USTUR 500: ELECTRODEPOSITION OF AMERICIUM, PLUTONIUM, THORIUM, AND URANIUM

Purpose	Electrodeposition of americium, plutonium, uranium, and thorium	**Method Number**	USTUR 500
Original Date	10/10/95	**Author**	USTUR Radiochemistry Staff
Revision Number	1	**Approved By**	Jim Elliston
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SAFETY NOTE: Before beginning this procedure, read all of the Material Safety Data Sheets for the chemicals listed in Sec. 3. Read Sec. 4.3 of the EM-9 Safety manual for information on personal protective clothing and equipment.

1. Principle of Method

1.1. Radionuclides isolated using the anion-exchange procedures for americium, plutonium, thorium, and uranium are electrodeposited from a sulfate electrolyte solution onto stainless-steel disks.

2. Apparatus

2.1. Stainless-steel disks: 1.59-cm (5/8-in.)-diameter disk, fabricated from 20-mil 316 stainless steel cold-rolled or No. 4 finish.

2.2. Coin holders.

2.3. Electrolytic cell: polypropylene cell body and splatter guard (Fig. 1).

2.4. Teflon holding block for electrolytic cell.

2.5. Electrodeposition apparatus: dc power supply to provide 550 mA of regulated current to each electrolytic cell.

2.6. Hot plate: Corning PC-500 or equivalent.

2.7. Pasteur pipettes.

2.8. pH electrode, e.g., Beckman electrode

2.9. Ammonia gas generator (Fig. 2).

2.10. pH meter.

2.11. Platinum electrodes.

3. Reagents

NOTE: All reagents have a shelf life of six months.

3.1. Thymol blue indicator. Mix 0.2 g of thymol blue powder with 21.5 mL of 0.02 M NaOH and dilute to 500 mL with nanopure H_2O.

3.2. Sulfuric acid (0.18 M). Mix 10 mL of concentrated sulfuric acid with 990 mL of nanopure H_2O.

3.3. Ammonium hydroxide (concentrated, reagent-grade).

3.4. Ammonium hydroxide (1.5 M). Mix 100 mL of concentrated ammonium hydroxide with 900 mL of nanopure H_2O.

3.5. Ammonia gas from ammonium hydroxide solution. Pass a stream of air through concentrated ammonium hydroxide.

3.6. Sulfuric acid (9 M). Add 500 mL of concentrated sulfuric acid to 500 mL of nanopure H_2O.

CAUTION: Solution gets very hot. Mix in a Pyrex beaker and set in pan of cold water while mixing. Allow solution to cool if prepared in volumetric glassware before final volume adjustment.

3.7. Acetone.

3.8. Nitric acid (8 M). Add 500 mL of concentrated nitric acid to 500 mL of nanopure H_2O.

3.9. Concentrated hydrogen peroxide (30%) H_2O_2.

4. Daily Calibration of Electrode

4.1. Supplies

4.1.1. pH electrode and meter.

4.1.2. Small Kimwipes.

4.1.3. Barnstead 18 megaohm water (nanopure H_2O).

4.1.4. Saturated KCl solution.

4.1.5. Parafilm

4.1.6. 1 M HCl.

4.1.7. pH 2 buffer.

4.1.8. pH 4 buffer.

4.1.9. Tweezers.

4.1.10. Disposable pipettes.

4.2. Electrode setup

4.2.1. Remove Parafilm from both covered areas of electrode.

4.2.2. Clean any dried KCl from electrode using nanopure H_2O, Kimwipes, and tweezers.

NOTE: Avoid getting any water into the electrode.

4.2.3. Refill electrode using saturated KCl solution.

4.2.4. Turn on pH meter and allow to warm up.

4.2.5. Remove electrode from KCl and rinse with nanopure H_2O.

4.2.6. Press the [auto read] button, then the [cal] button on the meter and place the electrode into a sample of the pH 2 buffer.

4.2.7. Once the meter has beeped, remove the electrode and rinse with nanopure H_2O.

4.2.8. Press the cal button and place the electrode into a sample of the pH 4 buffer.

4.2.9. Once the meter has beeped, remove the electrode and rinse with nanopure H_2O.

4.2.10. Place the electrode into a saturated KCl solution to prevent the tip from drying.

4.3. Electrode storage

4.3.1. Turn off the pH meter.

4.3.2. Place the electrode into saturated KCl solution.

4.3.3. Wrap junction and the electrode opening with Parafilm to prevent excessive evaporation.

5. Preparation of Sample Disks and Electrodeposition Cell

5.1. Engrave the sample number and identifiers and date into the sample disk.

5.2. Rinse stainless steel cap once with 8 M HNO_3, twice with nanopure H_2O, and once with acetone. Shake off excess and let dry.

NOTE: All acetone waste is to be caught in a beaker and placed in an appropriate non-radioactive storage container for disposal.

5.3. Remove cover from disk using fine tipped tweezers and gloves.

5.4. Assemble cell by placing the prenumbered disk (numbered side down) into the bottom depression of the cap.

5.5. Insert quad ring and press into place with tweezers or dental pick. Make sure the edge of the disk is covered by the quad ring and that the quad ring is seated fully beneath the threading of the cap.

5.6. Screw a new 30 mL polypropylene small mouth bottle into the stainless steel cap. Remove the end of the bottle with an appropriate cutting instrument.

5.7. Number each cell to prevent mishandling.

6. Electrodeposition

6.1. Add 1.0 mL of 9 M H_2SO_4 to the sample (from USTUR 200 - 6.3.14; USTUR 300- 6.4.15; USTUR 400 - 6.3.11) and heat until the H_2SO_4 fumes. Continue heating for an additional 10 min. Allow the sample to cool.

6.2. Place cell in Teflon holder to prevent tipping.

6.3. Add 3 mL of nanopure H_2O and 1 drop of thymol blue indicator. If it is a thorium sample, add one drop of conc. H_2O_2.

6.4. Transfer the solution to an electrolytic cell using a disposable pipette. Rinse the beaker twice with 3 mL of 0.18 M H_2SO_4. Add the rinses to the electrolytic cell. Insert new pasture pipette into tubing on ammonia gas generator (Fig. 2).

6.5. Adjust the solution in the deposition cell to slightly past the salmon pink end point (pH 2-2.3) using NH_3 gas. Use the Teflon cell holder during pH adjustment

to prevent tipping the cell over. Check the pH with pH electrode. If pH is between 2 and 2.3 continue. If pH exceeds 2.3, reduce with 9 M H_2SO_4 (by drops) and repeat.

NOTE: Be sure to use a new Pasteur pipette for each sample.

6.6. Rinse pH electrode with nanopure H_2O and blot with Kimwipe between each sample.

6.7. Place the cell in the electrodeposition rack and cover with a splatter guard. Lower the electrode into the cell and adjust so that the platinum wire is approximately 11 mm from the planchet.

6.8. Turn on the main power switch of the electrodeposition unit. Make sure each unit is set to 0.55 A. Attach the positive lead to the platinum wire electrode and the negative lead to the steel cap.

6.9. Electrodeposit plutonium, thorium, and uranium for 2 h, and americium for 2.5 h. Combinations of these elements shall be deposited for the time specified for the longest element.

6.10. Upon completion of the time required for electrodeposition, fill the cell with 1.5 M NH_3 and continue depositing for 1 min.

6.11. Disconnect the leads.

6.12. Remove the cell from the rack and discard contents into the radioactive waste container. Do not add to waste from columns because of reaction between HCl and NH_3.

6.13. Rinse deposition cell three times with nanopure H_2O.

6.14. Disassemble the cell, rinse disk with nanopure H_2O, and air dry.

6.15. Place disk in 200° C oven for 10 min.

6.16. Place the disk in a coin holder, label the holder with sample set number, and submit to count room for alpha spectrometric measurement.

7. Cleaning Equipment

NOTE: Proper cleaning of equipment is necessary to prevent cross-contamination of radioactive materials between samples.

7.1. Soak the platinum wire electrodes in 8 M HNO_3 for at least two hours and in nanopure H_2O for at least 2 hours.

7.2. Rinse the pH electrode with nanopure H_2O and blot dry with a Kimwipe between each sample. For samples known or expected to be highly radioactive (>100 dpm), rinse the electrode with 1 M HCl, then nanopure H_2O. For very high samples (>1000 dpm), soak the electrode for 1 hour in 1 M HCl and rinse with nanopure H_2O before the next sample.

8. Proper Waste Disposal Practices

8.1. All cell waste and first acid rinses shall be caught and stored in a properly labeled radioactive waste container.

8.2. Electrodeposition supernatant should be stored in waste jug and disposed of in the following manner.

8.2.1. Place waste into a beaker which pours well, and has at least 1000 mL of space per 900 mL of waste.

8.2.2. Add 5 ml conc. HNO_3 and 15 ml conc. H_3PO_4 per 900 mL of waste.

8.2.3. Add 2 ml of 1 g $Ca(NO_3)_2$/mL per 900 mL of waste while stirring.

8.2.4. Add stirring bar and stir/heat to 75°-80° C.

8.2.5. Add conc. NH_4OH slowly until precipitate forms.

8.2.6. Add an additional 10 mL conc. NH_4OH per 900 mL of waste.

8.2.7. Stir/heat for one hour at 75°-80° C.

8.2.8 Remove stir bar and allow sample to cool and settle.

8.2.9. Decant and filter to remove precipitate from liquid.

8.2.10. Discard supernatant into (hot) sink.

8.2.11. Discard precipitate into solid radioactive waste.

8.3. Retain the plates for five years and then discard in a solid radioactive waste container, attaching a list of samples discarded in container.

9. Source Materials

9.1. H.A. Boyd, B.C. Eutsler, J.F. McInroy, "Determination of Americium and Plutonium in Autopsy Tissue: Methods and Problems," in *Actinides in Man and Animals*, Proceedings of the Snowbird Actinide Workshop, Oct. 14-17, 1979, M.E. Wrenn, scientific editor (R.D. Press, Salt Lake City, Utah, 1981), pp. 43-52.

9.2. N.A. Talvitie, "Electrodeposition of Actinides for Alpha Spectrometric Determination," *Anal. Chem.* **44**, 280-283 (1972).

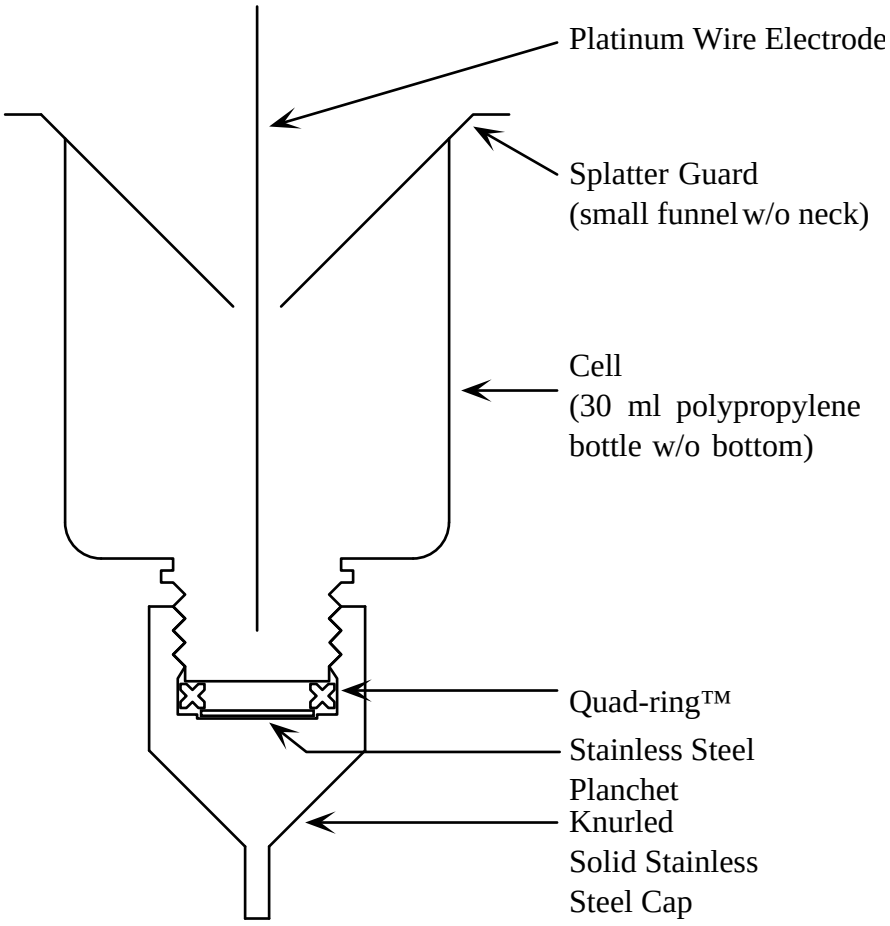

Fig. 1. Electrolytic cell.

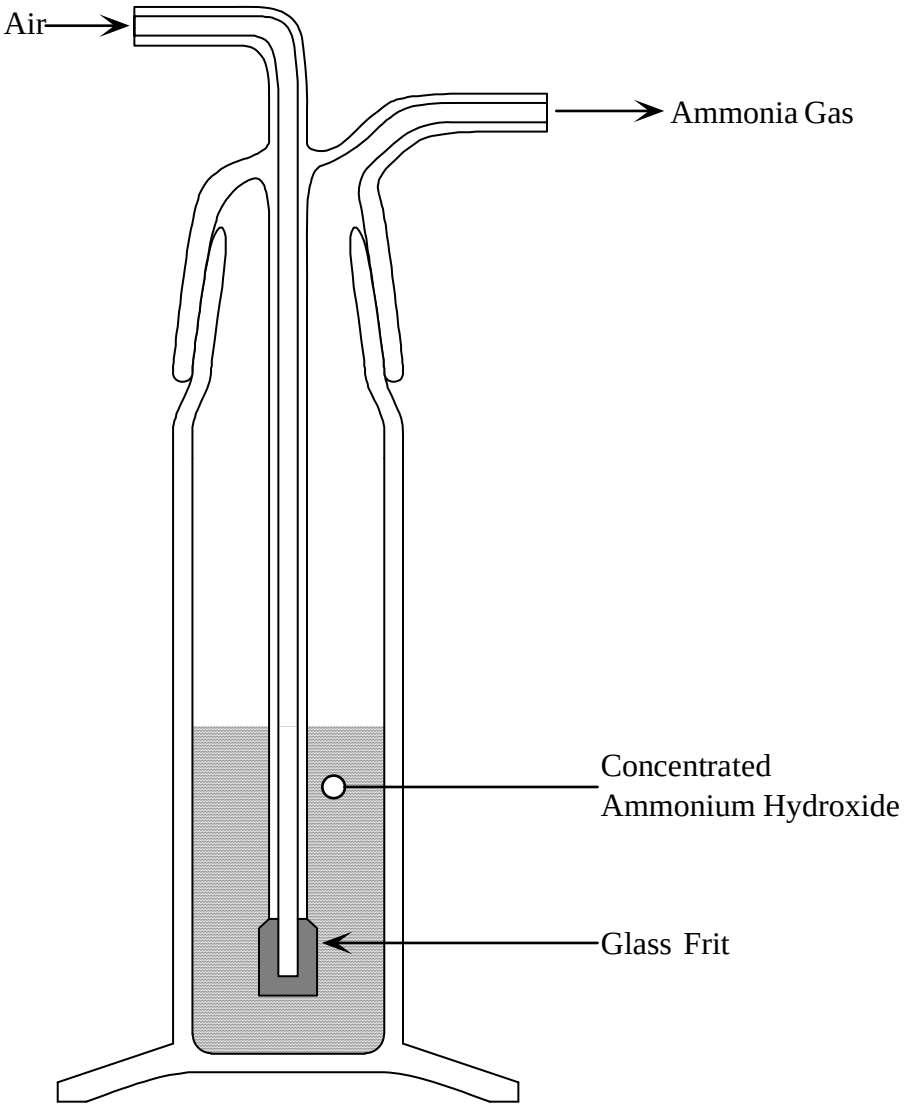

Figure 2. Ammonia Gas Generator.