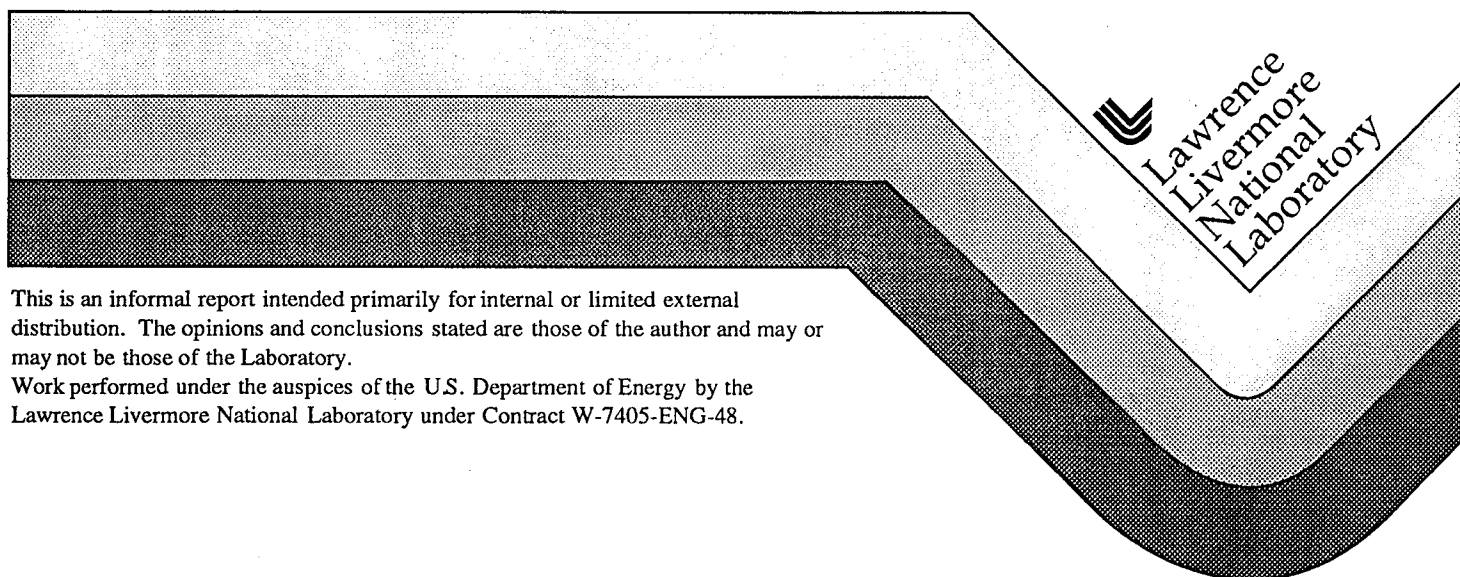


U235: A Gamma Ray Analysis Code for Uranium Isotopic Determination

DeLynn Clark

December 1996



DISCLAIMER

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

U235: A Gamma Ray Analysis Code for Uranium Isotopic Determination

DeLynn Clark

Isotope Sciences Division

Lawrence Livermore National Laboratory

University of California

December 1996

Abstract:

A ^{235}U analysis code, U235, has been written that can nondestructively determine the percentage of ^{235}U in a uranium sample from the analysis of the emitted gamma rays. The code is operational and work is now underway to improve the accuracy of the calculation, particularly at the high ($>90\%$) and low ($<0.7\%$) ^{235}U concentrations. A technique has been found to evaluate low ^{235}U concentrations that works well on the existing standards. Work is now under way to evaluate this technique for other detectors and other types of samples. Work is also proceeding on: (1) ways to better determine gamma backgrounds, (2) techniques to determine the equivalent thickness of the sample to correct for gamma attenuation, (3) evaluation of the existing data base of branching ratios of ^{235}U , ^{238}U and their daughters gamma rays to allow better results and (4) evaluation of the existing data base on the emission ratios for uranium, thorium, and protactinium x-rays.

Introduction:

Gamma ray spectrometry can be used to analyze uranium isotopic abundance ratios. The "standard" uranium enrichment meter relies on making standards of the various sample types of interest. Analyzing these standards with mass spectrometry to find the appropriate calibration factors is then done to calibrate out all the unknowns in the counting scheme. Then the strong ^{235}U gamma peak at 185.712 keV can be counted with a "simple" two channel analyzer to find the peak counts and background. The net 185.715 counts are used to calculate the enrichment. This technique works well but has the draw back that new "standards" have to be made for each different geometry and analyzed by mass spectrometry. This calibration process is often very time consuming and costly as well as being limited to "calibrated" geometries.

Accurate analysis of a radioactive sample by high resolution Germanium detector spectrometry requires correct information on the gamma ray and x-ray branching ratios for the radionuclides in the sample. ^{235}U and ^{238}U sample analysis is complicated in that the gammas observed often come from their radioactive daughters produced by successive alpha and beta decays. In addition to gamma decay these elements decay by internal conversion, IC, and subsequent

emission of daughter product x-rays. For example, when ^{235}U alpha decays the result is a radioactive ^{231}Th nucleus. This thorium isotope decays by both gamma emission and internal conversion (IC). Internal conversion results in an electron being ejected; usually from the K-shell; but L, M etc. shell conversion are also possible. This ejected electron leaves a hole in the atomic orbit that is filled by the fall of another electron from a higher electron orbital, giving rise to a thorium x-ray spectrum associated with the decay. In addition, x-rays are also produced by gammas interacting (via the photoelectric effect) in the material itself—so called fluorescent x-rays. In the case of a pure uranium sample these will be uranium x-rays. Internal conversion processes give rise to characteristic x-rays of the daughter product (not the parent) and are not proportional to the amount of material (the amount of thorium in a decaying sample of purified uranium is very small) in the sample. Internally conversion induced x-rays are proportional to the number of decays; i.e. each decay has a fractional output of x-rays of the daughter product regardless of the parent material present in the sample. This fact makes these x-rays usable for isotopic analysis if the sample has a very low concentration of daughter material (Th). To accurately use these IC x-ray peaks requires that the thorium present in very old “natural” uranium samples be removed. X-rays induced by the photoelectric effect (fluorescent x-rays) have energies characteristic of the bulk material—being proportional to the mass of material present in the source. The observed x-rays, from both fluorescent and internally converted sources, must originate within a mean free path of the surface to be easily observed.

Branching ratio and gamma, x-ray energy data have been published in various places^{1,2,3,4} for ^{235}U and ^{238}U and some of their daughter products; but this data is sometimes incomplete, or of inadequate accuracy. The current status of this data are summarized in Appendix A.

Figure 1 shows the main decay scheme for ^{235}U and ^{238}U . Pure ^{238}U emits only a 49.55 keV gamma that is so weak as to be almost useless for analysis (see Figure 2.). Fortunately, ^{238}U alpha (and beta) decays so that in within a few months it is in equilibrium with the ^{238}U decay and there are gammas from ^{234}Th , ^{234}Pa and ^{234}U available for analysis.(see Appendix E for a discussion of equilibrium). The small percentage (.0057%) of natural ^{234}U typically observed is due to the constant decay of ^{238}U . Similarly ^{235}U relatively quickly decays to equilibrium with its daughters, ^{231}Th and ^{231}Pa . Samples of uranium that have been enriched or separated can be analyzed for their ^{235}U concentrations by using these daughter product decays in all cases except very fresh (<2 month old) samples. At present the only way to accurately measure “fresh” samples before equilibrium is established is to use mass spectrometry.

Alternately, using high resolution gamma spectrometers, the spectra can be measured and the $^{235}\text{U}/^{238}\text{U}$ ratio determined by finding the peak intensities of neighboring gamma (or x-ray)

¹ Firestone, B. F., ed., “Table of Isotopes”, 8th Edition Lawrence Berkeley Laboratory, John Wiley & Sons, 1996

² Decay Data of the Transactinium Nuclides, Technical Report 261, IAEA, 1986

³Roy, J. C., et. al. *Int. J. Appl. Radiation Isotopes* , 35, pg 899, 1984

⁴Lammer, M. and Schwerer, O. Handbook of Nuclear Data for Safeguards”, INDC (NDS) - 248, IAEA, 1991

peaks from each isotope. By taking intensity ratios on gamma peaks very close to the same energy, the detector efficiency and gamma attenuation differences in the sample will be small and to first order cancel.

When referring to the ^{238}U peaks in the following discussion the assumption is made that the gamma spectrum is in equilibrium with daughters ^{234}Th (24.1 d), ^{234}Pa (6.70 hr.), $^{234\text{m}}\text{Pa}$ (1.17 min.), but not ^{234}U ($2.457\text{E}5$ yr.) and its daughters. Similarly, the ^{235}U spectrum is assumed to be in equilibrium with its daughter ^{231}Th (25.52 hr.), but not ^{231}Pa ($3.276\text{E}4$ yr.) and its daughters.

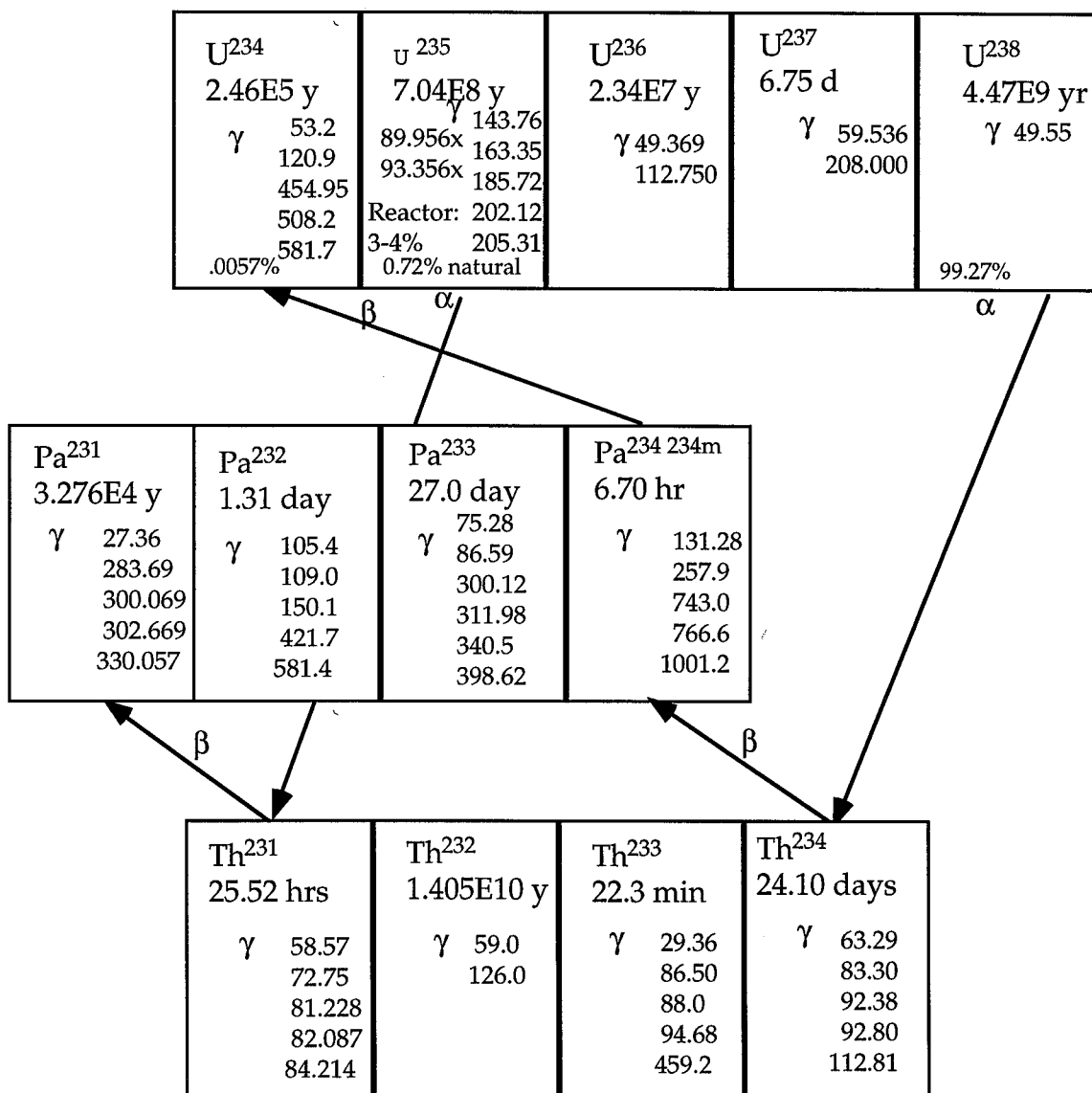


Figure 1. ^{235}U and ^{238}U decay scheme showing their principal daughters, their half-lives and their principal gamma rays.

The isotopic abundance is related to the observed peak intensities by the following relation:

$$I_1 = \lambda_1 A_1 B_1 \Omega_1 \epsilon_1 \tau_1 \text{ (counts/sec)}$$

Where:

I_1 = measured peak intensity of isotope one

$\lambda_1 = .6932/T_{1/2}$ = decay constant of isotope one

$T_{1/2}$ = material half-life (in seconds) of isotope one

A_1 = Number of atoms of isotope one

B_1 = branching ratio of isotope one

Ω_1 = Fractional solid angle of detector

ϵ_1 = Gamma counting efficiency of isotope one

τ_1 = Gamma transmission to detector

The isotopic ratio is given by the following equation:

$$A_1/A_2 = I_1 \lambda_2 B_2 \epsilon_2 \tau_2 / I_2 \lambda_1 B_1 \epsilon_1 \tau_1$$

Where:

A_1/A_2 = isotopic ratio

$\lambda_1 = .6932/T_1$ = decay constant

T_1 = material one half-life (in seconds)

I_1 and I_2 = measured peak intensities from isotope 1 and 2 respectively

T_1 and T_2 = half lives, in the same time units, of isotope 1 and 2 respectively

ϵ_1 and ϵ_2 = gamma counting efficiencies of isotope 1 and 2 respectively

B_1 and B_2 = branching ratios for characteristic gamma rays of isotope 1 and 2 respectively

Analysis is greatly simplified by the following observations:

$\epsilon_2 \tau_2 / \epsilon_1 \tau_1 \approx 1$ if the two gammas are close to the same energy

$\Omega_1 = \Omega_2$ The fractional solid angle of detector is the same for both gammas and cancels out.

λ_1 and λ_2 are known from the previously measured half lives.

B_1 and B_2 are known from the previously measured branching ratios

I_1 and I_2 have to be determined extremely accurately to get precise isotopic ratios.

The analysis proceeds on the assumptions that the solid angle terms cancel out and the half-lives and branching ratios of the respective gamma rays and x-rays can be determined. The efficiencies for detecting gamma rays are harder to determine; involving the intrinsic detector efficiencies and the overall detector and counting geometry used to obtain the data. Gamma and x-ray transmissions are nearly equal for energies close to each other. Fortunately, for gammas and x-rays close in energy the ratio of these terms, $\epsilon_2\tau_2 / \epsilon_1\tau_1 \approx 1$. Approximate detector efficiencies and gamma transmission corrections are used to make first order corrections to this ratio. The accuracy of determining the isotopic ratio, A_1/A_2 , is largely determined by the accuracy of determining the respective peak intensities, I_1/I_2 . The U235 code determines these peak intensities as accurately as possible and then applies the second order corrections for efficiency and transmission differences between the ratioed peaks to get as accurate an answer as possible.

A literature survey of ^{235}U and ^{238}U gammas and their daughter products found a number of peak groupings that could potentially provide information on isotopic ratios. These are listed in Appendix A. The survey also found that the state of information on the branching ratios and energies of ^{235}U and ^{238}U and their daughter products gammas (and x-rays) were of mixed quality.

The ^{235}U Analysis Code—U235

The ^{235}U Analysis Code—U235 was written to accomplish three main goals: first develop a tool that can accurately determine uranium isotopic ratios; second extend the applicability of the code to very low ^{235}U concentrations (depleted sources) and third to very high ^{235}U concentration (enriched) sources. Presently the code works for uranium samples that are 0.05% ^{235}U to 95% ^{235}U . Code algorithms have been found that very precisely subtract the "background" signal and fit the observed peak shapes. X-rays were fit with a Voigt profile, the shape resulting from the Lorentzian profile emitted by the x-rays and the gaussian detector response. Gammas were fitted with a gaussian profile and a low energy exponential tail. (see Figure 5 and 6 for examples of these profiles). Techniques were developed to unfold the complex peak multiplets observed in the spectra using mathematical descriptions of the peak shapes and utilizing Taylor series minimization to fit the observed data as accurately as possible.

There are several potential energy regions in the uranium gamma ray spectra that can be used to calculate isotopic abundance ratios. In this report only gammas (and x-rays) less than 300 keV are considered. This energy region is measured by a "typical" low energy Ge detector set with a gain of .075 keV/channel and 4096 channels of data. A number of $^{235}\text{U} + ^{238}\text{U}$ spectra were available for this detector arrangement, making it a "natural" first place to start analysis. The only serious limitation this energy range imposes is the relative few ^{238}U (and daughters) peaks less than 300 keV. Fortunately there are two relatively strong $^{238}\text{U}/^{234}\text{Th}$ lines at 92.365 and 92.790 keV and a relatively strong IC x-ray at 93.356 keV $^{235}\text{U}/\text{Th-}\alpha 1$. (See Appendix A) Higher energy analysis suffers from the opposite problem, there are very few useful ^{235}U peaks above 205 keV. One of the disadvantages of using gammas in the 80 to 300 keV range is their limited transmission through "thick" material. This restricts the applicability of the analysis procedures to homogenous sources or "thin" heterogeneous uranium sources.

The 20--80 keV Energy Region

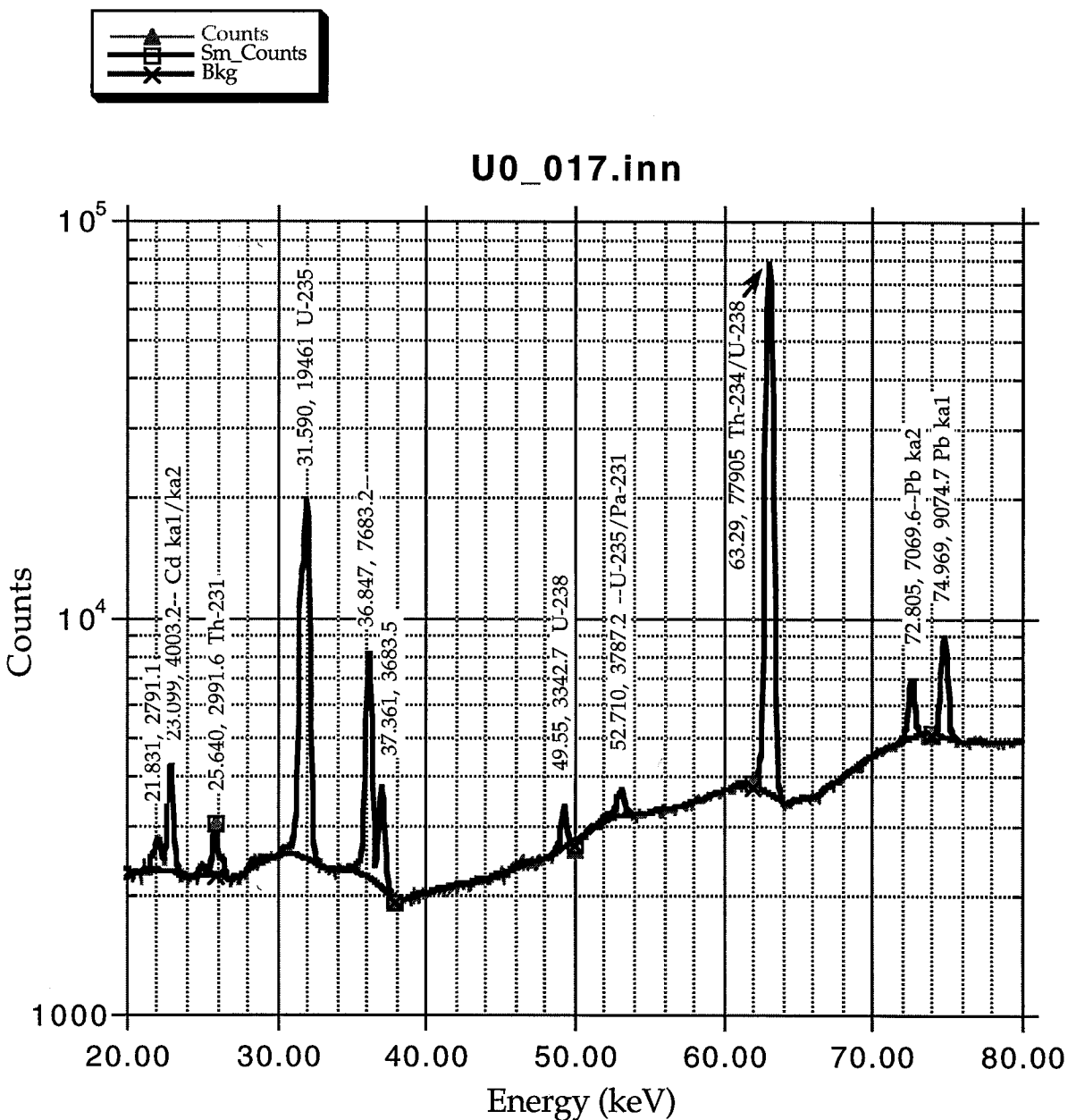


Figure 2. shows a plot of a 99.983% ^{238}U spectrum from 20 -80 keV. Clearly shown is the only gamma directly associated with the ^{238}U decay--the 49.55 keV peak. The strongest line in this region is the $^{238}\text{U} \rightarrow ^{234}\text{Th}$ daughter line at 63.29 keV. The first number on the peak labels above shows their energy, the second number their approximate peak counts and the third their origin.

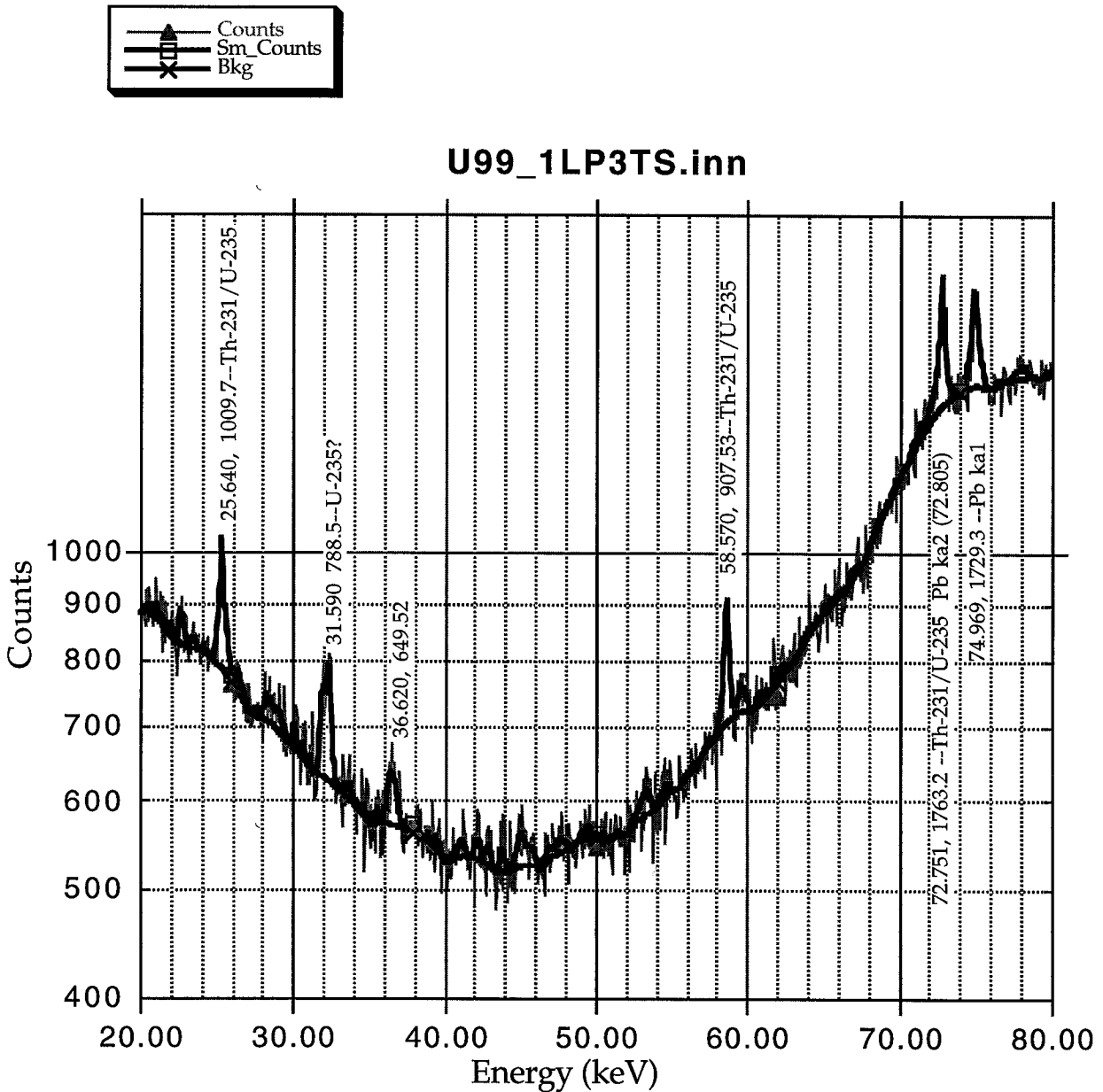


Figure 3. Shows the spectrum from a 99.1% sample of ^{235}U . It is considerably different than a ^{238}U spectrum shown above. There are no strong lines from ^{235}U or its daughters in this region. The lead $k\alpha_1$ and $k\alpha_2$ x-ray lines are a "typical" spectral contaminate resulting from fluorescent x-rays in the collimation etc.

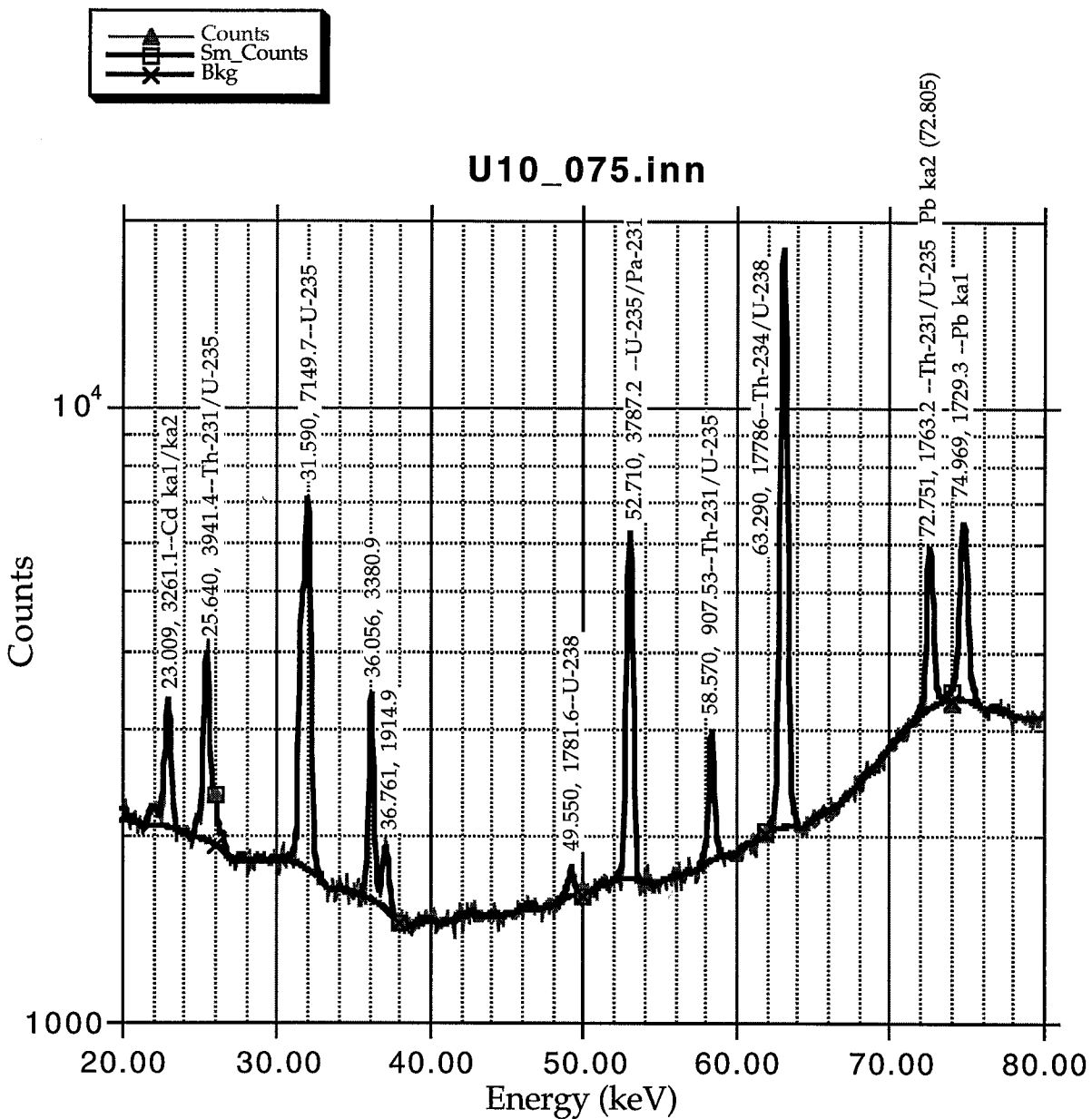


Figure 4.. Plot shows a 10.075% ^{235}U , 90.0 % ^{238}U spectrum from 20-80 keV. In this region is shown the only pure ^{238}U peak at 49.55 keV.

The 80-85 keV Energy Region

The lowest energy range of “practical” use is the 81-85 keV region. It contains peaks due to ^{235}U (81.228, 82.087, and 84.214 keV) as well as a 83.300 keV peak due to $^{238}\text{U}/^{234}\text{Th}$ decay. Figure 5. shows the spectrum of a 10.075% ^{235}U sample. Even though the lead x-rays are weak they are a typical “contaminant” to spectra in this region and have to be accounted for accurate peak intensity determinations. The 83.30 $^{238}\text{U}/^{234}\text{Th}$ peak is quite weak making its accurate determination difficult.

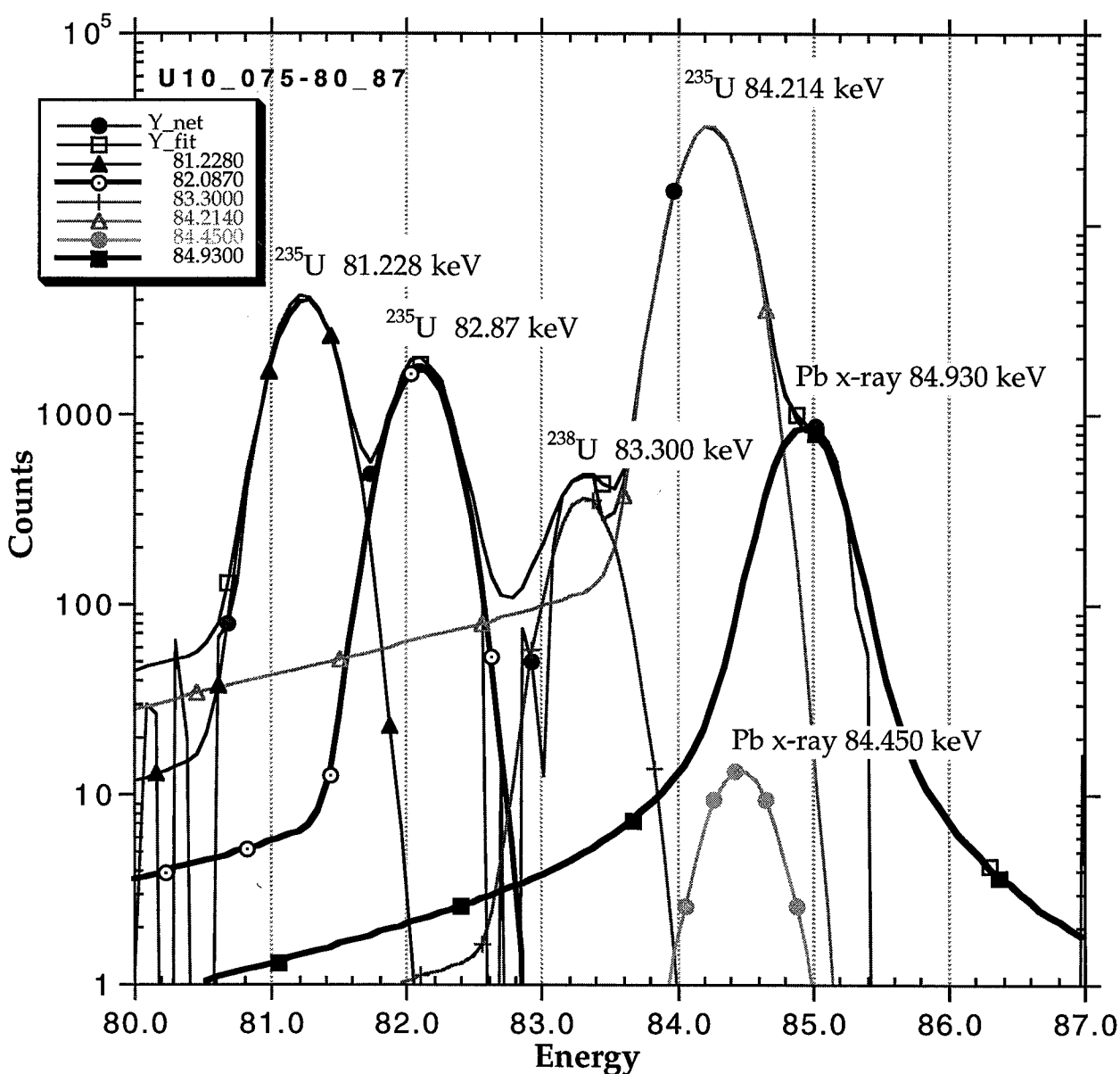


Figure 5. The net (minus background) uranium spectrum (10.075% ^{235}U) from 80 to 87 keV. As can be seen the $^{238}\text{U}/^{234}\text{Th}$ 83.300 peak is quite weak, making good peak intensity measurements difficult for this sample and/or lower concentrations of ^{238}U .

The 87-100 keV Energy Region

This region has three peaks due to ^{238}U , a number of ^{235}U peaks and the two strong U $\alpha 1$ and $\alpha 2$ x-ray peaks. The tight clustering of peaks requires careful peak fitting and analysis. For most concentrations this is the region of primary interest since the 92.365 and 92.790 ^{238}U peaks are very near the 93.356 Th- $\alpha 1$ / ^{235}U peak. The Th $\alpha 1$ and Th $\alpha 2$ x-ray peaks, due to ^{235}U decay, bracket the ^{238}U doublet. The ^{238}U 95.85 peak is so weak and has so much interference from the Pa $\alpha 1$ 95.89 keV peak as to be virtually useless as a diagnostic tool. The main limitations on using this energy range are that at both high ^{235}U and low ^{235}U concentrations the signals of either the ^{238}U peaks or the ^{235}U peaks are too small, respectively, to accurately determine.

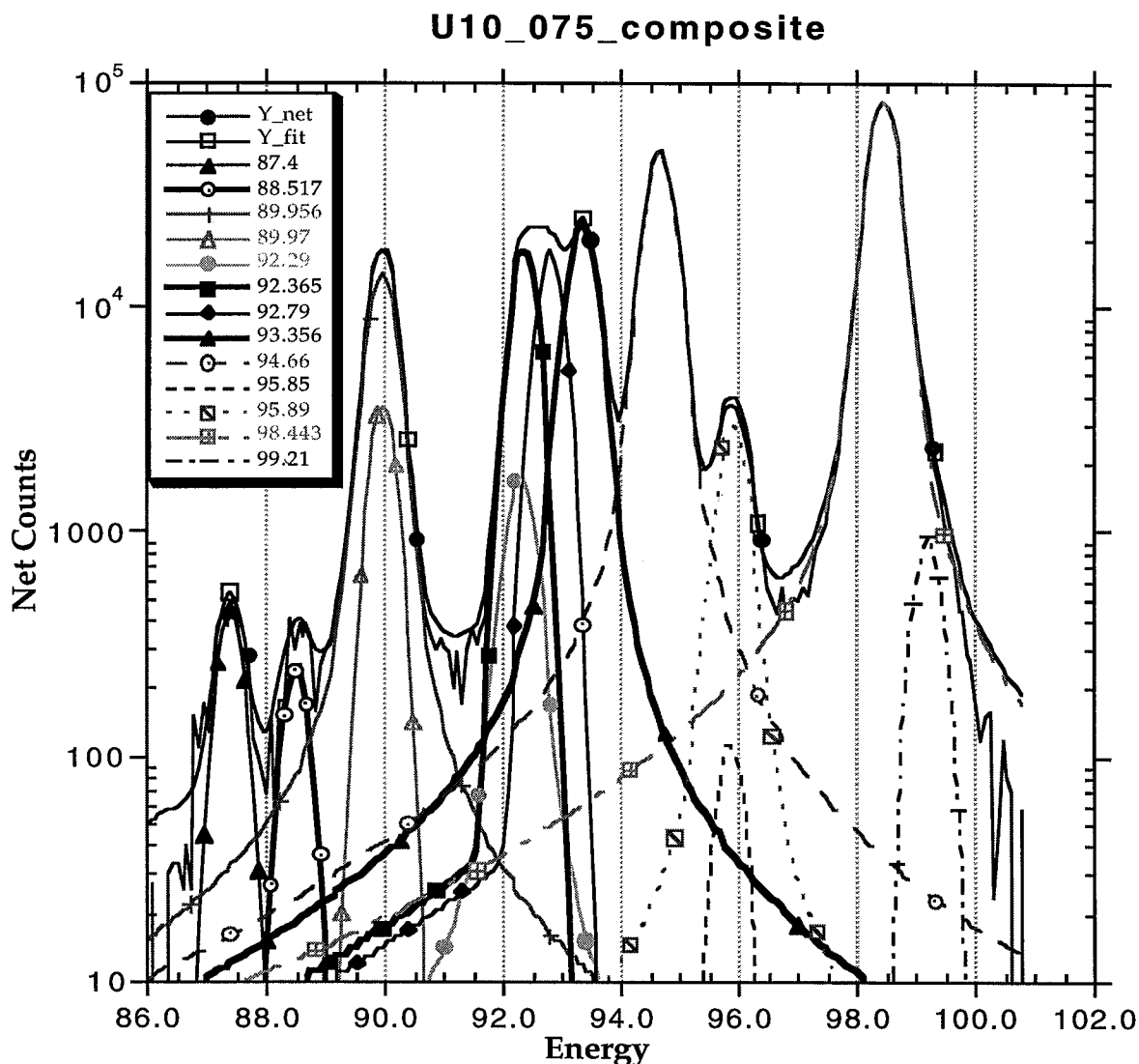


Figure 6a shows all 13 peaks used in fitting the data in the 86-102 keV region. Appendix 1 gives the identification of each of the energies and where they come from. Clearly seen is the gamma ray profile of the ^{238}U peaks and the much broader Voigt x-ray profile of the ^{235}U /Th x-ray daughter peaks and the uranium x-rays.

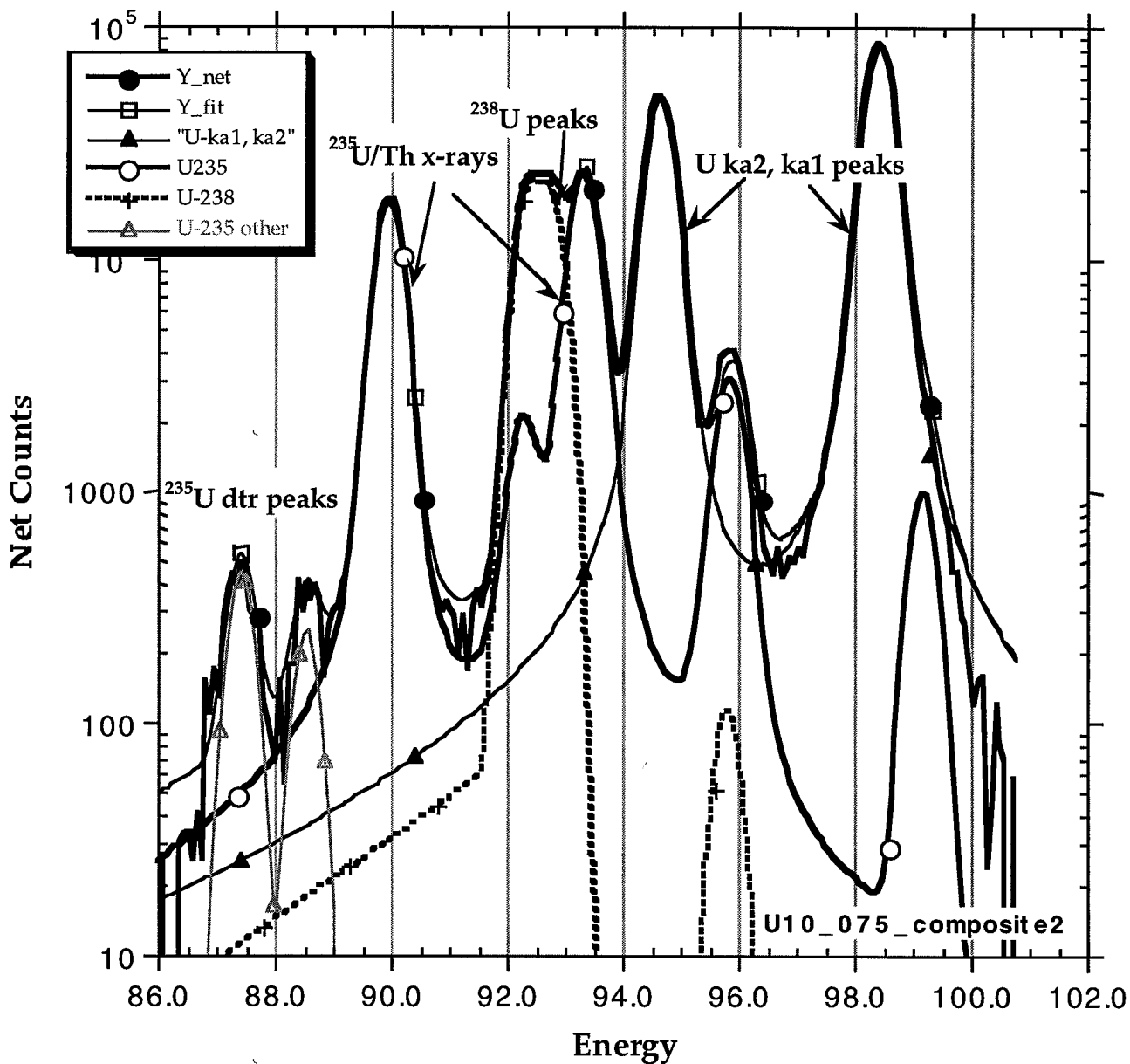


Figure 6b. The net count spectrum from 86 to 102 keV of a 10.075% ^{235}U sample with the peaks grouped into their respective components. At this ^{235}U concentration the ^{235}U and ^{238}U peaks are approximately equal. The fitting process uses both the protactinium and thorium x-rays from the ^{235}U daughters to find the best fit to the combined ^{235}U and ^{238}U spectrum.

The 100-118 keV Region

This region is very complex with 21 peaks containing all the $k\beta$ x-rays of U, Th and Pa plus a 109.2 keV gamma from ^{235}U and a 112.82 keV peak from ^{238}U . The large number of peaks and the overlap of peaks due to the wide Voight profile of the x-ray signals makes extracting useful peak ratios difficult. The Th and Pa x-ray peaks are tied to the ^{235}U decay and might give a useful data if good branching ratios were available—unfortunately they are not. Figure 7. shows the different x-ray multiplets in this region (each the sum of six x-ray peaks) and the two gamma rays. This energy region is not used in the analysis due to the difficult nature of the signals and the "poor" information available on branching ratios.

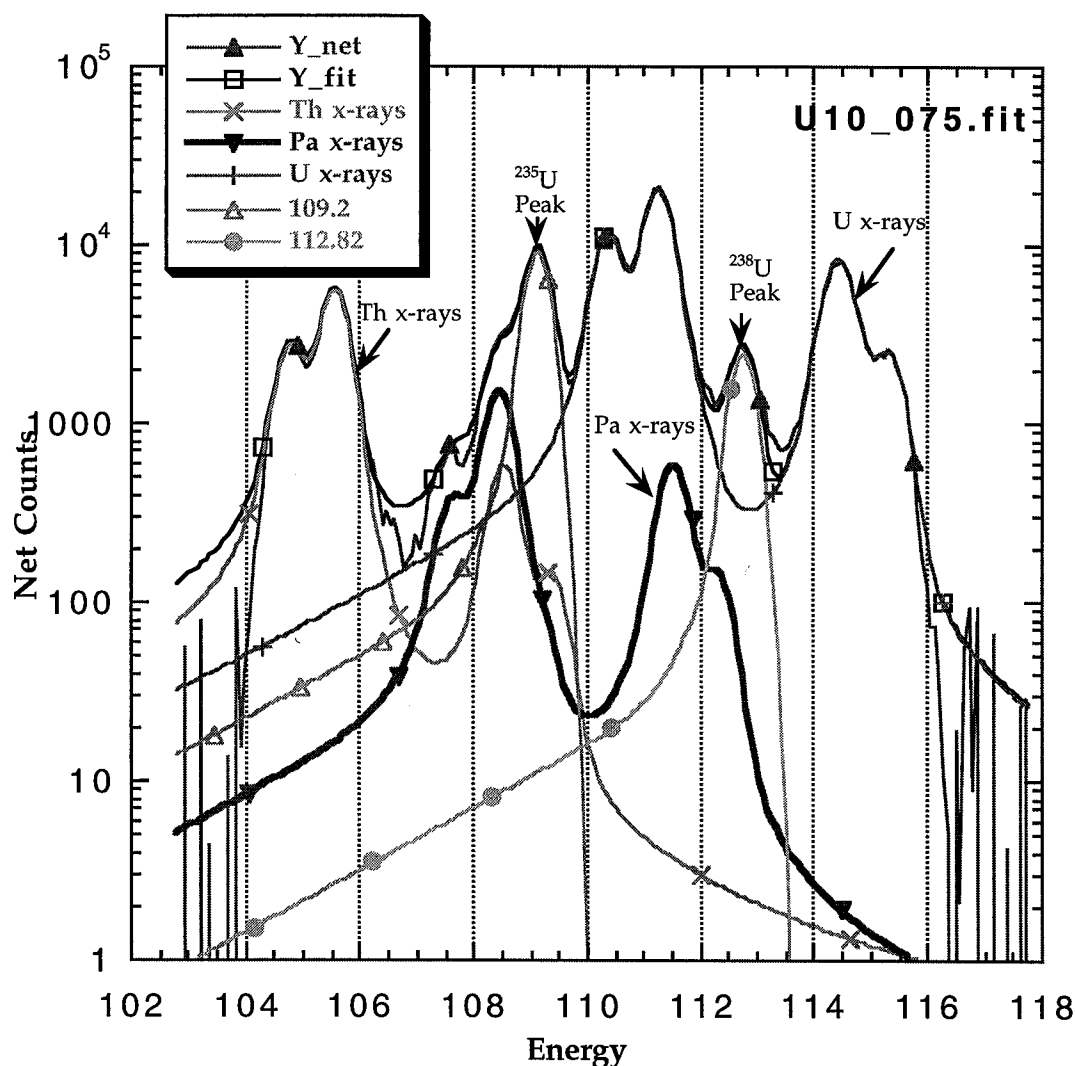


Figure 7. The net count spectrum from 102 to 118 keV of a 10.075% ^{235}U sample. In this energy range are 21 peaks, mostly thorium, protactinium and uranium $k\beta$ x-rays.

The 118-180 keV Region

The 118-180 keV region has relatively few peaks. The usually clean 120.90 keV peak of ^{234}U is useful for obtaining an estimate of that isotope. This peak suffers in that it is usually weak (sometimes too weak to analyze), giving poor statistic answers and there are no near by peaks to ratio it to. For good accuracy the 120.90 keV peak intensity needs to be corrected for efficiency and gamma transmission.

There are usually no ^{238}U /daughter peaks of sufficient intensity to be of interest in this region. The only exception is for depleted uranium spectra where the normally weak 131.300 keV $^{238}\text{U}/^{234}\text{Pa}$ peak is enhanced and the 143.760 keV ^{235}U peak is one of the cleanest ^{235}U peak available (see Figure 8b). Trying to dig the 93.35 keV $^{235}\text{U}/\text{Th}$ x-ray peak out of the 82-102 spectral regions is very inaccurate at very low ^{235}U concentrations and these isolated ^{235}U and ^{238}U /daughter peaks in the 118-180 keV region can be more accurately analyzed. The 143.76 and 163.33 keV ^{235}U peaks are also potentially useful for establishing the average material thickness in the sample by analyzing their relative intensities. Both of these techniques are utilized in the U235 code for low ^{235}U concentrations and transmission corrections.

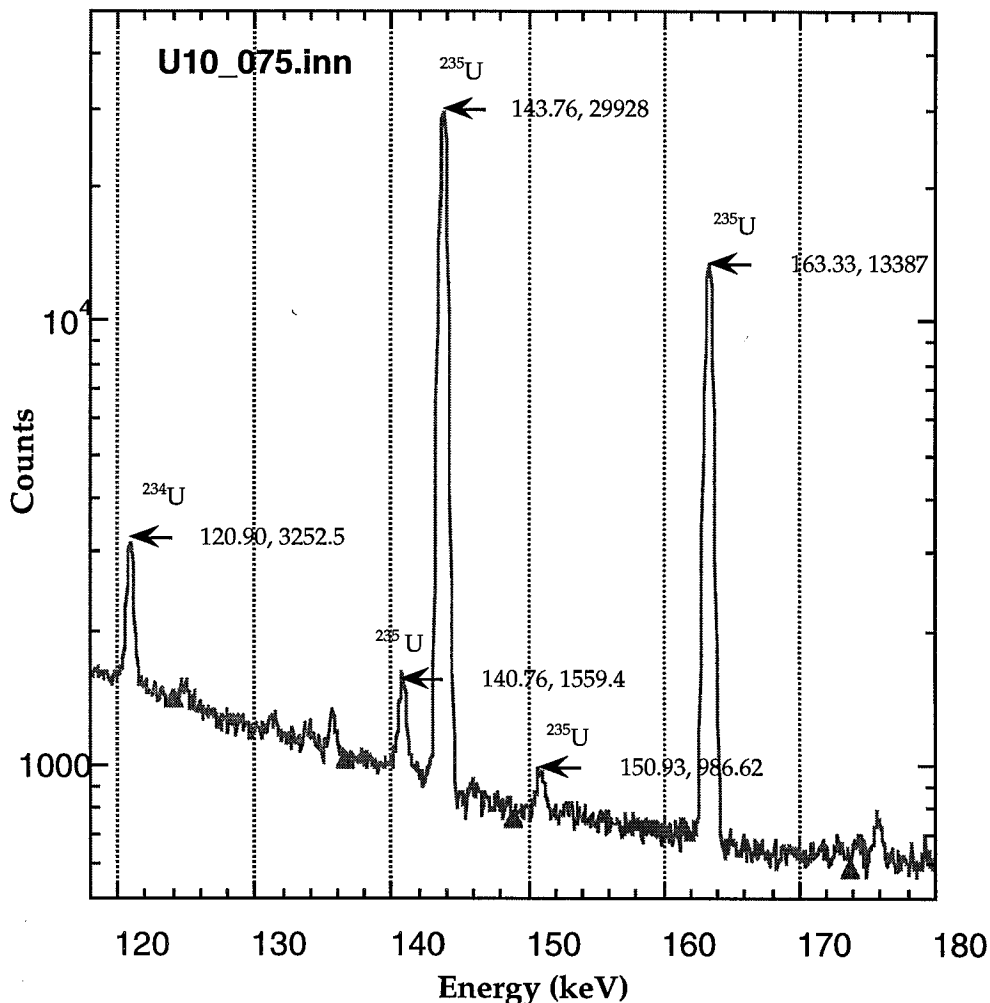


Figure 8a. The gamma spectrum from 118 to 180 keV of a 10.075% ^{235}U sample. Peaks are rather sparse in this region with usable ^{238}U peaks mainly showing up at low ^{235}U concentrations. The

^{234}U peak at 120.90 keV is usually quite weak but can usually be analyzed because of its isolation. Lack of good statistics on this peak may limit its accuracy.

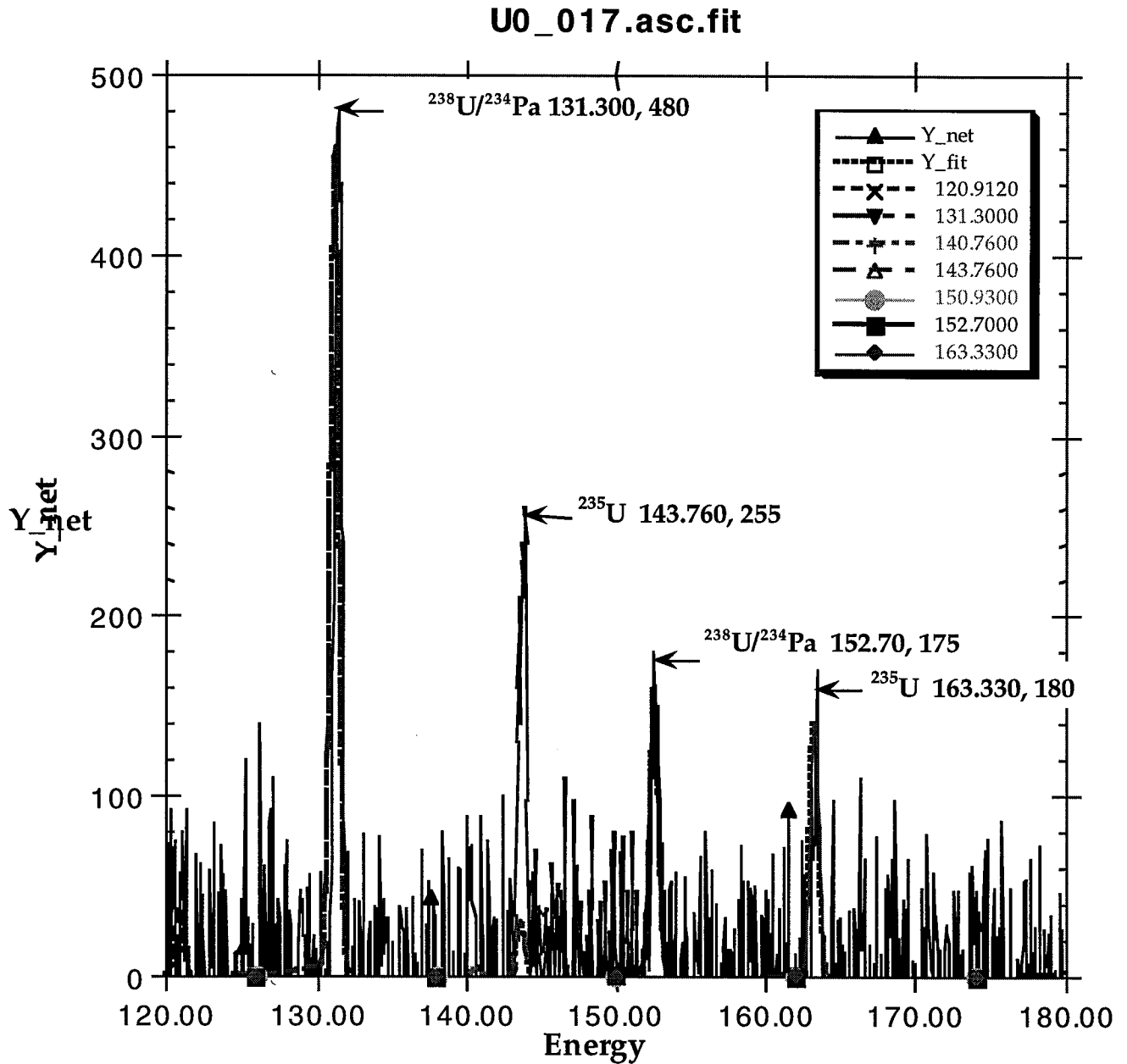


Figure 8b. showing the same 120-180 keV range for a 0.017 percent ^{235}U sample. this shows the ^{238}U peaks are enhanced; but the low count rate makes getting decent statistics for analysis very time consuming. The ^{234}U 120.90 keV peak is normally too weak to analyze at low ^{235}U concentrations.

The 180-210 keV Region

The 180-210 keV region has several prominent ^{235}U peaks including the most intense ^{235}U peak at 185.715 keV. This peak, in conjunction with the 98.443 keV uranium x-ray peak, is used to determine a more accurate gain and zero for the spectrum and to verify that ^{235}U is present in the spectrum. There are no easily observable ^{238}U peaks in this region. The only major uncertainty here is the 185.712 keV peak which has several other weak peaks around it that must be corrected for to get a good 185.715 keV peak intensity. One of the significant variations observed in this region is the 185.715 keV peak height to 188 keV background ratio. This ratio is found to vary from about 1000 at 90% enrichment to 1 at .02% enrichment. This change is attributed to the high energy gammas in ^{238}U decay and the contribution they make to the Compton continuum in this energy region. Further investigation is needed to see if these ratios are a function of sample, and detector type—or a universal characteristic of uranium spectra. This ratio is of potential usefulness in establishing a quick estimate of the ^{235}U enrichment. A spectrum with a high 185.712 to 188 keV ratio has almost certainly a "high" ^{235}U enrichment. Conversely a weak 185.715 peak with a high Compton continuum has a low ^{235}U enrichment.

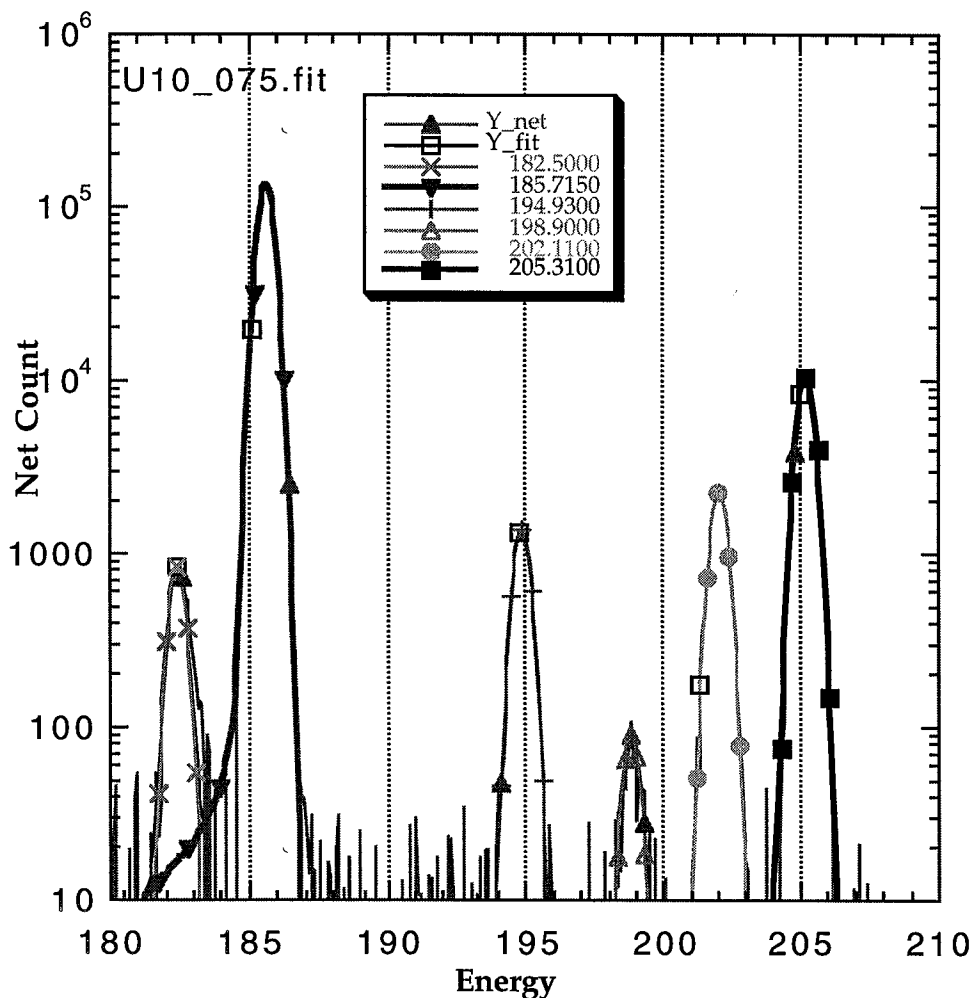


Figure 9. The net count spectrum from 180 to 210 keV of a 10.075% ^{235}U sample. In this region there are typically no ^{238}U peaks intense enough for any peak analysis.

The 210--300 keV Region

The 210-300 keV region only has one strong $^{238}\text{U}/^{234\text{m}}\text{Pa}$ peak at 258.2 keV. This peak is too weak to be of any great interest. The overall low intensity of this region lowers its utility in analyzing isotopic ratios.

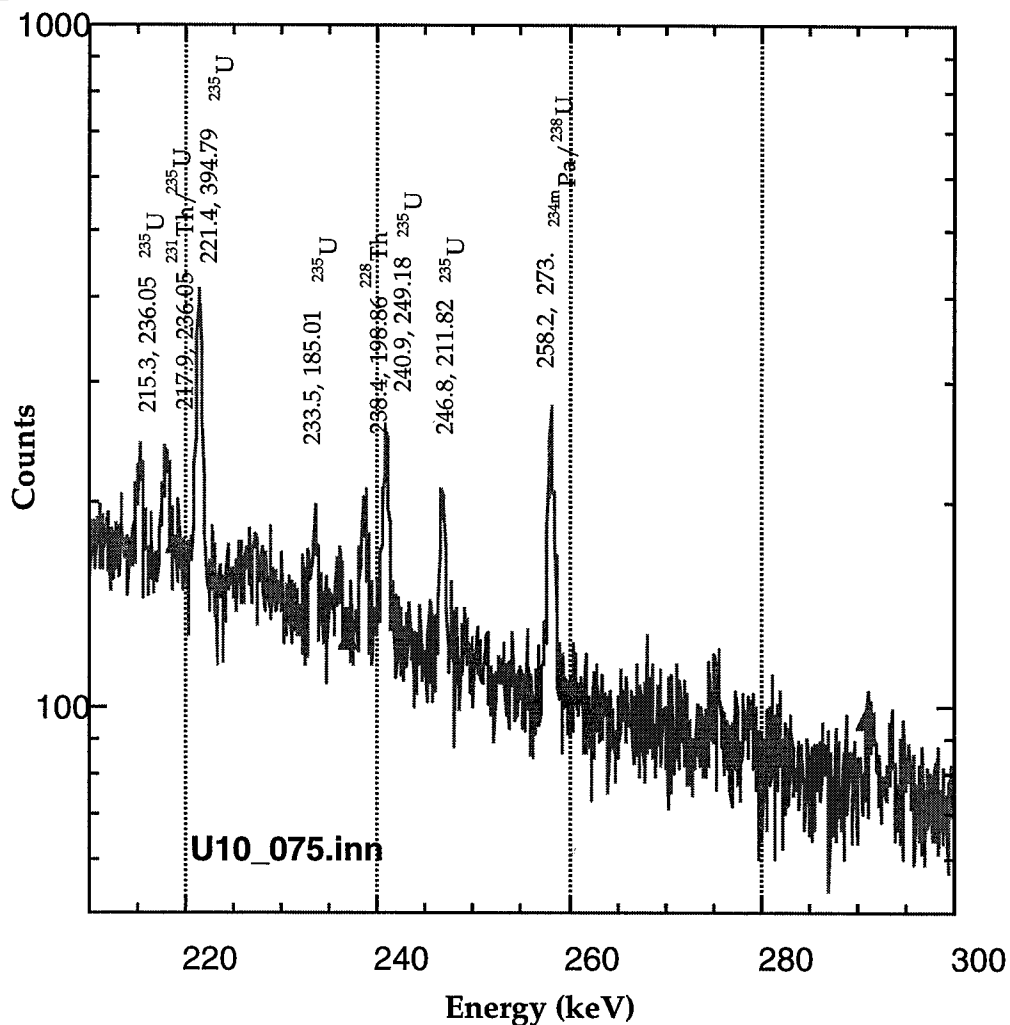


Figure 10.. The net count spectrum from 210 to 300 keV of a 10.075% ^{235}U sample. In this region there is only one ^{238}U peak of interest and a few ^{235}U peaks.

U235 Calculation Results

Table 1 shows the results of the U235 code calculations using the ^{238}U peaks at 92.29 and 92.7 keV, ratioed with the 93.356 keV $\text{Th } \alpha 1 / ^{235}\text{U}$ peak. The first column shows the calculated enrichment on a number of standard (known) sources with the isotopic composition shown in column 2-4. The data in the last column shows the percent difference between the measured and calculated enrichment. As expected the samples that are the most difficult to analyze are where there is either very small ^{235}U or ^{238}U signals.

The code has been modified at low ^{235}U concentrations to use the 131.3 keV $^{238}\text{U} / ^{234}\text{Pa}$ line and the ^{235}U lines at 143.76 and 163.33 keV to obtain a more accurate estimate of the $^{235}\text{U} / ^{238}\text{U}$ ratio for low ^{235}U concentrations. Normally the 131.3 keV $^{238}\text{U} / ^{234}\text{Pa}$ line is too weak to analyze; but at low concentrations the long counting time required to accumulate enough counts combined with the low ^{235}U emission rate make this line analyzable. It has the additional advantage that there are no competing lines near it; hence even though weak, it is unambiguous to analyze.

Table 1
U235 Code Preliminary Calculation Results

Calc. U235	Standard % ^{235}U	Standard % ^{234}U	Standard % ^{238}U	Std-Calc % diff.
0.0169	0.017	0.001	99.982	-0.59%
0.5133	0.483	0.005	99.512	6.27%
1.0126	0.991	0.008	99.001	2.18%
2.0302	2.013	0.016	97.971	0.85%
2.9991	3.009	0.031	96.960	-0.33%
5.0127	4.949	0.041	95.010	1.29%
10.0806	10.075	0.077	89.848	0.06%
49.1959	49.380	0.672	49.948	-0.37%
75.4537	75.130	0.584	24.286	0.43%
92.982	93.076	1.452	5.472	-0.10%

Summary:

Algorithms are being developed to better subtract backgrounds and improve the high concentration ^{235}U results. Future plans include expanding the coverage to higher than 300 keV for more complicated spectra and expanding the code to handle mixed Pu and U spectra better.

Appendix A
Gamma and X-ray Decay of ^{235}U and ^{238}U and their Daughters
(data from 49-300 keV)

The gammas energies listed in bold are used with these branching ratios listed in column three to determine $^{235}\text{U}/^{238}\text{U}/^{234}\text{U}$ ratios by the U235 code. All x-rays listed as IC-decay are internally converted in the isotope and decay with the isotopes decay characteristics—half-life and isotopic composition. All x-rays labeled as fluorescence are caused (nearly completely) by photoelectric absorption in the material and subsequent L-K shell electron decay. These x-rays are characteristic of the physical properties of the material and not its isotopic composition. Gammas and x-rays are listed by energy. This should allow quicker identification of observed spectra and may help pinpoint potential interference in a given measurement. Only the "strongest" lines are listed, many other gammas in this range are normally too weak to observe. These lines will occur with different intensities depending upon the isotopic concentration being observed. The branching ratios listed in column three are the ones presently used at LLNL.

						URADOS ^c	IAEA ^a
				Source	Parent	Branch Ratio - BR x 100	Branch Ratio x100
Group-1							
	E(keV)	Branch Ratio — BR x 100	g=0 x=-1				
1	49.550	0.064	0	U-238	U-238		0.064 ± 0.008
2	53.200	0.123	0	U-234			0.123 ± 0.002
3	58.570	0.500	0	Th-231	U-235	0.46 ± 0.060	0.5 ± 0.05
4	63.290	4.470	0	Th-234	U-238	3.94 ± 0.010	4.47 ± 0.88
5	72.751	0.260	0	Th-231	U-235		0.26 ± 0.02
6	72.804	27.700	-1	Pb- α 2	fluorescence		
7	73.920	0.202	0	Pa-234m	U-238		
8	74.000	0.036	0	Pa-234	U-238		
9	74.910	0.510	0	U-235	U-235		
10	74.969	46.200	-1	Pb- α 1	fluorescence		
11	81.228	0.850	0	Th-231	U-235		0.85 ± 0.03
12	82.087	0.370	0	Th-231	U-235		0.37 ± 0.02
13	83.300	0.073	0	Th-234	U-238	0.064 ± 0.10	0.073
14	84.214	6.710	0	Th-231	U-235		6.71 ± 0.1
15	84.450	5.580	-1	Pb- α 3	fluorescence		
16	84.930	10.700	-1	Pb- α 1	fluorescence		
17	87.300	3.910	-1	Pb- α 2	fluorescence		
Group-2							
	E(keV)	BR x 100	g=0,x=1	Source	Parent	URADOS	IAEA
1	87.700	0.050	0	Pa-231	Th-231/U-235		
2	88.500	0.030	0	Th-227	U-235		
3	89.956	3.360	-1	Th α 2	U-235 IC-decay	3.17 ± 0.08	3.4 ± 0.8
4	89.970	0.742	0	Th-231	U-235	0.97 ± 0.05	
5	92.290	0.470	-1	Pa α 2	U-235 IC-decay	0.451 ± 0.036	0.39 ± 0.03
6	92.365	2.600	0	Th-234	U-238	2.52 ± 0.06	2.60 ± 0.53
7	92.790	2.560	0	Th-234	U-238	2.50 ± 0.06	2.56 ± 0.52
8	93.356	5.500	-1	Th α 1	U-235 IC-decay	5.22 ± 0.14	5.6 ± 1.3
9	94.660	9.161††	-1	U α 2	fluorescence	61.2 (norm)	28.2 ± 0.6

10	94.700	0.0321		Pa-234	U-238		
11	95.850	0.0024	0	Th-234	U-238		
12	95.860	0.880	-1	Pa $k\alpha_1$	U-235 IC-decay	0.776 ± 0.043	0.63 ± 0.05
13	98.443	14.800††	-1	U $k\alpha_1$	fluorescence	100.0 (norm)	45.1 ± 0.9
14	99.270	0.400	0	Th-231	U-235	$0.14 \pm .03$	

Group-3	E(keV)	BR x 100	g=0,x=1	Source	Parent	URADOS	IAEA
1	102.270	0.400	0	Th-231	U-235		0.40 ± 0.02
2	104.819	0.137	-1	Th $k\beta_3$	IC-decay		
3	105.604	0.262	-1	Th $k\beta_1$	IC-decay		
4	106.239	0.009	-1	Th $k\beta_5$	IC-decay		
5	107.595	0.022	-1	Pa $k\beta_3$	IC-decay		
6	108.422	0.042	-1	Pa $k\beta_1$	IC-decay		
7	108.582	0.100	-1	Th $k\beta_2$	IC-decay		
8	108.955	0.003	-1	Th $k\beta_4$	IC-decay		
9	109.072	0.002	-1	Pa $k\beta_5$	IC-decay		
10	109.160	1.540	0	U-235	U-235		1.54 ± 0.05
11	109.442	0.022	-1	Th KO2_3	IC-decay		
12	110.480	0.555	-1	U $k\beta_3$	fluorescence		
13	110.500	.0043	-1	U-238	U-238		
14	111.350	1.000	-1	U $k\beta_1$	fluorescence		
15	111.486	0.017	-1	Pa $k\beta_2$	IC-decay		
16	111.870	0.001	-1	Pa $k\beta_4$	IC-decay		
17	111.964	0.037	-1	U $k\beta_5$	fluorescence		
18	112.380	0.004	-1	Pa KO2_3	IC-decay		
19	112.820	0.040	0	Th-234	U-238		0.256 ± 0.054
20	114.540	0.388	-1	U $k\beta_2$	fluorescence		
21	114.844	0.011	-1	U $k\beta_4$	fluorescence		
22	114.900	0.0064	0	Pa-234m	U-238		
23	115.377	0.089	-1	U KO23	fluorescence		

Group-4	E(keV)	BR x100	g=0,x=1	Source	Parent	URADOS	IAEA
1	120.900	0.0342	0	U-234		0.041 ± 0.006	0.0342 ± 0.0005
2	124.914	0.0600	0	Th-231	U-235		0.06 ± 0.003
3	131.300	0.0286	0	Pa-234	U-238		
4	134.030	0.0250	0	Th-231	U-235		0.025 ± 0.005
5	135.664	0.0840	0	Th-231	U-235		0.084 ± 0.007
6	140.760	0.2200	0	U-235			0.22 ± 0.02
7	143.760	10.9600	0	U-235		10.95 ± 0.15	10.96 ± 0.08
8	150.930	0.0800	0	U-235			$.08 \pm 0.02^d$
9	152.700	0.0083		Pa-234	U-238		
10	163.330	5.0800	0	U-235		5.11 ± 0.05	5.08 ± 0.04

Group-5	E(keV)	BR x100	g=0,x=1	Source	Parent	URADOS	IAEA
1	182.610	0.3400	0	U-235		0.37 ± 0.02	0.34 ± 0.02
2	183.500	0.0329	0	U-235			.0329
3	184.800	0.2200	0	Th-234	U-238		
4	185.715	57.2000	0	U-235		57.2 ± 0.02	57.2 ± 0.5

5	185.900	0.0039	0	Pa-234	U-238	3.89E-3
6	194.940	0.6300	0	U-235		0.630 ± 0.01
7	198.900	0.0420	0	U-235		0.042
8	202.110	1.0800	0	U-235		1.080 ± 0.02
9	205.311	5.0100	0	U-235		5.010 ± 0.05

Group -6	E(keV)	BR x100	g=0,x=1	Source	Parent	URADOS	IAEA
1	215.30	0.0288	0	U-235			
2	217.94	0.0370	0	Th-231	U-235		0.037 ± 0.001
3	221.38	0.1200	0	U-235			0.12 ± 0.01
4	226.63	0.0059	0	Pa-234	U-238		
5	227.17	0.0055	0	Pa-234	U-238		
6	233.50	0.0290	0	U-235			
7	238.50	0.0092	0	Th-231	U-235		
8	240.85	0.0540	0	U-235			
9	246.84	0.0530	0	U-235			
10	258.20	0.0730	0	Pa-234m	U-238		
11	291.63	0.0180	0	U-235			
12	293.90	0.0039	0	Pa-234	U-238		

a. "Handbook of Nuclear Data for Safeguards", INDC (NDS) - 248, IAEA, 1991

b. GAMGEN code LLNL

c. Presented in CEA meeting by DAMPRI-LPRI May 1996

d. "Decay Data of the Transactinium Nuclides", Report # 261, IAEA, 1986

²³⁸U and Daughters— ²³⁴Pa and ²³⁴Th

GAMGEN Calculation showing gammas/sec/gram of ²³⁸U (g/s/gm) at 5 years since separation and the implied Branching Ratio.(Branching Ratio normalized to 2.60% at 92.3 keV)

E(keV)	g/s/gm	g=0,x=1	BR x100	Source	Parent1	Emitter2	Parent2
62.9	2.36	0	0.0182	Th-234	U-238		
73.9	1.36	0	0.0105	Pa-234m	U-238		
74.0	5.30	0	0.0408	Th-234	U-238		
83.3	8.71	0	0.0670	Th-234	U-238		
92.3	338.0	0	2.6000	Th-234	U-238		
92.8	335.0	0	2.5769	Th-234	U-238		
94.7	21.6	0	0.1662	Pa-234m	U-238	Pa-234	U-238
95.9	1.62	0	0.0125	Th-234	U-238		
110.5	2.98	0	0.0229	U-238	U-238		
114.9	4.32	0	0.0332	Pa-234m	U-238	Pa-234	U-238
131.3	3.23	0	0.0248	Pa-234	U-238		
152.7	1.08	0	0.0083	Pa-234	U-238		
184.8	1.49	0	0.0115	Th-234	U-238		
258.2	9.02	0	0.0694	Pa-234m	U-238		

Appendix B

Uranium and Daughter X-rays*

Th X-rays		Pa X-rays		U X-rays		
E(keV)	%	E(keV)	%	E(keV)	%	
93.350	45.400	95.863	45.3	98.434	45.1	kα1
89.957	28.100	92.282	28.1	94.654	28.2	kα2
105.604	10.700	108.422	10.7	111.298	10.7	kβ1
108.582	4.100	111.486	4.163	114.445	4.15	kβ2
104.819	5.610	107.595	5.64	110.421	5.65	kβ3
108.955	0.110	111.870	0.11	114.844	0.12	kβ4
106.239	0.380	109.072	0.389	111.964	0.397	kβ5
109.442	0.900	112.380	0.93	115.377	0.95	k02_3

% refers to percent decay per 100 k-shell vacancies

* Browne, E. and Firestone, R., "Table of Radioactive Isotopes", LBL, 2986, pg. C-23

X-rays associated with Uranium decay—sorted by energy.

Uranium x-ray fluorescence intensity is set to 1.00 for U-kα1 in this comparison. The other branching ratios are derived from the observed decay of ²³⁵U. The Pa branching ratios determined from the protactinium fluorescent decay ratios normalized to a 0.042 branching ratio for the Pa kβ1 line at 108.422 keV. The thorium branching ratios determined from the Th fluorescent decay ratios normalized to a .503 branching ratio for the Th kα1 line at 93.350 keV. [All data preliminary.]

E(keV)	%	Branch Ratio	Type x-ray		
89.957	28.100	0.3113	Th	kα2	IC-decay
92.282	28.100	0.1103	Pa	kα2	IC-decay
93.350	45.400	0.5030	Th	kα1	IC-decay
94.654	28.200	0.6253	U	kα2	fluorescence
95.863	45.300	0.1778	Pa	kα1	IC-decay
98.434	45.100	1.0000	U	kα1	fluorescence
104.819	5.610	0.0622	Th	kβ3	IC-decay
105.604	10.700	0.1185	Th	kβ1	IC-decay
106.239	0.380	0.0042	Th	kβ5	IC-decay
107.595	5.640	0.0221	Pa	kβ3	IC-decay
108.422	10.700	0.0420	Pa	kβ1	IC-decay
108.582	4.100	0.0454	Th	kβ2	IC-decay
108.955	0.110	0.0012	Th	kβ4	IC-decay
109.072	0.389	0.0015	Pa	kβ5	IC-decay
109.442	0.900	0.0100	Th	k02_3	IC-decay
110.421	5.650	0.1253	U	kβ3	fluorescence
111.298	10.700	0.2373	U	kβ1	fluorescence
111.486	4.163	0.0163	Pa	kβ2	IC-decay
111.870	0.110	0.0004	Pa	kβ4	IC-decay

111.964	0.397	0.0088	U	kβ5	fluorescence
112.380	0.930	0.0037	Pa	k02_3	IC-decay
114.445	4.150	0.0920	U	kβ2	fluorescence
114.844	0.120	0.0027	U	kβ4	fluorescence
115.377	0.950	0.0211	U	k02_3	fluorescence

E(keV)	Th X-rays measured*	Norm. meas.	Intensity %	Scofield Calculation	Th X-rays %		Calc-Meas. % diff
93.350	93.348	100	45.4	100	45.40	kα1	0
89.957	89.957	61	27.694	61.9	28.10	kα2	0
105.604	105.606	19	8.626	22.35	10.70	kβ1	2.47
108.582	108.471	10	4.54	8.5601	4.10	kβ2	2.5
104.819	104.822			11.466	5.61	kβ3	3.53
108.955					0.11	kβ4	
106.239				0.8247	0.38	kβ5	0.68
109.442					0.90	k02_3	

Pa X-rays

E(keV)					%		
95.863	95.867	100	45.3	100	45.300	kα1	0
92.282	92.284	62	28.086	62.2	28.100	kα2	-0.1
108.422	108.418	24	10.872	22.45	10.700	kβ1	2.36
111.486			0	8.6882	4.163	kβ2	2.62
107.595	107.586	11	4.983	11.472	5.640	kβ3	3.86
111.870					0.110	kβ4	
109.072				0.8441	0.389	kβ5	0.78
112.380					0.930	k02_3	

U X-rays

E(keV)					%		
98.434	98.435			100	45.100	kα1	0
94.654	94.656			62.5	28.200	kα2	0.02
111.298	111.300			22.6	10.700	kβ1	2.25
114.445				8.7462	4.150	kβ2	2.35
110.421	110.416			11.549	5.650	kβ3	3.82
114.844					0.120	kβ4	
111.964	112.043			0.8656	0.397	kβ5	0.77
115.377					0.950	k02_3	

	kα2/kα1	kβ1/kα1	kβ3/kβ1	kβ5/kβ1
Th	0.619	0.224	0.513	0.0369
Pa	0.622	0.225	0.512	0.0376
U	0.625	0.226	0.511	0.0383

Scofield, J. D. "Relativistic Hartree-Slater Values of the K and L X-ray Emission"

Atomic Data and Nuclear Data Tables 14, 121-137(1974)

*Barreau, G. et. al., "Z. Phys. A. Atoms and Nuclei" 308, 209-213 (1982)

Appendix C

^{235}U Daughter— ^{231}Th Gammas, Pa x-rays, and Branching Ratios

E(keV)	$\pm\Delta E$	IAEA E(keV)	$x=-1$	* Rel. Int ^b	$\pm\Delta I$	Notes:	Branch Ratio (IAEA)	Imp. Branch Ratio ^a
26.560		25.640		202	20		0.146	0.135542
44.100	0.3			0.06	0.04			4.03E-05
58.470	0.05	58.570		7.2	0.7		0.005	0.004831
63.700	0.2			0.68	0.14			0.000456
72.660	0.06	72.751		4	0.4		0.0026	0.002684
73.000	0.1			0.1	0.04			6.71E-05
81.180	0.05	81.228		14.2	1.4		0.0085	0.009528
82.020	0.06	82.087		7.2	0.7		0.0037	0.004831
84.170		84.214		100		Reference^a	0.0671	0.067100
89.940	0.05			15.3	1.5			0.010266
92.230	0.05		-1	6	0.6	Pa $k\alpha_2$		0.004026
93.300	0.1			0.5	0.05			0.000336
95.870	0.05		-1	10.3	1	Pa $k\alpha_1$		0.006911
99.300	0.05			2.1	0.2			0.001409
102.300	0.05	102.270		6.7	0.7		0.004	0.004496
105.730	0.1			0.14	0.02			9.39E-05
106.580	0.1			0.34	0.04			0.000228
107.620	0.1		-1	1.29	0.14	Pa $k\beta_3$		0.000866
108.490	0.1		-1	2.43	0.24	Pa $k\beta_1+5$		0.001631
111.590	0.1		-1	0.9	0.1	Pa $k\beta_2$		0.000604
112.460	0.1		-1	0.34	0.04	Pa k_0		0.000228
115.500	0.2			0.04	0.01			2.68E-05
116.910	0.05			0.39	0.04			0.000262
125.100	0.05	124.914		0.95	0.09		0.0006	0.000637
134.140	0.08	134.030		0.42	0.05		0.00025	0.000282
135.770	0.06	135.664		1.3	0.1		0.00084	0.000872
136.780	0.2			0.09	0.03			6.04E-05
145.150	0.3			0.12	0.03			8.05E-05
146.000	0.07			0.58	0.06			0.000389
163.160	0.06			2.6	0.03			0.001745
164.940	0.1			0.06	0.03			4.03E-05
169.580	0.1			0.03	0.01			2.01E-05
174.190	0.08			0.31	0.03			0.000208
183.470	0.07			0.57	0.06			0.000382
188.770	0.2			0.08	0.01			5.37E-05
218.000	0.07	217.94		0.67	0.07		0.00037	0.00045
236.170	0.07			0.18	0.02			0.000121
240.400	0.2			0.005	0.0005			3.36E-06
242.600	0.1			0.013	0.0006			8.72E-06
249.800	0.3			0.01	0.002			6.71E-06
250.500	0.3			0.011	0.002			7.38E-06
267.800	0.07			0.023	0.0006			1.54E-05
308.900	0.3			0.008	0.001			5.37E-06
311.000	0.1			0.054	0.005			3.62E-05
318.000	0.4			0.002	0.0002			1.34E-06
320.200	0.3			0.004	0.0003			2.35E-06

a. Normalized to 0.0671 for 84.17 keV transition.

b. Browne, E and Asaro F. (Phys Rev C V7n6 p2545) find the 84.17 keV transition branching ratio = 0.070 ± 0.003

²³⁵U and Daughters Gammas

keV	Branch ratio*	g/s/gm	Emitter1	Parent1	Emitter2	Parent2
11.400	0.03050	2.40E+03	Th-231	U-235		
13.000	0.22367	1.76E+04	U-235	U-235	Ac-227	U-235
13.700	0.49817	3.92E+04	Th-231	U-235		
14.500	0.00224	1.76E+02	U-235	U-235	Ac-227	U-235
15.000	0.00407	3.20E+02	Th-231	U-235		
16.100	0.15250	1.20E+04	U-235	U-235		
16.600	0.37617	2.96E+04	Th-231	U-235		
17.200	0.00224	1.76E+02	Th-231	U-235		
19.100	0.02643	2.08E+03	U-235	U-235		
19.800	0.07523	5.92E+03	Th-231	U-235		
25.600	0.14869	1.17E+04	Th-231	U-235		
42.000	0.00061	4.80E+01	U-235	U-235		
42.800	0.00059	4.64E+01	Th-231	U-235		
58.600	0.00488	3.84E+02	Th-231	U-235		
72.700	0.00112	8.80E+01	U-235	U-235		
72.800	0.00255	2.01E+02	Th-231	U-235		
74.800	0.00061	4.80E+01	U-235	U-235		
81.200	0.00915	7.20E+02	Th-231	U-235		
84.200	0.06710	5.28E+03	Th-231	U-235		
90.000	0.03419	2.69E+03	U-235	U-235		
90.000	0.00956	7.52E+02	Th-231	U-235		
92.300	0.00397	3.12E+02	Th-231	U-235		
93.400	0.05592	4.40E+03	U-235	U-235		
95.900	0.00641	5.04E+02	Th-231	U-235		
96.200	0.00087	6.88E+01	U-235	U-235		
99.300	0.00122	9.60E+01	Th-231	U-235		
102.300	0.00417	3.28E+02	Th-231	U-235		
105.400	0.02008	1.58E+03	U-235	U-235		
108.200	0.00231	1.82E+02	Th-231	U-235		
109.000	0.00671	5.28E+02	U-235	U-235		
109.200	0.01563	1.23E+03	U-235	U-235		
111.900	0.00077	6.08E+01	Th-231	U-235		
116.100	0.00071	5.60E+01	U-235	U-235		
124.900	0.00057	4.48E+01	Th-231	U-235		
135.700	0.00079	6.24E+01	Th-231	U-235		
140.800	0.00224	1.76E+02	U-235	U-235		
143.800	0.11133	8.76E+03	U-235	U-235		
150.900	0.00081	6.40E+01	U-235	U-235		
163.100	0.00158	1.24E+02	Th-231	U-235		
163.300	0.05160	4.06E+03	U-235	U-235		
182.600	0.00346	2.72E+02	U-235	U-235		
185.700	0.58077	4.57E+04	U-235	U-235		
194.900	0.00641	5.04E+02	U-235	U-235		
198.900	0.00427	3.36E+02	U-235	U-235		
202.100	0.01098	8.64E+02	U-235	U-235		
205.300	0.05096	4.01E+03	U-235	U-235		
221.400	0.00122	9.60E+01	U-235	U-235		
240.900	0.00055	4.32E+01	U-235	U-235		

* Normalized to .0671 at 84.214 keV

²³⁸U and Daughters Gammas

E(keV) *	Branch Ratio % *	uncert. % *	g/s/gm	Emitter1	Parent1
63.24	3.6000	3	4.73E+02	Th-234	U-238
131.31	0.0286	1.4	3.23E+00	Pa-234	U-238
152.76	0.0083	3.7	1.08E+00	Pa-234	U-238
203.12	0.0027	8	3.37E-01	Pa-234	U-238
226.85	0.0167	1.3	9.54E-01	Pa-234	U-238
249.21	0.0035	4.7	4.53E-01	Pa-234	U-238
258.26	0.0730	0.46	9.02E+00	Pa-234m	U-238
272.20	0.0018	9.1	1.62E-01	Pa-234	U-238
293.74	0.0049	3.1	6.31E-01	Pa-234	U-238
369.52	0.0044	3.5	4.69E-01	Pa-234	U-238
372.02	0.0023	6.9	2.10E-01	Pa-234	U-238
450.96	0.0030	5.2	3.36E-01	Pa-234m	U-238
453.58	0.0019	8.4	2.71E-01	Pa-234m	U-238
458.63	0.0020	8	2.43E-01	Pa-234	U-238
468.44	0.0023	6.8	2.63E-01	Pa-234m	U-238
475.75	0.0023	6.5	3.18E-01	Pa-234m	U-238
506.70	0.0035	5.5	2.59E-01	Pa-234	U-238
543.98	0.0036	4.7	4.14E-01	Pa-234m	U-238
569.30	0.0203	1.3	1.73E+00	Pa-234	U-238
654.37	0.0022	7.6	9.70E-02	Pa-234	U-238
666.42	0.0015	9.8	2.59E-01	Pa-234	U-238
669.64	0.0017	8.9	2.26E-01	Pa-234	U-238
691.08	0.0090	2.1	8.75E-01	Pa-234m	U-238
699.02	0.0059	2.6	7.44E-01	Pa-234	U-238
702.05	0.0071	2.4	8.59E-01	Pa-234m	U-238
705.90	0.0065	2.4	9.16E-01	Pa-234	U-238
733.38	0.0115	1.5	1.39E+00	Pa-234	U-238
737.88	0.0021	8.3	1.62E-01	Pa-234	U-238
739.95	0.0118	2.1	1.13E+00	Pa-234m	U-238
742.77	0.0946	0.7	9.38E+00	Pa-234m	U-238
755.00	0.0021	8.1	1.62E-01	Pa-234	U-238
766.37	0.3220	0.65	3.29E+01	Pa-234m	U-238
781.73	0.0078	2.2	8.43E-01	Pa-234m	U-238
786.25	0.0554	0.93	5.67E+00	Pa-234m	U-238
796.42	0.0054	4.3	6.14E-01	Pa-234	U-238
805.74	0.0088	1.8	1.04E+00	Pa-234	U-238
808.20	0.0026	10	3.60E-01	Pa-234m	U-238
819.21	0.0037	3.9	4.20E-01	Pa-234	U-238
824.94	0.0068	2.6	6.47E-01	Pa-234	U-238
831.39	0.0078	1.9	8.89E-01	Pa-234	U-238
851.57	0.0070	2	6.88E-01	Pa-234m	U-238
875.94	0.0042	3	6.47E-01	Pa-234	U-238
880.45	0.0212	0.9	1.46E+00	Pa-234	U-238
883.22	0.0211	0.9	2.13E+00	Pa-234	U-238
887.28	0.0071	1.8	8.27E-01	Pa-234m	U-238
898.52	0.0059	2.2	6.63E-01	Pa-234	U-238
921.70	0.0127	1.1	1.32E+00	Pa-234m	U-238
924.98	0.0142	1.2	1.78E+00	Pa-234	U-238
926.61	0.0192	1.1	1.60E+00	Pa-234	U-238

941.94	0.0025	4.2	3.45E-01	Pa-234m	U-238
945.90	0.0335	0.86	2.44E+00	Pa-234	U-238
947.43	0.0031	4.4	1.29E+00	Pa-234	U-238
980.42	0.0045	3	4.85E-01	Pa-234	U-238
984.09	0.0030	4.2	3.07E-01	Pa-234	U-238
994.93	0.0057	2.1	4.61E-01	Pa-234m	U-238
1000.99	0.839	0.56	1.03E+02	Pa-234m	U-238
1041.70	0.0012	8	1.54E-01	Pa-234m	U-238
1061.89	0.0023	5.2	2.23E-01	Pa-234m	U-238
1084.25	0.0012	7.5	2.22E-01	Pa-234	U-238
1124.93	0.0042	3.1	3.34E-01	Pa-234m	U-238
1193.69	0.0135	0.96	1.43E+00	Pa-234m	U-238
1220.37	0.0009	10.2	1.11E-01	Pa-234m	U-238
1237.24	0.0053	1.8	5.73E-02	Pa-234m	U-238
1292.66	0.0009	11.2	9.70E-02	Pa-234	U-238
1352.80	0.0019	4.1	2.75E-01	Pa-234	U-238
1393.57	0.0039	2.5	4.85E-01	Pa-234	U-238
1413.88	0.0023	4.2	2.39E-01	Pa-234m	U-238
1434.13	0.0097	1.3	9.07E-01	Pa-234m	U-238
1452.63	0.0012	7.3	1.62E-01	Pa-234	U-238
1510.20	0.0129	1.2	1.45E+00	Pa-234m	U-238
1527.27	0.0024	3.7	2.47E-01	Pa-234m	U-238
1548.12	0.0014	5.9	2.07E-01	Pa-234m	U-238
1553.74	0.0081	1.6	1.00E+00	Pa-234m	U-238
1570.67	0.0011	7.8	1.21E-01	Pa-234m	U-238
1591.65	0.0019	5.2	4.30E-01	Pa-234m	U-238
1593.88	0.0027	3.6	9.70E-02	Pa-234	U-238
1668.44	0.0012	6.2	1.94E-01	Pa-234	U-238
1694.08	0.0013	5.9	1.94E-01	Pa-234	U-238
1737.73	0.0212	1.1	2.26E+00	Pa-234m	U-238
1759.81	0.0014	4.4	2.55E-01	Pa-234m	U-238
1765.44	0.0087	1.4	9.71E-01	Pa-234m	U-238
1809.04	0.0037	2.1	4.77E-01	Pa-234m	U-238
1819.69	0.0009	7.3	1.32E-01	Pa-234m	U-238
1831.36	0.0172	1.3	1.78E+00	Pa-234m	U-238
1863.09	0.0012	4.3	1.35E-01	Pa-234m	U-238
1867.68	0.0092	1.4	8.43E-01	Pa-234m	U-238
1874.85	0.0082	1.5	8.75E-01	Pa-234m	U-238
1877.21	0.00165	3.4	3.07E-02	Pa-234	U-238
1893.50	0.00219	2.9	2.39E-01	Pa-234m	U-238
1911.17	0.0063	1.6	5.89E-01	Pa-234m	U-238
1925.42	0.0005	10.1	8.08E-02	Pa-234	U-238
1937.01	0.0029	2.3	3.34E-01	Pa-234m	U-238

* Scott, H. L. and Marlow, K. W., NIM A286(1990) 549-55

²³⁸U and Daughters—²³⁴Pa and ²³⁴Th

E(keV)	g/s/gm	Emitter	Parent1	Emitter2	Parent2
62.9	2.36	Th-234	U-238		
73.9	1.36	Pa-234m	U-238		
74.0	5.30	Th-234	U-238		
83.3	8.71	Th-234	U-238		

92.3	338.0	Th-234	U-238		
92.8	335.0	Th-234	U-238		
94.7	21.6	Pa-234m	U-238	Pa-234	U-238
95.9	1.62	Th-234	U-238		
110.5	2.98	U-238	U-238		
114.9	4.32	Pa-234m	U-238	Pa-234	U-238
131.3	3.23	Pa-234	U-238		
152.7	1.08	Pa-234	U-238		
184.8	1.49	Th-234	U-238		
258.2	9.02	Pa-234m	U-238		

²³⁸U Daughter— Protactinium: Gammas and Branching Ratios

^{234m}Pa IC-decay*			
IC-decay prob%			
E(keV)	X1000	uncert. ±	
257.90	57.000	0.230	Pa234m
691.00	5.500	0.500	Pa234m
701.60	5.400	0.500	Pa234m
740.10	7.100	0.700	Pa234m
743.00	56.600	0.230	Pa234m
766.60	207.800	0.800	Pa234m
782.30	5.300	0.500	Pa234m
786.40	34.200	0.130	Pa234m
887.50	5.200	0.500	Pa234m
922.30	8.300	0.800	Pa234m
946.30	7.000	0.700	Pa234m
1001.20	590.000		Pa234m
1738.20	14.200	0.600	Pa234m
1831.50	11.200	0.400	Pa234m
1868.20	5.300	0.500	Pa234m
1911.80	3.700	0.400	Pa234m
1937.70	2.100	0.200	Pa234m

NORMALIZED 1001. = 590 (0.59%x1000)

* Ardisson G. and Marsol c., Nuovo Chimie 11v28A 155(75)

²³⁴ Pa IC-decay **		
IC-decay prob%		
E(keV)		
63.0	4.10	Pa 234
131.3	20.00	Pa 234
153.0	6.60	Pa 234
226.9	11.50	Pa 234
569.3	13.80	Pa 234
699.1	4.75	Pa 234
805.5	3.10	Pa 234
824.7	3.70	Pa 234
831.1	5.10	Pa 234
926.7	16.80	Pa 234
945.8	18.40	Pa 234
980.5	3.90	Pa 234
1394.1	2.40	Pa 234

** Radiochem. Radioanal Lett. 221 357(75)

Appendix D

Uranium Standard Measurements

LANL Uranium Standards: mass spec. results

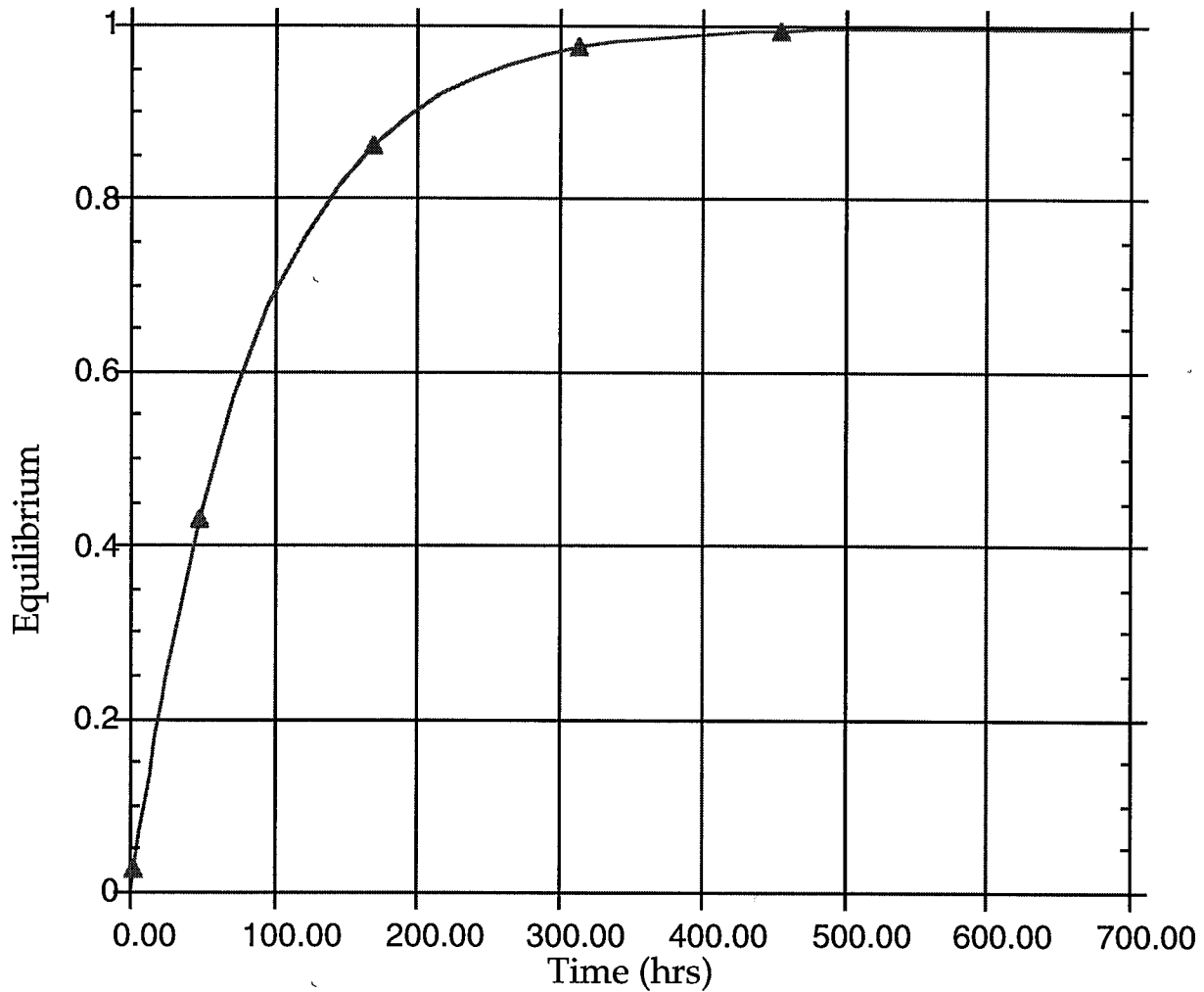
<u>U238</u>	<u>U235</u>	<u>U-234</u>	<u>U-236</u>	<u>u234/5</u>	<u>U236/5</u>	<u>U234/6</u>
%	%	%	%	ratio	ratio	ratio
99.275	0.7194	0.0050	0.0010	0.0070	0.0014	5.0000
99.272	0.7225	0.0050	0.0010	0.0069	0.0014	5.0000
99.267	0.7257	0.0050	0.0020	0.0069	0.0028	2.5000
99.267	0.7257	0.0050	0.0020	0.0069	0.0028	2.5000
98.024	1.9608	0.0160	0.0090	0.0082	0.0046	1.7778
98.022	1.9632	0.0160	0.0090	0.0082	0.0046	1.7778
96.895	3.0637	0.0250	0.0160	0.0082	0.0052	1.5625
96.894	3.0637	0.0250	0.0170	0.0082	0.0056	1.4706
89.664	10.1923	0.0530	0.0910	0.0052	0.0089	0.5824
89.662	10.1961	0.0520	0.0910	0.0051	0.0089	0.5714
89.627	10.2273	0.0540	0.0920	0.0053	0.0090	0.5870
87.883	11.9281	0.0730	0.1160	0.0061	0.0097	0.6293
86.730	13.0841	0.0840	0.1020	0.0064	0.0078	0.8235
82.278	17.4129	0.1400	0.1690	0.0080	0.0097	0.8284
72.499	26.9932	0.2370	0.2710	0.0088	0.0100	0.8745
62.677	37.8417	0.2630	0.2180	0.0070	0.0058	1.2064
46.749	52.4462	0.5360	0.2690	0.0102	0.0051	1.9926
32.833	66.3254	0.5830	0.2590	0.0088	0.0039	2.2510
7.342	91.4086	0.9150	0.3340	0.0100	0.0037	2.7395

<u>Standard</u>	<u>U234</u>	<u>U235</u>	<u>U236</u>	<u>U238</u>
ID#	(at. %)	(at. %)	(at. %)	(at. %)
u0_017	0.0010	0.0170	0.0000	99.9710
CRM U002	0.0002	0.0176	0.0000	99.9823
u0_483	0.0050	0.4830	0.0008	99.4970
NBS U-005a	0.0034	0.5064	0.0012	99.4890
AI-409	0.0050	0.7190	0.0010	99.3000
AI-408-2	0.0050	0.7226	0.0010	99.3000
AI-1127-2	0.0050	0.7256	0.0020	99.3000
AI-1127-1	0.0050	0.7258	0.0020	99.3000
u0_991	0.0080	0.9910	0.0019	98.9930
NBS U-010	0.0054	1.0037	0.0068	98.9840
AI-1125-1	0.0160	1.9610	0.0090	98.0000
AI-1125-2	0.0160	1.9620	0.0090	98.0000
u2_013	0.0160	2.0130	0.0044	98.0070
u3_009	0.0310	3.0090	0.0070	96.9830
CRM U030-A	0.0278	3.0404	0.0006	96.9312
AI-1126-1	0.0250	3.0630	0.0160	96.9000
AI-1126-2	0.0250	3.0650	0.0170	96.9000
u4_949	0.0410	4.9490	0.0128	94.9630
NBS U-050	0.0279	5.0100	0.0480	94.9150
U10_075	0.0770	10.0750	0.0299	89.8880
CRM U100	0.0676	10.1900	0.0379	89.7040
AI-324-2	0.0530	10.2000	0.0910	89.7000
AI-324-1	0.0520	10.2020	0.0910	89.7000

AI-323-1	0.0540	10.2200	0.0920	89.6000
UIISO-12	0.0730	11.9300	0.1160	87.8000
UIISO-13	0.0840	13.0890	0.1020	86.7000
UIISO-17	0.1400	17.4200	0.1690	82.2000
UIISO-27	0.2370	27.0000	0.2710	72.7000
UIISO-38	0.2630	37.8480	0.2180	61.6000
u49_38	0.6720	49.3800	0.2009	50.0740
UIISO-52	0.5360	52.4260	0.2690	46.2000
UIISO-66	0.5830	66.3170	0.2590	32.8000
U75_13	0.5840	75.1300	0.3320	23.5120
CRM U750	0.5923	75.3570	0.2499	23.8010
UIISO-91	0.9150	91.4190	0.3340	7.3300
u93_076	1.4520	93.0760	0.4291	5.4720

Appendix E

The approach to daughter product equilibrium.



Plot of the equilibrium buildup of ^{231}Th ($T_{1/2} = 25.52$ hours) from the decay of ^{235}U ($T_{1/2} = 7.037 \times 10^8$ years). Full (99.98%) equilibrium is seen in 30 days. At equilibrium, $N_U \lambda_U = N_{\text{Th}} \lambda_{\text{Th}}$ where N_U is the number of uranium atoms and N_{Th} is the number of thorium atoms; λ_U is the decay constant of ^{235}U and λ_{Th} is the decay constant of ^{231}Th . At equilibrium $N_{\text{Th}}/N_U = 4.137 \times 10^{-12}$.

$$\frac{N_d}{N_p} = \frac{\lambda_p \cdot N_p}{\lambda_d - \lambda_p} \cdot (e^{-\lambda_p t} - e^{-\lambda_d t}), \quad N_d \approx \frac{\lambda_p}{\lambda_d - \lambda_p} \cdot N_p e^{-\lambda_p t}$$

At equilibrium

$$N_d \lambda_d = N_p \lambda_p$$

a. Reference: Glasstone, S., Sesonske, A., *Nuclear Reactor Engineering*, 3rd Ed. p42(1991)

Appendix F

Uranium General Properties

Atomic number: 92
 Standard atomic weight: 238.0289
 Specific gravity: 18.95
 Symbol: U

General information

Standard state: solid
 Color: metallic gray
 Discoverer: Martin Klaproth
 Date discovered: 1789
 Discovered at: Germany
 Meaning of name: Planet Uranus

Radii (pm)

Atomic: 138.5
 van der Waals:
 Covalent: 142
 Metallic: 143

Isotopes

Uranium has sixteen isotopes, all of which are radioactive. Naturally occurring uranium nominally contains

99.28305% by wt ^{238}U
 0.7200% ^{235}U
 0.0054% ^{234}U

X-ray properties

Data for U ; Z = 92
 atomic weight = 238.070; density = 19.0500
 K-edge at: 115.603 keV
 L-edges at: 21.7560, 20.9470, 17.1670 keV
 M-edge at: 5.54900 keV
 K- α 1, K- β at: 98.4280 111.289 keV
 L- α , L- β 1 at: 13.6130 17.2180 keV
 K, L1, L2, L3 cross section jumps:
 Inf NaN 1.41000 2.29200
 Fluorescence yield for K, L1, L2, L3: 0.9720, 0.1760, 0.4670, 0.4890
 Calculations are based on data compiled By W. H. McMaster et. al.
 Fluorescence yield data by M. O. Krause, J. Phys. Chem. Ref. Data. 8, 307(1979).

Electron Configuration: -21-9-2
 heat Vapor: 110
 heat Fusion: 2.7

Electrical Conductivity: 0.034
 Thermal Conductivity: 0.064

Specific Heat Capacity: 0.028

Temperatures °K

melting: 1405.5
 boiling: 4028
 Debye: 200
 superconduction: 0.68

Uranium Isotope Properties

Z	Abund	mass (AMU)	half-life
232	0		69.8 yr.
233	0		1.592E5 yr.
234	0.0054	234.040946	2.457E5 yr.
235	0.720	235.043924	7.037E8 yr.
236	0	236.045562	2.342E7 yr.
237	0	237.04827	6.75 DA
238	99.275	238.050784	4.468E9 yr.
239	0		23.47 min.
240	0		14.1 hr

92-uranium-235

(U235 is also called actinouranium)
 U235(n,f) is standard for neutrons below 20 MeV.

General Properties

Atomic Mass:
 235.0439222 \pm 0.0000021 AMU
 Excess Mass: 40913.215 \pm 2.002 keV
 Binding Energy:
 1783871.153 \pm 2.028 keV
 Beta IC-decay energy: β -
 -123.716 \pm 0.869 keV
 Atomic Percent Abundance: 0.720%
 Spin: 7/2-
 Half life: 703.8E+6 Y (0.0710 %)
 Mode of IC-decay: Alpha to Th-231
 IC-decay energy: 4.6787 MeV
 Mode of IC-decay: Spontaneous fission

Branch ratio: $7.0\text{E-}9\%$

Fission spectrum avg. = 89.07 mb
g-factor = 0.9898

Meta state at 0.0768 keV

Spin: $1/2^+$

Half life: 25 M

Mode of IC-decay: Internal Transition

Total Cross Section

Cross Section at 0.0253 eV = 698.2 b

Maxwell avg. at 0.0253 eV = 608.4 b

Cross Section at 14 MeV = 5.865 b

Fission spectrum avg. = 7.705 b

g-factor = 0.9833

Elastic scattering Cross Section

Cross Section at 0.0253 eV = 15.04 b

Maxwell avg. at 0.0253 eV = 14.95 b

Cross Section at 14 MeV = 2.871 b

Fission spectrum avg. = 4.566 b

g-factor = 1.1215

Total inelastic Cross Section

Cross Section at 14 MeV = 350.3 mb

Fission spectrum avg. = 1.804 b

(n,2n) Cross Section

Cross Section at 14 MeV = 542.9 mb

Fission spectrum avg. = 11.56 mb

(n,3n) Cross Section

Cross Section at 14 MeV = 41.79 mb

Fission spectrum avg. = 7.074 μb

Total fission Cross Section

Cross Section at 0.0253 eV = 584.4 b

Maxwell avg. at 0.0253 eV = 506.8 b

Resonance integral = 278.1 b

Cross Section at 14 MeV = 2.056 b

Fission spectrum avg. = 1.235 b

g-factor = 0.9786

(n,4n) Cross Section

Fission spectrum avg. = 0.008408 μb

Radiative capture Cross Section

Cross Section at 0.0253 eV = 98.81 b

Maxwell avg. at 0.0253 eV = 86.67 b

Resonance integral = 133.0 b

Cross Section at 14 MeV = 0.1607 $\mu\text{ barn}$

92-uranium-238

(²³⁸U is also called uranium I)

General Properties

Atomic Mass:

238.0507835 ± 0.0000022 AMU

Excess Mass:

47304.541 ± 2.024 keV

Binding Energy:

1801693.796 ± 2.050 keV

Beta IC-decay energy:

β⁻ -145.345 ± 1.351 keV

Atomic Percent Abundance:

99.2745% 15

Spin: 0+

Half life: 4.468E+9 Y (0.0671 %)

Mode of IC-decay: Alpha to Th-234

IC-decay energy: 4.270 MeV

Mode of IC-decay: Spontaneous fission

Branch ratio: 5.45x10⁻⁵ %

Mode of IC-decay: Beta

Branch ratio: 2.2x10⁻¹⁰ %

Total Cross Section

Cross Section at 0.0253 eV = 12.09 b

Maxwell avg. at 0.0253 eV = 11.77 b

Cross Section at 14 MeV = 5.805 b

Fission spectrum avg. = 7.786 b

g-factor = 1.0994

Elastic scattering Cross Section

Cross Section at 0.0253 eV = 9.360 b

Maxwell avg. at 0.0253 eV = 9.356 b

Cross Section at 14 MeV = 2.704 b

Fission spectrum avg. = 4.804 b

g-factor = 1.1279

Total inelastic Cross Section

Cross Section at 14 MeV = 698.3 mb

Fission spectrum avg. = 2.595 b

(n,2n) Cross Section

Cross Section at 14 MeV = 910.0 mb

Fission spectrum avg. = 12.07 mb

(n,3n) Cross Section

Cross Section at 14 MeV = 350.0 mb

Fission spectrum avg. = 61.90 μ barn

Total fission Cross Section

Cross Section at 0.0253 eV = 11.77 μ barn

Maxwell avg. at 0.0253 eV = 10.45 μ barn

Resonance integral = 2.020 b

Cross Section at 14 MeV = 1.136 b

Fission spectrum avg. = 308.4 mb

g-factor = 1.0015

Radiative capture Cross Section

Cross Section at 0.0253 eV = 2.717 b

Maxwell avg. at 0.0253 eV = 2.414 b

Resonance integral = 278.1 b

Cross Section at 14 MeV = 1.943 mb

Fission spectrum avg. = 66.40 mb

g-factor = 1.0024

Isotope[1]		Jpi[2]	delta MeV)[3]	T1/2 or Abundance[4]	Decay Mode[5]	
Z	El A					
92	U 218	0+	21.88s	1.5 ms +73-7	A	
	219			23.2s	42 us +34-13	A
	220	0+		23.0s		
	221			24.5s		
	222	0+		24.3s	1.0 us +10-4	A
	223			25.82	18 us +10-5	A
	224	0+	25.70	1.0 ms 4	A	
	225			27.37	95 ms 15	A
	226	0+	27.32	0.20 s 5	A	
	227	(3/2+)	29.00		1.1 m 1	A
	228	0+	29.22		9.1 m 2	A > 95%,EC < 5%
	229	(3/2+)	31.204	58 m 3	EC ~ 80%,A ~ 20%	
	230	0+	31.603	20.8 d		A
	231	(5/2-)	33.78		4.2 d 1	EC
	231	(3/2+,5/2+	33.78		4.2 d 1	A ~ 4E-3%
	232	0+	34.601	68.9 y 4	A,Ne 9E-10%	
	233	5/2+	36.912	1.592E+5 y 2	A,SF <6.0E-9%	
					Ne 7E-11%	
	234	0+	38.140	2.455E+5 y 6	A,SF 1.7E-9%,Mg 1E-11%	
				0.0055% 5	Ne 9E-12%	
	235	7/2-	40.913	703.8E+6 y 5	A,SF 7.0E-9%,Ne 8E-10%	
				0.720% 1		
	235M	1/2+	40.913	~ 25 m	IT	
	236	0+	42.440	2.342E7 y 3	A,SF 9.6E-8%	
	236M			42.440	121 NS 2	SF 0.013%
	237	1/2+	45.385	6.75 d 1	B-	
	238	0+	47.305	4.468E+9 y 3	A,SF 0.5E-4%	
				99.2745% 15		
	238M	0+	47.305	267 NS 3	SF 0.015%	
	239	5/2+	50.570	23.45 m 2	B-	
	240	0+	52.708	14.1 h 1	B-,A	
	242	0+	58.830C	16.8 m 5	B-	

Explanation of Table

• Column 1, Isotope (Z, El, A)

Nuclides are listed in order of increasing atomic number (Z), and are subordered by increasing mass number (A). All isotopic species are included as well as all isomers with half-life ≥ 0.1 s, and some other isomers which decay by SF or alpha emissions. A nuclide is included even if only its mass estimate or its production cross section is available. For the latter nuclides T_{1/2} limit is given[8] .

Isomeric states are denoted by the symbol "m" after the mass number and are given in the order of increasing excitation energy.

The ²³⁵U thermal fission products, with fractional cumulative yields $\geq 10^{-6}$, are italicized in the table. The information on fission products is taken from the ENDF/B-VI fission products file[11].

The names for elements Z=104-109 are those adopted by the American Chemical Society Nomenclature Committee. The symbols Rf (Rutherfordium) and Ha (Hahnium) have, not been accepted internationally due to conflicting claims about the discovery of these elements.

• Column 2, J_{pi}

Spin and parity assignments, without and with parentheses, are based upon strong and weak arguments, respectively. See the introductory pages of any January issue of Nuclear Data Sheets[2] for description of strong and weak arguments for J_{pi} assignments.

• Column 3, Mass Excess, Delta

Mass excesses, M-A, are given in MeV with Delta(¹²C)=0, by definition. For isomers the values are obtained by adding the excitation energy to the Delta(g.s.) values. Wherever the excitation energy is not known, the mass excess for the next lower isomer (or g.s.) is given. The values are given to the accuracy determined by uncertainty in Delta(g.s.) (maximum of three figures after the decimal). The uncertainty is ≥ 9 in the last significant figure. An appended "s" denotes that the value is obtained from systematics.

• Column 4, T_{1/2} or Abundance

The half-life and the abundance (in bold face) are shown followed by their units ("%" symbol in the case of abundance) which are followed by the uncertainty, in italics, in the last significant figure. For example, 8.1 s \pm 1.0 s. For some very short-lived nuclei, level widths rather than half-lives are given. There also, the width is followed by units (e.g., eV, keV, or MeV) which are followed by the uncertainty in *N* italics, if known.

• Column 5, Decay Mode

Decay modes are given in decreasing strength from left to right, followed by the percentage branching, if known ("w" indicates a weak branch). The percentage branching is omitted where there is no competing mode of decay or no other mode has been observed.

The various modes of decay are given below:

B-	beta- decay
E	epsilon (electron capture), or epsilon+beta+, or beta+ decay
IT	isomeric transition (through gamma or conversion-electron decay)
n, p, A, ...	neutron, proton, alpha, decay
SF	spontaneous fission
2B-, 3A, ...	double beta- decay (beta-beta-), decay through emission of 3 alpha's,
B-N, B-P,	delayed n, p, alpha, ... B-A, ..emission following beta decay
EP, EA, ESF, ...	delayed p, alpha, SF, decay following epsilon or beta+ decay

The appendices conform to the Fundamental Physical Constants[13]. For properties of the elementary particles and for the astrophysical constants see the Review of Particle Properties, Physical Review D50, 1173 (1994) and its subsequent biennial updates. See also the World Wide Web at URL: <http://pdg.lbl.gov/>

References:

1. **Evaluated Nuclear Structure Data File**- a computer file of evaluated experimental nuclear structure data maintained by the National Nuclear Data Center, Brookhaven National Laboratory (file as of June 1995).
2. **Nuclear Data Sheets** - Academic Press, San Diego. Evaluations published by mass number for $A = 45$ to 266. See page ii of any issue for the index to A-chains.
3. **Nuclear Physics** - North Holland Publishing Co., Amsterdam - Evaluations by F. Ajzenberg-Selove and by D. R. Tilley, H. R. Weller, C. M. Cheves, and R. M. Chasteler for $A = 3$ to 20.
4. **Energy Levels of $A = 21-44$ Nuclei (VII)**, P. M. Endt, Nuclear Physics A521, 1 (1990).
5. **Nuclear Science Reference File**- a bibliographic computer file of nuclear science references continually updated and maintained by the National Nuclear Data Center, Brookhaven National Laboratory. Recent literature scanned by S. Ramavataram.
6. **Table of Isotopes** (1996), 8th edition, Editors: R. B. Firestone, et al., John Wiley, New York.
7. Spontaneous Fission, D. C. Hoffman, T. M. Hamilton, and M. R. Lane, Rept. LBL-33001 (1992).
8. **NUBASE: A Database of Nuclear and Decay Properties**, G. Audi, O. Bersillon, J. Blachot, and A. H. Wapstra, Intl. Symposium on Radionuclide Metrology and its Applications (1995).
9. **The 1993 Atomic Mass Evaluation**, G. Audi and A. H. Wapstra, computerized list of recommended values based on authors' publication Nuclear Physics A565, 1 (1993)
10. **Table of the Isotopes**, N. E. Holden, Rept BNL-61460 (1995) and private communication.
11. **Evaluation and Compilation of Fission Product Yields 1993**, T. R. England and B. F. Rider; Rept. LA-UR-94-3106 (1994). ENDF/B-VI evaluation; MAT #9228, Revision 1.
12. **Table of Isotopes** (1978), 7th edition, Editors: C. M. Lederer, V. S. Shirley, Authors: E. Browne, J. M. Dairiki, R. E. Doebler, A. A. Shihab-Eldin, J. Jardine, J. K. Tuli, and A. B. Buyrn, John Wiley, New York.
13. **The Fundamental Physical Constants**, E. R. Taylor and B. N. Taylor, Physics Today BG9 (August, 1995).

Technical Information Department • Lawrence Livermore National Laboratory
University of California • Livermore, California 94551

