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SPECTROPHOTOMETRIC DETERMINATION OF PLUTONIUM-239 BASED ON THE SPECTRUM OF PLUTONIUM(III) CHLORIDE

by

D. J. Temer and L. F. Walker

ABSTRACT

This report describes a spectrophotometric method for determining plutonium-239 (Pu-239) based on the spectrum of Pu(III) chloride. We used the sealed-reflux technique for the dissolution of plutonium oxide with hydrochloric acid (HCl) and small amounts of nitric and hydrofluoric acids. To complex the fluoride, we added zirconium, and to reduce plutonium to Pu(III), we added ascorbic acid. We then adjusted the solution to a concentration of 2 M HCl and measured the absorbances at five wavelengths of the Pu(III) chloride spectrum. This spectrophotometric determination can also be applied to samples of plutonium metal dissolved in HCl.

1. INTRODUCTION

A method of spectrophotometric determination¹ was originally developed for measuring high-temperature, calcined plutonium-238 (Pu-238) oxide that had been dissolved, by the sealed-tube technique,² with hydrochloric acid (HCl) and a small amount of perchloric acid (HClO₄) or nitric acid (HNO₃). We applied spectrophotometry, as it is described in the current report, to Pu-239 oxide. After having been fired up to temperatures of 950°C, Pu-239 oxide was dissolved in HCl and small amounts of HNO₃ and hydrofluoric acid (HF) by the sealed-reflux technique.³ We also applied spectrophotometry to plutonium metal dissolved in HCl. Because it involves relatively few steps, the spectrophotometric determination has proved to be a simple and rapid method of measurement; moreover, it is also robust, accurate, and precise, and it produces minimal waste.

According to this method, the spectrophotometric measurement is made at a plutonium concentration that ranges between 2 and 4 mg/mL of solution. A sample of Pu-239 oxide weighing approximately 100 mg is dissolved for analysis. All the elements that are likely impurities in plutonium oxide samples have been considered in a separate interference study.⁴ Uranium can be tolerated at levels exceeding 100 mg/g of plutonium. Tolerated

without effect at levels of 1 wt% relative to the plutonium content are neptunium, americium, iron, thorium, gallium, and aluminum. Chromium, rhodium, palladium, neodymium, ruthenium, and gold are elements that interfere at 10 mg/g of plutonium. Two elements from the second group, ruthenium and gold, also interfere at levels of 1 mg/g of plutonium. Tolerances for anions are 0.01 M F⁻, 0.5 M SO₄⁼, and 0.05 M NO₃⁻. Phosphate and oxalate do not interfere at concentration levels of 1.6 mM.

The plutonium oxide samples we tested did not show any evidence of the elements mentioned above; they either did not contain such elements or the levels at which the elements were present were below the level at which interference is observed. The value for nitrate represents the equivalent amount of HNO₃ that is added for the dissolution of plutonium oxide. HNO₃ converts to various species in mixture with HCl during the heated reactions of dissolution. Refer to Section 7 for a consideration of these tolerances relative to the amounts of HF and HNO₃ used for the dissolution of plutonium oxide samples.

2. APPARATUS

2.1. Dissolution Apparatus

Used for the sealed-reflux dissolution of plutonium oxide, this apparatus incorporates a No. 18 semiball clamp, a polyethylene or rubber stopper, and a fused silica tube into which the plutonium oxide sample and acids are inserted (see Fig. 1 and Refs. 2 and 3).

2.2. Filtration Apparatus

For preparing the plutonium reference solution, we use an apparatus with a 2-in., .45- μ m hydrophilic filter (e.g., Millipore HVLP).

2.3. Spectrophotometer

The Hewlett Packard Model 8452 Diode Array Scanning Spectrophotometer, or equivalent instrument, is interfaced with a computer that has floppy-disk storage. At Los Alamos, we currently use a spectrophotometer that has been modified; it now incorporates a tungsten lamp and a fiber-optic interface manufactured by Custom Sensors and Technology.* The interface sits where a standard cell holder used to be placed. The fiber-optic cables of the interface—two 600-micron, single-strand, shielded cables with SMA connections—are connected to the cell that sits in a cell holder placed in the containment box. The fiber-optic interface allows the 80- μ L flow cell to be placed in the plutonium containment box, while the spectrophotometer remains outside the containment box.

*Custom Sensors and Technology, Inc., 7534 Watson Rd., St. Louis, MO 63119.

2.4. Transfer Device

Figure 2 shows a simple device we have built for transferring dissolved samples to a 25-mL volumetric flask.

2.5. Volumetric Flasks

We keep a large supply of class A, 25-mL, calibrated volumetric flasks.

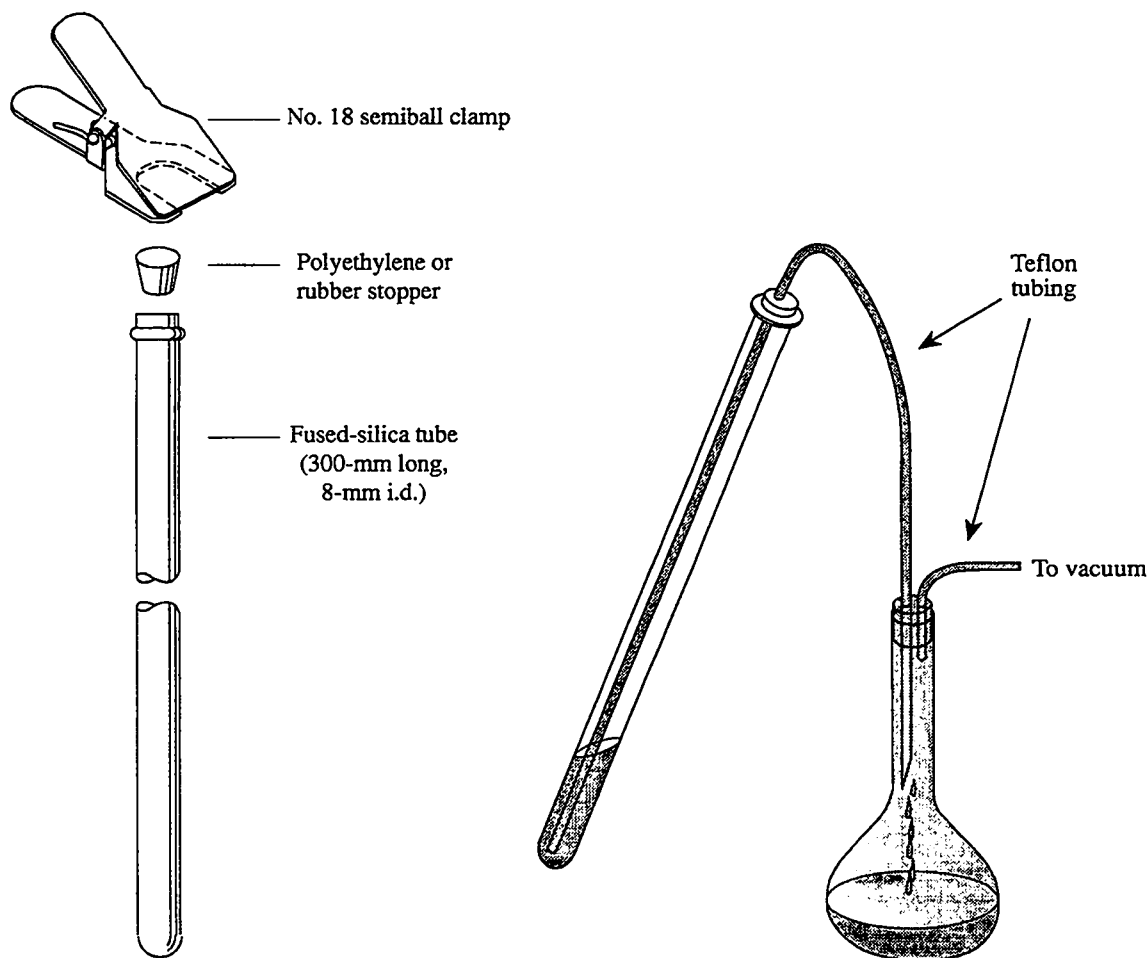


Fig. 1. The dissolution apparatus.

Fig. 2. The transfer device.

3. REAGENTS

Note: Distilled or deionized water must be used for all reagents.

3.1. Reducing Reagent

The reducing reagent we use is composed of ascorbic acid (L-, $C_6H_8O_6$) and aminoguanidine bicarbonate, each at a concentration of 0.40 M. We prepare this reagent

fresh daily by dissolving 7 g of ascorbic acid and 5.5 g of aminoguanidine bicarbonate in 100 mL of 1 M HCl. We then transfer the resulting reagent to a dispensing bottle with a 2-mL delivery.

3.2. Hydrochloric Acid (12 M)

This is an analytical reagent-grade acid, whose concentration is between 36.5% and 38.0%.

3.3. Hydrochloric Acid (1 M)

To obtain this reagent, we dilute to volume 83 mL of 12 M HCl with water in a 1-L volumetric flask.

3.4. Hydrofluoric Acid (1.3 M)

Using a plastic pipette, we transfer 4.8 mL of 48% 28 M HF to a 100-mL plastic, graduated cylinder and dilute to volume with water in order to obtain the desired reagent.

3.5. Nitric Acid (15.9 M)

This is an analytical reagent-grade acid, whose concentration is between 69% and 71%.

3.6. Plutonium Reference Solution

This solution contains 50 mg of plutonium per gram of 1 M HCl. To obtain the plutonium reference solution, we must strictly observe the following procedure:

3.6.1. Accurately weigh to 0.1-mg sensitivity approximately 6 g of high-purity plutonium metal, transfer to a tall-form beaker, and place a watch glass on top.

3.6.2. Add 20 mL of 6 M HCl through the beaker spout; after the plutonium metal dissolves completely, rinse the watch glass with 1 M HCl, delivered from a dispensing bottle, and collect the rinses in the beaker.

3.6.3. Filter the solution through a .45- μ m hydrophilic filter (we use a Millipore HVLP) and then completely transfer the solution to a polyethylene dispensing bottle previously tared to 0.1-mg sensitivity. Rinse the Millipore filtration apparatus thoroughly, at least three times, with 1 M HCl and combine the rinses with the solution in the polyethylene dispensing bottle.

3.6.4. Add 1 M HCl to a solution weight of 120 g, weigh to 0.1-mg sensitivity, and mix well. Currently, we use controlled potential coulometry (CPC) to standardize the reference solution of high-purity plutonium metal (>99.95%) against the New Brunswick Laboratory certified reference material No. 126 (NBL CRM 126)—plutonium metal standard.

3.6.5. Calculate the plutonium concentration of the solution by

$$C = 1,000 \text{ PMI} / (W_2 - W_1) ,$$

where C = plutonium concentration (milligram of plutonium per gram of solution);
 P = purity (weight fraction) of the plutonium metal reference solution;
 M = mass (gram) of the plutonium metal reference material;
 W_2 = weight (gram) of the polyethylene dispensing bottle and the solution;
and
 W_1 = tare (gram) of the polyethylene dispensing bottle.

3.6.6. While dispensing about 80 aliquants into individual glass vials, weigh each of them to 0.1-mg sensitivity—each aliquant contains approximately 60, 70, 80, 90, or 100 mg of plutonium—and then seal the vials to prevent contamination and evaporation. The reference solution should be completely dispensed the same day that it is prepared to ensure that any weight loss of the solution by transpiration is not significant.

3.6.7. Calculate the mass of plutonium in each aliquant by

$$M = C(W_B - W_A) ,$$

where M = mass (milligram) of the plutonium in an aliquant;
 C = plutonium concentration (milligram of plutonium per gram of solution; see previous computation);
 W_B = weight (gram) of the polyethylene dispensing bottle and the solution before an aliquant is dispensed; and
 W_A = weight (gram) of the polyethylene dispensing bottle and the solution after an aliquant has been dispensed.

3.7. Zirconium Solution (0.75 M)

This solution is obtained by dissolving 120.5 g of zirconyl chloride octahydrate ($ZrOCl_2 \cdot 8H_2O$) in a final volume of 500-mL 1 M HCl. The solution must be filtered if it contains particulate matter. Then, the solution is transferred to a dispensing bottle having a 2-mL delivery.

4. PROCEDURE

For the spectrophotometric measurement, we prepare one reagent blank, a set of calibration standards, and two dissolved weighed portions of each sample.

4.1. Reagent Blank

One reagent blank is processed with each series of reference material aliquants or group of samples. In a 25-mL volumetric flask, we mix 2 mL of 12 M HCl, 1 drop of 15.9 M HNO₃, and 1 drop of 1.3 M HF. We then proceed according to Steps 4.2.2 to 4.2.4 of the calibration procedure described below.

4.2. Calibration

Every other month, we observe the following procedure:

4.2.1. Process five dispensed aliquants of plutonium reference solution containing 60, 70, 80, 90, and 100 mg of plutonium. Quantitatively transfer each aliquant to a 25-mL volumetric flask, rinsing at least three times with 1 M HCl. Add 1 drop of 15.9 M HNO₃, 1 drop of 1.3 M HF, and 2 mL of 12 M HCl to simulate the sample conditions. Use a minimum amount of rinsing so that, during a 1- to 2-h waiting period, the plutonium will oxidize and resemble the oxide dissolutions used for the samples.

4.2.2. Add 2 mL of zirconium solution and mix. Zirconium(IV) is used to complex the fluoride. If zirconium is not added, the fluoride will prevent the complete reduction of plutonium to Pu(III). For uniformity, zirconium is added to all blanks, reference materials, quality controls, and samples, even when fluoride is absent.

4.2.3. Add 2 mL of reducing agent and make up to volume with 1 M HCl. Plutonium reduces to Pu(III), which shows in the characteristic blue color. The aminoguanidine bicarbonate* in the reducing reagent helps stabilize the ascorbic acid reductant.

4.2.4. Put the stopper on the volumetric flask and mix the solution by inverting the flask at least ten times. Gently lift the stopper after the first two and last inversions to relieve any pressure that may have been generated.

4.2.5. Measure absorbances at 516, 560, 586, 600, and 640 nm versus those of the reagent blank. Measurements should be completed within 4 hours so as to avoid the possible degradation of the reagents.

4.2.6. Compute the calibration coefficients for each of the four net absorbances ($A^{560} - A^{516}$, $A^{560} - A^{586}$, $A^{600} - A^{586}$, and $A^{600} - A^{640}$) by using data from the five standard aliquants. For each set of coefficients, compute a linear, least-squares fit of the net absorbances. Because standard and sample absorbances are measured against the absorbances of a reagent blank, no blank correction is needed.

*At Los Alamos National Laboratory, Peter DelMar performed experiments that proved the usefulness of aminoguanidine bicarbonate.

4.2.7. Using the linear coefficients found in the previous step, calculate the apparent plutonium concentration relative to the true plutonium concentration for each standard at each of the four net absorbances. Calculate a standard deviation of these data for each of the four net absorbances.

4.2.8. If the relative standard deviation (RSD) is greater than 0.2% at any of the four net absorbances, check for errant data or a possible outlier or, alternatively, obtain more calibration data before continuing.

We observe the following calibration procedure whenever we use the spectrophotometric method:

4.2.9. Along with the samples analyzed each day, process three aliquants of the plutonium reference solution containing 70, 80, and 90 mg of plutonium. (Follow Steps 4.2.2 through 4.2.8 described before.)

We use the latter calibration procedure to verify the more extended five-point calibration done every other month.

Note: The three-point calibration can only be run after a five-point calibration has previously been run. On the day the five-point calibration is run, there is no need for the additional three-point calibration.

4.3. Samples

We weigh two portions of each sample of Pu-239 oxide. Eight samples—two portions of each—can be analyzed daily.

4.3.1. Transfer approximately 100 mg of the sample to a pan and record the weight of the pan plus that of the sample.

4.3.2. Transfer the sample from the pan to a small sealed-reflux tube³ by tapping the tube while holding the pan firmly in contact with it.

4.3.3. Weigh the empty pan and compute the weight of the sample transferred to the tube by subtracting from the weight of the pan and the sample referred to in Step 4.3.1. Repeat Steps 4.3.1 through 4.3.3 to obtain a second portion of the same sample.

4.3.4. Add 2 mL of 12 M HCl, 1 drop of 15.9 M HNO₃, and 1 drop of 1.3 M HF (see Ref. 5).

4.3.5. Seal the tube and heat at 140°C for 2 hours or until the sample dissolves completely. If dissolution is not complete after overnight heating, evaluate the possibility that the undissolved material may not be plutonium and then continue with the procedure.

4.3.6. Using the vacuum transfer device, quantitatively transfer the solution to a 25-mL volumetric flask; rinse the tube at least four times with 1.0 M HCl and transfer the rinses to the same volumetric flask. The volume of the rinses must be kept minimal so that the total solution in the flask should not exceed 20 mL.

Once the solution has been transferred to the volumetric flask, follow Steps 4.2.2 through 4.2.5 of the calibration procedure.

5. CALCULATIONS

A computer program enables us to calculate, in two steps, the plutonium content of a sample.

5.1. First Set of Calculations

We calculate the plutonium content measured spectrophotometrically in the 25-mL volume for the four net absorbances (see Fig. 3) by

$$\begin{aligned} P_1 &= F_1 (S^{560} - S^{516}) , \\ P_2 &= F_2 (S^{560} - S^{586}) , \\ P_3 &= F_3 (S^{600} - S^{586}) , \text{ and} \\ P_4 &= F_4 (S^{600} - S^{640}) , \end{aligned}$$

where P_1 = plutonium content (milligram) in the 25-mL volume for the 560-nm peak minus the 516-nm baseline,
 P_2 = plutonium content (milligram) in the 25-mL volume for the 560-nm peak minus the 586-nm valley,
 P_3 = plutonium content (milligram) in the 25-mL volume for the 600-nm peak minus the 586-nm valley,
 P_4 = plutonium content (milligram) in the 25-mL volume for the 600-nm peak minus the 640-nm baseline,
 F_1 – F_4 = calibration coefficients at the subscript net absorbance, and
 S = absorbance of the sample at the superscript wavelength.

Next, we compute the average value \bar{P} and the standard deviation for values P_1 through P_4 .

5.2. Second Set of Calculations

We calculate the plutonium content of the sample by

$$V = 100 \bar{P} (N_S / N_R) / W ,$$

where V = plutonium content (weight percent) of the sample,
 \bar{P} = average value of the plutonium content (milligram) in the 25-mL volume,
 N_S = atomic weight of the plutonium in the sample,

N_R = atomic weight of the plutonium in the reference material of plutonium metal, and

W = weight (milligram) of the sample that was dissolved and transferred to the 25-mL volumetric flask for spectrophotometric analysis.

A baseline correction routine, which corrects for broad spectral interferences (sloping baseline), is automatically applied at 0.3% RSD.

6. RELIABILITY

For over 30 years, CPC^{6, 7} has been the Laboratory's method of choice for determining the total amount of plutonium in plutonium oxide samples. This well-documented method is robust, accurate, and precise, but the preparation of the sample, which is required for analysis, is labor intensive.

The spectrophotometric method, as described in this report, has been evaluated against CPC. A series of plutonium oxide samples were dissolved by the sealed-reflux technique, and aliquants from the same solution were analyzed both by CPC and the spectrophotometric method. The results of this comparison are presented in Table I.

Table I. Results of Comparison Between the Coulometric and Spectrophotometric Methods Used for Determining Pu-239

Sample Number	CPC % Pu	Spectrophotometry (S) %Pu	Ratio S/CPC
1	85.67	85.9	100.27
2	86.45	86.4	99.94
3	86.32	86.6	100.32
4	86.30	86.0	99.65
5	87.41	87.4	99.99
6	87.63	87.5	99.85
7	86.66	86.6	99.93
8	85.73	85.7	99.97
9	84.46	84.3	99.81
10	85.25	85.4	100.18

A comparison of the results obtained by using the two methods shows that the recovery is 99.99% (ratio of spectrophotometric result versus coulometric result) with a standard deviation of 0.21. Since 1987, we have run a control sample (high-purity plutonium oxide) both by the coulometric and Pu(III) spectrophotometric methods. The difference for the average plutonium value obtained by the two methods is 0.03% absolute, which once again indicates that the two methods yield the same results. Precision on typical samples is demonstrated in Table II, which shows the results of a study of 36 pairs of analyses on typical oxide samples obtained by calcining oxalate precipitate products; the average range for the 36 samples was 0.12% absolute.

Table II. Spectrophotometric Sample Data

Sample Number	Assay No. 1	Assay No. 2	Range	Sample Number	Assay No. 1	Assay No. 2	Range
1	87.75	87.94	0.19	19	87.39	87.43	0.04
2	87.92	88.05	0.13	20	86.90	86.82	0.08
3	87.87	87.93	0.06	21	87.26	87.34	0.08
4	87.70	87.69	0.01	22	87.75	87.55	0.20
5	87.94	87.93	0.01	23	87.31	87.26	0.05
6	86.91	86.74	0.17	24	87.49	87.43	0.06
7	83.04	83.29	0.25	25	83.34	83.50	0.16
8	74.47	74.69	0.22	26	87.49	87.46	0.03
9	87.84	87.77	0.07	27	87.69	87.86	0.17
10	87.78	87.62	0.16	28	86.36	86.54	0.18
11	87.81	87.61	0.20	29	87.12	87.17	0.05
12	87.32	87.09	0.23	30	87.37	87.42	0.05
13	87.76	87.49	0.27	31	86.87	86.93	0.06
14	86.50	86.15	0.35	32	87.22	87.27	0.05
15	87.14	87.05	0.09	33	87.11	87.09	0.02
16	87.44	87.37	0.07	34	85.44	85.09	0.35
17	87.08	87.07	0.01	35	86.91	86.81	0.10
18	87.42	87.36	0.06	36	86.88	86.96	0.08

7. CONCLUSIONS

The spectrophotometric method involves relatively few operations. Approximately 100 mg of plutonium oxide is dissolved by the sealed-reflux dissolution technique and quantitatively transferred to a 25-mL volumetric flask. Most samples will dissolve in 1 to 2 hours. However, samples that are fired at greater than 500°C may require approximately 14 hours of overnight heating for best results.

Some samples may contain residual silicates or nonplutonium material; generally, this does not present a problem because particulates either settle before the sample is drawn into the cell or are filtered out by the in-line glass-wool filter that is placed before the cell.

To complex the fluoride, we add zirconium, and to reduce plutonium to Pu(III), we add ascorbic acid. The solution is then diluted to a 25-mL volume with 1 M HCl, and the resulting solution has a concentration of 2 M HCl. Absorbances are measured at five wavelengths—516, 560, 586, 600, and 640 nm—with a computer-controlled photodiode array spectrophotometer having a 1-cm path-length flow-through cell and data storage on a floppy disk. A computer program is used for the calculation of results.

Figure 3 shows the spectrum of Pu(III) in 2 M HCl for the wavelength region of 510 to 650 nm. Four peak-minus-baseline and peak-minus-valley net absorbances (designated B - A, B - C, D - C, and D - E) provide four measures of the amount of plutonium in the sample that is analyzed.

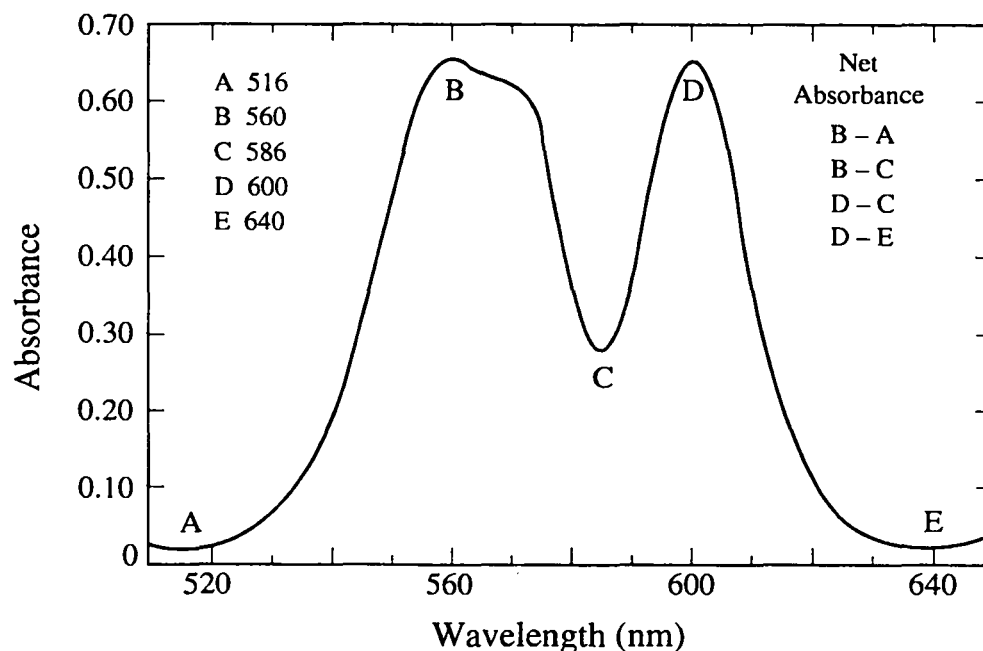


Fig. 3. Spectrum of Pu(III) in 2 M HCl.

Metallic elements that interfere have overlapping spectral absorbances at one or both of the two Pu(III) peaks (560 and 600 nm), at the valley (586 nm), and at one or both of the two baselines (516 and 640 nm). The tolerances given for americium, iron, gallium, neptunium, thorium, and uranium do not have a significant effect on any of the four peak-minus-baseline and peak-minus-valley net absorbances. Anionic interferences result from the complexation of higher oxidation states of plutonium, especially Pu(IV), to hinder the reduction to Pu(III) by ascorbic acid. Another type of anionic interference can be an oxidant, not reduced or only partially reduced by ascorbic acid, that catalyzes the oxidation of Pu(III). Fluoride, sulfate, and phosphate are anions that will complex the higher oxidation states of plutonium. Oxidants encountered in the spectrophotometric method are products resulting from the reactions of HNO₃ in mixtures with HCl during the sealed-reflux dissolution of plutonium oxide at elevated temperatures. The complexing interference of fluoride is decreased by the addition of zirconium. The solution that is measured spectrophotometrically contains 0.09 M zirconium and 2 M HCl; the zirconium in the solution will complex only up to 0.01 M F⁻.

The spectrophotometric measurement is made at a plutonium concentration ranging between 2 and 4 mg/mL of solution. As described in Ref. 5, 100-mg samples are accurately weighed and dissolved with 2 mL of 12 M HCl, 1 drop of 16 M HNO₃, and 1 drop of 1.3 M HF by using a small-sized sealed-reflux tube. The dissolved solution is transferred to a 25-mL volumetric flask, after which zirconium, ascorbic acid, and aminoguanidine are added; the solution is then diluted to 25 mL with 1.0 M HCl. The resulting fluoride and equivalent nitrate concentrations are 0.0026 M F⁻ and 0.03 M NO₃⁻, once again below the interfering levels of 0.01 M F⁻ and 0.05 M of equivalent nitrate. The disadvantages of using a smaller sample are a lower level of material representativeness and an increased weighing error.

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