Nuclear Fuel Reprocessing

By

Daniel Bolgren

Jeff Menees

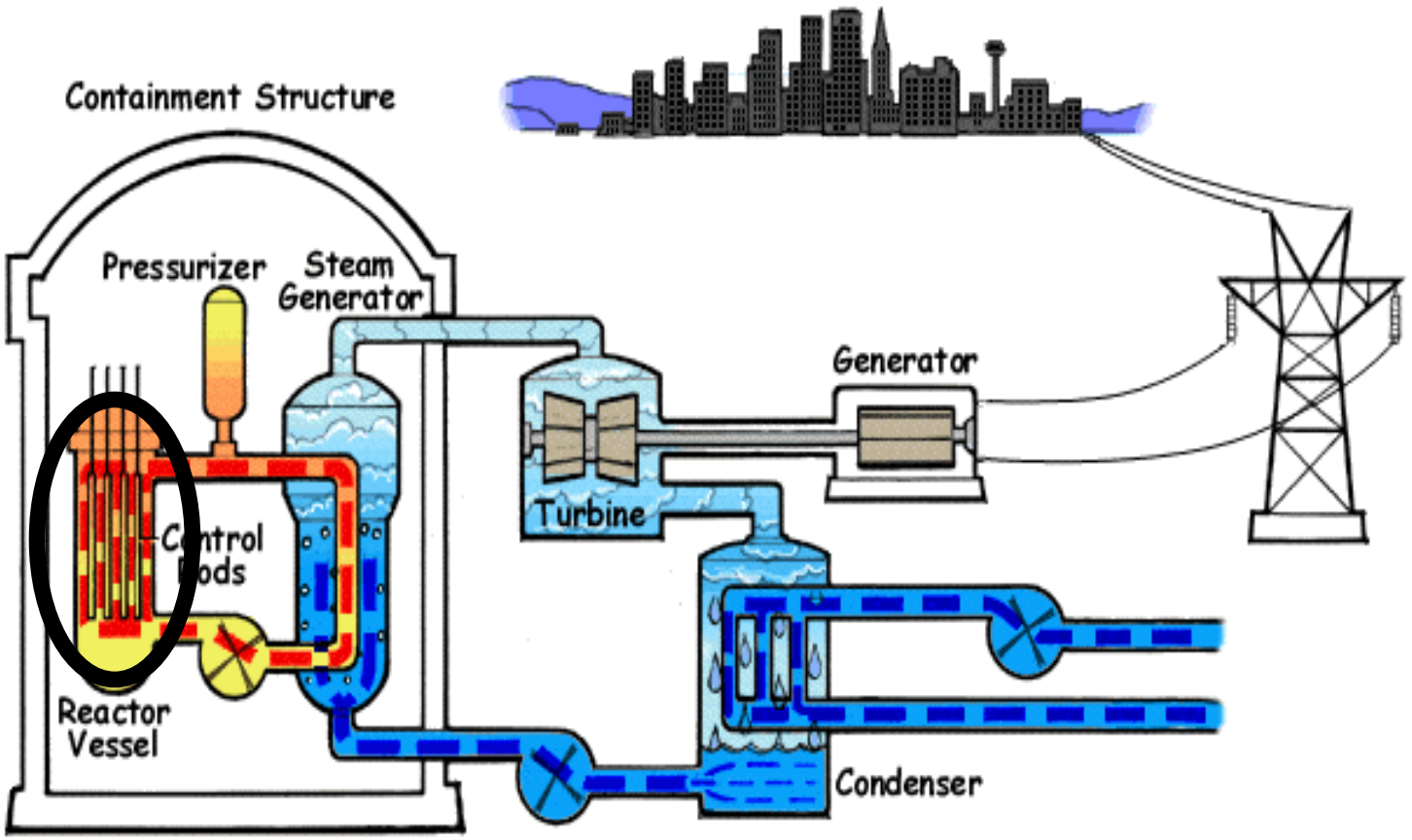
Goals of the Project

1. Develop a reprocessing technique that can:
    1. Reprocess used nuclear fuel.
    2. Reduce proliferation concerns.
2. Optimize a reprocessing location using:
    1. Current storage location.
    2. Transportation feasibility.

Overview

- Briefly explain of Nuclear Fission
- Background of Nuclear Fuel Reprocessing
- Nuclear Fuel Cycle
- Alternative Reprocessing Technique
  - Crown Ether Extraction Process
- Proposed Reprocessing Facility
  - Location optimization
  - Transportation feasibility
- Long Term Storage
  - Yucca Mountain

Nuclear Energy



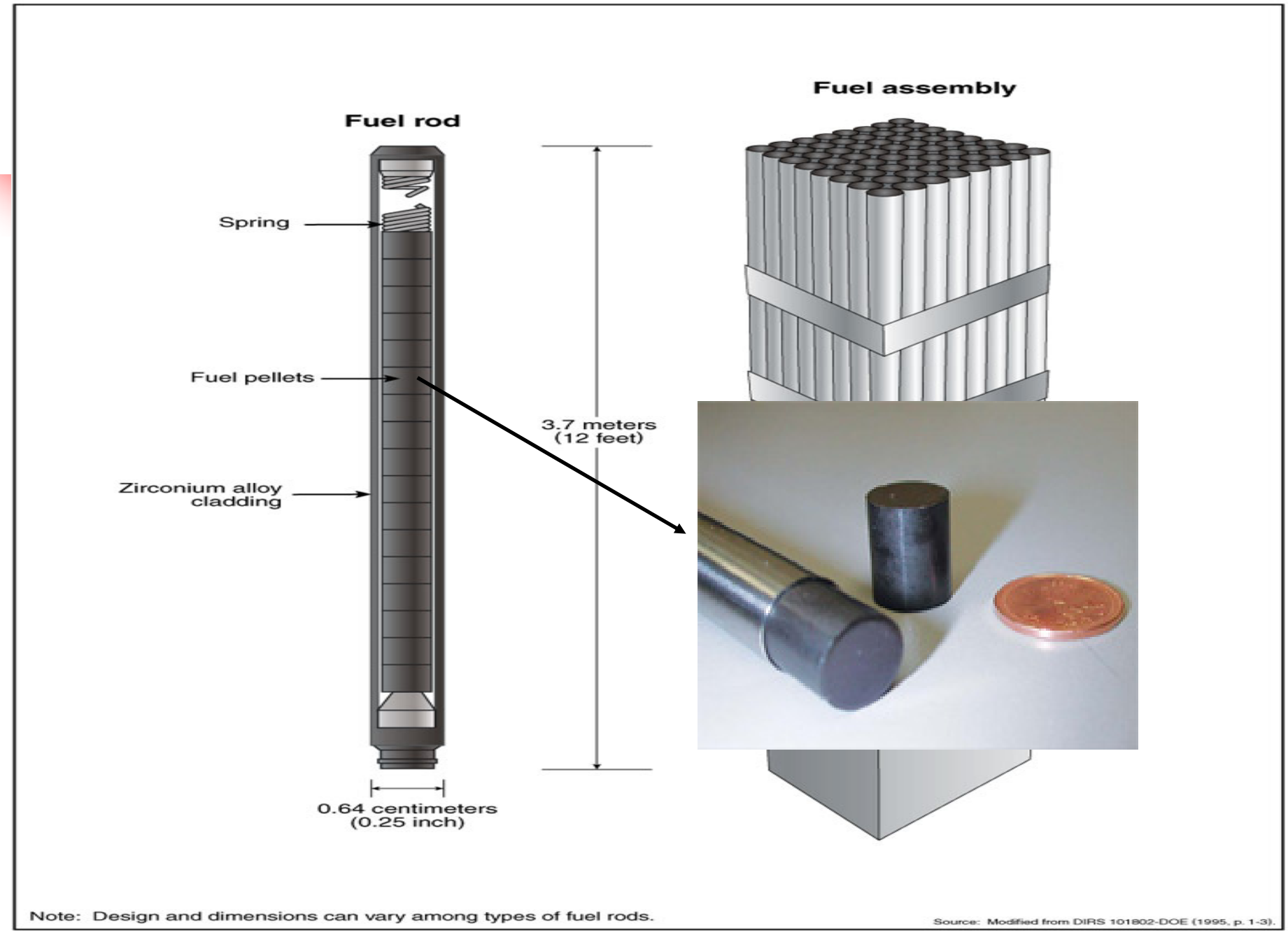
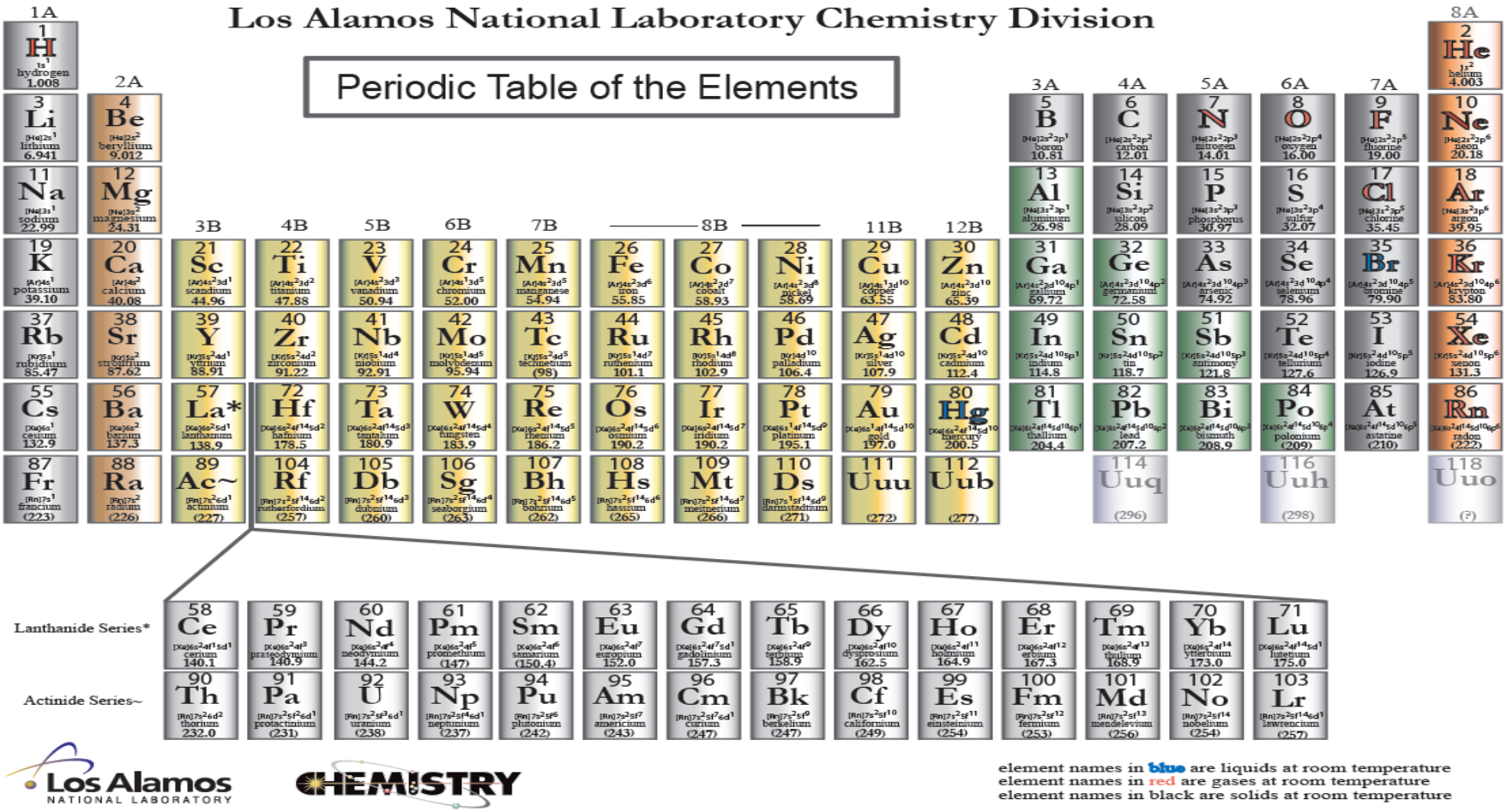


Figure 1-2. Typical commercial nuclear fuel assembly and rod.

Nuclear Chain Reaction?

Fission Products



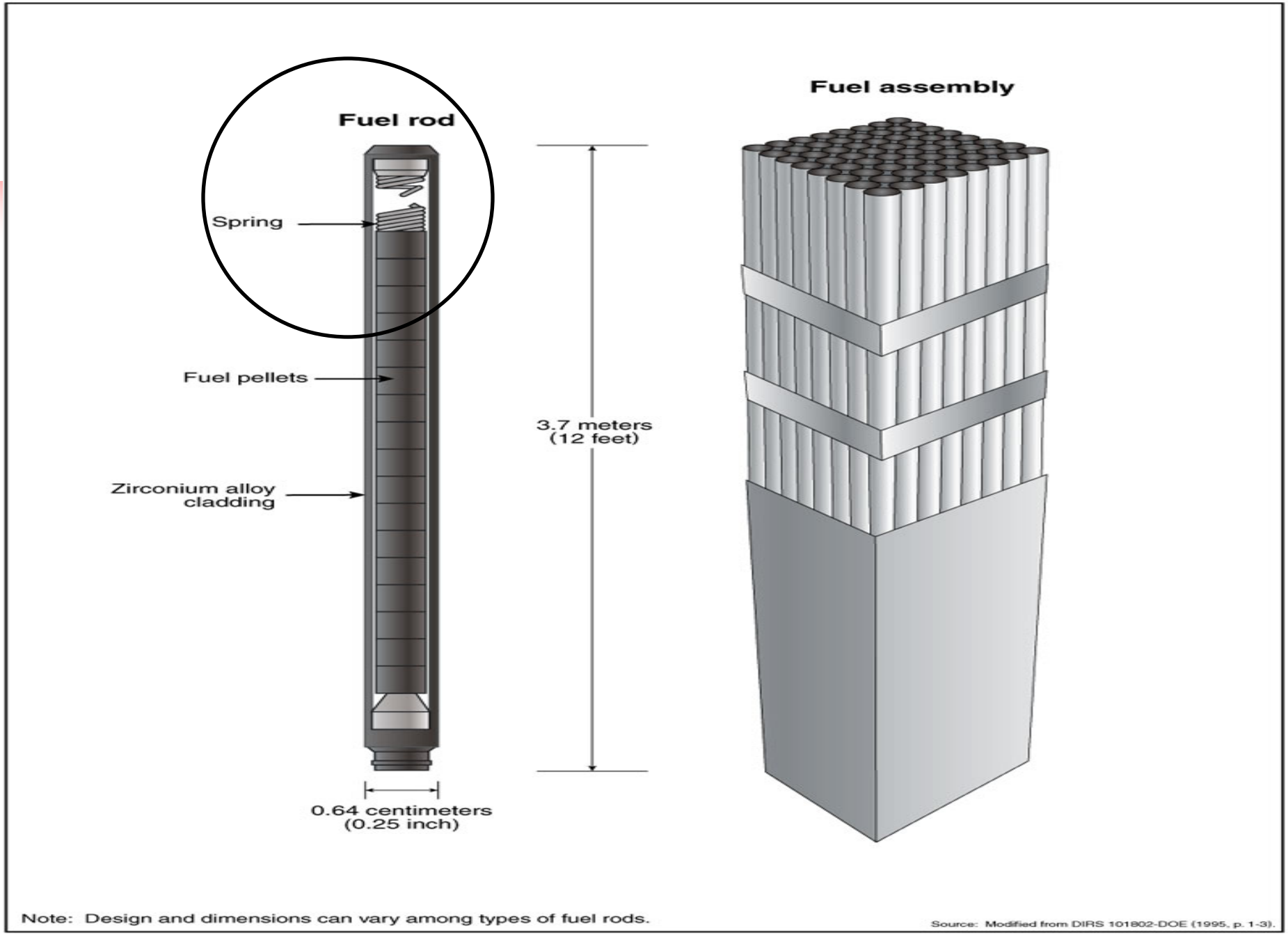
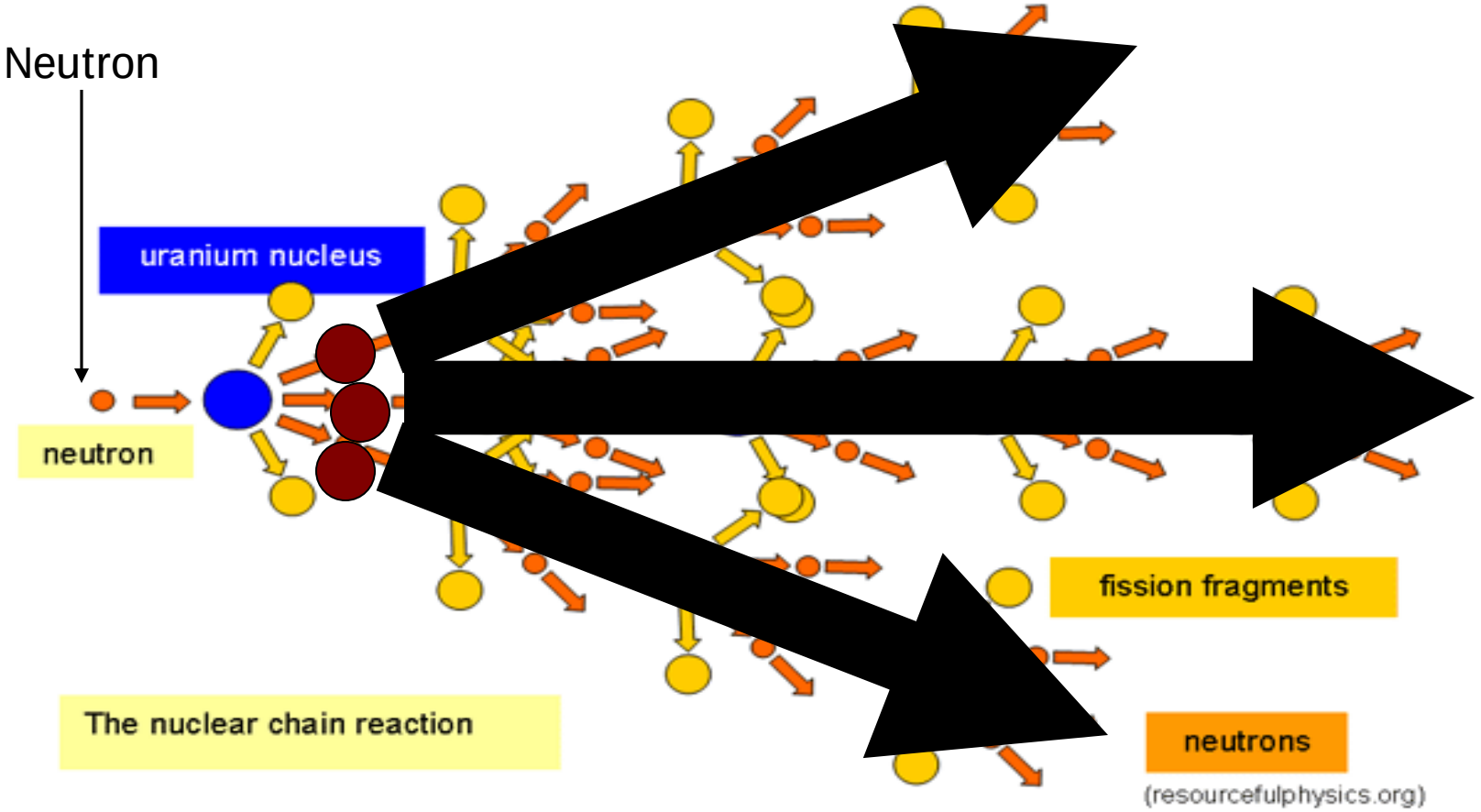
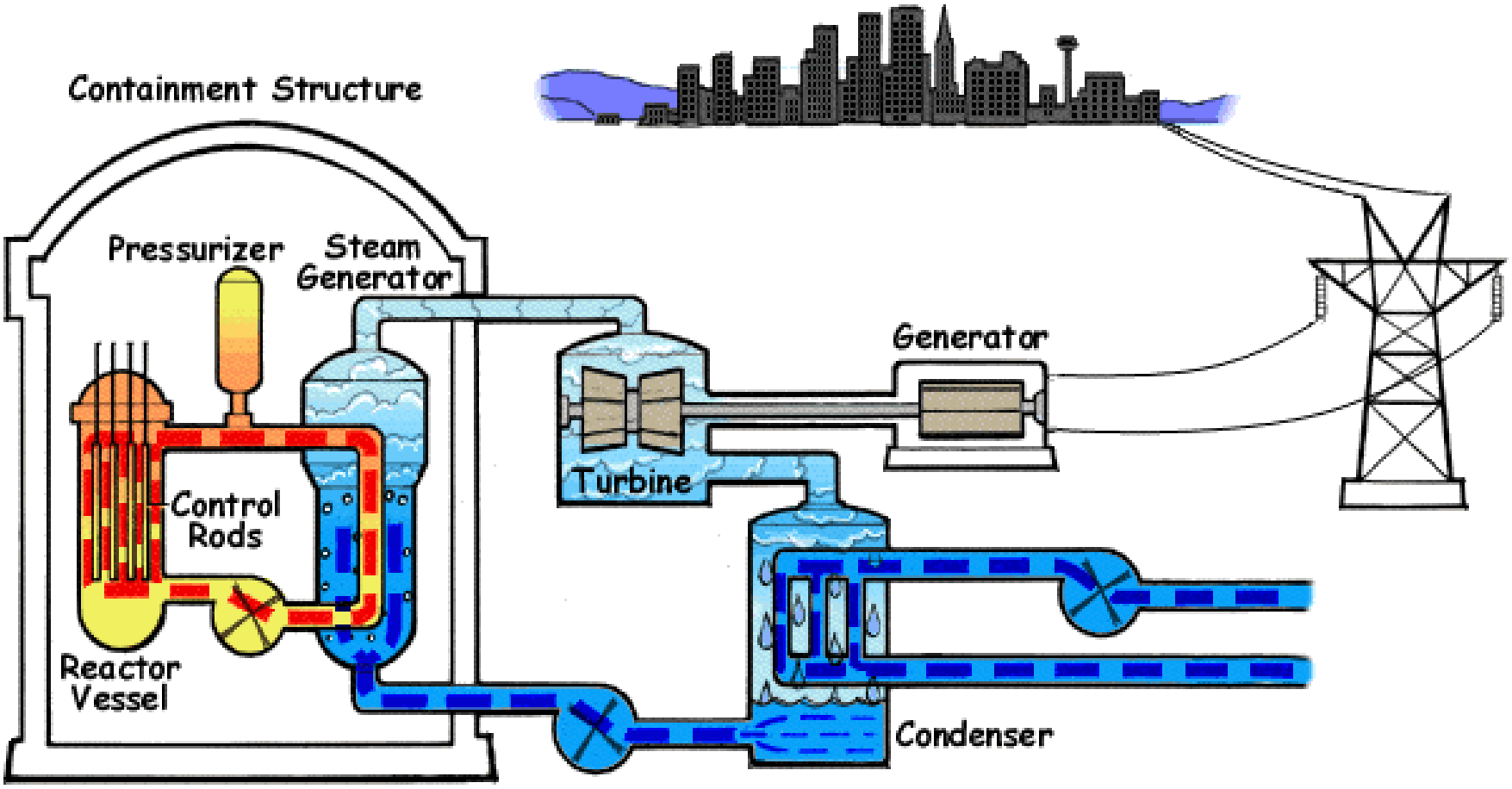


Figure 1-2. Typical commercial nuclear fuel assembly and rod.

Fission Efficiency



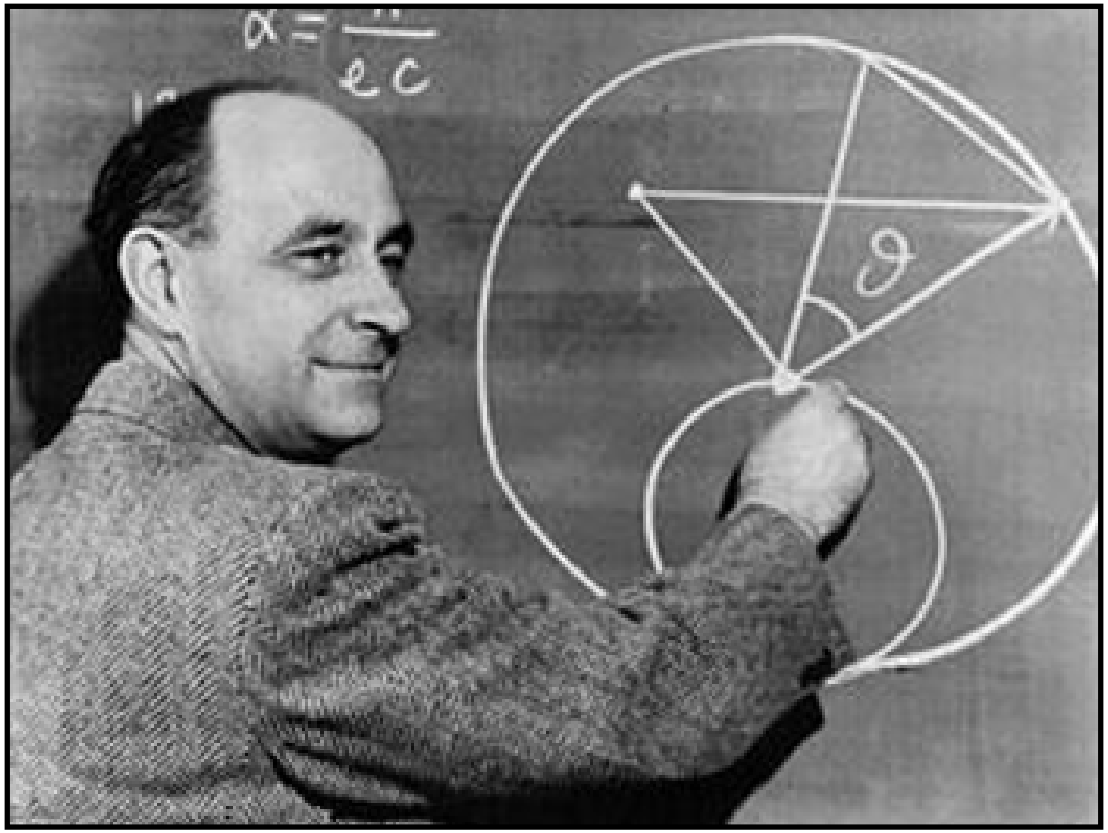
Reprocessing-Re-using Nuclear Fuel



Background of Reprocessing

- Began in 1940's
  - Fission Byproduct
    - Plutonium
- Nuclear Weapons
- Nuclear Proliferation
  - 1977
    - Presidential Directive
- Interest in next generation reactors
  - Reprocessed Uranium

Enrico Fermi



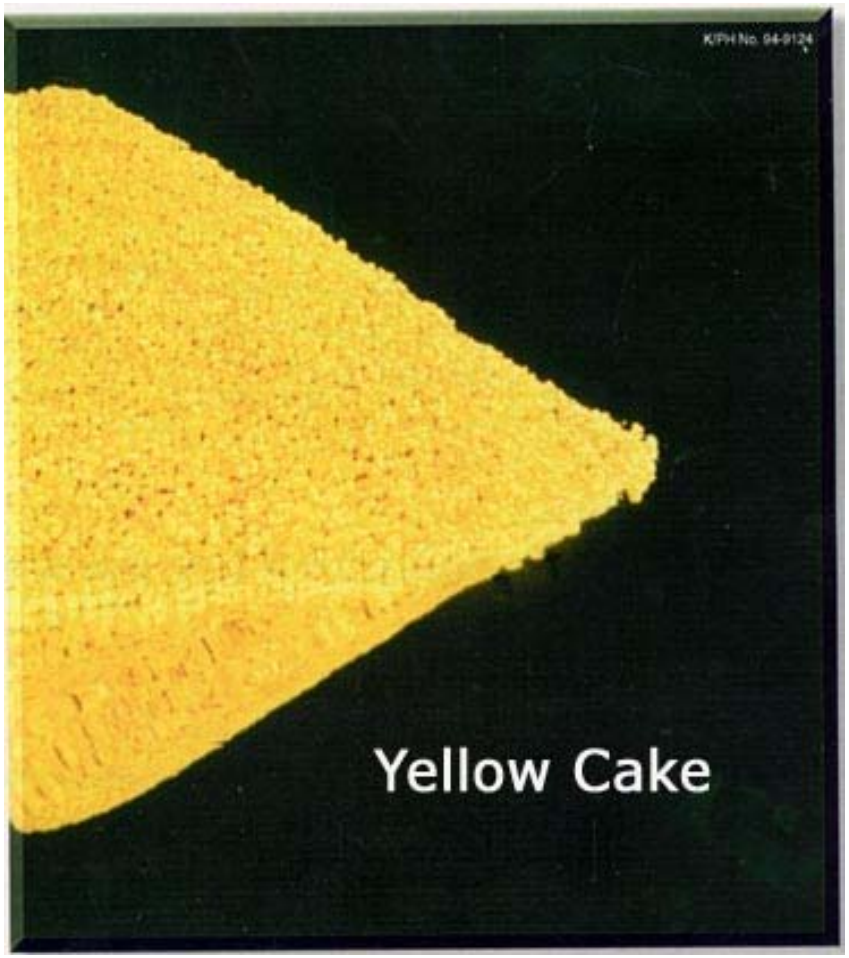
NUCLEAR FUEL CYCLE

Nuclear Fuel Cycle

- Uranium Ore
  - Starting raw material for nuclear fuel
- Typically contains .05 to .3 wt% U_3O_8
- Available isotopes U_{238} and U_{235}
- Approximately 99.28% U_{238} and .71% U_{235}

Nuclear Fuel Cycle

- Mined uranium ore is milled to isolate the U_3O_8
- Milling is typically accomplished through chemical leaching
- Produces Solid U_3O_8 commonly referred to as “Yellow Cake”

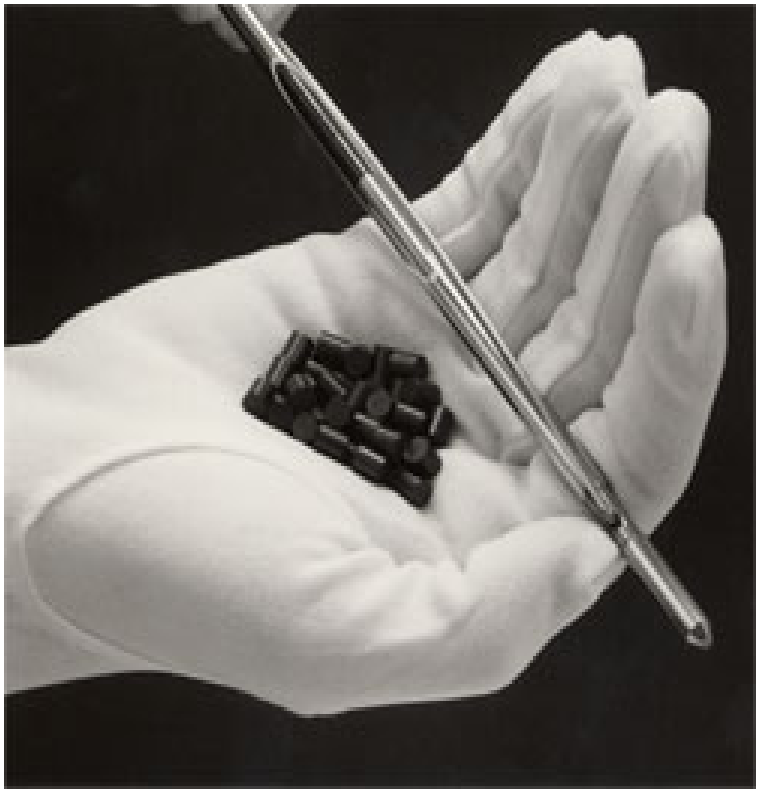


Nuclear Fuel Cycle

- Uranium Conversion
  - Required by enrichment facilities
  - Uranium hexafluoride
    - UF_6
    - Typically enrichment from .71 to 3.5% U_{235} depending on reactor specifications.
- Alternative Uranium Conversion
  - Ceramic Grade Uranium dioxide UO_2
    - CANDU-Reactors

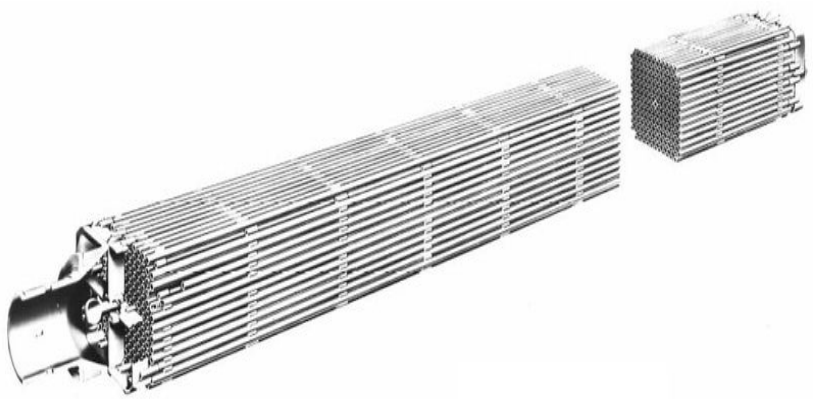
Nuclear Fuel Cycle

- Enrichment
  - Fabrication
- Enriched Uranium Pellets
  - Generally placed in fuel rods to meet specific core specifications
- Fuel rod casing
  - Stainless Steel
  - Zirconium
    - Fuel Rod Bundles

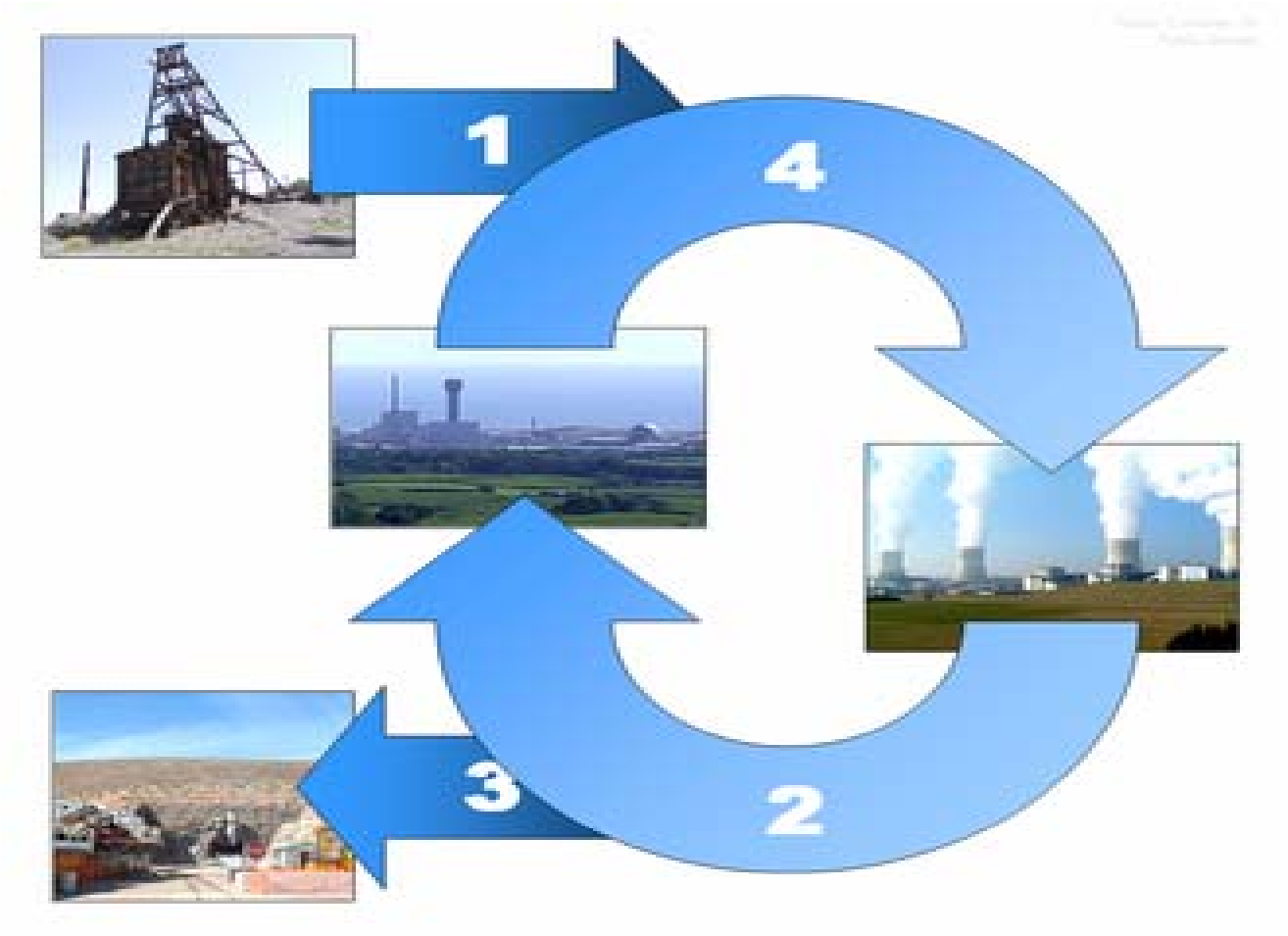


Nuclear Fuel Cycle

- PWR/BWR most common fuel rod configuration
- Typically put into bundles of 6 to 8 individual fuel rod assemblies
- Depending on Energy Production requirements
  - 2 to 6 year life span
- Fuel rod adjustments
  - Often require adjusting during operation



Recycle Nuclear Fuel

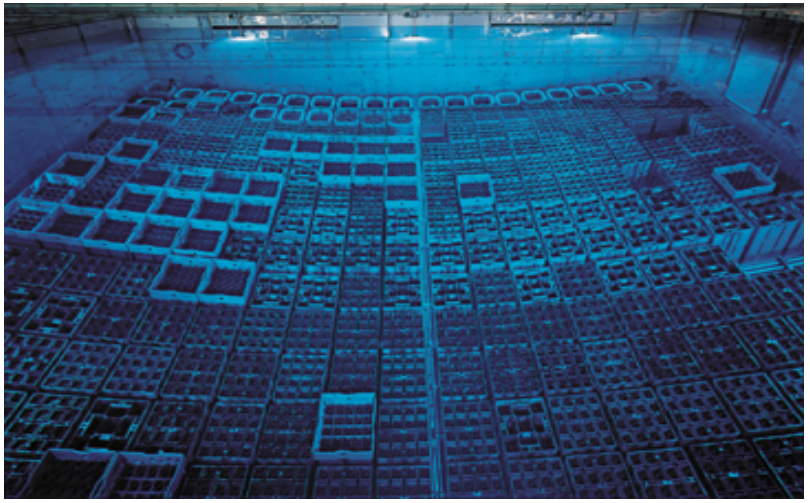


Options for “Spent Fuel”

- Storage
  - Short Term storage
    - Spent Fuel Pool
    - Dry Cask
  - Long Term Storage
    - Yucca Mountain
      - Environmental Concerns
      - Transportation
- Reprocessing
  - Environmental Concerns
  - Economical-Political
    - Includes Long Term Storage

Nuclear Fuel Cycle

- Spent Fuel Storage
- Continues to generate heat after removal from reactor
  - Spent fuel pool
    - Storage time ranges from 1-5 years depending on initial reactor operating conditions



Nuclear Fuel Cycle

- Dry Cask Storage
- Required after spent fuel pool
- Generally stored on reactor site
- Approx: 6 dozen fuel bundles/cask
  - Inert Gas
- Long term storage of a uniform container in Yucca mountain by 2017.

Federal Spent Fuel Repository

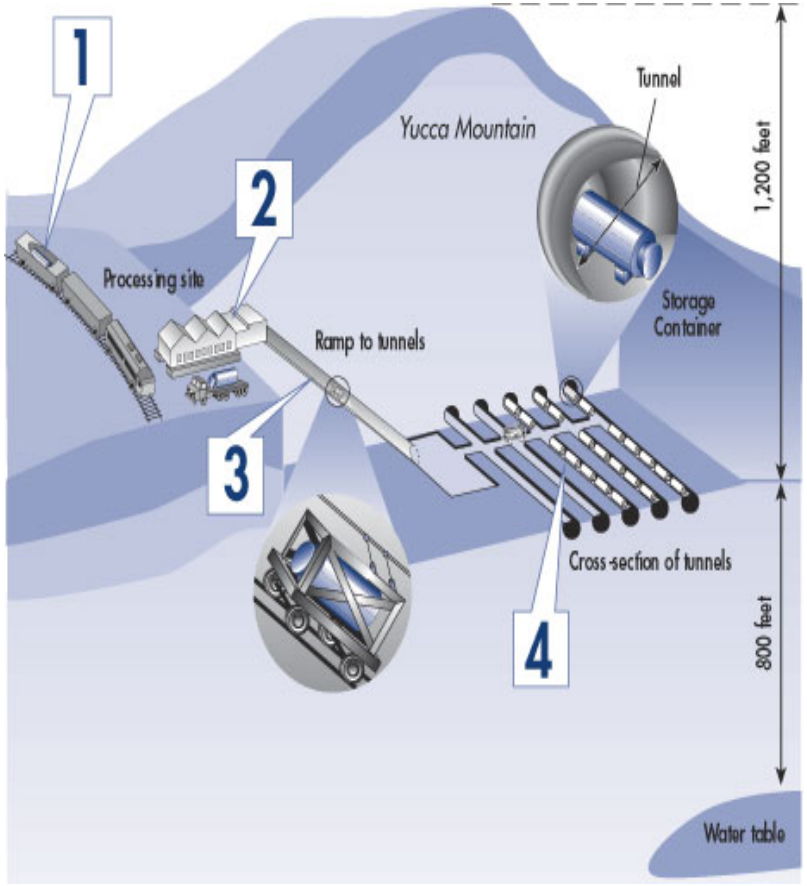
- Current U.S. policy dictates nuclear repository a better option than nuclear reprocessing
- Propose a single depository for all nuclear waste
  - Currently 126 separate repository locations nationwide
  - Costs of a single location will be less than many
- Yucca Mountain

Yucca Mountain

- Proposed National Repository
- Located in SW Nevada
- On a tectonic ridgeline
- March 31, 2017
  - Projected operation start date
- Est. total cost of 50-100 billion dollars

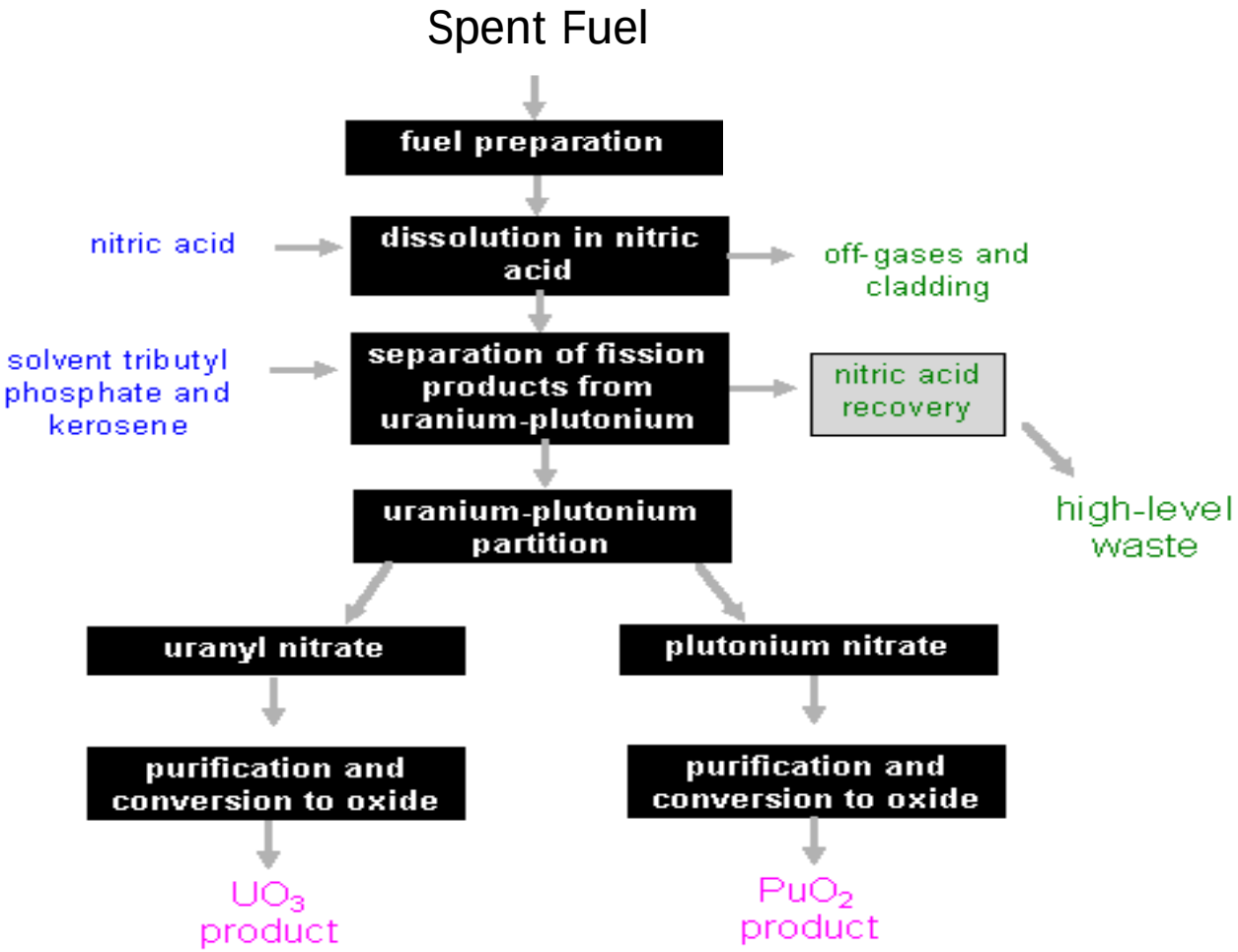
Yucca Mountain

- The Plan
  - Store spent fuel and nuclear waste 1000 ft below surface
  - Waste to be stored in individual “galleries” or alcoves
- Foreseeable Problems
  - Continued funding
  - Local and national opposition
  - Endless supply to a limited space
  - Water table

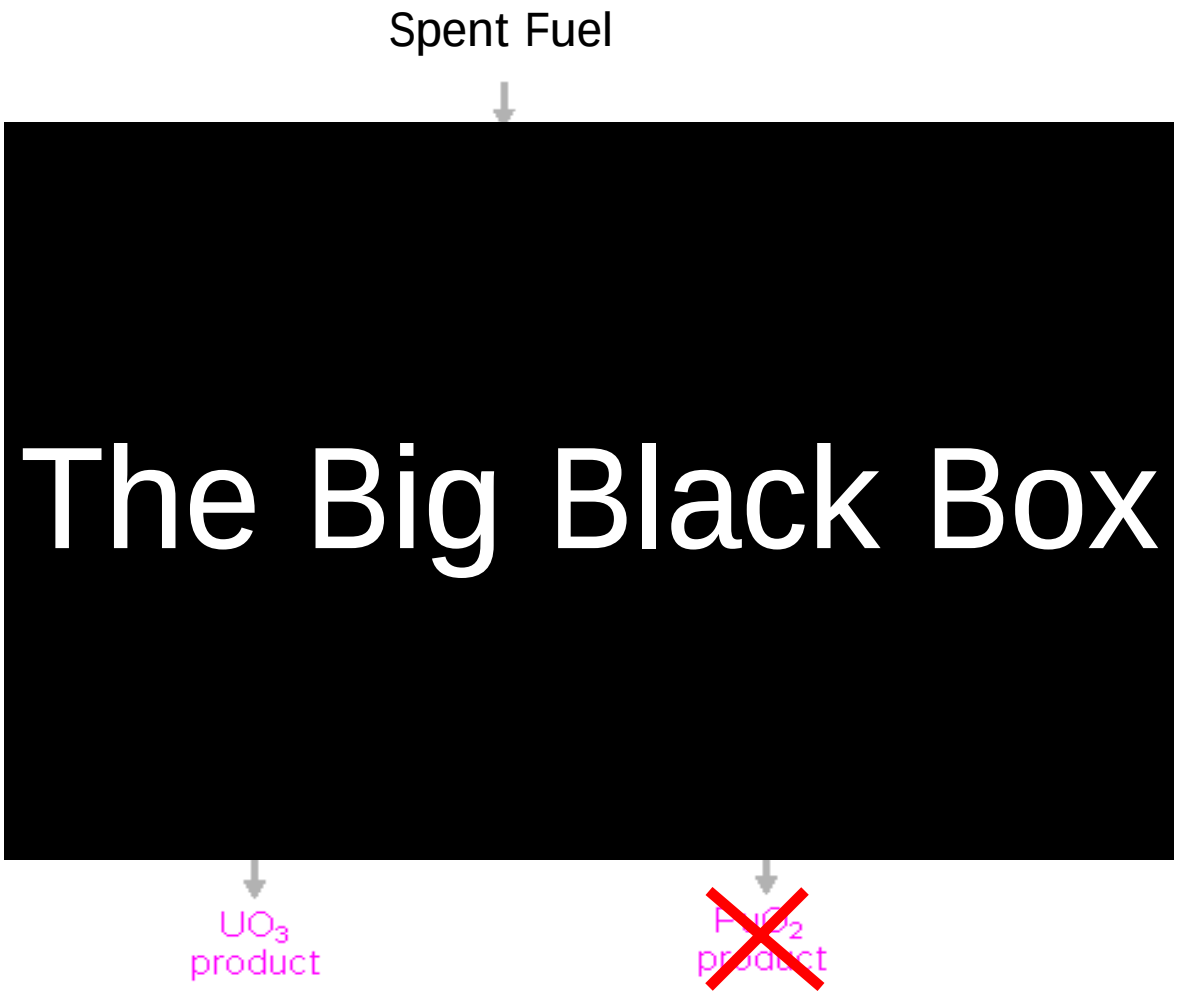


NUCLEAR FUEL REPROCESSING

The Purex Process

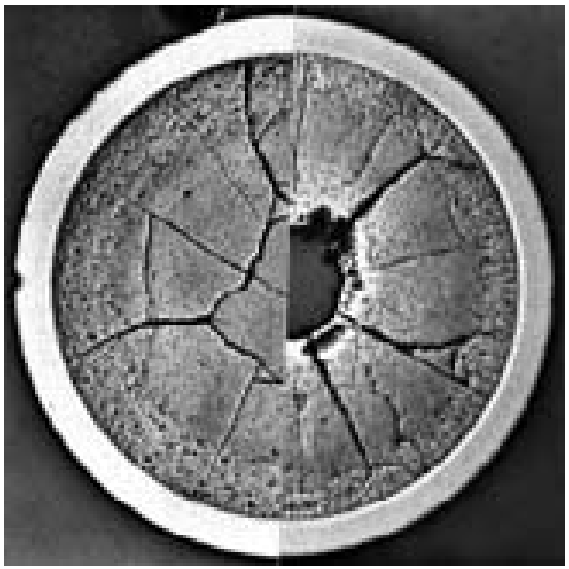


Reprocessing Technique



What we want!

- Fission Products
  - Un-used uranium

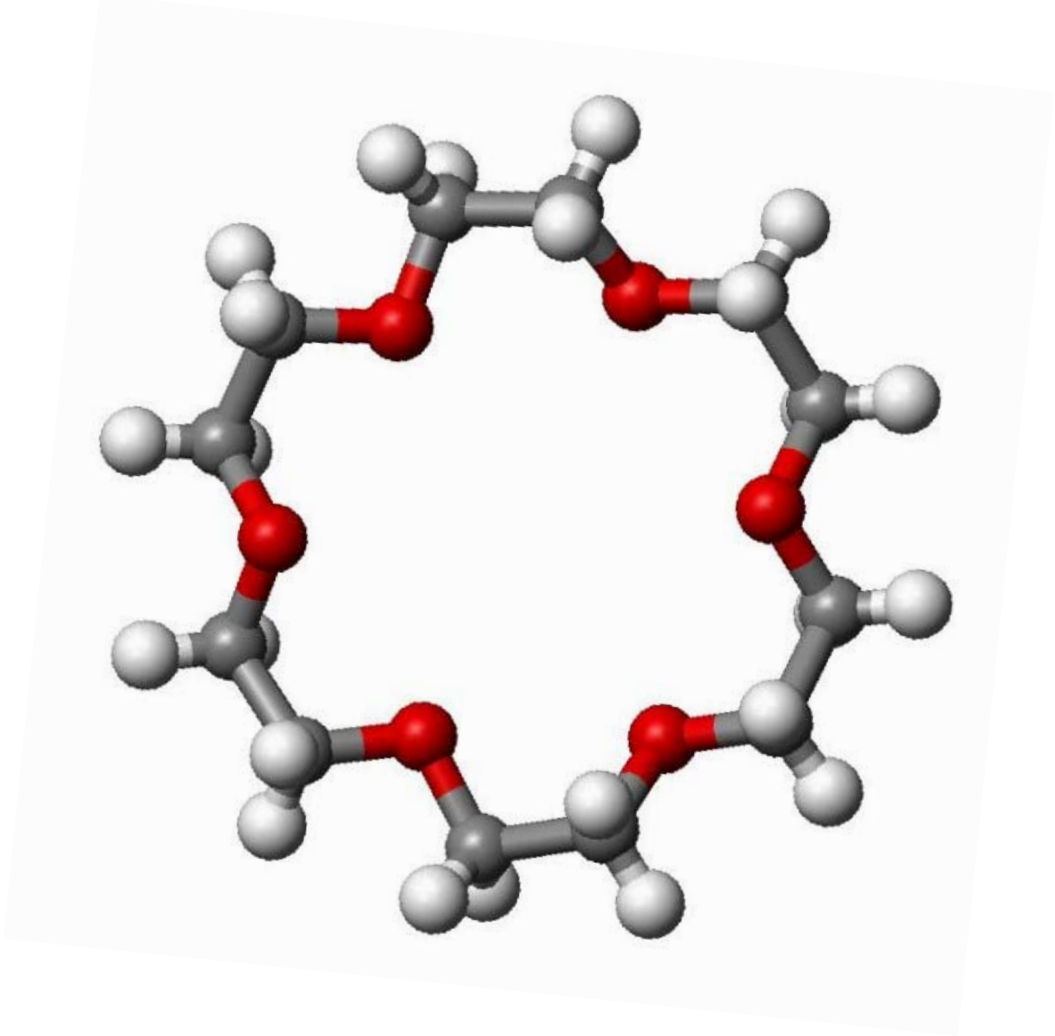


$$\longrightarrow \quad UO_2^{2+}$$

Uranyl ion

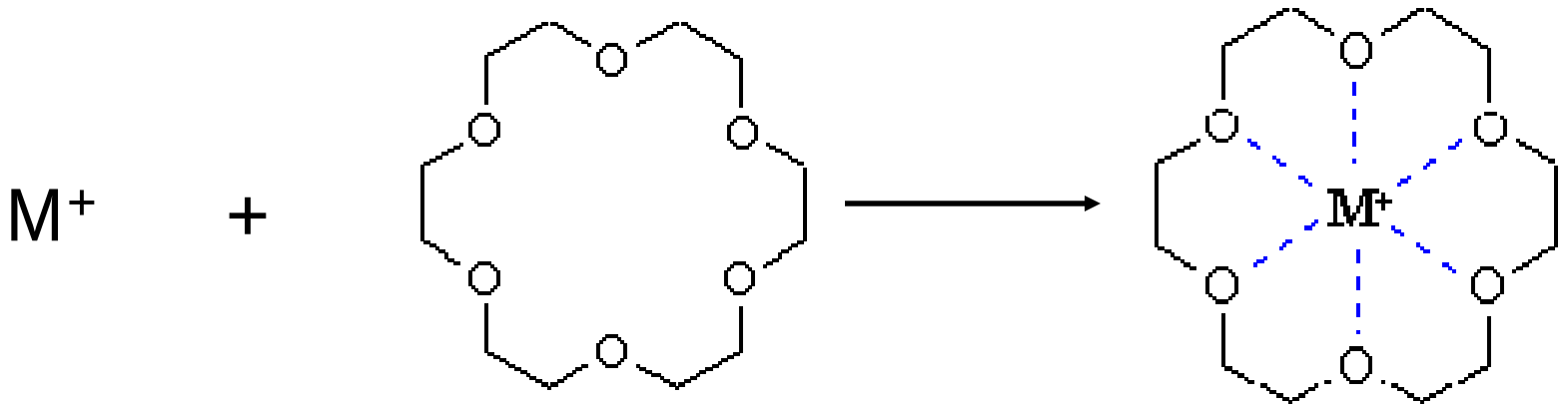
Our Solution!

Crown Ethers

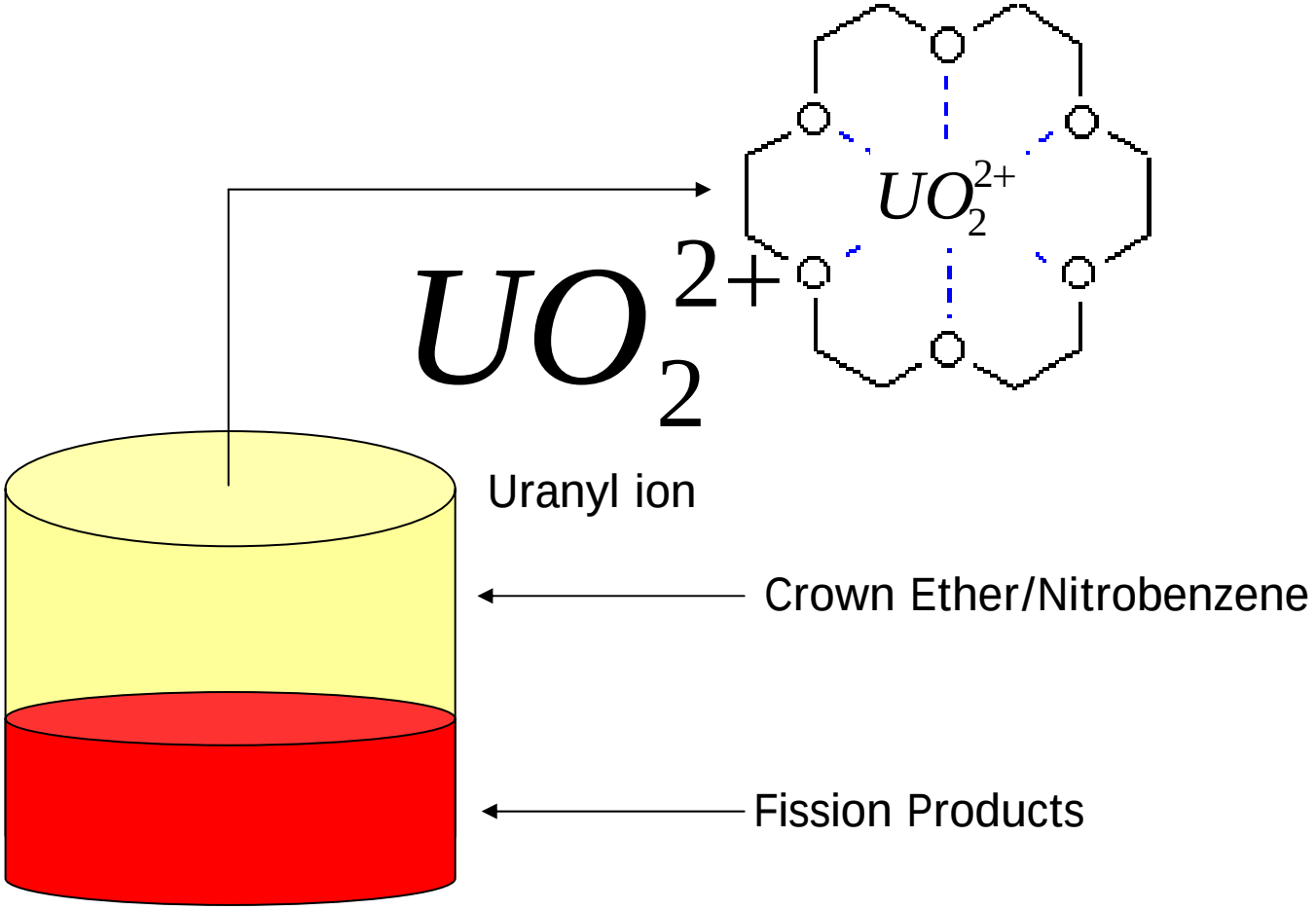


Crown Ethers

- Developed in 1960's
  - Noble Prize-1987
- Heterocyclic Chemical Compounds
  - Capable of transferring cations from an aqueous solution into an organic solution.



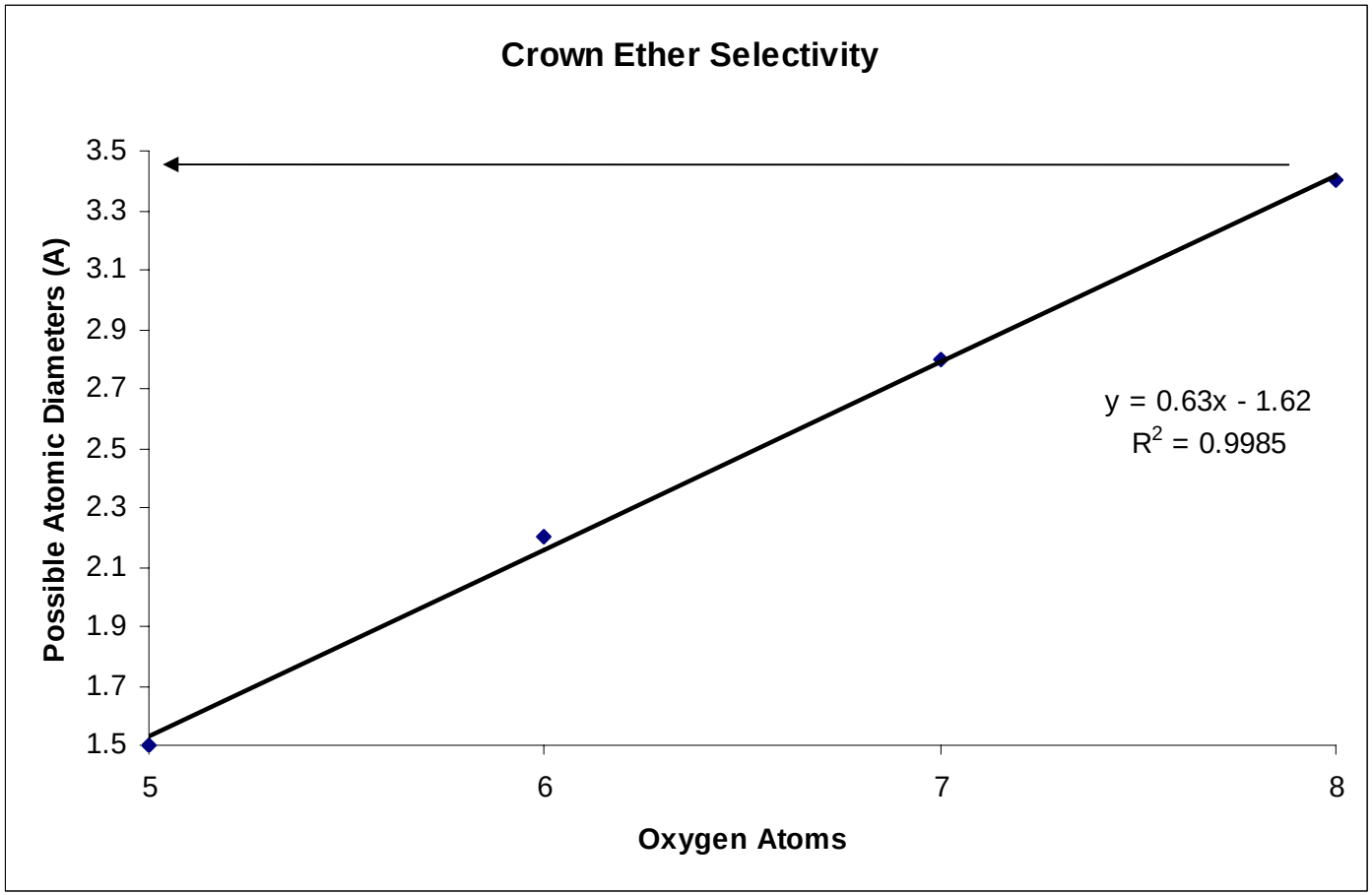
Why they will work!



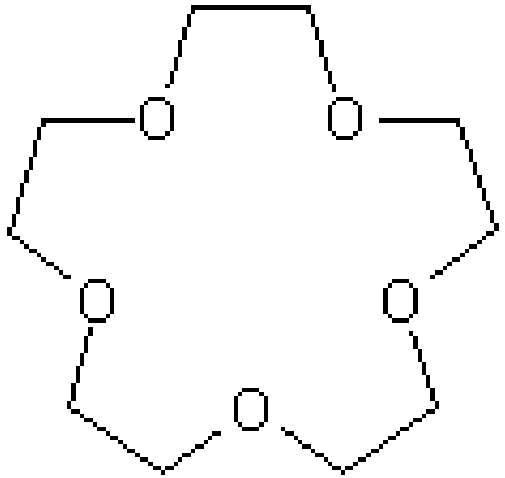
Crown Ether Selectivity

- Cation Selectivity
  - Oxygen Atoms in the ring
    - Determine atomic diameter range
- Extraction Improvement
  - Cyclohexane Rings
  - Benzene Rings

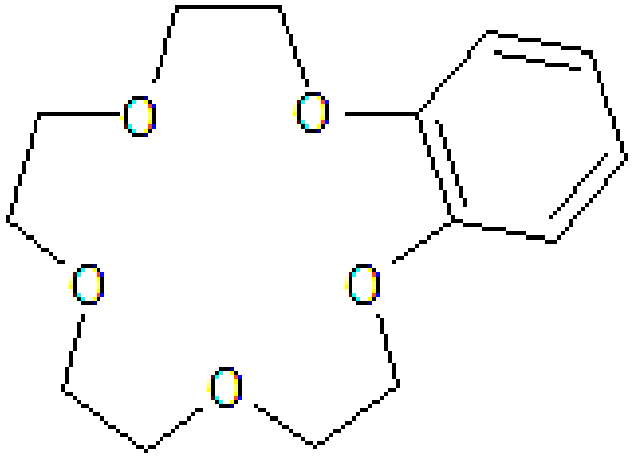
Crown Ether Characteristics



We have several Possibilities!

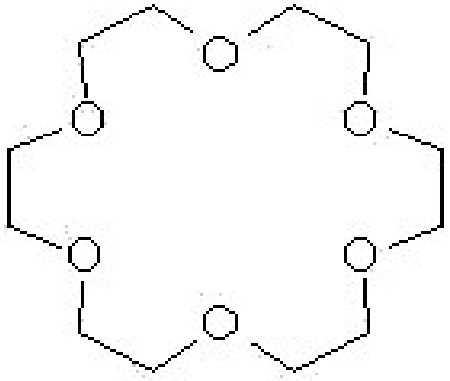


15-Crown-5

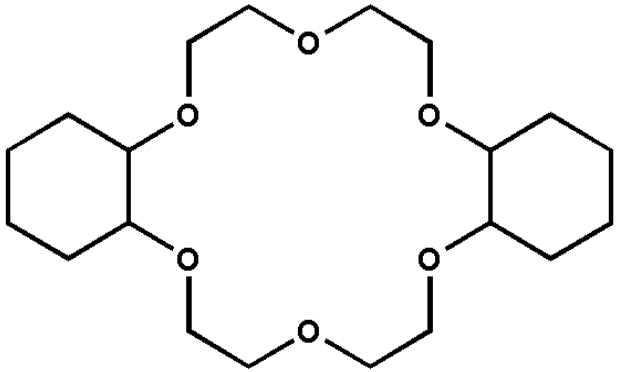


Benzo-15-Crown-5

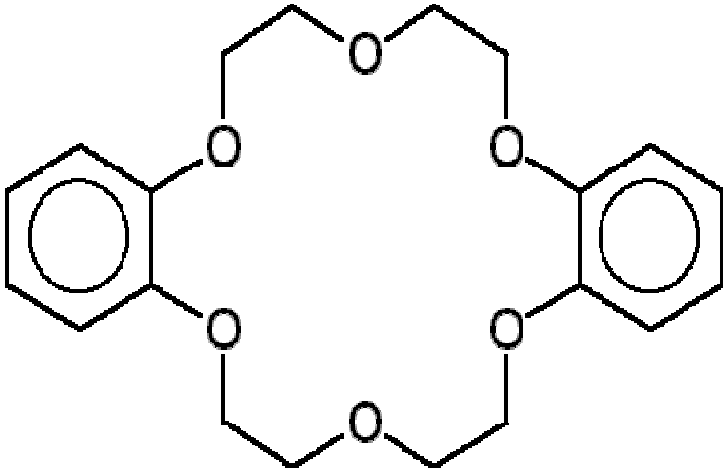
We have several Possibilities!



18-Crown-6

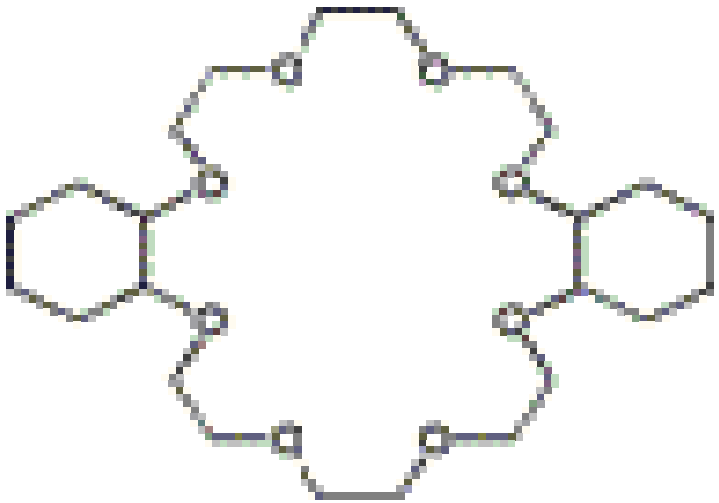


Dicyclohexane-18-Crown-6

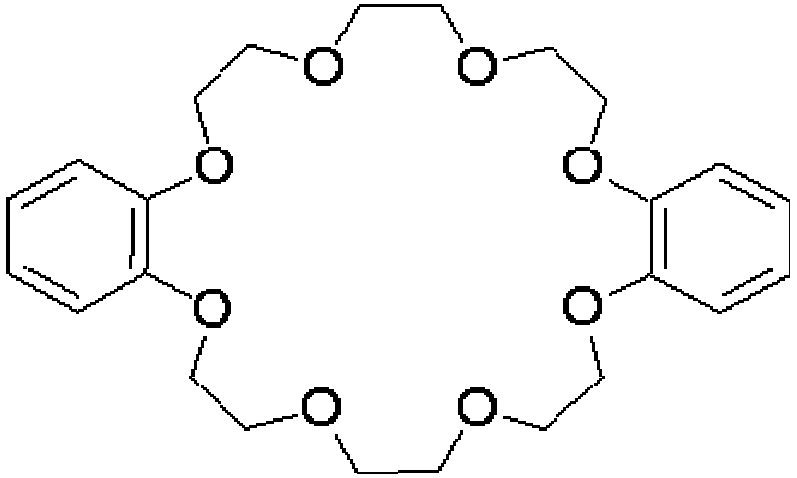


Dibenzo-18-Crown-6

We have several Possibilities!



DC-24-Crown-8

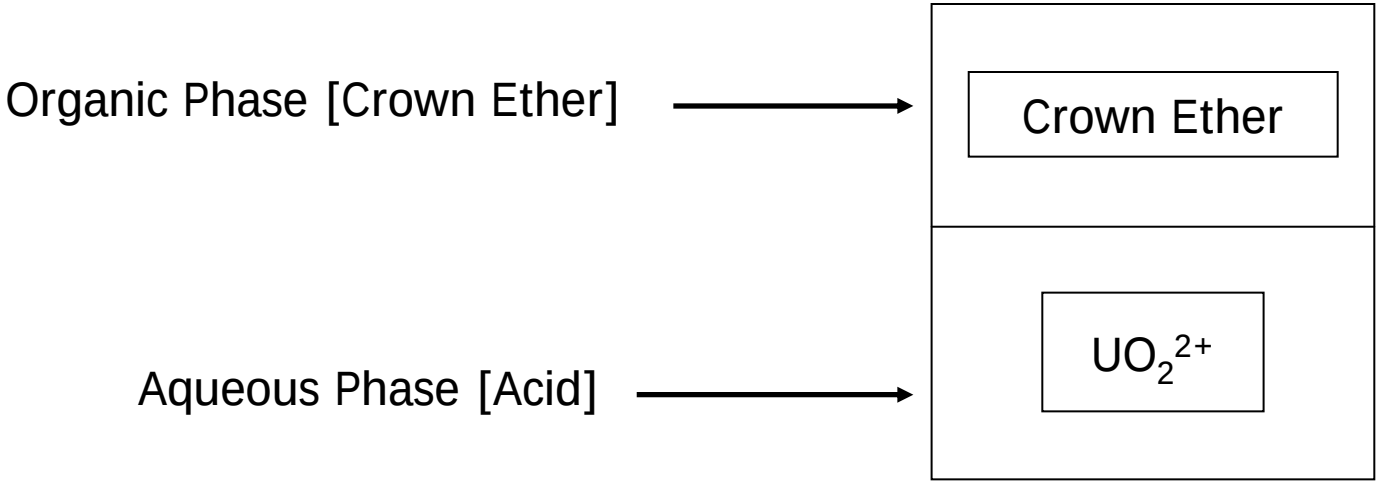


DB-24-Crown-8

Our Proposed Plan

- Dissolve Uranium Metal in a strong Acid.
  - HBr
- Combine this aqueous solution with various crown ethers
- Determine efficiency of this process based on:
  - Concentration HBr
  - Concentration of the Crown Ether in the Nitrobenzene

The Proposed Design



*Varied the concentration of Acid

*Varied the concentration of Crown Ether

Fundamental Equation

Partition Coefficient

$$K = \frac{[Concentration\ \text{Solute}]_{\text{Organic}}}{[\text{Concentration}\ \ Solute]_{Aqueous}}$$

**Organic=Crown Ether*

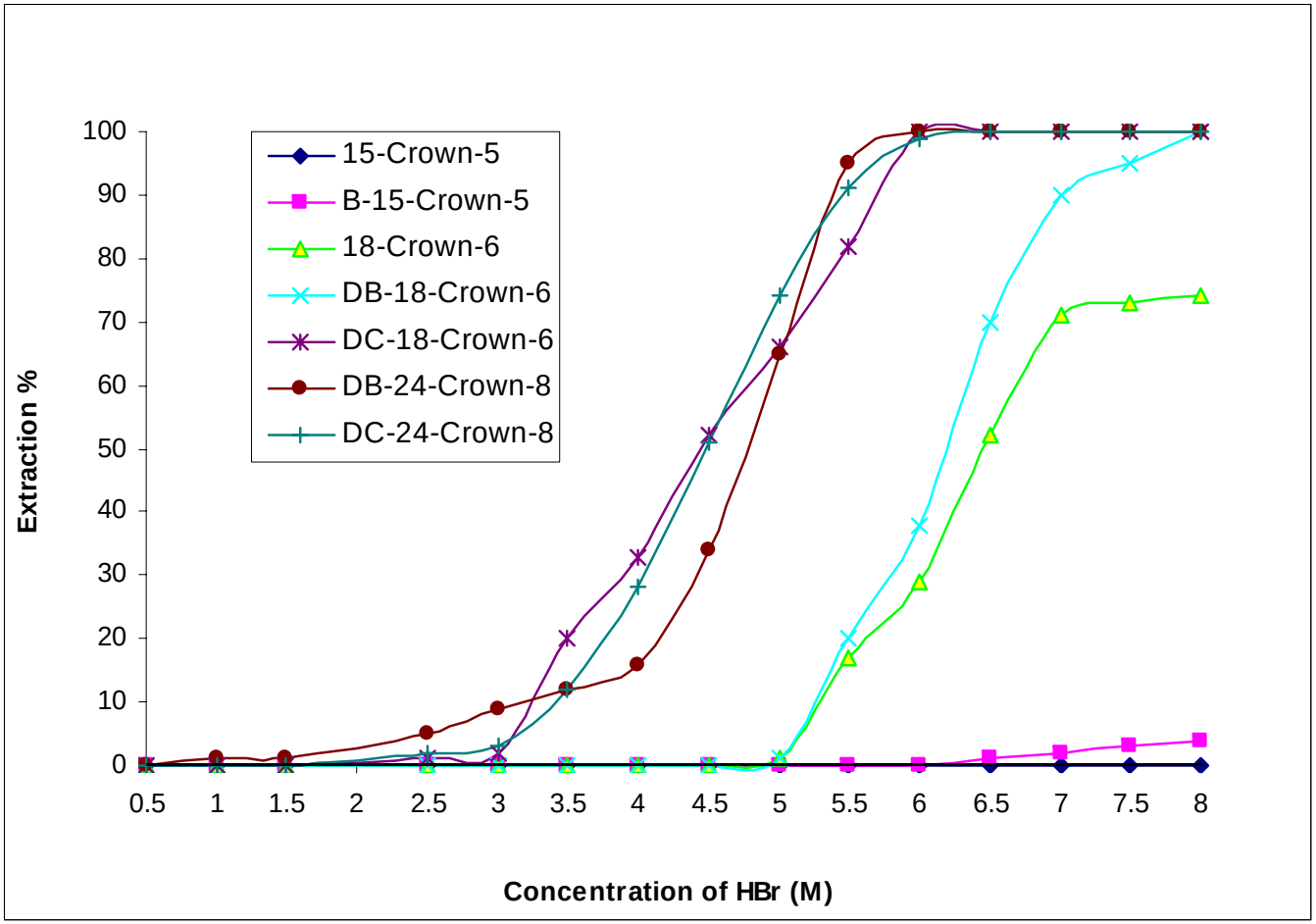
***Aqueous=Dissolved uranium in Acid

Partition Coefficient

$$K = \frac{[Concentration\ \text{Solute}]_{\text{Organic}}}{[\text{Concentration}\ \ Solute]_{Aqueous}}$$

- Extracting cation out of an aqueous solution
  - K >> 1
- Stripping cation from the crown ether
  - K << 1

Experimental Data



15-Crown-5

15-Crown-5		*Varying the concentration of HBr		
HBr				
Nitro-Benzene				
Conc: HBr*[mol/l]	$[Conc]_{aq}$	$[Conc]_{org}$	Partition Coef: K	log[HBr]*[mol/L]
0.5	4.2012E-07	0	0	-0.301029996
1	4.2012E-07	0	0	0
1.5	4.2012E-07	0	0	0.176091259
2	4.2012E-07	0	0	0.301029996
2.5	4.2012E-07	0	0	0.397940009
3	4.2012E-07	0	0	0.477121255
3.5	4.2012E-07	0	0	0.544068044
4	4.2012E-07	0	0	0.602059991
4.5	4.2012E-07	0	0	0.653212514
5	4.2012E-07	0	0	0.698970004
5.5	4.2012E-07	0	0	0.740362689
6	4.2012E-07	0	0	0.77815125
6.5	4.2012E-07	0	0	0.812913357
7	4.2012E-07	0	0	0.84509804
7.5	4.2012E-07	0	0	0.875061263
8	4.2012E-07	0	0	0.903089987

Benzo-15-Crown-5

Benzo-15-Crown-5		*Varying the concentration of HBr			
HBr					
Nitro-Benzene					
Conc: HBr*[mol/l]	$[Conc]_{aq}$	$[Conc]_{org}$	Partition Coef: K	log (K)	log[HBr]*[mol/L]
0.5	4.20117E-07	0	0	N/A	-0.301029996
1	4.20117E-07	0	0	N/A	0
1.5	4.20117E-07	0	0	N/A	0.176091259
2	4.20117E-07	0	0	N/A	0.301029996
2.5	4.20117E-07	0	0	N/A	0.397940009
3	4.20117E-07	0	0	N/A	0.477121255
3.5	4.20117E-07	0	0	N/A	0.544068044
4	4.20117E-07	0	0	N/A	0.602059991
4.5	4.20117E-07	0	0	N/A	0.653212514
5	4.20117E-07	0	0	N/A	0.698970004
5.5	4.15916E-07	4.20117E-09	0.01010101	-1.9956	0.740362689
6	4.11715E-07	8.40234E-09	0.020408163	-1.6902	0.77815125
6.5	4.11715E-07	8.40234E-09	0.020408163	-1.6902	0.812913357
7	4.07513E-07	1.26035E-08	0.030927835	-1.5097	0.84509804
7.5	4.03312E-07	1.68047E-08	0.041666667	-1.3802	0.875061263
8	4.03312E-07	1.68047E-08	0.041666667	-1.3802	0.903089987

18-Crown-6

18-Crown-6		*Varying the concentration of HBr			
HBr					
Nitro-Benzene					
Conc: HBr*[mol/l]	$[Conc]_{aq}$	$[Conc]_{org}$	Partition Coef: K	log (K)	log[HBr]*[mol/L]
0.5	4.20117E-07	0	0	N/A	-0.301029996
1	4.20117E-07	0	0	N/A	0
1.5	4.20117E-07	0	0	N/A	0.176091259
2	4.20117E-07	0	0	N/A	0.301029996
2.5	4.20117E-07	0	0	N/A	0.397940009
3	4.20117E-07	0	0	N/A	0.477121255
3.5	4.20117E-07	0	0	N/A	0.544068044
4	4.20117E-07	0	0	N/A	0.602059991
4.5	4.20117E-07	0	0	N/A	0.653212514
5	4.20117E-07	0	0	N/A	0.698970004
5.5	3.40295E-07	7.98222E-08	0.234567901	-0.629731418	0.740362689
6	2.89881E-07	1.30236E-07	0.449275362	-0.347487397	0.77815125
6.5	1.97455E-07	2.22662E-07	1.127659574	0.052178012	0.812913357
7	1.21834E-07	2.98283E-07	2.448275862	0.388860351	0.84509804
7.5	1.13432E-07	3.06685E-07	2.703703704	0.431959096	0.875061263
8	1.0923E-07	3.10886E-07	2.846153846	0.454258372	0.903089987

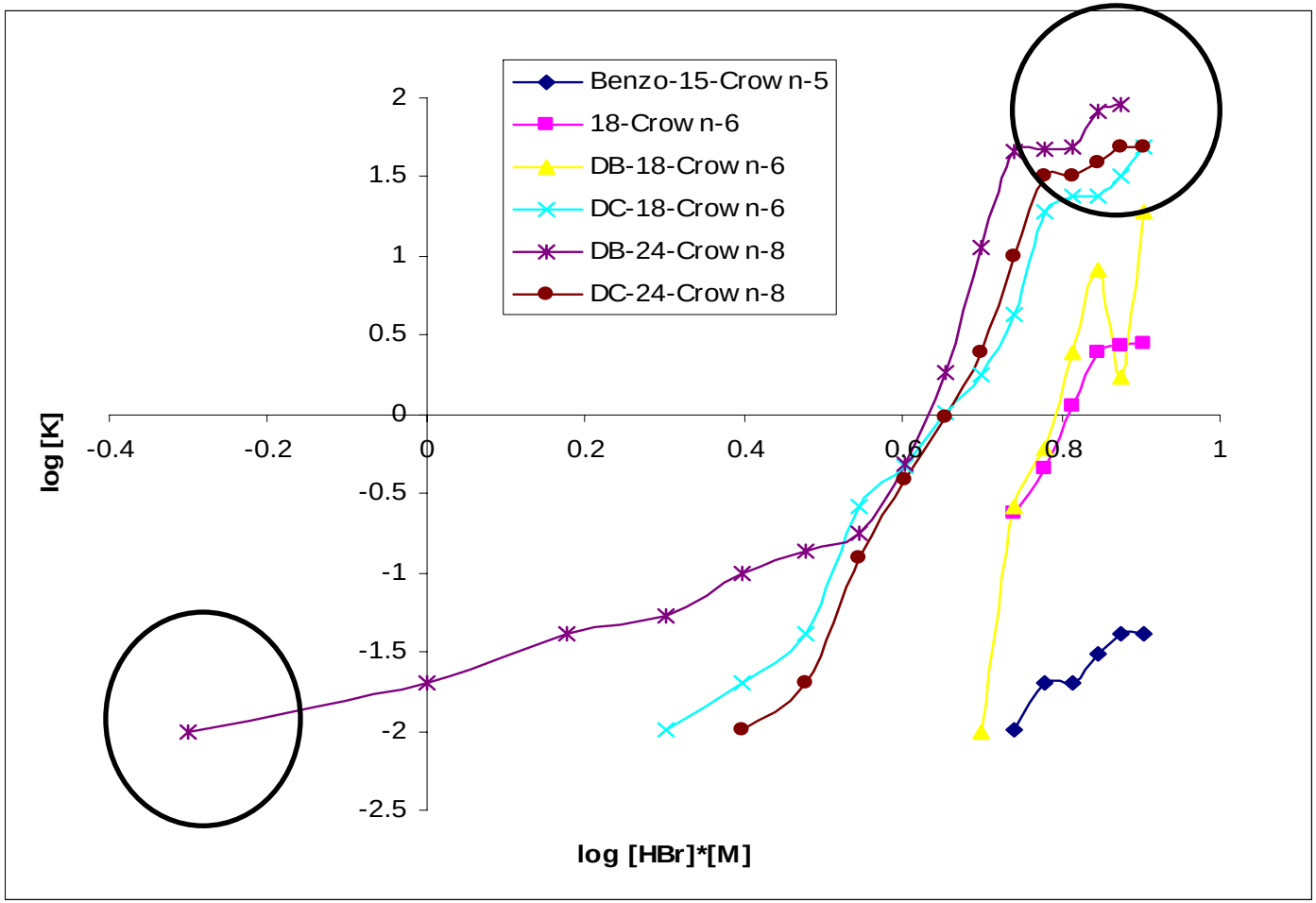
Dibenzo-18-Crown-6

DB-18-Crown-6		*Varying the concentration of HBr			
HBr					
Nitro-Benzene					
Conc: HBr*[mol/l]	$[Conc]_{aq}$	$[Conc]_{org}$	Partition Coef: K	log (K)	log[HBr]*[mol/L]
0.5	4.20117E-07	0	0	N/A	-0.301029996
1	4.20117E-07	0	0	N/A	0
1.5	4.20117E-07	0	0	N/A	0.176091259
2	4.20117E-07	0	0	N/A	0.301029996
2.5	4.20117E-07	0	0	N/A	0.397940009
3	4.20117E-07	0	0	N/A	0.477121255
3.5	4.20117E-07	0	0	N/A	0.544068044
4	4.20117E-07	0	0	N/A	0.602059991
4.5	4.20117E-07	0	0	N/A	0.653212514
5	4.20117E-07	4.20117E-09	0.01	-2	0.698970004
5.5	3.31892E-07	8.82245E-08	0.265822785	-0.575407797	0.740362689
6	2.60472E-07	1.59644E-07	0.612903226	-0.212608093	0.77815125
6.5	1.21834E-07	2.98283E-07	2.448275862	0.388860351	0.812913357
7	4.62129E-08	3.73904E-07	8.090909091	0.907997321	0.84509804
7.5	1.55443E-07	2.64674E-07	1.702702703	0.231138825	0.875061263
8	2.10058E-08	3.99111E-07	19	1.278753601	0.903089987

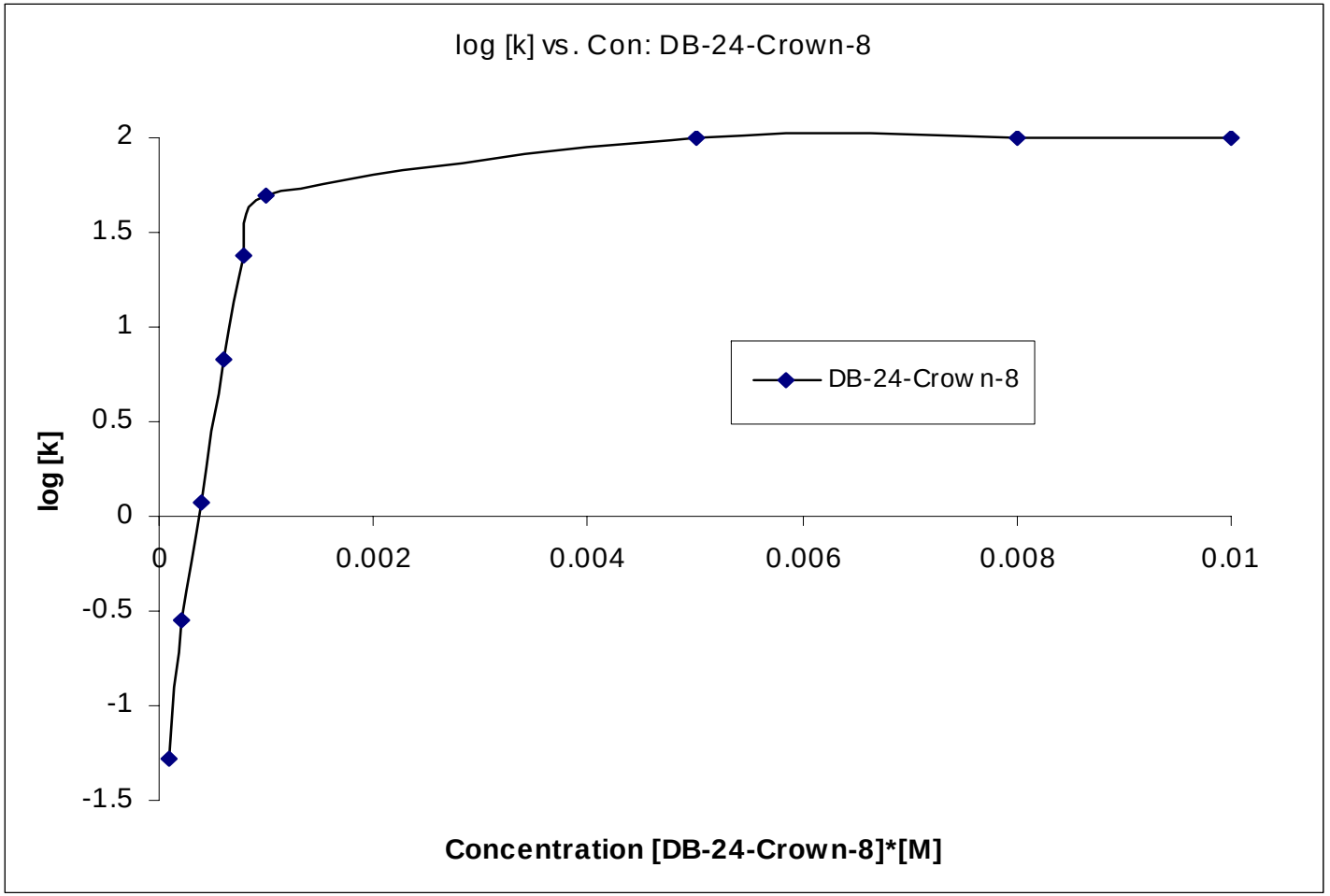
Dibenzo-24-Crown-8

DB-24-Crown-8		*Varying the concentration of HBr			
HBr					
Nitro-Benzene					
Conc: HBr*[mol/l]	$[Conc]_{aq}$	$[Conc]_{org}$	Partition Coef: K	log (K)	log[HBr]*[mol/L]
0.5	4.20117E-07	0	0	N/A	-0.301029996
1	4.20117E-07	4.20117E-09	0.01	-2	0
1.5	4.20117E-07	8.40234E-09	0.02	-1.698970004	0.176091259
2	4.03312E-07	1.68047E-08	0.041666667	-1.380211242	0.301029996
2.5	3.99111E-07	2.10058E-08	0.052631579	-1.278753601	0.397940009
3	3.82306E-07	3.78105E-08	0.098901099	-1.004798883	0.477121255
3.5	3.69703E-07	5.0414E-08	0.136363636	-0.865301426	0.544068044
4	3.57099E-07	6.30175E-08	0.176470588	-0.753327667	0.602059991
4.5	2.81478E-07	1.38639E-07	0.492537313	-0.307560863	0.653212514
5	1.47041E-07	2.73076E-07	1.857142857	0.268845312	0.698970004
5.5	3.36094E-08	3.86508E-07	11.5	1.06069784	0.740362689
6	8.86447E-09	4.11252E-07	46.39336493	1.666455873	0.77815125
6.5	8.82245E-09	4.11294E-07	46.61904762	1.668563397	0.812913357
7	8.40234E-09	4.11715E-07	49	1.69019608	0.84509804
7.5	5.0414E-09	4.15075E-07	82.33333333	1.915575699	0.875061263
8	4.62129E-09	4.15496E-07	89.90909091	1.953803606	0.903089987

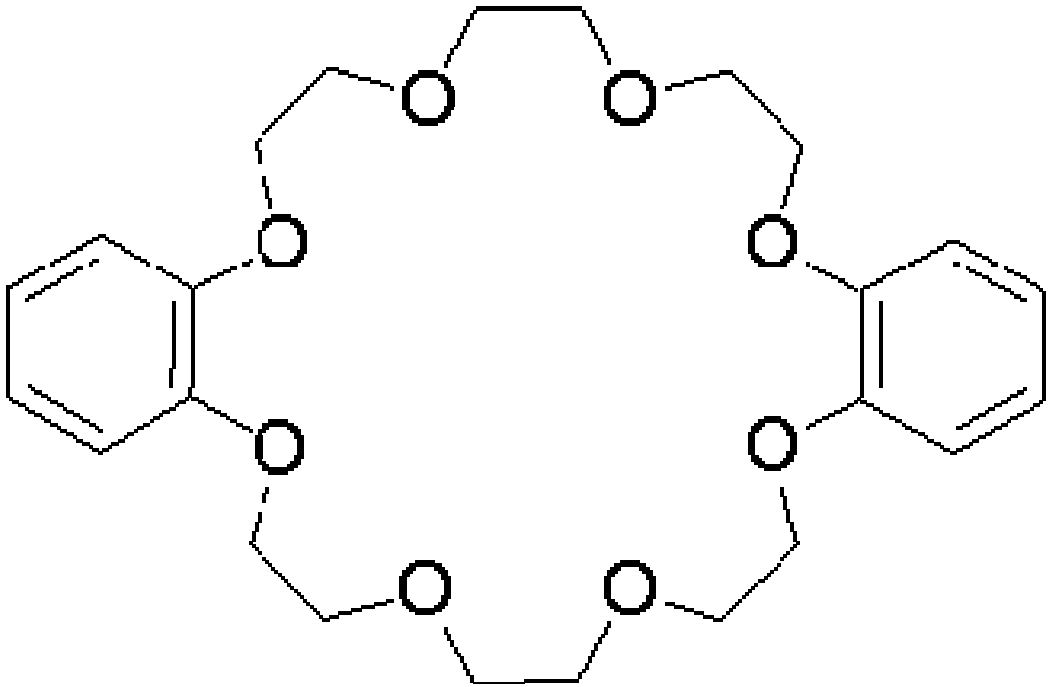
Partition Coeff: vs. [HBr]



Partition Coeff: vs. [DB-24-Crown-8]



As Expected!!!!



Optimal Extraction

$$HBr + H_2O \rightarrow Br^- + H_3O^+$$

[7.5 M]

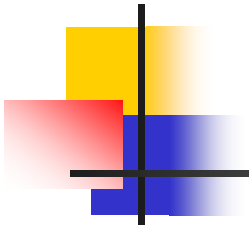
$$DiBenzo - 24 - Crown - 8$$

[.01 M]

$$UO_2^{2+} + 2Br^- + DB24C8 \xleftrightarrow{25^\circ C} UO_2(DB24C8)_2 + Br_2$$

$$K = \frac{[Concentration\ \text{Solute}]_{\text{Organic}}}{[\text{Concentration}\ Solute]_{Aqeous}} = 82.33$$

Optimal Stripping [Reverse Reaction]



$$HBr + H_2O \rightarrow Br^- + H_3O^+$$

[.45 M]

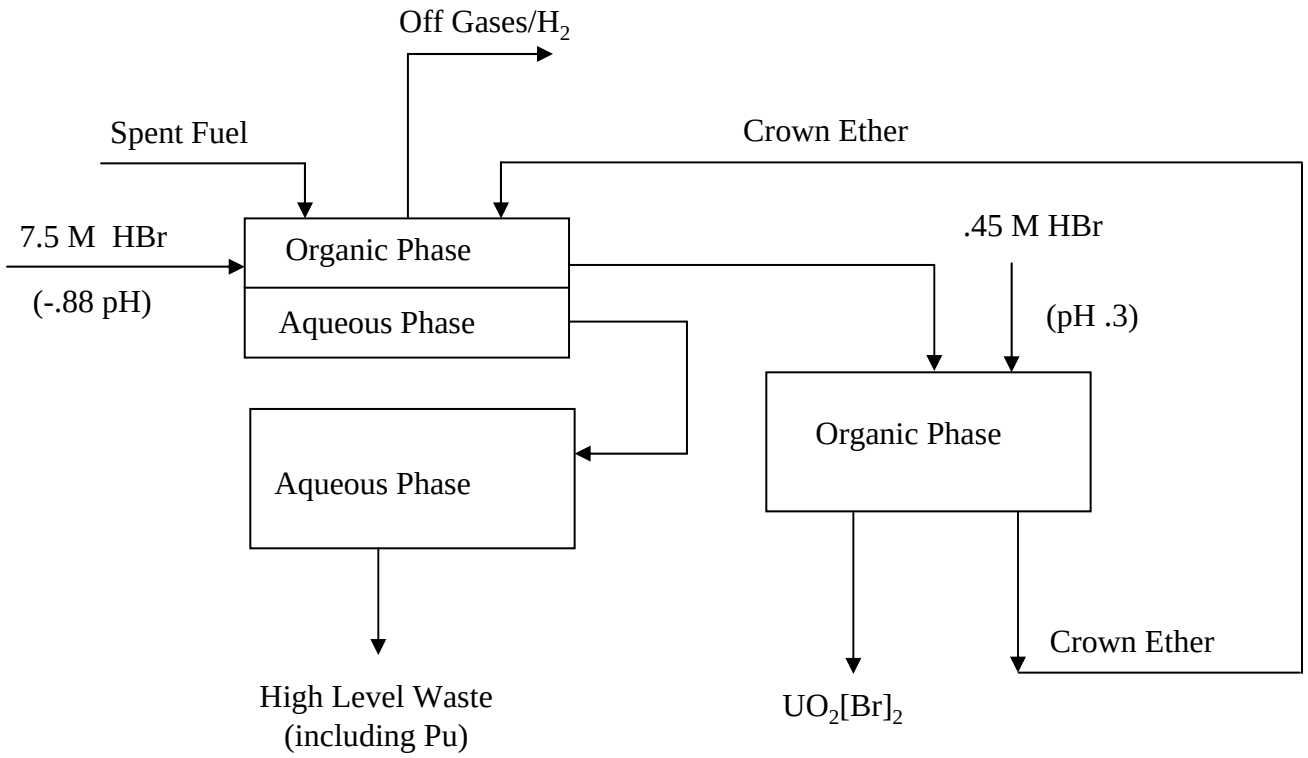
$$DiBenzo - 24 - Crown - 8$$

[.01 M]

$$UO_2^{2+} + 2Br^- + DB24C8 \xleftrightarrow{25^\circ C} UO_2(DB24C8)_2 + Br_2$$

$$K = \frac{[Concentration\ \text{Solute}]_{\text{Organic}}}{[\text{Concentration}\ Solute]_{Aqeous}} = .01$$

Proposed PFD

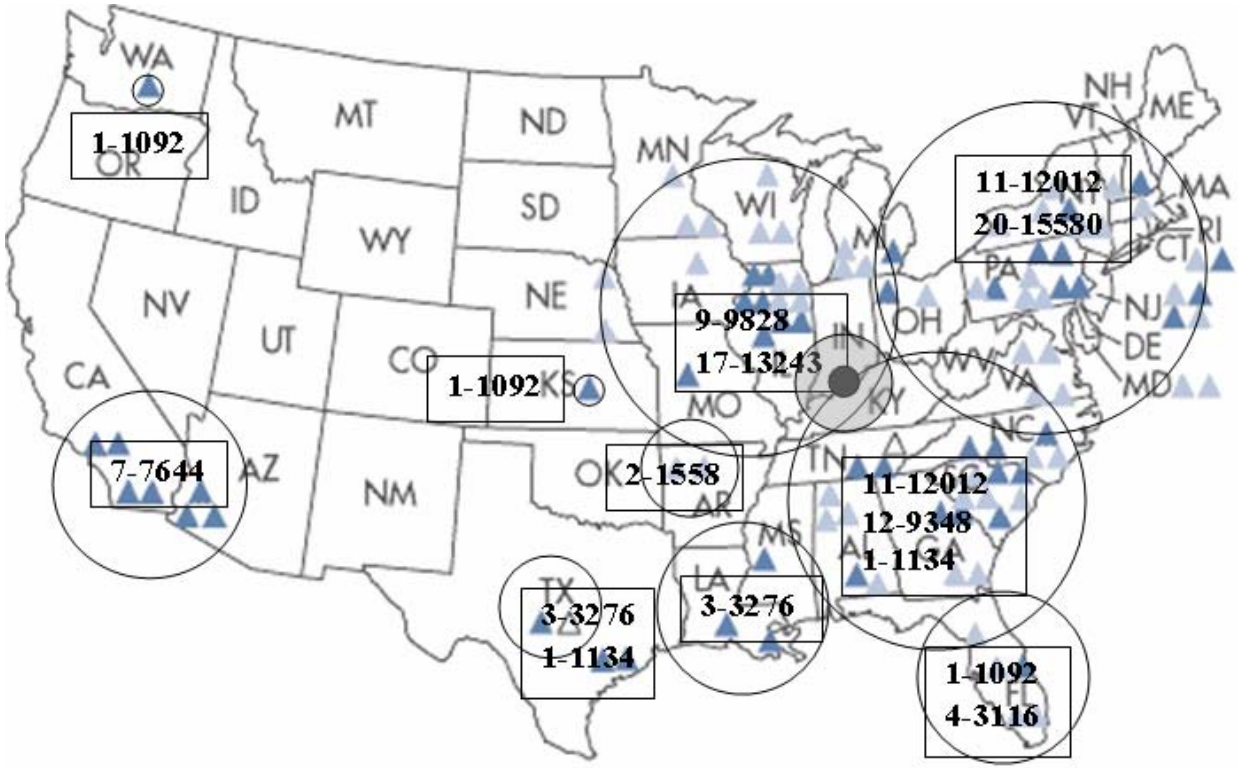


Site Location-Economics

Reprocessing Site Location

- Key Factors to Analyze
  - Relation to all of the nuclear facilities
    - Distance from the sites
    - Amount of spent fuel to be reprocessed from each site
  - Proximity to populous regions
  - Geography
  - Distance from major interstates
  - Proximity to the railroad system

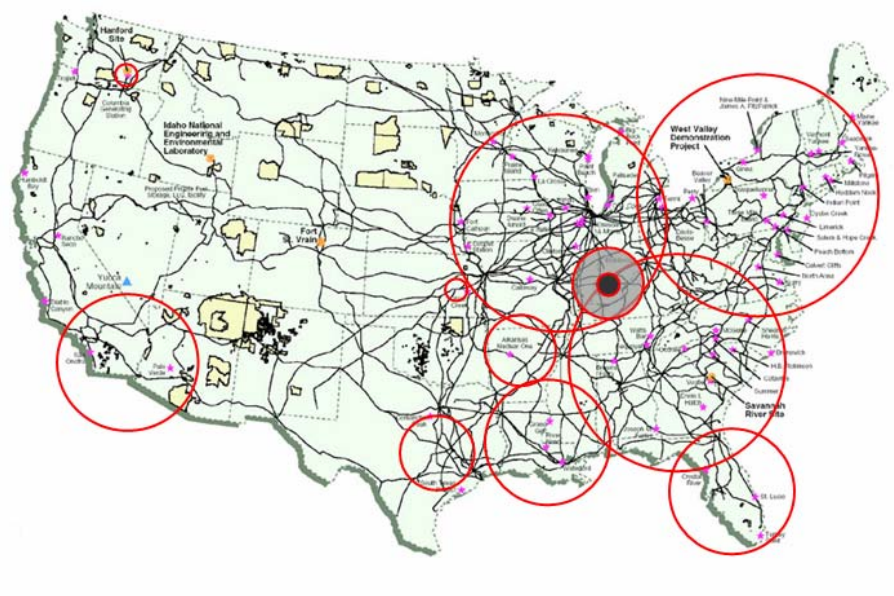
Reprocessing Site Location



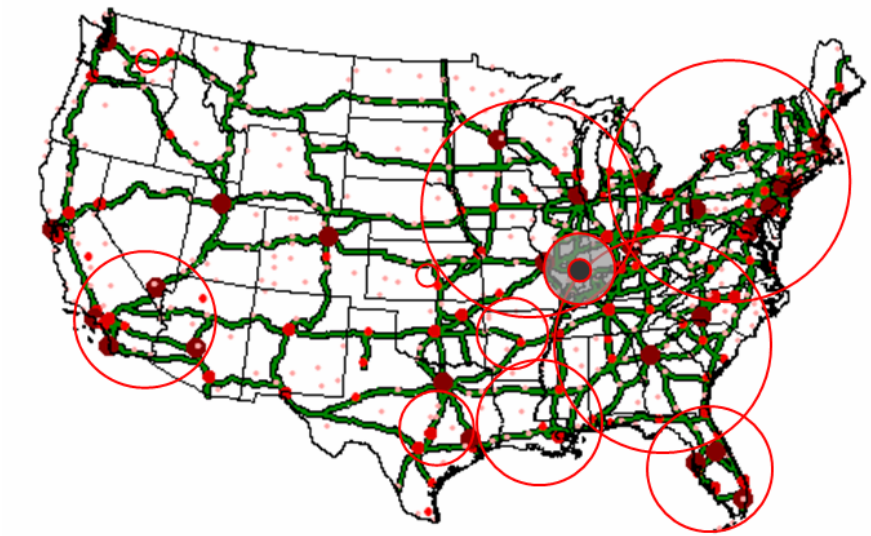
-General vicinity found by equating centralized point in relation to all nuclear reactors in the United States.

Reprocessing Site Location

U.S. Railroad System

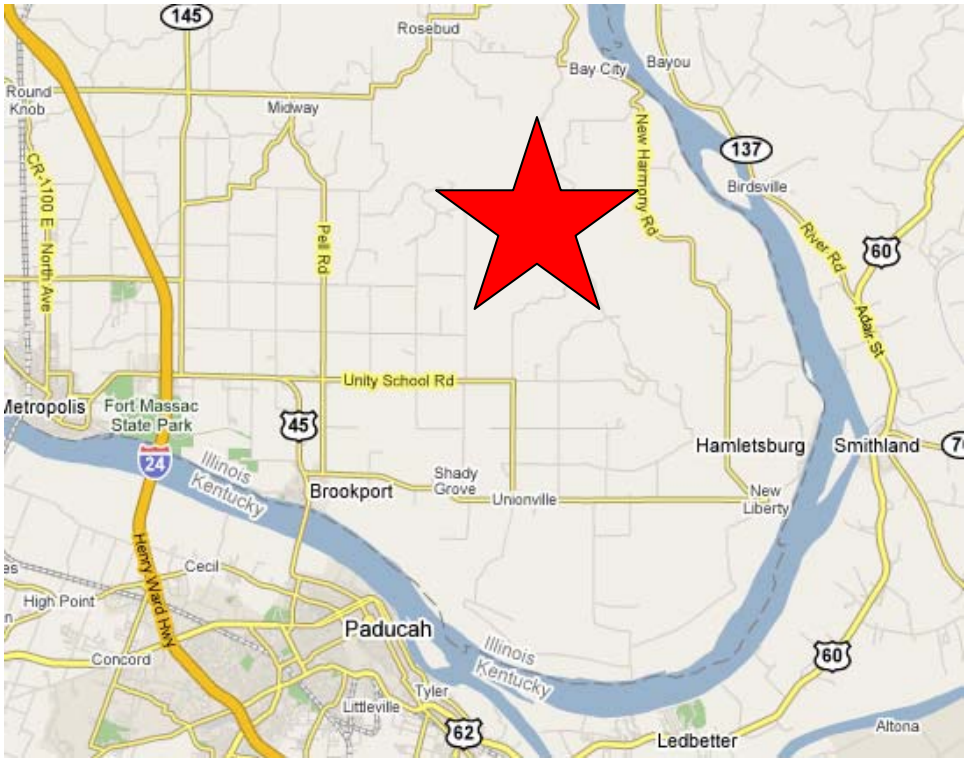


U.S. Interstate System



Metropolis, IL

- Remote Location
- Interstate-24
- Ohio River
- Feeder Railroads into St. Louis



Projected Cost

- Difficult to gauge
- How do we approach the development of an accurate budget?
  - Look at current and past reprocessing facilities built in other countries
  - Focus on the building infrastructure
    - This cost will far outweigh the associated equipment costs

Projected Cost Cont.

Rokkasho, Japan

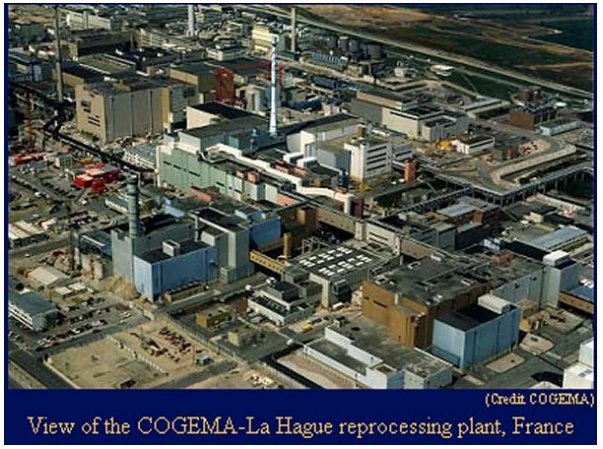
2005

Capacity: 800 metric tons/yr

TCI: $21 billion

Operational By: ??

La Hague, France



1976

Capacity: 1700 metric tons/yr

TCI: $14 billion

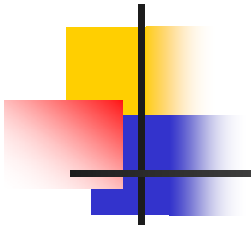
(several plant capacity expansions)

Projected Cost Cont.

Total Capital Investment (7500 metric ton/yr capacity)	
Direct Costs	**$31,434,562,500.00**
Purchased Equipment/Instrumentation & Controls	$39,250,000.00
Installation	$13,125,000.00
Building/Piping/Insulation	$28,050,000,000.00
Electrical	$876,562,500.00
Service/HBR holding facilities	$2,454,375,000.00
Land ($2000/acre) (625 acres)	$1,250,000.00
Indirect Costs	**$12,207,327,500.00**
Engineering and Supervision	$2,805,000,000.00
Legal Expenses	$15,605,000.00
Construction expense and contractor's fee	$4,511,722,500.00
Contingency	$4,875,000,000.00
Fixed Capital Investment	**$43,641,890,000.00**
Working Capital	$6,015,630,000.00
Total Capital Investment	**$49,657,520,000.00**

Recommendations

- Explore different Crown Ethers
- Explore various Acids
- Explore different design and economic aspects of the crown ether reprocessing

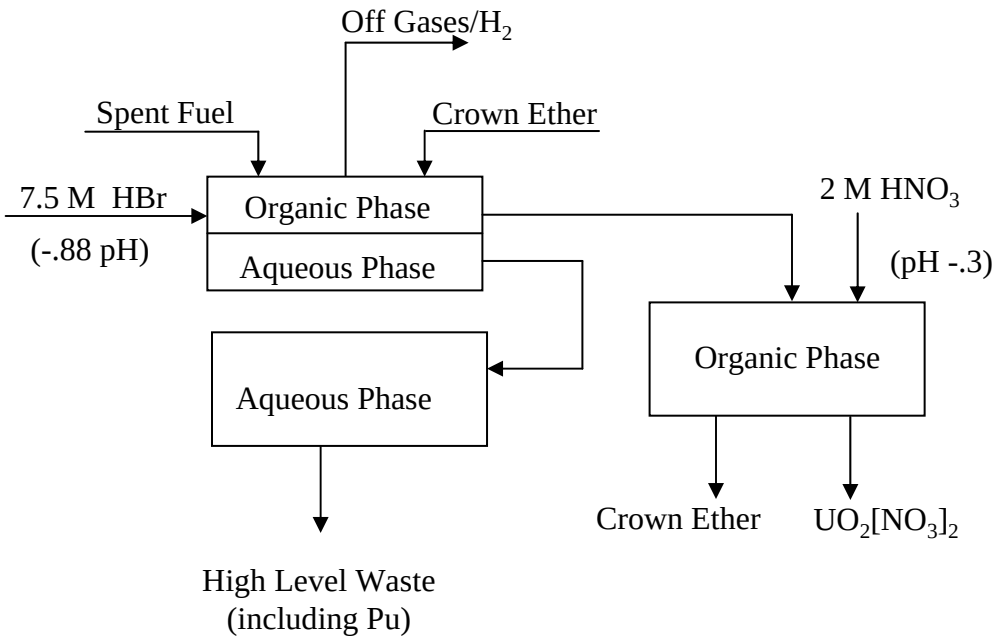


Special Thanks To!

- Dr. Glatzhofer
  - University of Oklahoma
- Dr. Nicholas
  - University of Oklahoma
- Dr. Taylor
  - University of Oklahoma
- Dr. Morvant
  - University of Oklahoma

Questions?

Proposed PFD



Why change Purex?

- Nuclear Proliferation
  - Produces weapons grade Plutonium
- Currently designed to separate U and P.
  - 30% TBP-Solvent
- Liquid-Liquid Extraction
  - Highly inefficient
  - Requires multiply recycle streams
- HLW
  - Produces large quantities of HLW disposal