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IN VIVO CALIBRATION MEASUREMENTS FOR MIXED OXIDE NUCLEAR REACTOR FUEL

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ABSTRACT

The introduction of mixed oxide (MOX) nuclear fuel containing enriched uranium and isotopes of plutonium presents a challenge for monitoring occupational exposure using direct *in vivo* measurements because photons from ^{235}U and ^{241}Am will confound evaluation of plutonium in the lungs. MOX is a unique blend of enriched uranium and plutonium that can be used in place of traditional enriched uranium in commercial pressurized water reactors. The plutonium in MOX fuel may be derived from either surplus weapons-grade plutonium or reprocessed commercial nuclear fuel. Thus, the final isotopic composition of MOX depends upon the origin of the plutonium, how well chemical and radiological impurities are removed from the plutonium prior to fuel fabrication, the time between fuel fabrication and use, and (for spent fuel) the total time of irradiation in the reactor. All of these factors will also have a significant influence on the risk associated with occupational exposure to MOX and the optimum procedure for detecting MOX *in vivo*. Calibration of an array of high resolution detectors to perform *in vivo* measurements for MOX in the lungs was performed using the Lawrence Livermore thoracic phantom and a set of lungs containing a homogeneously distributed mixture of ^{238}U, ^{235}U, ^{241}Am, and ^{238}Pu to simulate conditions that might arise following inhalation of a typical MOX aerosol. Activity associated with photons from ^{235}U and ^{241}Am were easy to quantify. Low energy x-rays associated with plutonium decay were detected with reasonable certainty for a chest wall thickness below 3 cm after applying spectrum deconvolution to remove the interference from higher energy photons associated with ^{235}U and ^{241}Am. Analysis must depend upon low abundant, higher energy plutonium photo peaks whenever chest wall thickness exceeds 3 cm because the low energy uranium L x-rays are greatly attenuated by overlying muscle tissue in the anterior thorax.

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INTRODUCTION:

Background:

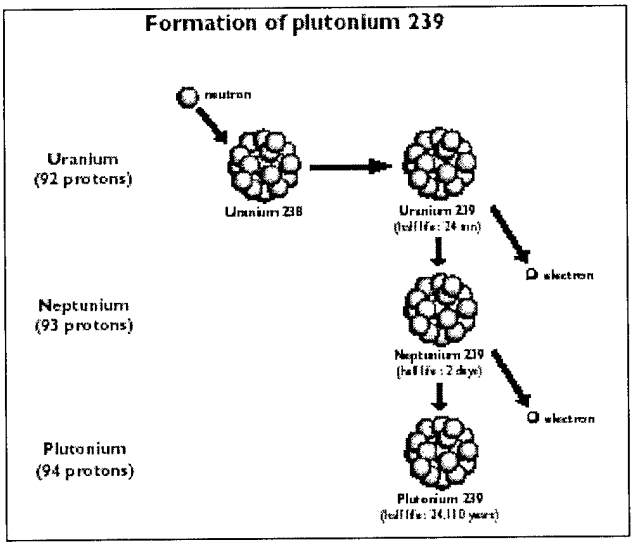
In 1993, the *Non-Proliferation and Export Control Policy* was issued by President Clinton due to the increased threat of nuclear weapons proliferation. In conjunction, Russia's President Yeltsin and President Clinton issued a shared statement between the United States (US) and Russia on *Non-Proliferation of Weapons of Mass Destruction and Their Means of Delivery* in 1994. The focus of the joint efforts was to ensure safe, secure, long-term storage and disposition of surplus weapons grade plutonium and enriched uranium. In 1998, both countries signed a 5-year agreement to provide a technical basis for how to manage the weapons grade isotopes with the intention of removing 50 tons of plutonium from each countries stockpile (62 FR 28009). The Department of Energy (DOE) published *Record of Decision for the Surplus Plutonium Disposition Final Environmental Impact Statement* (62 FR 28009) in 2000, which pursued both the immobilization and mixed oxide (MOX), fuel approaches to contend with the surplus.

MOX fuel is a combination of plutonium oxide and uranium oxide used in nuclear reactor fuel rods. The DOE decided to pursue the use of MOX fuel in an attempt to dispose of approximately 33 of the 50 total tons of excess weapons grade plutonium (^{239}Pu) that was produced during the *Cold War* era at the DOE Hanford and Savannah River facilities. The fundamental purpose of the MOX implementation is to ensure that

the plutonium, which was produced for nuclear weapons and declared excess to national security needs now and in the future, is never again used for nuclear weapons (62 FR 28009). The plutonium will essentially become unrecoverable after it completes a once-through reactor fuel cycle as opposed to raw plutonium, which can immediately be put to use in a crude, dirty or nuclear weapon. MOX usage will be a deterrent for theft of surplus raw plutonium oxide (PuO_2).

^{239}Pu was created in nuclear reactors at the facilities mentioned above from 1944 - 1986. The formation of plutonium starts with uranium-238 (^{238}U) absorbing a neutron from fission products to form uranium-239 (^{239}U). ^{239}U has a half-life a 24.4 minutes and β- decays into neptunium-239 (^{239}Np). ^{239}Np has a half-life of 2.35 days and also β- decays to form ^{239}Pu. The optimal irradiation of the uranium fuel to produce the desirable ^{239}Pu was approximately 3 months. Other isotopes of plutonium, namely plutonium-240 (^{240}Pu) and plutonium-241 (^{241}Pu) were also produced by absorption of fission neutrons (UIC). Figure 1 illustrates the production of ^{239}Pu (Cogema. Plutonium).

Figure 1: ^{239}Pu Production



Plutonium is separated from spent fuel by the PUREX process as a nitrate through a series of mechanical and chemical processing operations. It is then converted into oxide (PuO_2) chemically nicknamed green cake.

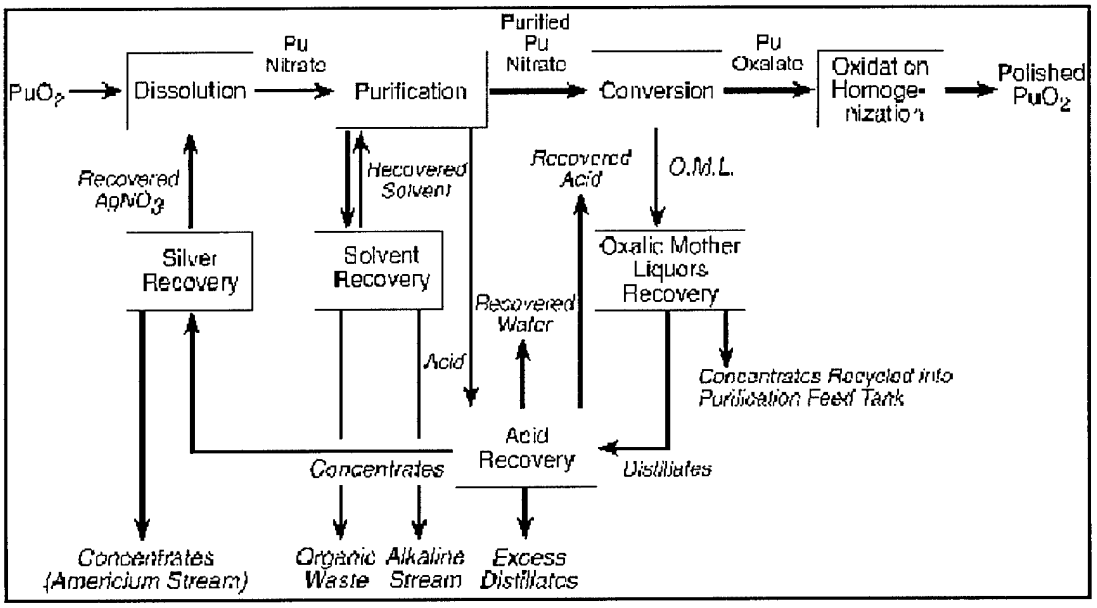
In 1999, the DOE awarded a contract to the consortium Duke Cogema Stone and Webster (DCS) to dispose of 33 tons of surplus weapons grade plutonium by irradiating it in the form of MOX in four U.S. commercial pressurized-water reactors (PWRs) pending NRC approval. The reactor sites chosen were McGuire 1 & 2 and Catawba 1 & 2, both of which are located in North Carolina. Initial MOX fuel usage is estimated to start in 2007.

Cogema has completed MOX fuel fabrication and spent fuel recycling for thirteen years at COGEMA-La Hague processing plant in France. An estimated 80% of the world's reprocessing is undertaken at the Cogema facilities (Cogema. Reprocessing). Twenty-seven foreign power companies from Belgium, Germany, the Netherlands, Switzerland and Japan reprocess their spent nuclear fuel at La Hague. In addition to these countries, Great Britain also uses MOX fuel for their PWRs. Thirty-Five reactors were loaded with MOX fuel in Europe as of September 2001, including twenty in France. Eight additional French reactors are technically ready for MOX and fifty European reactors are expected to convert to MOX over the next few years (UIC). Japan intends to use MOX fuel in all of their reactors by 2010 and is currently developing their own reprocessing plant using French technologies.

Fabrication of U.S. weapons grade plutonium into MOX will be completed at the Savannah River Site (SRS) in Georgia. The MOX facility will have two primary

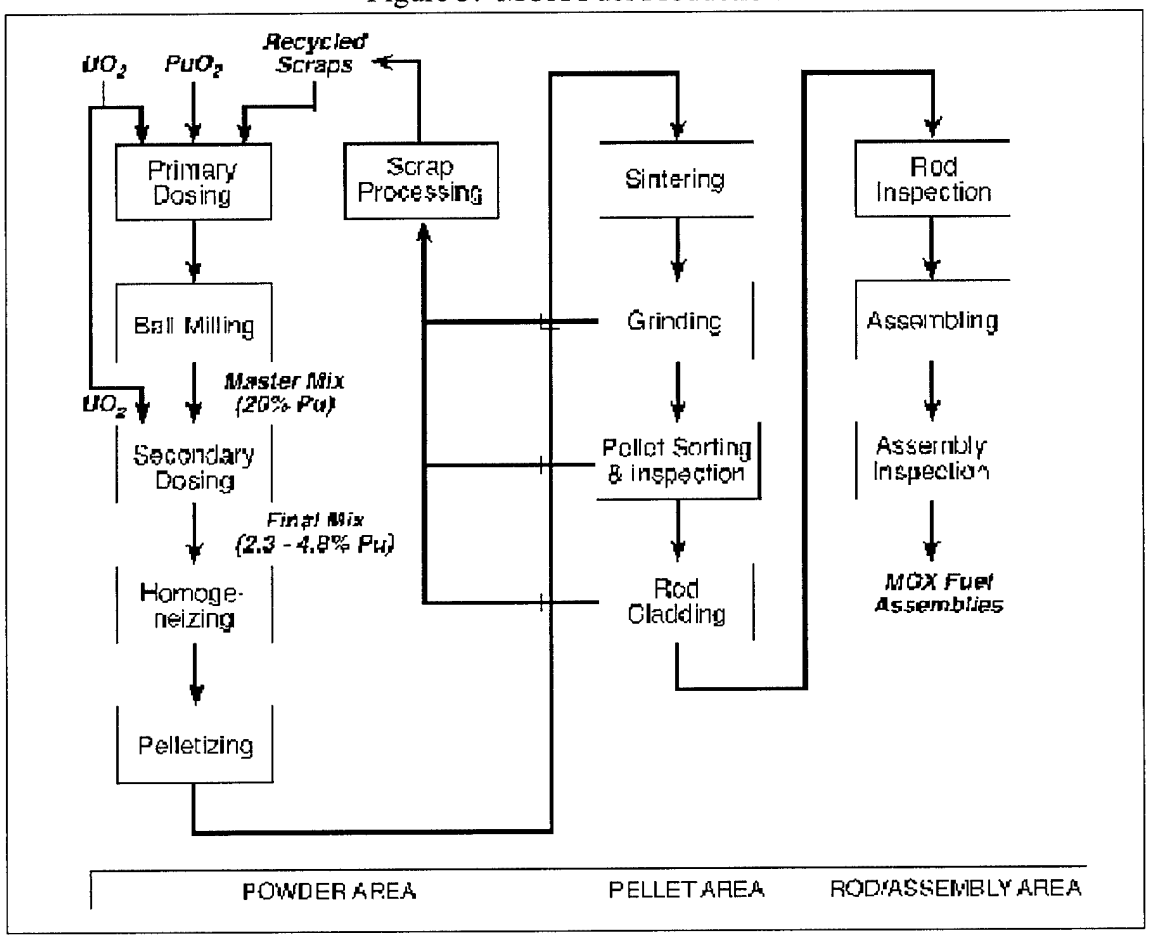
operations for the production of MOX fuel. The first process of fabrication is called aqueous polishing (AP). AP purifies the plutonium by removing chemical and radioactive impurities (NRC). This solvent extraction process is primarily used to remove gallium. Gallium is used to stabilize the crystalline structure of plutonium in nuclear weapons (Lyman 2000). Figure 2 is a schematic illustration of the polishing process to ready the PuO_2 for MOX fuel production. France, United Kingdom, Russia and Japan conduct a similar process in prepping their plutonium resources prior to MOX production. These process operations are all conducted within hot cells and gloveboxes because the plutonium is pyrophoric, which spontaneously ignites at about 150^0 C (IEER).

Figure 2: Aqueous Polishing



The second key process at SRS is the MOX production process. The production of MOX resembles uranium oxide (U_3O_8) fuel fabrication. Purified PuO_2 and natural U_3O_8 will be measured, mixed, crushed and blended. The mixture is then pressed into small cylindrical pellets that meet specific criteria for different fuel types. The pellets are then sintered at 1750° C in a reducing atmosphere for conversion into a ceramic. The pellets will be ground to the specific circumference requirements and inspected to ensure conformance. All powder operations are conducted under a nitrogen gas atmosphere. Figure 3 illustrates a schematic flow diagram of MOX fuel production.

Figure 3: MOX Fuel Production



MOX fuel will displace a fraction of the low enriched uranium (LEU) fuel that reactors currently use. The typical commercial nuclear fuel rod is composed of 96% ^{238}U and 4% ^{235}U. The composition of MOX fuel rods will be 93-95% ^{238}U and 5-7% plutonium. Thus, the plutonium will replace the LEU in a percentage of the reactor fuel rods. Because ^{239}Pu and ^{241}Pu have fissile properties similar to ^{235}U, the radioisotopes can be substituted into the fuel rods replacing ^{235}U and still maintain the performance desired from the nuclear reactor operations. It is estimated that DCS will operate with 40% of the reactor vessel being MOX fuel.

The terminology weapons-grade MOX (WG-MOX) and reactor-grade MOX (RG-MOX) have been used to describe the isotopic differences between the plutonium compositions that will eventually be irradiated as MOX fuel. WG-MOX will consist primarily of ^{239}Pu. It has been estimated by Lyman (2000), that approximately 93% of the plutonium used in WG-MOX will be ^{239}Pu. 6.5 % of the plutonium abundance will be ^{240}Pu, which was inadvertently created during plutonium production. These plutonium abundances will be used to create the MOX fuel used by the United States and Russia. Under Lyman's assumption, the surplus United States and Russian plutonium will be polished and used as MOX three years after creation. See Table 1 for MOX isotopic comparisons.

RG-MOX will comprise of plutonium that originated from reprocessing of spent nuclear fuel, which was irradiated for three to four years in a commercial reactor (Cogema. Reprocessing). The plutonium is created via ^{238}U neutron absorption as mentioned above, however the residence time in the reactor is significantly longer, thus

allowing for the greater growth of ^{240}Pu and ^{241}Pu. The abundance of ^{240}Pu and ^{241}Pu in RG-MOX will be 24% and 9% respectively and having the remaining 56% ^{239}Pu. There is also a slight abundance of americium-241 (^{241}Am) in the RG-MOX. ^{241}Am is progeny from the short-lived ^{241}Pu which β- decays in 14.4 years. See Table 1 for the isotopic comparisons of the three reactor fuel types. For monitoring purposes and actinide abundance in the calculations of absorbed dose there is a significant difference between the two MOX types due to the relatively short half-lives of ^{241}Pu and ^{241}Am and subsequently higher specific activity. Regardless of the plutonium isotopic composition differences, both MOX fuel types remain at 93-95% ^{238}U and 5-7% plutonium (Lyman 2000).

Table 1: Initial Isotopic Composition Within Various Fuel Configurations

Uranium Composition	*LEU*	*WG-MOX*	*RG-MOX*
^{234}U	0.04	0.002	0.002
^{235}U	4.25	0.20	0.20
^{236}U	0.01	0.001	0.001
^{238}U	95.7	99.797	99.797
Plutonium Composition			
^{238}Pu	-	0.04	2.30
^{239}Pu	-	92.37	56.2
^{240}Pu	-	6.49	24.2
^{241}Pu	-	0.24	9.00
^{242}Pu	-	0.10	6.90
^{241}Am	-	0.76	1.4

With the exception of ^{241}Pu, alpha emission is the primary mode of radioactive decay of the radionuclides in MOX. Only ^{235}U emits a photon with an energy and abundance that can easily be detected *in vivo*. Since there is no fixed or standard isotopic ratio for MOX fuel, occupational exposure monitoring can be quite complicated because all the

uranium and plutonium isotopes, as well as ^{241}Am, must be evaluated in order to accurately estimate dose. The isotopic composition of the MOX will change in a predictable manner as the fuel ages before irradiation. On the other hand, the isotopic composition of irradiated or spent MOX fuel will present a complicated scenario for evaluation since the isotopic composition will essentially be unknown. Any biological or environmental samples collected for indirect analysis post exposure would likely be costly and time consuming to analyze because of the need to perform high sensitivity, isotopic analysis of uranium, plutonium, and americium. Direct *in vivo* measurements will most likely utilize the 54% abundant 185 keV photon from ^{235}U to obtain a rapid, quantitative estimate of intake. Conventional methods for spectrum deconvolution are available to quantify the amount of plutonium and americium deposited in an organ or tissue.

Objective:

The objective of this study was to evaluate the ability to measure plutonium, uranium and americium in lungs by direct *in vivo* measurement as a result of occupational exposure to MOX fuel. Because nearly all the photons emitted by uranium, americium, and plutonium are low in energy and abundance, a second objective was to determine whether ^{235}U and ^{241}Am would confound direct *in vivo* measurement of plutonium associated with MOX fuel.

Results of direct, *in vivo* measurement of photons in the body are easily confounded if a worker has been exposed to more than one radionuclide. The interference produced by multiple energy photons is a predominant concern for the radionuclides emitting the

lowest energy photons, especially if these photons are of low abundance. Several methods of spectrum deconvolution have been developed to compensate for interferences arising from overlapping photon energies.

Direct *in vivo* measurement of plutonium, americium, and uranium in MOX fuel is particularly susceptible to confounding by higher energy photons because attenuation of the overlying chest wall tissue may effectively degrade resolution and decrease the sensitivity of the measurement.

MATERIALS AND METHODS

Direct *in vivo* measurement for MOX in the lungs requires a calibration standard that contains uranium, plutonium and americium in proportions that simulate the isotopic abundance expected in the fuel. A set of surrogate lungs for use in the Lawrence Livermore National Laboratory (LLNL) thoracic phantom (Griffith 1978) were fabricated at the University of Cincinnati using a method described by Spitz et. al. (1994). This method replicates the methods and materials used in the original design specifications for the LLNL thoracic phantom. Standard solutions of 93% enriched uranium, ^{241}Am, and ^{238}Pu were prepared from radionuclides traceable to the National Institute for Standards and Technology and added during lung fabrication to produce a set of lung phantoms containing homogeneously distributed radioactive material. Table 2 lists the activity of each isotope added to the phantoms. Because all commercial solutions of ^{239}Pu contain some ^{241}Am, it was decided to use a pure solution of ^{238}Pu that had no ^{241}Am, even though ^{238}Pu is not expected to be present in significant quantities in MOX fuel.

Table 2: MOX Phantom Lung Activity			
	Activity (nCi)		
	^{241}Am	^{238}Pu	^{235}U
Right Lung	178.33	654.13	1.155
Left Lung	144.71	537.8	0.971
Total Activity (nCi)	323.04	1191.93	2.126
Total Activity (Bq)	1.20E+04	4.41E+04	7.87E+01

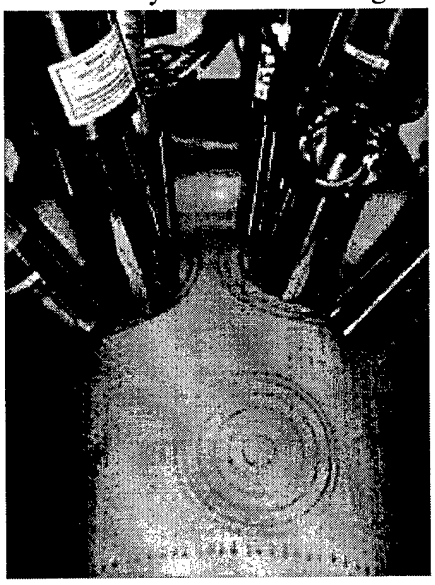
The MOX lungs were inserted into the LLNL thoracic phantom and counted for up to 50 minutes using an array of four 3800 mm^2 high resolution germanium detectors placed on the anterior thorax of the phantom. Thin, low Z entrance windows on each detector allow measurement of low energy photons and x-rays associated with the decay of plutonium. Efficiency calibration measurements were performed for chest wall thicknesses ranging from 1.56 to 4 cm by adding individual overlays of increasing thickness on top of the phantom thorax. The chest wall overlays were muscle equivalent (MEQ) at 1.04 g/cm^3 (Kahn). Figure 4 illustrates a similar counting arrangement using six 2000 mm^2 detectors.

Data collection and analysis was performed using the Canberra GENIE spectroscopy system with Canberra ABACOS whole body counter software. A library of relevant photon energies and abundances was developed expressly for the MOX lungs (Table 3).

Except for ^{57}Co, all of the isotopes listed in Table 3 have a 13 ± 1 keV photon. But due to the attenuation via CWT and correction for interference, it is not practical to measure this photon energy.

Table 3: Software Identification Library				
Nuclide	T 1/2	Energy (keV)	% Abundance	Key Line
Co-57	270.9 D	14.4	9.54	No
		122.1	85.60	Yes
		136.5	10.60	No
Th-234	24.10 D	63.29	3.83	No
		92.35	2.72	Yes
		92.78	2.69	No
U-235	7.03E+08 Y	143.79	10.50	No
		185.74	53.10	Yes
Pu-238	87.74 Y	17.13	5.17	Yes
		43.47	0.04	No
		99.85	0.01	No
Am-241	432.2 Y	59.54	35.70	Yes

Fig 4: Detector Array for In Vivo Lung Monitoring



By observing Table 3, it can be noted that four emissions lines are very close in energy, namely 16.12, 17.13, 17.61 and 16.16 keV. Even with the optimal resolution detectors used in this experiment, these energies will result as a multiplet peak with the possible appearance of a shoulder or two. The calculated total activity of ^{238}Pu will be attempted by subtracting the other three isotopes, in a method called peak stripping, leaving just the 17.13 keV for analysis. The activity ratios calculated by this method will be compared to a standard measure of the 43.53 and 99.48 keV emissions from ^{238}Pu. There is higher detector efficiency with these energies, however the abundance of these photon admissions is only 0.04 and 0.01% respectively (Browne).

In comparison, the 17.13 keV emission of ^{238}Pu is of lower energy, which will result in higher chest wall attenuation. The energy is also within 1 keV of three other isotopes known to be in the spectrum. 17.13 keV is most abundant photon from ^{238}Pu, making this energy the most appealing to use in spectral analysis.

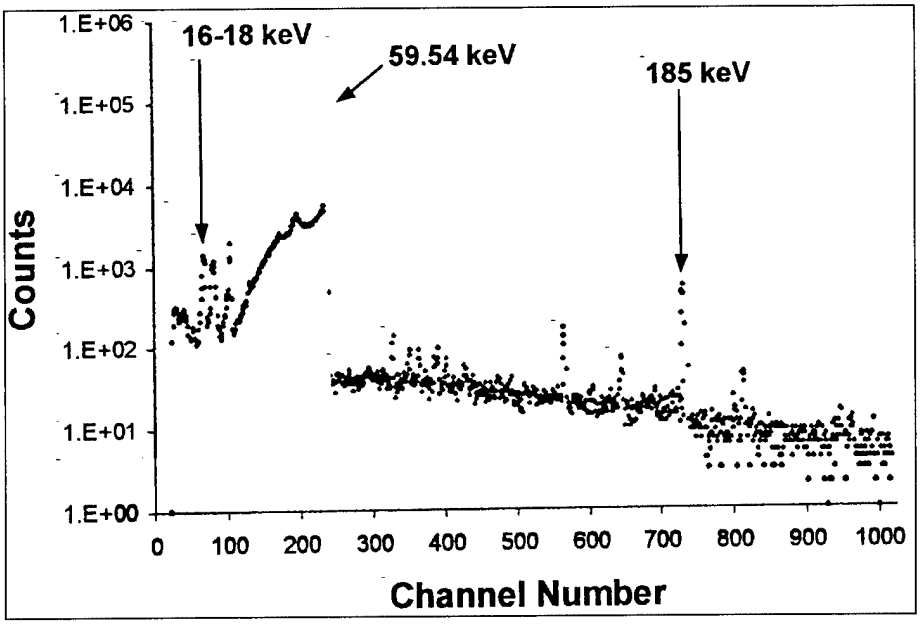
The low abundant emissions of 43.26 and 99.32 keV photons will result in less CWT attenuation and are unique energies in the spectrum making them favorable energies as well for analysis. The known verses measured activity ratios will be compared for these three energies emissions of ^{238}Pu in an attempt to determine the best energy for quantitative analysis.

RESULTS:

Figure 5 shows the complex photon spectrum obtained from a 50-minute calibration measurement using the LLNL thoracic phantom with lungs containing ^{238}U, ^{235}U, ^{238}Pu,

and ^{241}Am to simulate mixed oxide fuel. The muscle equivalent chest wall thickness for the phantom used in this measurement was only 2.56 cm. The 16.12 keV, 17.13 keV, and 17.61 keV peaks appear as a single wide peak in the 16 to 18 keV region of the spectrum.

Figure 5: Spectrum of Mixed Oxide Lung Set in LLNL Phantom (2.56 MEQ-CWT)



The compensation method used by the Canberra Genie system corrects for two or more radionuclides having at least one common peak and the peak cannot be easily resolved into multiplet peaks. A linear least squares solution is applied to a matrix of the branching ratios divided into a vector of the counts per second divided by the efficiency. Since the efficiency is already corrected for effect of attenuation no additional corrections are needed.

The least squares method utilizes the efficiency calibration errors. Since the error of calibration for a low energy *in vivo* measurement system will dramatically increase as a function of chest wall thickness, the comparison routine can potentially provide greater errors in the correction.

Without interference correction, the results for ^{238}Pu at 17.14 keV ranged from 1.5 to 6.4 times the known activity in the lung sets. The ratio of measured to known ^{238}Pu activity increased dramatically beyond 3 cm CWT. With interference correction the ^{238}Pu results ranged from 1.5 to 1.9, with the highest ratio of measured activities occurring above 3 cm CWT.

Figure 6 represents the known activity ratios for ^{238}Pu using the 17 keV energy peak as a function of CWT. Table 4 lists the activity concentrations for all the isotopes without interference corrections. A more extensive table of activity concentrations with uncertainties has been placed in Appendix B.

Figure 6: Activity Ratios for ^{238}Pu (17 keV) as a Function of CWT

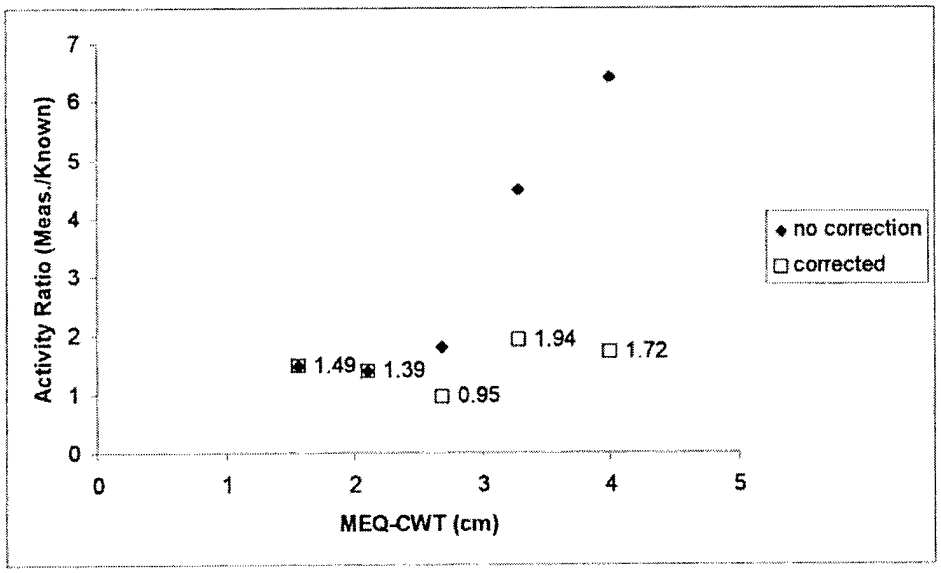
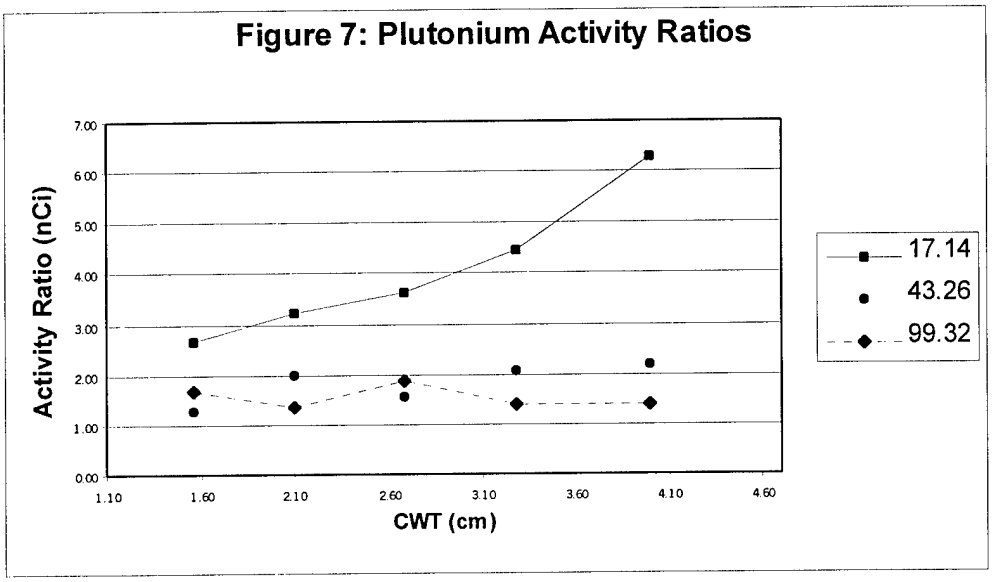


Table 4: Activity Concentrations as a Function of CWT (No interference Corrections)								
Count Scenario			Activity (nCi)					
Chest wall thickness (cm)	Description	Time (minutes)	Pu-238 (17.14 keV)	Pu-238 (43.26 keV)	Pu-238 (99.32 keV)	Am-241 (59.48 keV)	U-235 (143.85 keV)	U-235 (185.81 keV)
1.56	Lung set core	30.04	3.198E+03	1.528E+03	1.972E+03	330	2.44	2.60
2.10	1st Overlay	50.04	3.834E+03	2.351E+03	1.595E+03	325	2.36	2.46
2.68	2nd Overlay	40.02	4.334E+03	1.851E+03	2.217E+03	306	1.90	2.34
3.28	3rd Overlay	50.04	5.302E+03	2.461E+03	1.645E+03	323	2.31	2.43
4.00	4th Overlay	50.06	7.507E+03	2.61E+03	1.674E+03	306	1.82	2.25

Upon closer observation of the measurement results, it was determined that the GENIE system deconvoluted the ^{241}Am 17.5 keV from the multiplet peak occurring in the 16 to 18 keV when the CWT was less than 3 cm. Thus leaving only uranium isotopes as the interfering radionuclides to the measurement of ^{238}Pu. Since the activity of uranium was much lower than that of ^{241}Am and ^{238}Pu, the interference contribution was also much lower, resulting in much lower degrees of interference correction.

Beyond 3 cm CWT, the ^{241}Am peak was not deconvoluted out of the 16 to 18 keV peak by the software, therefore the observed 4 to 7 fold estimated increase in the ^{238}Pu activity.

Table 4 can be compared to the known activities listed in Table 2. Only the lower energy of ^{238}Pu (17.14 keV) showed noticeable differences as CWT increases. ^{241}Am and ^{235}U were easily and accurately measured using the 59.5 and 185.5 keV energies respectively. Figure 7 shows the activity ratios for the three plutonium emissions of concern.



A complete review of the activity ratios is listed in Table 5. Table 5 does not show the interference correction for the 17.14 keV emission. The 43.26 and 99.32 keV emissions had very low measured vs. known activity ratios. The 43.26 keV peak had a mean activity ratio of 1.81 for the five CWT scenarios. The 99.32 keV emission had a ratio of only 1.52 for the 5 cm chest wall scenarios.

Table 5: Activty Ratios at Various Energies as a function of CWT			
Chest wall thickness (cm)	Pu-238 (17.14 keV)	Pu-238 (43.26 keV)	Pu-238 (99.32 keV)
1.56	2.68	1.28	1.65
2.10	3.22	1.97	1.34
2.68	3.64	1.55	1.86
3.28	4.45	2.06	1.38
4.00	6.30	2.19	1.40

DISCUSSION:

With thinner chest wall thickness, there is a better chance for the 16-18 keV group of x-rays to be deconvoluted as a multiplet peak. The attenuation of the 12-14 keV peaks would be more severe and result in a higher uncertainty. The ability of a system to deconvolute the 16-18 keV group of peaks is a function of the detector resolution and the calibration of the system. When multiplet deconvolution does not occur, above 3 cm CWT for this study, and no compensation for interferences is applied, the measured values for ^{238}Pu was observed to be 4 to 6 times higher (Table 5) than the known value of ^{238}Pu in the lungs. When the 17.5 keV of ^{241}Am is deconvoluted out of the 16-18 keV multiplet, the measured values for ^{238}Pu was observed to be within 1.3 and 2.7 times the known value of ^{238}Pu (Figure 6). The deconvolution of the 17.5 ^{241}Am peak tended to occur at CWT's below 3 cm.

When the interference correction was applied to CWT's below 3 cm (i.e., to counts where the ^{241}Am 17.5 keV had been resolved out of the multiplet), the interference correction provided estimates of ^{238}Pu (based on 17 keV peak) that were –5% to +50% of the known ^{238}Pu activity. Improvement in the effectiveness of the interface correction was observed as the estimate of the ^{241}Am improved. Thus, when the 17.5 ^{241}Am peak was deconvoluted out of the 16-18 keV multiplet, the effectiveness of the GENIE interference correction is dependant on how well the deconvolution removes ^{241}Am counts from the multiplet. The more accurate the estimate of ^{241}Am activity as determined from the deconvolution 17.5 peak out of the 16-18 keV multiplet, the better the estimate of ^{238}Pu activity when the interference correction is applied. This

observation agrees with the computational method, since ^{241}Am, when the counts from ^{241}Am 17.5 peak are deconvoluted from the 16-18 keV multiplet, ^{241}Am is no longer considered to be part of the interfering radionuclides. If the deconvolution fails to resolve all of the counts from ^{241}Am these counts are added to the counts in the multiplet peak, and higher ^{238}Pu estimates occur.

Extrapolation of the efficiency below the lowest calibration energy line also becomes critical. This extrapolation, depending on the method used to extrapolate, can provide unrealistic values for the low energy efficiency.

The 43.26 and 99.32 keV emission peaks showed even a higher certainty in the activity ratios (Figure 7). Using the 99.32 keV emission line provide the best activity ratio of only 1.52. The 44.26 keV emission was slightly higher at 1.81.

Using either method to determine the amount of plutonium present provided acceptable results. Difficulty arises in predicting the amount of competing isotopes when using the 17.14 keV peak stripping method. The accident scenario that leads to individual exposure, will be most useful in predicting the isotope concentrations prior to measurement. For example, in the production of MOX, there should only be a slight interference from ^{235}U and ^{241}Am. ^{235}U is present in natural uranium, but only at 0.7%. The determination of ^{241}Am will be dependant on the processing procedure, specifically pre or post AP. But if there were ever a loss of coolant accident (LOCA) where MOX fuel in present within the reactor vessel, there would be numerous variables for interference corrections as the fission products will make the spectrum even more complex. In such a scenario, bioassay and longer *in vivo* measurements will be used as supplemental data for dose calculations.

REFERENCES:

1. 62 Fed Reg. 28009. U.S. Department of Energy (DOE), *Record of Decision for the Surplus Plutonium Disposition Final Environmental Impact Statement*, Federal Register: (Volume 65, Number 7). January 11, 2000

2. Cogema. Plutonium (Pu): Information brief on the properties and uses of plutonium. Available at: http://www.cogema.com/cogema/uk/fs_accueil.htm. Accessed 15 April 2003.

3. Cogema. Reprocessing. Available at: http://www.cogema.com/uk/fs_accueil.htm. Accessed 15 April 2003.

4. Uranium Information Centre (UIC) Ltd. Mixed Oxide Fuel (MOX), Nuclear Issues Briefing Paper 42 February 2002. Available at: http://www.uic.com.au/nip42.htm. Accessed 15 April 2003.

5. NRC. Chemical and Process Safety. Available at: http://www.nrc.gov/materials/fuel-cycle-fac/mox/chemical.pdf. Accessed 15 April 2003.

6. Lyman ES. Public Health Risks of Substituting Mixed-Oxide For Uranium Fuel in Pressurized Water Reactors. Science and Global Security, Volume 9, pp 1-47, 2000

7. Institute for Energy and Environmental Research (IEER). Properties of Plutonium Metal. Available at: http://www.ieer.org/sdafiles/vol_3/3-2/putables.html. Accessed 15 April 2003.

8. Griffith, R.; Dean, P.; Anderson, A.; Fisher, J.: Fabrication of a tissue-equivalent torso phantom for intercalibration of *in vivo* transuranic nuclide counting facilities. In Proceedings: IAEA International Symposium on Advances in Radiation Protection Monitoring. Vienna: IAEA; IAEA-SM-229/56; UCRL-80343; 493-504; 1978

9. 10. Edgordo Browne, Richard B. Firestone, Table of Radioactive Isotopes, 1st Edition, Wiley-Interscience, July 16, 1986

10. Spitz, H.; Glover, S.; Liu, N.; Smith, B.; Hickman, D.; Kruchten, D.; Anderson, L.: Measurement of the attenuation coefficient for Livermore thoracic phantom lungs fabricated using contemporary materials. Health Phys. 67(1): 39-46; 1994.
Browne, E.; Firestone, R. Shirley, V.: Table of Radioactive Isotopes. John Wiley (New York) 1986.

11. Kahn, F.: The Physics of Radiation Therapy, Second Edition. Lippincott Williams & Wilkins (Baltimore) 1994.

Appendix A
Isotopes with Photon Emissions and Yields
(Browne)

Am-241		
Photon Emission Products		
Type	Energy(keV)	Yield
γ	26.345	0.024
x-ray	20.997	0.052
x-ray	13.927	0.130
x-ray	17.611	0.202
γ	59.537	0.357

U-234		
Photon Emission Products		
Type	Energy(keV)	Yield
γ	53.200	0.0011
γ	120.912	0.0410

U-235		
Photon Emission Products		
Type	Energy(keV)	Yield
x-ray	12.95	0.220
x-ray	16.12	0.150
x-ray	19.12	0.026
γ	89.96	0.036
γ	93.35	0.055
γ	143.79	0.105
γ	185.74	0.531
γ	205.33	0.047

U-238		
Photon Emission Products		
x-ray	12.952	0.030
x-ray	16.161	0.041

Pu-238		
Photon Emission Products		
Type	Energy(keV)	Yield
x-ray	13.60	0.0420
x-ray	17.13	0.0520
γ	43.26	0.0004
γ	99.32	0.0001

Th-234		
Photon Emission Products		
Type	Energy(keV)	Yield
x-ray	13.274	0.0390
x-ray	16.572	0.0440
γ	63.000	0.0380
γ	92.350	0.0272
γ	92.780	0.0690

Appendix B
Observed Activity of MOX Lung Set

Observed Activity and Uncertainty for ^{241}Am, ^{238}Pu, and ^{235}U for *In Vivo* Analysis of MOX Lungs

			Activity (nCi)											
Chest Wall Thickness (cm)	Description	Time (minutes)	Pu-238 (17.14 keV)	% Error (2 SD)	Pu-238 (43.26 keV)	% Error (2 SD)	Pu-238 (99.32 keV)	% Error (2 SD)	Am-241 (59.48 keV)	% Error (2 SD)	U-235 (143.85 keV)	% Error (2 SD)	U-235 (185.81 keV)	% Error (2 SD)
1.56	Lungset Core	30.04	3.198E+03	3.2	1.528E+03	54.7	1.792E+03	38.1	330	0.7	2.44	16.7	2.60	5.8
2.10	1st Overlay	50.04	3.834E+03	3.9	2.351E+03	40.5	1.595E+03	36.5	325	0.6	2.36	17.3	2.46	4.9
2.68	2nd Overlay	40.02	4.334E+03	9.1	1.851E+03	65.1	2.217E+03	32.2	306	0.8	1.90	25.2	2.34	6.7
3.28	3rd Overlay	50.04	5.302E+03	15.9	2.461E+03	56.5	1.645E+03	53.9	323	0.8	2.31	22.7	2.43	6.6
4.00	4th Overlay	50.06	7.507E+03	38.0	2.612E+03	78.0	1.674E+03	50.3	306	0.9	1.82	30.7	2.25	7.5