}Y). The resulting efficiency curve (Fig. 4) was then compared to a measured efficiency curve generated with a multiline standard. The dead layer was then varied until a match was found at a dead layer of 0.045 cm.

Once the MCNP model was found to match the measured curve, a series of runs were made, varying the density of the sample in the Marinelli beaker from 0.2 to 1.6 g/cc (Fig. 5). This is the complete range of densities that are commonly found in food products.

Discussion

The 500 mL Marinelli beaker is a favorable geometry for samples of varying densities as the physical depth of the sample is small at any location. Consequently, there is little chance for self-absorption of energetic photons in the sample. A review of Fig. 5 reveals that the efficiency of the detector for 662 keV gammas from ^{137}Cs varies from 2.2 to 2.6 % across the whole range of food densities. When compared to the normal statistical uncertainty for low level environmental and food samples, the density correction is small and could safely be ignored. However, for the very low photon energy for ^{241}Am (59.5 keV), even this, the most favorable counting geometry, resulted in photopeak efficiencies from 3 to 4.2 %. Food density corrections are necessary for isotopes with low energy photons, even when the geometry is favorable. For unfavorable geometries (e.g. large bins of food set atop the crystal), density corrections are necessary at all densities and energies.

Validation

The MCNP model was validated by spiking food samples with standards of ^{241}Am and ^{137}Cs, and then comparing the results with and without a food density correction. The reference calibration of the gamma spectrometer was for the 500 mL Marinelli beaker with a water equivalent sample at 1.0 g/cc. Measured results with no correction, and those with density corrections derived from the MCNP

Table 1 Results of food samples spiked with ^{241}Am and ^{137}Cs standards

Food product	Density (g/cc)	Isotope	Spiked activity (Bq)	Measured activity (uncorrected)	% Error	Corrected activity (Bq)	% Error
Rice Crackers	0.245	^{241}Am	2.22	2.78	25	2.28	2.7
		^{137}Cs	2.22	2.11	5.0	1.94	1.2
Chaga Mushrooms	1.054	^{241}Am	8.46	7.87	7.0	7.96	5.9
		^{137}Cs[a]	–	7.60	–	7.60	–
Soy Sauce	1.18	^{241}Am	2.22	1.98	10.8	2.07	6.8
		^{137}Cs	2.22	2.09	5.8	2.13	3.8
Syrup	1.39	^{241}Am	8.89	7.20	20.0	7.96	10.4
		^{137}Cs	2.41	2.24	6.4	2.35	2.5

The uncorrected results are those provided by the spectrometer calibrated with a 1.0 g/cc multiline standard in a 500 mL Marinelli beaker. The corrected results were developed using the MCNP generated efficiency curves shown in Fig. 5 to correct for self absorption of the relevant photons in the sample matrix

[a] No spike added. Sample had 15 mBq/g ^{137}Cs in it on receipt

efficiency curves shown in Fig. 5. The results of the validation runs are shown in Table 1. The uncorrected results for the ^{241}Am spiked samples were quite poor for densities that varied significantly from the reference curve (1.0 g/cc). The density correction improved all of these results to within counting error. As expected, the accuracy of the ^{137}Cs results was not significantly improved with a food density correction as the photopeak efficiency for the 662 keV gamma did not change significantly across the normal density variation for foods counted in this geometry (500 mL Marinelli beaker).

Conclusion

An economical method for food density correction was developed using MCNP6 and a visual builder, Moritz, to speed the development of the input geometry for the transport code. The method was able to determine the unknown dead layer of a crystal by iteratively fitting MCNP developed efficiency curves to a measured one at a single sample density. Once the MCNP generated curve matched the measured curve at one density, the density of the sample was varied across the range of expected densities from foods and the MCNP generated efficiency curves were tabulated and plotted. Validation runs with spiked foods indicated that the method improved the accuracy of results for isotopes where the food product produced significant self-attenuation of the photons used to quantify the activity of the contaminant in the food.

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