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UCID - 16553

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**ERROR ANALYSIS OF THE METHOD FOR THE DETERMINATION
OF URANIUM BY CONTROLLED-POTENTIAL COULOMETRY**

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ERROR ANALYSIS OF THE METHOD FOR THE DETERMINATION OF URANIUM BY CONTROLLED-POTENTIAL COULOMETRY

ABSTRACT

The sources and magnitudes of the systematic and random errors associated with this method are outlined. A determination of uranium in solution is described in which the chemical and electrical calibrations of the coulometric instrument are pooled to calculate the quantity of uranium in the sample. These results are evaluated statistically.

INTRODUCTION

Fission cross section measurements for a number of isotopes of uranium and plutonium are presently being made at LLL using the 100-MeV electron linear accelerator as a pulsed source for neutrons.¹ For these measurements, accurate chemical analyses of the purified isotope solutions are required. The determination of the uranium concentration in one of these solutions by controlled-potential coulometry has prompted a careful examination of the errors associated with this technique. Coulometry is somewhat unusual because the analytical results can be obtained both by standardization with National Bureau of Standards (NBS) reference materials and by calculation from fundamental physical quantities, i.e., on an absolute basis, like gravimetry. We are interested in how these two approaches can be combined to obtain increased accuracy.

Not all of the procedural details and equipment used in this study have been optimized or refined in accuracy and precision. One of the purposes of this evaluation was to learn where the technique could be improved. This report examines the sources and magnitudes of the systematic and random errors associated with the uranium analysis as it is ordinarily done to provide a relative accuracy of 0.05-0.1%.

In the error analysis, two types of errors were distinguished: (1) the random error, which was obtained numerically as the relative standard deviation of the estimated mean value of the concentration of uranium in an

analyzed solution; and (2) the systematic error, which was evaluated as the worst-case error or bias that could be associated with the mean value of the uranium concentration when all the possible individual biases influencing the mean value were taken into account.

A similar error analysis was performed for the determination of plutonium by controlled-potential coulometry (UCID-16554).²

METHOD FOR THE DETERMINATION OF URANIUM

In the standard method^{3,4} for the determination of uranium by controlled-potential coulometry, U(VI) is reduced to U(IV) at a mercury cathode in a medium of 0.5 M H_2SO_4 and 0.1 M $\text{NH}_2\text{SO}_3\text{H}$. The current consumed in a complete electrolysis of an aliquot of sample solution is integrated electronically as a measure of the amount of uranium present. The current integral $\int i dt$ appears as a voltage at the output of the integrator. To calculate the quantity of uranium in the sample aliquot, the integrator must be calibrated in terms of weight of uranium per volt readout, either by electrolyzing uranium solutions of known concentration (chemical calibration) or by calibrating the integrator electrically in coulombs per volt and relating this value to the quantity of uranium via Faraday's Law. The latter procedure is independent of a chemical analytical standard (it is an absolute technique), but it assumes 100% current efficiency, after blank correction, for the electrolysis.

As employed here, the electrolysis procedure was to pre-electrolyze the solutions at +0.085 V vs SCE until the current decreased to 5 μA , zero the integrator, then reduce the U(VI) at -0.325 V until the current decreased to 10 μA , and record the integrator output voltage. To obtain the blank correction, a supporting electrolyte that contained no uranium was electrolyzed in the same manner and for the same length of time as the uranium aliquots. Details of the coulometric instrumentation and the

electrolysis cell are given in Refs. 5 and 4, respectively. The voltage measuring device was a Dana Laboratories Model 5000 digital voltmeter.*

CHEMICAL CALIBRATION

The NBS has two chemical standards available for the determination of uranium: SRM 950a, U_3O_8 ; and SRM 960, uranium metal. Occasionally, other laboratories have reported a significant disagreement between the chemical and electrical calibration values when using U_3O_8 ; however, this has not been encountered at LLL. It is suspected that some of the discrepancies have been due to either neglecting or improperly correcting for the blank. Because our analyses have always shown agreement within 0.1%, we have continued to use U_3O_8 as the chemical standard.

To prepare the standard solution for the present work, approximately 1 g of NBS U_3O_8 was ignited at $920 \pm 20^\circ C$ in an open crucible for 2 h and cooled in a desiccator. The material was weighed to the nearest 0.01 mg on a Mettler Type H16 balance, transferred to a Corning No. 5680 certified volumetric flask, and dissolved in 3 M HNO_3 . Dilution of the solution to volume gave a concentration of approximately 10 mg U/cm³. For each coulometric run, an aliquot of this solution was taken using a 1-cm³ Misco micropipet previously calibrated with mercury.

The chemical calibration value for each run, Q_c , is calculated from the following equation:

$$Q_c \text{ (mg U/volt)} = \frac{[\text{wt } U_3O_8 \text{ taken (mg)}] \left[\frac{3U}{3U + 8O} \text{ (gravimetric factor)} \right] [U_3O_8 \text{ purity}]}{\text{flask volume (cm}^3\text{)}} \times \frac{\text{Volume of sample taken (cm}^3\text{)}}{\text{Net integrator readout (V)}} \quad (1)$$

The weight of the U_3O_8 taken was obtained by difference weighing of the crucible. The net integrator readout equals the readout for the uranium aliquot minus the readout for blank.

*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Atomic Energy Commission to the exclusion of others that may be suitable.

The errors and uncertainties associated with the quantities in Eq. (1) are as follows:

- Weight of U_3O_8 Taken — The accuracy of the balance at 1 g is generally ± 0.1 mg. Two weighings are required to obtain the net weight of U_3O_8 , and the standard deviation associated with each is estimated to be 0.03 mg. Because the density of U_3O_8 is approximately 8.3, about the same as that of the weights, no air buoyancy correction is required.
- Gravimetric Factor — The atomic weight of natural uranium is 238.029 with a "reliability" of 4 ppm. That of oxygen is 15.9994 with a "reliability" of 19 ppm. The worst-case error in the factor would be $(3 \times 4) + (8 \times 19)$ ppm = 164 ppm (see Eq. (1)).
- U_3O_8 Purity — The percent assay is given by the NBS as 99.94% with an "uncertainty" of 0.02%; however, data obtained at the New Brunswick Laboratory show that the percent assay is more likely 99.93%.⁶
- Flask Volume — Corning states that the limit of error of the 100-cm³ certified flask is ± 0.08 cm³.
- Volume of Sample Taken — Precision grade micropipets of 1-cm³ volume are specified as accurate to 0.001 cm³. A number have been calibrated at LLL in the past 15 years and none has been found outside this range.
- Net Integrator Readout — The accuracy of the digital voltmeter is specified for 6 months as within 0.01% of reading $\pm 0.002\%$ of range. The instrument was calibrated within 6 months of use and was used on the 10-V range where the resolution is 0.0001 V. The voltages recorded for the standard uranium aliquot and the blank were about 9.7 and about 0.006, respectively.

Table 1 summarizes estimates of the worst-case errors in the chemical calibration value. Where the quantity used to compute Q_c is obtained by

Table 1. Worst-case systematic errors in the determination of the coulometric chemical-calibration value, Q_c .

Quantity	Approximate value	Estimated error	Estimated relative error (%)
Wt of U_3O_8 taken	1.0 g	0.26 mg	0.026
Gravimetric factor	0.848 mg U/mg U_3O_8	—	0.016
U_3O_8 purity	99.93%	—	0.02
Flask volume	100 cm ³	0.08 cm ³	0.08
Pipet volume	1 cm ³	0.001 cm ³	0.10
Net integrator readout	9.7 V	0.001 V	0.01
Sum of errors, $\Delta_c =$			0.25%

a difference measurement, the systematic error in the difference is taken to be that of a single measurement if the same range of the instrument is used in both measurements, or twice that of a single measurement if different ranges are used. The random error(s) (expressed as the standard deviations, if known) are then added to obtain the total error.

For this particular analysis, one standard solution was made up, and two aliquots of this solution were taken for coulometric analysis. Approximately 10 mg of uranium were measured per run and corrected for a blank of 0.006 mg. Approximately 13 years experience with this uranium determination has shown that the standard deviation of the blank is 0.001 mg, which is 0.01% of the total uranium, and that the relative standard deviation of the uranium measurement at the 10-mg level is 0.04%. The relative standard deviation of a single measurement of Q_c therefore is $[(0.04)^2 + (0.01)^2]^{1/2} = 0.04\%$. The two values obtained and their mean are as follows:

$$\begin{array}{r} 0.45545 \\ 0.45553 \\ \hline \text{Mean, } \bar{Q}_c = 0.45549 \text{ mg U/volt,} \\ \text{with } s_{Q_c} = 0.04\%. \end{array}$$

The relative standard deviation of the mean is $s_{\bar{Q}_c} = s_{Q_c} / \sqrt{n_c}$. Because only one sample of U_3O_8 was analyzed, there was only one independent determination of Q_c in this group of experiments. The determination is generally known to be reliable, but to be on the conservative side, n_c will be taken to be 1 in computing $s_{\bar{Q}_c}$.

ELECTRICAL CALIBRATION

In this technique of electrical calibration,⁷ the potentiostat is used as a constant-current source, and the time required for the integrator output voltage to traverse two levels is measured. The voltage drop across a precision resistor is measured to obtain the value of the current. This yields the calibration value in coulombs per volt. Then, from the value of the Faraday constant and the atomic weight of uranium, a calibration in mg U/volt is obtained. The overall equation is:

$$Q_e, \text{ mg U/volt} = \frac{\left(\begin{array}{c} \text{voltage} \\ \text{on std} \\ \text{resistor} \end{array} \right) \left(\begin{array}{c} \text{elapsed} \\ \text{time, s} \end{array} \right) \left(\begin{array}{c} \text{at. wt. of U} \\ 2, \text{ g/mole} \end{array} \right) 1000}{\left(\begin{array}{c} \text{value} \\ \text{of std} \\ \text{resistor, } \Omega \end{array} \right) \left(\begin{array}{c} \text{integrator} \\ \text{readout} \\ \text{voltage} \\ \text{difference} \end{array} \right) \left(\begin{array}{c} \text{value of} \\ \text{Faraday} \\ \text{constant, C/mol} \end{array} \right)} \quad (2)$$

The errors and uncertainties associated with the quantities in Eq. (2) are as follows:

- Voltage on Standard Resistor and the Integrator Readout Voltage Difference — Both were measured with the Dana Model 5000 digital voltmeter which has accuracy specifications as given above. For measurement of the standard resistor, the voltmeter was used on the 1-V range, where the resolution was 1 μ V.
- Elapsed Time — A Standard Electric Company Model S-100 clock with a resolution of 0.2 s was used. Its accuracy is specified as 0.2 s; however, its accuracy is also dependent on the shortterm accuracy of the line frequency. The Pacific Gas and Electric Company guarantees the frequency to be accurate to $\pm 0.08\%$, averaged over 24 h. Several short term tests in the Building

131 instrument shops have revealed no deviations greater than 0.05%. Periods measured for Q_e were 900-1100 s.

- Value of Standard Resistor — A 155.00- Ω , Model SR-1, standard wire-wound resistor made by Electro-Scientific Industries was used. Long-term drift of the value of this resistor is advertised to be less than 0.005%. It was last calibrated in 1966.
- Atomic Weight of Uranium — The atomic weight of natural uranium is 238.029 with a "reliability" of 4 ppm. It is divided by $n = 2$ to obtain the equivalent weight of uranium.
- Value of Faraday Constant — The current value of the Faraday constant is given as $96,484.6 \pm 0.3$ C/mol.⁸

Table 2 summarizes estimates of the worst-case errors in the determination of the electrical calibration value.

The coulometric instrumentation is calibrated electrically at approximately one to two year intervals. It is important to know if the calibration values are drifting, but thus far there is no evidence of this. Table 3 lists the past electrical calibration values for one of the instruments. The equipment and technique were the same for all but

Table 2. Worst-case systematic errors in the determination of the coulometric electrical calibration value, Q_e .

Quantity	Approximate value	Estimated error	Estimated relative error, %
Voltage on std resistor	1.35 V	0.1 mV	0.01
Integrator readout voltage difference	11 V	1 mV	0.01
Elapsed time	1000 s	—	0.05
Value of std resistor	155 Ω	—	0.005
At. wt. of uranium	238 g/mole	—	0.0004
Value of Faraday constant	96,500 C/mol	0.3 C/mol	0.0003
Sum of errors, $\Delta_e = 0.076\%$			

Table 3. Electrical calibration values of controlled-potential coulometry system, Serial No. 1. (Reduction current, 27- Ω range.)

Date of calibration	Mean value, $\bar{Q}_e$, C/V
November, 1966	0.36922
April, 1969	0.36903
July, 1970	0.36927
December, 1971	0.36921
February, 1974	0.36925

the most recent calibration, when the more accurate Dana digital voltmeter was used and the voltage on the standard resistor was measured with the digital voltmeter rather than a portable potentiometer. Thus, even though the 1974 data are believed to be more accurate than the 1966-1971 data, there is still no discernible drifting trend.

The most recent value of $\bar{Q}_e$ (Table 3), obtained 2 months after the instrument was used for the analysis of the unknown uranium solution and the standard uranium solution, was computed from a set of 11 measurements of Q_e . The relative standard deviation of a single measurement, computed from

$$s_{Q_e} = \left[\frac{\sum_{i=1}^{11} (\bar{Q}_e - Q_e)^2}{10} \right]^{1/2} \cdot \frac{100}{\bar{Q}_e}, \quad (3)$$

is 0.026%. For most calibration sets, s_{Q_e} is usually in the range of 0.02-0.04%. The relative standard deviation of the mean is then

$$s_{\bar{Q}_e} = s_{Q_e} / \sqrt{n_e}.$$

The electrical calibration value for uranium, computed from the most recent calibration data and using Eq. (2), is $\bar{Q}_e = 0.45546$ mg U/volt.

POOLING THE TWO CALIBRATION VALUES

In pooling the two independent calibration values, $\bar{Q}_c$ and $\bar{Q}_e$, it is assumed, as discussed earlier, that there is no systematic bias between the

two and that this has been adequately proven. These results give no evidence to the contrary.

The pooled value for the instrument calibration for uranium, $\bar{Q}$, could be obtained by computing a mean from $\bar{Q}_c$ and $\bar{Q}_e$, weighted in proportion to the variances of Q_c and Q_e . However, because their variances are not greatly different and because the value of $\bar{Q}_c$, although less precise, tends to cancel chemical and technique errors, the values of $\bar{Q}_c$ and $\bar{Q}_e$ will be pooled with equal weights. Therefore, our best estimate of the calibration value is:

$$\begin{aligned}\bar{Q} &= \frac{\bar{Q}_c + \bar{Q}_e}{2} = \frac{0.45549 + 0.45546}{2} \\ &= 0.45547_5 \text{ mg U/volt.}\end{aligned}\tag{4}$$

In computing the worst-case error in $\bar{Q}$, the errors Δ_c and Δ_e must be added, and an additional increment must be included because the integrator output voltage divider is used at one setting for the electrical calibration and at a different setting for the chemical calibration and the uranium samples. This affects the uranium results in the same way as an error in Q_c . This error can be estimated as 0.01% from the non-linearity specification of the General Resistance voltage divider.

The worst-case systematic error in $\bar{Q}$ would therefore be equal to the mean of the individual errors in Q_c and Q_e , taken from Tables 1 and 2, and the voltage divider error. Thus

$$\Delta_{\bar{Q}} = \frac{\Delta_c + \Delta_e + 0.01}{2} = \frac{0.25 + 0.076 + 0.01}{2} = 0.168\%.\tag{5}$$

The variance or relative standard deviation of $\bar{Q}$ can be calculated using the relation

$$\text{Var}(y) = a^2 \text{Var}(x_1) + b^2 \text{Var}(x_2),\tag{6}$$

where

$$y = ax_1 + bx_2. \quad (7)$$

From Eq. (4), then

$$\text{Var}(\bar{Q}) = (1/2)^2 \text{Var}(\bar{Q}_c) + (1/2)^2 \text{Var}(\bar{Q}_e) \quad (8)$$

$$\text{Var}(\bar{Q}) = 1/4 \left[\frac{(\sigma_{Q_c})^2}{n_c} + \frac{(\sigma_{Q_e})^2}{n_e} \right], \quad (9)$$

and the standard deviation of the mean $\bar{Q}$ is:

$$\sigma_{\bar{Q}} = [\text{Var}(\bar{Q})]^{1/2}. \quad (10)$$

The estimate of σ_{Q_c} is $s_{Q_c} = 0.04\%$, and $n_c = 1$. The estimate of σ_{Q_e} is $s_{Q_e} = 0.026\%$ from Eq. (3) and $n_e = 11$. Thus the estimate of the relative standard deviation of the mean is:

$$\begin{aligned} s_{\bar{Q}} &\approx 1/2 \left[\frac{(0.04)^2}{1} + \frac{(0.026)^2}{11} \right]^{1/2} \\ &\approx 0.020\%. \end{aligned} \quad (11)$$

This value is combined with the systematic error, Eq. (5), to obtain the worst-case total error in $\bar{Q}$ and with the standard deviation of the uranium measurement to obtain the precision of the uranium-concentration estimate for the unknown solution.

MEASUREMENT OF URANIUM IN UNKNOWN SOLUTION

The sample, as received, was a solution of isotopically nearly pure U-238 in HNO_3 at a concentration of about 50 mg/cm^3 . Three 0.5-cm^3 aliquots of this solution were transferred to 50-cm^3 flasks with a calibrated micropipet. One coulometric run was carried out on each of the three diluted solutions using a calibrated, Class A, 7-cm^3 "to deliver" pipet.

Of the three flasks, two were Corning Class A with a specified error limit of 0.05 cm^3 , and one was a certified flask of the same tolerance.

The 7-cm³ pipet was calibrated four times with water and found to have a mean volume of 7.004 cm³, with a relative standard deviation of about 0.005 cm³. The 0.5-cm³ pipet was a Misco micropipet calibrated with mercury.

As was done for the chemical calibration, the integrator readout for the uranium solution was corrected for a blank equivalent to 0.006 mg U. The concentration of uranium in the unknown solution was calculated from the equation:

$$\begin{array}{l} \text{Concentration} \\ \text{of unknown} \\ \text{solution,} \\ \text{mg U/cm}^3 \end{array} = \frac{(\text{net integrator})}{\text{readout, V}} \frac{(\text{flask volume, cm}^3)(\bar{Q}, \text{mg U/volt})}{(\text{coulometric})}{(\text{aliquot volume, cm}^3)(\text{volume of original})}{(\text{solution aliquot, cm}^3)} \quad (12)$$

Table 4 summarizes estimates of the worst-case errors in the uranium concentration value. The worst-case error in $\bar{Q}$ is taken to be the sum of the systematic error (from Eq. (5)) and the random error (from Eq. (11)).

The results of the three coulometric determinations for separate aliquots of the original solution, based on $\bar{Q} = 0.45547_5$ mg U/volt, are

$$\begin{array}{r} 49.724 \\ 49.810 \\ 49.810 \\ \hline \text{Mean} = 49.78 \text{ mg U/cm}^3 \end{array}$$

$$s_m = 0.10\%$$

$$n_m = 3.$$

The approximately 0.01% difference between the mean atomic weight of natural uranium and that of isotopically pure U-238 was not taken into account, nor was the uncertainty in the isotopic analysis.

The relative standard deviation of the mean value of the uranium concentration, $s_{\bar{U}}$, can be found by combining the variance of the three coulometric measurements, $(s_m)^2$, with the variance for $\bar{Q}$, $(s_{\bar{Q}})^2$:

Table 4. Worst-case systematic errors in the coulometric determination of uranium in unknown solution.

Quantity	Approximate value	Estimated error	Estimated relative error (%)
Volume of original solution taken	0.5 cm ³	0.0005 cm ³	0.10
Flask volume	50 cm ³	0.05 cm ³	0.10
Coulometric aliquot volume	7 cm ³	0.005 cm ³	0.07
Net integrator readout	7 V	0.001 V	0.01
$\bar{Q}$, calibration factor	0.45 mg U/V		0.188 ^a
Sum of errors, Δ_U =			0.47%

^aSum of systematic and random errors.

$$\begin{aligned}
 s_{\bar{U}} &= \left[\frac{(s_m)^2}{n_m} + (s_{\bar{Q}})^2 \right]^{1/2} \\
 &= \left[\frac{(0.10)^2}{3} + (0.020)^2 \right]^{1/2} \\
 &= 0.061\%.
 \end{aligned} \tag{13}$$

SUMMARY AND CONCLUSIONS

The mean value of the three determinations and the 95% confidence limits are

$$\bar{U} = 49.78 \text{ mg U/cm}^3 \pm 0.26\%. \tag{14}$$

The confidence limits are computed from $\bar{U} \pm t_a s_{\bar{U}}$, taking $t_a = 4.3$ from the t-distribution table for $n_m = 3$. This expresses the effect of random error on the measured value of the uranium concentration.

The worst-case, or maximum expected, systematic error, which results from the factors in Table 4, is 0.47%. This is to be regarded as the worst possible error or bias; that is, it is virtually certain that the

true error does not exceed this value. If this error is intolerable, a new, smaller error limit could be formulated, but it would have a lower probability attached to it. Note that all the errors were assumed to be of the same sign, an unlikely but not impossible event. If any of the systematic errors are of different sign, they will tend to cancel and reduce the overall error.

In the chemical calibration and determination of uranium in the sample, the dominant systematic errors result from the uncertainties in the measurement of the solution volumes. This points out the value of using weighed-solution techniques, but this was not possible here. Use of the chemical and the electrical calibration values together also is apparently worthwhile, provided they agree fairly closely. Time is the principal measurement that could be refined in the determination of the electrical calibration value; this could be accomplished by the use of a gated, crystal-controlled counter/timer.

Combination of the electrical and chemical calibration values to obtain an estimate for use in the uranium measurement decreases the random error in $\bar{Q}$, compared to that obtained by using the chemical calibration value alone. This is shown by Eq. (11). This procedure also lends intuitive confidence to the results.

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