Extraction of ^{229}Th from ^{233}U for Medical Research Applications

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Why ^{229}Th?

- **Parent of Decay Chain That Includes ^{213}Bi**
  - **Bismuth used for alpha-immunotherapy research**
- **Long-lived Daughter of ^{233}U**
  - **Half-life of ^{229}Th = 7,300 years**
- **ORNL Extracts ^{225}Ac from ^{229}Th**
  - **Half-life of ^{225}Ac = 10 days**
- **Medical Research Center Extracts ^{213}Bi from ^{225}Ac**
  - **Half-life of ^{213}Bi = 46 minutes**

Example Application

- **Treatment of Acute Myeloid Leukemia**
  - **Memorial Sloan-Kettering Cancer Center, New York**

- **Outline of Protocol**
  - **Receptor: CD33 surface protein**
  - **Antibody: HuM195**
  - **Linker: DPTA**
  - **Cytotoxic agent: ^{213}Bi**

- **Proceeding Through Human Trials**
  - **Phase 1 trials completed in 1999**
  - **Phase 1/2 trials on modified protocol beginning in 2001**

Current Capability and Need

- **Previous Separation of 90 mCi ^{229}Th**
  - **"X-40" material left over from ^{233}U processing in 1970s**
- **Can Provide Single Shipments of 35 mCi ^{225}Ac**
  - **Will ship over 420 mCi in FY-2001**
  - **All significant FY-2001 batches are claimed**
- **Cannot Meet Request for Shipment of 50 mCi ^{225}Ac**
  - **Hard to accommodate new small users**

^{233}U Inventory at RDF

- **Inventory**
  - 446 kg of ^{233}U
  - Over 1000 packages
  - ^{232}U contamination ranges from 2-6 ppm to almost 200 ppm
- **Storage Concerns**
  - Material has been in static storage up to 35 years
  - DNFSB Recommendation 97-1
- **Inspection and Repackaging Project**
  - Evaluate condition of existing packaging

Mound ^{233}U Material

- **Well-characterized material**
  - **3.4 kg ^{233}U with 72 mCi ^{229}Th**
  - **Put in storage in 1996**
  - **One package removed in 1998**

- **Obtained Approval to Remove From Storage Prior to Inspection Project**

Process Block Diagram

- **John Keller's block diagram**

Glove Box Used for 2 ppm ^{232}U Material

- **Photo of Scott at the 2026 glove box**

Incoming Uranium Oxide

- Photo of klinker on a spoon; or photo of material in beaker

Separation Column

- Photo of column and flask, or photo showing green band

Recovered Uranium Oxide

- Photo of good oxide on scoop

Thorium Separation Results

- **Thorium Recovered from Four Batches of ^{233}U**
  - **Containing 2 ppm ^{232}U**

  - **RAML-01 2.7 mCi**
  - **RMAL-02 2.4 mCi**
  - **RMAL-01-01 3.1 mCi**
  - **RMAL-01-02 7.0 mCi**

- **Total of 15 mCi ^{229}Th**

Operating Experience

- Column Operation Nearly Quantitative
- Mound ^{233}U Contains Significant Plutonium Contamination
- Plutonium Carries Into ^{229}Th Product
- Some Residual Uranium Carried In ^{229}Th Product

Typical Composition of Recovered ^{229}Th

• Data in mg	RMAL-01	RMAL-02
– ^{229}Th	12.5	11.2
– Total Thorium	13.9	20.0
– Total Uranium	64.3	38.9
– Total Plutonium	18.7	92.5

RMAL Hot Cells

- Photo of RMAL hot cells

Preparations for ^{233}U with 6 ppm ^{232}U

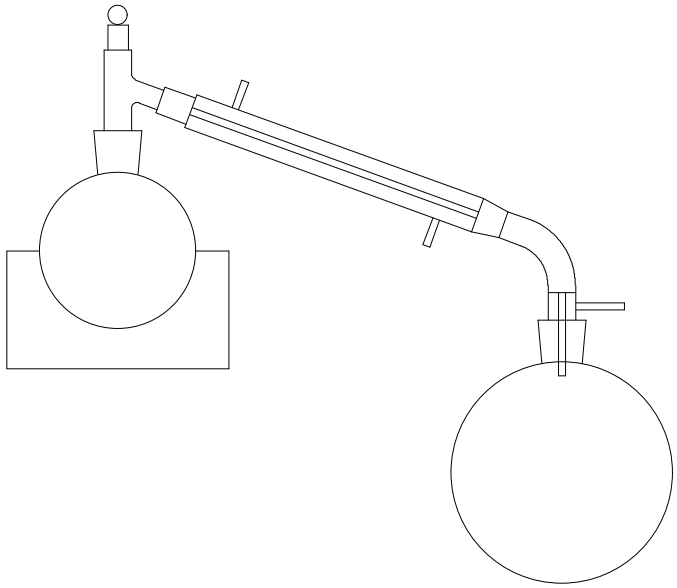
- **Remote Operation in Hot Cells**
  - **Hot cell preparation nearly complete**
  - **Possible continuation of uranium conversion in glove box**
- **Review Selection of Resin**
  - **Now using Dowex**
  - **Evaluating MP-1**
- **Replace Filtration with Centrifuge**

Thorium Purification Glove Box Installation

- Photo of Miting at the glove box

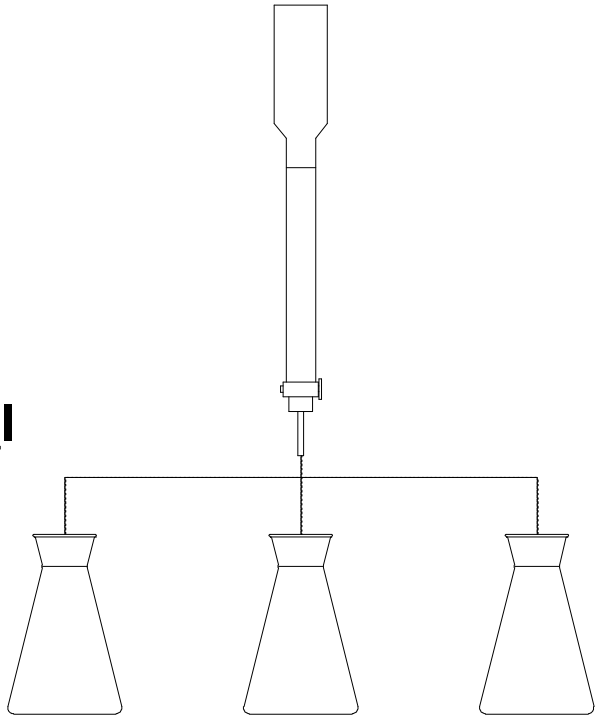
Thorium Purification Process

- **Adjust Plutonium Valence and Prepare Column Feed**
  - **Adjust solution to 0.5 M HNO_3**
  - **Adjust plutonium valence with Na_2SO_3 and $NaNO_2$**
  - **Concentrate to 10 M HNO_3**



Thorium Purification Process, cont.

- **Extraction of Thorium from Uranium and Plutonium**
  - Add feed to 10 ml of MP-1 resin
  - Wash uranium with 10 M HNO_3
  - Elute thorium with 10 M HCl
  - Elute plutonium with 10 M HCl and NH_4I



- **Continue Removal of Trace Pu**
  - Add feed to 2 ml of MP-1
  - Wash thorium with 10 M HCl
  - Complete when U and Pu are not detected

Thorium Inventory in Alpha Cell

- **Photo of alpha cell**

Preparation of New ^{225}Ac Cow

- **Begin With Small Batch of "X-40" Material**
  - Waste material with high stable ^{232}Th content
  - Demonstrate successful radium/actinium extraction
- **Add Increments of Newly-Separated Thorium**
  - Evaluate extraction process performance
  - Evaluate thorium losses as ^{232}Th content falls
- **Add Other Material Separated in Earlier Campaigns**
  - Bring new ^{229}Th cow up to 25 mCi
  - Add 10 mCi ^{225}Ac production capability
  - Bring single ^{225}Ac source capability up to 45 mCi

Improvements to RDL Facilities

- **New Glove Box Laboratory**
  - **Used for actinium extraction and thorium purification**
  - **Allows handling of larger radium/actinium batches**

- **Need to Address Growing Radiation Levels**
  - **Local shielding for concentrated source terms**
  - **Automation of process elements**
  - **Tongs/manipulators for semi-remote operation**

  - **Combine cleanup process with thorium separation in RMAL hot cells**

Future Plans

- **Continue Separation of ^{229}Th from Mound ^{233}U**
  - **Additional 55 mCi of ^{229}Th from Mound ^{233}U**
- **Process Other ^{233}U as Inspection Project Proceeds**
  - **Near-term potential for over 100 mCi of ^{229}Th**
- **DOE-ORO Request for Proposals for Disposition of ^{233}U Inventory at RDF**
  - **Thorium separation, lease**
  - **Continue ORNL operation as bridge?**