LOW ENERGY GAMMA SPECTROMETER NAI(TL) (FIDLER) MEASUREMENT AND VERIFICATION OF URANIUM BUILDING SURFACE FINAL SURVEY FOR FREE RELEASE FOR UNRESTRICTED USE DATA

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ABSTRACT

Final surveys for free release for unrestricted use of building surfaces are subject to technical review. Alpha and beta surveys of building surfaces are affected by many factors such as calibration isotope, calibration source size, source composition, contaminant distribution, and surface condition correction factors. Reasonable disagreement between a licensee and an oversight group can occur. Demonstration of the validity of alpha measurements with a second monitoring technique is an effective method of preventing disagreement. The case study will illustrate how a Low Energy Gamma Spectrometer NaI(Tl) is an effective tool for the verification of alpha surface measurements and surface status assumptions for a facility with a wide range of uranium enrichments. Under the correct conditions the use of Low Energy Gamma Spectrometry NaI(Tl) for direct measurement and free release of uranium activity located beneath or within painted building surfaces is demonstrated to be feasible.

INTRODUCTION

BWXT Services has developed and the Nuclear Regulatory Commission has accepted, a practical, and cost effective technique to verify alpha surface Final Survey for Free Release for Unrestricted Use measurements conducted in support of the decommissioning of a site with depleted uranium (DU), Low Enriched Uranium (LEU), High Enriched Uranium (HEU), and Mixed Oxide (MOX) facilities. The surface measurement verification was a critical component of the Final Surveys. Uranium isotopes were the source of the surface alpha emission rate in the uranium buildings and limited areas of the site MOX facility. Variable or unknown uranium enrichments including Depleted Uranium (0.3% enriched), Low Enriched Uranium (3% enriched), and High Enriched Uranium (97.6% enriched) were present at many locations. Small quantities of fission and activation products were also present throughout the majority of the site.

An integral portion of the final survey are the assumptions made in the final survey process, particularly source composition, contaminant distribution, and surface condition correction factors. If these assumptions are not representative, the accuracy of the final survey results is open to question. Verification of final survey results with additional alpha and beta measurements addresses the thoroughness of the survey, and survey instrument performance, but does not test the validity of the assumptions made in the final survey process. Demonstration of the validity of assumptions concerning contamination location or other surface conditions by a second type of monitoring technique is a more effective method of preventing disagreement, as it tests the assumptions made in the final survey process. If the assumptions are correct both monitoring methods will yield consistent results.

In situ Low Energy Gamma Spectrometry (using a Bicron G5 Field Instrument for Detection of Low Energy Radiation (FIDLER) in combination with a Canberra Multi Channel Analyzer (MCA) - Model 3502) was selected as the second monitoring technique. L Shell X-rays and low energy gammas were measured using the FIDLER. The method was chosen because X-rays and gammas are insensitive to surface roughness and surface material type, when compared with alpha emissions. In addition, direct measurement through paint is feasible, allowing verification of building surface contamination history. In our experience these issues are key debatable issues in the facility final survey. The final survey guidance documents MARSSIM "Multi-Agency Radiation Survey and Site Investigation Manual" and Draft NUREG-1507 "Minimum Detectable Concentrations with Typical Radiation Survey Instruments for Various Contaminants and Field Conditions" identify surface roughness and surface composition corrections as the critical factors in interpreting final survey data. NUREG-1507 demonstrates that invalid assumptions for surface roughness and surface material corrections can each cause non-conservative errors on the order of a factor of 2-3. The presence of paint over contamination may completely block detection of alpha emitters.

The FIDLER is a large area (126 cm^2) thin-window (1.6 millimeter) NaI scintillation detector that is specifically designed for low energy X-ray and gamma radiation monitoring. Thin window NaI probes have high intrinsic efficiency for X-rays and gammas of these energies. The probe is effective in detecting 10 keV to 100 keV X-rays and gammas. It is ineffective in detecting photons of greater energy. This is a distinct advantage as it greatly reduces the interference from higher energy gammas and X-rays, which is present in thicker detectors. The FIDLER was used as a gamma spectrometer covering the energy spectrum up to 100 keV. Two regions of this spectrum are of particular interest for monitoring surfaces in uranium facilities facility. They are the L Shell X-ray region between 10-20 keV, and the uranium and uranium daughter K shell X-ray region (63 to 100 keV). The FIDLER was used with a MCA to record the number of counts in these regions of interest (ROIs) and the entire energy spectrum. Biased survey locations were chosen throughout the facility for the verification measurements. The survey consisted of the following principal steps:

- Determination of Background and Regions of Interest: Determination of the gamma and x-ray emissions of interest in the 10 keV to 100 keV range for licensed materials and for relevant background emissions.
- Setup and Calibration of the In Situ Low Energy Gamma Spectrometry System: Calibration isotopes and method were selected and applied.
- Selection of Survey Locations: Biased survey locations were chosen for the verification measurements.
- Survey: Conduct of the survey.
- Data Analysis: Analysis of the spectra and the net count rate. If present, net X-ray peaks were identified and quantified.

METHODOLOGY

Determination of Background and Regions of Interest

Measurements in this verification survey were of Uranium L Shell X-rays. The original survey was of alpha emissions. Data from this survey would determine if the assumption that direct alpha measurements properly characterized the radiological status of each grid was correct. The uranium isotopes and their immediate daughters emit significant quantities of 10-20 keV X-rays, (see Tables I and II). Table I, *" Site Uranium X-ray Abundance and Typical Detection Efficiency,"* shows the calculation of X-ray detection efficiency for the principal facility uranium enrichments. The data includes the L Shell X-ray energy abundance by energy and isotope, and isotopic abundance for each isotope in the Parks facility uranium suites. Figures 1, *"Comparison of Background Concrete vs. Natural Uranium Spectra"* and Figure 2, *"Comparison of Spectra from Background Concrete and Uranium in The L Shell X-ray Region,"* compares a typical measurement spectrum from a background area to that of natural uranium. Figure 1 shows the spectrum that is typical of the floor material background and a natural uranium spectrum over the approximate range of 10 to 100 keV. Figure 2 provides a similar plot over the range of 10 to 30 keV (approximate). It includes the counting window used for the L Shell X-ray monitoring.

A background location in the site MOX fuel manufacturing facility was selected for an initial spectrum collection with a Bicron G5 FIDLER. The natural uranium spectrum was collected using a 4287 dpm NIST traceable area source. The FIDLER is a large area (126 cm^2) thin window (1.6 millimeters) NaI(Tl) scintillation detector for low-energy X-ray and gamma radiation monitoring. Thin window NaI(Tl) probes have high intrinsic efficiency for X-rays and gammas of these energies. The probe is effective in detecting 10 keV to 100 keV X-rays and gammas. It is ineffective in detecting photons of greater energy. The spectra from the FIDLER used with a Canberra Multi Channel Analyzer (MCA) Model 3502 are shown in Figures 1 and 2.

Uranium isotopes emit L Shell X-rays in the 10-20 keV range. The emission rates range from 19.8% for Depleted Uranium to 13.7% for High Enriched Uranium (see Table 1). The emission rates are drawn from ENDF/B-VI decay data. The rough equality of the X-ray production over the entire range of uranium enrichment allowed the use of a single efficiency for all uranium measurements and eliminated the need to determine the isotopic distribution of uranium at measurement locations.

Figure 1 contains the uranium spectrum, which has three significant features, overlaid upon the background spectrum. The uranium source and the background spectra have the same primary features, which are a low background region in the lower channels, a 25-30 keV complex, and a Th-234 peak at 63.3 keV. The Th-234 peak lies upon a background complex of 60-100 keV emissions. As a result, background for the K Shell X-ray region is very high compared with the remainder of the spectrum. The second feature is the L Shell X-ray peak region, which is in the low count rate region of the background spectrum. The third is the 25-30 keV complex, which is derived from multiple sources. Figure 2 focuses on the L Shell X-ray region of the uranium X-ray spectrum and the low count rate region, which coincide. Figures 1 and 2 show the rationale for the selecting the L Shell X-ray peak region as the primary ROI, instead of the K Shell X-ray peak region.

Setup and Calibration of the In Situ Low Energy Gamma Spectrometry System

The instrument was calibrated by a Certified Health Physics Engineer with the assistance of the Director of the Analysis Laboratory. Rapid temperature changes were avoided. The base data are:

- Sensitive range for the FIDLER 10 to 100 keV
- Maximum Voltage 1600 Volts
- Operating Range 39- 109 degrees Fahrenheit
- Temperature Rate of Change 18 degrees Fahrenheit per hour.

The uranium 16 (10-20) keV X-ray complex and the 60 –100 keV X-ray complex gamma complex are the two regions of interest. Before use a source measurement were performed and recorded for each region of interest and for at least channels 2 through 80. The post measurement spectrum on the MCA was observed for visible peaks and indications of unexpected isotopes in the spectrum.

The FIDLER was calibrated with an Am-241 area source and a natural uranium area source. The Am-241 L Shell X-rays are a reasonable surrogate for the uranium L Shell X-rays, and the 59.5 keV emission is similar to the Th-234 63.3 keV. The windows were set using the Am-241 source, as the range of L Shell X-ray energies for Am-241 is greater than that of the uranium. This allowed dual use of the instrument for transuranic and uranium work. The Am-241 source was used as the daily check source as it had a much higher emission rate than the uranium source. Efficiency was determined using the natural uranium source. The efficiencies for the site uranium suites were determined as shown in Table 1. Isotopic abundance and isotopic emission rates are reflected in the calculated efficiency. Cs-137 was used to over check the energy calibration, as it has useful 32 keV K Shell X-rays.

The use of the multichannel analyzer allowed a precise window (region of interest) setting, window corrections to allow for instrument drift, accurate peak location, visual confirmation of the presence or absence of peaks of interest, and continuous monitoring of the spectrum. These features ensured the quality of the survey measurement and detection of possible activity that might be hidden in the background.

Selection of Survey Locations

Five survey locations were selected for measurement and spectra recording.. The locations were on painted walls. The FIDLER measurements were conducted to verify the judgement that activity was not present beneath the paint, and therefor that alpha measurements taken on the painted surface were representative of the actual contamination level. Microshield V 5.03 was used to determine the effects of the paint on the X-ray transmission to the detector. The paint thickness had been determined as a part of the site lead paint characterization. All radioactive material was assumed to be beneath the paint. The surface conditions at the selected sites were considered typical for the uranium areas of the facility. The sites were inspected and found free of dust, water et al, that might interfere with the measurements. Each location was 50 cm or further from the floor or a second wall. Previous measurements had found

that survey measurements in locations closer than 30 cm to a second concrete surface were distorted and elevated, primarily in the 50-100 keV region.

Survey

The survey was conducted by placing the FIDLER directly on the floor or against the wall within each selected grid at the grid center, where the alpha measurements were taken. A semi-portable steel framework jig was used to allow reliable long term counts on wall surfaces from 30 cm to 250 cm from the floor. The weight of the jig ensured its stability in a constant position. A source check was conducted with an Am-241 source, to ensure proper instrument response and to confirm the validity of the counting window settings. The response in the L Shell X-ray and K Shell X-ray Regions Of Interest (ROIs) were recorded. The channels in which the L Shell X- rays, the Am-241 gamma (if found), and the Th-234 63.3 keV gamma occurred were recorded. The MCA was set to the survey time to be used and the count started. The counting times and resulting Minimum Detectable Activities (MDA) are in Tables IV and V. The counting times used were 600 minutes. Measurements were begun and completed between 0700 and 2400 hours, to avoid temperature changes greater than the manufacturer's recommendations.

After the count was complete, the data were recorded and the spectrum was visually inspected. The presence and location (or absence) of a L Shell X-ray peak, an Am 241 59.5 keV peak, and the location of the Th 234 63.3 keV peak were recorded.

Data Analysis

The data analysis consisted of the following primary steps:

- Spectrum Inspection for Uranium. Each recorded spectrum was plotted and inspected for the presence of definitive uranium peaks (see Figures 1 and 2). These were the Th-234 63.3 keV gamma peak and the 16 keV L Shell X-ray peak in the 12-20 keV L Shell X-ray region. The 16 keV L Shell X-ray peak comprises the large majority of the uranium X-ray emissions for all enrichments.
- General Spectrum Analysis. The spectra were reviewed for the presence of other gamma emitters such as Cs-137, which has emits K Shell X-rays of 31.8 and 32.2 keV (ENDF/B-VI Decay Data) of 5.9% total abundance. Due to the resolution of NaI(Tl), the two emissions comprise a single peak.
- Spectrum Stripping. The L Shell X-ray spectrum was stripped by subtracting the lowest channel cpm count rate (see Figure 3 *Gross and Net L Shell X-ray Spectra*) for a typical spectrum.
- Alpha Activity Level Estimation. The slope of the 25-30 keV complex was inspected. The slope inflection point was determined. The inflection point was treated as the beginning of the detectable presence of L Shell X-rays. The gross area of a "peak"was defined by the channel with a net zero count rate and the inflection point channel was determined. The "background count rate for the "peak" was determined by multiplying the inflection point count rate by the number of channels in the "peak" and dividing the result by two. The net count rate for the "peak" was then determined

by subtracting the background count rate from the gross count rate. The net count rate with the peak efficiency from the instrument calibration and the probe sensitive area (126 cm^2) was then used to calculate the dpm uranium alpha emitters per 100 cm^2. As the site used Depleted Uranium (0.3% enriched), Low Enriched Uranium (3.0% enriched), and High Enriched Uranium (97.6 % enriched), the lowest applicable efficiency, that of High Enriched Uranium was used. The efficiency was adjusted for absorption of X-rays by the paint.

- The estimated uranium alpha emitting activity levels were compared with the direct alpha measurements (see Table III). Four of the results where consistent with background. One location had a result of 6.8Bq (405 dpm)/100 cm^2. The location was sampled and found to be at background activity on a per gram basis. The activity present was 8 Bq (480 dpm)/100 cm^2 (MDA).

DISCUSSION

Direct low energy Region of Interest FIDLER measurements for uranium on site concrete surfaces are an effective method of verifying base final survey assumptions, including assumptions on the location of surface contamination on a wall (above or below the paint layers).

In addition, in the correct circumstances direct final survey using this method is a practical alternative to remediation of surfaces. Measurement times of 15 minutes per location achieve MDAs (after allowance for X-ray absorption by paint) of approximately 1000 dpm alpha per 100 cm^2 . Uranium L Shell X-ray measurement is a feasible method for proving that a given location meets release criterion, if the Regulatory Guide 1.86 Uranium alpha activity limit of 83 Bq or 5000 dpm per 100 cm^2 applies. The MDAs were calculated using the lowest efficiency of the site uranium mixtures, that of High Enriched Uranium. Due to variance in material background reliance on spectrometry and X-ray peaks is recommended.

L Shell X-ray spectroscopy needs to consider the poor resolution of the X-ray peaks. The L Shell X ray "peaks" are broad, flat and are more accurately described as distortions of the background spectrum (30% Full Width at Half Maximum). The L Shell X-rays were selected as they are less subject to background interference than the K Shell X-rays. This includes cases where measurements were attempted at a wall-to-wall or floor-to-wall joint, where the remainder of the spectrum was significantly affected by emissions from the second surface. Use of K Shell X-rays gamma to detect compliance level uranium activity was infeasible at release criteria levels because of significant interference from background emissions, predominantly Th-234, a U-238 daughter, and other background isotopes. Th-234 was so reliably abundant that its location in the spectrum was adopted as an important quality control check.

The physical conditions under which the FIDLER is used are important. NaI(Tl) is temperature sensitive. The instrument is best used under steady state temperature conditions. Daily temperature change effects can be controlled by scheduling measurements in the relatively constant temperature periods of the day. In large concrete structures, these periods of relatively constant temperature proved to be from approximately 0700 to 1600 hours and from 1600 hours to 2400 hours. Overnight measurements in June and July failed as the temperature change was greater than the equipment

manufacturer's specifications. Postponing measurements from periods of unusual or unstable cold or heat should be considered when feasible. The mid summer measurement efficiencies are one-third to one-half of those realized in cooler periods.

CONCLUSION

NaI(Tl) Low Energy Spectrometry for verification or direct measurement of surface uranium activity is feasible and useful. It readily provides sufficient data to test assumptions concerning the appropriateness of surface roughness correction factors, and surface activity location assumptions. The L Shell X-rays are insensitive to surface roughness and surface material type, when compared to alpha emissions and can be detected at shallow depths, such as that of paint on walls. The X-ray results can be used as a standard, against which the assumptions can tested. If the predicted results are achieved the assumptions are correct, and the survey is valid. If disagreement exists, the cause must be determined and resolved.

In addition, if 10 – 20 minute counting times are acceptable, direct Final Survey Free Release for Unrestricted Use measurements may be taken using this methodology. The technique would be useful in circumstances where walls were painted to control contamination. This was a common practice in older uranium facilities.

Table I: Site Uranium X-ray Abundance and Typical Detection Efficiency

Isotope	**10-12 keV**	**12-14 keV**	**14-16 keV**	**16-18 keV**	**18-20 keV**	**Total X-ray Abundance**	**Intrinsic Efficiency[(1)]**	**Weighted Efficiency**
U-238	----------	----------	0.0000922	0.08	----------	0.08	0.245	0.0196
Th-234	----------	----------	----------	0.10	----------	0.10	0.245	0.0245
Pa-234M	----------	----------	----------	0.0047	----------	0.0047	0.245	0.00115
Pa-234	----------	----------	----------	1.12	----------	1.12	0.245	0.10976
U-235	----------	----------	----------	0.31	----------	0.31	0.245	0.0760
Th-231	----------	----------	----------	0.962	0.002	0.964	0.245	0.2353
U-234	0.0022	0.0366	----------	0.0487	0.010	0.098	0.245	0.02401

Table II:Weighted L Shell Efficiency Vs Uranium Enrichment (Mass)

Isotope	**Total Efficiency**	**0.3% U-235 Abundance**	**0.3% Total Efficiency**	**0.7% U-235 Abundance**	**0.7% Total Efficiency**	**3.0% U-235 Abundance**	**3.0% Total Efficiency**	**97.6 % U-235 Abundance**	**97.6% Total Efficiency**
U-238	0.0196	0.914	0.018	0.49	0.010	0.206	0.004	0.0003	0.000
Th-234	0.0245	0.914	0.022	0.49	0.012	0.206	0.005	0.0003	0.000
Pa-234M	0.0012	0.913	0.001	0.49	0.001	0.206	0.000	0.0003	0.000
Pa-234	0.0110	0.00119	0.001	0.0005	0.001	0.0003	0.000	----------	0.000
U-235	0.0760	0.017	0.001	0.023	0.002	0.041	0.003	0.033	0.003
Th-231	0.02353	0.017	0.004	0.023	0.005	0.041	0.010	0.033	0.008
U-234	0.02401	0.068	0.002	0.49	0.012	0.753	0.018	0.967	0.023
Total	**----------**	**----------**	**0.050**	**----------**	**0.042**	**----------**	**0.040**	**----------**	**0.034**

Table III: Verification Measurement Results[1]

Grid	**Room**	**Direct Alpha Measurement Bq (dpm)/100 cm^2**	**L Shell X-ray measurement[2] Bq (dpm)/100 cm^2**
Q3	A56	0.2 (12) (PAINTED SURFACE)	1.0 (56)
I3	A56	0 (0) (PAINTED SURFACE)	1.3(78)
Y3	A56	0.2 (12) (PAINTED SURFACE)	1.4(83)
AA3	A56	0 (0) (PAINTED SURFACE)	1.3(79)
AG1	A56	0.5 (28) (PAINTED SURFACE)	6.8(405)[3]

1.0 Release criterion was 83 Bq (5000 dpm) uranium alpha per 100 cm^2.

2.0 The L Shell X-ray results were taken to determine if elevated activity levels of uranium, an isotope used in the area prior to the wall painting were present.

3.0 A wall scabble (volumetric) sample was taken at this location. The activity level was 480 dpm/100 cm^2 (MDA).

4.0 All activities were calculated assuming the activity was due to High Enriched Uranium(HEU). The most likely contaminant is Depleted Uranium, which has a 44% higher efficiency.

Table IV: Counting Times and MDAs for FIDLER X-ray Window Peak Counting

Window	CPM Gross Background (1)	Count Time Minutes	Location	Efficiency	MDA Bq(dpm)/100 cm^2
X Ray	393740	600	Q3 –A56	0.02	0.8 (46)
X Ray	513205	600	I3-A56	0.02	0.9 (52)
X Ray	528588	600	Y3-A56	0.02	0.9 (54)
X Ray	447928	600	AA3-A56	0.02	0.8 (54)
X Ray	544535	600	AG1-A56	0.02	0.8 (48)

1.0 The background varies directly with counting efficiency.

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Table V: Counting Times and MDAs for The FIDLER X-ray Window Gross Counting

Window	CPM Gross Background (1)	Count Time Minutes (2)	Location	Efficiency (3)	MDA Bq (dpm)/100 cm^2
X Ray	393740	600	Q3 –A56	0.031	0.5 (27)
X Ray	513205	600	I3-A56	0.031	0.5 (31)
X Ray	528588	600	Y3-A56	0.031	0.6 (40)
X Ray	447928	600	AA3-A56	0.031	0.6 (29)
X Ray	544535	600	AG1-A56	0.031	0.8 (32)

1.0 The background varies directly with counting efficiency.

Table VI: Statistical Data for Background

Data Type	**Background (cpm)**
Average	425
Median	428
Population Standard Deviation	22.9
Minimum Value	383
Maximum Value	455
Measurement Range	72
95% Lower Limit	374
95% Upper Limit	476
95% Confidence Range	102
T value	2.228

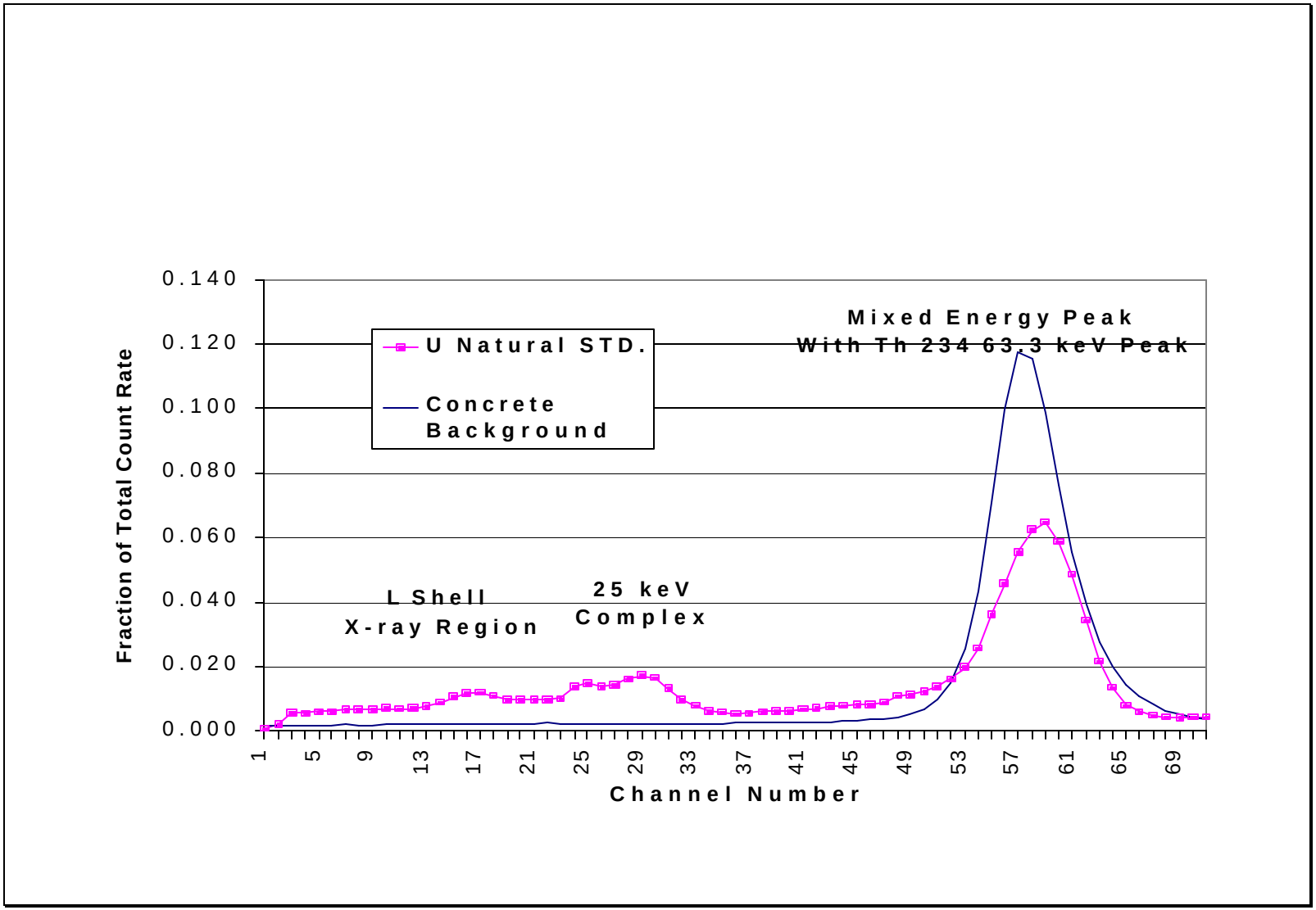

Fig. 1. Comparison of Background and Natural Uranium Spectra

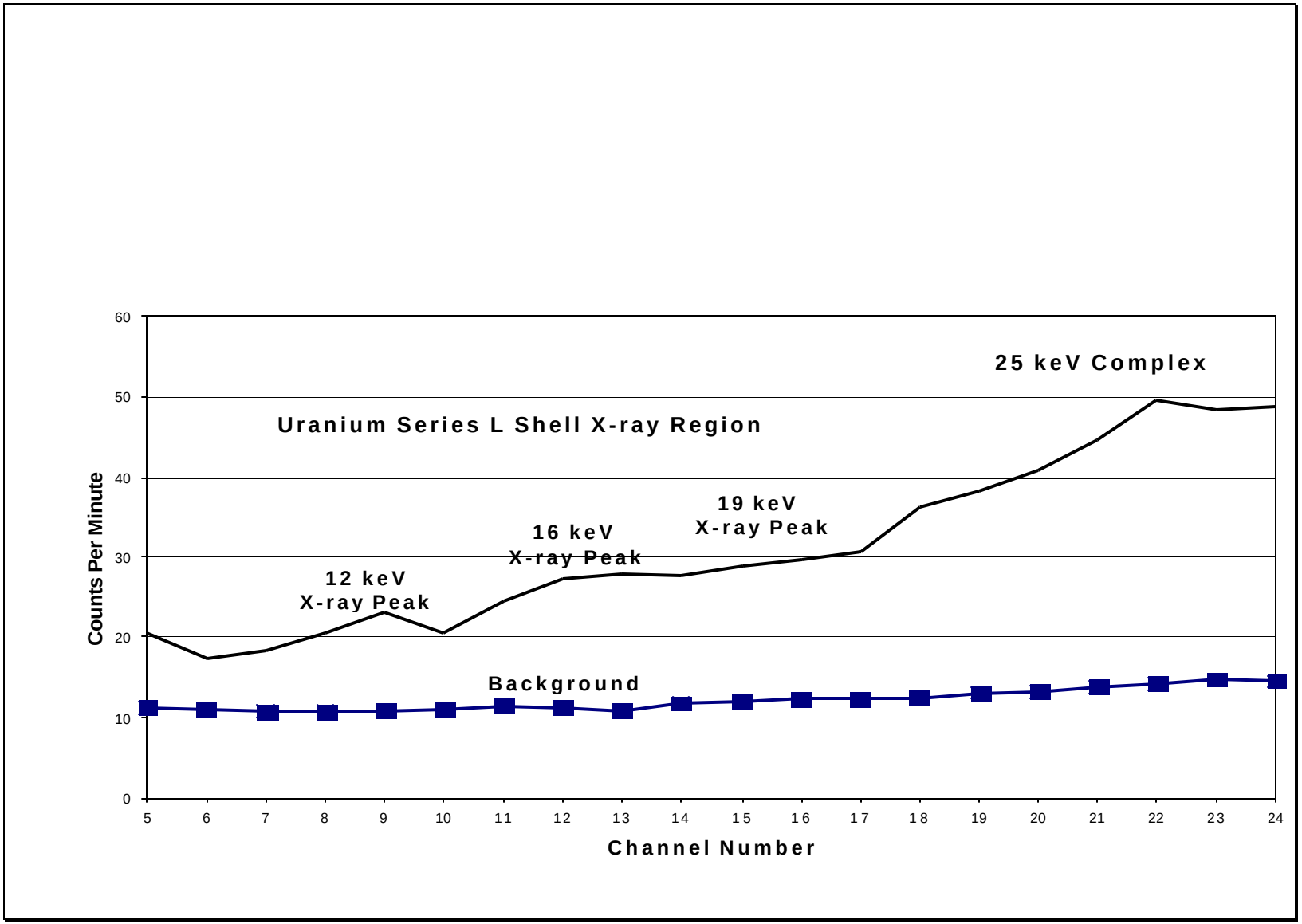

Fig. 2: Comparison of Spectra From Background Concrete and Natural Uranium in The L Shell X-ray Region

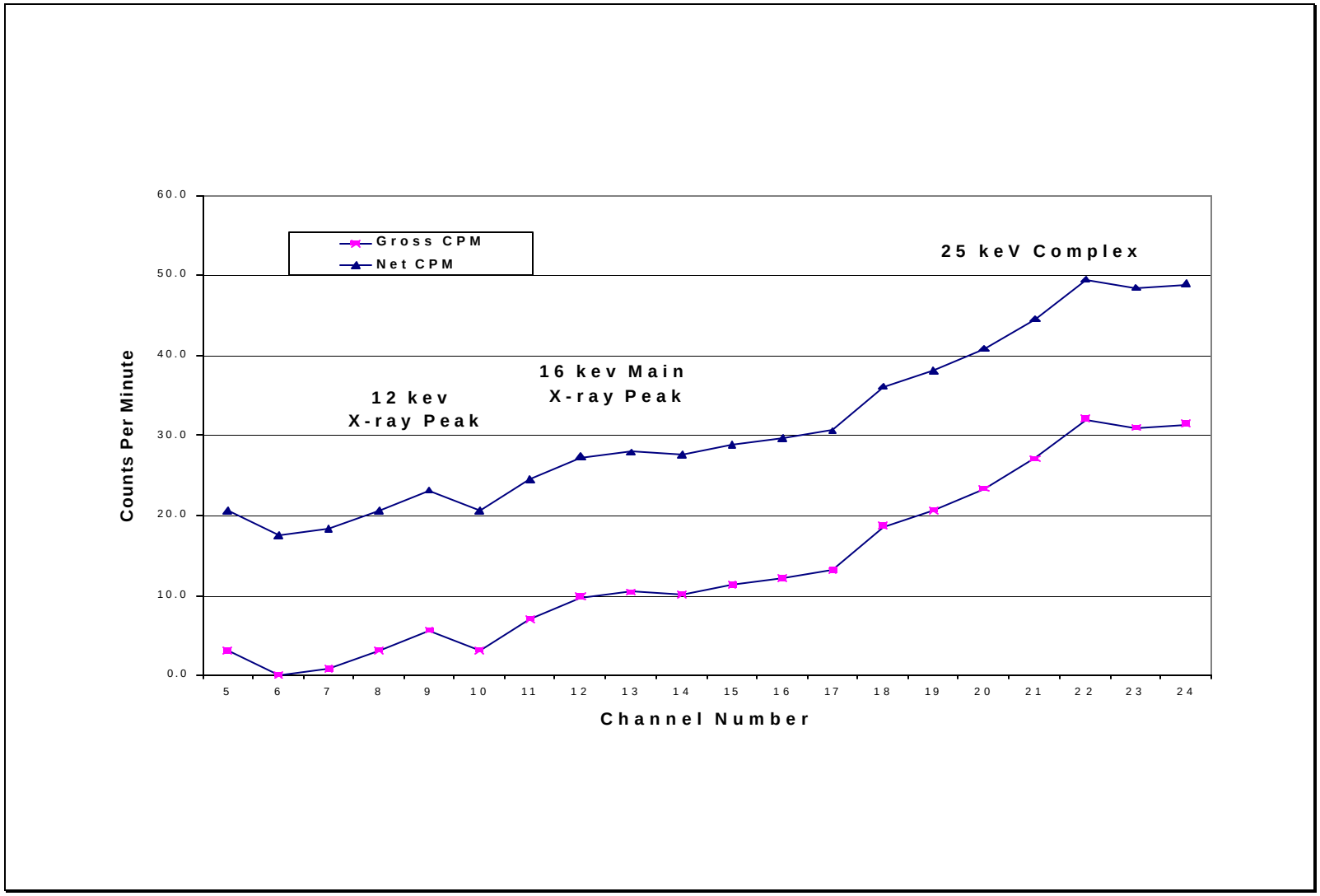

Fig. 3. Gross and Net L Shell X-ray Spectra