FIELD MEASUREMENT OF AM ^{241} AND TOTAL URANIUM AT A MIXED OXIDE FUEL FACILITY WITH VARIABLE URANIUM ENRICHMENTS RANGING FROM 0.3% TO 97% U ^{235}

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ABSTRACT

The uranium and transuranic content of site soils and building rubble can be accurately measured using a NaI(Tl) well counter, without significant soil preparation. Accurate measurements of total uranium in uranium-transuranic mixtures can be made, despite a wide range (0.3% to 97%) of uranium enrichment, sample mass, and activity concentrations. The appropriate uranium scaling factors needed to include the undetected uranium isotopes, particularly U ^{234} can be readily determined on a sample by sample basis as a part of the field analysis, by comparing the relative response of the U ^{235} 186 keV peak versus the K shell X rays of U ^{238} , U ^{235}, and their immediate ingrowth daughters. The ratio of the two results is a sensitive and accurate predictor of the uranium enrichment and scaling factors. The case study will illustrate how NaI(Tl) gamma spectrometry was used to provide rapid turnaround uranium and transuranic activity levels for soil and building rubble with sample by sample determination of the appropriate scaling factor to include the U^{234} and Uranium^{238} content.

INTRODUCTION

BWXT Services has developed, a practical and cost effective technique to determine soil and rubble TRU and uranium content to support remediation and Final Survey for Free Release for Unrestricted Use surveys 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. Transuranics (predominantly Am ^{241}, Pu ^{238}, Pu ^{239}, Pu ^{240}, and Pu ^{241}) were present in quantity throughout the majority of the site. Small quantities of fission and activation products (Cs^{137} and Co^{60}) were also present throughout the majority of the site. Uranium isotopes were common throughout the site and dominant in sections of the facility. 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. The site consisted primarily of Depleted Uranium and High Enriched Uranium use areas. The majority of the site had been exposed to all three uranium enrichments. An enrichment over check by means of comparing the Uranium series K shell X rays to the U^{235} activity level based on the U ^{235} 186 keV peak was done routinely to ensure the application of the correct total uranium to U ^{235} scaling factor. The primary goal of the enrichment over check was to ensure that a conservative scaling factor was applied to the U ^{235} measurement. Applicable total uranium to U ^{235} scaling factors (22, 24.4, 39.2 and 58.8) at the site varied considerably.

The equipment used consisted of a Canberra MCA coupled to a Bicron Model 2M2/2 2" by 2" NaI(Tl) crystal. The system was operated as a set of up to 5 counting windows (Am^{241}, Uranium related K shell X rays (89-120 keV), U ^{235} (186 keV peak), Co ^{60}, and Cs^{137}). The equipment was set up and operated in small site trailer. Shielding consisted of 0.75 inches of steel and fifty 40-pound bags of white play sand. Sample preparation was limited to filling a Marinelli beaker with sample, and pouring off standing liquid. Shielding analysis (Microshield V 5.03) had demonstrated that the wet soil fluence rate for this configuration was within 3% of the dry soil fluence rate. Data was collected as integral and peak counts in a Region of Interest.

Determination of Regions of Interest (ROI)

The X ray and gamma emissions of interest were determined and Regions of Interest (ROI)s set for them. ROIs were set up for Am ^{241}, Uranium related K shell X rays (89-120 keV), U ^{235} (186 keV peak), Co ^{60}, and Cs ^{137}.

Setup and Calibration of the NaI(Tl) Gamma Spectrometry System

Calibration isotopes and method were selected and applied. Two U ^{235} windows were established in order to allow determination of Uranium enrichment.

Sample Requirements

Sample requirements were minimal as the sample measurement technique was intended to immediately support ongoing remediation guide and conservatively estimate the activity present in order to minimize laboratory grade sample analysis.

Data Analysis

Analysis of the counting results and uranium enrichment over check. If present, net peaks were identified and quantified. An initial estimate of the uranium enrichment was performed**.**

METHODOLOGY

Determination of Regions of Interest

Measurements were of Am ^{241}, U ^{235}, Cs ^{137}, Co ^{60}, and Uranium K Shell X-rays. Regions of interest or counting windows were set about the Am ^{241} 59.5 keV, U ^{235} 186 keV, Cs ^{137} 662 keV, Co ^{60}, and Uranium K Shell (89-120 keV) X ray and gamma peaks. NIST traceable Marinelli soil samples containing known activities of 97.6 % enriched Uranium and Am ^{241} were used to set the and U ^{235} ROIs. A resin Marinelli gamma spectroscopy calibration source was used to set the Co ^{60} and Cs ^{137} windows. The regions of interest were set by eye and verified using figure of merit calculations. The Canberra 3502 allows the long-term setting of ROIs. The unit determines net peak counts and gross counts. Net peak data is determined by determining the count above a line drawn between the beginning and end channels of the ROI.

The uranium isotopes and their immediate daughters emit significant quantities of X-rays and limited gammas, in the energy range from 89-120 keV (see Tables I and II). Table I, ***"Isotopic Abundance Vs U ^{235} Enrichments,"*** shows the activity distribution of the uranium isotopes vs. mass enrichment and the fractional abundance of 89– 120 keV emissions. Table II "***K Shell X Ray (89-120 keV) Fractional Abundance vs Isotope and U ^{235} Enrichment"*** shows the total X-rays emitted in the 89 –120 keV energy range and the marked decrease of the photon abundance in this range as mass enrichment increases. The emission rates range from 5.9 % for Depleted Uranium (0.3% U ^{235}) to 0.53 for 90 % enriched Uranium (see Table II). The emission rates are drawn from ENDF/B-VI decay data. The steep decline in X ray production in the 89 to 120 keV range as uranium enrichment increases yields comparative results that are diagnostic of the uranium enrichment.

Setup and Calibration of the NAI(Tl) Gamma Spectrometry System

A Certified Health Physics Engineer calibrated the instrument with the assistance of the Director of the Analysis Laboratory. The unit was located in a small site trailer to avoid rapid temperature changes. The system consisted of a Canberra 3502 Multi Channel Analyzer, and a 2" by 2" NaI(Tl) detector. The detector was inside 0.75" of steel shielding and 15 inches of white sand. A standard count time of 1000 seconds was selected after an analysis of background and empirical testing. 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. The detector sensitive range was 15 keV to 3 meV.

The system was calibrated to both peak and integral counts for each isotope. Each calibration was to activity per gram, as the compliance standards were in terms of activity per gram. Invariant and mass corrected soil background were subtracted from the raw data. Mass corrected background was determined by counting a soil sample known to be similar to the site background soil activity levels.

Cs ^{137} and Co ^{60}Calibration

Calibration of the counter for Cs ^{137} and Co ^{60} was achieved by placing a NIST traceable Marinelli resin source in the counter. The system efficiency assumed to be constant across a wide range of activity levels and for sample masses of 500-1200 grams.

Am ^{241}Calibration

Three NIST traceable Am ^{241} Marinelli soil samples were counted in the system. The source Am ^{241} activities ranged from 6 to 45 pCi per gram. Linear regression was applied to the resulting data to yield gross (integral) and net peak efficiency equations, due to concerns about possible variation in response with activity concentration. The correlation between the net count rate per gram and the activity per gram was good. Coefficients of Determination (r^2) of 0.99 or better were achieved for the integral and peak counting modes. The resulting efficiency equations were assumed to be valid for sample masses of 500 to 1200 grams.

U ^{235} Calibrations

Four NIST traceable U ^{235} Marinelli soil samples were counted in the system. The U ^{235} source was 97.6% enriched uranium. The source U ^{235} activities ranged from 0.80 to 4.95 pCi per gram. Total uranium activity levels were 6 to 196 pCi/gram. Linear regression was applied to the resulting data to yield gross (integral) and net peak efficiency equations, due to concerns about possible variation in response with activity concentration. The correlation between the net count rate per gram and the activity per gram was good. Coefficients of Determination (r^2) of 0.98 or better were achieved for the U ^{235} 186 keV peak and integral equations. Coefficients of Determination (r^2) of 0.96 or better were normally achieved for gross counts in the U ^{235} K shell X-ray 89-120 keV ROI. Peak fit results were poorer, with typical Coefficients of Determination of 0.90. The resulting efficiency equations were assumed to be valid for sample masses of 500 to 1200 grams. The X ray window was calibrated to 97.6% enriched uranium in order to allow a comparison between the "U ^{235}" results of the two ROIs. Due to the increase in photons in the energy range of interest (89-120 keV) as the degree of U ^{235} enrichment decreases and the U ^{238} activity level increases, the response of the U ^{235} X ray ROI increases above that of the U ^{235} 186 keV ROI. The ratio between the two windows is diagnostic of the U ^{235} enrichment.

Figure 1 " ***Uranium K Shell/U ^{235} Ratios vs. U ^{235} Enrichment*** " and Table II "***K Shell X Ray (89-120 keV) Fractional Abundance vs. Isotope and U ^{235} Enrichment***" demonstrates the relationship between the U ^{235} enrichment and the relative response of the two U ^{235} ROIs.

Sample Requirements

Sample requirements were minimal as the sample measurements were intended to provide immediate support to ongoing remediation and conservatively estimate the activity present in order to minimize laboratory grade sample analysis. The samples were required to be in Marinelli containers. Sample mass was required to be within the range 500 to 1200 grams per sample and standing water was not allowed to be in the sample. No drying or other processing was conducted.

Data Analysis

The data analysis consisted of the following primary steps;

- Spectrum Inspection. The technicians operating the system were trained review the spectra and notify supervision of unusual peaks. Daily background and source checks for Am ^{241}, U ^{235}, Co ^{60}, and Cs ^{137} were performed to provide assurance of the stability of the system, and the utility of the ROIs.

- The background and sample data was analyzed in Excel to determine the estimated activity per gram for each isotope in both gross count and peak count mode. If positive results were obtained, the higher of the two results was assigned as the analysis result. If LLD results were obtained, one half of the LLD value was used as the analysis result. NaI(Tl) on site measurement of soil samples for Am ^{241}, U ^{235}, Co^{60}, and Cs ^{137} were sensitive to well below compliance levels. ". Typical LLDs for U ^{235}, Co ^{60}, and Cs ^{137} were several tenths of a pCi/g. Am ^{241} typical LLD s were 0.5-2 pCi/g.

- The U ^{235} K shell X ray window "U ^{235} " results were divided by the main (186 keV) U ^{235} to yield a ratio that corresponded to U ^{235} enrichment. Due to statistical and counting error concerns, minimal sample preparation, the slow change of uranium scaling factors over the range 3 to 50% U ^{235} enrichment (see Figure 2 " *Scaling Factors Vs U 235 Enrichment*"), and the presence of three distinct enrichments at the site, the ratios were interpreted broadly. Values in excess of 5 were routinely assumed to represent Depleted Uranium. Values between 2 and 5 were routinely assumed to represent Low Enriched Uranium (LEU). Values below 2 were interpreted as representing High Enriched Uranium (HEU). When the detected enrichment had a scaling factor greater than that normally used for survey unit in question, the larger scaling factor was applied to the data to determine the total Uranium activity level.

DISCUSSION

NaI(Tl) on site measurement of soil samples for Am ^{241}, U ^{235}, Co ^{60}, Cs ^{137}, and approximate uranium enrichment was successful. The system as expected, reliably detected these isotopes to well below compliance levels for the site (respectively 25, 30, 8, and 15 pCi/gram). Compliance levels for Am ^{241}, and U ^{235}, were effectively reduced to 5 – 10 pCi/g and 0.5 – 1.3 pCi/g respectively, due to the presence of additional transuranic and uranium isotopes, that were not readily detectable, but were part of the release criteria. As scaling factors varied over the site , the Am ^{241}, and U ^{235} activity concentrations equivalent to

the compliance criteria similarly varied. Typical detection levels were several tenths of a pCi/g for the listed isotopes, with the exception of Am^{241}. Am^{241} typical detection levels were typically 0.5 to 2 pCi/g.

The U^{235} activity was needed to reliably determine a useful ratio for the determination of the degree of U^{235} enrichment approximately 0.7- 0.8 pCi/g U^{235}. Two thirds of the samples with 0.7 to 1.0 pCi/g U^{235} yielded usable uranium enrichment ratios. The proportion increases as the U^{235} increases. Only the gross or integral count rate calibration was useful for this purpose. Peak results for the K shell X ray ROI yielded ratios that were incorrect. The peak result was attributed to the multiple separate emissions in the K Shell X ray window and the consequent effect on net peak results. Eight tenths of a pCi/g of U^{235} is equivalent to 18 to 47 pCi total uranium per gram depending on isotopic enrichment. Given this level of U^{235} activity, the method works and was useful in preventing under reporting of the total uranium activity levels present, and in validating the appropriateness of the scaling factors used. Use of the data was limited to identifying the uranium as Depleted Uranium, Low Enriched Uranium (3-5%), or High Enriched Uranium. Distribution of HEU and LEU in MOX (Mixed Oxide) (where only DU was expected) and DU areas was more widespread than expected. . Cs^{137} or Co^{60} activity levels of more than 1-2 pCi/g rendered the use of K Shell X ray window for isotopic identification infeasible. Am^{241} activity levels of 5-10 pCi/g also distort the results.

The dual gross count and peak counting systems were useful. The two results per ROI served as a quality control measure. If they agreed the result was accepted as valid. The results were investigated if they were contradictory. Peak counting was significantly more resistant to interference from higher energy isotopes. However the gross or integral counting was more sensitive by a factor of approximately 2. Cs^{137} or Co^{60} activity levels of more than 1-2 pCi/g rendered gross counting for Am^{241} and, U^{235} infeasible.

The physical conditions under which the NaI(Tl) counting system was used were important. NaI(Tl) is temperature sensitive. The instrument is best used under steady state temperature conditions. Installing air conditioning, heating, and barriers to rapid temperature changes can control daily temperature change effects. These features were put in place after difficulties in maintaining calibration and shifts in instrument response were detected and traced to temperature fluctuations. Maintenance of the facility at a constant temperature for 24 hours a day was found to be useful. The steel shielding and sand shielding together retained nighttime temperatures for several hours after sunrise. When the mass equilibrated to daytime temperatures, it was consistently found that the system was significantly different in energy response, then early in the morning.

Shielding is necessary. It allows the reduction of the counting time to a period short enough to allow screening of final survey samples, and deliver prompt data needful to support ongoing remediation work. In addition, it prevents the analysis from being affected by the frequent changes in background. Large amounts of material, soil, waste, et al are constantly being moved during a major decommissioning project, with consequent changes in background levels that can affect results of a counting system that is not adequately shielded. The replacement of concrete block shielding with 50 bags of playground sand and the addition of 0.75" of steel shielding halved the required counting time and eliminated analysis errors due to sudden shifts in background.

CONCLUSION

On site NaI(Tl) soil sample monitoring for Transuranics, Uranium, Co^{60}, and Cs^{137} works. Sensitivity is sufficient to meet typical compliance levels. Turnaround time is short enough to allow effective (same day) remediation support. If approximately 0.7-0.8 pCi/g U^{235} is present, the uranium enrichment can usually be determined with sufficient accuracy to ensure the application of a conservative scaling factor for the

determination of total uranium activity. Sample preparation beyond the requirements that the material be placed into a Marinelli beaker, have a mass of 500 grams or more, and be free of standing water was not necessary. Significant shielding is required to reduce counting time and to eliminate sudden spikes in background due to shipping, excavation, and waste handling activities. Maintenance of a constant temperature in the counting facility prevents down time and counting error.

Table I. Isotopic Abundance Vs U^{235} Enrichments

ISOTOPE	K X RAY FRACTION	DECAY CHAIN FRACTION	0.3%	0.711%	2%	3%	5%	10%	20%	50%	90%	97%
U^{238}	0.00125	1	0.9140	0.4900	0.2726	0.2059	0.1330	0.0661	0.0284	0.0059	0.0005	0.0000
Th^{234}	0.0543	1	0.9140	0.4900	0.2726	0.2059	0.1330	0.0661	0.0284	0.0059	0.0005	0.0000
Pa^{234} IT	5.93E-3	0.9987	0.9140	0.4900	0.2726	0.2059	0.1330	0.0661	0.0284	0.0059	0.0005	0.0000
PA^{234}	0.5712	1.30E-03	0.9140	0.4900	0.2726	0.2059	0.1330	0.0661	0.0284	0.0059	0.0005	0.0000
U^{235}	0.1376	1	0.0170	0.0230	0.0365	0.0410	0.0451	0.0473	0.0458	0.0385	0.0311	0.0330
Th^{231}	0.0295	1	0.0170	0.0230	0.0365	0.0410	0.0451	0.0473	0.0458	0.0385	0.0311	0.0330
U^{234}	8.80E-05	1	0.0680	0.4900	0.6853	0.7531	0.8219	0.8867	0.9258	0.9558	0.9685	0.9685

Table II. K Shell X Ray (89-120 keV) Fractional Abundance vs. Isotope and U^{235} Enrichment

ENRICHMENT	0.3%	0.711%	2%	3%	5%	10%	20%	50%	90%	97%
U^{238}	0.0001	0.0001	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Th^{234}	0.0496	0.0266	0.0148	0.0112	0.0072	0.0036	0.0015	0.0003	0.0000	0.0000
Pa^{234} IT	0.0054	0.0029	0.0016	0.0012	0.0008	0.0004	0.0002	0.0000	0.0000	0.0000
PA^{234}	0.0007	0.0004	0.0002	0.0002	0.0001	0.0000	0.0000	0.0000	0.0000	0.0000
U^{235}	0.0023	0.0032	0.0050	0.0056	0.0062	0.0065	0.0063	0.0053	0.0043	0.0045
Th^{231}	0.0005	0.0007	0.0011	0.0012	0.0013	0.0014	0.0014	0.0011	0.0009	0.0010
U^{234}	0.0000	0.0000	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
Sum of K Shell X rays	0.0587	0.0338	0.0228	0.0195	0.0157	0.0120	0.0095	0.0069	0.0053	0.0056
Ratio to 97.6% Enrichment	10.48	6.04	4.07	3.48	2.81	2.15	1.69	1.23	0.95	1.00

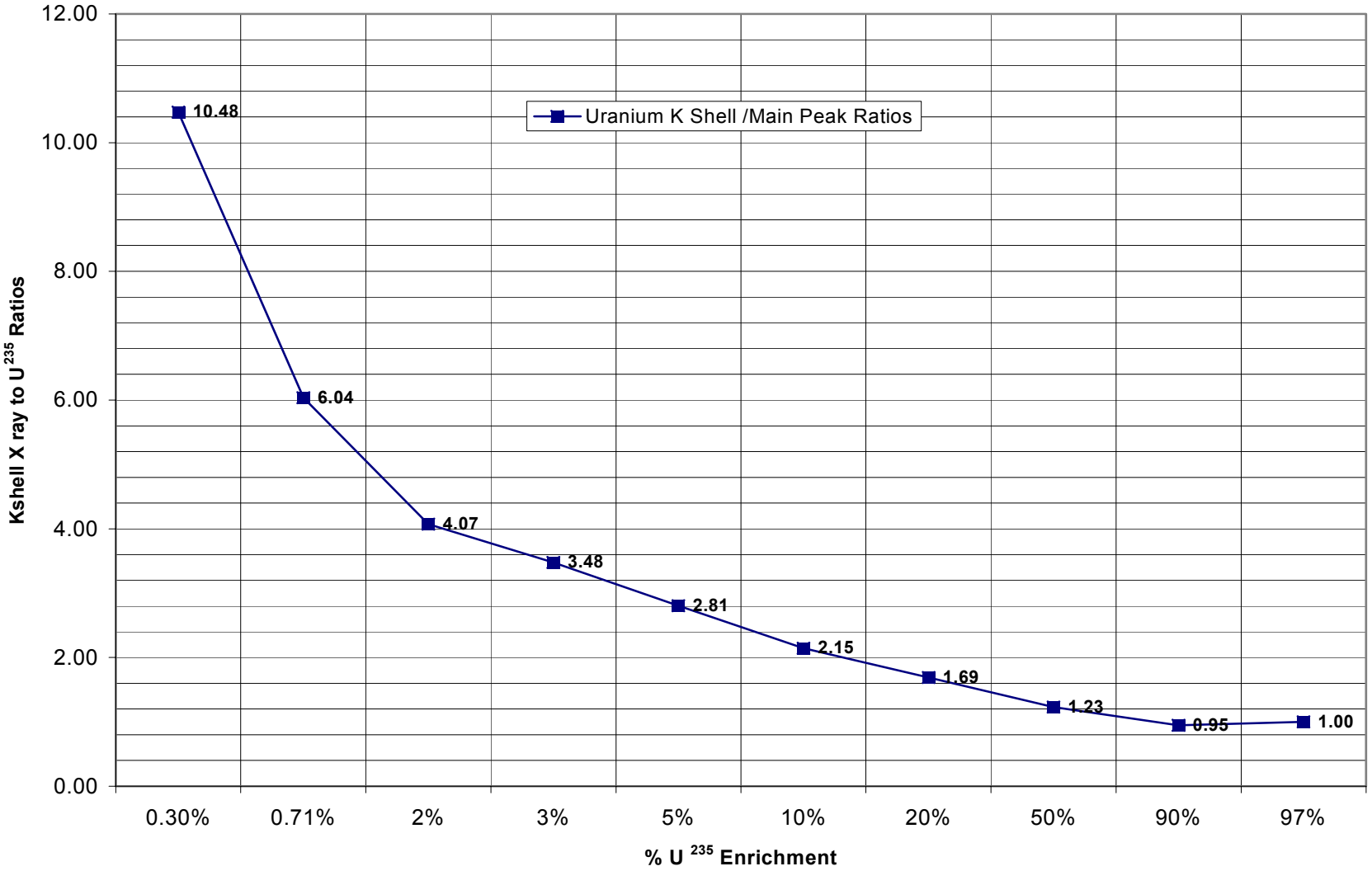

Fig. 1. Uranium K Shell/U^{235} Ratios Vs U^{235} Enrichment

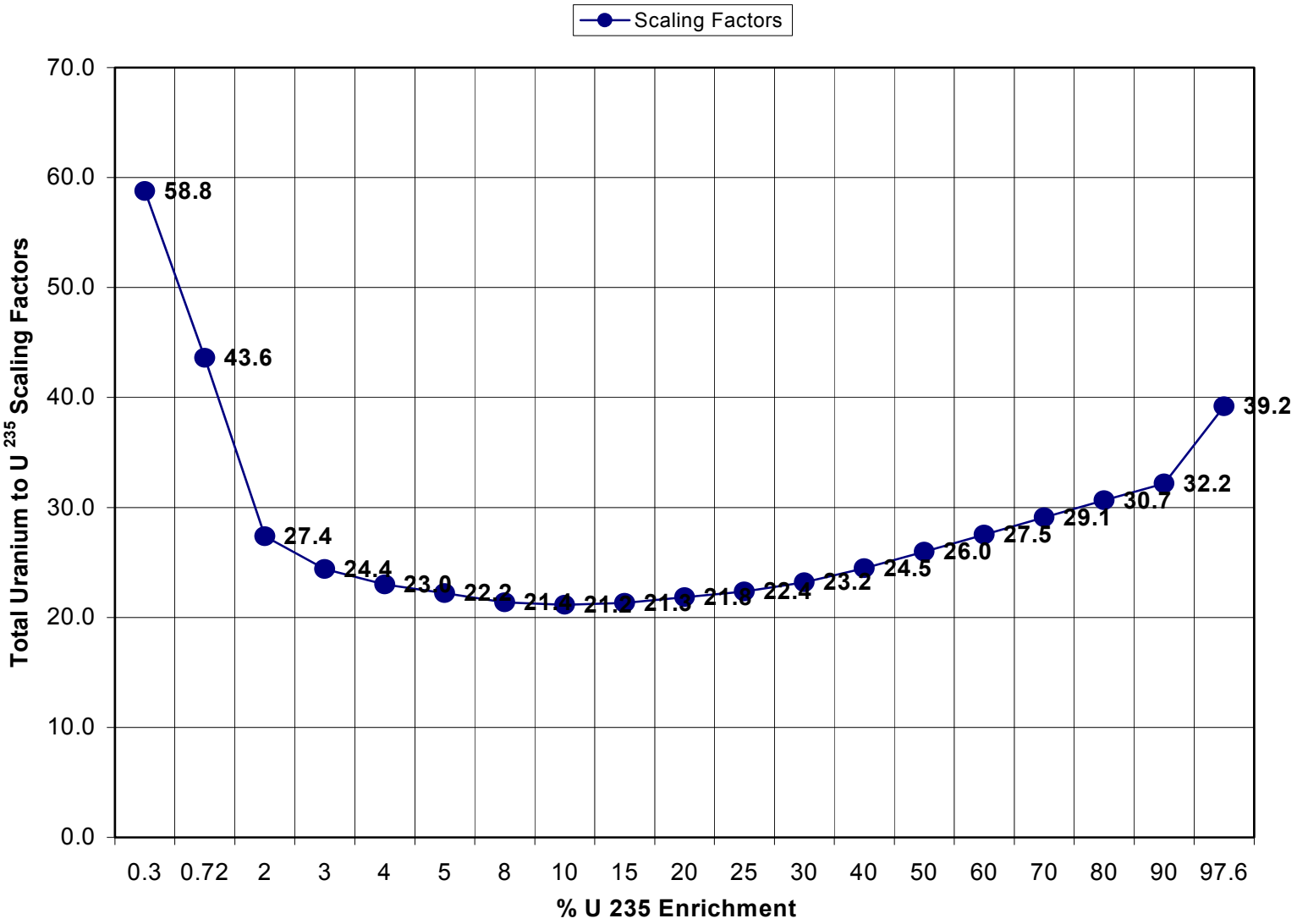

Fig. 2. Scaling Factors Vs U^{235} Enrichment