Y/DW-1340

Y-12

**OAK RIDGE
Y-12
PLANT**

MARTIN MARIETTA

MERCURY DETECTION WITH THERMAL NEUTRONS

Z. W. Bell
Computing & Telecommunications Services Division

Date of Issue: September 19, 1994

Prepared by the
Oak Ridge Y-12 Plant
managed by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400

DS

MASTER

MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

875

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Table of Contents

Introduction	1
Neutron Source Design	3
HPGe Detector	10
Collimator Design	10
List of Procured Equipment	12
Cost to Restart and Complete	12
Remaining Tasks	13
References	18

List of Figures

Figure 1. Cup geometry.	14
Figure 2. Multiple cup geometry, conical ends.	14
Figure 3. Starting geometry for detailed study of scattering and solid angle.	15
Figure 4. Final configuration of moderator.	15
Figure 5. HPGe spectrum showing peaks due to Ge, Fe+ ²¹⁴ Pb, and Hg.	16
Figure 6. Collimator layers.	16
Figure 7. Geometry of collimator placement.	17
Figure 8. Geometry of moderator placement.	17

Final Report: Mercury Detection with Thermal Neutrons

Introduction

This report describes the work performed to design a gauge to detect mercury concealed within walls, floors, pipes, and equipment inside a building. The project arose out of a desire to decontaminate and decommission (D&D) a building in which mercury had been used as part of a chemical process. The building contains plumbing and equipment, some with residual mercury even after draining, sumps, and hollow walls. So that releases of mercury to the environment might be minimized during D&D activities, it was considered advisable to locate pockets of mercury that may have collected in concealed spaces so that they might be drained in a controlled fashion prior to the application of the wrecking ball or sledge hammer.

The detection of such pockets within a building presents some problems not ordinarily encountered in a laboratory environment. Often, only a single side of a wall or pipe is accessible. This condition disqualifies transmission gauges (such as conventional x radiography) in which a probe is sent through the volume under test (VUT) from one side and its passage or attenuation is detected on the opposite side. A robust, one-sided system was needed.

Because mercury is a high-Z metal with high density, gauges that relied on x rays were considered first. Gauges based on the detection of Compton-scattered photons and fluorescent x-rays were evaluated. A gauge based on Compton scattering can be used to infer the average electron density in a VUT. This is accomplished by collimating the source and the detector so that the intersection of their lines of sight defines a fixed (or at least calculable) volume. In principle the presence of a globule of mercury in an otherwise homogeneous matrix would be detectable if the VUT was not excessively large compared to the size of the globule. However, when the matrix consists of a non-uniform mixture of materials of widely differing Z and density (such as re-bar and/or copper conductors in concrete) such a gauge will have difficulty distinguishing between large moderately dense inclusions and small pockets of mercury. In addition, such a gauge is hampered by the relatively low cross section for Compton scattering (versus photoelectric absorption) at the energies required to penetrate concrete walls or steel pipe.

A gauge based on K x-ray resonance fluorescence (KXRF) was then considered. This device would operate by irradiating a VUT with x rays from an x ray tube with energy above the K edge in mercury (83.1 keV). Absorption of these x rays results in the ejection of K shell electrons and would leave the atom in a highly excited state. The mercury atom then would deexcite by the emission of its characteristic x rays, which would be detected. The highest energy emissions in mercury are due to L shell to K shell electronic transitions which result in the emission of 70 keV x rays. Although these x rays can penetrate a few centimeters of concrete, they are almost completely absorbed by 6 mm of steel. In addition, because the detector would be required to be at an extreme backward angle with respect to the direction of propagation of the probing x ray beam, the gauge would not have a useful depth of field. Furthermore, because it would be necessary for the continuous radiation scattered by the VUT to have energy below that of the mercury K x rays, the maximum energy of the probing energy would be limited to about 90 keV. At this energy, significant absorption of the incident beam would decrease noticeably the x ray flux available to induce fluorescence at the VUT.

Gauges using ultrasonic probes were rejected because concrete, being an aggregate material, would present an inhospitable medium through which to propagate sound. Such a gauge would rely on detecting reflections of sound from pockets of mercury and would be confused by reflections from the normal inclusions found in concrete. In addition, layers of paint over walls and pipes attenuate the sound and decrease the maximum depth that can be probed.

Chemical "sniffers" were rejected for two reasons. First, painted walls and sealed pipes present effective barriers to the rapid migration of mercury vapor. Thus, mercury contained in pipes and equipment or behind painted walls is invisible to a sniffer. Second, high levels of mercury vapor already present in the air in the building were expected to make it extremely difficult to locate a pocket by following a plume of vapor.

A perusal of the literature on the nuclear physics of mercury resulted in the discovery that natural mercury possesses a thermal neutron capture cross section 100 times greater (384 barns as opposed to 1 to 5 barns) than most structural materials. This cross section is due almost entirely to a single isotope, ^{199}Hg , which comprises 17% of natural mercury. It was also found that in 77% of captures of neutrons by this isotope, a gamma ray with energy 368 keV is emitted. This gamma ray is attenuated by only 40% by passage through 0.25 inch wall steel pipe and its range is several centimeters in concrete. Because thermal neutrons also can penetrate several centimeters of steel and concrete, it seemed reasonable to investigate the development of detector based on this principle.

A feasibility study was performed at the start of the project and published¹. During this study, the scientific literature concerning the interaction of mercury and structural materials with neutrons was examined. It was found that only one naturally radioactive material, ^{214}Pb (a decay product of ^{238}U and ^{226}Ra) and that on irradiation only one stable element, iron, produce gamma rays near the energy of the mercury gamma ray. The proximity of these gamma rays necessitated the use of a high purity germanium (HPGe) detector, rather than sodium iodide, for spectroscopy.

For a flux of neutrons, ϕ , irradiating a mass of mercury, M , the rate, r , of captures is given by

$$r = \frac{\phi \cdot \sigma \cdot M \cdot N_A}{W} \quad (1)$$

where N_A is Avogadro's number, σ is the capture cross section, and W is the atomic weight of mercury. Thus from a measurement of r , and a knowledge of ϕ , M can be inferred by inverting equation 1.

Experiments were also performed to demonstrate this principle of operation². Mercury-containing batteries were placed inside a steel pot (0.25 inch walls) and irradiated with a moderated Am-Li neutron source. Spectra which clearly showed the mercury gamma ray in the presence of those from ^{214}Pb (a contaminant in the detector shielding) and iron were obtained with an HPGe detector shielded with lead bricks, polyethylene, and cadmium. It was estimated that mercury in the presence of iron is detectable at the level of 460 ppm. It can be inferred from this work that given the background and source (25,000 neutrons/sec isotropically escaping the moderator) in this experiment, in the absence of significant absorption, approximately 13 grams of mercury can be detected in 800 seconds of counting. The source designed for the actual gauge would be expected to lower the limit of detection to 3 grams in 800 seconds. The reader is referred to the report for more details of the experiment.

In the remaining sections of this report, the reader will find descriptions of the considerations going into the design of the neutron source, the detector, and the detector's collimator. In addition, an inventory of equipment already procured, and an estimate of the cost to complete the construction of the gauge is given. A list of tasks necessary for the fielding of the gauge will also be found.

Neutron Source Design

The neutron source selected for this gauge is an Am-Li source containing 10 curies of ^{241}Am and producing approximately 500,000 neutrons/sec. This source was in storage at the Y-12 plant when the project was started; its identification saved the approximately \$12,000 cost of a new source.

A radioactive source (such as this one) produces an omnidirectional (and nearly isotropic) neutron fluence. In addition, the spectrum of neutrons is continuous with a maximum energy of approximately 1.5 MeV and an average energy of 340 keV. Consequently, for efficient sensing of mercury, it is necessary to slow down (moderate) the neutrons to thermal energies (0.025 eV) while simultaneously directing as great a number toward the VUT. To this end, a moderator composed of plastic was designed with the MCNP³ computer code.

MCNP is a Monte Carlo simulation code that solves neutron (and photon) transport problems by tracking hundreds of thousands (or more) particles through a user specified geometry. The usual quantities of interest are numbers of particles crossing critical surfaces or entering volumes, the distribution of velocities or directions relative to critical surfaces, and the energy deposition by particles in critical volumes. The results are expressed as the probability per source particle of the value (or range of values) of the quantity of interest.

The initial designs centered around an existing 50,000 neutron/second (n/s) source contained in a 6 inch diameter by 8 inch high cylindrical polyethylene (PE) moderator surrounded by a 0.25 inch thick steel shell. This moderator had channels cut into it to accommodate a detector, cables and a sample for irradiation. Simulations, however, were performed for a solid cylinder containing the 0.75 inch diameter by 0.75 inch high source capsule. The results indicated that approximately half of the source neutrons were captured by the hydrogen in the PE and that the remainder escaped. The surfaces of the moderator were essentially Lambertian, with a slight decrease in brightness toward the intersection of the end caps with the cylinder wall. In addition, a significant portion of the escaping neutrons were not thermalized. Hence, it was necessary to increase the amount of moderator material.

The large numbers of captured neutrons led to simulations, with carbon replacing much of the PE, in which the source was placed inside a cylinder 18 inches in diameter by 11.25 inches high. Carbon was chosen because it has a very low capture cross section, moderate scattering cross section and low atomic weight. With the moderator composed entirely of carbon, only 3% of the neutrons were captured (mostly by the steel shell), but there was almost no degradation of neutron energy, and no directionality was imparted to the escaping neutrons.

Simulations were then run for geometries in which the moderator was constructed of 5 layers of PE and carbon. The top and bottom layers were always carbon, and the remaining layers were either PE or carbon. The slab containing the source was split into an inner and outer ring whose compositions could be set to PE or carbon independently. It was found, as expected, that the addition of PE was crucial to the moderating properties and that 7 - 10 cm of PE effectively blocked neutrons from

exiting the assembly through the PE. This observation led to the investigation of "cup" geometries, as shown in Figure 1.

Cup geometries are arrangements of concentric cups of moderator material surrounding the source. Alternating cups of carbon and PE nested up to 4 levels were placed around a source. The source was completely enclosed in a PE cylinder, the thicknesses of whose top and bottom (dimensions A and B, respectively, in Figure 1) were varied independently between 1 and 2 inches. Typically, these arrangements resulted in 50% to 66% of the source neutrons escaping the container.

A single PE cup (0.5 inch wall) was found to enhance the emission of neutrons out the open bottom of the cup by approximately 50% over the emissions either out the closed top or round wall. This result was dependent, however, on the amount of PE in direct contact with the source: It was found that thinning the top (dimension A in Figure 1) doubled this enhancement by decreasing the number of captured neutrons. These "uncaptured" neutrons were then free to scatter from the closed ends of the carbon and PE cups and then meander toward the open ends. Replacement of the PE with beryllium was found not to affect the directionality of the moderator while instead significantly hardening the energy spectrum.

The use of two concentric PE/carbon cups was found to affect the amount of leakage of neutrons out the round side or closed end (total of 18%) rather than the numbers exiting the open end (still 40%). This is attributable to the increased mass of PE between the source and those outside surfaces. The addition of two more PE cups (for a total of four) resulted in a 10% loss of desired neutrons compared with the two cup case and a 40% loss of leakage neutrons. This geometry resulted in emissions of 36% through the bottom, 3% through the side, and 8% through the top.

Simulations were then run with cups that had conical or spherical closed ends, such as in Figure 2. No significant differences were observed between these two geometries. However, the leakages through the top and side for these shaped 2-cup geometries were identical to that for 4-cup geometries, while the flux of neutrons out the bottom was that of the flat 2-cup geometries.

When all the results so far described were considered as a whole, it was noticed that the 40% of source neutrons exiting the moderator out the open end of the cups was consistent with what might be expected by consideration of the solid angle subtended by that face with respect to the source. Therefore a systematic study was performed to distinguish the effects of geometry from those of reflector material.

The solid angle with respect to a point source subtended by a surface can be calculated by the Monte Carlo method simply by counting the number of simulated particles emitted by the source that cross the surface. An MCNP input file was prepared that described a point source in the center of a PE cylinder which, in turn, was within a 44 cm diameter empty cylinder. Neutrons crossing the flat bottom of this empty cylinder were counted and compared with the calculated solid angle subtended by it. For the purposes of the solid angle calculation, the bottom of the cylinder and the point source define a cone.

The solid angle subtended by the open end of radius r of a cone of height y is given by

$$\Omega = 2\pi \left(1 - \frac{y}{\sqrt{y^2 + r^2}}\right) \quad (2)$$

The fraction, f , of neutrons emitted by a point source at the vertex of the cone that exit through the open end is then given by

$$f = \frac{\Omega}{4\pi} = \frac{1 - \frac{y}{\sqrt{y^2 + r^2}}}{2} \quad (3)$$

The geometry used for this calculation is shown in Figure 3. The solid angle was computed for a cone of height $1 + X$ and base radius of 22 cm.

The results of the MCNP runs are summarized in table 1.

Table 1. Neutron tally (effective solid angle) v. actual solid angle.

X	TALLY	$\Omega/4\pi$
1	0.4435	0.4547
2	0.4127	0.4324
3	0.3810	0.4106
4	0.3280	0.3892
5	0.2680	0.3684

It is apparent from the table that as more PE is added to the moderator, more neutrons are absorbed or scattered away from the bottom of the structure than are being scattered into the desired direction (down). To construct a more efficient moderator, it is necessary to either decrease the amount of PE in the structure at the expense of a harder energy spectrum, or to add reflectors to direct stray neutrons toward the bottom. The addition of reflector material was the original logic behind the cup designs described above.

It was determined from the MCNP runs that 5 cm of PE was the minimum necessary to be interposed in the line of sight between the outside world and the source. This thickness guarantees a 70% probability that a neutron scatters at least once in the moderator. Further calculations were run with $X = 5$ cm.

A slab of PE (thickness T) was now added to the top of the PE cylinder and runs were made for T between 1 and 3 cm. The reflector was placed in contact with the top surface of the PE cylinder and had a radius of 22 cm.

Table 2. Effects of different thickness of top reflector

T	TALLY
1	0.3065
2	0.3244
3	0.3336

It is seen that even the addition of 1 cm of reflector dramatically increases the fluence in the desired direction (0.2680 v. 0.3065). It is also seen that each additional centimeter of reflector does not proportionately increase the fluence. This is attributed to the fact that although a fast neutron might penetrate a large distance into the reflector and then be scattered back toward the bottom, the extra PE effectively shields the bottom of the structure from these neutrons. Two centimeters of reflector was considered adequate.

The effect of an empty channel between the top reflector and the cylinder was considered next. There are two ways to make this channel: The reflector may be backed away ("raising the roof") or the top of the cylinder may be thinned. In the tables below, H is the width of the empty channel.

Table 3. Effect of thinning the top of the PE cylinder.

H	TALLY
1	0.3288
2	0.3292
3	0.3329
4	0.3318

Table 4. Effect of "raising the roof."

H	TALLY
1	0.3191
2	0.3150
3	0.3088
4	0.3040
5	0.3017

In all instances it is seen that thinning the top of the cylinder produces better results than raising the roof. This occurs because the further the roof is from the bottom surface of the assembly, the smaller

is the solid angle subtended by any area on the bottom surface with respect to any point on the reflector. However, thinning also results of a harder neutron spectrum.

Since PE contains hydrogen which has a small, but non-negligible capture cross section, the effect of substituting carbon for PE was investigated. Both a carbon reflector and a single carbon cup were examined.

Table 5. Effects of different thickness of carbon top reflector (no channel).

T	TALLY
2	0.3041
3	0.3145
4	0.3219

Table 6. Effects of different thickness of carbon cup.

T	TALLY
2	0.3452
3	0.3730
4	0.3952

It is seen from Table 5 that carbon is inferior to PE as a reflector material. This is because although carbon has essentially no capture cross section, its scattering cross section is a factor of two smaller than hydrogen's. Thus the neutrons saved from capture are outweighed by those that simply escape the system.

The carbon cup, on the other hand provides a 20% improvement over the PE reflector results (Table 3). In this case some neutrons that would otherwise have simply escaped out the sides have been scattered downward in the desired direction. Since PE would be expected to be a better reflecting material, it too was tried as a cup material.

Table 7. Effects of different thickness of PE cup.

T	TALLY
2	0.4160
3	0.4465
4	0.4571

Use of PE offers a 15% improvement over carbon. Note that these results are comparable to those obtained for the concentric cup geometry. This fact that should not be entirely unexpected since the

amounts of PE in both problems are similar. Thus, the number of channels is not as important as the probability of scattering.

The effect of providing a channel between the PE cylinder and the cup was now investigated. The channel was created by raising the roof.

Table 8. Effect of "raising the roof" and a 4 cm PE cup

H	TALLY
0	0.4571
1	0.4608
2	0.4567
3	0.4572
4	0.4545
5	0.4495

The row for H=0 is simply the result for T=4 in Table 7. Apparently, there is an optimum channel width of approximately 1 cm. Beyond that, the effects of decreased solid angle overcome any possible improvements due to the increased channel width. Calculations were also performed for geometries that included a 1 cm thick carbon liner inside a 3 cm thick PE cup, and various arrangements of cups placed within the cup and liner. None of the results were significantly different than that shown in the line for H=1 in table 8 above.

All the calculations so far described were based on an existing source contained in a stainless steel can 1.5 inches in diameter by 1.5 inches high. When the source to be used for this project was located and its drawings examined, it was found that the actual dimensions were 1.25 inches diameter by 4.25 inches high. The actual volume occupied by the source material was 1 inch in diameter by 2.925 inches high. Therefore it had been inappropriate to treat the source as a point; rather, a volumetric source should have been used. In addition, the source container was itself held in a tungsten shield (because of the 60 keV ^{241}Am gamma rays). The larger dimensions of the actual source also meant that it would be more advantageous to rotate the source by 90° so that it lay on its curved side.

When all these new developments were taken into account, the numbers of neutrons exiting the bottom of the assembly was found to have been cut in half. This was determined to have occurred because of the increase in the amount of PE surrounding the source and because of the orientation of reflecting surfaces with respect to the exit surface. Part of the effects of the former could be ameliorated by removing the part or all of the back (with respect to the exit surface) of the of the central PE cylinder.

It had been observed during the previous calculations that neutrons exiting a flat surface of a moderator had a distribution of directions varying approximately as the cosine of the angle with respect to the surface normal. This behavior is explained by noting that the distance from a point within a moderator to the surface increases according to the cosecant (1/cosine) of the angle between the normal to the surface and the velocity vector and that the probability of a neutron escaping

decreases as the negative exponential of the distance to the surface. Thus, the velocity vectors of emerging neutrons are weighted toward the normal to the surface. Hence, to increase the neutron fluence out the bottom surface of the moderator assembly, it is necessary to have reflector surfaces facing the bottom surface. This scheme suggests erecting a tent over the source as shown in figure 4.

A systematic analysis of the effects of various dimensions of the assembly was performed. It was found that by optimizing the width of the feet of the moderator (at 6 cm), and the height of the moderator above the source, an efficiency of 0.3358 could be achieved. That is, 33.58% of the source neutrons could be expected to exit the moderator out the bottom and that over 70% of those would be thermalized.

MCNP provides tallies of the numbers of neutrons lost to capture sorted both by location of the capture event and by the element effecting the capture. It was found that 63.34% of all source neutrons were captured in the moderator. Of those, hydrogen accounted for 42% of all source neutrons. This was not surprising considering the amount of PE in the system. It was also found that the tungsten shield accounted for 20% of all source neutrons. This was a surprise because the shield is not that large (only about 1 cm thick walls). However, a little prior research would have shown that tungsten has a capture cross section of 20 barns. This implies that the mean free path of thermal neutrons is approximately 0.8 cm, meaning that significant numbers of thermal neutrons entering the shield are captured. A calculation run with a lead shield found that the yield was boosted to 40% (from 33.6%). Hydrogen in the PE accounted for remaining losses. Thus, as an aside, one should not shield a neutron source with tungsten.

The realities of construction of the tent moderator were now tackled. PE is not the easiest of materials to machine into complex shapes. For this reason, it was suggested by engineering staff to examine the possibility of substituting polymethylmethacrylate (PMMA) plastic (Plexiglas) or polycarbonate (LEXAN). When the entire moderator was made of these substitutes, it was found that although the total number of neutrons exiting the bottom was 20% higher than in case of PE, the number of thermals was not significantly different. In addition, the number of neutrons escaping the system in undesirable directions was 3 to 6 times larger for PMMA and LEXAN, respectively. This occurs because the ratio of hydrogen to the sum of carbon plus oxygen atoms in LEXAN is lower than it is for PMMA, which in turn has a lower ratio than does PE.

Tent geometries were then run with PE, PMMA, and LEXAN paired as the cradle and reflector. The following results were obtained:

Table 9. Effects of plastic combinations

Reflector → Cradle ↓	LEXAN	PMMA	PE
LEXAN	4287 (2229)	4161 (2167)	4051 (2185)
PMMA		3957 (2305)	3906 (2344)
PE			3358 (2242)

Materials were never paired such that the H:(C+O) ratio of the reflector was lower than that of the cradle. The numbers in parentheses are the numbers of thermals ($E < 10$ eV) escaping through the bottom of the moderator. In all cases, a 0.020 inch thick layer of cadmium was placed around the outside surface of the reflector. It is seen that the PE-PMMA combination produces the largest fluence and it was found that this combination did not produce significantly more escapes out the other sides of the moderator. Consequently the final design called for this configuration and a set of drawings, M2E-920300-A263 through M2E-920300-A268 were issued.

HPGe Detector

Because of the nearness in energy of the decay gamma rays from ^{214}Pb , and the capture gamma rays from iron to that of mercury, it was necessary to give serious consideration to the detector. Sodium iodide (NaI) offers the advantages of high efficiency and being relatively inexpensive. However, the energy resolution of even small crystals is insufficient to resolve the gamma rays of interest. HPGe, on the other hand, is considerably more expensive but resolves even the 366 keV iron gamma ray from the 368 keV mercury gamma ray.

In order to decide if NaI could be used for the subject application, simulated data smeared by the resolution function of a typical detector (7%) and corrupted by additive Poisson noise was analyzed by a sum-of-gaussians non-linear curve fitting program⁴. The results indicated that it would be impossible to distinguish the peaks at 352, 366 and 368 keV with any degree of certainty. This was not wholly unexpected because the most widely spaced peaks differ in energy by only 4%.

Commercially available HPGe detectors typically have energy resolution between 1 and 2 keV (about 0.3% - 0.6%) at 368 keV. Thus, there is no question that these detectors can resolve the relevant gamma rays. Consequently, an ORTEC HPGe detector was obtained with a Ge crystal 70 mm in diameter and 78 mm deep. This detector was quoted to be 67% as efficient as a 76 mm by 76 mm NaI detector at 1.33 MeV. At 368 keV, the efficiencies are comparable.

The detector's energy resolution was tested by irradiating with a neutron source mercury-containing batteries held within a steel pot. The resolution as a function of amplifier shaping (signal integration) time was measured by acquiring pulse height spectra for shaping times from 1 s to 8 s. The optimum value of 0.41 keV was determined to occur with 3 s shaping time.

Figure 5 shows a typical spectrum obtained after 800 seconds of counting. The leftmost peak is caused by neutrons captured by Ge in the detector crystal itself and is useful as a flux monitor. The middle peak is caused by the 352.4 keV iron capture gamma ray and the 351.9 keV gamma ray from ^{214}Pb present in the shielding material. The rightmost peak results from mercury capture gamma rays at 368.1 keV. The background on which all these peaks sit is caused primarily by the registration of gamma rays that undergo Compton scattering in the HPGe detector and then escape the Ge crystal.

Collimator Design

HPGe detectors are omnidirectional; they do not distinguish between photons entering the crystal from different directions. Consequently, it is necessary to place a collimator around the detector so that it becomes sensitive only to a particular direction. A passive collimator was chosen for this application because of its simplicity.

The original design called for a graded cylindrical collimator with walls containing 2 inches of tungsten and sleeves of 0.020 inch tin and copper as shown in Figure 6. The tungsten's job is to attenuate high energy photons in order to remove as much Compton continuum as possible. This improves the signal-to-noise ratio and decreases the time required to make a decision concerning the presence of mercury.

High energy photons entering the tungsten are absorbed via photoelectron production and pair production. The latter results in the generation of 511 keV photons which are absorbed by photoelectrons. Photoelectron production results in the emission of 70 keV x rays characteristic of tungsten. These x rays have only a short range in tungsten and are absorbed by the tin layer which converts them into tin x rays with a peak energy of about 29 keV. The tin x rays are in turn absorbed by the copper layer which then emits its own characteristic x rays with maximum energy of about 9 keV. In this way, high energy photons are converted into photons whose energies are far below the detection capabilities of the HPGe crystal.

Because the detector was to operate in a neutron field, and neutrons damage HPGe crystals, the entire collimator was to be surrounded by a cadmium sheath around the tungsten. Neutrons stopped in the cadmium produce a 570 keV photon which is easily absorbed by the rest of the collimator.

Cadmium, however, is inappropriate over the front of the collimator because this capture gamma ray would be seen by the detector. Therefore, a disc of ${}^6\text{LiF}$ bound with epoxy was designed to fit over the opening of the collimator. A 5 mm thick layer of ${}^6\text{LiF}$ absorbs all thermal neutrons and emits no photons.

Although tungsten was considered to be the material of choice for this collimator, its cost was determined to be too high to be practical. The HPGe crystal is 3.5 inches in diameter and 3.1 inches in length. For the collimator to be reasonably directional, it would need to be 9 inches in length, thus requiring approximately 340 pounds of tungsten. With the cost of tungsten between \$30 and \$50 per pound, this collimator would be prohibitively expensive. A lead collimator, using 4 inch walls, and weighing about the same, was quoted to cost only \$2000. Since all other considerations would be unchanged, it was decided to substitute lead for the tungsten.

The placement of the collimator relative to the neutron source determines the field of view of the system. Schematically, this is shown in Figure 7. The basic problem is to determine the position, (x,y) , of the crystal, the orientation, ψ , of the collimator, and the offset, h , of the detector from the front of the collimator, given the half-width of the moderator, W , the inner and outer radii of the collimator, and the desired depth of view relative to the moderator.

The geometry shown in Figure 7 implies 5 conditions on the various parameters:

1. $X_1 \leq -W$, the collimator does not interfere with the source.
2. $Y_2 \geq 0$, the collimator is not in front of the source.
3. $X_3 \leq -W$, the source does not block the field of view
4. $Y_4 \geq -L_1$, the field of view covers the desired region.
5. $Y_5 \leq -L_2$, the field of view covers the desired region.

These conditions lead to a set of non-linear inequalities that define regions in (h, ψ, x, y) space.

$$\begin{aligned}
 R\cos\psi + h\sin\psi + X + W &\leq 0 \\
 -R\sin\psi - h\cos\psi + Y &\geq 0 \\
 X + Y\tan(\psi + \theta) + W &\leq 0 \\
 X\cot(\psi + \theta) + Y + L_1 &\geq 0 \\
 X\cot(\psi - \theta) + Y + L_2 &\leq 0
 \end{aligned}
 \tag{4}$$

A simple solution can be obtained by abutting the collimator and moderator. Then, selecting ψ such that the line through $(0, -L_1)$ and $(-W, 0)$ pass through the center of the rear of the crystal, determines a minimum condition on h .

Because the open face of the moderator is approximately 18 inches square, and it is desired to detect puddles of mercury at floor level (9 inches below the centerline of the moderator), the moderator must be angled downward as shown in Figure 8, and L_1 should be 10.7 inches. Since concrete blocks are typically 8 inches in depth, with 1 inch thick walls, L_2 needs to be at approximately 17.7 inches. Given the inner and outer radii of the collimator of 1.75 and 6 inches, the setback, h , must be at least 5 inches for all conditions to be satisfied. Since the crystal is approximately 3 inches deep, placing it so that the front is 3 inches from the front face of the collimator satisfies the condition on h .

List of Procured Equipment

The following items were procured for this project:

- 1 ORTEC HPGe detector, GEM50195P, SN 34-TP40465A.
- 1 ORTEC NOMAD portable detector electronics package, SN 140.
- 1 ORTEC 3.0 liter dewar, DWR-3.0G.
- 1 set of moderator drawings, M2E-920300-A0263 through M2E-920300-A0268.

The ORTEC equipment has been tested and presently resides in Building 9203. A total of 9 sets of the drawings have been distributed.

Cost to Restart and Complete

All development work on this project has been halted. Neither the collimator nor the moderator have been assembled. An Am-Li neutron source, presently in storage, has been identified, but has not been claimed. It will remain in the custody of health physics personnel. Should the source be claimed for some other project, its loss will result in a cost of between \$10,000 and \$15,000 for a replacement. An additional \$20,000 may need to be allocated for redesign of the moderator, should a new source not have the same physical characteristics (such as dimensions, spectrum, component materials).

Should the project be restarted the following costs (FY94 dollars) might be expected:

Moderator construction	\$25,000
Collimator construction	2,000
^6LiF disc	1,200
Labor	250,000
Total	\$278,200

These costs are to complete the project and are independent of the costs associated with the possible purchase of a new source and/or redesign of the moderator.

Remaining Tasks

The following activities have been completed for this project:

- Feasibility study for mercury detection with neutrons, including literature survey.
- Estimates of the limit of detection have been obtained.
- A report describing feasibility study has been issued.
- The equipment required to fabricate the detector system has been identified.
- The detector and associated electronics have been procured.
- A neutron source has been identified (but not claimed because of the suspension of the project).
- The source moderator has been designed.
- The detector collimator has been designed.
- Proof of principle testing has been performed and reported (see reference 2).

The following activities have not been completed:

- Preparation of test plan for bench scale evaluations.
- Bench scale evaluation with actual source, moderator, detector, electronics, and collimator.
- Document bench scale evaluation.
- Identify changes and modify system for field application/demonstration.
- Install detector at site.
- Prepare field test procedures/operating procedures for Y-12 D&D personnel.
- Perform system field test.
- Demonstrate process to DOE, Y-12 D&D, and ER management.
- Document results and transfer instrument to Y-12 D&D operations.
- Train operating personnel.

Note that the much of the laboratory and all of the design work has been completed. None of the fabrication or field testing has been started. Should the project be restarted, the sponsor could expect completion approximately 18 months after funding is in place.

Figure 1. Cup geometry.

Figure 2. Multiple cup geometry, conical ends.

Figure 3. Starting geometry for detailed study of scattering and solid angle.

Figure 4. Final configuration of moderator.

Figure 5. HPGe spectrum showing peaks due to Ge, Fe+²¹⁴Pb, and Hg.

Figure 6. Collimator layers.

References

1. Z. W. Bell, *Feasibility of Using Prompt Neutron Capture Gamma Rays to Detect Mercury*, Martin Marietta Energy Systems, Inc., Internal Report Y/DW-1250, October, 1993.
2. Z. W. Bell, *Detection of Concealed Mercury with Thermal Neutrons*, in Review of Progress in Quantitative Nondestructive Evaluation, Vol. 14, eds. D. O. Thompson and D. E. Chimenti (Plenum, New York), to be published. Also available as Martin Marietta Energy Systems, Inc., Report Y/DW-1304/R2, August, 1994.
3. J. F. Briesmeister, *MCNP 4A CCC-200*, (Radiation Shielding Information Center, Oak Ridge National Laboratory, 1993).
4. The curve fitting program GAUSSN was supplied by C. M. Davenport and implemented on the Y-12 Development Division VAX. Information on the method can be found in [davenportcm]gaussn.hlp on the Development Division VAX.

Distribution

Z. W. Bell, 9203, MS-8084
Y. C. Childs, 9766, MS-8112 (10)
J. L. Cochran, 9203, MS-8084
K. A. Kitzke, 9203, MS-8084
A. K. Lee/DOE-OSTI (2), 9731, MS-8175
J. M. Smith, 9103, MS-8142
S. G. Snow, 9203, MS-8084
S. A. Wallace, 9203, MS-8084