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FEASIBILITY OF USING PROMPT NEUTRON CAPTURE GAMMA RAYS TO DETECT MERCURY

Z. W. Bell

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Abstract

This report describes a study to determine the feasibility to use neutrons to probe hidden spaces within buildings for the presence of mercury. The study was performed in four phases: First a search of the scientific literature was performed to ascertain the behavior of mercury subsequent to the capture of a thermal or near-thermal neutron. Second, a Monte Carlo investigation (using the code MCNP) of the effects of neutrons on materials expected to be found near and/or surrounding the mercury was undertaken. Third, a Monte Carlo study of the shielding and beam forming properties of various configurations of moderator material was started. Lastly, a Monte Carlo analysis of a likely field situation involving mercury behind 1 inch and 2 inch thicknesses of concrete was performed.

Introduction

Mercury can be a serious contaminant of a building's air, plumbing, walls, and floors. It is much heavier than water, readily vaporizes at room temperature, and finds its way into the ecosystem by respiration, ingestion, and diffusion. It is toxic to humans and many animals. Consequently, if it is known or suspected that the material is present at a decontamination site, it is necessary to find the locations of deposits so that large quantities are not spilled or otherwise released to the environment.

Mercury occurs naturally as several isotopes, among them ^{199}Hg . This isotope represents 17% of natural mercury and possesses a thermal neutron capture cross section of 2200 barns. Thus, a layer of mercury 3.0 mm thick captures 99% of the thermal neutrons incident on it. In contrast to this, most structural materials have capture cross sections in range of 1-10 barns. Because of this, interrogation with thermal neutrons of a volume containing mercury would yield a significant number of gamma rays unique to mercury. These gamma rays are expected to have sufficient energy to penetrate any wall, pipe or floor hiding mercury from visual or other surface inspection. Furthermore, because of the relatively low absorption cross of most building materials, neutrons can penetrate walls, pipes and floors to reach and react with the mercury.

Neutrons interact with matter via nuclear reactions in which the incident neutron is either scattered or absorbed. Scattering may be either elastic or inelastic. In the latter case, the incident neutron survives, and the target nucleus may emit a proton, alpha particle, deuteron, neutron, one or more gamma rays, or a combination of these. The incident neutron also survives the former situation, but there is no signature of the event other than the change of energy and/or direction of the neutron. Typically inelastic scattering requires neutrons with energies of several keV.

The absorption process always results in the capture of the incident neutron by the target nucleus. This results in the emission of prompt (typically within nanoseconds) capture gamma rays that are characteristic of the target nucleus in same way the optical spectrum of the sun is characteristic of the elements present in the heliosphere. The major difference, however, is that the capture gamma rays are unique to each isotope of each element while the emissions from all isotopes of the same element are essentially identical. Detection of the gamma rays can lead to a quantitative measure of the amount of target material present.

Neutron absorption also may lead to a radioactive product. This is the process of neutron activation. In this case, the emanations from the product may be detectable and also may lead to a quantitative measure of the amount of target present. A product with detectable radiation is generally useful for several half-lives.

Neutron activation (which is not the thrust of this investigation) is a well established method of trace analysis. Usually a sample is placed in a nuclear reactor where it is bathed in the neutron flux. The sample captures some neutrons, and some of the material is converted to a radioactive isotope of the same element. The sample then is withdrawn from the reactor and the activity is measured. From a knowledge of the reactor's flux, and the neutron absorption cross sections, estimates of the amounts of the species present are made. This scheme depends on the presence of materials that upon absorption of a neutron become radioactive with a half-life that ranges from seconds to years (at most). Therefore, one cannot use this method to detect hydrogen, because deuterium is stable. On the other hand, it is possible to detect arsenic in this way because ^{76}As has a half-life of 26 hours and emits detectable radiation. This is what was done at Oak Ridge National Laboratory with samples of hair and finger nail from Zachary Taylor.

Absorption and scattering are competing nuclear processes whose likelihood of occurrence is measured by the cross section for the event. Cross sections are measured in units of barns (10^{-24} cm^2), and, for the purposes of this report, may be considered to be the area of a nucleus presented to an incident neutron. The cross section for different processes are different; a nucleus that is prepared to absorb a neutron can be viewed as having a different area than a nucleus that is prepared to scatter a neutron. In this model, then, the probability of a "scattering" nucleus colliding with a neutron (and, therefore, scattering it) is proportional to its cross section. Similarly, the probability of an "absorbing" nucleus interacting with a neutron is proportional (with the same constant of proportionality) to its cross section. Tables and charts of neutron interaction cross sections are widely available^{1,2,3}.

Cross sections are functions of the energy of the incident neutron. At high energies (above 1 Mev), the cross section is typically small (few barns) and relatively structureless. Important processes are inelastic scattering, and knockout reactions. At intermediate energies (1 eV to 1 MeV) the cross sections of many isotopes contain large, rapid, narrow fluctuations (resonances) at the energies of which strong absorption or scattering may occur. In addition, large, narrow minima in the cross sections occur that allow neutrons at selected energies unhindered passage. Below 1 eV, most cross sections are relatively featureless, with the cross section rising as the inverse of the neutron's velocity. The dominant processes are elastic scattering and absorption. Typically, the cross section at low energy is larger than the average cross section in the resonance region (except, perhaps, at a resonance energy), which is larger than the average cross section in the high energy region. Thus, the likelihood of interaction is largest for low energy neutrons.

Although it is advantageous to use the lowest possible neutron energy, it is not possible to generate neutrons whose energy, on average, is less than the thermal energy of surrounding atoms. The reason for this is easily understood upon consideration a group of neutrons whose average energy is larger than that of the surrounding atoms. As this group collides with its surroundings, scattering will serve to share the energy of the neutrons with their surroundings, thus lowering the average energy over time. The distribution of the speeds of the atoms in the surrounding medium is determined by the

temperature in the medium. The average of this distribution is not zero. Consequently, repeated collisions between neutrons and atoms "cools" the neutrons and "heats" the surroundings. For the case of a group of neutrons whose average energy is less than the thermal energy of atoms in the surrounding medium, collisions tend to "cool" the surroundings and "heat" the neutrons. In either case, the distribution of neutron speeds tends toward that of the surrounding atoms. Therefore, the lowest practical neutron energies obtainable are "thermal" and correspond to a distribution of energies typical of a gas at room temperature (mean value of 0.0253 eV). Part of this study involved simulations to determine an optimum configuration of moderator materials to thermalize as many neutrons as possible from a radioactive neutron source.

The thermal neutron absorption cross section of mercury is 384 barns and is due almost entirely to the 2200 barn cross section of the isotope ^{199}Hg . A cross section of this magnitude implies that a layer only 3 mm thick, absorbs over 99% of the thermal neutrons incident on it. However, the resultant ^{200}Hg is stable, and therefore cannot be detected by activation analysis. On the other hand, 77% of the deexcitations of the ^{200}Hg involve the emission of a 368 keV gamma ray. This gamma ray is moderately penetrating, and is easily detected by commercially available high-purity germanium detectors. Part of this study involved the determination of the state of knowledge of the deexcitation of ^{200}Hg .

A device using thermal neutrons as a probe has the advantage that neutrons are not strongly absorbed by materials such as concrete and steel. Consequently, neutrons are able to penetrate to a depth of a few inches in such materials and they can interrogate the hollow spaces of tile walls, cinder block, and pipes. Furthermore, because the energy of the deexcitation gamma ray is so high, it can penetrate the walls of tile, block and pipe and be detected. The device can conduct a one-sided measurement; access to both sides of a pipe or wall (as would be necessary for radiography) is not necessary since the capture gamma rays are emitted isotropically. Also, measurements are not expected to be affected by surface preparation (paint) or texture. Part of this study involved simulations to estimate the detectability of the mercury gamma ray.

Among the disadvantages are the need for heavy and bulky shielding around the neutron source and detector. Personnel must also be trained in the use and handling of radioactive material (the neutron source), and they must also be monitored for radiation exposure. It is also possible to induce radioactivity in the area where the source is used. (This topic is treated at length below.) Simulations were performed, as part of this study, to determine optimum shielding for both personnel and the gamma ray detector.

Review of the Literature

Although there have been no previous investigations of a prompt capture gamma ray based detector for mercury, neutrons have been investigated as a possible probe for mercury. A group at MIT⁴ has reported using neutron activation analysis to determine the levels of airborne mercury. Unfortunately, the 77 keV gamma rays detected in this experiment are of insufficient energy to penetrate much concrete or steel. In addition, the experiment required a nuclear reactor, and several hours of counting time to make a determination. A Soviet paper abstracted in Chemical Abstracts⁵ implies that the high cross section of mercury makes it feasible to determine the amount present from a measurement of the attenuation of a thermal neutron beam. This, of course, requires a source and a detector to straddle a prepared sample. The method is not suitable in

situations where there is little control over the non-mercury sample content or where access to both sides of the sample is denied.

A review of the scientific literature on the capture of neutrons by mercury has revealed many investigations into the absorptive properties of ^{199}Hg with respect to neutrons. Only a few of them will be described here.

Breitag, Casten, and Cole⁶ measured the capture gamma rays from the capture of thermal neutrons and at the resonances at 33.5, 129.7 and 175.1 eV using a bent-crystal spectrometer, and germanium detectors. They observed over 500 transitions, and devised a level scheme containing 60 levels below 3.3 MeV. They found that the first excited state, at 368 keV, was populated in 77% of the captures. This occurred because the deexcitation of ^{200}Hg proceeds via gamma ray cascades, many of which pass through the first excited state. The next most common transition has an energy of 1693 keV and occurs in 11 % of all captures. It is caused by a transition to the first excited state, and is in coincidence with the 368 keV gamma. The most common transition that does not go through the first excited state has an energy of 1570 keV and occurs in 6% of captures.

Lone, Earle, and Bartholomew⁷ used a chopper at a reactor to study the resonances at 33.5, 129.7, and 175.1 eV. They found that the deexcitation of ^{200}Hg from capture by these resonances also proceeded largely through the first excited state. They report the 368 keV gamma ray occurring in 81, 77, and 82 % of captures at the three resonance energies, respectively.

Schmorak⁸ has reviewed the literature through 1979, evaluated the data, and generated a consistent level scheme and deexcitation scheme for ^{200}Hg . He has adopted much of the work of Breitig, Casten, and Cole, and Lone, Earle, and Bartholomew, and does not disagree with the intensity of the 368 keV line assigned by those authors.

Reference 1 indicates that the thermal neutron capture cross section of ^{199}Hg is 2200 barns. Coupling this information with the high probability of the 368 keV gamma ray makes the detection of mercury by capture gamma ray detection an attractive possibility.

Tuli⁹ has produced a compendium of thermal neutron capture gamma rays sorted by energy and isotope. This table was searched for gamma rays of energy near 368 keV produced by the capture of neutrons by other nuclides. In the region from 360 keV to 374 keV, gamma rays are produced by Ir, Re, Ta, Ag, Os, Rb, Sm, As, Yb, Nd, Br, Fe, Lu, Pt, Am, Tl, Au, Dy, Ho, and Ga, in addition to Hg. Of these elements, only iron is of practical significance because it is the main constituent of steel.

Iron produces a gamma ray with energy 366.8 keV in approximately 1.5 % of neutron captures¹⁰. This gamma ray is sufficiently close in energy to that of mercury, that the peaks due to these gamma rays would not be well separated in the spectrum from a germanium detector. However, iron also produces a capture gamma at 352.4 keV in 10.9 % of neutron captures. Therefore, accurate measurement of this gamma ray implies that a correction to the data from 368 keV is possible. Tuli's table indicates that there are no competing gamma rays in the region around 352 keV that would be of significance to this study. Spectra showing these gamma rays are discussed below.

Experimental Study

An experiment was conducted in which an Am-Li neutron source housed in a polyethylene moderator was used to irradiate a 3300 gram stainless steel sample and a 13 gram liquid mercury (housed in a thermometer bulb) separately and together. The neutron flux was estimated to be about $15/\text{cm}^2/\text{s}$. The gamma ray detector was a 15% relative efficiency high purity germanium detector. Figure 1a shows a capture gamma ray spectrum obtained from the steel alone. The 352 keV and 366.8 keV gamma ray peaks are clearly visible. Figure 1b shows the spectrum obtained from the same piece of steel, but with the mercury sample present also. The peak at 352 keV is seen to be unobstructed, while the peaks at 366.8 and 368 keV are merged. It is clear that the existence of the mercury peak can be determined because of the distortion of peak shape. The two peaks are sufficiently well separated to obtain estimates the contributions of each. Alternatively, the iron contribution to the merged peak can be inferred from the 352 keV peak.

Estimation of Neutron Activation

Because neutrons are to be used by this instrument, it is reasonable to be concerned that unwanted activation of structural materials may occur. Therefore, an effort was made to estimate the level of activation of various materials. Concrete, iron, nickel, stainless steel, mercury, copper, aluminum, air, and lithium were considered. The table below shows the elements present in these materials and gives some relevant physical constants for each.

Element	Mass	Abund.	Cross Section (barns)	Unstable Product
Cr	50	.0435	15.8	^{51}Cr
	52	.8379	0.8	
	53	.95	18.0	
	54	.0236	0.36	^{55}Cr
Mn	55	1.00	13.3	^{56}Mn
Fe	54	.058	2.3	^{55}Fe
	56	.917	2.6	
	57	.022	2.5	
	58	.003	1.28	^{59}Fe
Ni	58	.683	4.6	^{59}Ni
	60	.261	2.9	
	61	.011	2.4	
	62	.036	14.6	
	64	.009	1.55	^{63}Ni ^{65}Ni
Cu	63	.692	4.47	^{64}Cu ^{66}Cu
	65	.308	2.17	
C	12	.989	0.0035	^{14}C
	13	.011	0.0014	

N	14	.9963	1.83 (n, p) 0.078 (n, gamma) 0.00002	¹⁴ C ¹⁶ N
	15	.0037		
Ca	40	.969	0.41	⁴¹ Ca
	42	.006	0.7	
	43	.001	6.0	
	44	.021	0.84	⁴⁵ Ca
	46	.00004	0.7	⁴⁷ Ca
	48	.002	1.1	⁴⁹ Ca
O	16	.998	0.0002	
	17	.0004	0.0004 (n, gamma) 0.24 (n, alpha)	¹⁴ C ¹⁹ O
	18	.002	0.0002	
H	1	.99985	0.333	
	2	.00015	0.00051	³ H
Si	28	.922	0.177	
	29	.047	0.10	
	30	.031	0.107	³¹ Si
S	32	.9502	0.53 (n, gamma) 0.004 (n, alpha)	
	33	.0075	0.45 (n, gamma) 0.14 (n, alpha) 0.002 (n, p)	³³ P ³⁵ S ³⁷ S
	34	.0421	0.23	
	36	.0002	0.23	
	24	.790	0.053	
	25	.100	0.18	
Mg	26	.110	0.038	²⁷ Mg
	23	1.00	0.53	²⁴ Na
	24			
K	39	.9326	2.1 (n, gamma) 0.0043 (n, alpha) 30.0 (n, gamma) 4.4 (n, p) 0.39 (n, alpha)	³⁶ Cl
	40	.0001		
	41	.0673	1.46	⁴² K
	6	.075	0.00039 (n, gamma) 941.0 (n, alpha)	³ H ⁸ Li
	7	.925	0.45	
Li (nat)	196	.0015	3220.0	¹⁹⁷ Hg
	198	.101	1.9	
	199	.170	2200.0	

	200	.231	< 60	
	201	.132	8.0	
	202	.300	4.9	²⁰³ Hg
	204	.068	0.4	²⁰⁵ Hg
Al	27	1.00	0.233	²⁸ Al

Most of the above materials (H, O, C, Na, Mg, Al, Si, S, K, Ca, Fe) are constituents of concrete. Iron is present all steels. Nickel, manganese, and chromium are the major additives to iron in stainless steels. It was assumed that wiring is made of copper and that the other materials may be found.

In all calculations, absorption cross sections were taken from reference 1. Calculations were done for a source emitting 100,000 thermal neutrons/sec, and all neutrons were absorbed in the material. All calculations of activity level are for 1 hour of irradiation. This is probably a longer time that will be used in the field.

Concrete

The concrete formulation used was for an "ordinary" concrete¹¹ with density 2.2 g/cc. In the table below, the fraction absorption represents the fraction of neutrons captured in a block of concrete by each constituent element. The last column indicates the induced activity (in pC) in 1 cubic meter (2200 kg) of concrete due to each product after 1 hour of irradiation.

Element	% by weight	Fraction absorption	Activity after 1 hr.	
H	0.85	0.357	None	
C	0.48	0.000178	0.000176	¹⁴ C
O	50.64	0.000112	0.00009	¹⁴ C
			0.0052	¹⁹ O
Na	1.66	0.0486	6204.0	²⁴ Na
Mg	0.23	0.000756	132.6	²⁷ Mg
Al	4.44	0.0486	137114.	²⁸ Al
Si	30.49	0.236	3003.	³¹ Si
S	0.12	0.00248	0.000230	³³ P
			0.043	³⁵ S
			0.617	³⁷ S
K	1.87	0.127	1.8x10 ⁻⁷	³⁶ Cl
			91.35	⁴² K
Ca	8.03	0.109	0.000226	⁴¹ Ca
			2.22	⁴⁵ Ca
			0.127	⁴⁷ Ca
			1565.	⁴⁹ Ca
Fe	1.19	0.0692	0.0300	⁵⁵ Fe
			0.190	⁵⁹ Fe

Bear in mind that these activities are in picocuries. On a per gram of concrete basis, the ²⁸Al yield is only 0.062 pC/gram. This approximately 1/600 the allowed limit of ²³⁸U in trash. All other yields are significantly smaller.

The worst offenders in this list are ^{24}Na , ^{27}Mg , ^{28}Al , ^{31}Si , ^{42}K , and ^{49}Ca . The following table shows the residual activity (pC) in the hypothetical concrete block (1 cubic meter, 2.2 g/cc) at various times after irradiation.

	Half Life	1 hr	1 day	3 days	7 days
^{28}Al	2.25 min	0.0013	0	0	0
^{49}Ca	8.72 min	13.28	0	0	0
^{27}Mg	9.45 min	1.63	0	0	0
^{31}Si	2.62 hr.	2305	5.25	0	0
^{42}K	12.4 hr.	86.36	23.78	1.61	0.0074
^{24}Na	15.0 hr.	5923	2042	221	2.60

Thus, it is clear that the already low activity levels drop rapidly with time and induced activity in concrete would not be expected to be a problem.

All results below were calculated with the neutron transport code MCNP, written at Los Alamos National Lab, and obtained from the Radiation Shielding Information Center at ORNL.

Iron

The following results are for a 2.54 cm (1 inch) thick iron slab, 30 cm (1 foot) wide and 300 cm (10 feet) long, surrounded by air, irradiated by 100,000 neutrons/sec over a 15 cm diameter area at the center of a 30 cm surface for 1 hour. (Note that this source produces 44 times the flux as did the source used for the concrete block.) This is intended to simulate a steel girder, flooring, or part of an I-beam. It is not expected that the results would differ significantly for a pipe. ^{54}Fe would be expected to produce 2.01 pC of ^{55}Fe while ^{58}Fe would be expected to produce 1.19 pC of ^{59}Fe uniformly distributed in 14 kg iron. This mass of iron is just the material irradiated by the source, rather than the entire bar. The activity per gram is 0.0002 pC/g.

Nickel

The following results are for a 2.54 cm (1 inch) thick nickel slab, 30 cm (1 foot) wide and 300 cm (10 feet) long, surrounded by air, irradiated by 100,000 neutrons/sec over a 15 cm diameter area at the center of a 30 cm surface for 1 hour. ^{58}Ni was found to induce 0.00117 pC of ^{59}Ni ; ^{62}Ni was found to induce 0.147 pC of ^{63}Ni ; ^{64}Ni was found to induce 1213 pC of ^{65}Ni .

^{65}Ni has a 2.52 hour half-life, and its behavior is identical to that of ^{31}Si above: In 1 - 2 days, the activity due to this isotope disappears.

Stainless Steel (-304)

The following results are for a 2.54 cm (1 inch) thick stainless steel slab, 30 cm (1 foot) wide and 300 cm (10 feet) long, surrounded by air, irradiated by 100,000 neutrons/sec over a 15 cm diameter area at the center of a 30 cm surface for 1 hour.

Isotope	Activity (pC)	Product
⁵⁰ Cr	70.5	⁵¹ Cr
⁵⁴ Cr	836	⁵⁵ Cr
⁵⁵ Mn	29555	⁵⁶ Mn
⁵⁴ Fe	1.28	⁵⁵ Fe
⁵⁸ Fe	0.758	⁵⁹ Fe
⁵⁸ Ni	0.000153	⁵⁹ Ni
⁶² Ni	0.0192	⁶³ Ni
⁶⁴ Ni	158	⁶⁵ Ni

⁵⁶Mn has a half-life of 2.6 hours; it decays away in 1 - 2 days. ⁵⁵Cr has a half-life of 3.5 minutes; it decays away in 1 - 2 hours. ⁶⁵Ni has been discussed above.

⁵¹Cr has a half-life of 27.7 days, and is potentially worrisome if bars of chromium are present at the site. As a component of stainless steel, however, 70.5 pC / 14000g (0.005 pC/g) it should not be considered a hazard.

Aluminum

As seen above, any activity induced in aluminum decays with a 2.25 minute half-life. Consequently, any activated aluminum is cold within an hour after irradiation.

Copper

The following results are for a cable 2.54 cm thick, 300 cm long, surrounded by air, irradiated by 100,000 neutrons/sec over a 15 cm diameter area at the center of the cable for 1 hour. This geometry approximates an electrical cable.

⁶³Cu induces 14876 pc of ⁶⁴Cu; ⁶⁵Cu induces 60589 pc of ⁶⁶Cu. As in the case of ²⁷Mg and ⁴²K, these products decay away rapidly.

Nitrogen

The source of nitrogen captures is the nitrogen in the air. From the calculations of the iron, copper, and nickel, the production of ¹³C from (n, p) reactions caused by neutrons escaping the bars might be expected to yield 0.000019 pC in the air within a 5 foot radius of the source. This translates to 13x10⁻¹² pico curies per liter, which is unmeasurable.

The reaction ¹⁵N(n, γ)¹⁶N is insignificant because of the abundance (or, rather, lack of it) of ¹⁵N. In addition, ¹⁶N has a half-life of only 7 seconds, and decays away in minutes.

Mercury

Since it is impossible to predict the exact geometry of mercury deposits, the calculation was performed for a 1 cm cube (13.55 g) irradiated with the same flux (10 neutrons/sec/cm²) as irradiated the concrete block for 1 hour.

¹⁹⁶Hg induces 0.57 pC of ¹⁹⁷Hg; ²⁰²Hg induces 0.0099 pC of ²⁰³Hg; ²⁰⁴Hg induces 0.299 pC of ²⁰⁵Hg. These activities are similar to the activities generated in the concrete block and are negligible.

Lithium

Lithium is more difficult to assess because the enrichment in ${}^6\text{Li}$ is unknown. The ${}^7\text{Li}(n, \gamma)$ reaction is not significant because the ${}^8\text{Li}$ product has a half-life of only 0.844 second. Consequently in a short amount of time, any activity becomes unmeasurable.

The ${}^6\text{Li}(n, \alpha){}^3\text{H}$ is only significant if appreciable quantities of ${}^6\text{Li}$ are present. The cross section for this reaction is so large, that essentially all neutrons incident on material containing significant quantities of ${}^6\text{Li}$ are absorbed. Assuming a completely absorbing material, a 1 hour irradiation time by 100,000 thermal neutrons/sec, would induce 0.0000174 pC of tritium activity. This is an insignificant quantity.

In summary, the above calculations show that while radioactivity is induced in materials expected to be found at a site, the levels generated are, for the most part, insignificant. Furthermore, those species that do give rise to measurable activities all have short half-lives. Consequently, in a matter of hours, or at most days, all activity will have decayed away to background levels.

All calculations were done with a source that represents an upper limit on the number of thermal neutrons/second to be emitted. Similarly, 1 hour of irradiation is probably too large by a factor of 4 to 6. In actual practice, it is more likely that the actual levels would be lower by a factor of 10 to 20 from those quoted above.

Monte Carlo Study of Detector Shielding

It will be important to shield the germanium detector from stray neutrons and gamma rays. Neutrons can activate the detector itself, and excess gamma rays cause unwanted background. A cylindrical shield with four layers was considered. An outermost layer, 0.5 cm thick, composed of ${}^6\text{LiF}$ stops all thermal neutrons and generates no gamma rays (as does cadmium and ${}^{10}\text{B}$). The next inner layer was 10 cm of W/Ni/Fe alloy, which stops essentially all gamma rays below about 2 MeV. The next inner layer was 0.5 cm of tin, to filter the fluorescent x rays from tungsten. Although cadmium is often used for this purpose, tin was selected here because it is not considered an environmental problem. The innermost layer was 0.5 cm of copper, to remove the tin x rays. The fluorescent x rays from copper are below the detection threshold of the germanium detector. This shape of shield also serves a collimator and gives directionality to the field of view of the detector. It will also be sufficiently massive to support and protect the detector.

Monte Carlo Study of Source Moderator

Neutrons are not the result of any naturally occurring radioactive decay. Hence, they must be produced by a nuclear reaction. Although a nuclear reactor is an excellent source of neutrons, it would be somewhat impractical to employ one in this application. Accelerator-based sources make use of the (γ, n) or (p, n) reaction, but again, this type of apparatus is not suitable for a field instrument. Radioactive neutron sources that employ spontaneous fission (${}^{252}\text{Cf}$), or the $\text{Be}(\gamma, n)$ or an (α, n) are widely available, compact, and easily shielded. Fission sources produce, in addition to neutrons, many high energy gamma rays, and would therefore cause problems in shielding the instrument's gamma ray detector and operators. $\text{Be}(\gamma, n)$ sources are typically large

because they rely on the absorption of gamma rays by Be (which has a small absorption coefficient) to produce neutrons. Therefore, it was decided to investigate (α , n) sources.

Americium is often used as the source of alpha particles for a radioactive neutron source. Typically an intimate mixture of the oxide with a low Z material such as Li, B, Be, or C, or an alloy of the low Z material with the Am is used. The close proximity of the target to the source of alpha particles ensures reasonable efficiency.

The most prolific of such sources is the Am-Be combination. Unfortunately, each neutron is accompanied by a 4.4 MeV gamma ray which is difficult to shield. Am-¹³C sources produce a 6 MeV gamma ray. Therefore, these sources were rejected.

A further consideration is the energy distribution of the neutrons from the source. All of these sources produce a spectrum of neutrons whose upper end may lie at several MeV. Since for this study, thermal neutrons are desired, it is desirable to choose a source with the most thermal spectrum. The Am-Li source produces a spectrum whose highest energy is approximately 1.5 MeV with a most probable energy of about 200 keV. Therefore, this source was chosen for further study.

As described above, neutrons are thermalized by their collisions with nuclei in their surroundings. The best moderator is hydrogen, which has a scattering cross section and a small capture cross section. At thermal energies, scattering is favored over capture 60 to 1. Other materials, such as deuterium, and carbon have much smaller capture cross sections than does hydrogen, but the average energy loss per collision is much smaller, and the probability of collision between a neutron and either of these materials is much smaller than that of hydrogen.

For the purposes of this gauge, it is necessary to direct the neutrons toward a wall, for example, to be interrogated. It is necessary for the moderator to be as efficient as possible in thermalizing neutrons while not capturing neutrons. Simulations were performed with a cylindrical moderator assembly (the "pig") encased in 0.25 inch of steel. The hydrogen was in the form of polyethylene. Various configurations of polyethylene and graphite were evaluated, and statistics that measured the probability of neutrons exiting pig in the desired direction were accumulated. The source was always modelled as a point source within an otherwise empty cylinder 2 cm in diameter, and 2 cm long.

Slab geometries, in which the neutron source is sandwiched between slabs of graphite and/or polyethylene, were examined first. Figure 2 shows the general layout of the moderator material; the steel case is omitted. Material in the cells labelled 1 through 6 are graphite or polyethylene. Cells 3 and 4 are annuli; cells 1, 2, 5, and 6 are disks. Best results were obtained when materials 2, 3 and 5 were polyethylene and had a thickness of no more than about 5 cm. The remaining material was graphite with similar dimensions. This seemed to minimize the absorption of neutrons while still allowing significant numbers to escape. The fraction of source neutrons escaping was typically near 50%. However, this geometry did not preferentially channel neutrons toward any surface and a significant number of escapes occurred through the round side.

A geometry of concentric cylinders, as shown in figure 3, was investigated. Cell 1 contained graphite and cell 2 contained polyethylene. The dimensions of cell 2 were varied while keeping the outer radius of cell 1 fixed. It was found that the highest flux of outward bound neutrons, through the bottom surface, was obtained when distance, A,

between the bottom of the polyethylene cylinder and the bottom of the graphite cylinder, was set to zero. It was still necessary to use approximately 5 cm of polyethylene to adequately thermalize the neutrons. With the bottom of the two cylinders coincident, approximately half of the escaping neutrons escaped through the bottom. Capture still accounted for approximately half of all source neutrons.

The latter geometry, while promising, still resulted in the loss of half the escaping neutrons. It would be very desirable to reflect the neutrons that were heading for the top and side of the pig back into the interior. To this end, simulations were performed with "cup" geometries in which the central cylinder was surrounded by concentric cups of graphite and polyethylene as shown in figure 4. Figure 4 shows an arrangement of polyethylene cups at the edge of the pig and midway between the edge of the pig and the outer edge of the thermalizing cylinder.

The theory behind this geometry is that polyethylene has a somewhat higher scattering cross section than does graphite. Graphite also has a very small capture cross section. Consider a graphite rod surrounded by polyethylene. Neutrons that are heading parallel to the axis of the rod can travel essentially unhindered. However neutrons that reach the outer surface of the rod, are quickly scattered by the polyethylene. There is a high probability that the neutron will be scattered back into the rod; thus increasing the number of neutrons available to travel down the rod.

Three such geometries were investigated: A single polyethylene cup (1 cm thick) placed at the outer edge of the pig (but inside the steel), two cups arranged as in figure 4, and 4 cups arranged symmetrically between the inner cylinder and outer wall. The central cylinder was thinned slightly above the source, and in radius, to compensate for the extra polyethylene of the cups.

With the single cup at the outside edge of the pig and the thinned thermalizing cylinder, neutron escapes jumped to 61.5%. Of those escapes, 67% were directed out the bottom, 22.5% went out the top, and the remainder went out the side. Net, 41% of the source neutrons exited the bottom of the pig, while 20.6% of the neutrons exited the top or side.

Adding a second cup, as shown in figure 4, reduced the percentage of escaping neutrons to 58%. However, of those escapes, 70% were now exiting the bottom of the pig. Net, 40.2% of the source neutrons went out the bottom of the pig, and 17.4% exited the top or side. Thus the 3% additional captures are seen to be preferentially from those neutrons that would have been lost to the system.

Adding two additional cups midway between the cups shown in figure 4 dramatically decreased the number of escapes to 47.2%. This was reflected in the absolute percentage of source neutrons escaping through the bottom (36.2%). However, neutrons exiting through the bottom now made up 76.7% of all escaping neutrons. The percentage of neutrons escaping through the side or top was 10.9%. Again, a much larger effect was seen in the lost neutrons than in the useful neutrons. Simulations were performed for geometries using a pig with two polyethylene cups and a mercury button, 0.3 cm thick and 1 cm radius (3.2 grams), hidden behind 2.54 cm and 5.08 cm of concrete. The button was placed on the centerline of the cylinder of the pig. Through 2.54 cm and 5.08 cm of concrete, the simulations showed that the probability of capture was 0.0588% and 0.0324%, respectively, per source neutron. This means that for a source strength of 100,000 neutrons/second, 58.8 and 32.4 captures/second could be

expected. Assuming a detector efficiency of 0.2%, count rates of 0.09 and 0.05 per second in the 368 keV peak might be expected. Thus, in only about 15 minutes, a significant indication of the presence of the mercury would be seen.

Conclusions and Summary

The scientific literature indicates that mercury has a very high thermal neutron absorption cross section that is almost entirely due to the isotope ^{199}Hg . Because ^{200}Hg is stable, activation analysis is rejected as a detection tool. However, the deexcitation of ^{200}Hg proceeds with the emission of a 368 keV gamma ray in 77% of decays. Consequently, a gauge based on the detection of that prompt gamma ray was investigated.

Monte Carlo simulations of various source/moderator configurations have shown that geometries based on a source surrounded by alternating concentric cups of graphite and polyethylene can channel 35% to 40% of the source neutrons toward a target, while minimizing the number of neutrons exiting in undesirable directions.

Safety and waste disposal concerns dictated consideration of the possibility of unintentional activation of materials exposed to the neutron source. Calculations indicated that the most of the significant activity would be caused by materials with short half-lives (minutes to hours). Consequently, any such induced activity would die away within a few day's time. Chromium was found to be the only exception to this, and special care would need to be taken if significant quantities (such as bars or ingots) of the pure metal were present. As a constituent of stainless steel, however, only 0.005 pC/g might be expected. This is not a significant quantity.

The effects of interposed concrete were investigated by Monte Carlo simulation. It was found that even through 5 cm (2 inches), only a few minutes would be needed to determine the presence of a few grams of mercury. An experiment testing the interference of capture gamma rays from iron confirmed that mercury could be detected at the level of 1 part in 200.

From the above considerations, it is reasonable to conclude that small drops of mercury should be visible to a gauge detecting prompt capture gamma rays, even if they are behind up to 5 cm (2 inches) of concrete. In the presence of iron or steel, however, careful analysis of the spectrum is necessary to account for the contribution of the 366 keV gamma ray from iron.

References

1. F. William Walker, Dudley G. Miller, and Dr. Frank Feiner, Chart of the Nuclides, 13th edition, General Electric Company, 1983.
2. D. I. Garber and R. R. Kinsey, Neutron Cross Sections, BNL 325, Third Edition, National Neutron Cross Section Center, Brookhaven National Laboratory, 1976.
3. The National Nuclear Data Center at Brookhaven National Laboratory maintains an online computer system that gives users access to its nuclear physics and

photo-atomic physics data bases and bibliographic services. TELNET to BNLND2.DNE.BNL.GOV and use the username NNDC.

4. Michael Ames, Ilhan Olmez, Sandra Meier, and Philip Galvin, A Methodology for Determining Vapor Phase Mercury by Instrumental Analysis, presented at the EPRI Second International Conference "Managing Hazardous Air Pollutants," July 13 - 15, 1993.
5. N. G. Malysheva, Possible use of neutron absorption for the determination of mercury enrichment products, Chem. Abstracts, Abstr. 1225, April 1964.
6. D. Breitig, R. F. Casten, and G. W. Cole, Physical Review C, 9, 366 (1974).
7. M. A. Lone, E. D. Earle, and G. A. Bartholomew, Nuclear Physics A243, 413 (1975).
8. M. Schmorak, Nuclear Data Sheets 26, 81 (1979).
9. J. K. Tuli, Thermal Neutron Capture Gamma Rays, Report NNDC/INT-91/1, National Nuclear Data Center, Brookhaven National Laboratory, 1991.
10. Nuclear Data Sheets, 20, 253 (1977).
11. Edwin J. Callan, Concrete for Radiation Shielding, in Concrete for Radiation Shielding, 2nd Edition, published by the American Concrete Institute, 1962, p. 13.

Figure 1. Spectra of 3300 gram stainless steel sample (a) alone, (b) with 13 grams of Hg.

Figure 2. Slab geometry for moderator design.

Figure 3. Slab/sandwich geometry for moderator design.

Figure 4. Concentric cup geometry for moderator design. Only two cups are shown here; other cups were placed symmetrically between the polyethylene cups shown here.