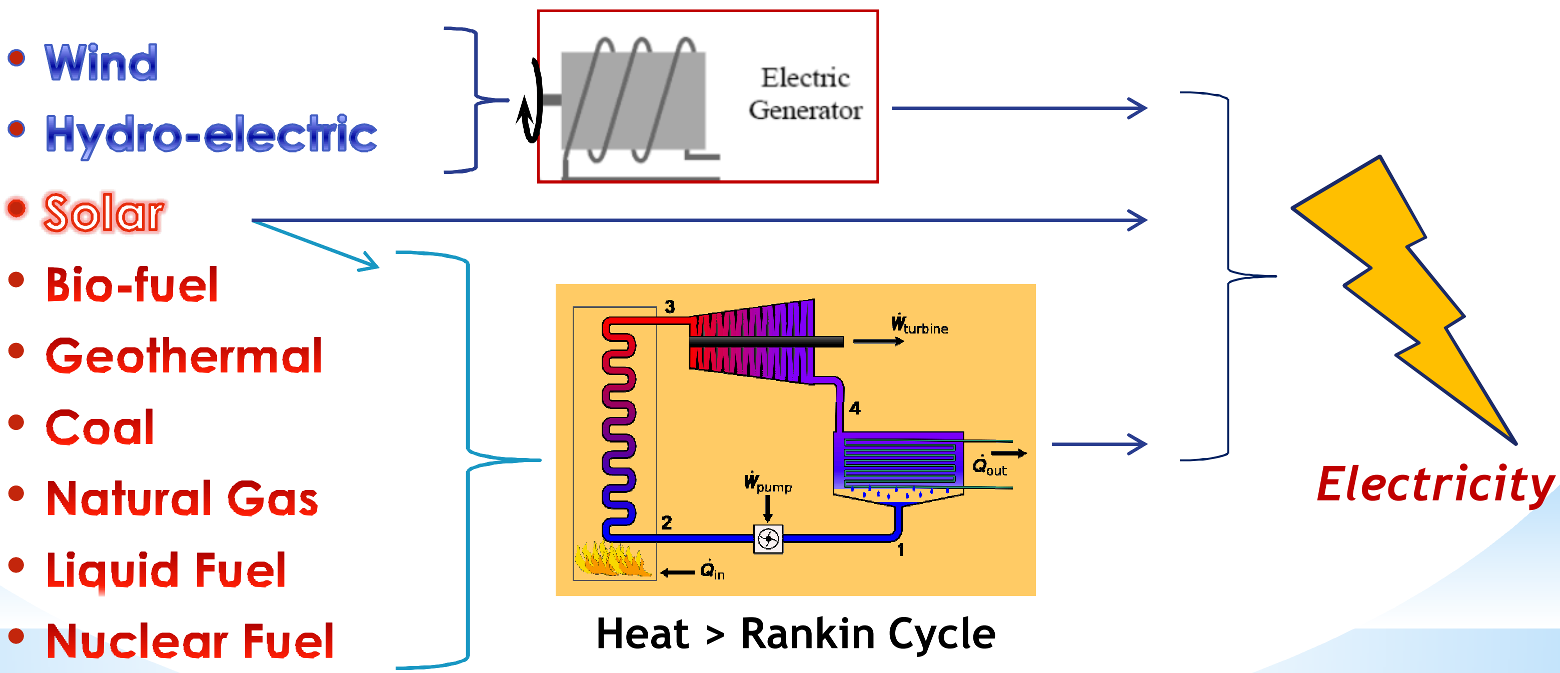
SAND2013-1038P

Nuclear Fuel Cycle

Dr. Robert Finch

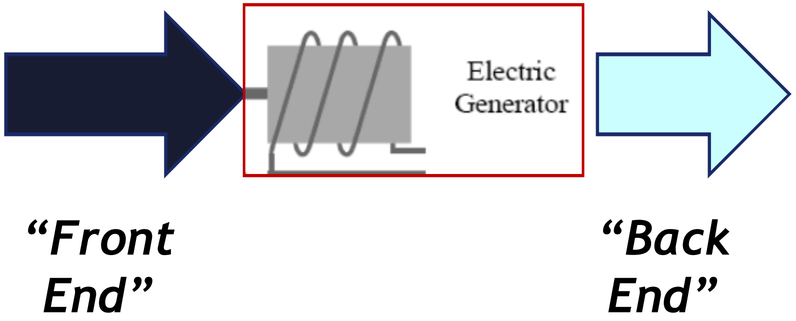
What is a “Fuel Cycle”?

- What generates the power (or heat) to produce electricity?
  - What is its source?
  - How is it used?
  - What happens after it is used?



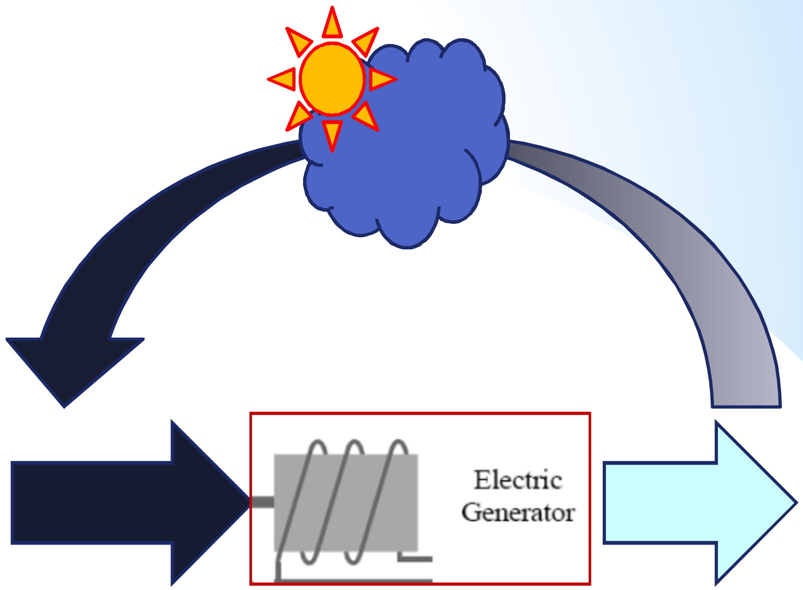
Generic Fuel Cycle

- Source
  - Where does it originate?
  - How is it extracted?
- Treatment
  - Can it be used directly?
  - Are manufacturing or other processes necessary?
- Use
  - Engineered power-generation system
- Waste stream
  - What happens after it can no longer generate electricity (if applicable)?
- Disposal
  - What to do with the waste product (if applicable)?



Renewable Energy Cycle

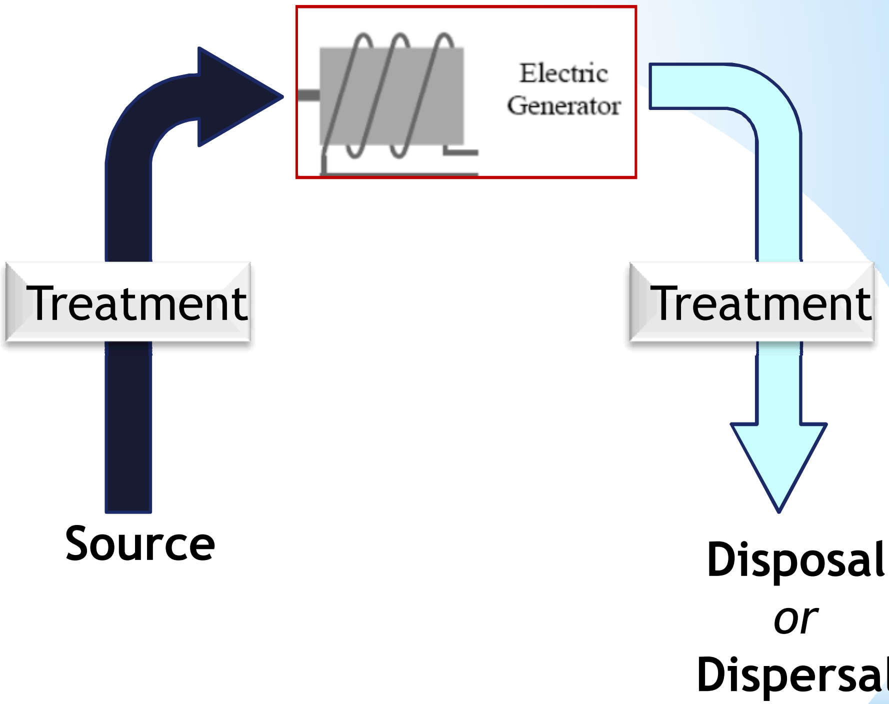
- Source
  - Regenerated naturally
    - Inexhaustible
  - Little or no resource management
- Treatment
  - Variable, but commonly used directly
    - water, wind, photovoltaic
- Use
  - Mechanical Energy
    - Wind & Hydro-electric
  - *Heat > Rankin Cycle*
    - Solar & Geothermal heat
  - *Photovoltaic*
    - Direct conversion of solar energy
- Waste stream
  - Negligible or non-toxic
- Disposal
  - Not applicable



- **Wind**
- **Hydro-electric**
- **Solar**
- **Geothermal**
- **Some Bio-fuels**

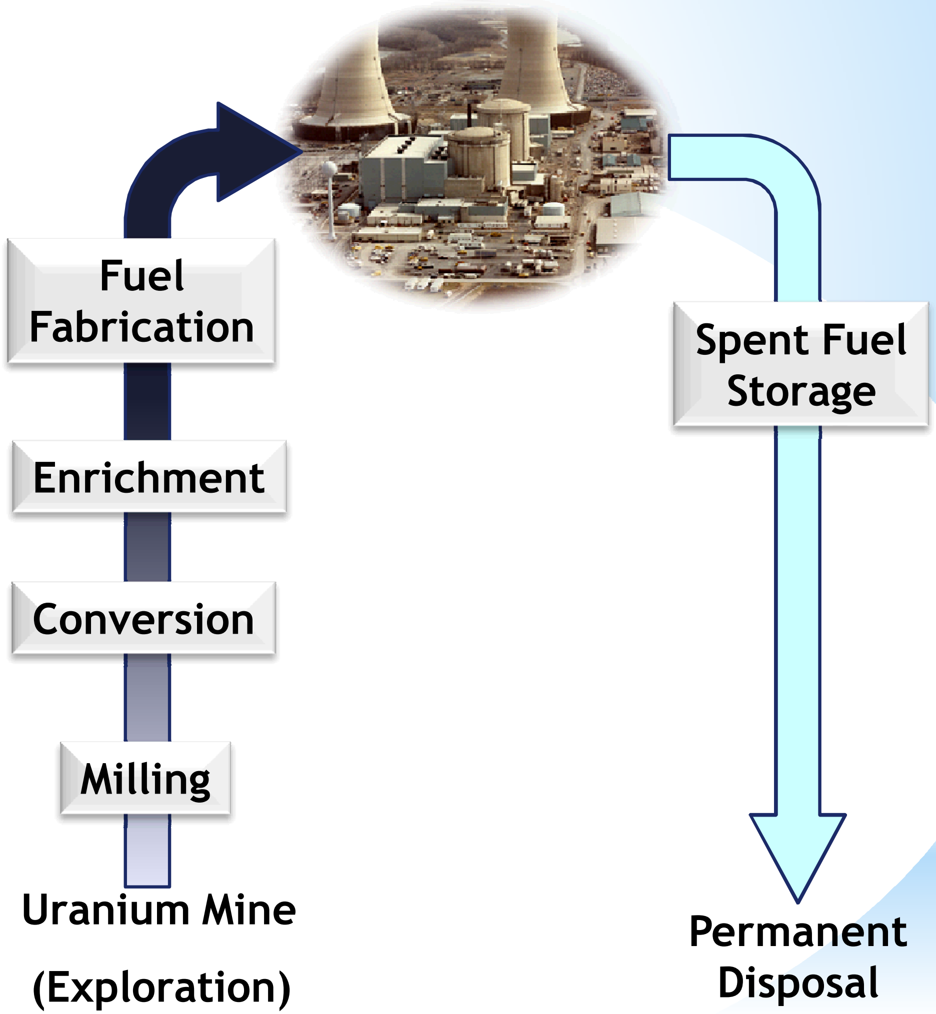
Non-Renewable Energy Cycle

- Source
  - Mine, well, farm, etc.
  - Must be managed
  - Exhaustible
- Treatment
  - Generally must be processed before use in a power plant
- Use
  - Rankin Cycle most common
- Waste stream
  - Variable (solid, liquid, gas)
  - Commonly toxic
- Disposal
  - Depends on waste stream and relative toxicity
  - Treatment is common
  - Burial is common



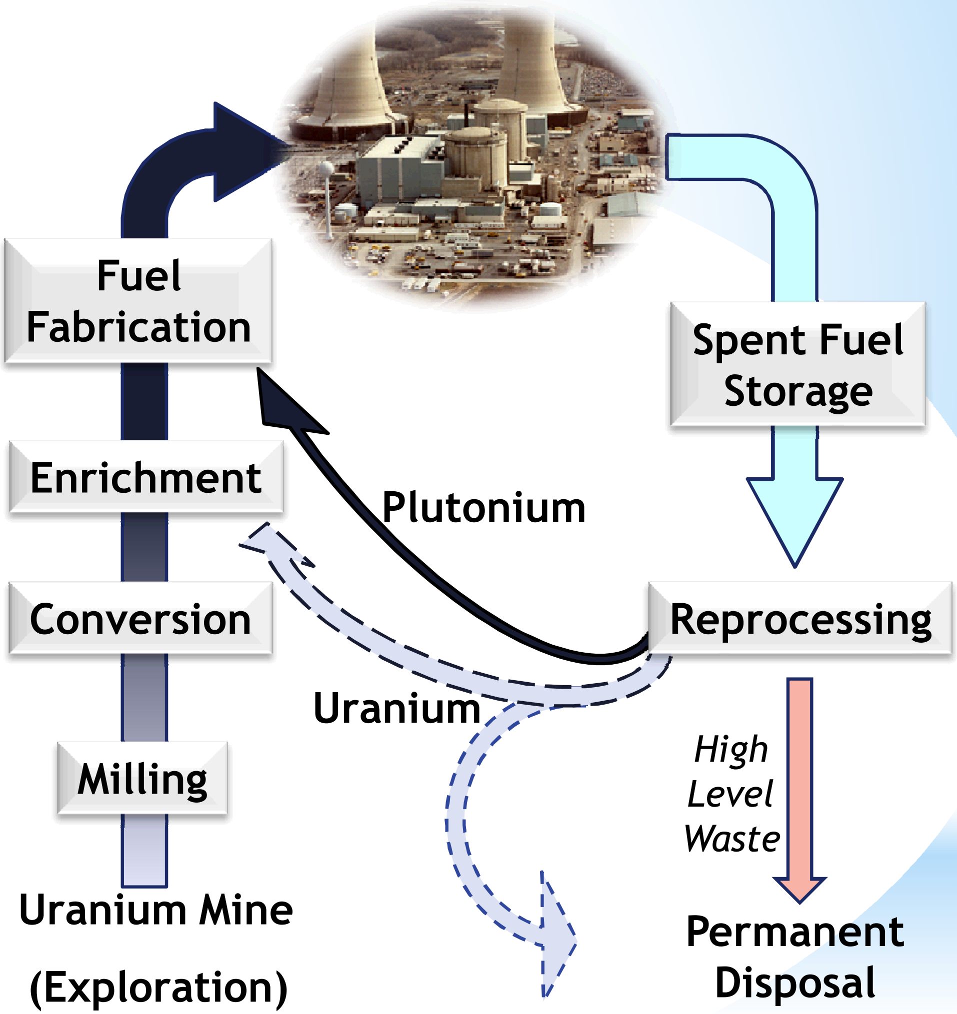
Nuclear Fuel Cycle: “Open” (Once-Through)

- Source
  - Mined
- Treatment
  - Milling
  - Conversion
  - Enrichment
  - Fabrication
- Use
  - Nuclear Reactor & Rankin Cycle
- Waste stream
  - Primarily solid
  - Highly toxic
- Disposal
  - Interim waste management
  - Deep Burial



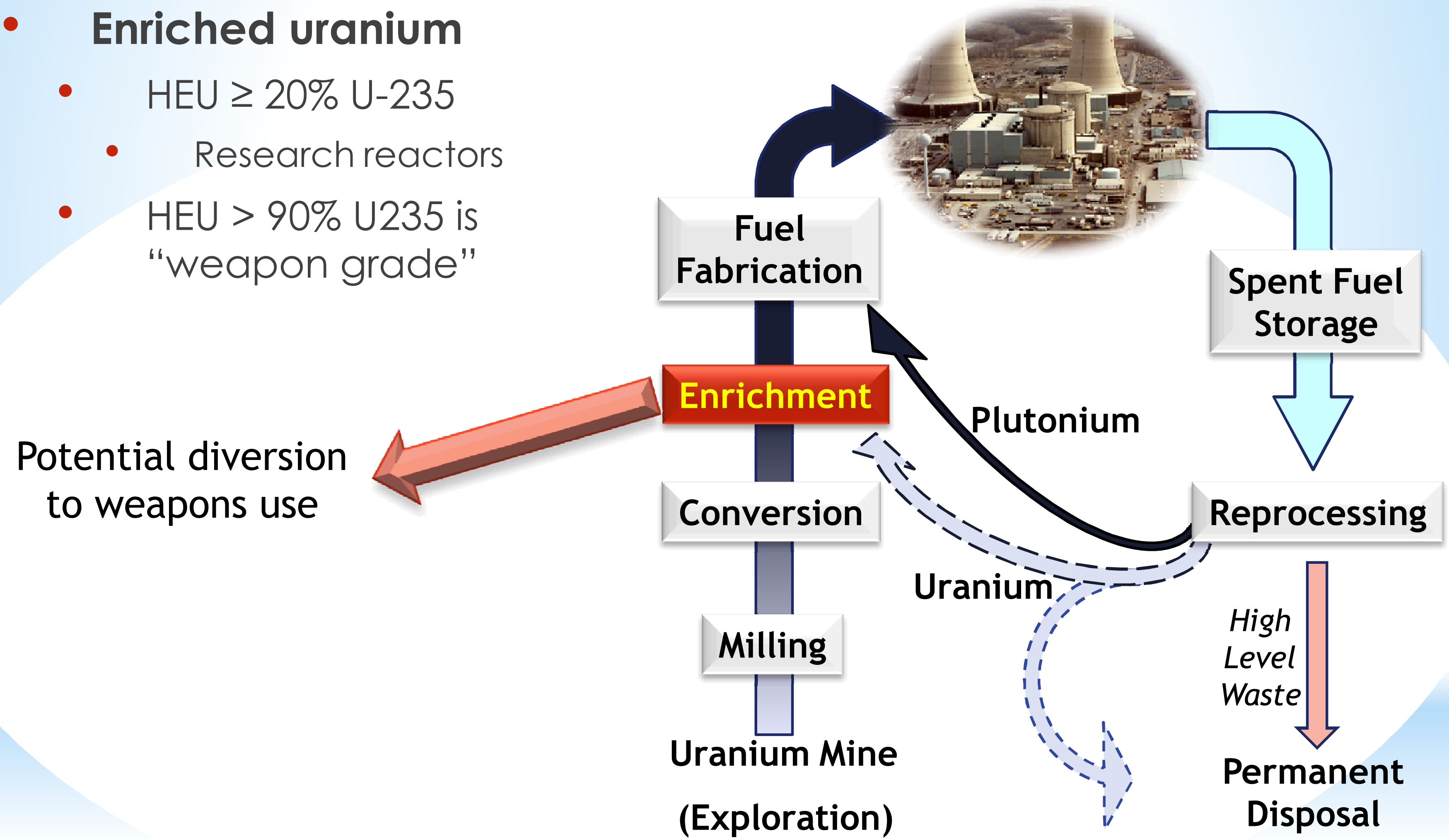
Nuclear Fuel Cycle: "Closed"

- Source
  - Mined
    - Reduced demand
- Treatment
  - Milling
  - Conversion
  - Enrichment
  - Fabrication
- Use
  - Nuclear Reactor & Rankin Cycle
- Waste stream
  - Solid & Liquid
  - Highly toxic
- Disposal
  - Interim storage
  - Reprocessing
  - Recycle (Pu, ±U)
  - Deep Burial (HLW)



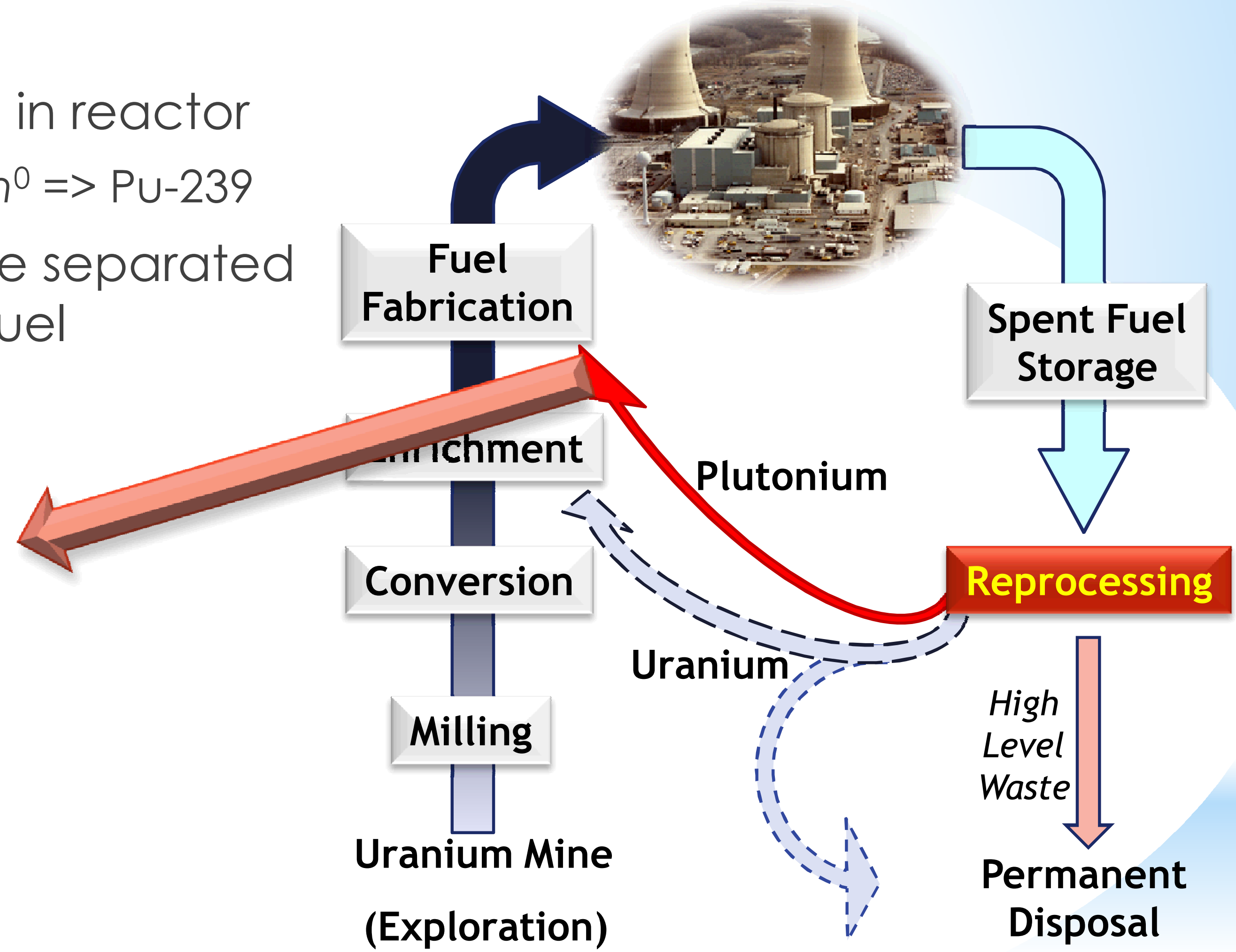
Special Nuclear Materials & Safeguards

- **Enriched uranium**
  - HEU ≥ 20% U-235
    - Research reactors
  - HEU > 90% U235 is "weapon grade"



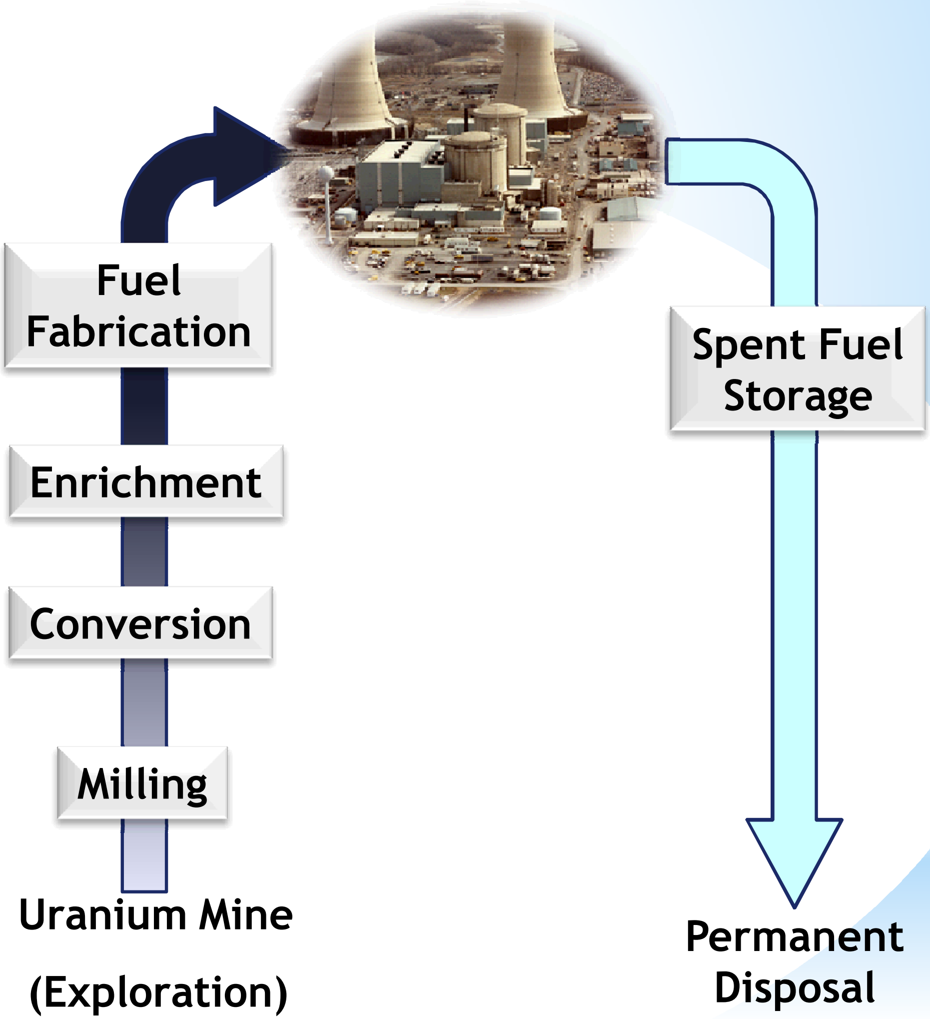
Special Nuclear Materials & Safeguards

- **Plutonium**
  - Produced in reactor
    - U-238 + n^0 => Pu-239
  - Pu must be separated from the fuel



Uranium Fuel Cycle & Light Water Reactors

- Most nuclear-power reactors are fueled by uranium
  - Uranium must be mined & processed before use
  - Thorium can also be used
- Most nuclear-power reactors are LWRs
  - Moderated & cooled by H_2O
  - "Thermal" Reactors
  - Most thermal power reactors use *uranium* oxide (UO_2) for fuel
  - Natural uranium can also be used
    - Heavy-water moderated (CANDU)
    - Graphite moderated (MAGNOX)



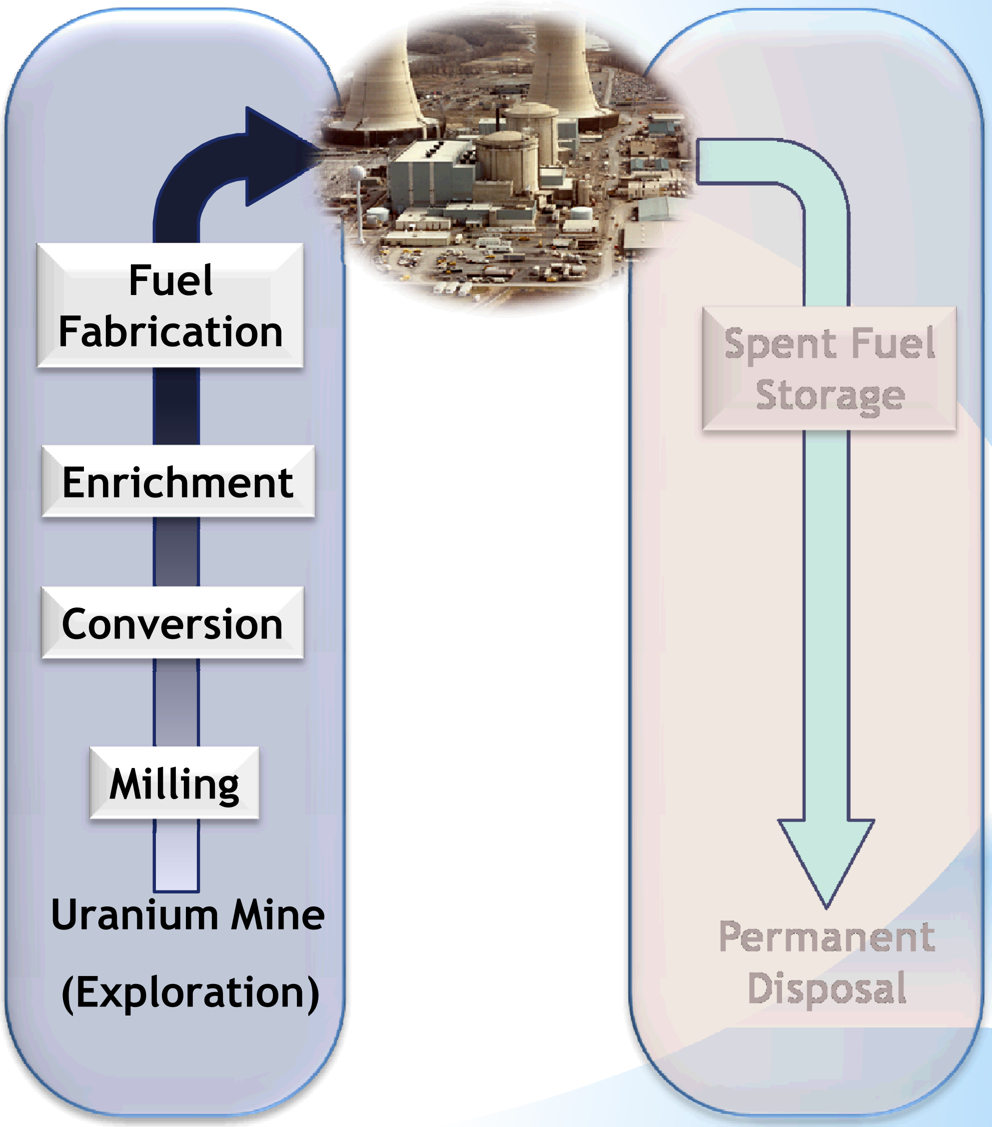
Uranium Fuel Cycle & Light Water Reactors

Front End

- Processes required to obtain, treat and manufacture nuclear fuel for use in a reactor

Back End

- Manage spent fuel and related waste streams
  - In-reactor changes to fuel during reactor operation
- Final waste disposal

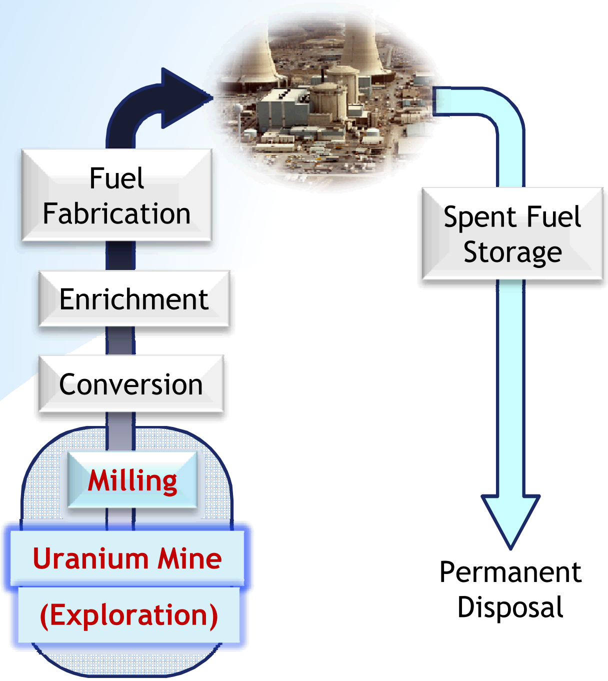


Uranium Ore to Uranium Fuel

U-235 content

Uranium Ore (0.7%)

Fuel Pellet (3.5%)



Uranium Mining & Milling

Uranium Exploration

- Radiation surveys
  - Airborne surveys
  - Surface surveys
- Geological studies
  - Knowledge about likely uranium-bearing rocks
- Hydro-chemical methods
  - Higher-than-average uranium concentrations dissolved in surface or ground waters
- Well logging
  - Drilling in promising geological terrain (expensive)
- Botanical methods
  - Some plants can concentrate uranium in their stems, leaves and roots

Sources of Uranium

- Uranium ore
  - Uranium that can be *economically* extracted
- Uranium is commonly associated w/organic matter
  - *Black shale* and *lignite* deposits
- Uranium recovered as a *by-product*
  - *fertilizer production*
    - *phosphorite deposits*

- Uranium assay
  - The amount of uranium in uranium ore
  - Usually reported as "U_3O_8"
    - U_3O_8 does not actually occur naturally; it is a man-made product

Uranium Supply

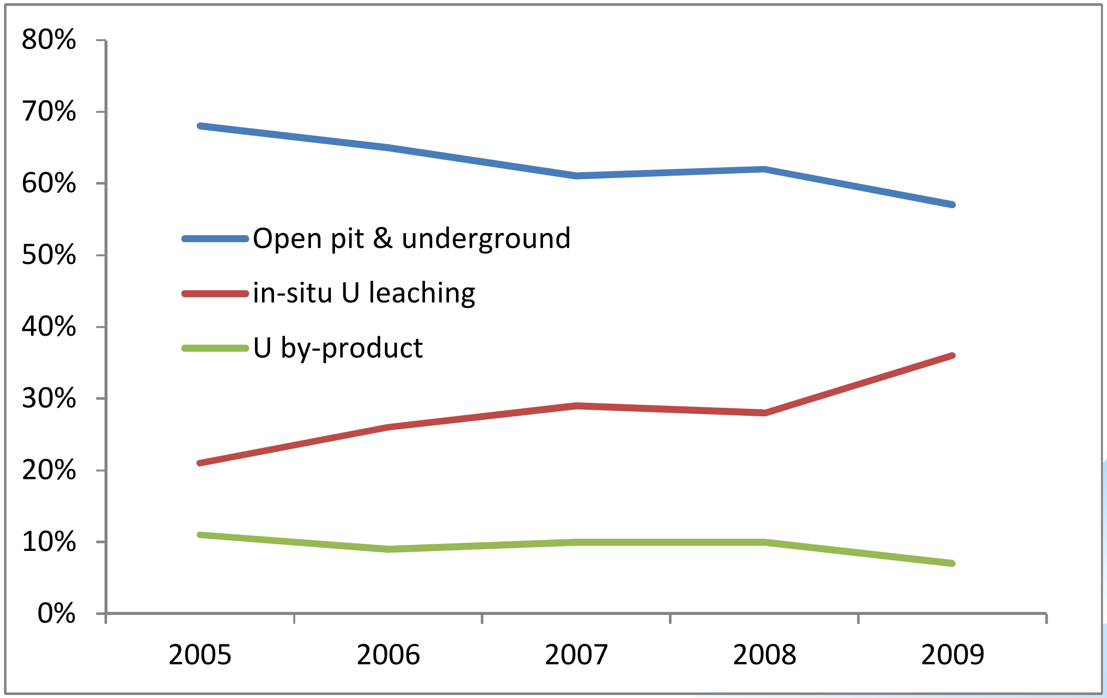
- Current known resources of uranium worldwide: **~5.4 million tonnes**
  - "Reasonably Assured Resources"
- Current *annual* demand in nuclear-power reactors
  - Approximately 68,000 tonnes (each year)
- Approximately 80 years of uranium remaining from known resources
  - Additional sources may appear with increased exploration
  - Higher prices can increase economically recoverable resources
  - Motivates some countries to recover fissile content (Pu & U) from spent fuel
    - "Closed Fuel Cycle"

Top ten uranium-producing countries:

1. Australia
2. Canada
3. Kazakhstan
4. Niger
5. United States
6. South Africa
7. Brazil
8. Russia
9. China
10. Ukraine

Uranium Mining

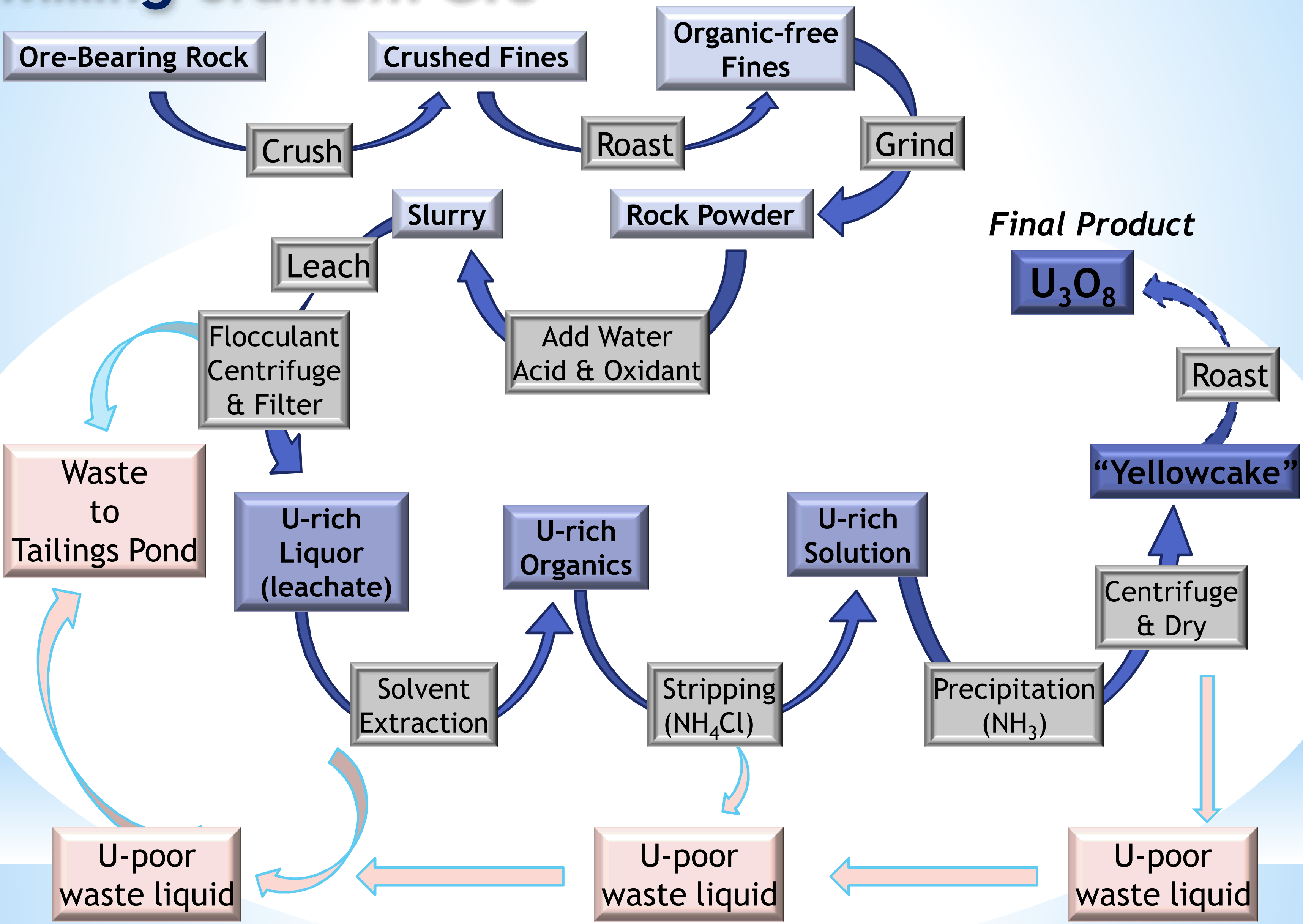
World Uranium Production (2009)	
Open pit & Underground	57%
In-situ leach (ISL)	36%
By-product	7%



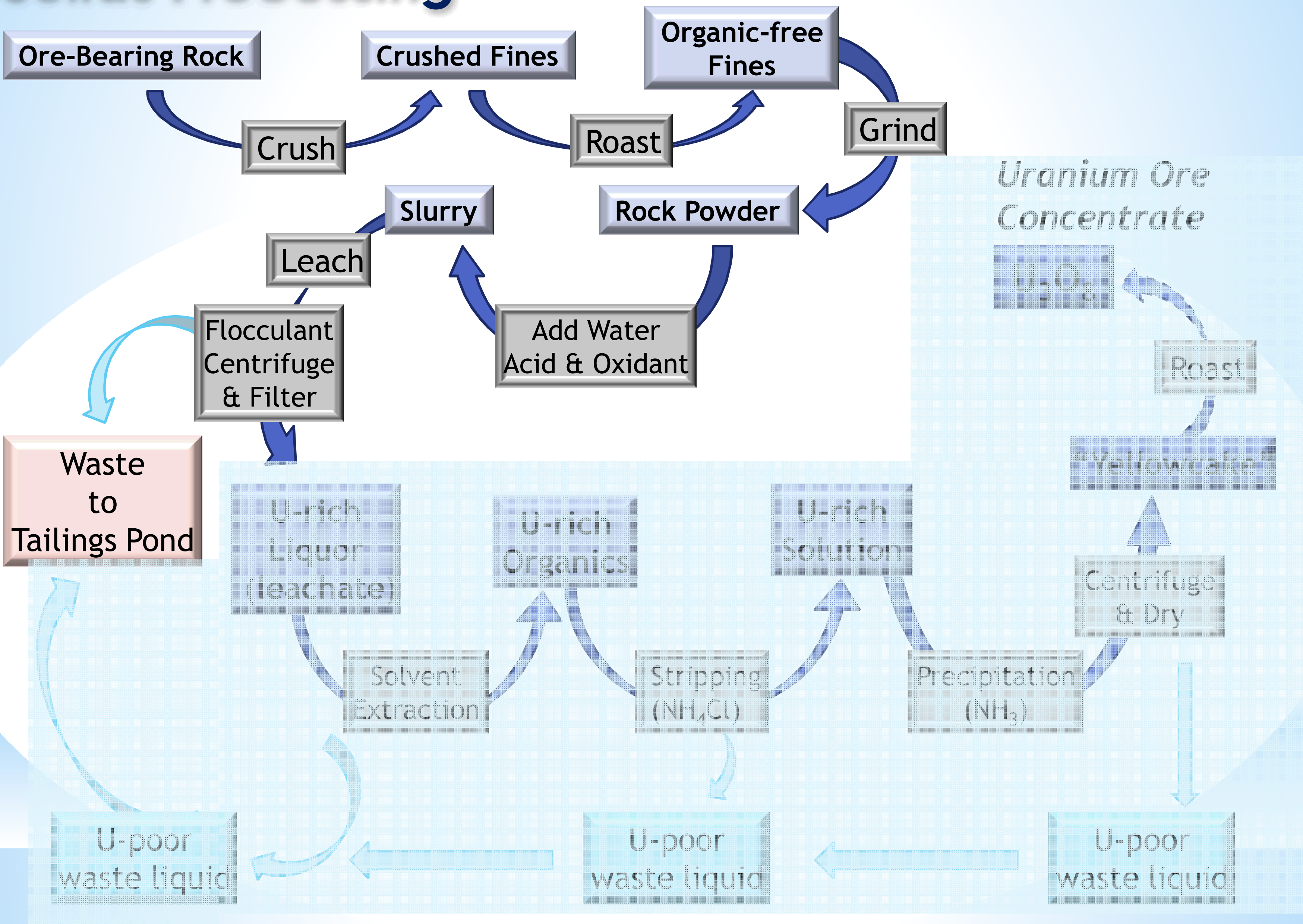
Open Pit Mining

Rossing mine in Namibia

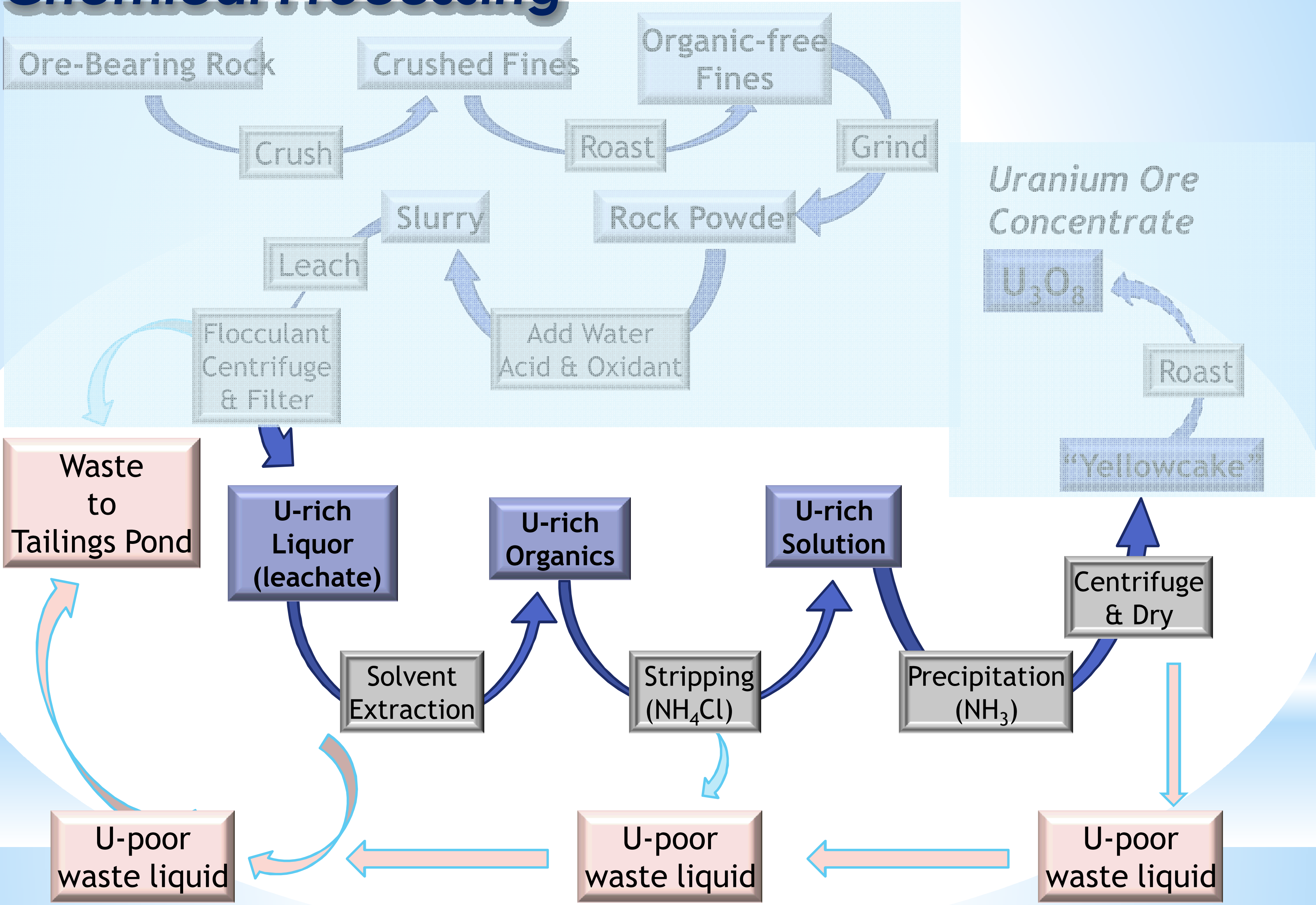
Milling Uranium Ore



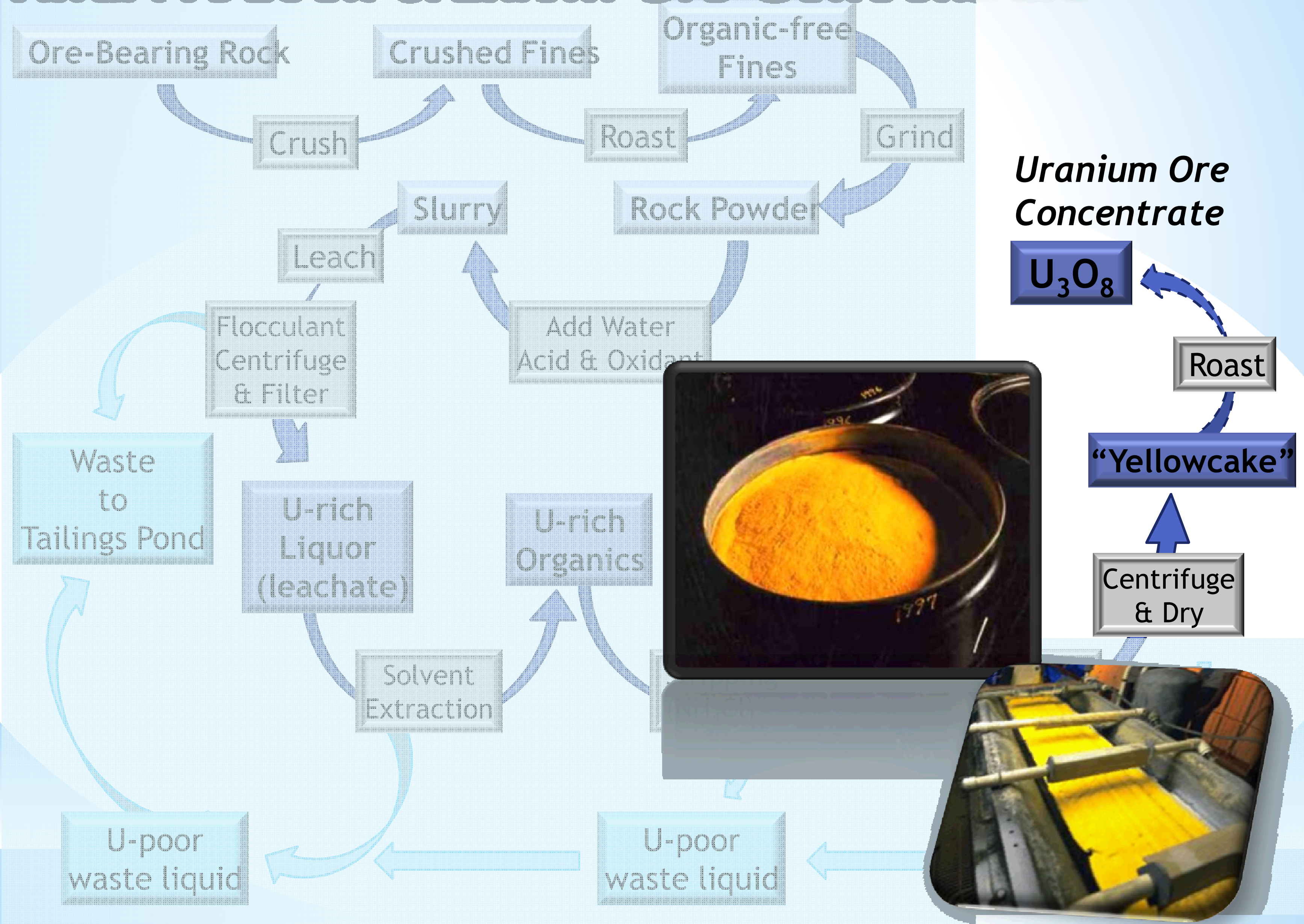
Solids Processing



Chemical Processing



Final Product: Uranium Ore Concentrate



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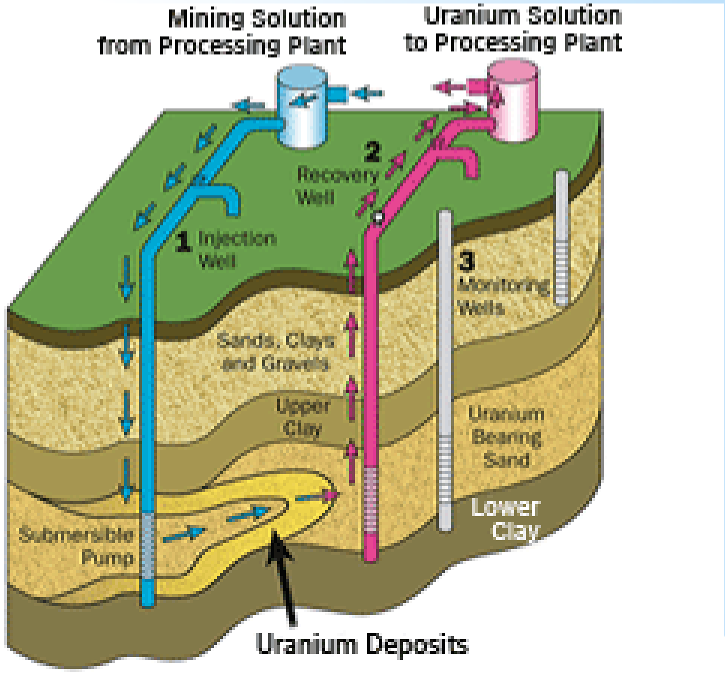
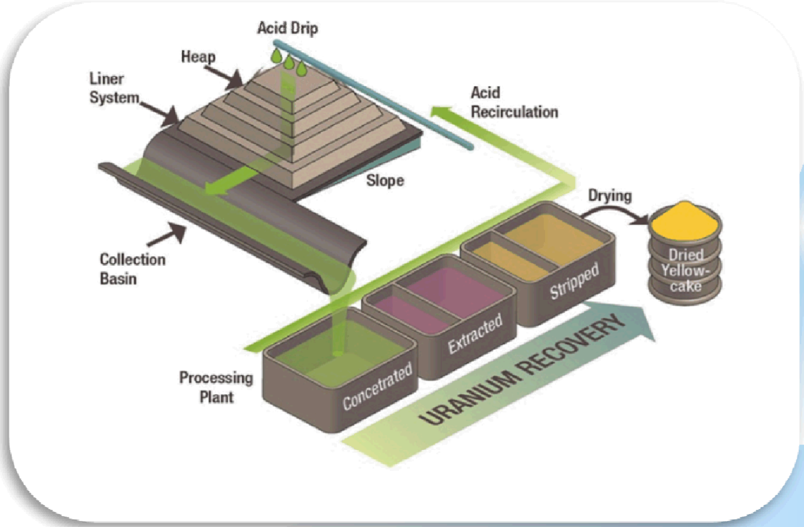
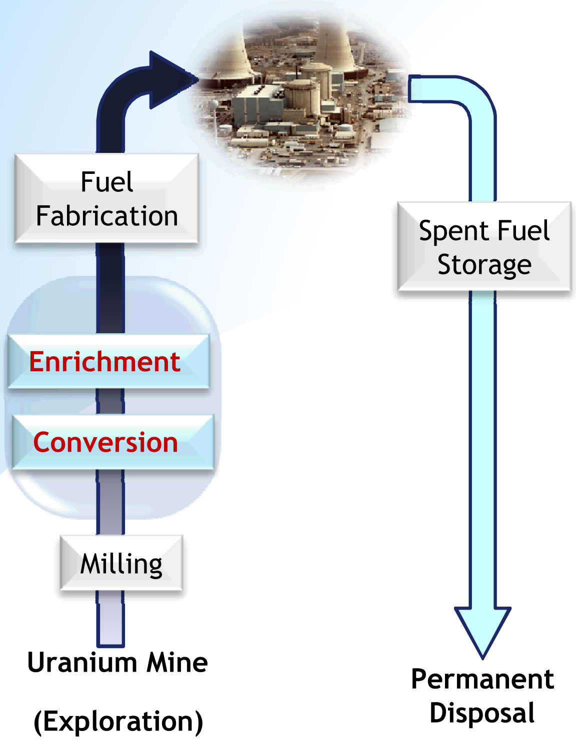


- Uranium milling procedures depend on the mining method
  - In-situ leaching does not require solids processing such as does open-pit and underground mining
  - Uranium is chemically dissolved out from the rocks underground and recovered directly in solution
  - Uranium ore concentrate is produced from the uranium-bearing solution



Heap Leach Recovery



Uranium Conversion & Enrichment

Uranium Conversion and Enrichment

LWRs require uranium fuel *enriched* in U-235

- Maintain a chain reaction with *thermal* neutrons
  - Some reactor types do not require enriched uranium
- Isotopes are chemically similar – use physical separation
  - Slight mass differences between U-235 & U-238

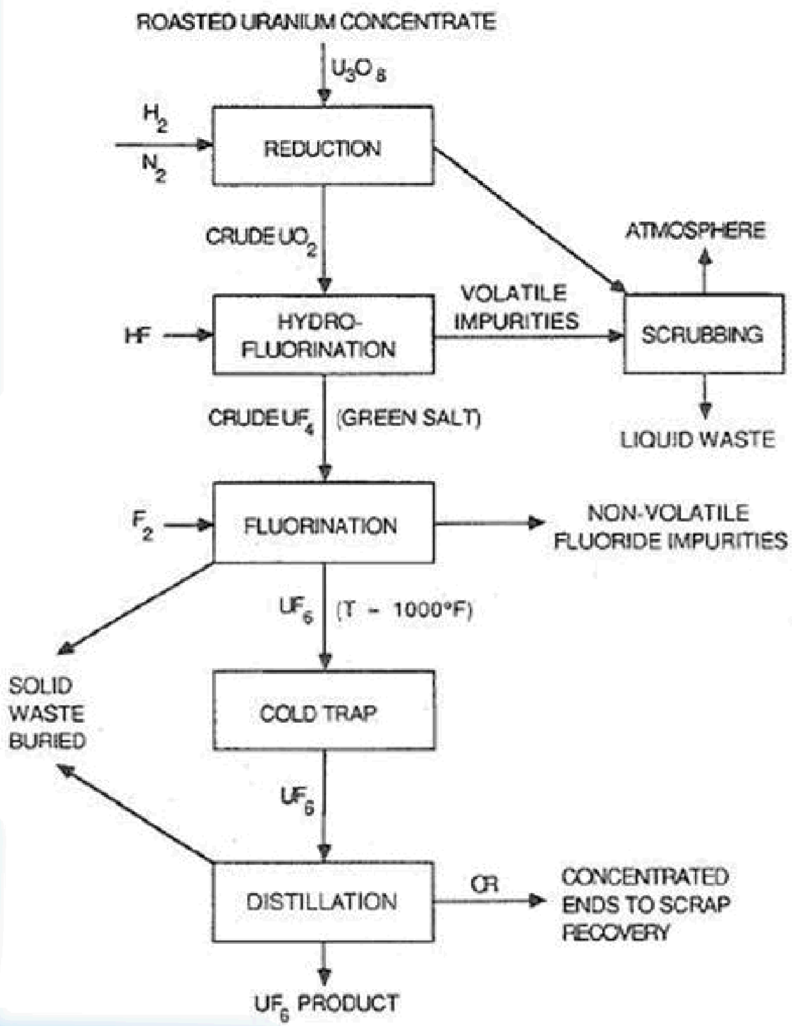
***Convert* uranium ore concentrate into more volatile form**

- Gaseous uranium can be enriched by using several processes
- Uranium ore concentrate is a non-volatile solid

Proliferation concerns

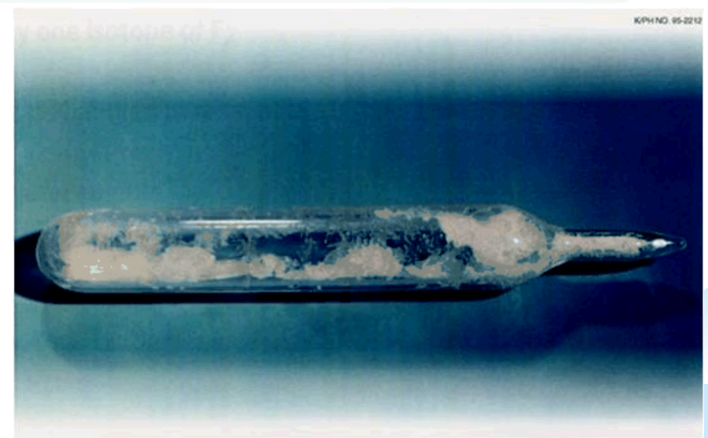
- Enrichment is a "sensitive technology"
- Nuclear weapons also require enriched U
  - Much higher level of enrichment than needed for commercial reactors
- International safeguards & export controls

Uranium Conversion



UF_4
(Green Salt)

1. **Reduction**
   $U_3O_8 + H_2 = UO_2 + H_2O$
2. **Conversion to tetrafluoride**
   $UO_2 + HF = UF_4 + H_2O$
3. **Oxidation to hexafluoride**
   $UF_4 + F_2 = UF_6$



UF_6

Uranium Isotopes

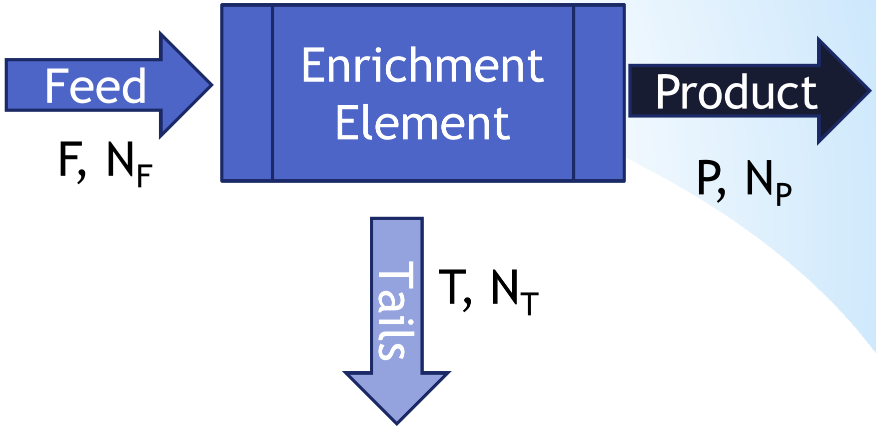
Natural Uranium

- ^{238}U (99.2745%)
- ^{234}U (0.0055%)
- ^{235}U (0.72%)
    - Fuel for Light Water Reactors (LWRs)
        - ^{235}U/ U(total) ~3.5 -5%
            - Five (5) to seven (7) times higher than in natural uranium

- Natural uranium can also be used as nuclear fuel
    - Heavy-water-moderated reactors
        - CANDU reactors
    - Graphite-moderated reactors
        - MAGNOX reactors

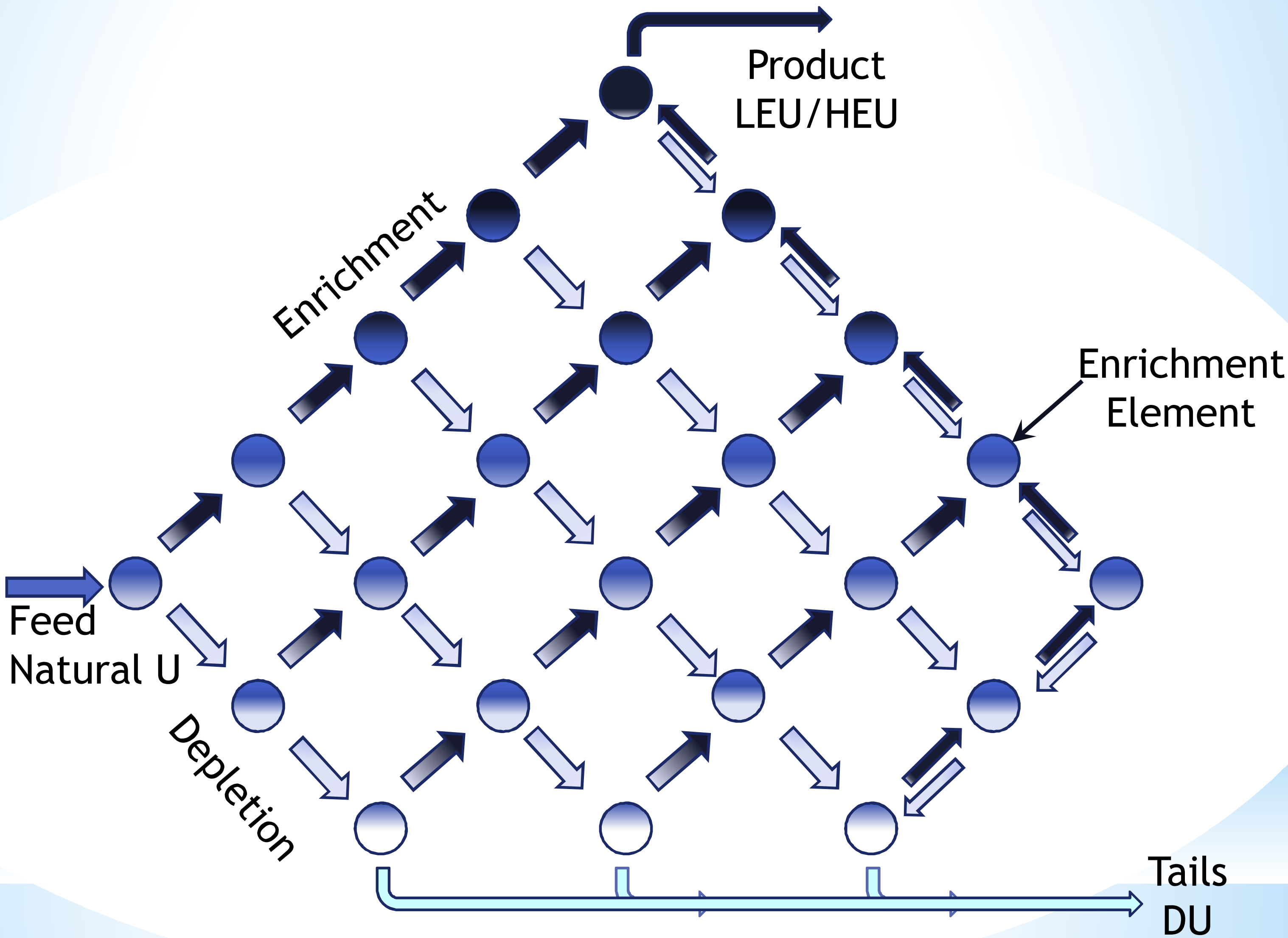
Enrichment Element

- Mass balance: $F = P + T$
- Feed
  - $N_F = \text{U-235}/(U_{Feed})$
- Product
  - $N_P = \text{U-235}/(U_{Product})$
- Tails
  - $N_T = \text{U-235}/(U_{Tails})$
- $N_P > N_F > N_T$



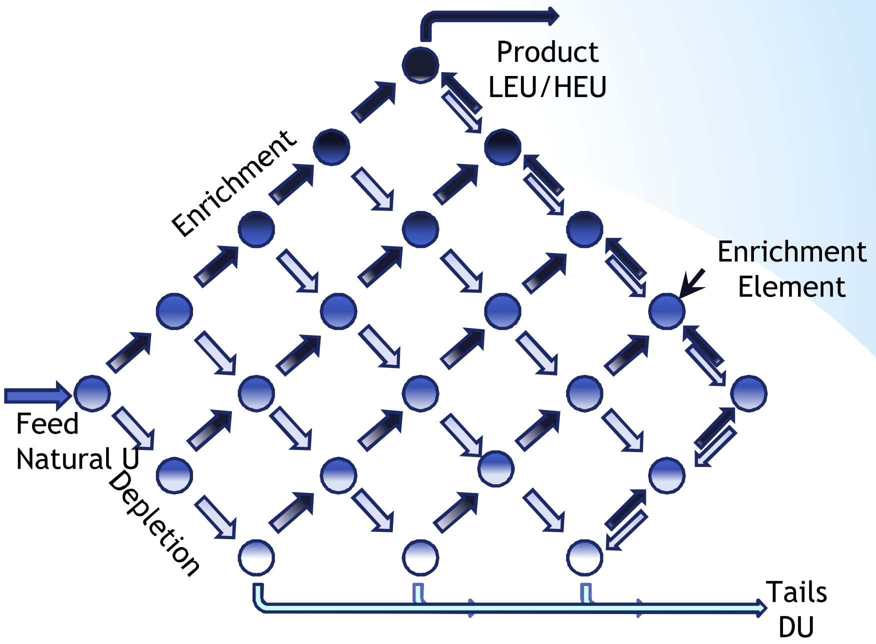
U-235 ≈ 1-U-238

Schematic Enrichment Cascade



Uranium Enrichment

- Input = Feed
  - Natural Uranium (NU)
  - 0.71% U-235
- Output = Product
  - Enriched Uranium
  - < 20% U-235 (LEU)
  - > 20% U-235 (HEU)
- Waste = Tails
  - Depleted Uranium
  - ~0.2% U-235 (DU)



Separating the uranium isotopes requires work!

Separative Work Units and Value

- The *Product value* is the Product mass times the value function for the Product stream

Product value = $\text{Mass}_{\text{Product}} \cdot \text{Value}_{\text{Product}}$

  - Feed Values and Tail Values are calculated the same way

- A ***Separative Work Unit*** (or **SWU**) is the sum of Product and Tail values *minus* the Feed value:

SWU = (*Product Value*) + (*Tails Value*) – (*Feed Value*)

$$\text{SWU} = \text{Mass}_{\text{Prod.}} \cdot \text{Value}_{\text{Prod.}} + \text{Mass}_{\text{Tails}} \cdot \text{Value}_{\text{Tails}} - \text{Mass}_{\text{Feed}} \cdot \text{Value}_{\text{Feed}}$$

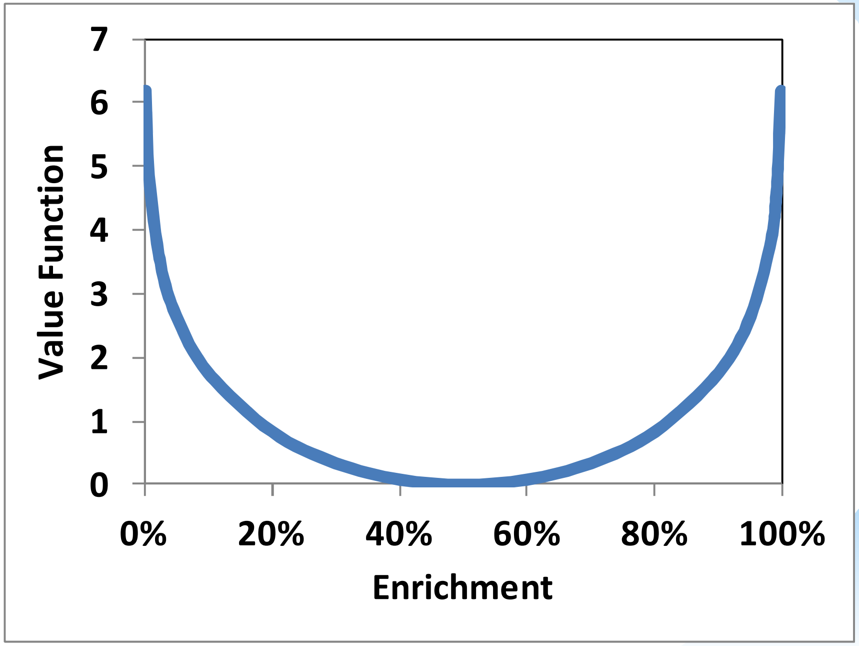
Separative Work and Value

The "Value" of an isotopic mixture relates to its Entropy

Value Function = $(2N-1) \cdot \ln[N/(N-1)]$

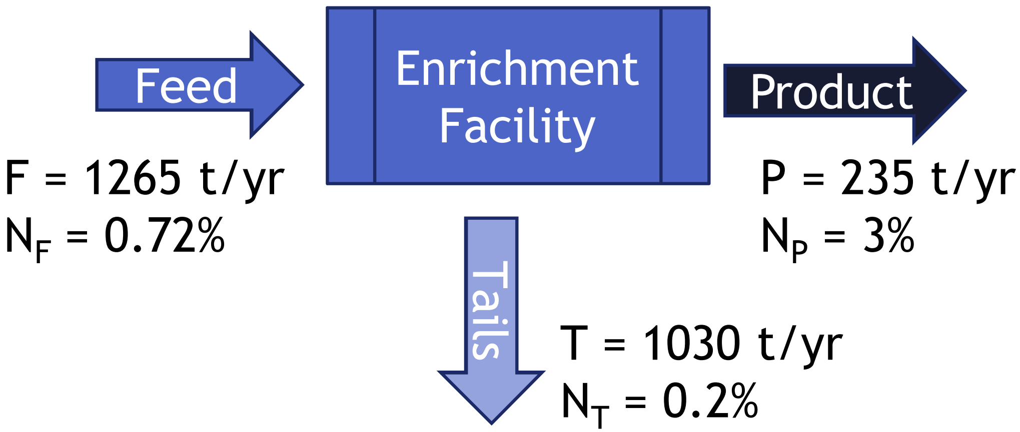
***N* = proportion of U-235 in Feed, Product, or Tails**

- Feed (NU)
  - U-235 = 0.72%
  - $Value_{Feed}$ = 4.86
- Tails (DU)
  - U-235 ~ 0.2%
  - $Value_{Tails}$ = 6.19
- Low-enriched uranium (LEU)
  - U-235 = 5%
  - $Value_{Product}$ = 2.65
- High-enriched uranium (HEU)
  - U-235 = 20%
  - $Value_{Product}$ = 0.83



Generic Enrichment Facility

- An *Enrichment Facility* can be described by its total *enrichment capacity*
  - E.g., Tonnes-SWU per year

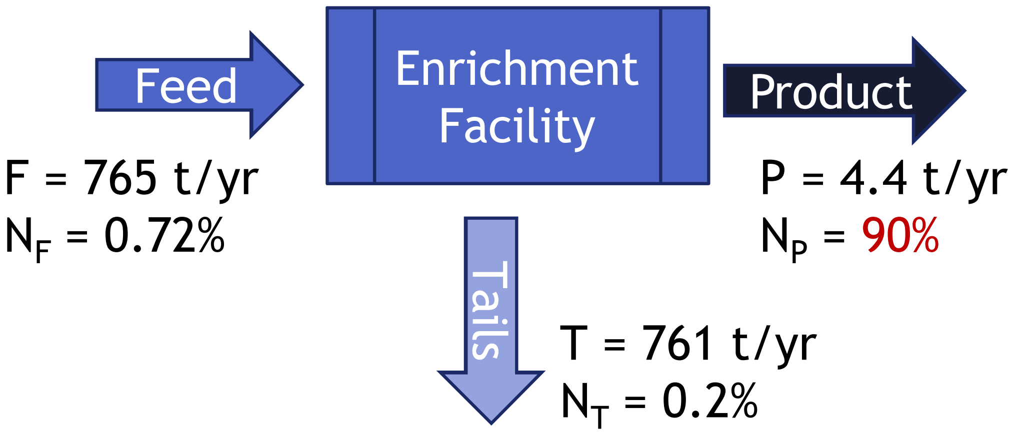


$$SWU = Mass_{Prod.} \bullet Value_{Prod.} + Mass_{Tails} \bullet Value_{Tails} - Mass_{Feed} \bullet Value_{Feed}$$

SWU ≈ 1,000 tonnes-SWU per year

Generic Enrichment Facility – HEU production

- An Enrichment Facility can also produce HEU
  - Same basic process, but important design changes needed

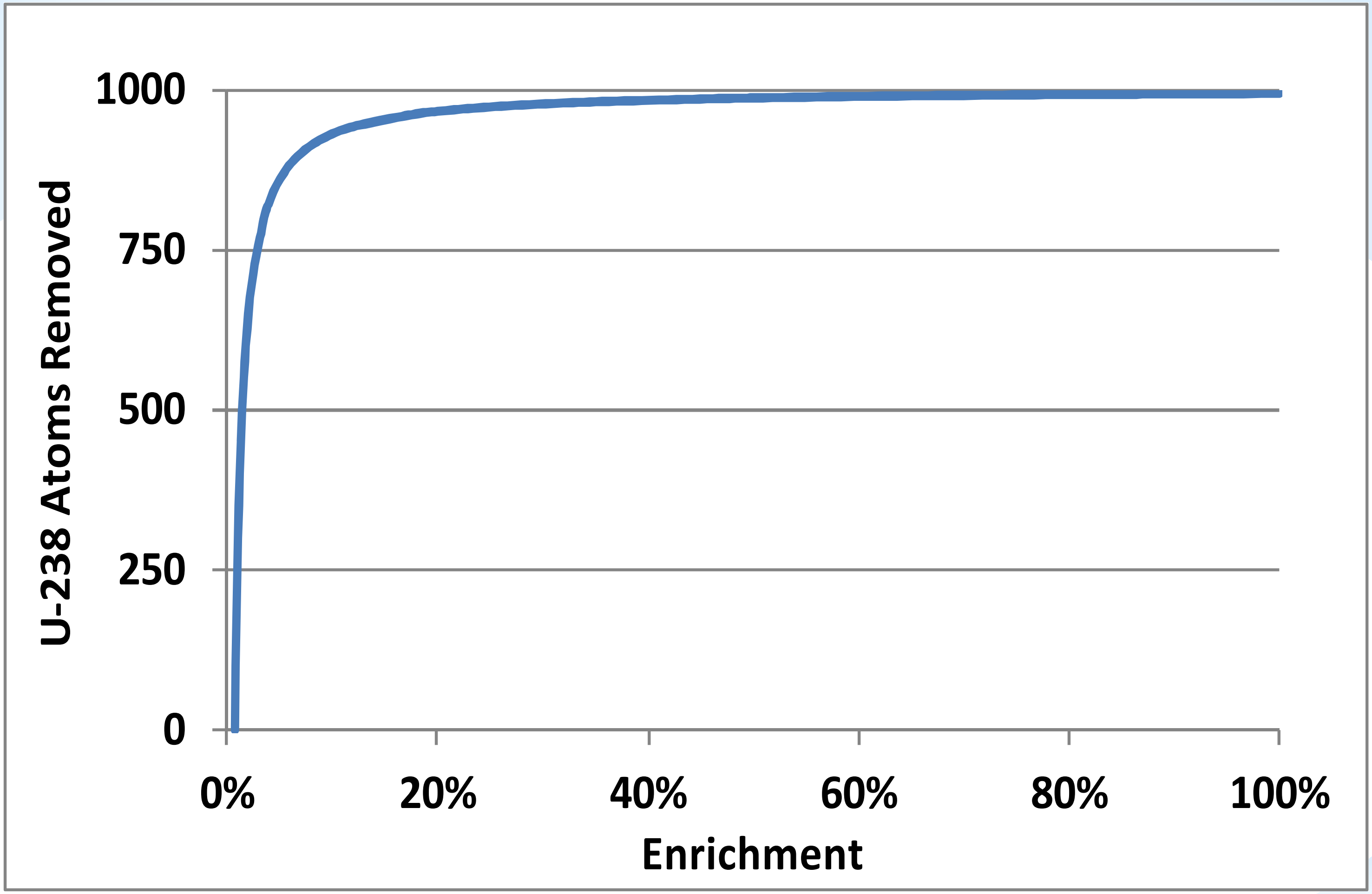


SWU ≈ *1,000 tonnes-SWU per year*

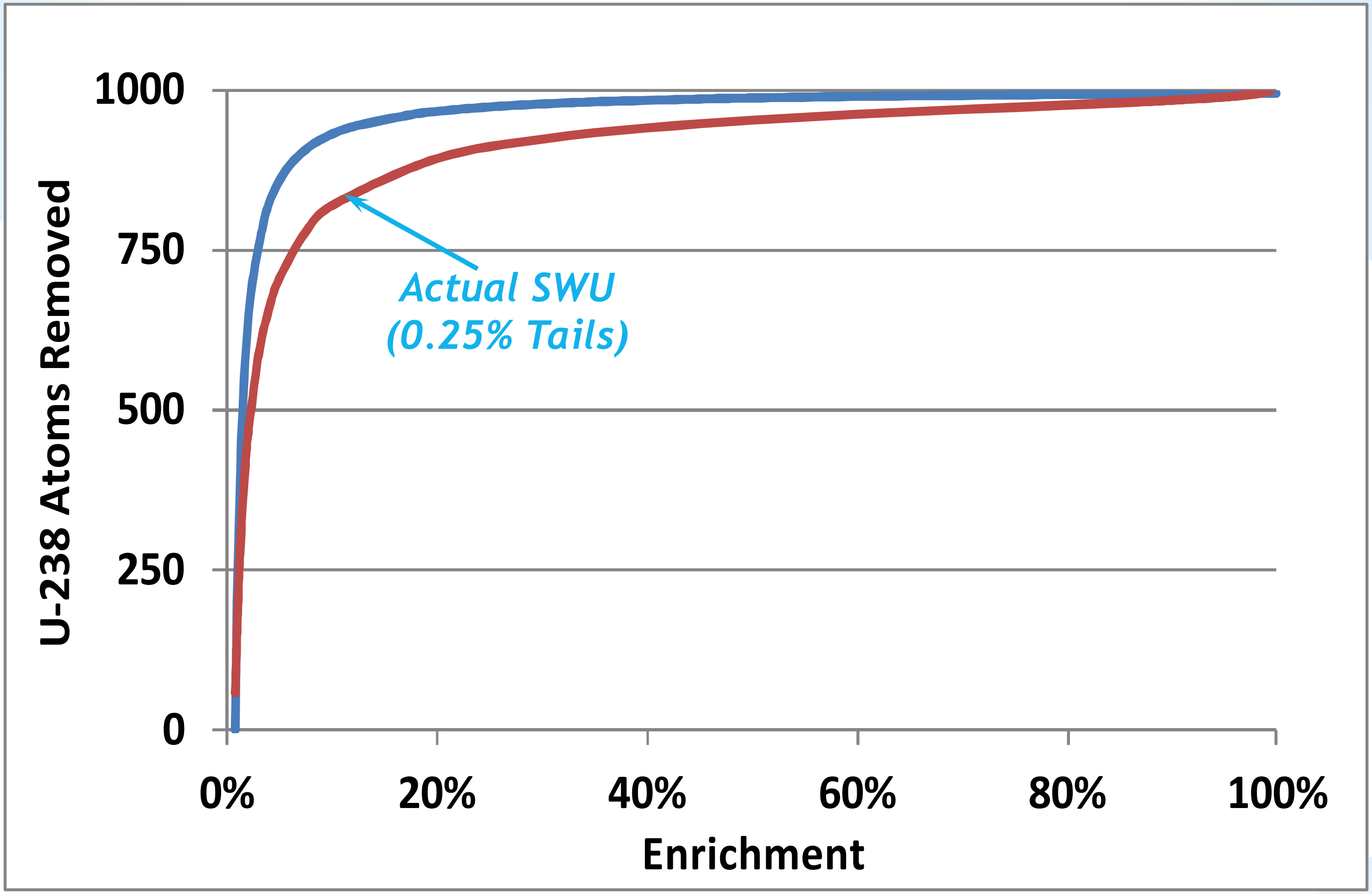
Conceptual Exercise on Separative Work

- Remove *only* U-238 atoms from the Feed
  - No U-235 in the tails → a "perfect" (& fictive) separation process
- Start with 1,000 atoms of Natural Uranium
  - 993 are U-238 (99.3%)
  - 7 are U-235 (0.7%)
- To achieve 5% enrichment from Natural Uranium
  - Must remove **860** U-238 atoms
  - Product: 1000 – 860 = 140 U atoms
    - No change to the original 7 U-235 atoms
  - Enrichment: 7/140 = 5%
- To achieve 20% enrichment (7/35)
  - From 5% LEU: Remove only **105** additional U-238 atoms
- To achieve 87.5% enrichment (7/8)
  - From 5% LEU: Remove **132** additional U-238 atoms
  - From 20% HEU: Remove only **27** additional U-238 atoms

Conceptual Exercise on Separative Work

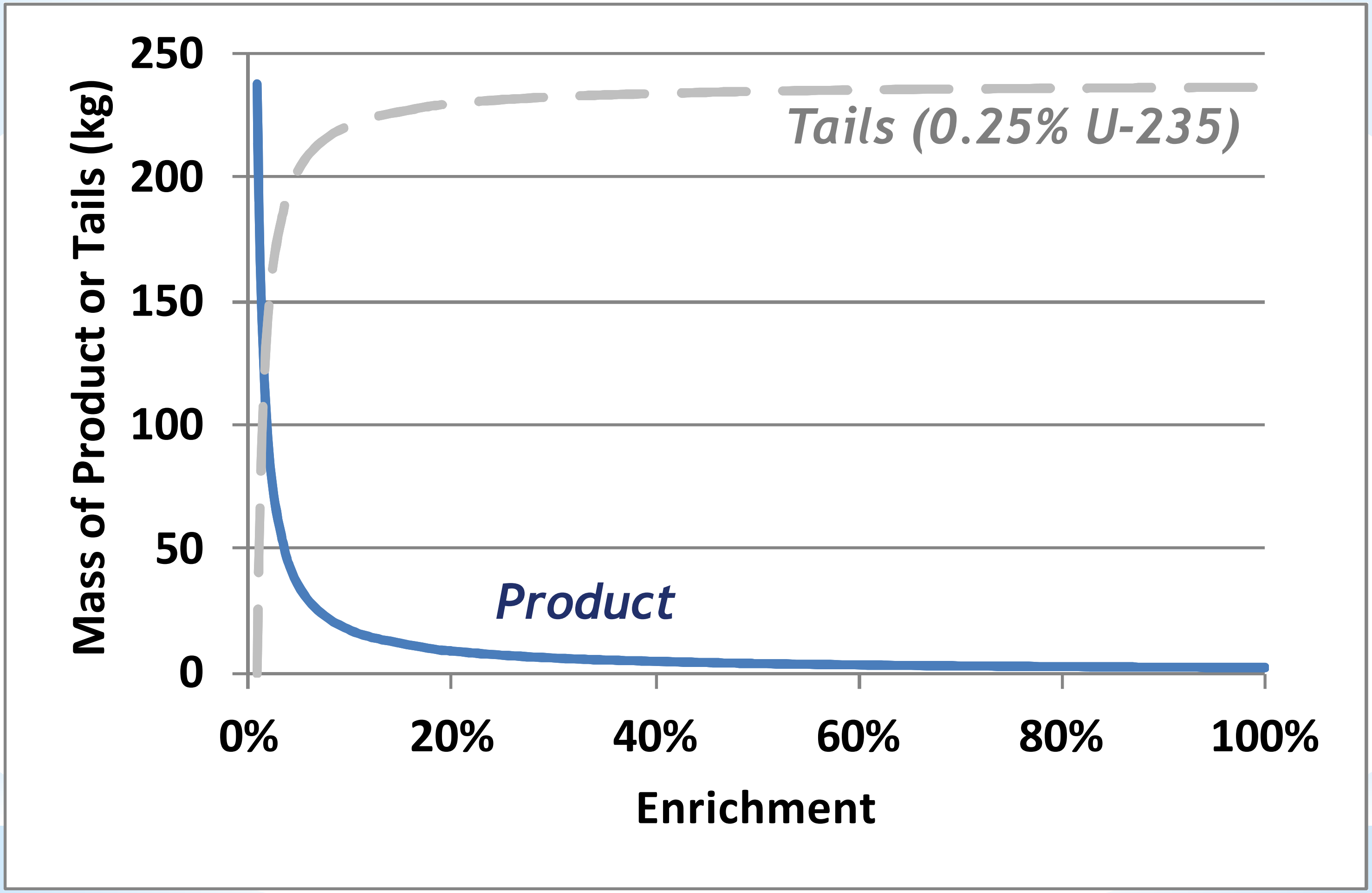


Conceptual Exercise on Separative Work



Conceptual Exercise on Separative Work

For a fixed mass of feed

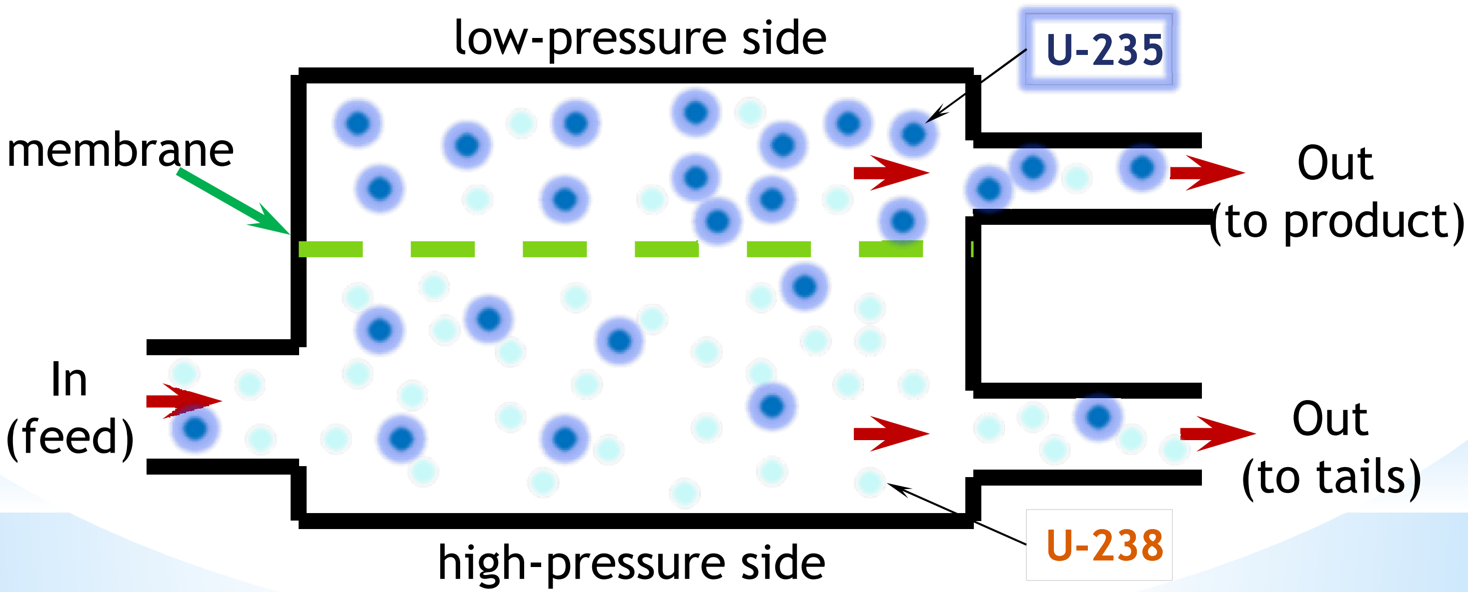


Uranium Enrichment

- Several technologies have been developed to perform isotopic enrichment
  - Gaseous Diffusion
  - Gas Centrifuge
  - Laser – AVLIS, MLIS, SILEX
  - Other – aerodynamic, electromagnetic, chemical

Gaseous Diffusion

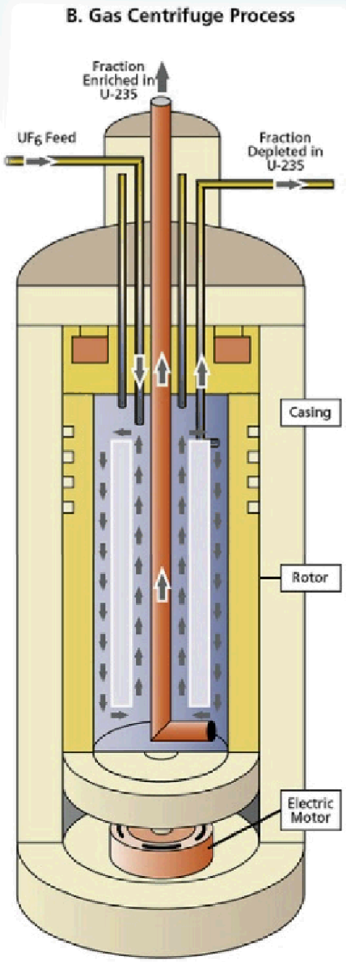
- Pressure difference across a membrane
  - lighter molecules ($^{235}UF_6$) move faster than heavy ones ($^{238}UF_6$)
  - gas passing through barrier slightly enriched in $^{235}UF_6$
- Separation factor 1.003
- 2.3–3.0 MW-hr per SWU



Centrifuge Enrichment

- Gas Centrifuge
- $^{238}UF_6$ experiences stronger *centripetal force* than $^{235}UF_6$
  - $\underline{F} = m\underline{a}$
- Countercurrent flow
  - Outer radius = ^{235}U-depleted stream
  - Inner radius = ^{235}U-enriched stream

Energy used per SWU is much less than that of gaseous diffusion



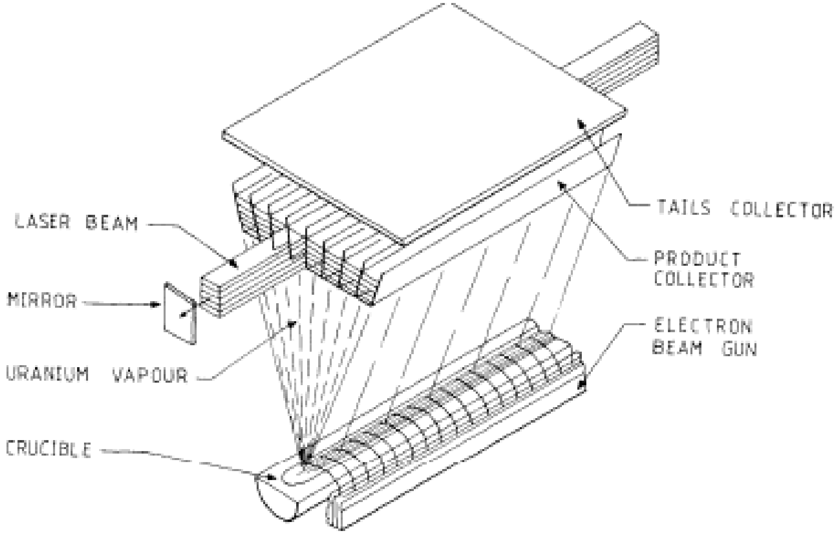
Gas Centrifuge

Laser Isotope Separation

- **Atomic Vapor Laser Isotope Separation (AVLIS)**
- **Molecular Laser Isotope Separation (MLIS)**
- Possible to selectively ionize one isotope
  - Frequency-dependent light absorption
- Ionized gas collected at negatively charged plate

Fewer stages possible

~0.3 MW-hr/SWU

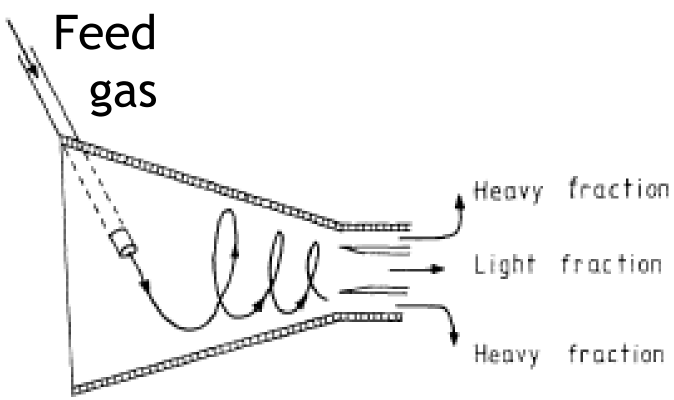


Aerodynamic Separation

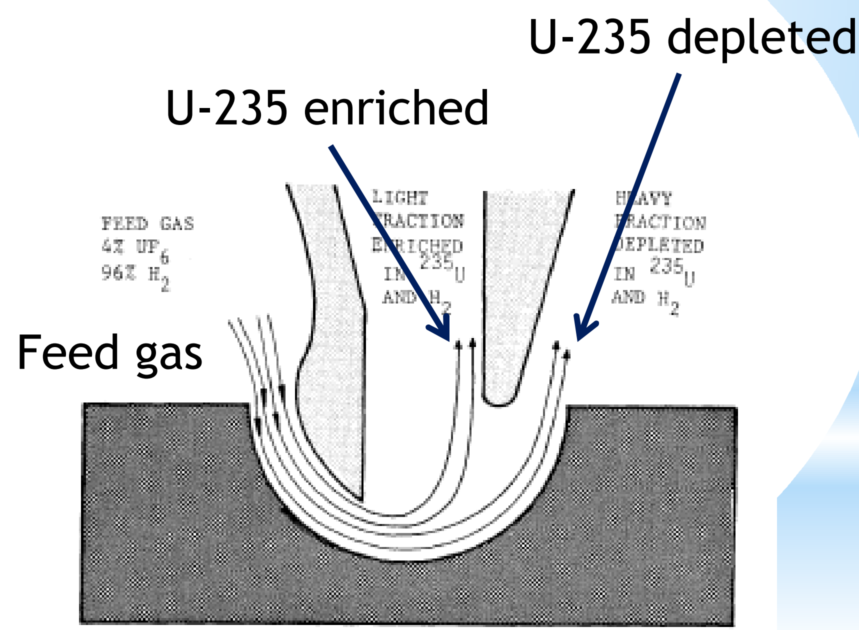
Jet Nozzle Process

Helikon Process (vortex tube)

- $^{235}UF_6$ and $^{238}UF_6$ experience different acceleration
  - Principle similar to centrifuge
  - Not commercially developed



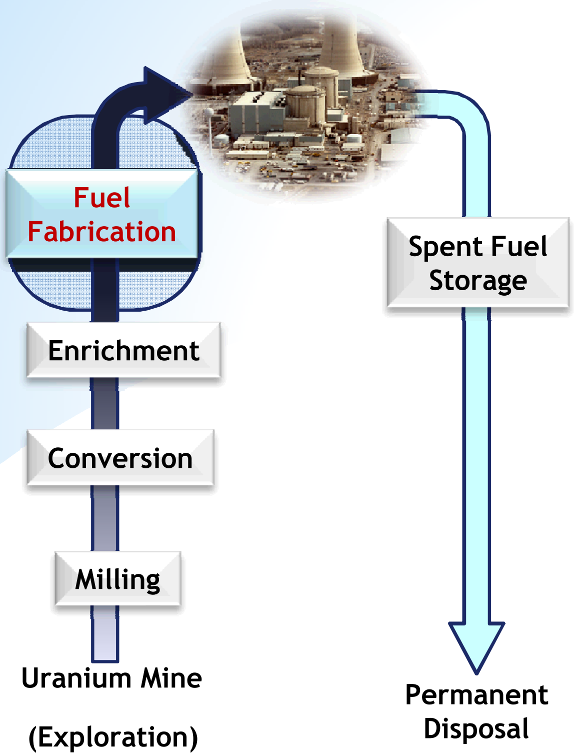
Vortex Tube



Jet Nozzle

Other Enrichment Processes

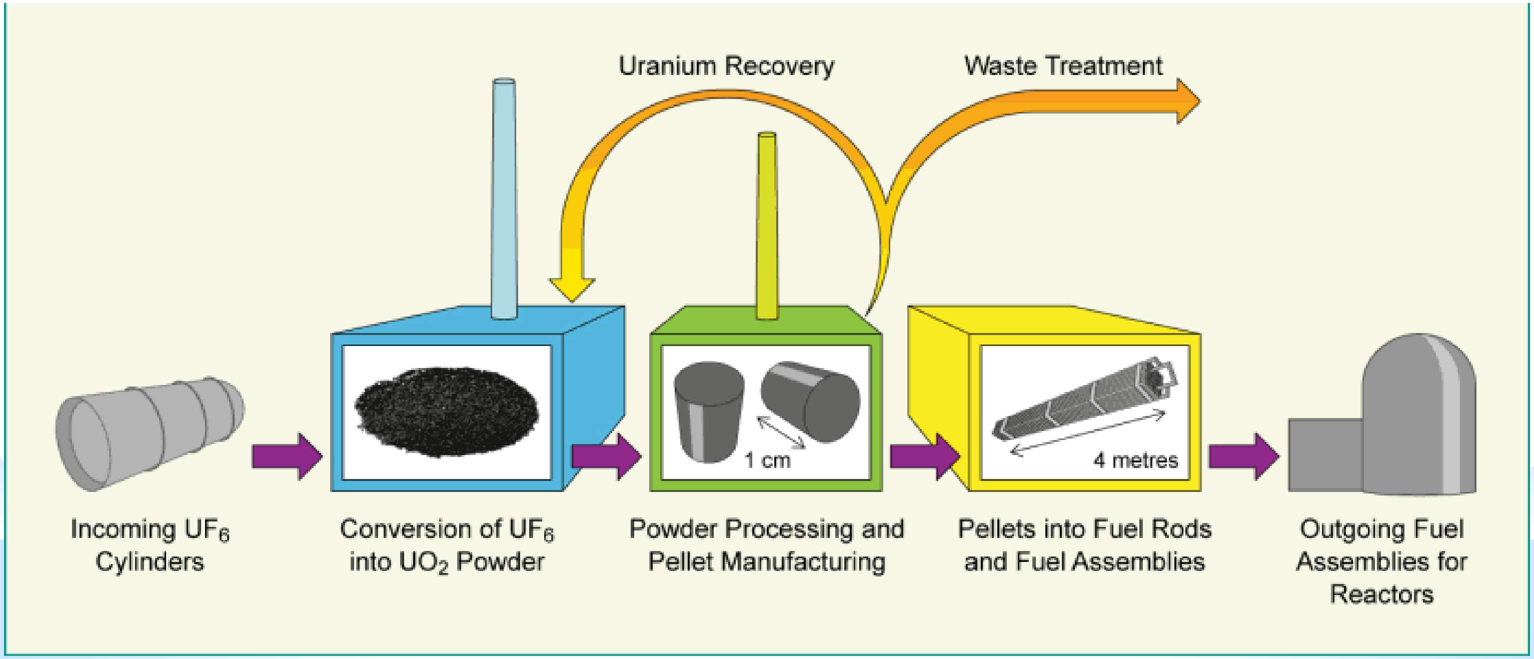
- Electromagnetic and plasma processes
- Chemical-exchange methods



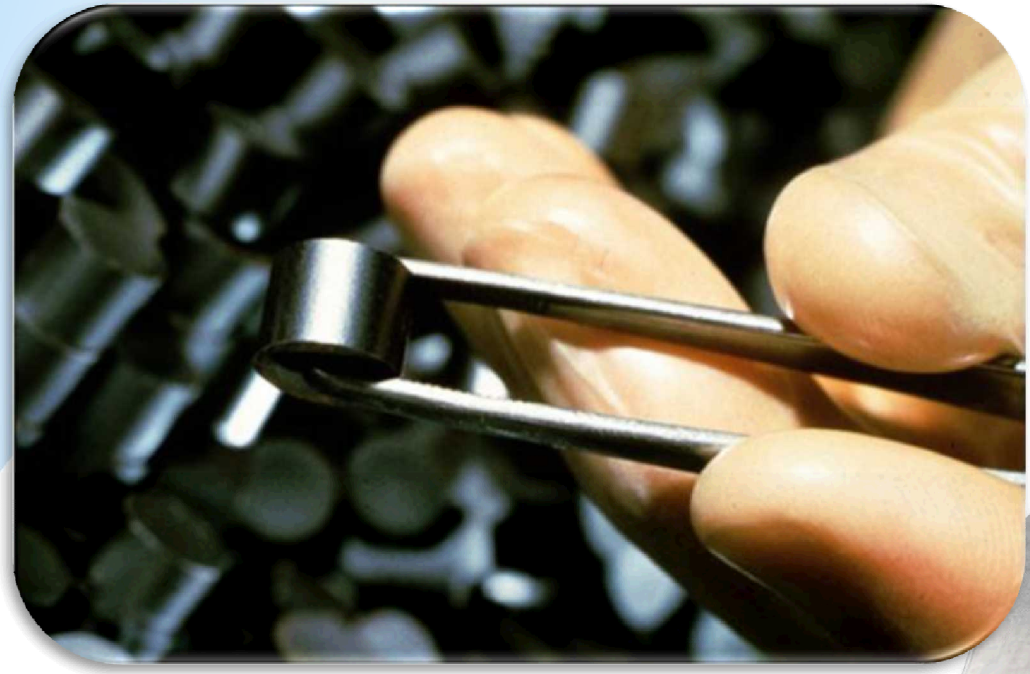
Fuel Design and Fabrication

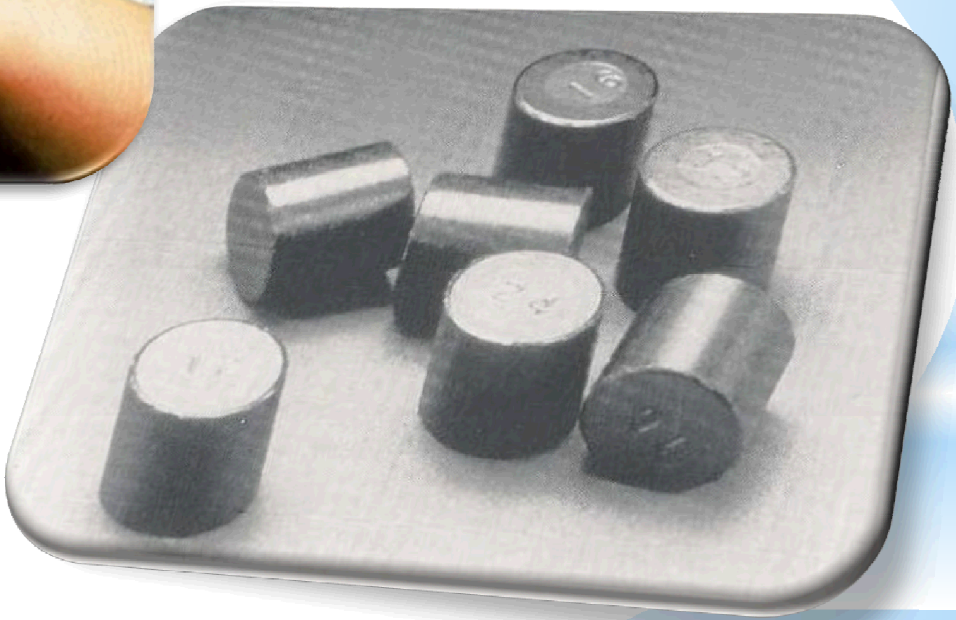
Oxide-Fuel Fabrication

1. **Uranium Hexafluoride** (UF_6) from enrichment plant
2. Uranium Hexafluoride converted to **Uranium Dioxide** (UO_2)
3. UO_2 is formed into dense **Fuel Pellets** (sintering in hydrogen atmosphere)
4. UO_2 pellets are inserted into **Fuel Rods** (fuel pins)
5. Multiple fuel rods make up a **Fuel Assembly**
6. Fuel Assemblies are shipped to reactors



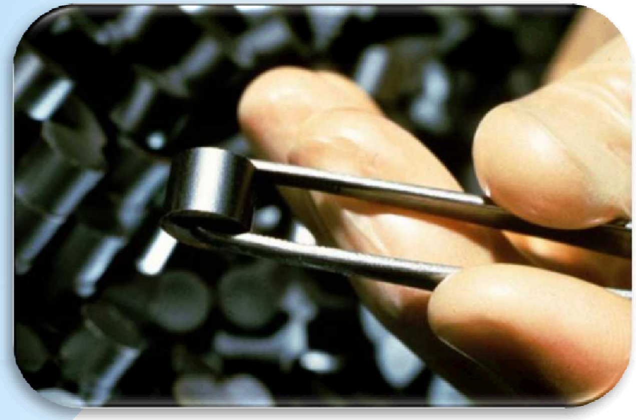
Uranium Dioxide (UO_2) Fuel Pellets



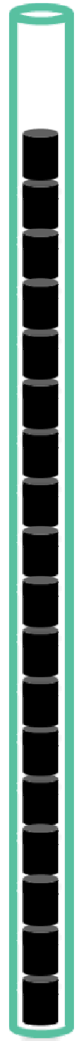


US NRC file photos
http://www.nrc.gov/images/reading-rm/photo-gallery/20100907-005.jpg)

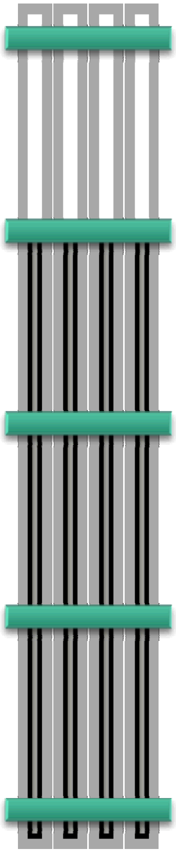
Fuel Assembly



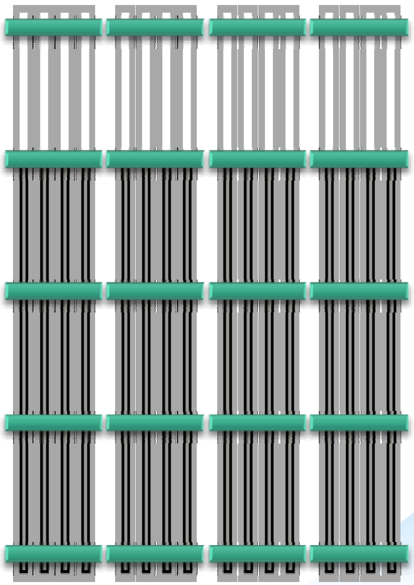
Ceramic **fuel pellets** stacked and sealed inside a metallic alloy tube (the **"cladding"**) to make each **fuel rod**



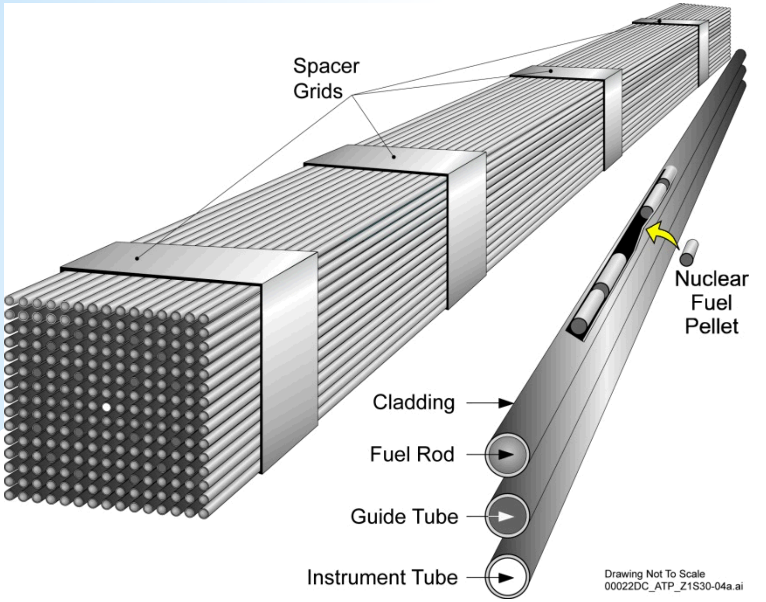
Fuel rods grouped in a regular array or **fuel assembly**



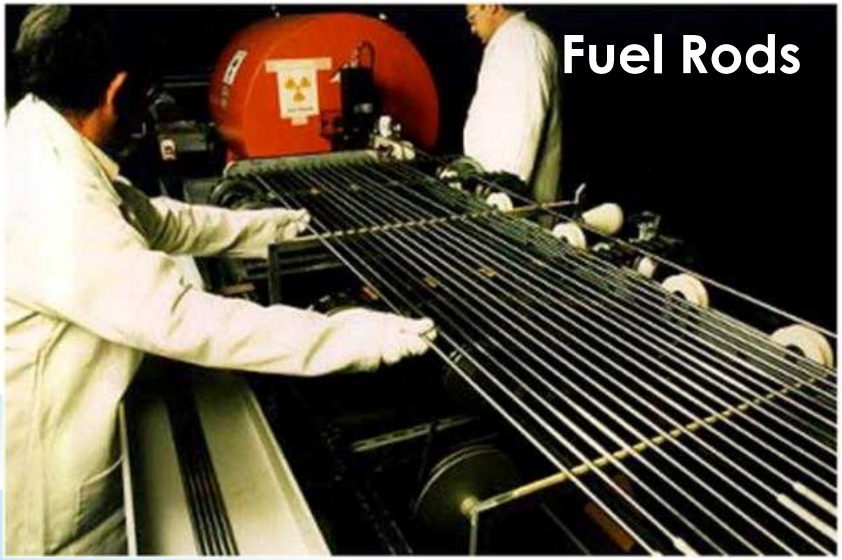
Fuel assemblies arranged in a larger regular array in a **reactor core**



Fuel Rods & Assemblies



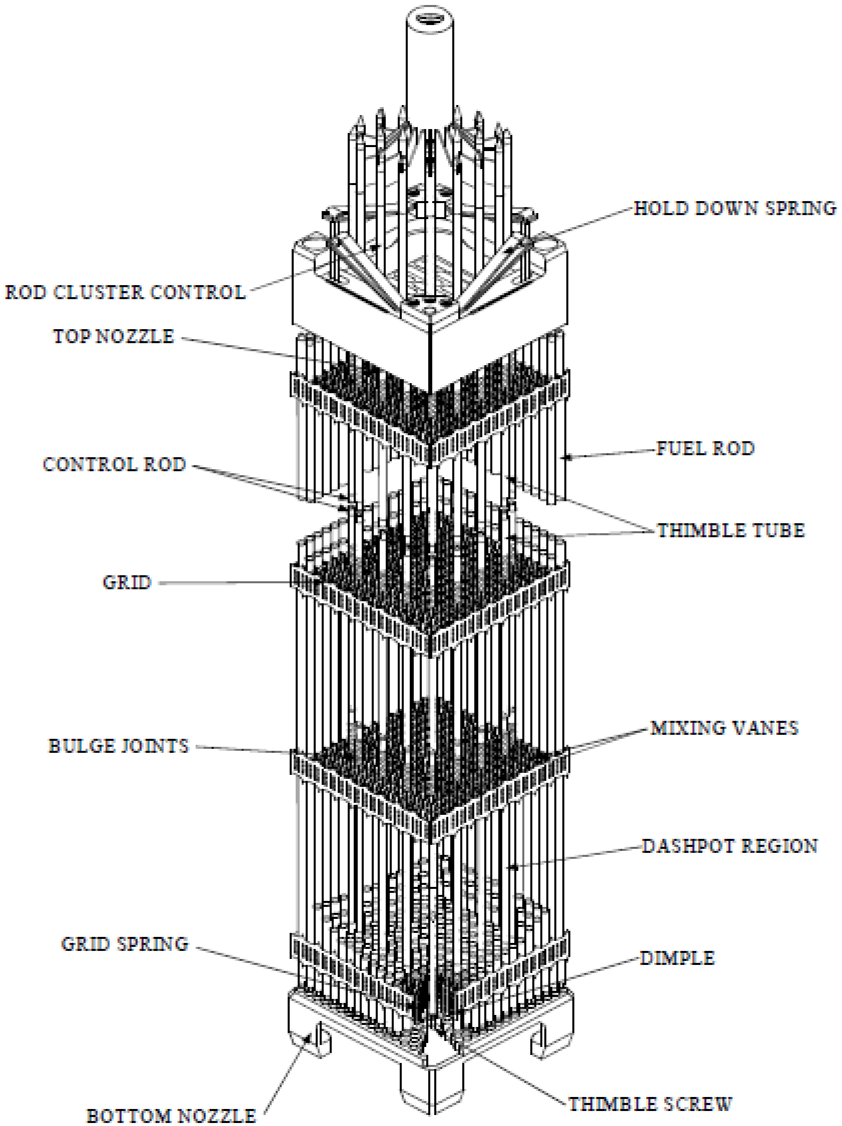
Fuel Rods

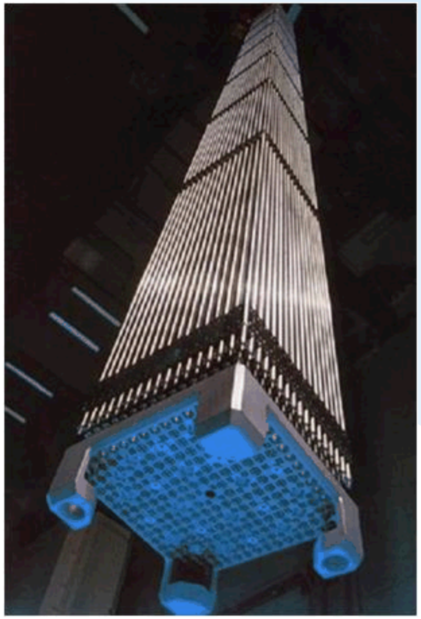


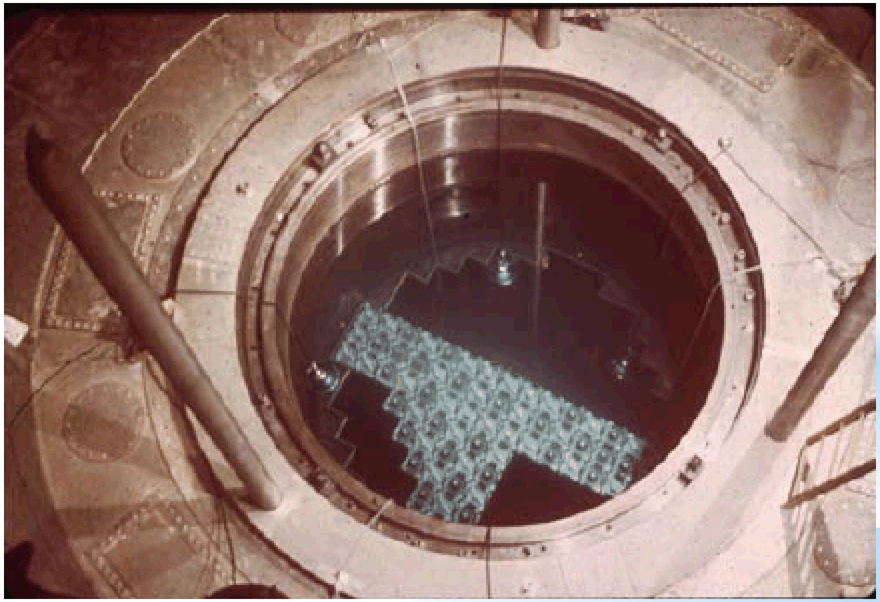
Fuel Assemblies

U.S. NRC file photos
(http://www.nrc.gov/images/reading-rm/photo-gallery/20071114-045.jpg, http://www.nrc.gov/images/reading-rm/photo-gallery/20100907-141.jpg)

Fuel Assembly & Reactor Core





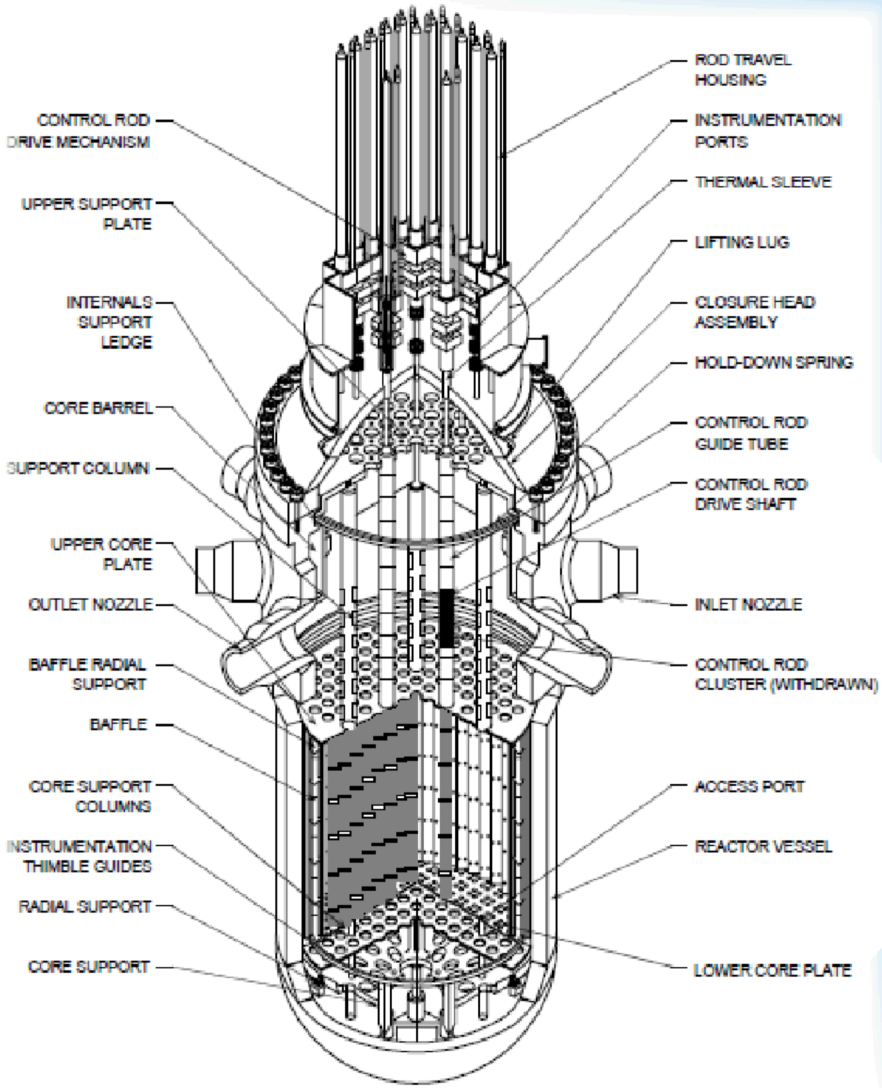


Reactor Vessel (PWR)

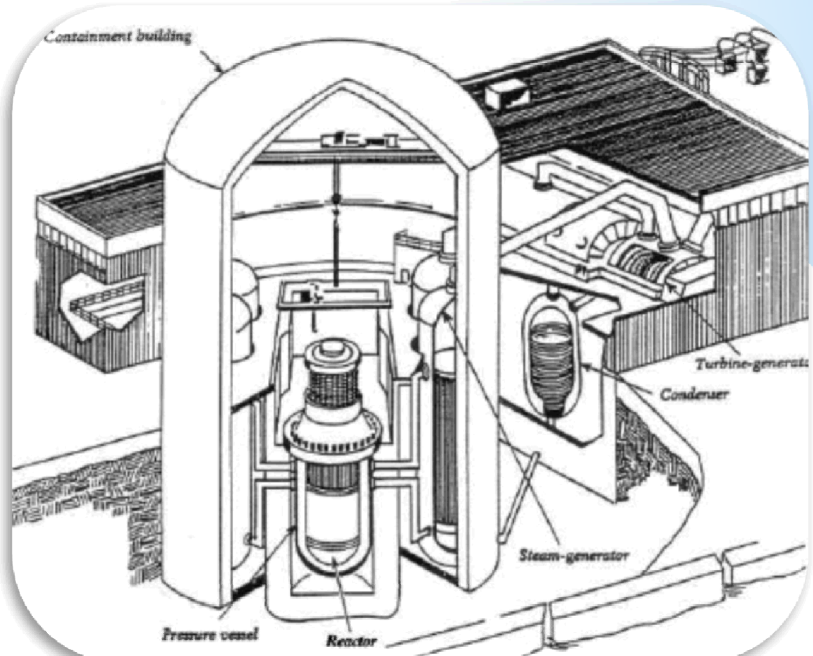
- Provides barrier to fission-product release
- Part of coolant system
- Pressure boundary

Typical 1000 MWe PWR

- ~ 70 tons uranium in core
  - ~160 fuel assemblies
- 18-month fuel cycle
  - ~1/3 fuel assemblies replaced each fuel cycle
- ~ 25 tons fresh fuel *added* and spent fuel *removed* every 18 months
  - "equilibrium fuel cycle"

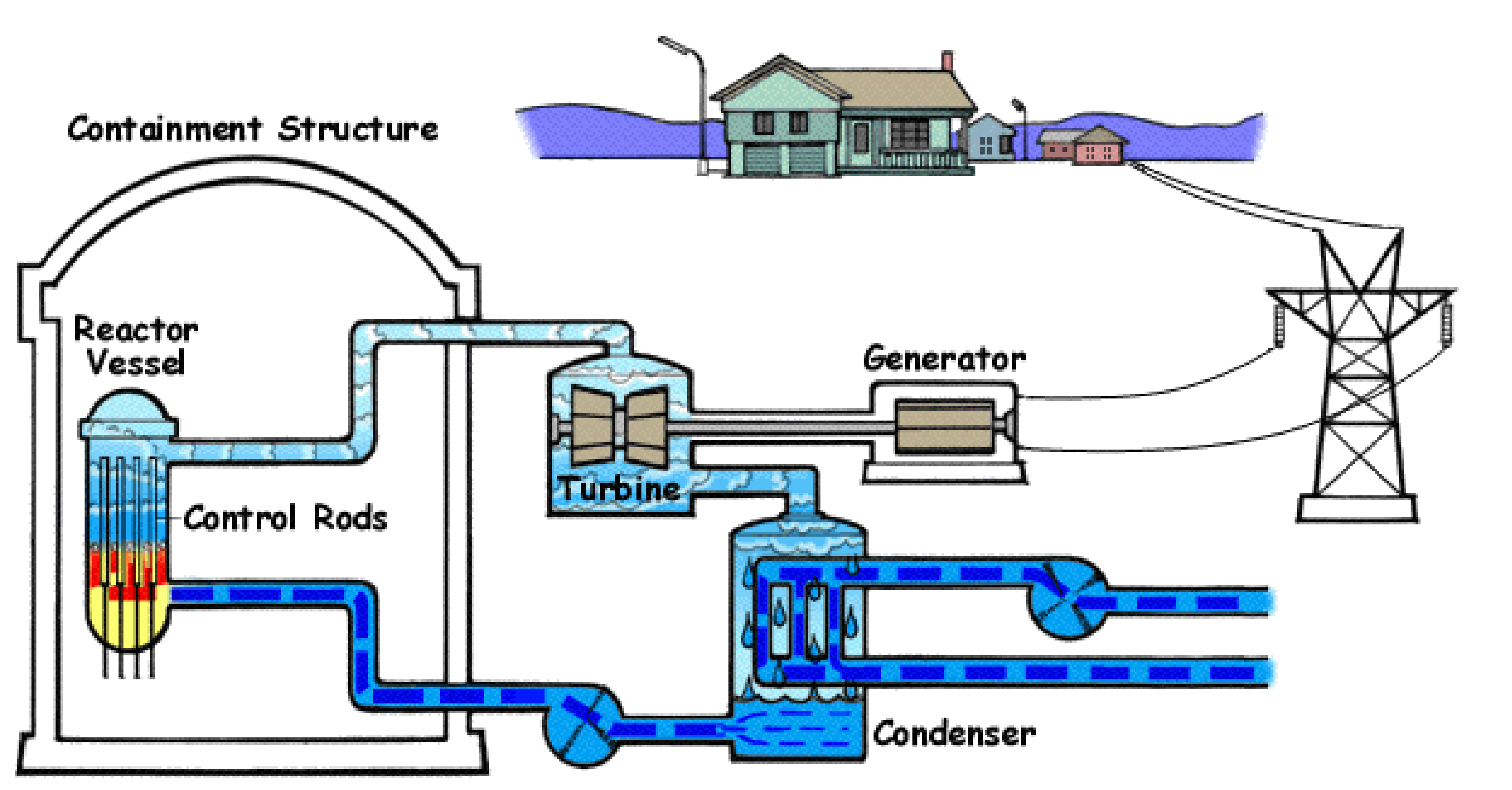


Fuel is "Burned" in Reactor Core



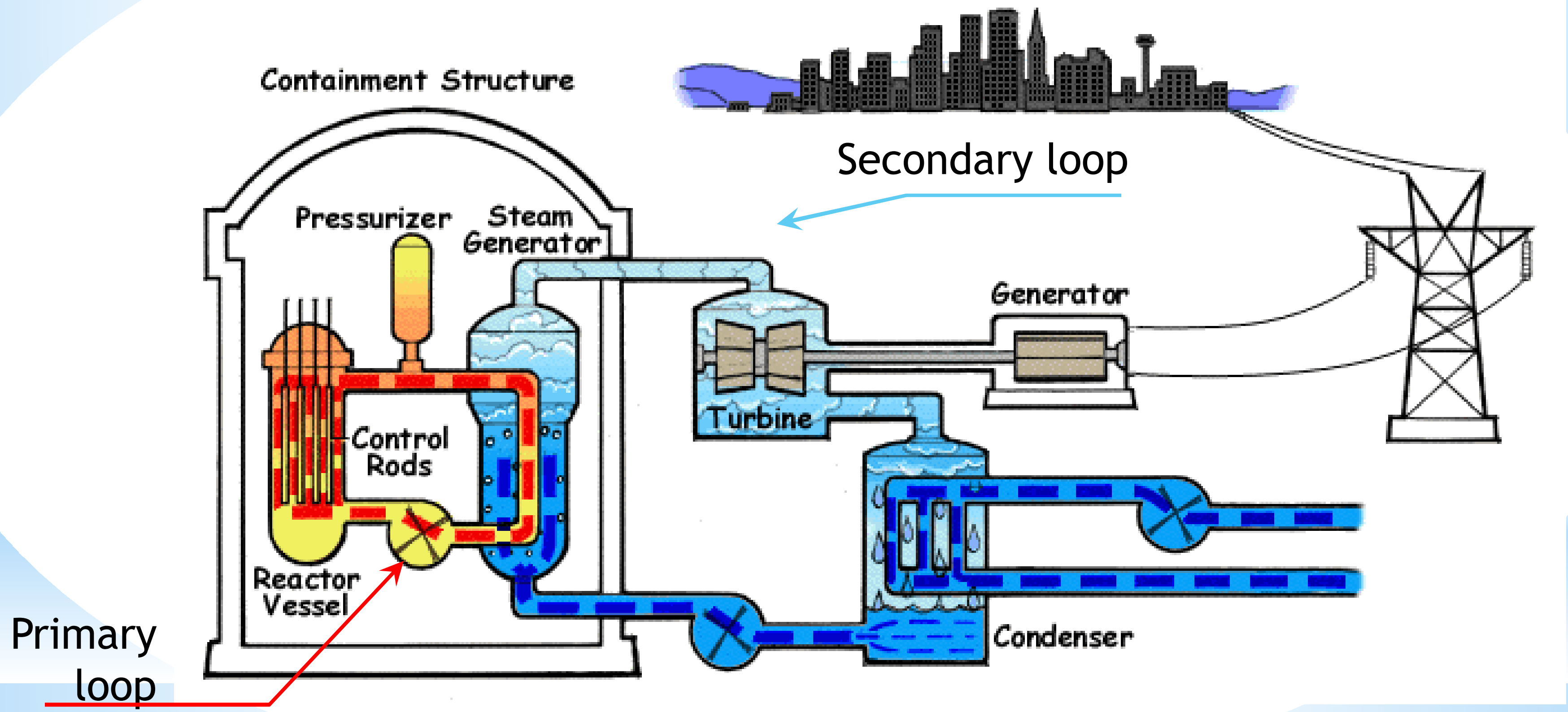
Boiling Water Reactor (BWR)

- BWR boils the water in the core
- Steam from core runs a turbine to produce electricity



Pressurized Water Reactor (PWR)

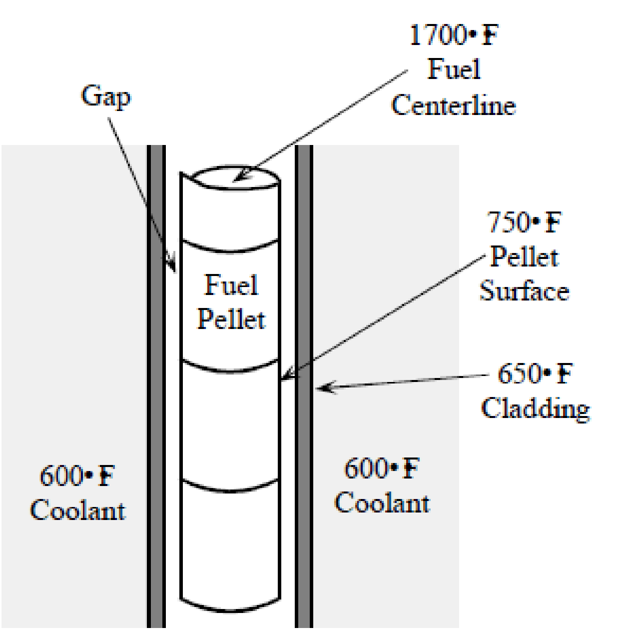
- Pressurized water heats, but does not boil
- "*Primary Loop*" generates steam in a "*Secondary Loop*"
  - Secondary loop runs turbine to produce electricity

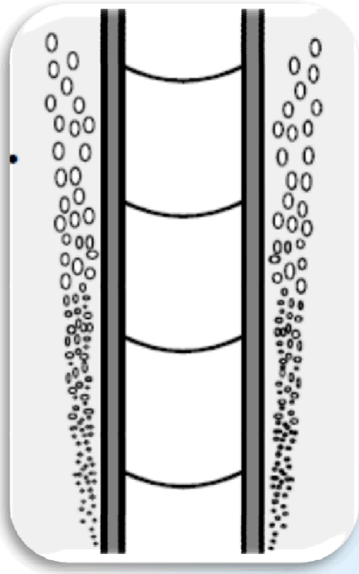


Source: NRC http://www.nrc.gov/reading-rm/basic-ref/students/animated-pwr.html

Nuclear Fuel Performance

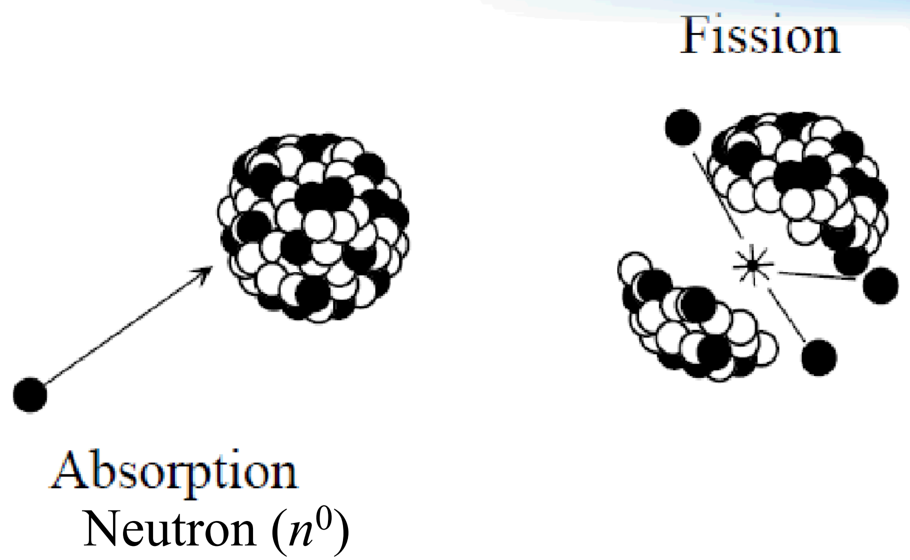
- Harsh in-reactor environment
  - High radiation
  - High temperature
- Nuclear power plants are costly
  - high capital costs
  - Low fuel cost compete with other electricity generation methods
- Fuel failures may shutdown a power plant
  - Fuel pins are the first barrier to radioactive releases





"Burning" Nuclear Fuel

- U-235 Readily absorbs a neutron
  - Becomes highly unstable U-236.
- U-236 has high probability to **fission**
  - ~80% of U-236 atoms fission
  - Produces fission products, more neutrons, and **energy**
  - Kinetic energy of products converted to heat
    - Heat is used to make high pressure steam and, ultimately, electricity.
- Fission of U-236 releases two or three additional neutrons
  - Can cause other atoms to fission
  - A chain reaction (criticality)

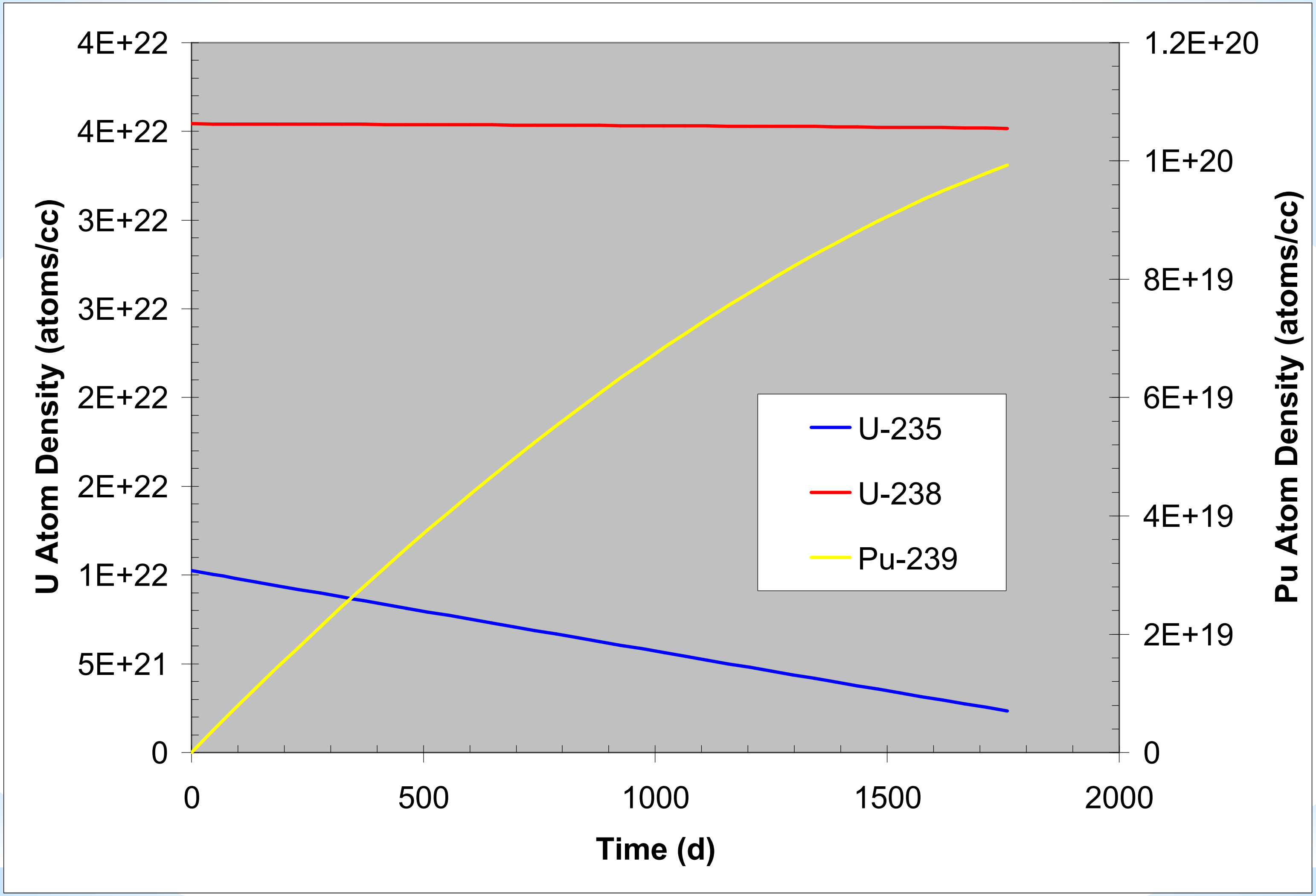


$$E = mc^2$$

The energy produced by fission is proportional to the *mass difference* between initial atom plus neutron and the products left after fission:

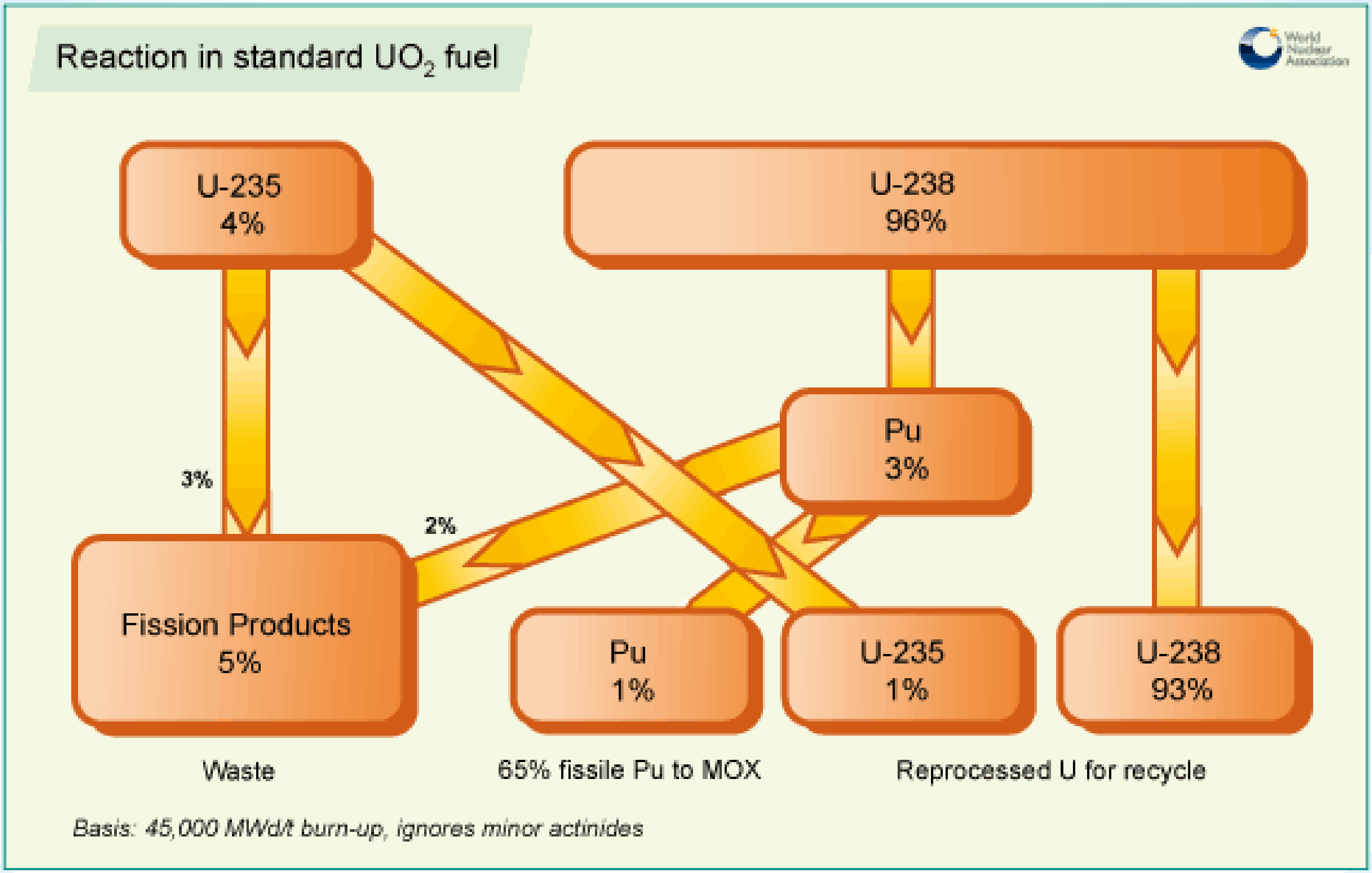
$$(U\text{-}235 + n^0) - (FPs + xn^0)$$

Actinide Buildup and Decay



Note different scales (Pu ~ 1/100th of U)

Irradiation of LEU UO_2 fuel



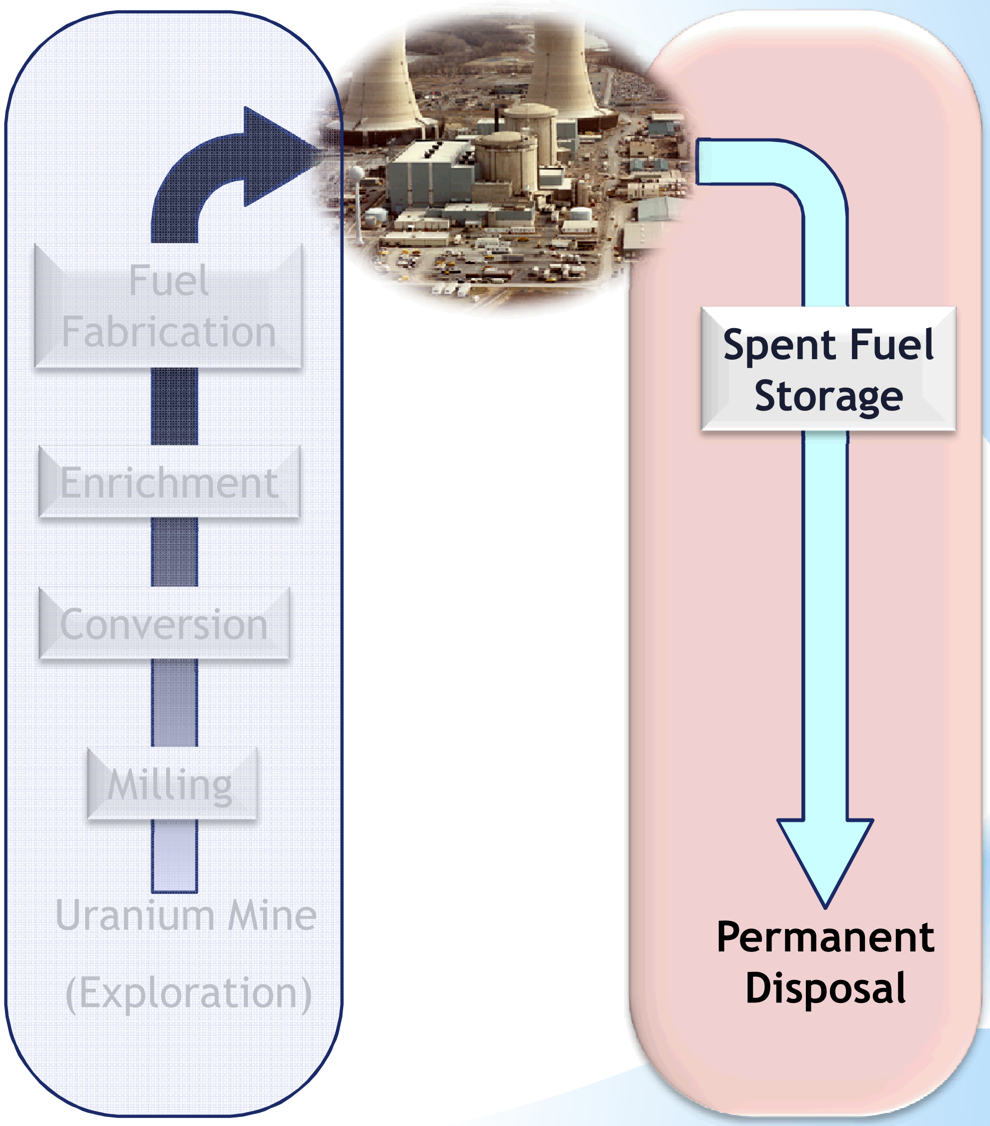
Uranium Fuel Cycle & Light Water Reactors

Front End

- Processes required to obtain, treat and manufacture nuclear fuel for use in a reactor

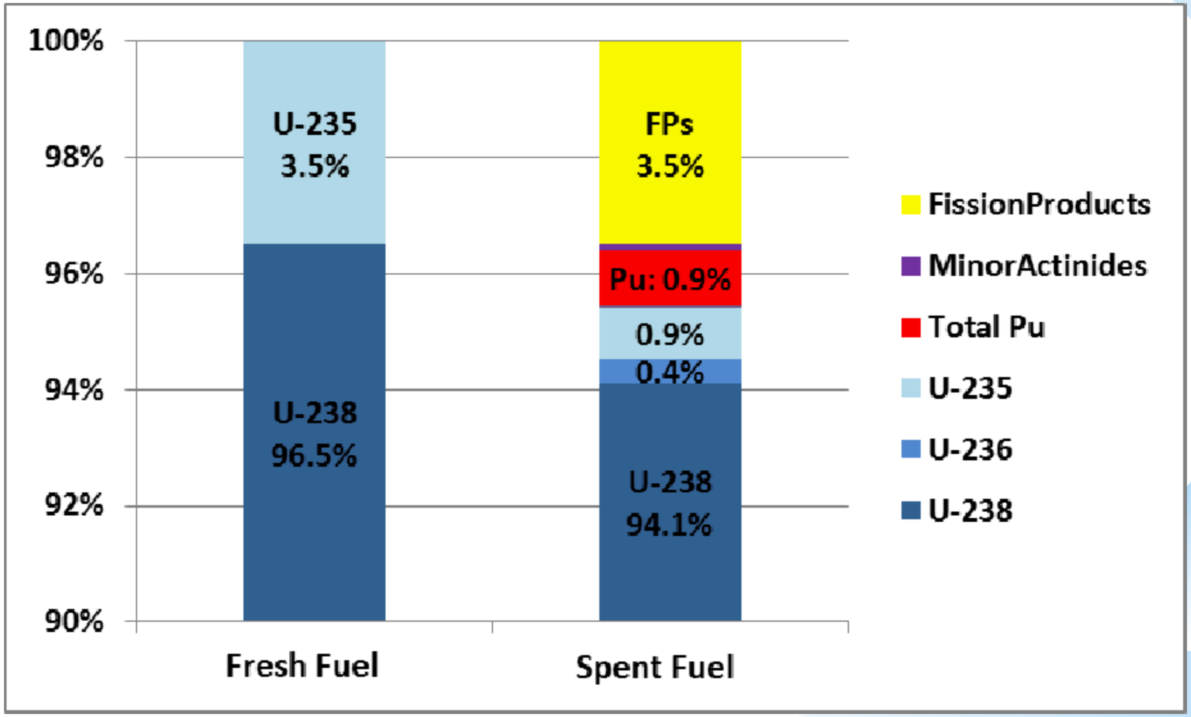
Back End

- **Manage spent fuel and related waste streams**
  - In-reactor changes to fuel during reactor operation
- **Final disposal**



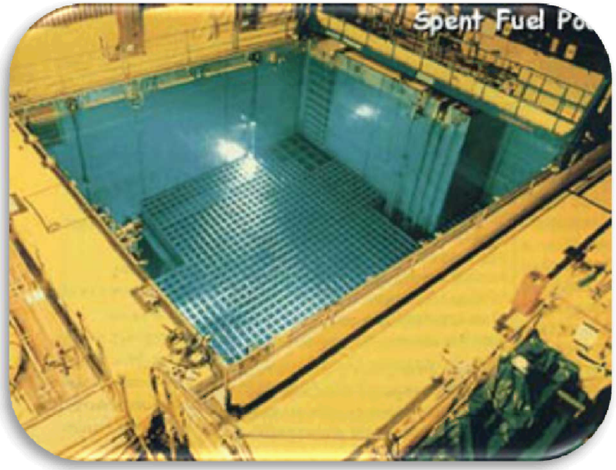
Spent Fuel

- Burnup
  - Power produced per mass of uranium
- Fission produces Fission Products (FPs)
  - Commonly radioactive
  - Neutron absorbers
    - "poison" the fuel



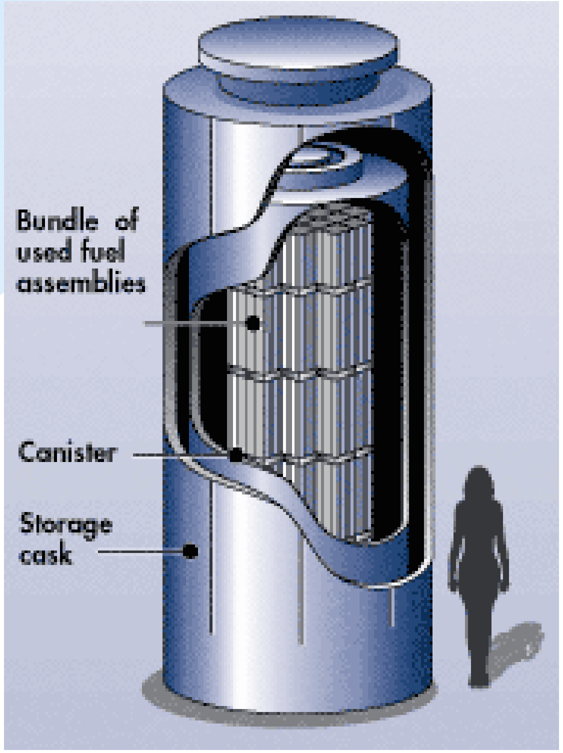
Storing Spent Fuel

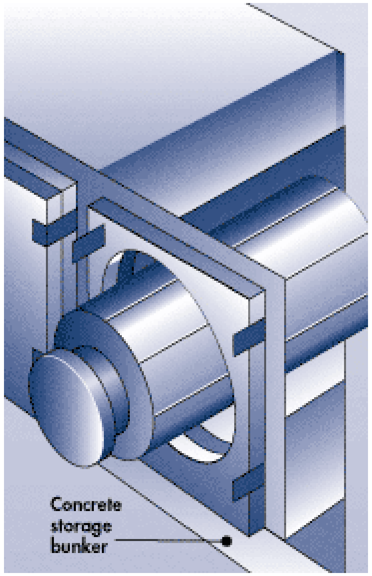
- Spent fuel is removed from the reactor after ~18 months
  - The spent fuel is HOT!
  - SNF is cooled in a large water pool connected to the reactor (wet storage)
  - Fission products decay, radiation & temperature decrease
- After several years, cooler fuel assemblies may be transferred into dry casks for long-term storage (dry storage)
  - At-reactor storage pool must retain space for newly discharged fuel



At-Reactor Spent Fuel Pools

Dry Storage Spent Fuel

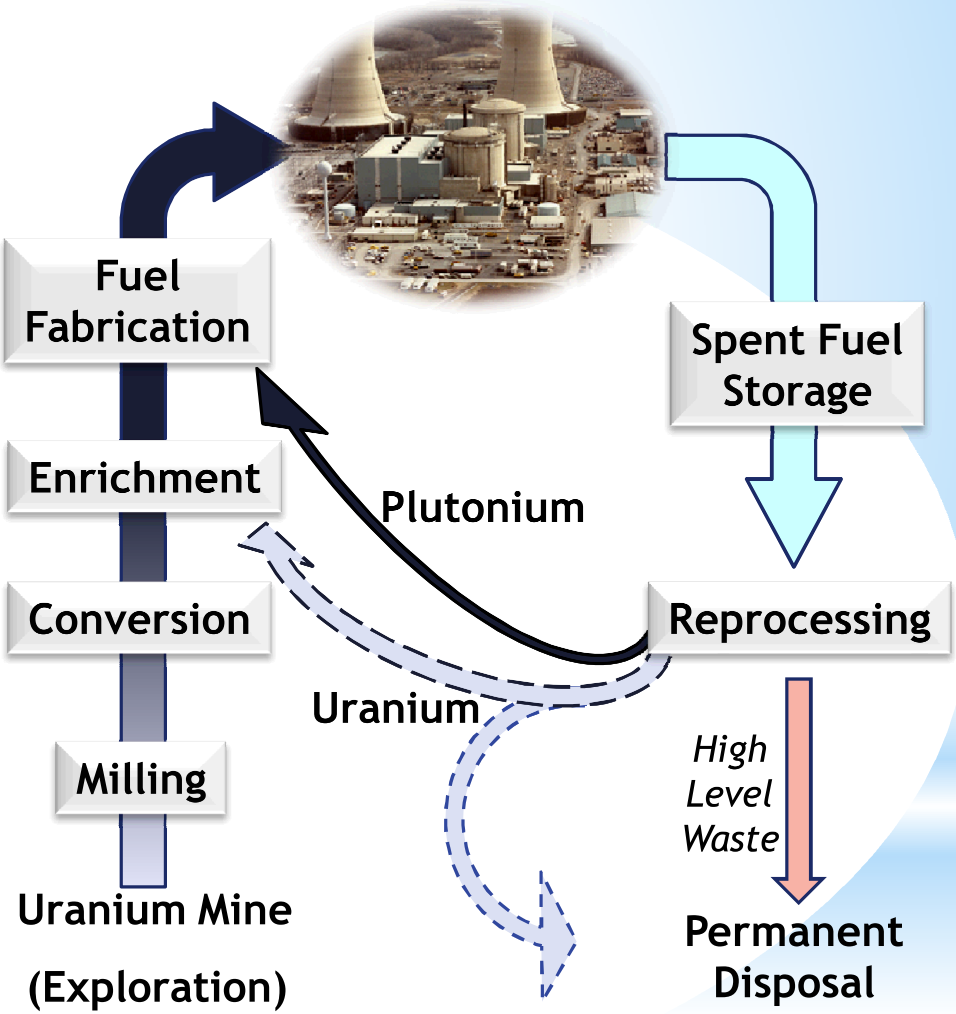




Dry-Storage Casks: Vertical and Horizontal Designs

"Closed" Fuel Cycle – Reprocessing & Recycle

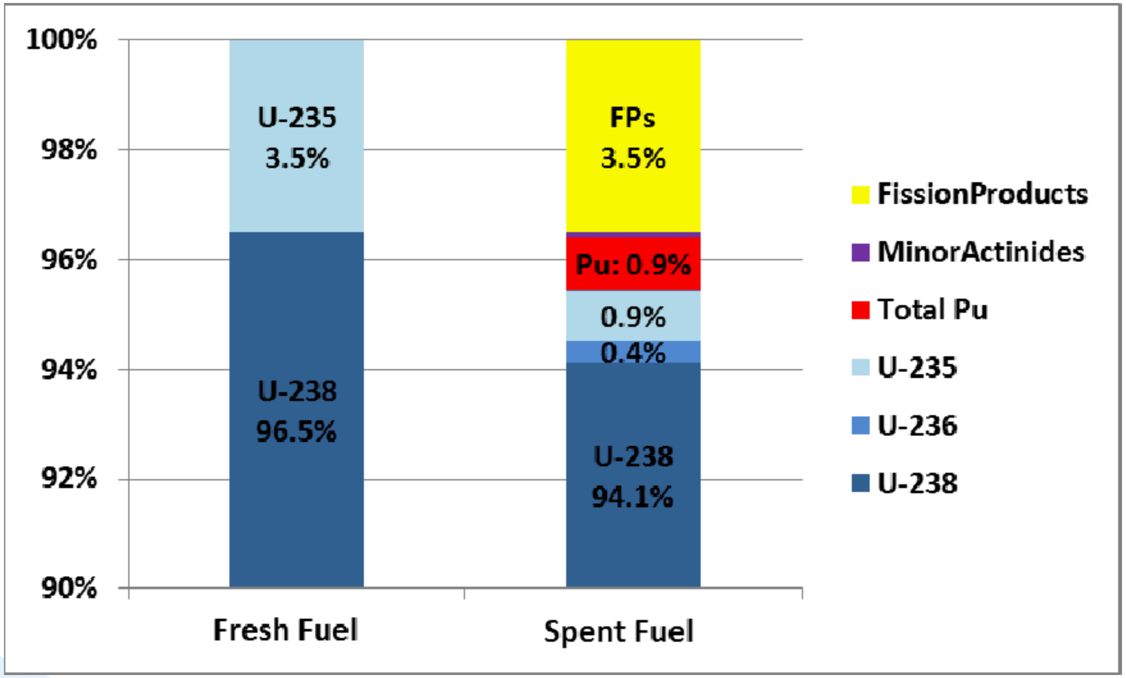
- More efficient use of resources
  - Cost vs. benefit
- Recover Fissile materials to reuse in a reactor
- Separate neutron poisons and dispose as high-level waste
  - Transmutation?
- Design waste forms for disposal
  - Enhanced durability in disposal environment



Spent Fuel Recycling

Spent fuel contains…

- Abundant ***fertile*** material
  - Uranium-238
- Appreciable concentrations of ***fissile*** material
  - U-235 and plutonium



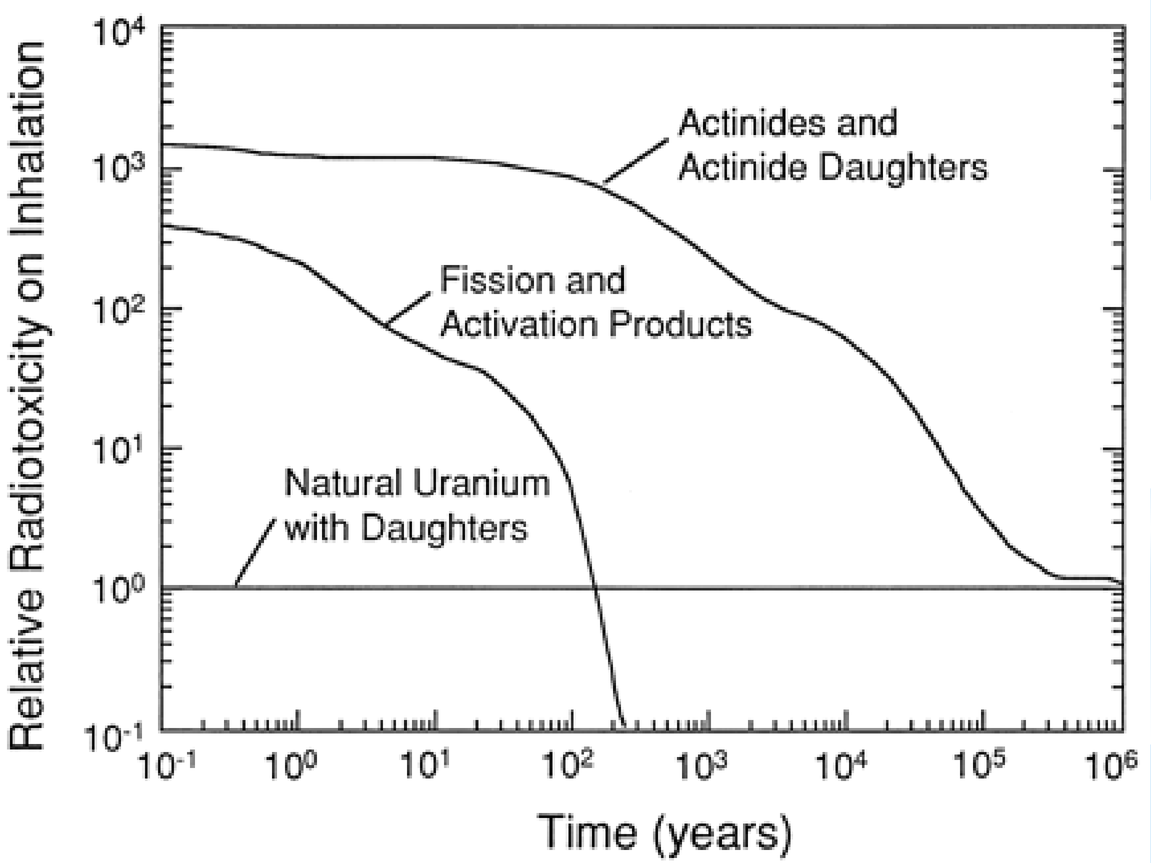
But also contains neutron-absorbing fission products

Spent Fuel Reprocessing - PUREX process

1. Cooled spent fuel is chopped into small pieces
   - mechanical shearing or sawing
2. Fuel and cladding are sent to a “dissolver” and reacted with nitric acid
   - Cladding hulls are recovered, washed, packaged, and shipped for eventual disposal
3. **Solvent extraction** separates >99% of fission products from plutonium and uranium
   - Tri-Butyl Phosphate (TPB) added to nitric acid solution
4. Plutonium & uranium chemically separated from each other
   - Pu (and potentially U) can be recycled into Mixed-Oxide Fuel (MOX)
     - Depleted uranium commonly added to MOX
   - MOX can be used in some LWRs

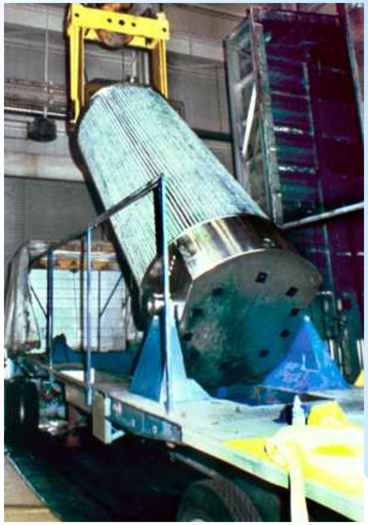
Actinide Management

- Reprocessing and recycling can reduce the quantity of actinides disposed as *waste*
- Recycle actinides as fuel
  - May need special reactors



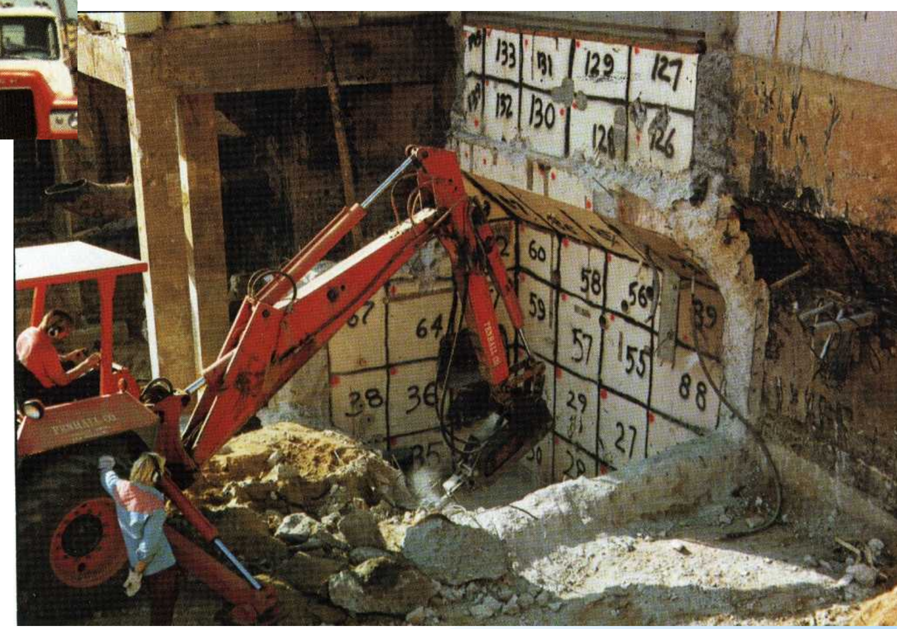
Transportation –HLW & Spent Fuel

Rail



Truck

Decommissioning

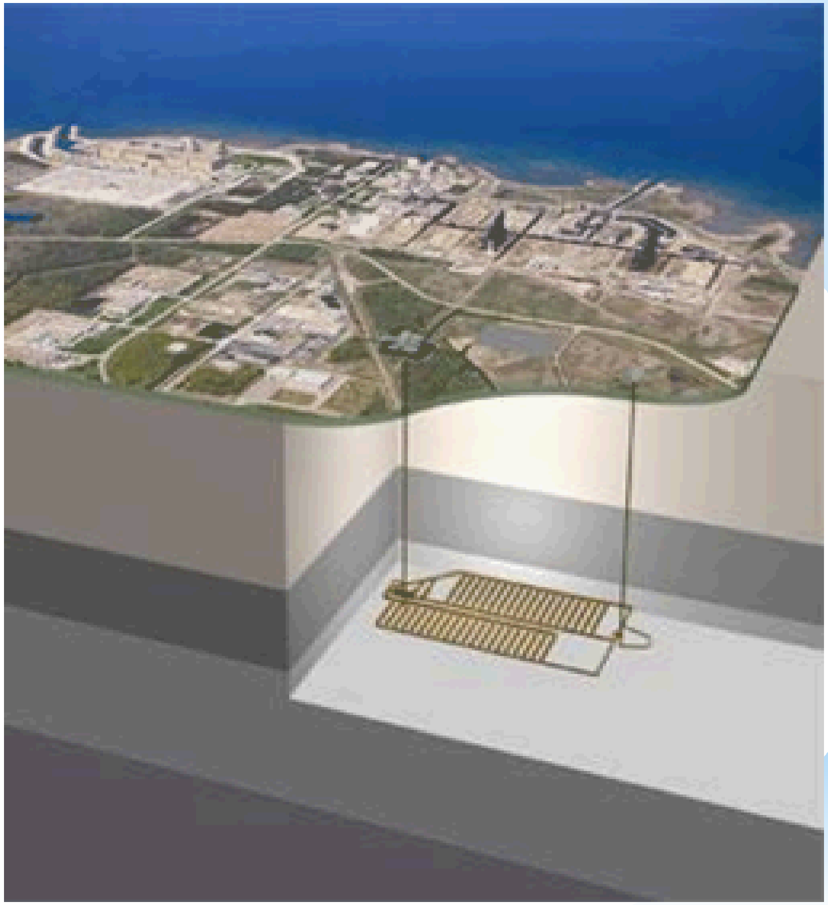


Low-Level Waste Disposal

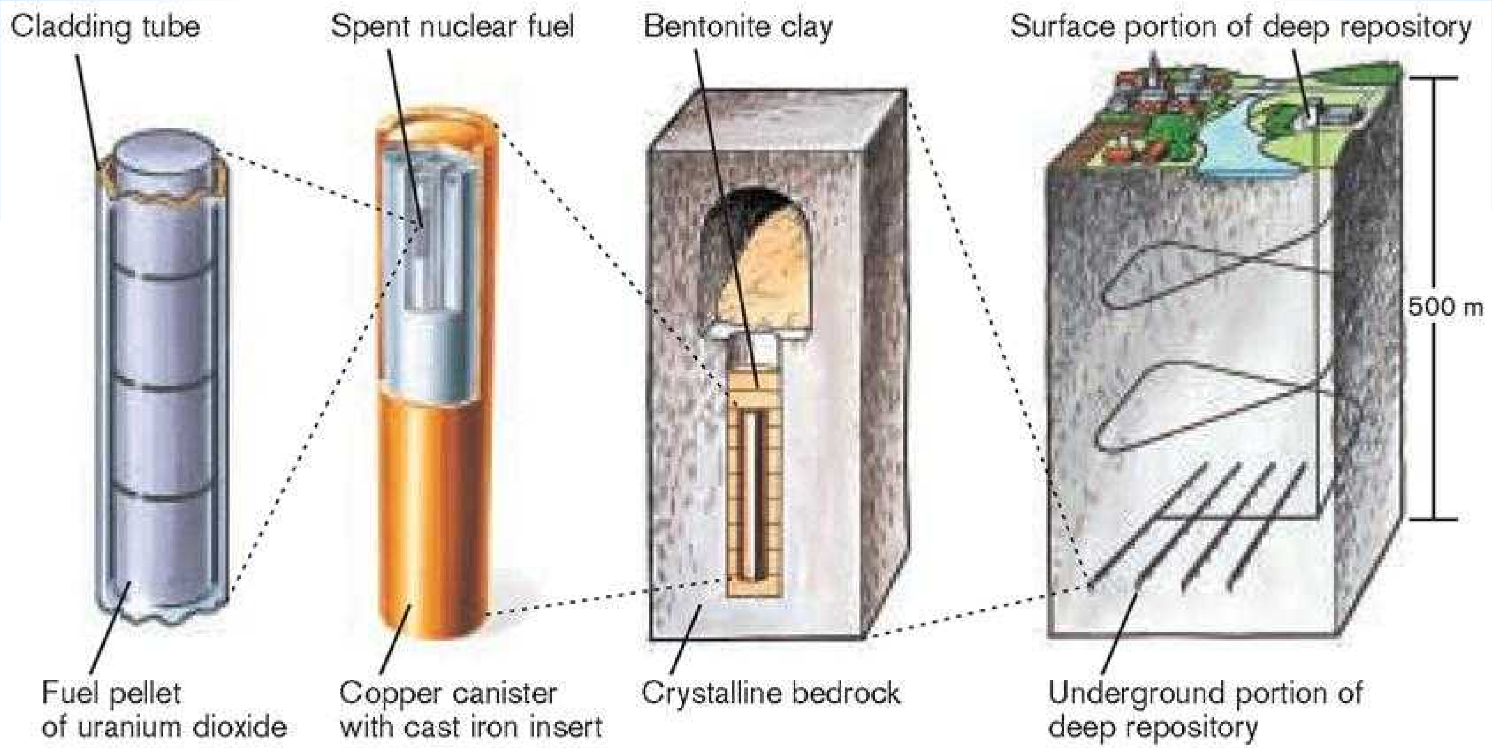
Shallow Burial

Disposal of High Level Waste and Spent Fuel

- All countries that have a plan, plan for deep disposal in a geologic repository
- Design variables
  - Waste Form
  - Rock type
  - Groundwater transport
  - Saturation
  - Waste Container
  - Potential Retrievability
- Major hurdles
  - Public acceptance
  - Site selection
  - Transportation plans
  - Long-term licence requirements

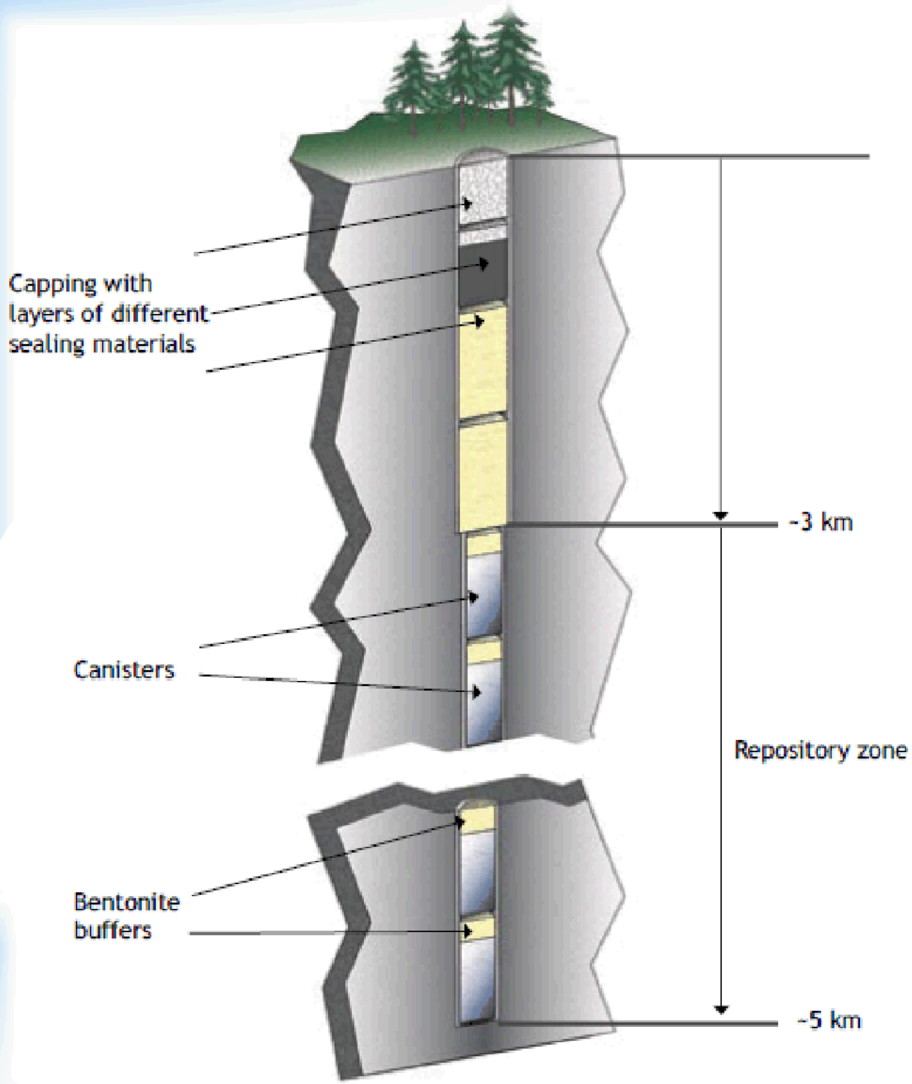


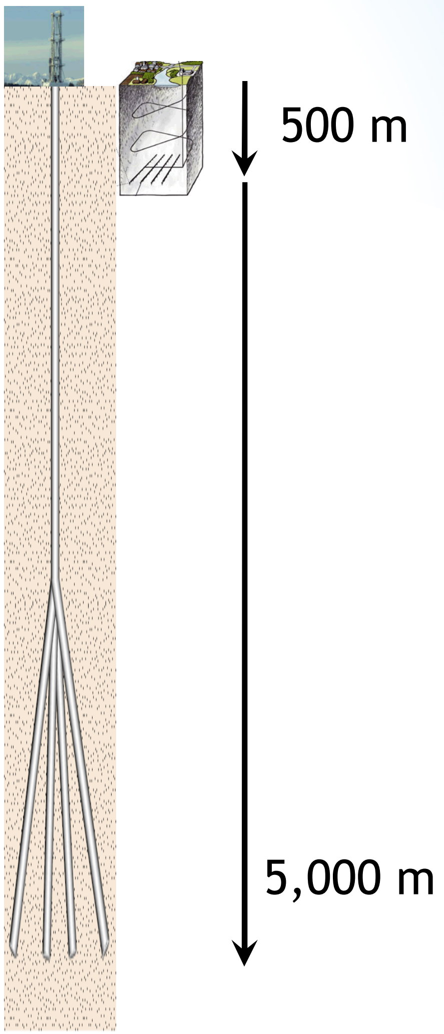
Permanent Disposal



Mined Geologic Repository

Deep Borehole Disposal





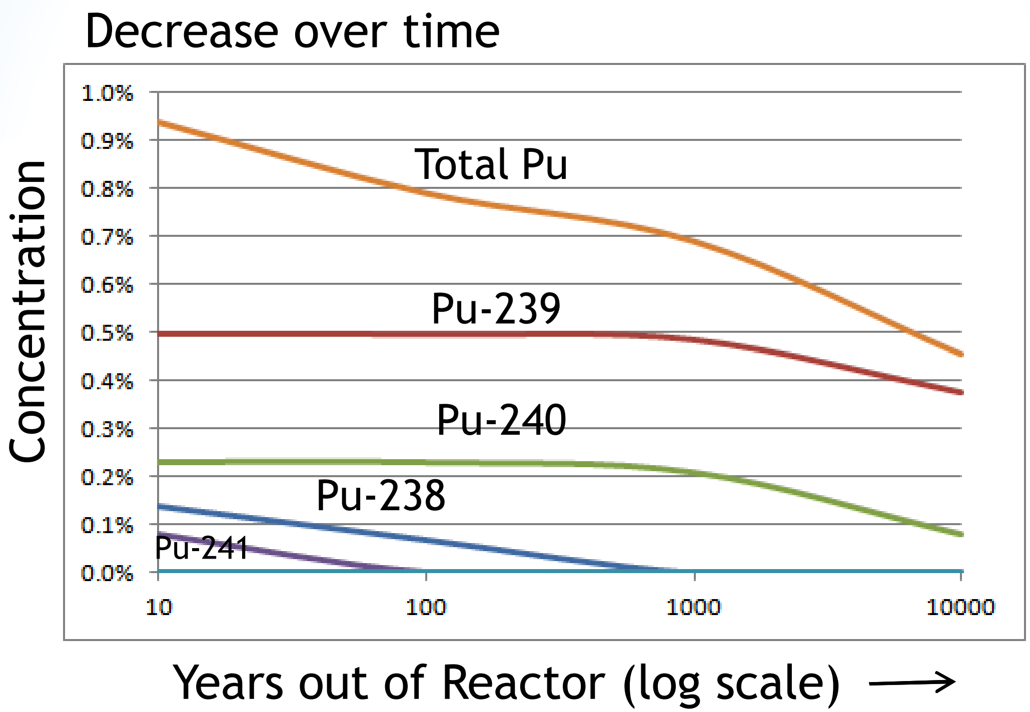
Source: M. Kårelind, in K.-I. Åhäll (2006) MKG Report 2, December 2006, pp. 7,9.

Desirable Characteristics of Waste Forms

- Solid
- Corrosion resistant
- Stability
  - Thermal
  - Chemical
  - Mechanical
  - Radiation-damage resistant
- Compatible with disposal environment

UO_2 fuel has many of these characteristics

Plutonium in Spent Fuel*



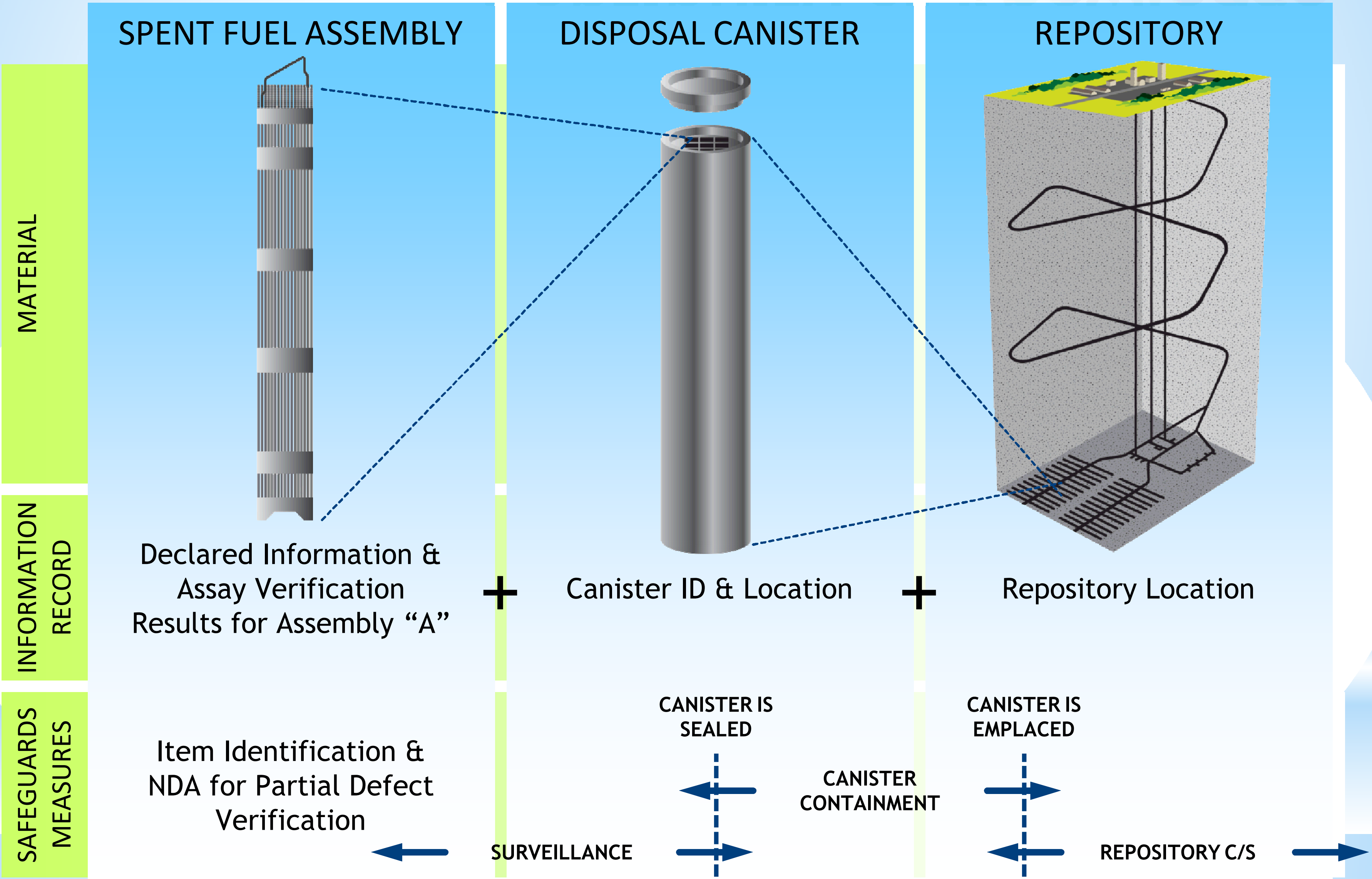
*PWR fuel with burnup = 33 MWd/kgU.
Data from Bruno & Cera in *Uncertainty Underground*

Repository Safeguards

- Design information verification (DIV)
  - Undeclared structures, rooms, tunnels, etc.
- Nuclear materials safeguards
  - Timely detection of diversion
    - Nuclear material accountancy (NMA)
    - Continuity of knowledge (CoK)
    - Containment & Surveillance (C/S)
- Undeclared activities
  - Reprocessing
  - Tunneling/mining
  - Tampering with or removing casks

➢ ***Safeguards on spent fuel remain in force for as long as a safeguards agreement remains in force***

Continuity of Knowledge



Suggested Reading

- U.S. NRC "Nuclear Reactor Concepts" Workshop Manual, http://www.nrc.gov/reading-rm/basic-ref/teachers/unit3.html
- Nuclear Power for Energy Generation http://www.nrc.gov/reading-rm/basic-ref/teachers/01.pdf
- The Fission Process and Heat Production http://www.nrc.gov/reading-rm/basic-ref/teachers/02.pdf
- Pressurized Water Reactor Systems http://www.nrc.gov/reading-rm/basic-ref/teachers/04.pdf
- DOE Fundamentals Handbook "Nuclear Physics and Reactor Theory", http://www.hss.doe.gov/nuclearsafety/ns/techstds/standard/hdbk1019/h1019v2.pdf