Development of Tc-99 Characterization Approach for the Portsmouth Gaseous Diffusion Plant

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ABSTRACT

A methodology to non-invasively characterize the Tc-99 inside the equipment at the Portsmouth Gaseous Diffusion Plant was developed. The sensitivity of the approach was examined through critical benchmarking measurements and computer simulations. From comparison of these measurements and computer simulations, uncertainties in the methodology were established. The study results have shown that, given simplified geometries of pipe wall and deposit materials, the Tc-99 bremsstrahlung can be used to characterize the level of contamination of Tc-99 in a uranium deposit assuming uniform distribution of Tc-99 in the deposit. The sensitivity of the approach is adequate to address worker safety limits based on calculations from the derived air concentrations. It is anticipated that the technique will be most successful in thin-walled containers, such as aluminum bellows or copper tubing. The reliability of the approach needs to be tested by conducting in situ measurements of known quantities of Tc-99 contamination.

INTRODUCTION

The Portsmouth Gaseous Diffusion Plant is located on a U.S. Department of Energy (DOE)-owned reservation in Pike County, south-central Ohio. Technetium was introduced to the process equipment as a contaminant that accompanied irradiated uranium fed into the Portsmouth plant. During the operating history at Portsmouth, about 1,100 metric tons of irradiated uranium with 60 to 90 kilograms (kg) of technetium were fed into the plant. As of March 1999, the technetium inventory at Portsmouth was estimated to be about 35 kg, or about 600 curies (Ci) 1. Tc-99 has a specific activity 10,000 times higher than uranium, so even small quantities of Tc-99 can pose significant worker safety concerns and waste disposal issues. There is currently no means to perform nondestructive assays of Tc-99 from outside of the equipment. This article describes the initial development of a methodology, conducted in the last year, to characterize the Tc-99 contamination inside the equipment at Portsmouth.

Tc-99 is almost strictly a beta emitter, with a maximum energy of 293.5 kilo-electron volts (keV). Betas do not travel far in other materials. For instance, a 100 keV beta will travel 0.07 millimeters (mm) in aluminum before stopping. This short range makes the characterization of Tc-99 from direct decay products nearly impossible if the beta must travel through any material, such as a pipe wall. As the betas slow down in material, they emit photons. The photons are called "bremsstrahlung," which is German for "braking radiation." Photons will travel through much more material than betas; on average, a 100-keV photon will travel through 22 mm of aluminum before interacting. Thus, it is conceivable that Tc-99 inside of pipes may be characterized by observing the indirect bremsstrahlung products.

The approach of characterizing Tc-99 by observing its bremsstrahlung products has been previously examined. Williams et al. studied the rate of Tc-99 bremsstrahlung from nickel metal ingots contaminated with Tc-99 2. The metal ingots were generated from scrap metal during diffusion plant upgrades. In addition to the Tc-99, the ingots were contaminated with low levels of uranium-235 (U-235), uranium-238 (U-238), neptunium-237 (Np-237), and plutonium-239 (Pu-239). The radiation from these transuranic radionuclides did not significantly interfere with the Tc-99 bremsstrahlung signal. They achieved a minimum detectable concentration of 5 ± 1 parts per million (ppm) with a 5-minute measurement.

Bremsstrahlung from Tc-99 has been observed at Portsmouth 3. The bremsstrahlung was typically observed in thin-walled containers, such as aluminum bellows or copper tubing. At that time of the observations, there was no attempt to develop a quantitative methodology for characterizing the Tc-99 inside the pipes.

It is helpful to understand a number of qualitative aspects of beta decay and bremsstrahlung production:

- The energy distribution of the betas from beta decay is continuous, and monotonically decreasing as the energy increases. The average energy of the beta distributions is about one-third of the maximum energy—for example, the average beta energy for the 293.5-keV decay of Tc-99 is approximately 100 keV.
- The bremsstrahlung production depends to a significant extent on the atomic number (Z) of the material in which the beta decay occurs. As Z increases, more bremsstrahlung photons are created, and these photons tend to have higher energies.
- The transport of the photons through material depends to a significant extent on the atomic number of the material through which the photons pass on the way to the detector. The higher the Z, the more low-energy photons are absorbed.
- Bremsstrahlung production is a highly inefficient process. For every one bremsstrahlung photon that reaches the detector, there are 10^4 to 10^5 beta-decaying parent nuclei.
- In very high-atomic-number materials, like lead and uranium, the K-edge has a significant impact on transport of photons through the material.

Each of these aspects plays a critical role in developing a method to characterize Tc-99 via its bremsstrahlung products.

The impact of the K-edge effects requires further discussion. Photons with energies around this edge are most likely to interact with matter via the photo-electric effect, in which the photon is absorbed by an atomic electron and frees that electron from its binding to that atom. For high-Z materials, the photoelectric effect is significantly dominated by the innermost electrons, the K-shell electrons.

This dominance of the photoelectric effect with K-shell electrons in high-Z materials has two impacts. First, as the bremsstrahlung photons drop below the energy necessary to free a K-shell electron from its atom, the reaction probability for those photons suddenly drops. This rapid drop in cross section as the photon energy decreases translates into a sudden increase in the transmission of the photons through a fixed amount of material. The second impact is the enhancement of x-rays from the parent nucleus. When a bremsstrahlung photon knocks out a K-shell electron, the parent nucleus emits x-rays as outer-shell electrons drop in energy to fill the void of the knocked-out K-shell electron. These x-rays are distinct from the x-rays emitted as the parent decays. In that case, the x-ray's energy is determined by the electromagnetic field of the daughter nucleus. This distinction is important because the rate of the x-rays from the parents can be enhanced by bremsstrahlung, whereas the rate of the x-rays from the daughters cannot.

For high-Z materials, the K-shell effects have a dramatic impact on the energy distribution of the observed bremsstrahlung photons as can be seen in Fig. 1a and b for the transmitted bremsstrahlung through aluminum and lead respectively. Lead provides a clean example of high Z material since it has little residual activity. The aluminum bremsstrahlung spectrum varies smoothly with a fairly low energy peak in its distribution. The lead spectrum, in contrast, has a significant structure due to the K shell effects. The K-edge for lead is 88.0 keV, and the four x-rays of lead are 72.8, 75.0, 84.8, and 87.6 keV. Both the sudden increase in the transmission of photons immediately below the K-edge and the enhancement of the lead x-rays are obvious. Without the Tc-99 source, the x-rays would be practically unobservable.

METHODOLOGY

This section describes a methodology for characterizing the Tc-99 contamination in deposits at the Portsmouth Gaseous Diffusion Plant from outside of the pipe. An emphasis has been placed on the contamination in deposits with high enrichments (greater than 20%), because it is known that the Tc-99 contamination tends to be more common in the highest enrichment segments of the plant. It is possible to extend this procedure for lower enrichments, but many of the uncertainties would have to be re-examined.

In developing this methodology, we assumed that:

- Measurements would be conducted on the outside of the Portsmouth equipment.
- The material, thickness, and geometry of the pipe are known.
- The enrichment of the deposit is known to 10% (relative). The enrichment can be determined from either historical data or from measurements at the site.
- The Tc-99 contamination is uniformly distributed throughout the deposit.
- The thickness of the deposit is unknown and must be determined.
- The deposit is large compared to the size of the detector.
- There is no prior knowledge of the radiation spectrum not related to possible Tc-99 sources, and there is no way to measure this "background."
- The level of U-232 contamination is known to 50 parts per trillion (ppt) of uranium.

The procedure for characterizing the Tc-99 contamination is fairly straightforward:

1. Measure the photon spectra of a possibly contaminated pipe or tubing with a high-purity germanium (HPGe) detector.
2. Determine the strength of the 144, 163, and 186 keV lines of U-235 in that spectrum, denoted ρ_{144}, ρ_{163}, and ρ_{186}, respectively.
3. Determine the strength of the spectrum over an energy range in which a significant bremsstrahlung signal is anticipated (e.g., 70 to 115 keV), denoted ρ_{brems}.
4. From knowledge of the pipe, the level of enrichment, and the strength of the 186 keV line, determine the thickness of the deposit in grams per square centimeter (g/cm^2) inside the pipe.

5. Form the ratio of the strength of the spectrum in the bremsstrahlung region to that of the gamma peaks (e.g., ρ_{brems}/ ρ_{186}). Examining at all three ratios will provide some additional energy dependence information helpful in checking some assumptions.
6. Compare these ratios to those determined from computer simulations of a similar environment and extract the ratio of Tc-99 to U-235.
7. Multiply this ratio by the enrichment and the thickness of the hold to determine the "thickness" of the Tc-99 contamination in grams per square centimeter.

This approach emphasizes the use of ratios to reduce systematic uncertainties when comparing the data and computer simulations. The bulk of the effort in the successful execution of these procedures occurs in step 6, the computer simulations of the measurement.

To validate the above approach, a series of laboratory measurements was conducted at the Pacific Northwest Division of Battelle (PNWD). Most of the measurements were conducted with a HPGe detector. In order to separate the contributions from the uranium and bremsstrahlung products, accurate measurements of the gammas lines of the various uranium isotopes will be required. Many of the gamma lines for U-235 are closely packed together. The need for high resolution to resolve these lines motivated the use of HPGe detectors

APPROACH

Measurements of realistic Portsmouth scenarios in the laboratory were not possible due to financial, temporal, and regulatory restrictions. Rather, the approach adopted was to conduct key benchmarking measurements in the laboratory that provided critical information on the processes involved, then to extend our understanding of these processes through computer simulations to scenarios related to Portsmouth. With this approach, the procedure for developing the methodology can be divided into two phases:

1. Benchmarking computer simulations with key measurements. Through this process, we can determine a level of confidence of the computer simulations in describing the measurements to be conducted at Portsmouth.
2. Systematically studying the variables for realistic scenarios via computer simulations. With systematic studies, we can determine the uncertainties of the methodology for the characterizing Tc-99 in situations relevant to Portsmouth.

The computer simulations were conducted using the Monte Carlo N-Particle (MCNP) Version 5 standard package developed at Los Alamos National Laboratory and distributed by the Radiation Safety Information Computational Center for describing low-energy particle interactions 4. It is widely used throughout the nuclear physics community.

As an initial effort, a short study was conducted to determine the sensitivity to the type of detector. Various sizes of NaI scintillating detectors as well as two HPGe geometries, coaxial and planar, were included in the study. For the laboratory measurements, it was found that the smallest NaI detector provided the best signal-to-noise, while the planar HPGe detector was considerably better because it was less susceptible to the high-energy background of the laboratory. The laboratory measurements were conducted with the coaxial HPGe detector because it provided nearly the same count rate as the NaI, resolution capable of distinguishing the x-ray structure of high-Z materials and was easier to operate than the planar HPGe detector, which required its dewar to be refilled a few times each day.

The primary Tc-99 source used in the project was constructed at the Radiochemical Processing Laboratory (RPL) at PNWD. It consisted of 2.1 mg of Tc-99 electroplated onto a steel disk, which was enclosed in a cardboard "microscope slide" type-holder. The steel disk sat in a recessed circular cavity of the slide. The disk was covered by a thin Mylar film, ~3 µm thick. The Mylar film did not significantly attenuate the Tc-99 betas because the continuous slowing-down approximation range of a 100-keV electron in Mylar is 70 µm. The effective activities of the RPL sources were determined by comparing the bremsstrahlung spectrum generated by these sources to that generated by the National Institute of Standards & Technology-traceable 1.005-µCi source from Isotope Products. The normalization of RPL Tc-99 source was determined by taking the ratio of the sums over 25 to 70 keV of the two spectra, yielding an activity of 32.7 ± 0.6 µCi.

Benchmarking

To develop a quantitative understanding of the Tc-99 bremsstrahlung signature at Portsmouth, two distinct physical processes were studied: the creation of the bremsstrahlung photons and the transport of those photons through various materials until they reach a detector. As part of the decay of the parent nucleus, a beta particle is emitted. This beta will not travel very far before it is stopped, emitting bremsstrahlung as it slows down. On rare occasions, that bremsstrahlung photon will be energetic enough to be observed in a detector. In order for those bremsstrahlung photons to be observed, they must pass through more material, such as additional deposit thicknesses and pipe walls. In this additional material, the photons can pass through without changing, can be completely absorbed, or can "down-scatter," emitting one to several photons with less energy than the original photon. Benchmarking measurements of both processes of bremsstrahlung production and photon transport were conducted on a range of materials relevant to the measurements at Portsmouth.

The production of bremsstrahlung was tested by measuring the photon spectrum generated by a Tc-99 source on the other side of a thin absorber from a coaxial HPGe detector. Aluminum, copper, and lead were used as absorbers. Aluminum and copper were chosen because they are directly relevant to applications at Portsmouth where this methodology is likely to succeed. Lead was chosen as a readily accessible non-radiating high atomic number (high Z) material. The absorbers must be thin to minimize the absorption of the bremsstrahlung photons. The properties of the absorbers are shown in Table I[1]. For aluminum and copper, the thicknesses were several times the range of a mean energy electron from Tc-99 decay while also much less than the mean free path of photons in that material. These thicknesses ensured that all the beta particles were stopped in the material and that few of the resulting bremsstrahlung photons were absorbed. The lead thickness was based on availability and cost.

The measurements were conducted with a Canberra GR4520 REGe (n-type) HPGe detector. The absorbers were placed immediately in front of the detector, while the 35.6 µCi Tc-99 source was placed 2 centimeters (cm) from the face of the detector. Measurements were conducted for one-half hour of live time, and background measurements (without the Tc-99 source) were conducted for each absorber.

Table I. Properties of Absorbers used for Bremsstrahlung Production Benchmarking

Material	Thickness (cm)	Thickness (g/cm^2)	Electron CSDA Range (g/cm^2)	Photon Mean Free Path (g/cm^2)
Aluminum	0.051	0.14	0.019	5.9
Copper	0.102	0.91	0.022	2.2
Lead	0.046	0.52	0.031	0.18

[1] The CSDA range is the range of the electron in the continuous-slowing-down approximation. The electron CSDA range and photon mean free path are given for particles with 100 keV kinetic energy.

Comparisons of the bremsstrahlung production measurements and MCNP simulations for aluminum and lead are shown in Figs. 1A and 1B. The MCNP results have been normalized to the effective yield of the fabricated 35.6-μCi Tc-99 source; these plots are an absolute comparison of data and MCNP. The agreement ranges from fair for the aluminum to good for the lead. For Portsmouth, the agreement for the high Z materials is important, as the bremsstrahlung production will occur mostly in the uranium in the deposit. The sharp peak at 20 keV in the aluminum-measured spectrum is related to the Tc-99 source. It is only visible in the aluminum spectra because it is absorbed by the other heavier materials.

A numerical comparison of the measured and simulated spectra is provided in Table II. The sum of events over the range of 50 to 200 keV was determined for both the measured and simulated spectra. This range of energies is relevant to the methodology for characterizing Tc-99. The ratio of these sums is shown in the table. While the agreement improves for the highest Z material (lead), the one most relevant to bremsstrahlung production at Portsmouth, the overall average was taken. It is important to note that the correction has the potential to be experimental setup-dependent (e.g., a different detector may have a different normalization).

The MCNP modeling of the transport of photons through various materials was also benchmarked against measurements. As the relevant range of gamma energies closely matches the spectra of highly enriched

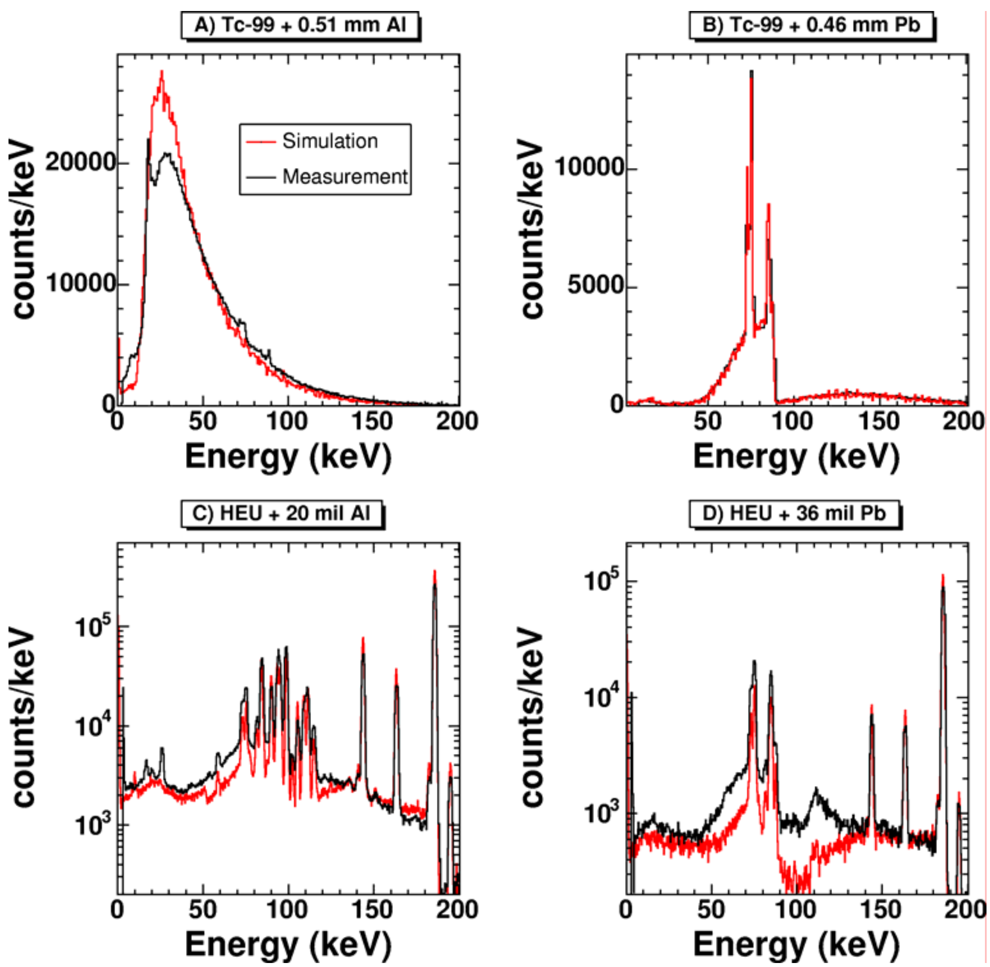

Fig. 1. Histograms comparing the bremsstrahlung production of a Tc-99 source behind aluminum and lead, A and B. Comparison of measured and simulated spectra to study photon absorption in various materials, C and D

Table II. Ratio Measurement to MCNP of Bremsstrahlung Production in Various Materials

Material	**Sum Measured/Sum MCNP**
Aluminum	1.26
Copper	1.42
Lead	1.12
Average	1.27 ± 0.15

uranium (HEU), a small disk of 99% enriched HEU (~0.2 grams of U-235) was used as a gamma source. This HEU was encased in thin layers of a metal, most likely aluminum. The Canberra HPGe detector detected the photons that were transmitted from the HEU disk through various materials and absorbers. Two thicknesses of aluminum, copper, and lead were used as absorbers, as seen in Table III. The thicknesses of aluminum and copper roughly span the range of thicknesses relevant to Portsmouth. The lead was a surrogate for uranium without the added complications of the additional γ-ray emissions of uranium.

As a first check, the spectra of only the HEU (no absorbers) from measurement and simulations were compared. The overall normalization of the simulations was determined by matching the counts in the 186 keV peak of U-235 because the mass of the HEU sample was not well known. The comparison between simulation and data is reasonably good except that the continuum strength is under-predicted below the 144 keV peak for all absorbers. The strength in the continuum is a result of down-scatter (the scattering of photons resulting in the lowering of the initial photon momentum) in the experimental setup and only partial energy deposited in the detector from incident photons. Examples of these comparisons are shown in Figs. 1C and D.

Possible sources of additional continuum strength were investigated in both the simulations and the experimental setup. Extra material in the experimental setup was minimized. The impact of thicker-than-specified detector windows and source-encasing material was also studied. None of these possibilities significantly decreased the difference in the continuum strength between measurement and simulations. The discontinuity of the continuum strength at the 186 keV peak suggests that the strength is a result of 186 keV gammas down-scattering. The observation of this discontinuity eliminates the possibility that U-232 and U-238, which were not included in the MCNP simulation, might explain the missing strength because they would contribute equally above and below the 186 keV peak.

The accelerated schedule of this task prevented further investigations into the origin of the difference in

Table III. Properties of Absorbers Used for the Photon Transport Benchmarking

<table>
<tr><th>Material</th><th>Thickness
(cm)</th><th>Thickness
(g/cm^2)</th><th>Photon Average Mean Free Path over 70-120 keV
(g/cm^2)</th><th>Photon Mean Free Path at 200 keV
(g/cm^2)</th></tr>
<tr><td rowspan="2">Aluminum</td><td>0.051</td><td>0.14</td><td rowspan="2">5.52</td><td rowspan="2">8.2</td></tr>
<tr><td>0.254</td><td>0.69</td></tr>
<tr><td rowspan="2">Copper</td><td>0.102</td><td>0.91</td><td rowspan="2">1.77</td><td rowspan="2">6.4</td></tr>
<tr><td>0.264</td><td>2.37</td></tr>
<tr><td rowspan="2">Lead</td><td>0.046</td><td>0.52</td><td rowspan="2">0.24</td><td rowspan="2">1.0</td></tr>
<tr><td>0.091</td><td>1.03</td></tr>
</table>

the continuum strength. Instead, the methodology was adjusted to account for this slight deficiency by allowing for a correction to the simulated results based on the benchmarking studies. The quantity to be determined is the ratio of the strength in the region in which there will be a significant bremsstrahlung signal to the strength of the 186 keV peak of U-235,

$$\rho = \Sigma_{brems} \;/\, \Sigma_{186}\,, \qquad \text{(Eq. 1)}$$

where the bremsstrahlung region is the energy range of 70 to 115 keV. A significant fraction of the bremsstrahlung signal of Tc-99 through uranium will be in this energy range. The use of ratios eliminates some of the normalization effects. The comparison of this ratio for simulations and for measurements will provide information on the reliability of the simulations and whether any corrections are necessary.

A list of simulated and measured ratios for the gamma transport studies using the HEU source is provided in Table IV. The super ratio ρ_{meas}/ρ_{sim} is remarkably consistent for the aluminum and copper absorbers, varying less than 1.5%, even though the individual ratios for the simulations and measurements vary by more than 70%.

From these results, we assign a one-standard deviation uncertainty of 1.5% to the description by MCNP of the effect of the photon transport through the pipe walls on ρ. The impact of the lead absorbers on the ratio ρ, however, is more disconcerting. The increase in the super ratio ρ_{meas}/ρ_{sim} indicates that the MCNP model is predicting too much strength in the bremsstrahlung region compared to the 186 keV peak. In Fig. 1D, it can be observed that while MCNP under-predicts the continuum strength in the bremsstrahlung region, it over-predicts the strength of the lead x-rays.

Lead is special in a number of ways. First, the K-edge for lead is at 88 keV, in the middle of the bremsstrahlung region. Thus, one can expect significant discontinuities over this region. In addition, the mean free path in lead (0.24 g/cm^2) is shorter than in uranium (0.36 g/cm^2). The photoelectric absorption for lead is considerably higher than for uranium, despite its lower atomic number. This difference is caused by the electron shell structure.

The results of the photon transport through lead are indicative of how MCNP will describe the photon transport through uranium. Understanding how well MCNP describes the photon transport through lead is critical in assessing the uncertainties in the methodology for different deposit thicknesses. The diverging differences between the MCNP results for ρ and the measured results for ρ suggest that a correction should be applied to the MCNP results for the predicted bremsstrahlung strength relative to the 186 keV peak. To determine the correction, we assume the following:

- The correction depends on the number of mean free paths length, *l*, of uranium the photon must pass through before reaching the detector. It was observed that the strengths in the bremsstrahlung region for the simulations and measurements diverge, whereas they are consistent for the 186 keV peak. For this reason, the average mean free path of photons in the

Table IV. Ratio of Strength in Bremsstrahlung Region to 186 keV Peak for the Photon Transport Study for Simulated and Measured Results

Absorber	No. of mean free path lengths	ρ_{sim}	ρ_{meas}	ρ_{meas}/ρ_{sim}
None	0.00	1.06	1.50	1.42
0.5 mm aluminum	0.03	1.06	1.51	1.42
2.5 mm aluminum	0.13	1.09	1.56	1.44
1.0 mm copper	0.51	0.85	1.20	1.42
2.5 mm copper	1.34	0.62	0.89	1.43
0.5 mm lead	2.17	0.56	1.05	1.87
1.0 mm lead	4.29	0.48	0.92	2.03

bremsstrahlung region, 70 to 115 keV, is used.

- The average mean free path over the 70 to 120 keV will be used.
- The correction to ρ_{sim} for a photon at depth l is linear in l, with the line described by the results of the super ratio ρ_{sim}/ρ_{meas} for no absorber and for the 1.0-mm lead absorber (i.e., the correction $D(l) = D_0 + \delta l$, where $D_0 = 1.42$ and $\delta = 0.14$).
- The uncertainty in the correction will vary, starting at 5% for $l = 0$ and 50% for $l = 4.29$. These uncertainties are large but appropriate considering that we do not have any uranium measurements. The relative uncertainty is parameterized as $\varepsilon(l) = \varepsilon_0 + \phi l$, where $\varepsilon_0 = 0.05$ and $\phi = 0.105$.
- The strength of the bremsstrahlung signal falls off exponentially. The scale of the falloff is determined by the decrease in the measured strength of the bremsstrahlung region for the 1-mm lead absorber, relative strength $\omega(l) = e{-}\alpha l$, where $\alpha = 0.38$.

The correction and uncertainty as specified above are for a photon originating at a given depth. The correction for a deposit of thickness l_0 must be averaged over all thicknesses up to l_0, weighted by the strength of the bremsstrahlung response at that depth. The correction to the simulated ratio ρ is for a deposit l thick is

$$\begin{aligned} T(l) &= \int_0^l \omega(t)D(t)dt \Big/ \int_0^l \omega(t)dt, \\ &= D_0 + \frac{\delta}{\alpha}\left(1 - \frac{\alpha l}{e^{\alpha l} - 1}\right), \end{aligned} \qquad \text{(Eq. 2)}$$

where the uncertainty is given by

$$\begin{aligned} \delta T(l) &= \int_0^l \omega(t)\varepsilon(t)D(t)dt \Big/ \int_0^l \omega(t)dt, \\ &= D_0\varepsilon_0 + \frac{D_0\phi + \varepsilon_0\delta}{\alpha}\left(1 - \frac{\alpha l}{e^{\alpha l} - 1}\right) + \frac{\phi\delta}{\alpha^2}\left(2 - \frac{\alpha l(\alpha l + 2)}{(e^{\alpha l} - 1)}\right). \end{aligned} \qquad \text{(Eq. 3)}$$

The dependence of the correction and its uncertainty on the mean free path has some interesting properties. Both the correction and its uncertainty have an asymptotic limit despite the unrestricted growth of the relative uncertainty of $D(l)$. This limit is reached because the regions in which the uncertainties are large only contribute a small amount to the overall uncertainty of the correction.

Additional benchmarking was done to check the sensitivity to the separation distance between the source and detector. A HEU source was placed at 2, 3, 4, 6, and 9 cm from the front face of an HPGe detector. The variation of the super ratio ρ_{sim}/ρ_{meas} was 1.2%. This variation is consistent with the reproducibility of the distance of the source from the detector. Unfortunately, these measurements were conducted with a different detector from the one used for the photon transport measurements, so that it is not possible to make a meaningful comparison of the average of these ratios.

The primary purpose of the benchmark studies was to determine the reliability of models used in the computer simulation. Table V summarizes the observed corrections and their uncertainties. The uncertainties were chosen to be conservative. For deposits with thickness roughly less than 1 mm (assuming a deposit density of about 6 grams per cubic centimeter [g/cm^3]), the uncertainty of the bremsstrahlung will be the leading uncertainty. For thicker deposits, the uncertainty of the absorption of the photons in the bremsstrahlung region (70 to 115 keV) through the uranium deposits will be the leading uncertainty.

Systematic Studies

The objective of the systematic studies was to provide information on the uncertainty of the methodology related to the deposit thickness and enrichment, two variables that could not be controlled in the laboratory measurements. In addition, the simulations allowed a study of the performance when the Tc-99 is uniformly mixed with the uranium deposit, as it would be at Portsmouth, rather than layered above the radiator as could be done in the laboratory.

The geometry used in the simulations was more closely related to the actual measurements at Portsmouth. For each simulated measurement, it was assumed that the uranium deposit was behind a metal sheet, either aluminum, copper, or steel. The thickness of the wall varied. The diameter of the deposit is three times the diameter of the detector to approximate a large deposit. The deposit is assumed to be solid uranium. The mixture of other, lower-atomic-number elements will have little impact on the bremsstrahlung production or photon absorption in the deposit. These processes are also largely independent of the isotopic nature of the deposit, so that it was assumed that the deposit was U-235. Rather than work with the linear thickness of the deposit, the areal thickness in grams per square centimeter was used. The areal thickness is the deposit density times the deposit thickness, and it is the relevant quantity for describing the photon absorption in the material. No collimation is assumed in the computer model.

The studies involved a large collection of MCNP simulated results. Simulations were separately run for U-232, U-235, U-238, and Tc-99. The uranium spectra thrown in the simulations were generated by SuperSynth 5 assuming a 30-year grow-in time. The spectra for U-235 and U-238 are largely independent of the grow-in time because the half-life of the parent is extremely long, while the daughters have half-lives less than a month, until they reached another long-lived (greater than 10,000-year half-life) daughter. The spectrum of U-232 is fairly sensitive to the grow-in because it has a half-life of 68.9 years. The daughters in the decay chain of U-232 are mostly short-lived, with the exception of the Th-228, which has a half-life of 1.9 years. Thus, the primary result of altering the grow-in time for U-232 beyond 10 years is a change in the overall intensity of the entire spectrum. Because the overall contamination of U-232 is not known and will have to be determined through an examination of the measured spectrum, the uncertainty in the grow-in time for U-232 is not important. The 30-year grow-in period for U-232 was selected as a reasonable value based on operating history at Portsmouth.

The statistics of the Tc-99 simulations were limited. To run these simulations, it was necessary to run MCNP in mode "PE", which calculates processes for both photons and electrons. This mode is considerably more time-consuming than the mode "P" used for the photon simulations. In addition, the bremsstrahlung production process is very inefficient, with about 1 out of 10^4 to 10^5 electrons creating a bremsstrahlung photon. While a typical uranium simulation would take a few hours, a Tc-99 simulation

Table V. Summary of Corrections and Uncertainties for the MCNP Model

	Correction	Uncertainty of Correction
Bremsstrahlung production	1.27	11.8%
Photon absorption in pipe	1	1.2%
Photon absorption in deposit	$T(l)$	5%< $\delta T(l)$ < 39%
Uncertainty in enrichment	1	7.3% E<5% 3.6% E>5%
^{232}U contribution	1	2.4% E<5% 1.0% E>5%

would require 50 hours on a 3-gigahertz (GHz) Pentium-4 machine, and the Tc-99 simulations would have considerably fewer counts than those for uranium.

An example of the effect of Tc-99 contamination within a deposit on the gamma spectra is shown in Fig. 2. This figure shows the simulated spectra for a deposit with and without a 0.01 atoms of Tc-99 per atoms of uranium. The Tc-99 bremsstrahlung signal is quite clear, and it has a noticeable impact below the 186 keV peak from U-235. In addition, a significant fraction of the bremsstrahlung lies in the region below 70 keV, suggesting that additional sensitivity may be obtained by increasing the bremsstrahlung region in the analysis to lower energies. The energy distribution below 70 keV is fairly sensitive to the wall material and thickness.

The uncertainties in general depend on the uranium enrichment of the deposit. For the purposes of assigning uncertainties, two regions of enrichment were considered: 0.7% to 5.0% and 5.0% to 100%. For simplicity, the uncertainties are assumed to be constant over each of these regions and are equal to the largest uncertainty in that region. In general, the uncertainties are largest for the smallest enrichment.

The first issue examined was the impact of the deposit thickness. This impact will vary with the photon energy of interest; for photons in the range of energies relevant to this methodology, more energetic photons are attenuated less. The difference in attenuation lengths for the bremsstrahlung region and for the 186 keV peak will lead to different depths in the deposit for which the measurements will be sensitive.

The impact of the attenuation of photons on the ratio ρ is shown in Fig. 3. The ratio ρ approaches the asymptotic limit faster than the strength of the 186 keV peak because of the higher attenuation for the photons in the bremsstrahlung region. Also shown in this figure are plots of ρ as a function of enrichments for three different deposit thicknesses on the inside of a 1.3-mm aluminum wall. Because

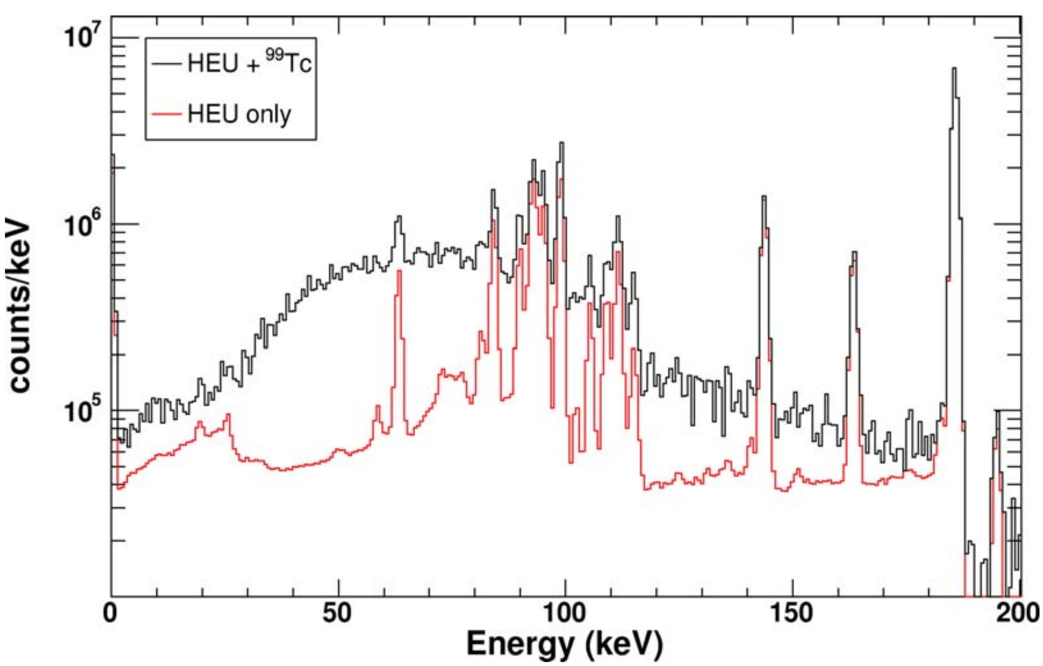

Fig. 2. Simulated spectra showing the effect of Tc-99 in the uranium deposit. This deposit used in the simulation is 5 mm thick with a 10% enriched uranium and a 50 ppt U-232 contamination. A 2.5-mm-thick aluminum wall separates the deposit and the detector. The red line is for the HEU signal alone, while the black line is for the HEU + Tc-99 bremsstrahlung signal for a 0.01 atoms of Tc-99 per atom of uranium contamination.

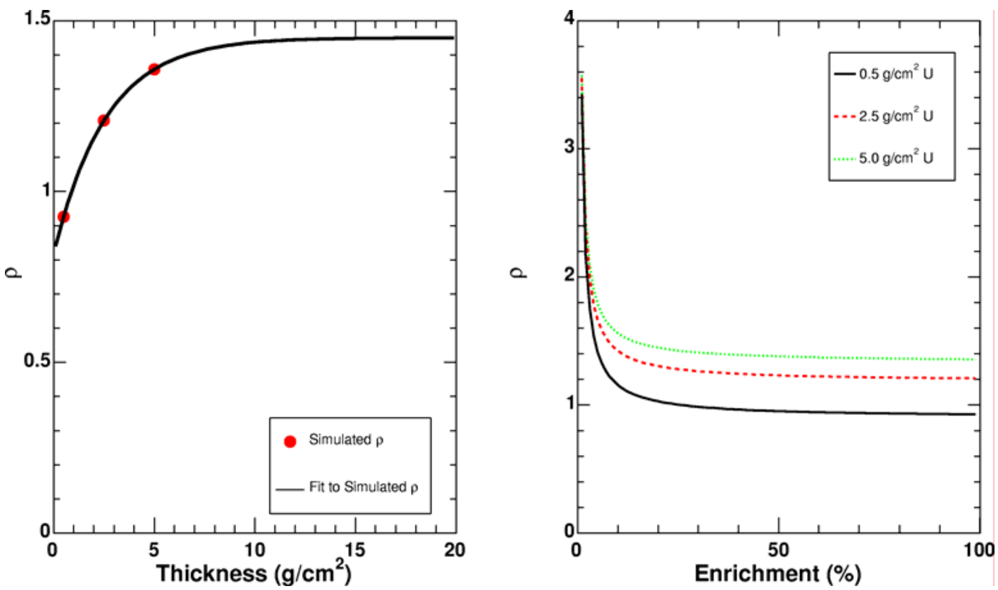

Fig. 3. Ratio of strengths in bremsstrahlung region to peak as a function of deposit thickness (left) and enrichment (right). For the dependence on the enrichment, three deposit thicknesses are shown. These results are from simulations and do not include any contributions from Tc-99.

most of the strength in both the bremsstrahlung region and the 186 keV peak comes from U-235, ρ is fairly constant until the enrichment falls below 10%, at which point the contributions from U-238 become significant.

The other issue examined was the impact of the enrichment of the deposit. As the enrichment varies, the aspects of the observed photon spectra will vary: the strength of the 186 keV peak, the strength of the gamma peaks of U-235 in the bremsstrahlung region, and the strength of the x-rays in the bremsstrahlung, both daughter- and parent-related. The contributions from U-232 and U-238 will be small compared to those from U-235 unless the enrichment is very low.

The ratio ρ is not very sensitive to the level of U-232 contamination. The typical U-232 level of contamination is 50 to 100 ppt of uranium. Despite this low level of contamination, the significantly shorter half-life of U-232 makes the activity due to U-232 in the range of a few percent of the more common uranium isotopes. For 0.7% enrichment, 50 ppt of U-232 contribute 2.1% of the strength in the bremsstrahlung region for a 0.5 g/cm^2 thick deposit and 2.4% for a 5.0 g/cm^2 thick deposit. At 5% enrichment, these values change to 1.0% and 0.9% for the 0.5 g/cm^2 and 5.0 g/cm^2 thick deposit, respectively. It is assumed that the U-232 contamination is known to the level of 50 ppt, and a thickness-independent uncertainty of 2.4% is assigned to the low enrichment region and 1.0% to the high enrichment region. Note that it would be fairly straightforward to measure the U-232 contributions directly with the 238 keV gamma line from its decay chain.

The sensitivity to the enrichment is larger. It is assumed that the enrichment is known to 10% relative uncertainty. For the 0.5 g/cm^2 thick deposit, the ratio ρ varied by 7.3% at 1% enrichment and 3.6% at 5% enrichment. As the enrichment increases, the variation in ρ decreases. For thicker deposits, these variations also decrease. Uncertainties of 7.3% and 3.6% are used for the low- and high-enrichment regions due to the 10% uncertainty in the level of enrichment.

The studies of the sensitivities of ρ to U-232 and enrichment were performed assuming no bremsstrahlung contributions from Tc-99. This approach is appropriate for determining minimum detectable levels of contamination. The inclusion of strength from bremsstrahlung from Tc-99 would simply reduce these relative uncertainties further because the contributions from the bremsstrahlung would dilute the effects of the variations. This reduction in uncertainties is not addressed in the establishing the uncertainties of the methodology.

RESULTS

The results of experiments used to measure Tc-99 using bremsstrahlung radiation are presented in this section. The minimum detectable level of Tc-99 contamination, measurement uncertainties, and worker safety are discussed.

Minimum Detectable Contamination

The minimum detectable contamination (MDC) of Tc-99 in the uranium deposit is based on a modified version of the Currie equation 6. The Currie equation assumes that the signal is extracted from two measurements, a background and a background plus signal measurement, which have approximately equal uncertainties. The Tc-99 characterization methodology requires only one measurement and a comparison with simulations. The uncertainties of the simulations have already been accounted for. For these reasons, the MDC is reduced by a factor of $\sqrt{2}$. To claim detection of Tc-99 at the 95% confidence level, the ρ_{meas} must be larger than ρ_{sim} for only uranium by $2 \times 1.645\sigma_\rho$, where σ_ρ is the uncertainty in ρ.

Plots of the MDC for various wall materials, deposit thicknesses, and wall thicknesses are shown in Fig. 4. Because the contributions for U-235 dominate the bremsstrahlung region and the 186 keV peak for most of the range of enrichments, the MDC curves are fairly linear. The MDC increases as the enrichment increases because those from U-235 dominate the contributions in the bremsstrahlung region; a correspondingly larger signal from the Tc-99 is required in order to achieve the MDC level. In the evaluation of the MDC, it was assumed that statistical uncertainties are very small compared to the systematic uncertainties, so they are ignored. This assumption is valid for long measurements, such as for 30 minutes, even for the 6.4-mm (0.25") iron pipe wall.

While the focus of this project has been on comparing the strength of the bremsstrahlung region to that of the 186 keV peak, there are other U-235 peaks to which a similar comparison can be made. This feature is important because it will provide a means to test for extremely large Tc-99 signals. The Tc-99 bremsstrahlung contributions will provide a strongly energy-dependent contribution to the measured spectra. For large Tc-99 contaminations, this will create observable energy dependence in the ratios of bremsstrahlung to peak strength, well beyond that expected simply from the difference in the strengths of the gamma peaks from bare HEU.

Measurement Uncertainties

The uncertainty in the characterization of the level of contamination of Tc-99 in the uranium deposit, in atoms of Tc-99 to atoms of uranium, is dependent on the uncertainties listed in Table V. To achieve a 95% confidence level in the characterization, the standard deviations of the uncertainties in this table must be multiplied by a factor of two. Thus, the measurement uncertainty for the Tc-99 contamination ranges from 27% for very thin deposits up to 82% for very thick deposits.

Worker Safety

The derived air concentration (DAC) is often used to measure the hazard of a radionuclide for workers. The DAC is defined as the "concentration of a given radionuclide in air which, if breathed by the reference man for a working year of 2,000 hours under conditions of light work (inhalation rate 1.2 cubic meters of air per hour), results in an intake of one ALI." An annual limit on intake (ALI) is the smaller value of intake that would result in a committed effective dose equivalent of 5 rem or a committed dose equivalent of 50 rem to any individual organ or tissue. In order to determine if the Tc-99 bremsstrahlung

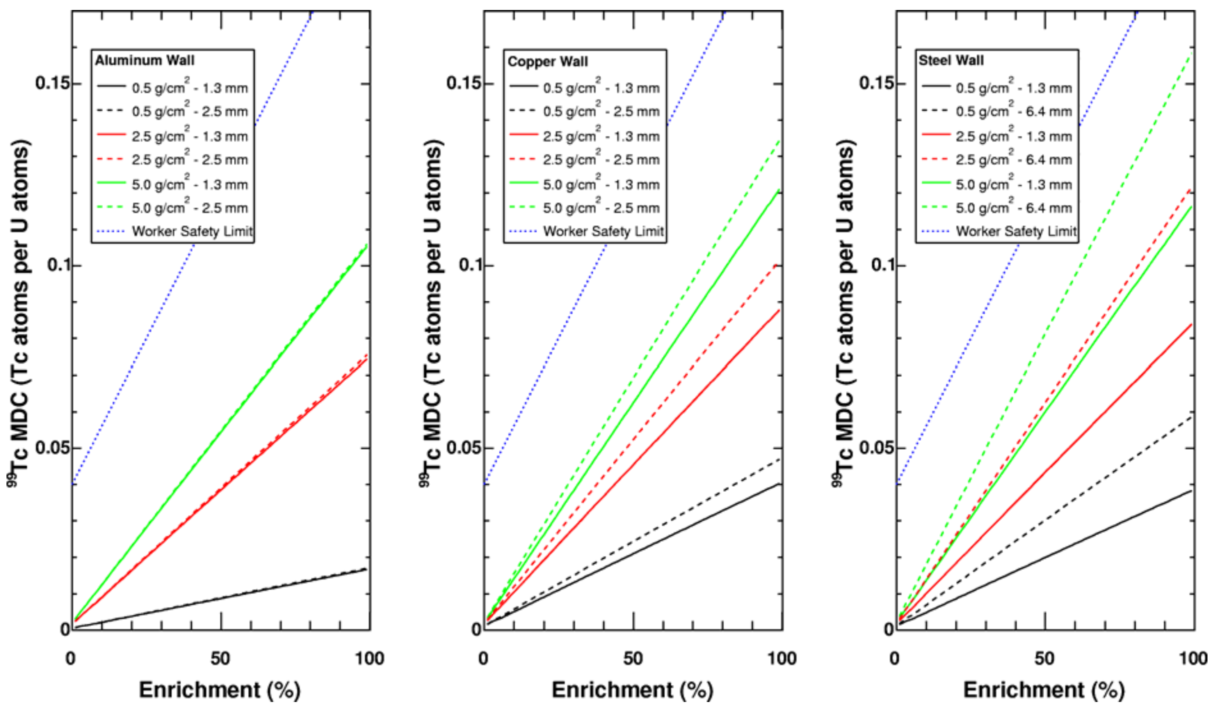

Fig. 4. Plots of MDC as a function of enrichment for different wall materials, wall thicknesses, and deposit thicknesses. From left to right, the three plots are for aluminum, copper, and steel (iron) pipe walls. The three different line colors represent different uranium deposit thicknesses, while the solid and dashed lines denote the thin- and thick-walled pipe. The dotted blue line represents the worker safety limit discussed in Section 4.3. In the legends, the first number is the uranium deposit thickness and the second number is the wall thickness.

measurement technique was sensitive enough for worker protection purposes, the ratios of the DACs for Tc-99, U-235, and U-238 were examined (Table VI) 7. For these ratios, the Tc-99 and uranium would equally contribute to the inhalation dose. The required level of sensitivity to the concentration of Tc-99 in the uranium deposit, if only Tc-99 and uranium contributed to the inhalation dose, assumed to be a concentration such that Tc-99 contributed only 20% of the total dose. This level of sensitivity is achieved by dividing the ratios in Table VII by a factor of four. For 100% enrichment, the required sensitivity is 1 atom of Tc-99 per 4 atoms of uranium, while for DU it is 1 atom of Tc-99 per 25 atoms of uranium. These required sensitivities are plotted in Fig. 4, along with the MDC for several deposit thicknesses and pipe wall types. From this figure, it is clear that the sensitivity of the Tc-99 characterization technique is adequate for worker protection purposes

CONCLUSION

A methodology to characterize the Tc-99 inside the equipment at the Portsmouth Gaseous Diffusion Plant was developed. The approach uses the bremsstrahlung radiation generated when emitted beta particles of Tc-99 decay are stopped in the surrounding material. The quantity used in the characterization is the ratio of the number of detected photons with energies between 70 and 115 keV, a range in which the bremsstrahlung contribution will be large, to the number of photons detected in the 186 keV peak of U-235. The observed ratio is compared to the ratio generated from computer simulations to quantify the technetium contamination in the deposit. The investigation of this methodology has reached the following conclusions:

Table VI. DAC Ratios for Tc-99, U-235, and U-238

Ratio	Activity Ratio	Atom Ratio
Tc-99:U-235	3,300	1.0
Tc-99:U-238	3,300	0.16

- The bremsstrahlung generated from the beta particles from the Tc-99 decay provides an observable signal that can be used to characterize the level of contamination of Tc-99 in a uranium deposit.
- The sensitivity of the approach is adequate to address worker safety issues.
- The sensitivity of the approach is higher for lower levels of enrichment.
- The sensitivity of the approach depends on the deposit thickness, wall material, and wall thickness.

The laboratory investigation of this methodology has assumed that the technetium contamination will be uniformly distributed throughout the uranium deposit. This assumption is reasonable if the migration of the Tc-99 through the equipment at Portsmouth is very slow, on the order of a year. If this assumption is not reasonable, then it is conceivable that a careful examination of the K-edge in the photon distribution would enable characterization of a layer of technetium within the uranium deposit. The K-edge is observed in measurements in which the uranium and technetium are distributed in layers, such as the benchmarking measurements. The edge disappears from the observed bremsstrahlung spectra when the technetium contamination is distributed uniformly throughout the deposit.

Improvements to the sensitivity of the characterization are possible. The leading limitations to the uncertainties are related to reliability of the computer simulations in describing the relevant physical processes. These reliabilities were established by comparing the simulations to several critical benchmarking measurements. Minor improvements in the computer simulations and additional measurements on various thicknesses of U-235 could improve the sensitivities by factors of 2 to 5. Because these processes are all electromagnetic and nuclear in nature, additional non-trivial refinements of the computer simulations could significantly improve the sensitivity further.

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