LA-UR-04-7880

Identification of an Unknown Material in a Radiation Shield Using the Schwinger Inverse Method

Jeffrey A. Favorite
X-5, Applied Physics Division, Los Alamos National Laboratory
Los Alamos, NM, USA

Keith C. Bledsoe
Nuclear Engineering Program, Ohio State University
Columbus, OH, USA

American Nuclear Society Winter Meeting
Washington, DC, November 14–18, 2004

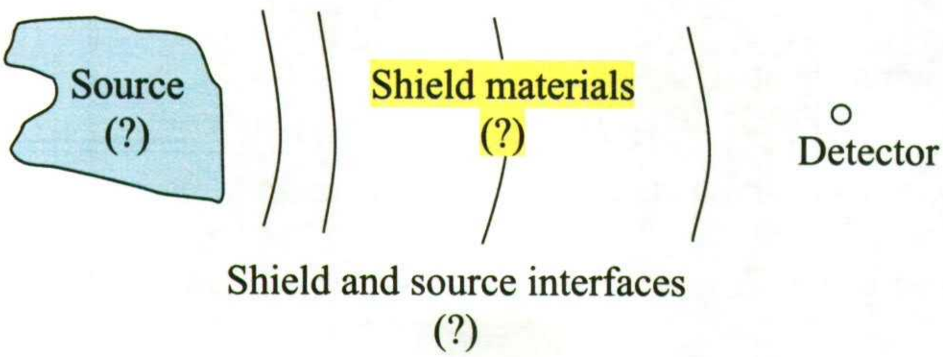
Abstract (not a viewgraph)

The Schwinger method for solving inverse gamma-ray transport problems was proposed in a previous paper. The method is iterative and requires a set of uncoupled forward and adjoint transport calculations in each iteration. In this paper, the Schwinger inverse method is applied to the problem of identifying an unknown material in a radiation shield by calculating its total macroscopic photon cross sections. The gamma source is known and the total (angle-independent) gamma leakage is measured. In numerical one-dimensional spherical and slab test problems, the Schwinger inverse method successfully calculated the photon cross sections of an unknown material. Material identification was successfully achieved by comparing the calculated cross sections with those in a precomputed material cross section library, although there was some ambiguity when realistic measurements were used. The Schwinger inverse method compared very favorably with the standard single energy transmission technique (SET).

These viewgraphs accompany a short summary, LA–UR–04–3968.

- Consider a radioactive object emitting γ rays of discrete energies that are well resolved using high-purity germanium (HPGe) detectors.

- We want to use γ leakage measurements to tell us what the system is.



- The Schwinger inverse method (Favorite, *Nucl. Sci. Eng.*, 2004) and the Newton-Raphson method (Favorite and Sanchez, ICRS-10/RPS2004) have been applied to the problem of determining internal interface locations.

- The Schwinger inverse method was applied to the problem of determining an unknown shield material, but the solution was not implemented numerically until now

- In this talk, we will:
  + Describe the Schwinger inverse method
  + Describe another inverse method for determining a composition, the single energy transmission technique (SET)
  + Demonstrate that the Schwinger inverse method converges to a set of cross sections
  + Demonstrate that the converged cross sections can be used to identify a material

- We consider only the transport of photons of discrete energies and assume that any scattered photons lose energy and are removed. The angular flux of photons at the discrete energy denoted by index g is given by

$$\hat{\mathbf{\Omega}} \cdot \vec{\nabla} \psi^{g}(r,\hat{\mathbf{\Omega}}) + \Sigma_t^g(r)\psi^g(r,\hat{\mathbf{\Omega}}) = q^g(r)$$

for $g = 1,\ldots,G$. (This equation represents the *forward problem.*)

- The adjoint equation is

$$-\hat{\mathbf{\Omega}} \cdot \vec{\nabla} \psi^{*g}(r,\hat{\mathbf{\Omega}}) + \Sigma_t^g(r)\psi^{*g}(r,\hat{\mathbf{\Omega}}) = q^{*g}(r),$$

where the source is to be defined (it will be the detector response function).

- These equations can be rendered in operator notation as

$$L^g \psi^g = q^g$$

and

$$L^{*g} \psi^{*g} = q^{*g}.$$

- Suppose the system leakage for each energy line g is measured at a detector. The quantity of interest is

$$\begin{aligned} M^g &= \int dV \int d\hat{\mathbf{\Omega}} \Sigma_d^g(r,\hat{\mathbf{\Omega}})\psi^g(r,\hat{\mathbf{\Omega}}) \\ &= \left\langle \Sigma_d^g \psi^g \right\rangle, \end{aligned}$$

where the detector response function $\Sigma_d^g(r,\hat{\mathbf{\Omega}})$ is

$$\Sigma_d^g(r,\hat{\mathbf{\Omega}}) \equiv \hat{\mathbf{\Omega}} \cdot \hat{\mathbf{n}}_d \delta(r - r_d),$$

where $\hat{\mathbf{n}}_d$ is the outward unit normal vector at point r on the surface $r = r_d$.

[• In this work, we use the approximation

$$\Sigma_d^g(r,\hat{\mathbf{\Omega}}) \equiv \begin{cases} A_d / V_d, & r_d - \delta r < r < r_d + \delta r \\ 0, & \text{otherwise,} \end{cases}$$

with $r_d = 100$ cm, $\delta r = 0.1$ cm, $A_d = 4\pi r_d^2$, and
$V_D = 4\pi[(r_D + \delta r)^3 - (r_D - \delta r)^3]/3$.]

- The perturbed quantity is the unperturbed quantity plus the perturbation; e.g., $\Sigma'^{g}_{t,n} = \Sigma^{g}_{t,n} + \Delta\Sigma^{g}_{t,n}$.

- The forward and adjoint equations for the perturbed case are

$$L'^{g}\psi'^{g} = q'^{g}$$

and

$$L'^{*g}\psi'^{*g} = \Sigma^{g}_{d},$$

respectively. The perturbed quantity of interest is

$$M'^{g} = \left\langle \Sigma^{g}_{d}\psi'^{g} \right\rangle.$$

(The leakage detector response is not perturbed.)

- A variational functional for M'^{g} is the Schwinger functional,

$$M'^{g}_{v}[\tilde{\psi}^{*g},\tilde{\psi}^{g}] = \frac{\left\langle \Sigma^{g}_{d}\tilde{\psi}^{g} \right\rangle \left\langle \tilde{\psi}^{*g} q'^{g} \right\rangle}{\left\langle \tilde{\psi}^{*g} L'^{g} \tilde{\psi}^{g} \right\rangle}$$
$$= \frac{\left\{\int dV \int d\hat{\Omega}\, \Sigma^{g}_{d}(r,\hat{\Omega})\tilde{\psi}^{g}(r,\hat{\Omega})\right\}\left\{\int dV \int d\hat{\Omega}\, \tilde{\psi}^{*g}(r,\hat{\Omega}) q'^{g}(r)\right\}}{\int dV \int d\hat{\Omega}\, \tilde{\psi}^{*g}(r,\hat{\Omega})\left[\hat{\Omega}\cdot\vec{\nabla}\tilde{\psi}^{g}(r,\hat{\Omega}) + \Sigma'^{g}_{t}(r)\tilde{\psi}^{g}(r,\hat{\Omega})\right]}.$$

- Using the unperturbed forward and adjoint fluxes as trial functions instead of the perturbed forward and adjoint fluxes reduces the Schwinger functional to

$$M'^{g}_{v} = M^{g}\frac{\int dV \int d\hat{\Omega}\, \psi^{*g}(r,\hat{\Omega}) q^{g}(r) + \int dV \int d\hat{\Omega}\, \psi^{*g}(r,\hat{\Omega}) \Delta q^{g}(r)}{\int dV \int d\hat{\Omega}\, \psi^{*g}(r,\hat{\Omega}) q^{g}(r) + \int dV \int d\hat{\Omega}\, \psi^{*g}(r,\hat{\Omega}) \Delta\Sigma^{g}_{t}(r)\psi^{g}(r,\hat{\Omega})}.$$

- In the Schwinger inverse method, we use a measurement for M'^g_S (call it M^g_0) and attempt to find $\Delta\Sigma^g_t$ and / or Δq^g:

$$M_0^g = M^g \frac{\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})q^g(r) + \int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\Delta q^g(r)}{\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})q^g(r) + \int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\Delta\Sigma_t^g(r)\psi^g(r,\hat{\Omega})}.$$

- Rearrange the above equation to obtain

$$\frac{1}{\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})q^g(r)}\left[\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\Delta\Sigma_t^g(r)\psi^g(r,\hat{\Omega}) - \frac{M^g}{M_0^g}\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\Delta q^g(r)\right] = \frac{M^g - M_0^g}{M_0^g}.$$

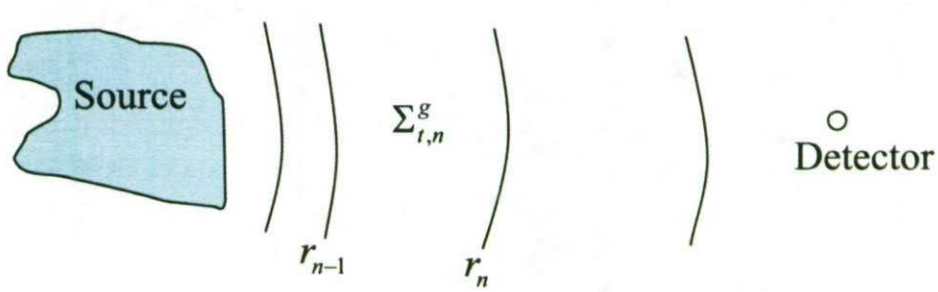
This equation is the essence of the Schwinger inverse method and was manipulated to solve a variety of inverse transport problems in *Nucl. Sci. Eng.*

- For the problem of an unknown shield material, $\Delta q^g(r) = 0$ and the second integral in brackets vanishes. The first integral in brackets is

$$\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\Delta\Sigma_t^g(r)\psi^g(r,\hat{\Omega}) = \sum_{n=1}^{N}\left(\Sigma'^g_{t,n} - \Sigma^g_{t,n}\right)\int_{r_{n-1}}^{r_n} dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\psi^g(r,\hat{\Omega}),$$

where

+ N is the number of unknown shield materials
+ $\Sigma'^g_{t,n}$ is the total cross section to be computed for region n, which is bounded by surfaces $r = r_{n-1}$ and $r = r_n$.



- The equation for $\Sigma'^{g}_{t,n}$ is

$$\sum_{n=1}^{N}\left(\Sigma'^{g}_{t,n}-\Sigma^{g}_{t,n}\right)\int_{r_{n-1}}^{r_n} dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\psi^{g}(r,\hat{\Omega})$$
$$= \int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})q^{g}(r)\left(\frac{M^{g}-M_{0}^{g}}{M_{0}^{g}}\right) \quad , \quad g=1,\ldots,G.$$

- There are G equations but $G \times N$ unknowns $\Sigma'^{g}_{t,n}$; thus, the equation cannot be solved unless there is only one unknown material in the shield.

- In this case, the equation becomes a set of G equations for the set of cross sections for region n,

$$\Sigma'^{g}_{t,n} = \Sigma^{g}_{t,n} + \frac{\int dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})q^{g}(r)}{\int_{r_{n-1}}^{r_n} dV \int d\hat{\Omega}\,\psi^{*g}(r,\hat{\Omega})\psi^{g}(r,\hat{\Omega})}\left(\frac{M^{g}-M_{0}^{g}}{M_{0}^{g}}\right) \quad , \quad g=1,\ldots,G.$$

- The G equations are completely uncoupled. Each of the cross sections will converge independently, at its own rate.

- To identify the unknown material, we compare the converged cross sections Σ'^{g}_{t} with those of 40 materials in a precomputed cross section library using the root mean square (RMS) for material m:

$$(\text{RMS})_m = \sqrt{\frac{1}{G}\sum_{g=1}^{G}\left(\Sigma'^{g}_{t}-\Sigma^{g}_{t,m}\right)^2}.$$

- A χ^2 norm would be more appropriate if there were estimates of the errors on the converged cross sections Σ'^{g}_{t}.

- The SET is a standard method of composition analysis. From the attenuation equation in a slab, $I_b^g = I_a^g e^{-\Sigma_t^g t}$, the total cross section is

$$\Sigma_t^g = \frac{1}{t} \ln \frac{I_a^g}{I_b^g}.$$

This equation is used to determine the composition of a two-component mixture.

- We compare the SET to the Schwinger method for a slab and a sphere. For the sphere, we use the approximation

$$\Sigma_t^g = \frac{1}{t} \ln \frac{J_a^g}{J_b^g},$$

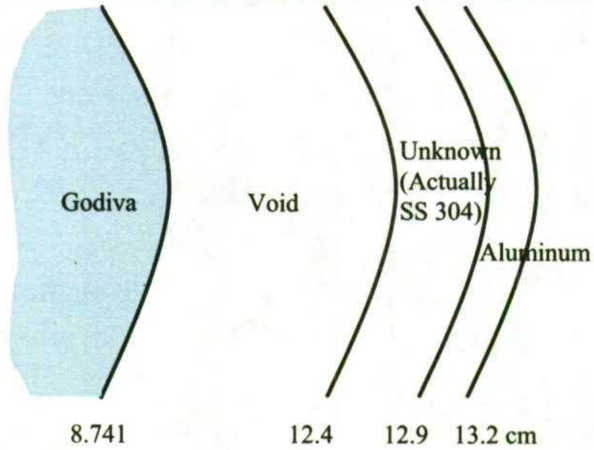
where J_a^g and J_b^g are the partial currents of gamma rays of energy g entering and exiting (respectively) the unknown material.

- In a multilayered slab, the monodirectional flux exiting the unknown region, I_b^g, can easily be reconstructed from the measured flux, M_0^g.

- In a multilayered sphere (with a source of finite radius), it is possible only to approximate the partial current exiting the unknown region, J_b^g.

  + We assume that there is a way of perfectly calculating the partial current J_b^g from the angle-integrated leakage measurement alone. This assumption is quite favorable to the SET.
  + In our implementation, J_b^g is calculated using the Schwinger-converged cross sections, guaranteeing consistency between J_b^g and M_0^g.
  + The partial current entering the unknown material, J_a^g, is, of course, available from a transport calculation of any model because there is no backscattering from the unknown material in this paper.

- Godiva model (spherical):

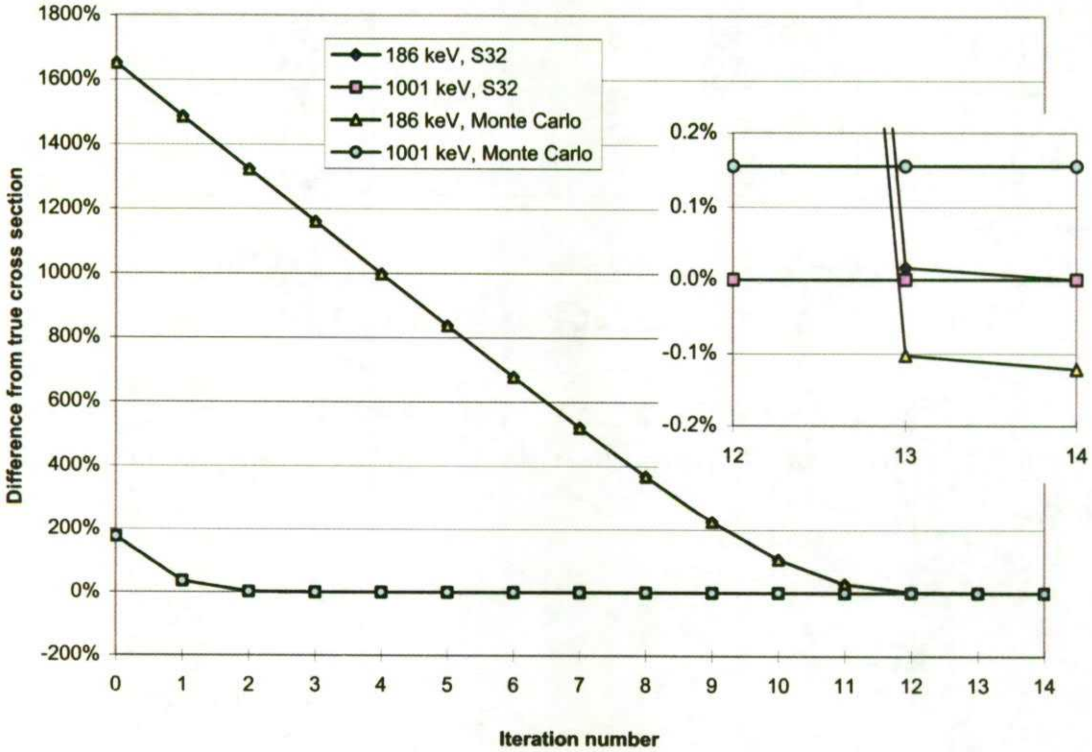


- The 186-, 766-, and 1001-keV uranium γ lines are used.

- Transport calculations used PARTISN, S_{32}.

- "Measurements" were obtained with PARTISN and MCNP.

- Initial guess was gold. Convergence of the 186- and 1001-keV lines:



- When S_{32} measurements were used, the three cross sections converged almost exactly to those for SS 304.
- When Monte Carlo measurements were used, the Schwinger method converged only to within ~0.2% of the true 186- and 1001-keV cross sections, and only to within 1.1% of the true 766-keV cross section.

- When S_{32} measurements were used, the RMS difference for SS 304 was 8.2×10^{-7}; the next smallest RMS difference was 1.9×10^{-3} for iron. The correct material was easily selected.

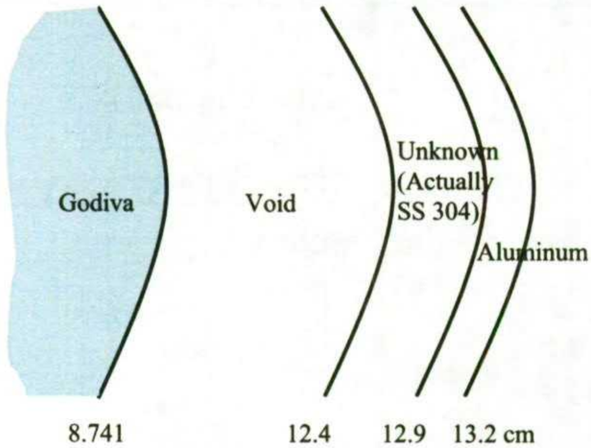
- When Monte Carlo measurements were used, there is no way to statistically tell the difference between SS 304 and iron:

	Material	Density (g/cm^3)	Problem 1 RMS Diff.
1	SS 304	7.86	3.597E–3
2	Iron	7.86	4.394E–3
3	Carbon steel	7.86	1.721E–2
4	SS 316	7.98	2.819E–2
5	Cobalt	8.90	1.102E–1
6	Copper	8.96	1.607E–1
7	Nickel	8.902	1.636E–1

- In the *Transactions*, only the 186- and 1001-keV lines were used, and iron had a smaller RMS than SS 304

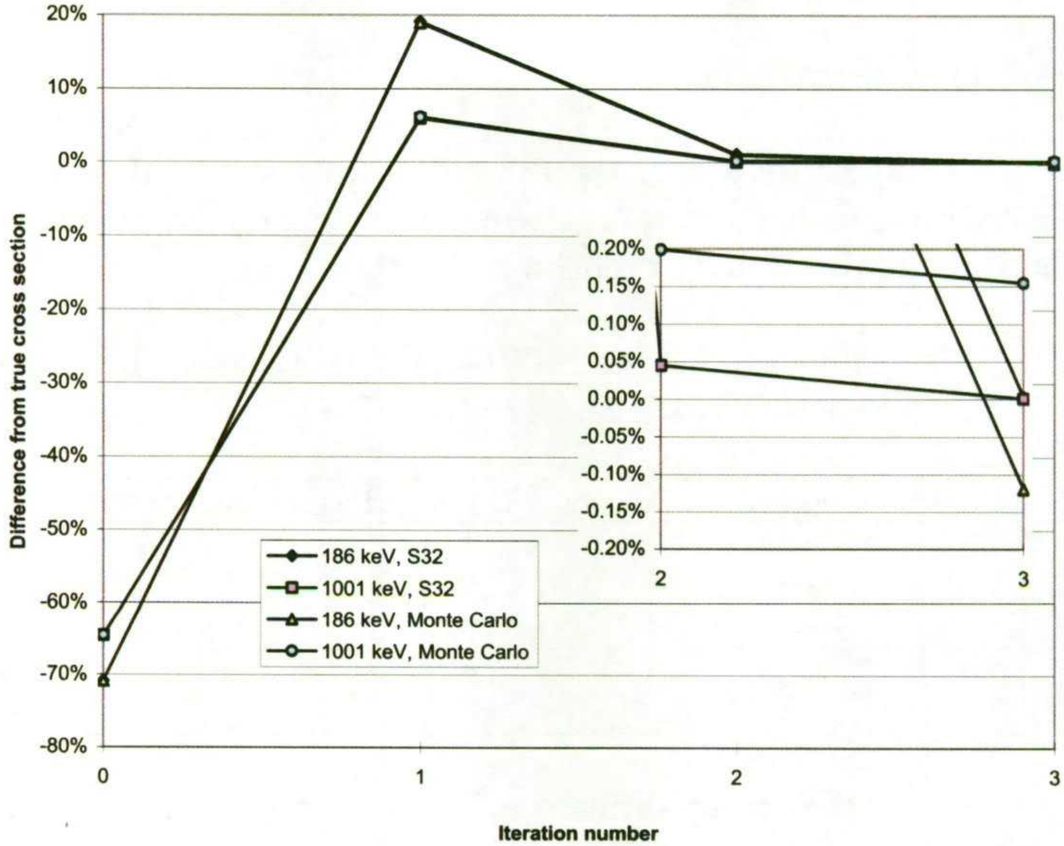
- Interestingly, cadmium, which is between SS 316 and nickel in density, has a much larger RMS difference (0.9537) than those materials for this problem.

- Godiva model (spherical):



- The 186-, 766-, and 1001-keV uranium γ lines are used.

- Initial guess was Al. Convergence of the 186- and 1001-keV lines:



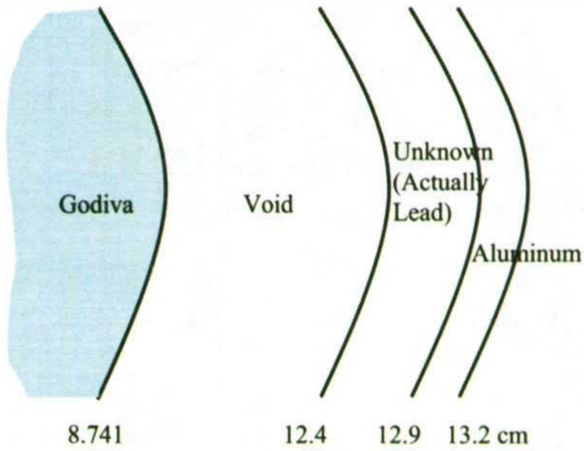
- The method converged to almost exactly the same cross sections as in problem 1, but much faster. Note:

Material	Density	Difference with SS 304
SS 304	7.86	
Al	2.70	–5.16
Au	19.3	11.44

Test problem 3 – Cross section convergence

- Godiva model (spherical):

- The 186-, 766-, and 1001-keV uranium γ lines are used.



- Initial guess was Al.

- No convergence!

  + A large difference in the 186-keV line caused a huge overcorrection in the new cross section, leading to zeros in the forward and adjoint fluxes and a problem with 0/0.

  + Using only the 766- and 1001-keV lines removes this problem.

- Note:

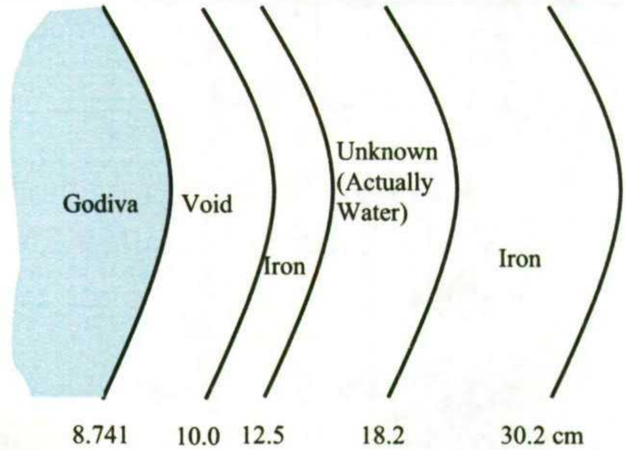
Problem	Material	Density	Difference with actual material
	SS 304	7.86	
1	Alum.	2.7	–5.16
2	Gold	19.3	11.44
	Lead	11.4	
3	Alum.	2.7	–8.7

- It is not just the density difference, but the direction that determines how the method will converge.

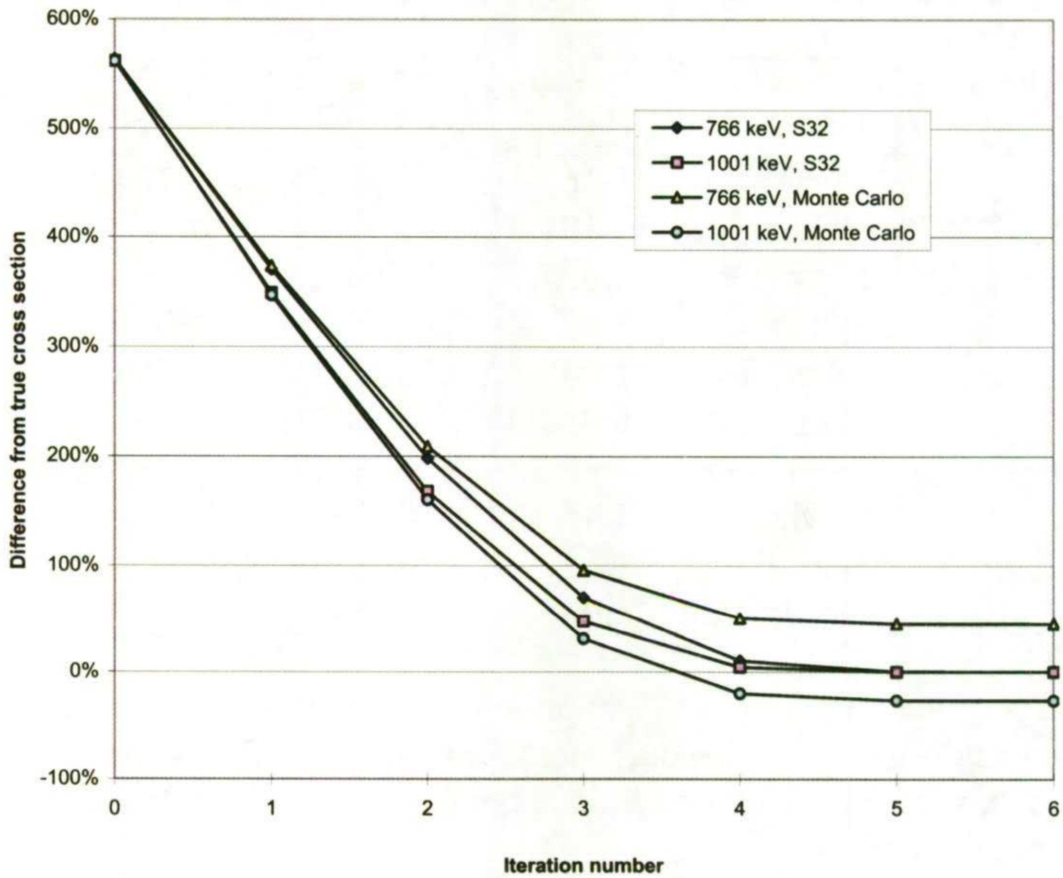
Test problem 4 – Cross section convergence

- Nuclear reactor shield with Godiva source (spherical):

- The 766- and 1001-keV uranium γ lines are used. The 186-keV line was used with S_{32} but not Monte Carlo measurements.



- Initial guess was iron. Convergence of the 766- and 1001-keV lines:



- When S_{32} measurements were used, the three cross sections converged almost exactly to those for water.
- When Monte Carlo measurements were used, the Schwinger method converged only to only to within 45% of the true 766-keV cross section and 27% of the true 1001-keV cross section

- S_{32} measurements:

  + For the Schwinger method, the RMS difference for water was 1.8×10^{-6}; the next smallest RMS difference was 2.2×10^{-3} for seawater.

  + For the SET, the smallest RMS was for rubber, 2.6×10^{-3}. The RMS for water was 8.2×10^{-3}; it was the fourth smallest.

- Monte Carlo measurements – The first 10 materials were the same for both methods:

Schwinger order	Material	Density (g/cm^3)	Schwinger RMS Diff.	SET RMS Diff.	SET order
1	Lucite	1.16	2.705E–2	3.057E–2	2
2	Rubber	1.10	2.714E–2	3.106E–2	4
3	Seawater	1.025	2.842E–2	3.369E–2	5
4	Water	1.00	2.896E–2	3.450E–2	6
5	Fiberglass	1.00	3.008E–2	3.605E–2	8
6	Wax	0.93	3.031E–2	3.635E–2	9
7	C-phenolic	1.40	3.066E–2	3.015E–2	1
8	Kevlar	1.45	3.184E–2	3.076E–2	3
9	Beryllium	1.85	3.741E–2	3.453E–2	7
10	Dry ground	1.90	5.303E–2	4.796E–2	10

- Under the very special assumption that partial currents can be obtained from an angle-integrated leakage measurement, the SET is about as good as the Schwinger inverse method for a spherical problem with large measurement error.

Summary and conclusions

- The Schwinger inverse method has been applied to the problem of material identification in a source / shield system.

- A material is identified by calculating its photon cross sections, then matching them with those in a precomputed table.

  + One issue that was addressed is whether the Schwinger method would converge to the correct cross section set. (It does.)

  + The second issue was whether a photon cross section set can be used to identify a material. (It can provide a reasonable range of candidate materials.)

- Some other method besides the Schwinger inverse method can be used to calculate the unknown cross sections: the SET, the Marquardt method, etc.

- The single energy transmission technique (SET) was compared with the Schwinger method. The SET was not designed for spheres.

  + It was assumed for the benefit of the SET that the exiting partial current can be calculated from the angle-integrated leakage measurement.

  + The SET was not very accurate when used with consistent S_{32} measurements.

  + The SET was about as accurate as the Schwinger method when used with inconsistent Monte Carlo measurements.

- We have assumed perfect knowledge of the source and geometry; we don't want to.

- We intend to extend this methodology to two-dimensional cylindrical systems.