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Calculation of Uranium Isotopic Activity Composition Based on Data from Various Assay Methods

T. L. Rucker, Ph.D. and C. M. Johnson, Jr.
Science Applications International Corporation
301 Laboratory Road, P.O. Box 2501, Oak Ridge, TN 37831, USA.

Introduction

Uranium is often characterized in the environment and in waste associated with fuel cycle and weapons facilities that may have processed uranium at various enrichments of the ^{235}U isotope. These characterizations are normally accomplished by making isotopic activity measurements by alpha spectrometry, by making mass measurements of total uranium (e.g., fluorometry or Kinetic Phosphorescence Analysis) and isotopic measurements by mass spectrometry, or by performing gamma spectrometry measurements of the activity of ^{235}U and ^{238}U, assuming equilibrium with the ^{238}U progeny. However, the mass spectrometry measurements often only report isotopic composition (weight percent) of ^{235}U and gamma spectrometry does not provide information on the ^{234}U activity. Although ^{238}U and ^{235}U comprise most of the mass, ^{234}U is a significant fraction of the alpha activity at any enrichment and varies significantly by enrichment. Since the radiological health risk is proportional to the alpha activity and not to the mass, there is often a need to calculate isotopic uranium activities from total uranium mass or gamma spectrometry data. If alpha spectrometry is performed, isotopic alpha activity data is directly obtained. However, there may still be a need to calculate the total uranium and enrichment values for reporting or comparison purposes for data validation and evaluation.

Some characterization studies have approached this problem by empirically determining a $^{234}U/^{235}U$ ratio from alpha spectrometry or mass spectrometry made on representative samples taken from the site. However, many sites associated with the uranium fuel cycle and weapons production have processed and handled uranium at various enrichments. Therefore, empirically determining a $^{234}U/^{235}U$ ratio representative of the environment or waste generated at these sites, or even areas of these sites, is very difficult if not impossible.

The other alternative is to model the relationship between ^{234}U and ^{235}U as a function of uranium enrichment. It is known that the ^{234}U isotope is enriched along with the ^{235}U isotope in conventional gaseous diffusion enrichment processes. This is natural since the gaseous diffusion process produces an enriched stream and a depleted stream based on the mass. It should be noted that the atomic vapor laser isotope separation (AVLIS) enrichment process enriches only the ^{235}U isotope. However, AVLIS enriched uranium is not currently available in most uranium fuel cycle and weapons facilities.

Literature Data Sources

A literature search was performed to find resources that describe the relationship between ^{234}U and ^{235}U as a function of uranium enrichment. The *Health Physics Manual of Good Practices for Uranium Facilities*[1] (EGG-2530) provides the following equation for calculating the specific activity (μCi/g) from the uranium enrichment (weight percent ^{235}U):

$$S = (0.4 + 0.38E + 0.0034E^2) \quad (1)$$

where:

S = specific activity (μCi/g), and
E = weight % ^{235}U.

This equation has been widely used, including being referenced in Nuclear Regulatory Commission regulations.[2] This manual also presents a graph that shows the percent of total alpha activity by isotope vs. % weight ^{235}U that is said to be related to the specific activity equation. However, no equation was provided to calculate the activity percentages and no data was provided that could be used to fit such an equation. Furthermore, the sources of data used to develop the relationships and the specific activity equation were not referenced such that the data could be retrieved. Therefore, this reference can only be used to predict the ^{234}U and ^{235}U activity ratio by interpolation from the graph, which is imprecise. Furthermore, this reference cautions that the specific activity equation will not be accurate when different enrichments of uranium have been blended together or when the enriched uranium has been produced by recycling of irradiated uranium. Therefore, the specific activities calculated from the equation should only be considered an approximation.

A report prepared at the Oak Ridge Gaseous Diffusion Plant, K-1839[3], provides a number of graphs that predict the ratio of the ^{235}U to ^{234}U concentration as a function of enrichment with various types of feed, product, and tails design concentrations. However, no equation was provided to calculate ratios and no data was provided that could be used to fit such an equation. Again, this reference can only be used to predict the ^{234}U and ^{235}U activity ratio by interpolation from the graph. It is obvious from this reference that there is considerable variation in the ratio of the ^{235}U to ^{234}U depending on whether the enrichment cascade is designed for low enriched uranium product (power reactor fuel), high enriched uranium product (weapons and navel reactor fuel), dual enriched uranium product, and the use of uranium other than natural as feed (previously enriched uranium or reactor fuel recycled material). Therefore, the use of one equation for all situations may not be advisable.

Another Oak Ridge report, K/HS/593[4], provides an equation for the calculation of the weight percent ^{234}U from the weight percent ^{235}U as follows:

$$\% \, ^{234}U = 0.0015 + 0.0058E + 0.000054E^2 \quad (2)$$

where:

E = % ^{235}U by mass.

No reference was provided for the source of the data used to derive this equation. This reference states that the ratio of ^{234}U to ^{235}U in enriched uranium can vary by 50% or more for a given level of ^{235}U.

A third Oak Ridge report, K/PO/SUB-79/31057/1,[5] provides $^{235}U/^{234}U$ mass ratios and weight percentages for each uranium isotope for nine specific enrichments of uranium. The data was obtained from personnel at an operating gaseous diffusion plant in Oak Ridge and, therefore, is likely to be applicable to uranium that was enriched there. We have fit this data with various equations to compare the results with the curves found in the previous references and to evaluate their usefulness for predicting the isotopic activity concentrations when not measured directly.

Comparison of Data From Different Sources

Figure 1 compares the percent of the total uranium activity by isotope versus enrichment from three sources of data. The data taken from K/PO/SUB-79/31057/1 was used directly to produce the data points shown assuming isotopic specific activities (Curies per gram of isotope) of 6.13E-3 for ^{234}U, 2.14E-6 for ^{235}U, and 3.33E-7 for ^{238}U. The data taken from EGG-2530 was extrapolated from the graph in that reference. The data taken from K/HS/593 was calculated using equation (2) and assuming a total uranium specific activity as would be calculated from equation (1). It can be seen that the data from the different sources do not completely overlap but follow the same general trend.

The $^{234}U/^{235}U$ activity ratio, the total uranium specific activity, and the isotopic specific activity (μCi/g of uranium) versus enrichment were also compared for data taken from K/PO/SUB-79/31057/1, data extrapolated from several graphs in K-1839 for ideal enrichment cascades, data taken from EGG-2530 calculated using equation (1), and data taken from K/HS/593 calculated using equation (2) and assuming the isotopic specific activities listed above. The data from K/PO/SUB-79/31057/1 and K-1839 provide similar $^{234}U/^{235}U$ activity ratio results over most of the enrichment range, but the data from the K/HS/593 equation provides a completely different general trend and differs from the ratios predicted by the other equations by 30% and even more in the depleted region. The curves for specific activity versus enrichment from different sources follow the same trend. However, the total uranium specific activity curve from the K/PO/SUB-79/31057/1 data appears to be more linear and the EGG-2530 crosses it at two points, at approximately 0.5 wt% ^{235}U and at approximately 70 wt% ^{235}U. The curve for the specific activity of ^{234}U generated from the K/HS/593 equation appears to correspond to the EGG-2530 equation fairly well even though the percent of the total uranium activity predicted by the two references shown in Figure 1 is not as comparable.

Development of Equations for Practical Application

Because the K/HS/593 equation does not produce comparable results to other sources of data when predicting the $^{234}U/^{235}U$ ratio, as demonstrated in the $^{234}U/^{235}U$ activity ratio versus enrichment comparison, and because of the difficulty in interpolating graphs from other sources, we have chosen to fit the data presented in K/PO/SUB-79/31057/1 to equations that will allow the calculation of various parameters depending on the parameters that have been measured. The

K/PO/SUB-79/31057/1 curve for the total uranium specific activity (μCi/g of uranium) versus enrichment was plotted and fitted with the following equation:

$$\text{Total Uranium } \mu\text{Ci/g U} = 0.19192 + 0.64333\ (\text{wt\% } ^{235}\text{U}) - 0.00025\ (\text{wt\% } ^{235}\text{U})^2 \quad (3)$$

This equation can be used to calculate the total uranium alpha activity when only the total mass concentration and weight percent ^{235}U have been measured. Although this equation is different than that given in EGG-2530, the results are similar and this equation is in better agreement with the data presented in K-1839, which is the best documented study of the relationship between the isotopes that we have found.

The results of the $^{234}U/^{235}U$ activity ratio versus enrichment data was also plotted and curve fitted. The equation for the fitted line is:

$$^{234}\text{U}/^{235}\text{U Activity Ratio} = 22.012 + 9.372\ (\log \text{wt\% } ^{235}\text{U}) - 3.516\ (\log \text{wt\% } ^{235}\text{U})^2 \quad (4)$$

This equation can be used to calculate the uranium isotopic activities when only the total mass concentration and weight percent ^{235}U have been measured. Likewise, the relationship of $^{234}U/^{235}U$ activity ratio versus the $^{238}U/^{235}U$ activity ratio was also plotted and fitted. The equation for the fitted line is:

$$^{234}\text{U}/^{235}\text{U Activity} = 27.18 + 0.3004\ (^{238}\text{U}/^{235}\text{U activity}) + 0.00143\ (^{238}\text{U}/^{235}\text{U activity})^2 \quad (5)$$

This equation can be used to calculate the ^{234}U activity when ^{235}U and ^{238}U activities have been measured by gamma spectrometry. Finally, the relationship of enrichment versus the $^{238}U/^{235}U$ activity ratio was plotted and curve fitted. The equation for the fitted line is:

$$\text{Weight \% } ^{235}\text{U} = 1.065 - 0.7317\ (\log\ ^{238}\text{U}/^{235}\text{U activity}) - 0.129\ (\log\ ^{238}\text{U}/^{235}\text{U activity})^2 \quad (6)$$

This equation can be used to calculate the weight percent ^{235}U when ^{235}U and ^{238}U activities have been measured by gamma spectrometry.

Results

These equations have been used to compare data from a large number of samples that have had the combination of isotopic uranium measurements performed by alpha spectrometry, ^{238}U progeny and ^{235}U measurements performed by gamma spectrometry, and total mass concentration and weight percent ^{235}U measurements performed by thermal ionization mass spectrometry (TIMS). The comparisons were performed to help validate and evaluate the data to determine if potential biases existed. Results for four representative Oak Ridge waste samples are shown in Table-1.

It can be seen in the table that some of the calculated results often compare favorably with the measured results when the measurement uncertainty is included, although some of the results are significantly different than what would have been expected. It appears that the poorest

agreement is between the enrichment measured by TIMS and that calculated from the gamma spectrometry data. This is likely due to the large relative uncertainties obtained for the gamma ^{235}U measurements.

It is important to incorporate in the comparison an evaluation of the uncertainty in the measurements. Unfortunately, uncertainties are not usually reported for the total uranium mass concentration measurements. It is obvious, however, that the uncertainty relative to the result increases as the uranium concentration decreases. Therefore, the results can be expected to not agree as well at lower concentrations. In addition to the measurement uncertainties, sampling and subsampling uncertainties must also be incorporated to an extent, depending on the heterogeneity of the matrix, since different subsamples are often processed for the different analyses. The uncertainty of the applicability of the derived equations to all sources of uranium must also be taken into account.

While exact agreements cannot be expected, general conclusions can be reached that can be valuable in detecting significant biases. By performing this type of comparison, we have identified situations where there was a significant bias in one method or another due to sample preparation or calibration deficiencies. It can also be concluded from the data presented in the table that the ^{235}U results by alpha spectrometry may have a positive bias. This is most likely due to the tail of the larger ^{234}U peak interfering in the spectral region used for the ^{235}U determination.

Conclusion

Both of the comparisons of the percent of the total uranium activity by isotope versus enrichment and the $^{234}U/^{235}U$ activity ratio versus enrichment have demonstrated that the various sources of data predicting the relationship of uranium isotopes evaluated in this report do not all agree. This may not be surprising since it is apparent that considerable variability in the relationships can occur depending on the design of the enrichment cascade where the uranium was enriched, the type and enrichment of the feed material, and on whether the uranium has resulted from mixing of different enrichments. We have chosen K/PO/SUB-79/31057/1 as the source of data that is most consistent and applicable to uranium generated from Oak Ridge. The equation for calculation of the weight percent ^{234}U from the weight percent ^{235}U from K/HS/593 appears to be particularly inappropriate for the calculation of ^{234}U activity concentrations.

The equations we have derived from that data have shown reasonable agreement with actual isotopic measurement data made on Oak Ridge waste samples. We believe it is appropriate to use these equations for general comparison of analytical results for validation and evaluation purposes. However, because the actual isotopic ratios vary not only with enrichment but also depend on the design of the enrichment cascade, the composition of the enrichment feed material, and on the amount of blending of enrichments that have taken place, there is considerable uncertainty in the calculated results using any equation. Therefore, it is recommended that measurements made for critical applications (e.g., health and environmental protection) be performed by alpha spectrometry so that a direct measurement of the uranium alpha activity can be obtained.

References

1 EGG-2530 UC-41, B. L. Rich et al., "Health Physics Manual of Good Practices for Uranium Facilities," Idaho National Engineering Laboratory, Department of Energy, June 1988, pp 2-10.

2 10 CFR 20 Appendix B, Code of Federal Regulations, "Standards for Protection Against Radiation, Concentrations in Air and Water Above Natural Background."

3 K-1839 UC-15, S. Blumkin and E. Von Halle, "The Behavior of the Minor Uranium Isotopes in Separation Cascades," Union Carbide Corporation Nuclear Division, Oak Ridge Gaseous Diffusion Plant, January 1976.

4 K/HS/593, R. E. Rodriguez and C. A. Johnson, "Results of the Outdoor Radiological Survey of Six Priority Areas at the K-25 Site," Martin Marietta Energy Systems, March 1995, pp 3-4.

5 K/PO/SUB-79/31057/1, "Airborne Uranium, Its Concentration and Toxicity in Uranium Enrichment Facilities," Envirosphere Company, February 1979.

Table 1. Calculation of Isotopic Uranium and Total Uranium Alpha for Comparison to Alpha and Gamma Spectrometry Results

	EXAMPLE 1		EXAMPLE 2		EXAMPLE 3		EXAMPLE 4	
Analyses	**Result (pCi/g)**	**Uncertainty (pCi/g)**	**Result (pCi/g)**	**Uncertainty (pCi/g)**	**Result (pCi/g)**	**Uncertainty (pCi/g)**	**Result (pCi/g)**	**Uncertainty (pCi/g)**
Total U by TIMS	3840 µg/g		3600 µg/g		490 µg/g		9.0 µg/g	
U-235 wt%	0.53	0.05	0.54	0.05	0.62	0.05	4.3	
Calculated Isotopic Uranium								
U-234 Calculated	**8.73E+02**		**8.40E+02**		**1.38E+02**		**2.25E+01**	
U-235 Calculated	**4.36E+01**		**4.16E+01**		**6.50E+00**		**8.28E-01**	
U-238 Calculated	**1.26E+03**		**1.18E+03**		**1.61E+02**		**2.84E+00**	
Total U Alpha (Calc)	**2.05E+03**		**1.94E+03**		**2.89E+02**		**2.66E+01**	
Alpha Spectrometry Results								
U-234	8.65E+02	6.90E+01	1.01E+03	7.30E+01	6.40E+01	1.36E+01	1.95E+01	3.10E+00
U-235	9.84E+01	2.30E+01	9.67E+01	2.20E+01	5.32E+00	2.25E+00	2.73E+00	1.10E+00
U-238	1.41E+03	8.70E+01	1.46E+03	8.70E+01	7.91E+01	1.64E+01	2.83E+00	1.20E+00
Total U Alpha Sum	**2.37E+03**	**1.13E+02**	**2.57E+03**	**1.16E+02**	**1.48E+02**	**2.14E+01**	**2.51E+01**	**3.50E+00**
Calculated Results from Gamma								
U-234	**7.86E+02**		**1.15E+04**		**9.69E+01**		**2.49E+01**	
U-235 wt%	**3.66E-01**		**4.78E-01**		**2.14E+00**		**6.14E+00**	
Gamma Spectrometry Results								
Th-234	1.37E+03	1.20E+01	1.41E+04	1.10E+03	2.09E+01	1.18E+01	1.78E+00	1.40E-01
U-235	4.17E+01	2.00E+01	5.61E+02	1.70E+02	3.79E+00	2.14E+00	9.35E-01	2.80E-02

Note: Data in bold are results calculated from curve fit equations.
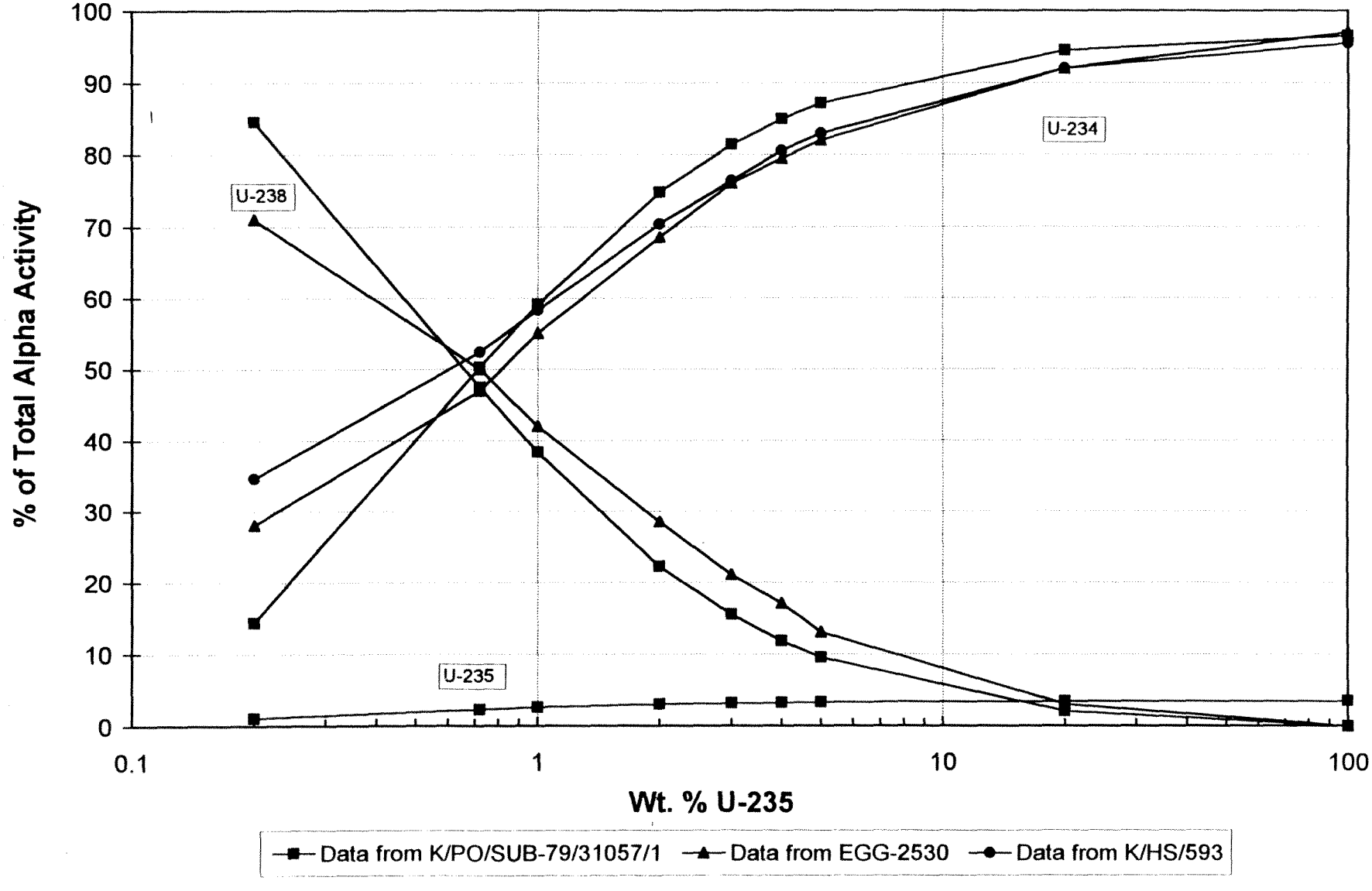
Figure 1. Percent of Total Uranium Activity vs. Enrichment by Isotope