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Aqueous Chloride Operations Overview: Plutonium and Americium Purification/Recovery

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Aqueous (Chloride) Operations: The Basics

We take things like *this*

Salt mixture, variable Pu content

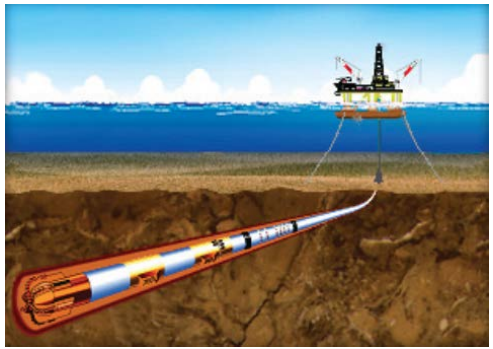
Purify it to *this*

Pu/Am Oxalate, high Pu/Am purity

And convert it to *this*

Pu/Am Oxide, high Pu/Am purity, highly stable/storable

Americium neutron source used for oil and gas exploration



So it can be used to do things like *this*

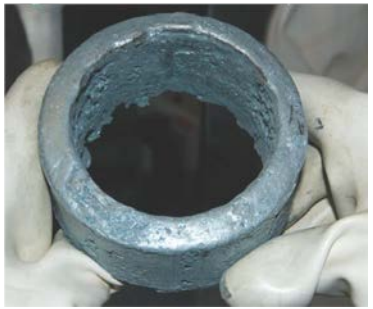
Aqueous Chloride Mission

- **Recover plutonium and americium from pyrochemical residues (undesirable form for utilization and storage) and generate plutonium oxide and americium oxide**
  - Plutonium oxide is recycled into Pu metal production flowsheet
  - Plutonium oxide is suitable for storage
  - Americium oxide is a valuable product, sold through the DOE-OS isotope sales program

What are pyrochemical residues? Pyrochemistry operations?

Pyrochemistry is…

- An operation that will be described in detail by Matt Jackson



- An operation that converts fairly pure Pu feed (oxide) into VERY pure Pu metal

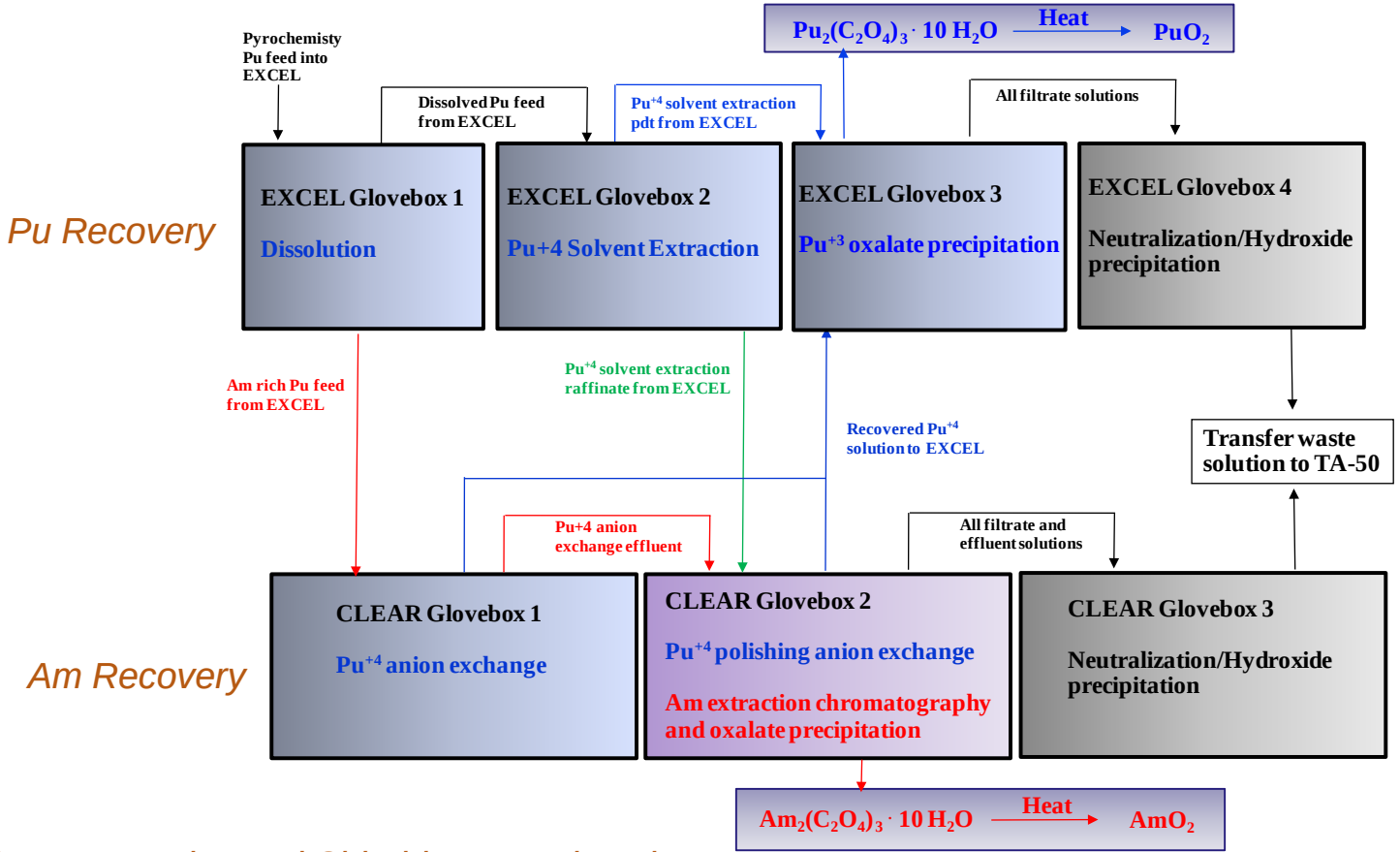
- An operation that generates residues that are composed of a lot of salt (mostly Cl-) and a little Pu

Can’t We Just Dispose of the Residues?

- **Sure! IF we had an *unlimited* supply of the following:**
  - Waste drums
  - Waste drum storage space (WIPP)
  - $$
- **LANL has a lot of pyrochemical residues**
  - Decades of past residues stored
  - New residues generated by current operations
- **Waste drums have limits on Pu content (or fissile material equivalent)**
  - More Pu = More waste drums = More waste storage space/$$
- **Consolidation of Pu = recovery/reuse of Pu, more waste that can go into a drum, less storage space needed**

Aqueous chloride operations recover the Pu (and Am) from lower Pu content residues

Aqueous Chloride and Americium Flowsheet



EXCEL = Experimental Chloride Extraction Line

CLEAR = Chloride Extraction and Actinide Recovery Line

First, Manage the Feed: Size Reduction

Block of feed (simulant), harder to remove Pu

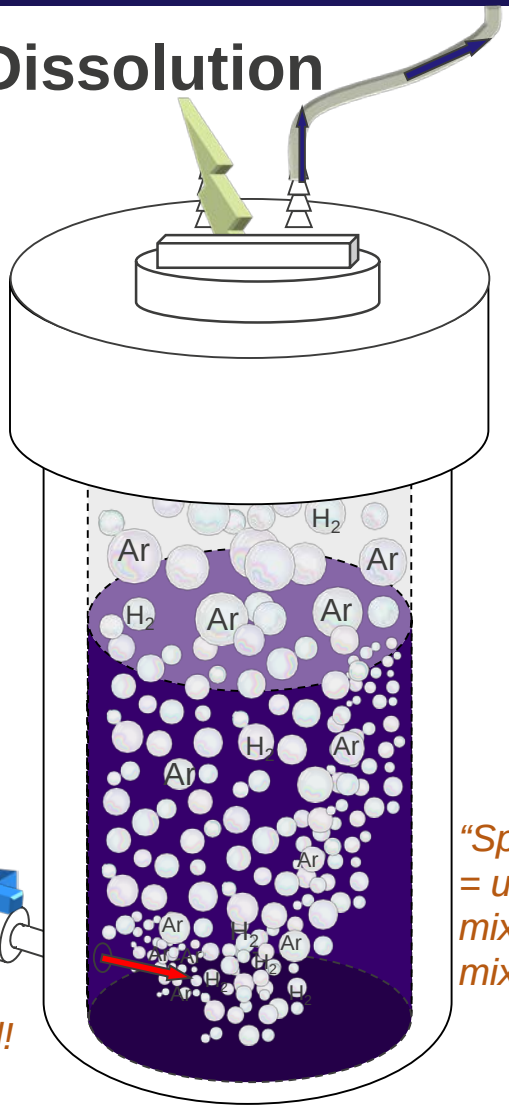
Crushed feed, plenty of surface area, ready for dissolution

Process Begins: Dissolution

Argon gas

12M HCl

Water

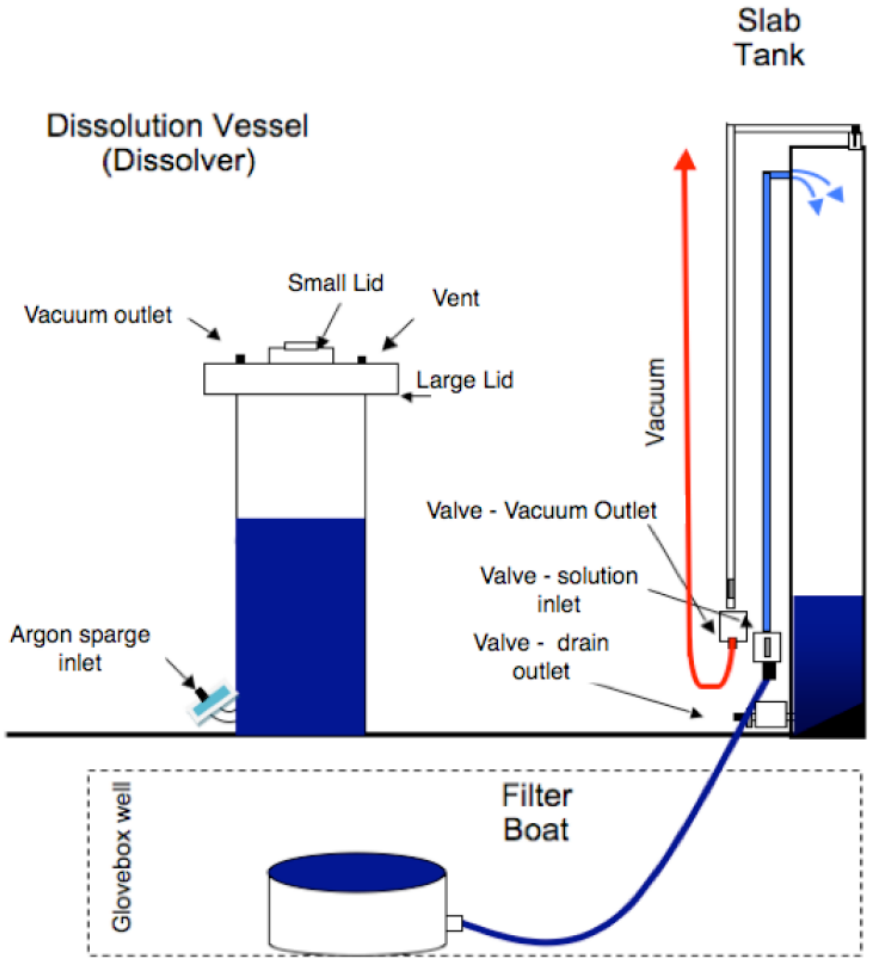


No heating required!

"Sparging" = using gas to mix/agitate the mixture

Looks like plastic? Kynar lined. Why?

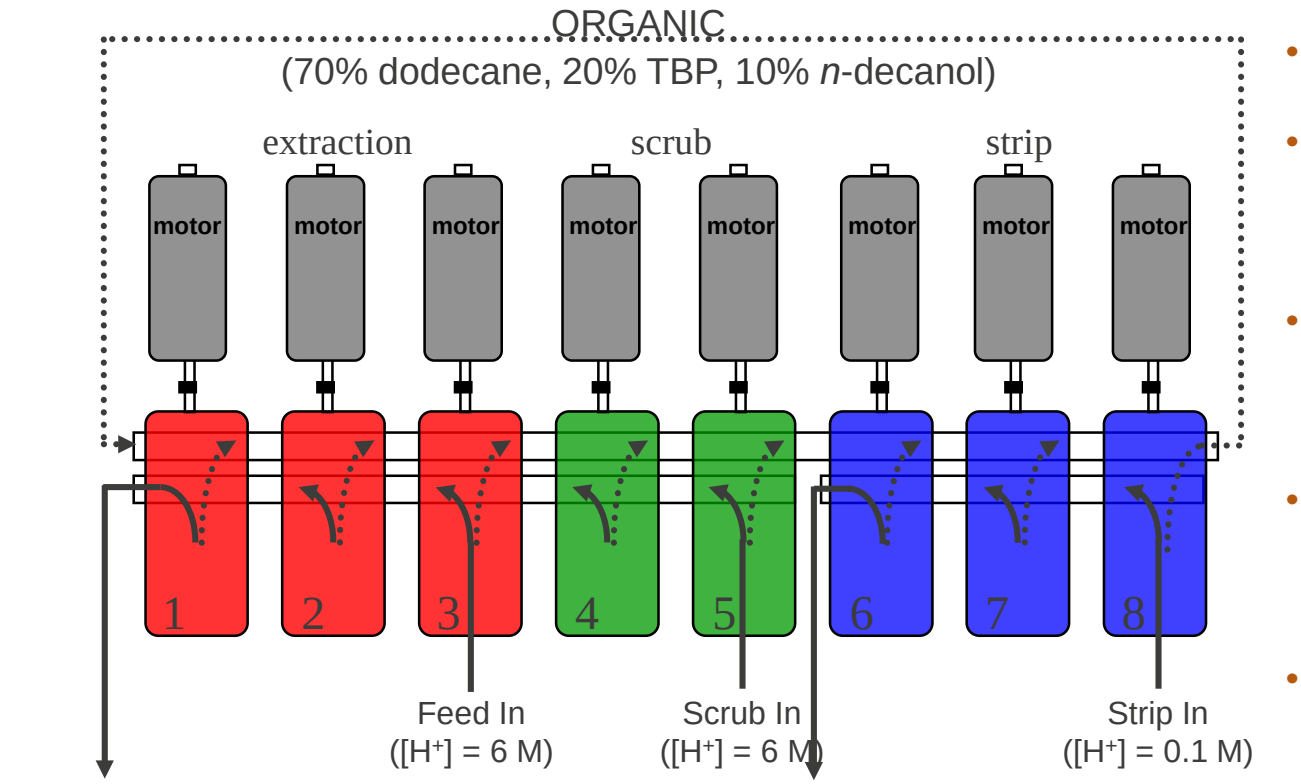
Dissolution, Continued…



Vacuum filter the dissolved material into a Kynar slab tank

Slab tank is "skinny" (~4" thick). Why? So those neutrons can leak!

Begin Purification: Solvent Extraction



"Raffinate" – remember this for later

Legend
aqueous
organic

- *Solvent Extraction: a liquid-liquid separation method*
- *Pu(+4) is pulled into the organic phase from the higher acid concentration (~6 M), aqueous phase*
- *Organic phase, containing the desired Pu, is washed, again with higher acid concentration* aqueous solution
- *Pu is removed from the organic phase with lower acid concentration (~0.5 M) aqueous solution.*
- *"Contactors" are specially designed chambers that mix and separate the phases, allowing separation to be "automated", with continuous flow.*

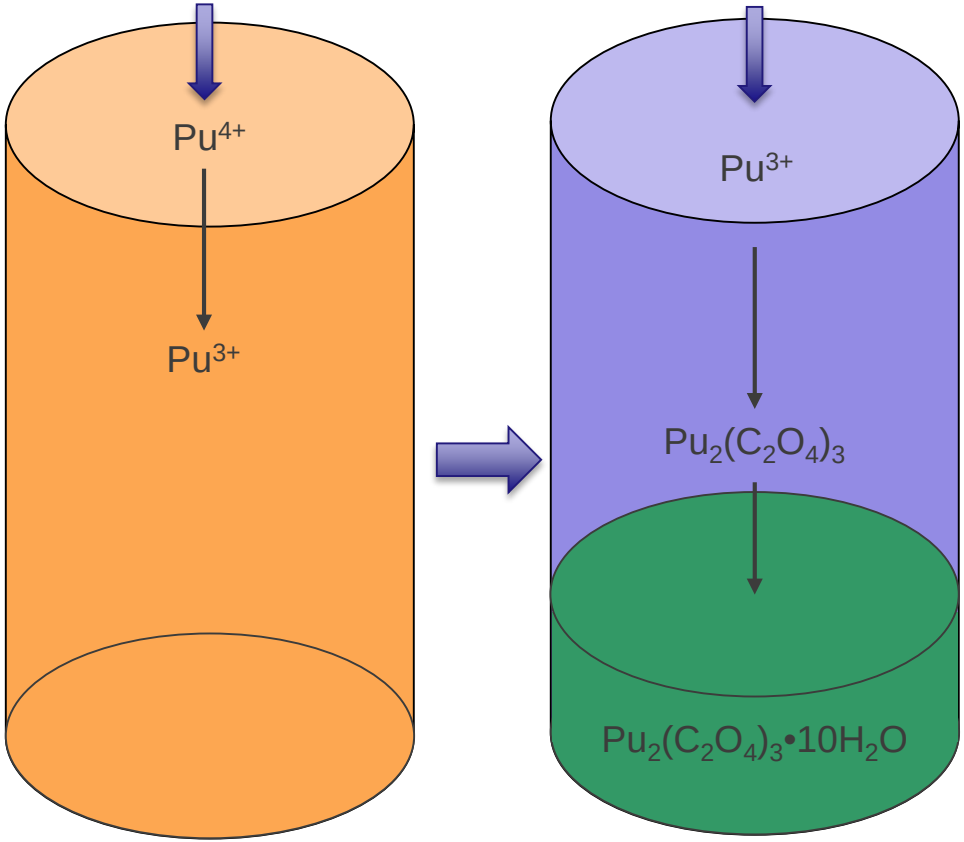
Purification, Continued: Oxalate Precipitation

$NH_2OH•HCl$

(hydroxylamine hydrochloride)

$H_2C_2O_4•2H_2O$

(oxalic acid)

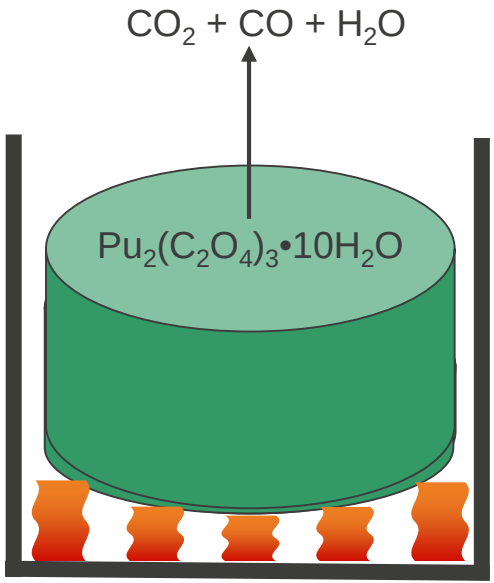


Precipitation, Filtration

Pu^{3+} makes a MUCH nicer oxalate precipitate than Pu^{4+}

Endgame: Calcination

Purified oxalate cake



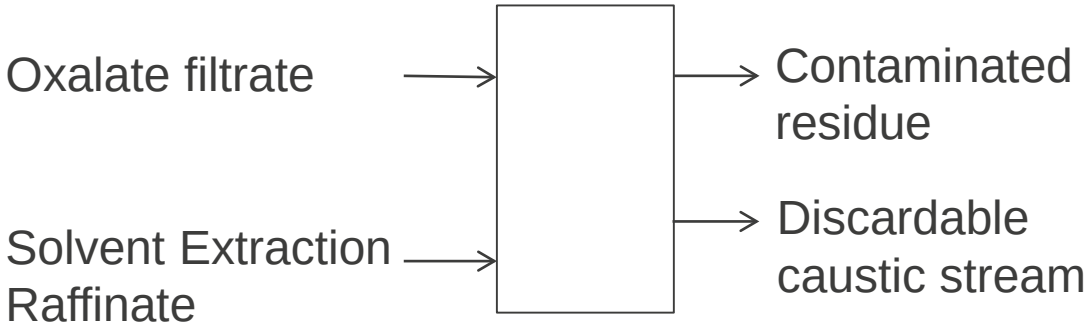
Oxidize = heat in an oven at high temp (aka "calcination")

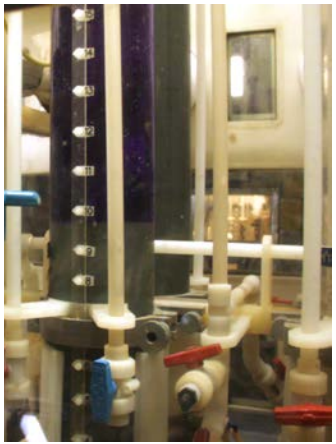
Purified, stable Pu oxide product

Pu oxide packaged for storage, use

What About the Waste Products? Hydroxide Precipitation

- **Feed: oxalate filtrate, solvent extraction raffinate***
  - *more on raffinate for americium recovery to come…
- **Product: contaminated residue, neutralized caustic solution that meets the discard criteria (to TA-50) requirements**





Raffinate = Higher acid molarity aqueous solution that was "washed" (Pu removed) by the organic phase in solvent extraction
…BUT, americium is here too (Am is not removed by the organic phase)
Important? You bet!

BOTTOM LINE: the bulk of the waste is now VERY low Pu level = fewer waste drums needed

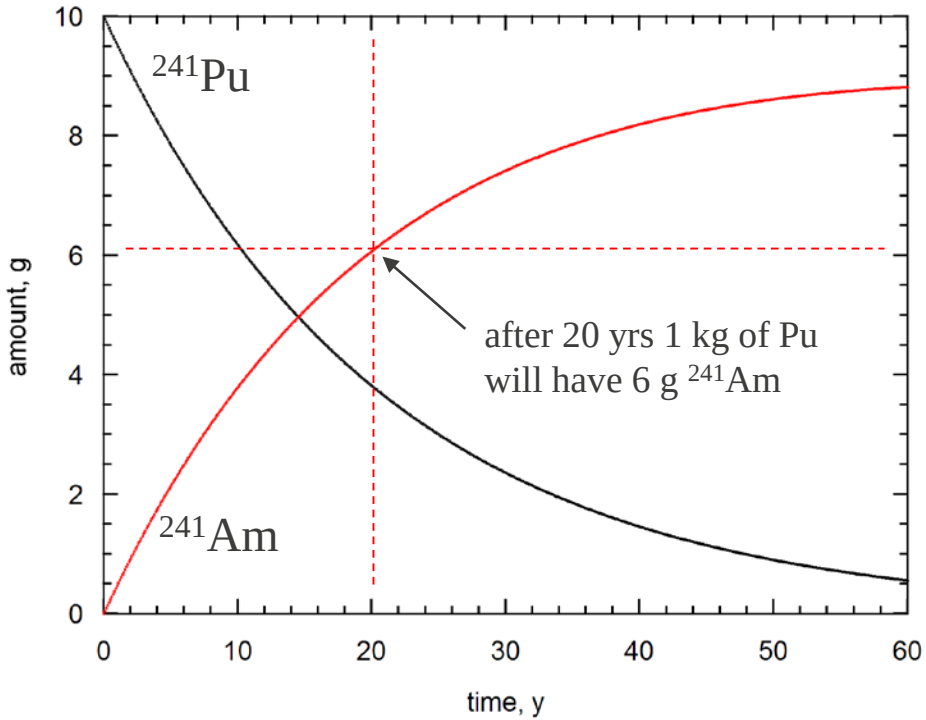
Didn't We Mention Americium Recovery?

- **Yes!**
- **Where does it come from? From the plutonium**
  - ^{241}Pu decays to ^{241}Am over time (always a little ^{241}Pu in ^{239}Pu supply)
- **Where do we have it now?**
  - OLD pyrochemistry residues
    - = Raffinate
    - …or Pu anion exchange

$$^{241}Pu \xrightarrow{t_{1/2}=14.4\,years} {}^{241}Am + \beta^-$$

$$^{241}Am \xrightarrow{t_{1/2}=432\,years} {}^{237}Np + \alpha + \gamma(59.5keV)$$

Dose!

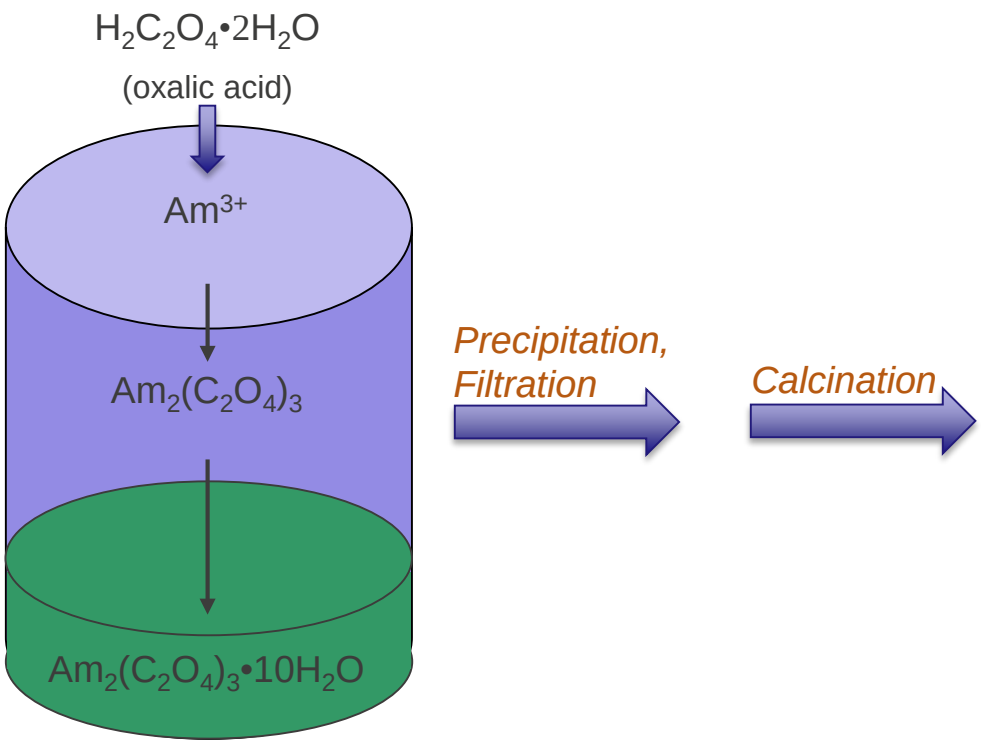


Americium Recovery: CLEAR Line

- **CLEAR: Chloride Extraction and Actinide Recovery Line**
- **Key methods: Pu anion exchange and extraction chromatography**
  - Pu and Am are separated on resin columns
  - Pu and Am “stick” to different solid “resin” material, while waste products do not (flow right through)
  - Recover each separately

Americium Recovery: Oxalate Precipitation and Calcination

- **Americium is precipitated similarly to Pu (easier, no reduction needed!)**
- **Calcination of americium is also analogous to Pu calcination (just a different furnace)**



Americium calcination is a MUCH smaller scale operation than Pu calcination. Why?
1. DOSE 2. Am is generated slowly (^{241}Pu decay)

LANL in the News: Americium Recovery

Physics Today 2015, 68(10), 20.

issues and events

Isotope program expands activities

To meet the growing demand for isotopes and lessen dependence on foreign sources, the Department of Energy has been realigning its capabilities and connecting with universities.

The US isotope program gets a healthy dose of kudos in a recent report by the Nuclear Science Advisory Committee Isotopes Subcommittee (NSACI). A key change in the program since it moved six years ago within DOE from the Office of Nuclear Energy to the nuclear physics program in the Office of Science is that it now has a research component. The report, *Meeting Isotope Needs and Capturing Opportuni-* tion. The program's mission includes developing new and better ways to produce more types and larger quantities of isotopes. Another goal is to train young scientists to do R&D on isotopes.

Filling a gap

At DOE national laboratories, isotope production is largely parasitic on machines that have other missions. The isotope program oversees production at

plete
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"Russia is the only supplier of americium-241, which is used in well logging to identify sites for drilling oil and gas. An industry consortium 'has provided the isotope program with money to develop a method of producing americium-241 here [in the US]. We're developing it at Los Alamos.'"

Questions?

“What’s this thing?”