A Monte Carlo Approach to Food Density Corrections in Gamma Spectroscopy

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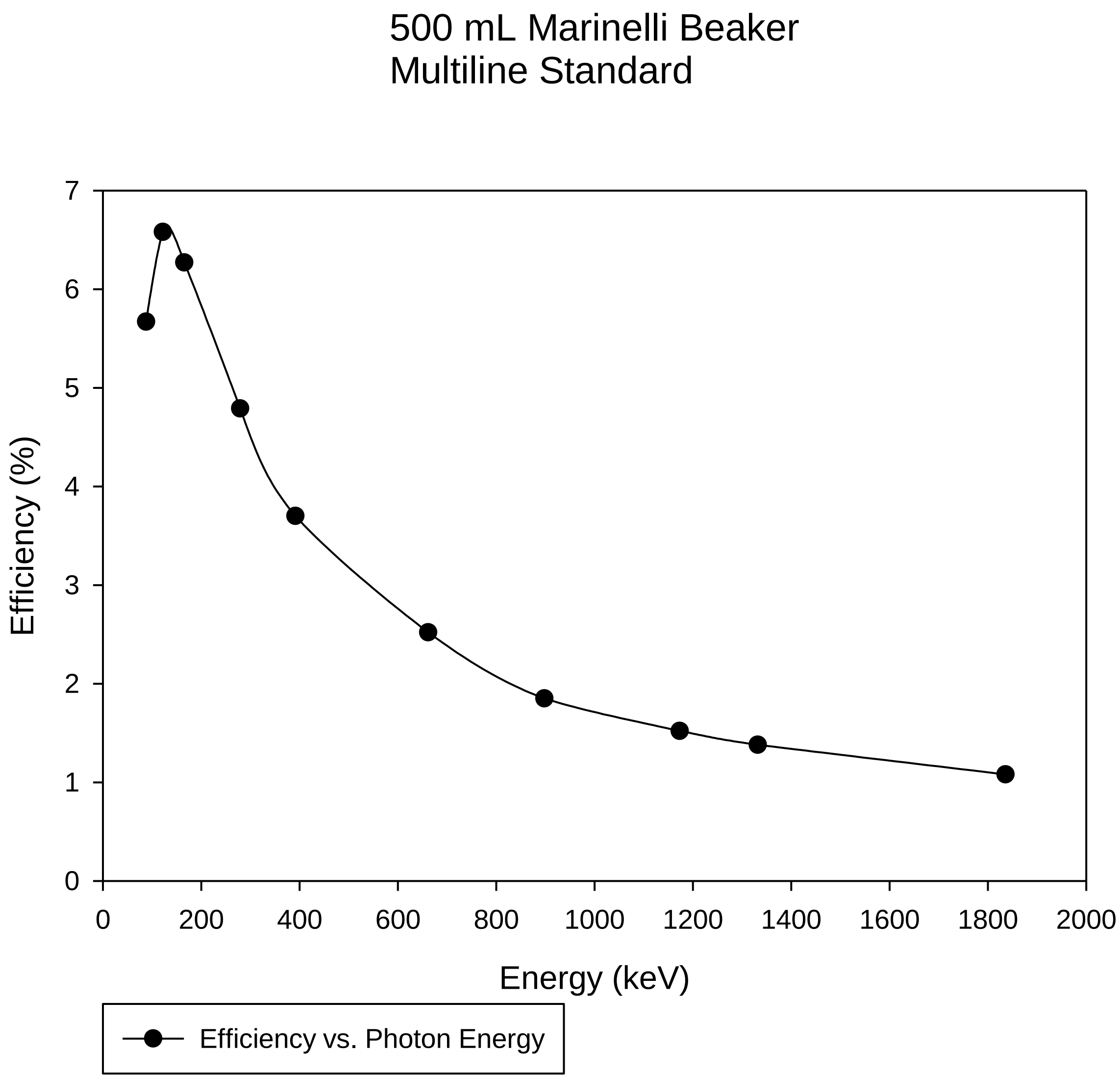
The Need for Food Density Corrections

- Self absorption of gammas from radionuclides in a sample can affect the efficiency curve used to quantify sample activity.
- The amount of correction can be trivial for isotopes with high energy photons (e.g. Cs-137) and favorable geometries (Marinelli Beakers).
- Alternately, isotopes with significant low energy photon emissions (e.g. Am-241), and unfavorable counting geometries, such as large buckets of the food product, can require significant corrections.

Correction Approaches - Analytical

- The most common technique is to simply order standards in the appropriate geometry at different densities over the expected range of densities seen and develop efficiency curves for each.
- The measured food product result is then corrected based on the interpolated value(s) of the efficiencies for the density of the food product under test.
- This approach is expensive and the standards cost about $690 each, with a disposal cost of $500 each every year.
- Cost is an important consideration in food testing as the margin is low for food products, and expensive test costs cannot be tolerated.

Measured Efficiency Curve



Correction Methods – ISOCS/LABSOCS

- Canberra markets a robust tool to develop geometries and generate efficiency curves for them.
- Based on a series of MCNP runs plus other methodologies.
- The dead layer is measured at the factory and then transport runs are then made for this particular crystal.
- Powerful and robust.
- Expensive.

Correction Methods - ANGLE

- Ortec/Ametek markets this technology based on extended attenuation curves for particular geometries.
- Requires detailed input on the geometry under study and the crystal dimensions.
- Requires an efficiency curve generated by one standard in the selected geometry.
- Iterates to find the dead layer.
- Can make multiple types of corrections (TCS, etc).
- Powerful and robust.
- Expensive.

Correction Methods – This Work

- Uses MCNP as a primary tool.
- MCNP6 is free!
- Speeds development of the inp file by using a graphical geometry composer, Moritz.
- Requires a standard calibration at one density.
- Iterates the thickness of the dead layer to match the measured calibration curve.
- Runs are then generated over the range of densities expected. This requires only one number be changed in the inp file.
- Validate with spiked food samples.

MCNP Method Advantages

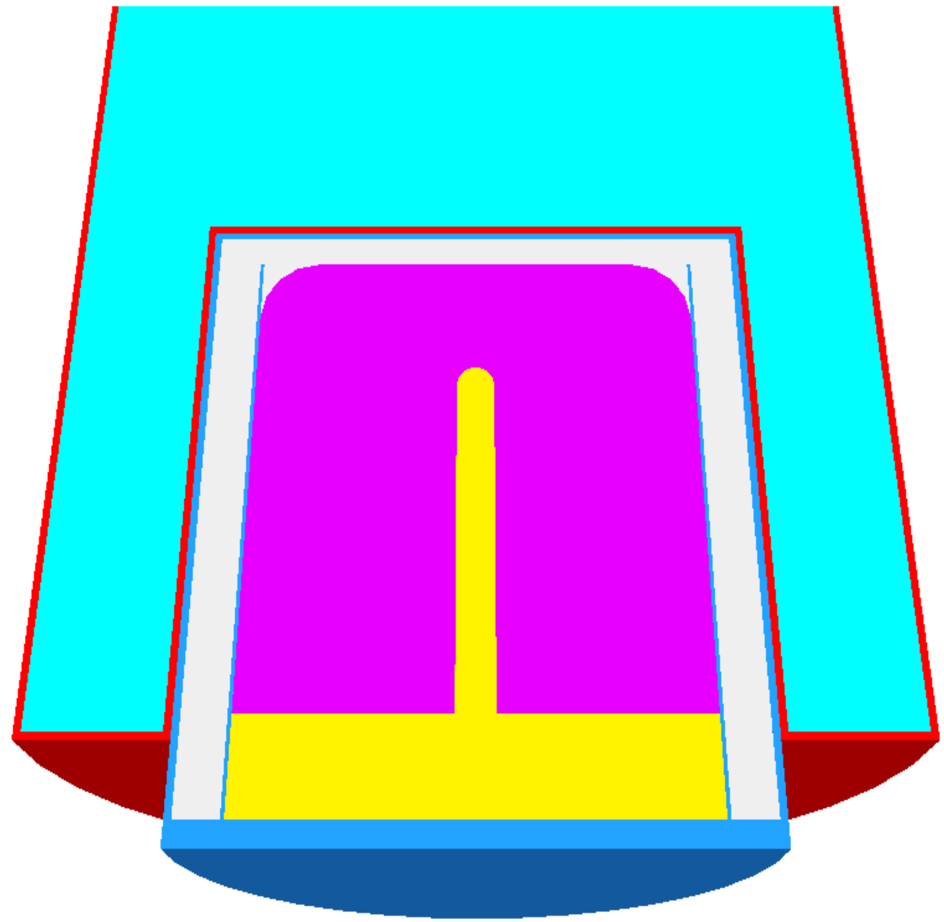
- Once the geometry is developed and verified, it will not change over the life of the crystal except to reflect changes in the thickness of the dead layer.
- For properly maintained crystals, the dead layer grows only slowly over the life of the crystal.
- Allows for a large number of density curves over the entire range of expected densities.
- Inexpensive.
- Can also be used for other materials (e.g. soil) and other corrections such as the peak to total values for True Coincidence Summing corrections.

MCNP Method Disadvantages

- Requires training to develop the inp file (or can be hired out).
- Must be verified by comparison of the MCNP generated efficiency triplets with those generated from a NIST traceable standard at one density. Most licensing authorities require one standard for each geometry.
- Iterative solution for the dead layer if the crystal has not already been characterized. Generally, starting with a .4 mm dead layer, it only requires three to four iterations to get the actual value.

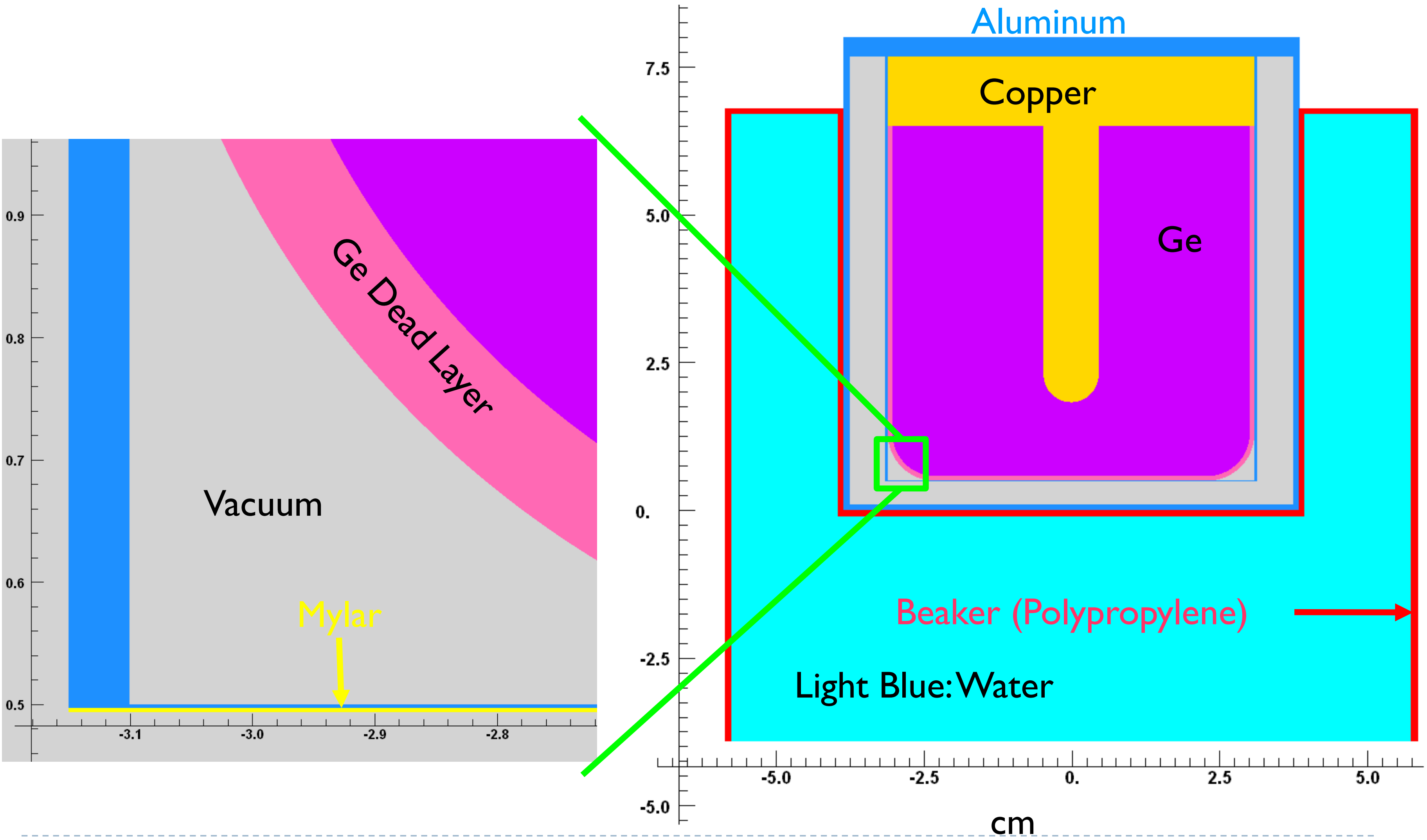
MCNP Method - Example

- Start using an uncharacterized P type HPGe coaxial crystal 62 mm in diameter and 60 mm in length, with a 3 inch endcap. 40% relative efficiency.
- For a geometry, use a 500 mL Marinelli beaker.

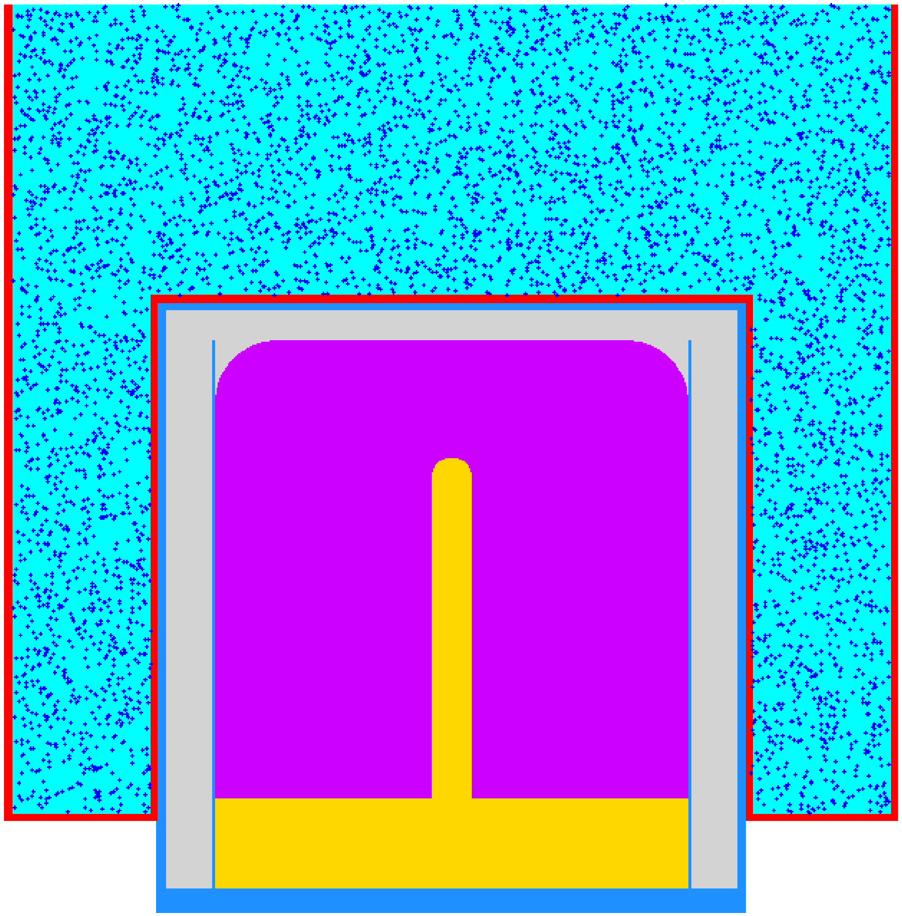


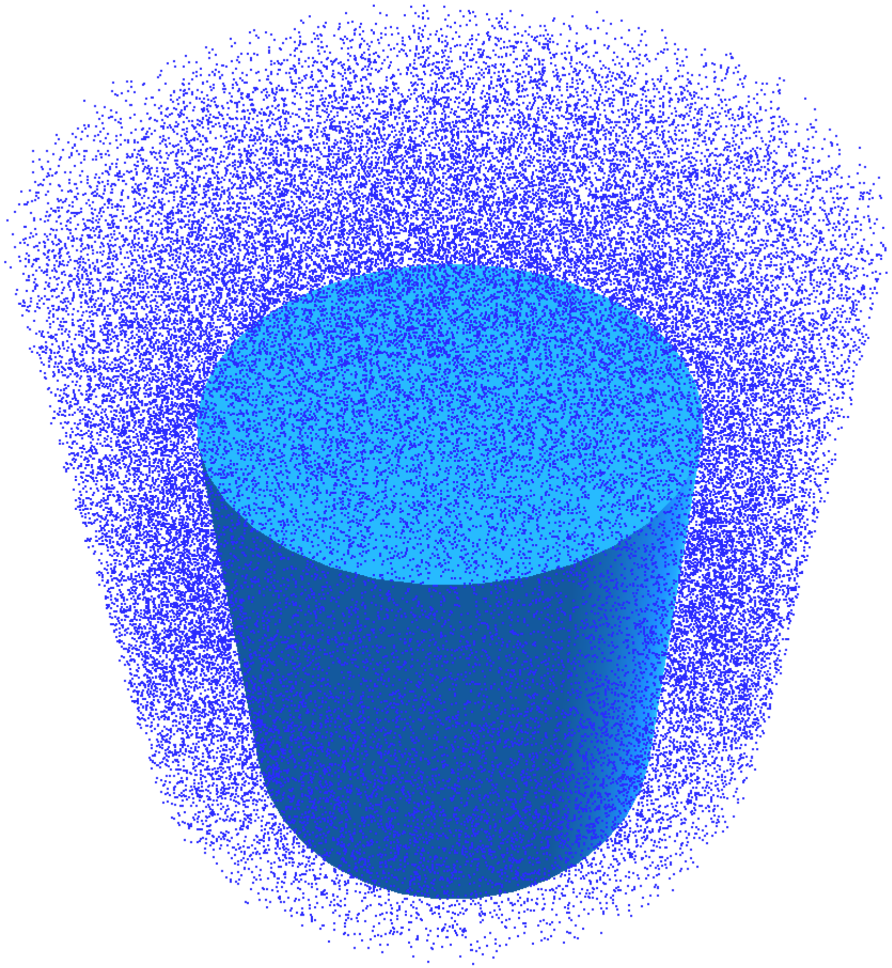
3D Cutaway of Detector & Beaker Model

MCNP Model: Materials

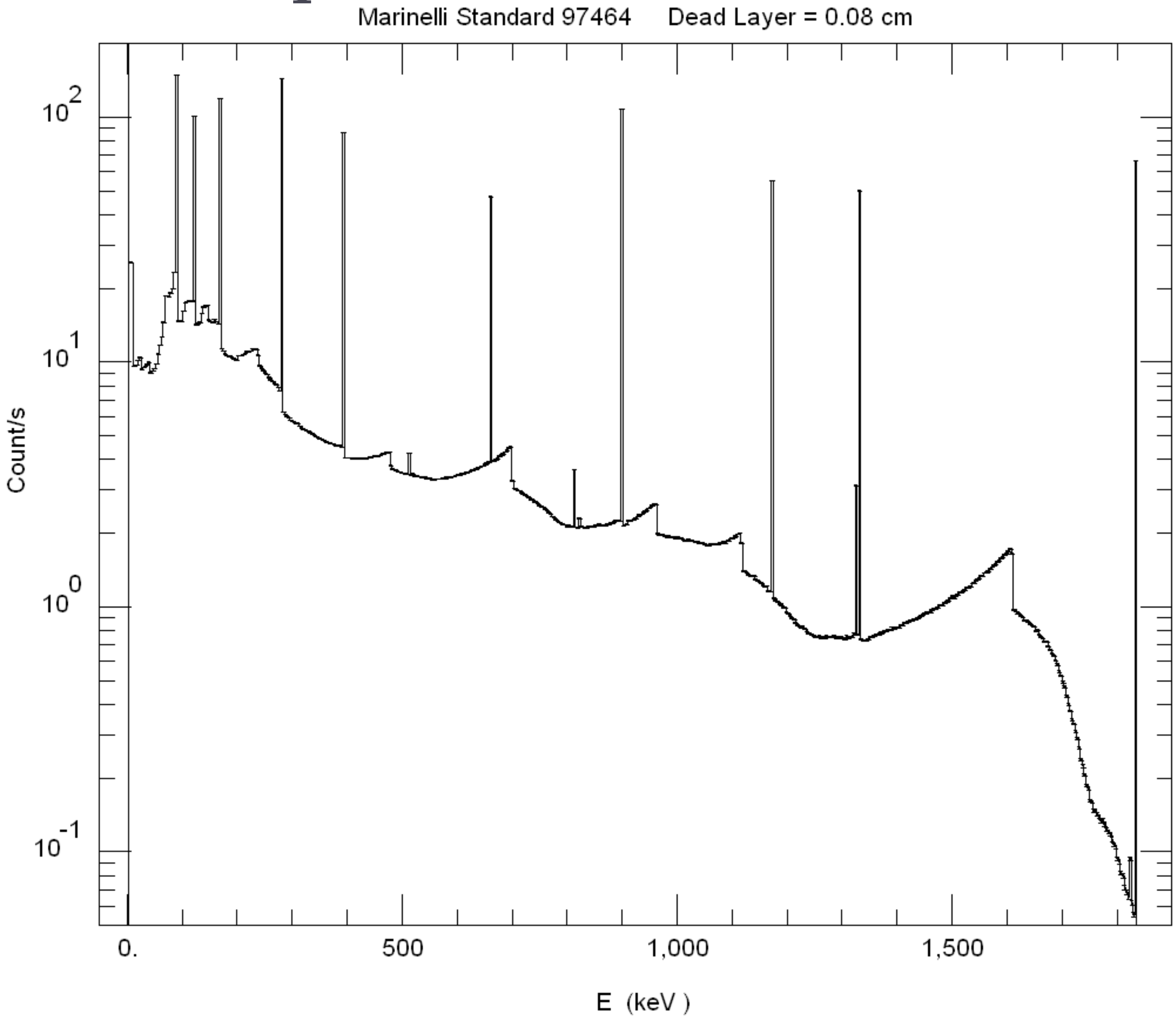


Monte Carlo Source Points





Calculated Spectrum – Multiline Standard



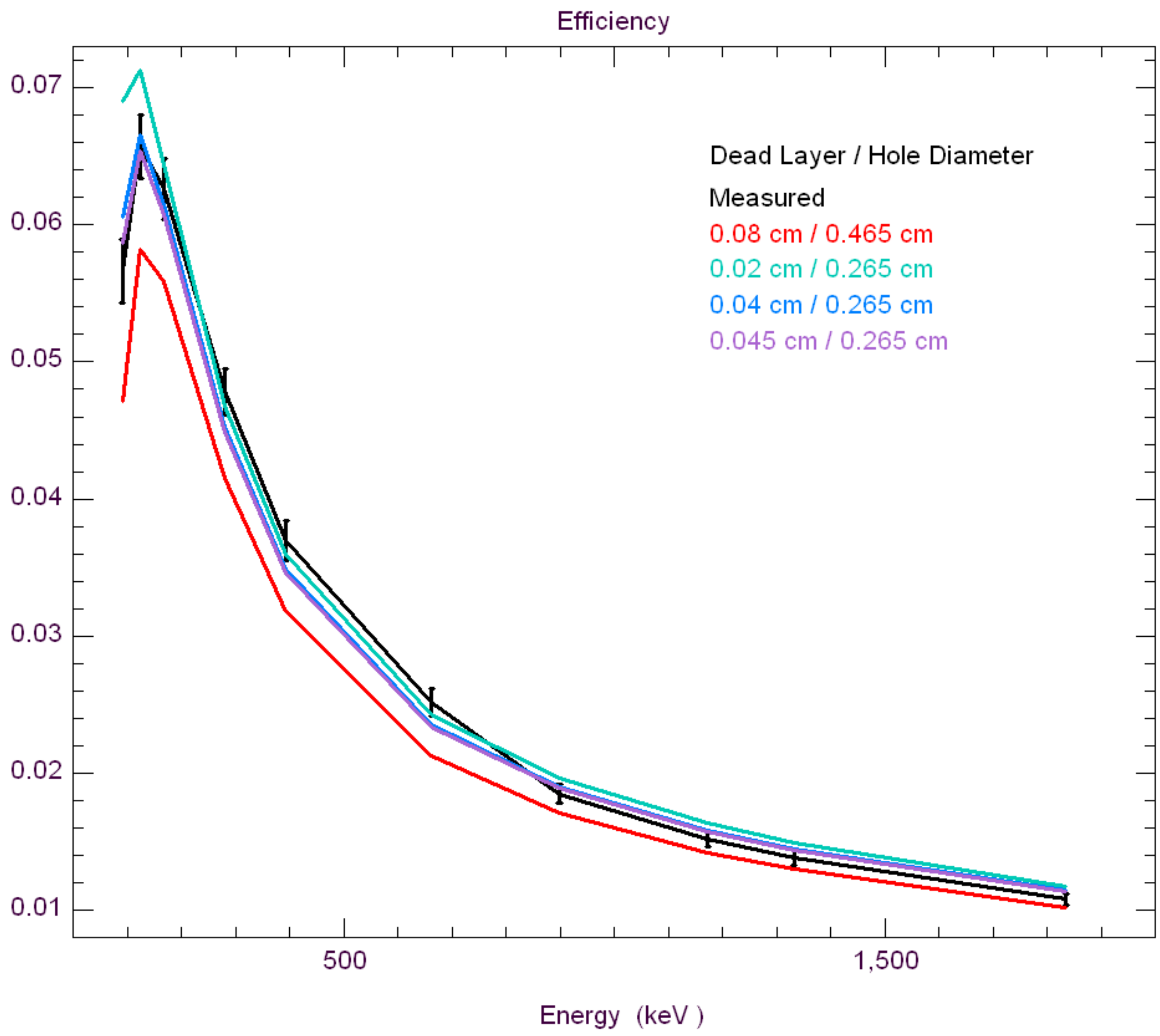
MCNP Example

- The example chosen is a very favorable geometry for food measurements and the thickest dimension of sample that photons must penetrate is only a little over a centimeter.
- Consequently little, if any, correction would be expected for energetic photons.
- However, we will show that even this most favorable geometry requires density corrections for low energy isotopes.
- The model will be validated using food samples spiked with Am-241 and Cs-137.

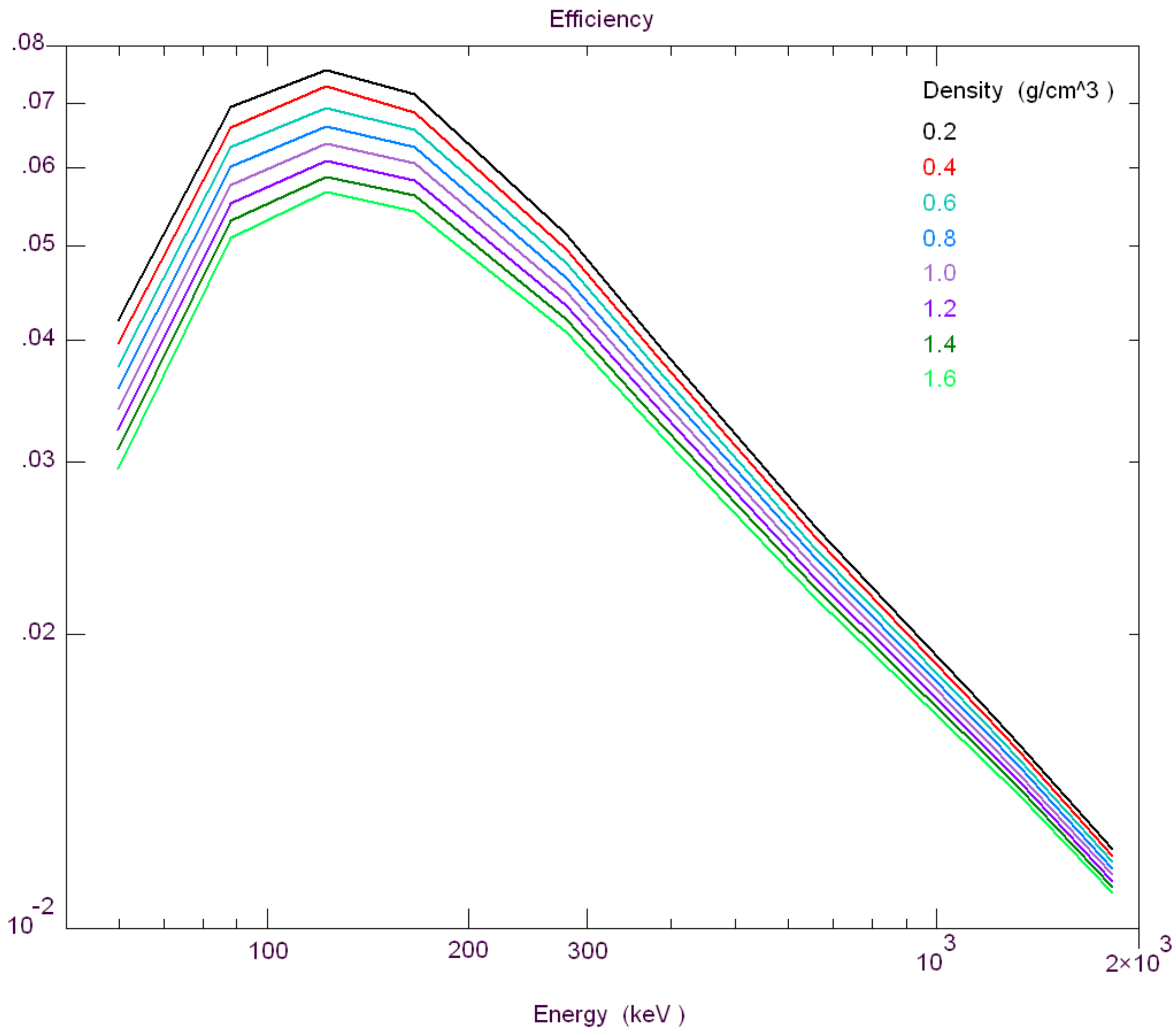
MCNP Model – Dead Layer Iteration

- The dead layer for the crystal chosen was not known, so the model was run for various dead layer thicknesses until the MCNP model matched the measured efficiency curve.
- Initial curve indicates that the assumed dead layer of 1.0 mm was too large.
- Additional runs at 0.4 and 0.45 mm were run.
- The 0.45 mm thickness for the dead layer provides the best match of the measured curve.

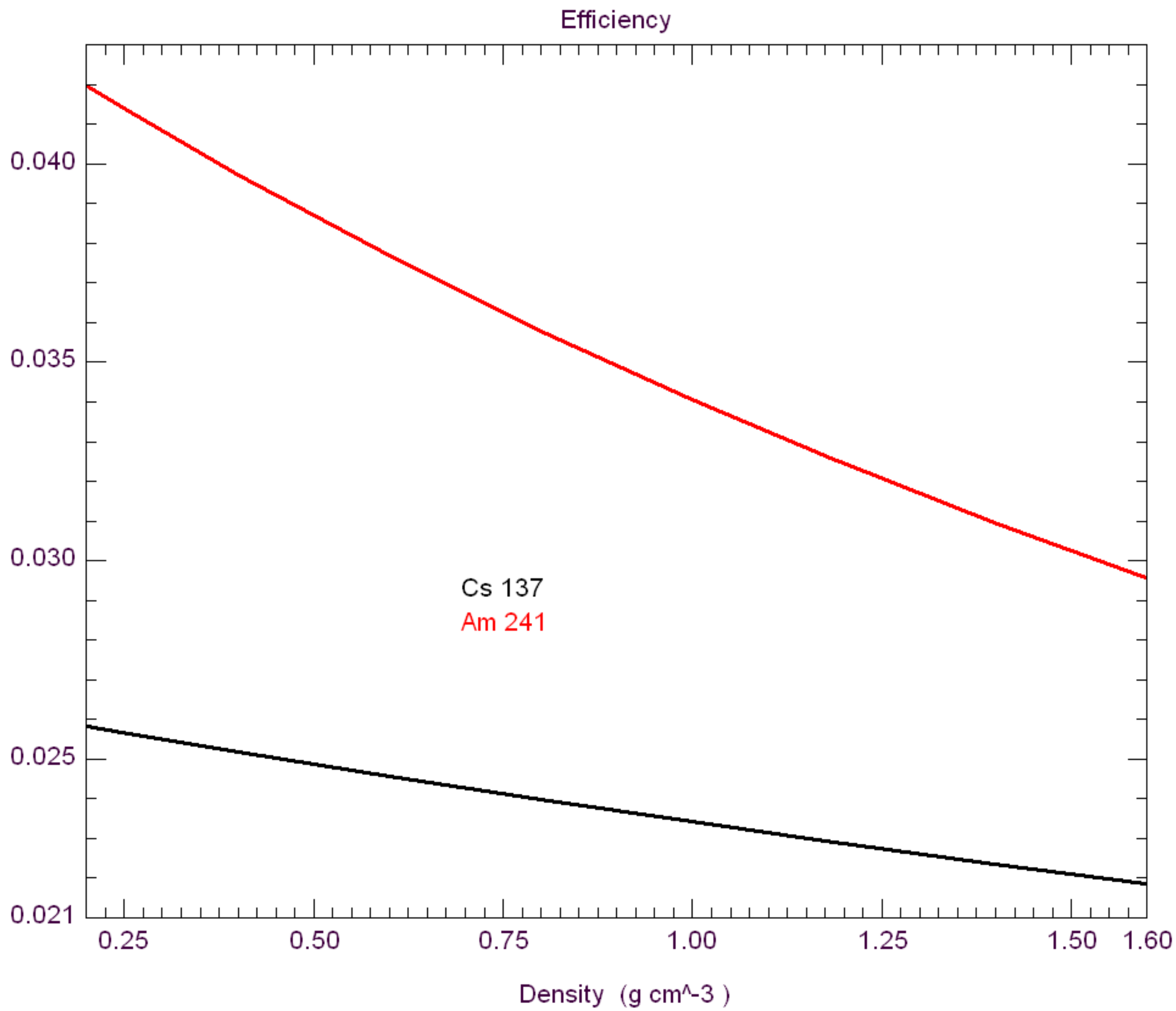
Iterative MCNP Efficiency Curve @ 1g/cc



MCNP Efficiency Curves – 0.2 to 1.6 g/cc



Efficiency Variation with Energy



Time to Develop Model

- Completely new detector design: 1 to 2 days
  - Assuming full details from vendor
- Modify existing model to change dead layer dimensions : 1 to ½ day
- Model new beaker, including source definition & verification: ¼ to ½ day
- Models can be parameterized to speed changing dimensions
  - %DLThick = 0.04 ; %OuterGeRadius = 3
  - Cylinder Defs: 1 CX %OuterGeRadius
  - 2 CX %OuterGeRadius - %DLThick

Time to Determine Actual Dead Layer

- Prepare MCNP source definition for peaks in standard
- Decay to measurement time
- MCNP calculation of spectrum
  - 10^8 histories (15 min w/ Marinelli beaker) sufficient
- Extract peaks & continuum, determine efficiency vs. energy
- Compare to measured efficiency curve, iterate as needed with different dead layer thickness
- For 10 peak standard & 6 iterations: 1 day

Run Times for 8 Densities & 10 Energies

- The total analysis time for the full set of efficiency curves over the range of expected densities: ½ day.
  - Includes preparing MCNP input files, MCNP runs (can do concurrently), extraction of results from output
- This makes the total time to develop and run the model starting with an uncharacterized crystal to be 2 to 4 days.
- Additional geometries for the characterized crystal can be done in 1 day.

Validation

- Existing food samples were spiked with standards of Am-241 and Cs-137. These isotopes represent the most difficult isotope for correction (Am-241) with a 60 keV gamma, and one of the most common high energy gamma emitters found in food products (Cs-137).
- The samples were counted on the characterized crystal and geometry (500 mL Marinelli) using the calibrated water efficiency curve (multiline standard).
- Results were corrected using the MCNP generated efficiency values for the observed food density and compared to the spiked values.

Validation - Rice Crackers

- An existing sample of rice crackers with a density of 0.245 g/cc was spiked with Cs-137 and Am-241 standards and was counted on the characterized crystal. Canberra Genie/Apex software was used.
- The values from the gamma analysis program were then corrected with the MCNP generated efficiency values at the lower density.
- The corrected results for Am-241were easily within the counting error.
- As expected, the Cs-137 values needed no correction across the full range of expected food densities for this geometry.

Validation – Rice Crackers

Isotope	Spike	Observed	Error (%)	Corrected	Error(%)
Am-241	0.12 pCi/g	0.15	25%	0.123	2.7%
Cs-137	0.12 pCi/g	0.114	5%	None	

Validation – Heavy Soy Sauce

- Soy sauce is one of the more common dense foods encountered at 1.18 g/cc.
- An existing sample of heavy soy sauce was spiked with Cs-137 and Am-241 standards and was counted in the Marinelli beaker geometry modeled.
- The results were then recorded and corrected using the MCNP generated efficiency values for the observed density.
- Again, the Cs-137 needed no correction.
- The error for the Am-241 result was essentially halved and was within counting error.

Validation- Heavy Soy Sauce

Isotope	Spike	Observed	Error (%)	Corrected	Error(%)
Am-241	0.12 pCi/g	0.107	10.8%	0.112	6.97%
Cs-137	0.12 pCi/g	0.113	5.8%	None	

Validation – Chaga Mushrooms

- Chaga mushrooms are a medicinal herb sold at health food stores. Density 1.054 g/cc
- The product is imported from Russia and contains about 0.541 pCi/g Cs-137. This is well below the DIL of 32 pCi/g.
- Mushrooms are a concentrator of Cesium isotopes.
- The sample was spiked with 0.457 pCi/g of Am-241 and was counted on the gamma spectrometer.
- The correction for the Am-241 was small due to the similar density to the water standard (reference), but did reduce the error.

Validation – Chaga Mushrooms

Isotope	Spike	Observed	Error (%)	Corrected	Error(%)
Am-241	0.457 pCi/g	0.425	7.5%	0.430	5.9%
Cs-137	None	0.41 pCi/g		None	

Validation: Syrup - Density 1.39 g/cc

Isotope	Spike	Observed	Error (%)	Corrected	Error(%)
Am-241	0.48 pCi/g	0.39	20%	0.43	12%
Cs-137	0.09 pCi/g	0.087	6.4%	0.09	2.2%

Validation: Ketchup – Density 1.106 g/cc

Isotope	Spike	Observed	Error (%)	Corrected	Error(%)
Am-241	0.61 pCi/g	0.50	17%	0.51	15%
Cs-137	0.111 pCi/g	0.111	6%	None	

Conclusion

- Monte Carlo derived food density correction values for a specific HPGe crystal and geometry, successfully corrected results to within counting error for all foods ranging from 0.24 to 1.6 g/cc.
- The model was developed quickly, and at low cost using an advanced visualization tool to speed the development of the MCNP input file.
- The unknown thickness of the dead layer was determined by comparison of the MCNP generated efficiency curve with an analytical curve from a water standard.

Conclusion

- The new method provides an inexpensive solution for food density corrections that is valid for the lifetime of the HPGe crystal.
- Requires a comparison of the MCNP generated efficiency curve with a measured standard at one density. This is normally required by most accreditation bodies.
- Can also be used to generate Peak to Total curves for True Coincidence Summing corrections.