Nuclear Fuel Performance

NE 533 Spring 2024

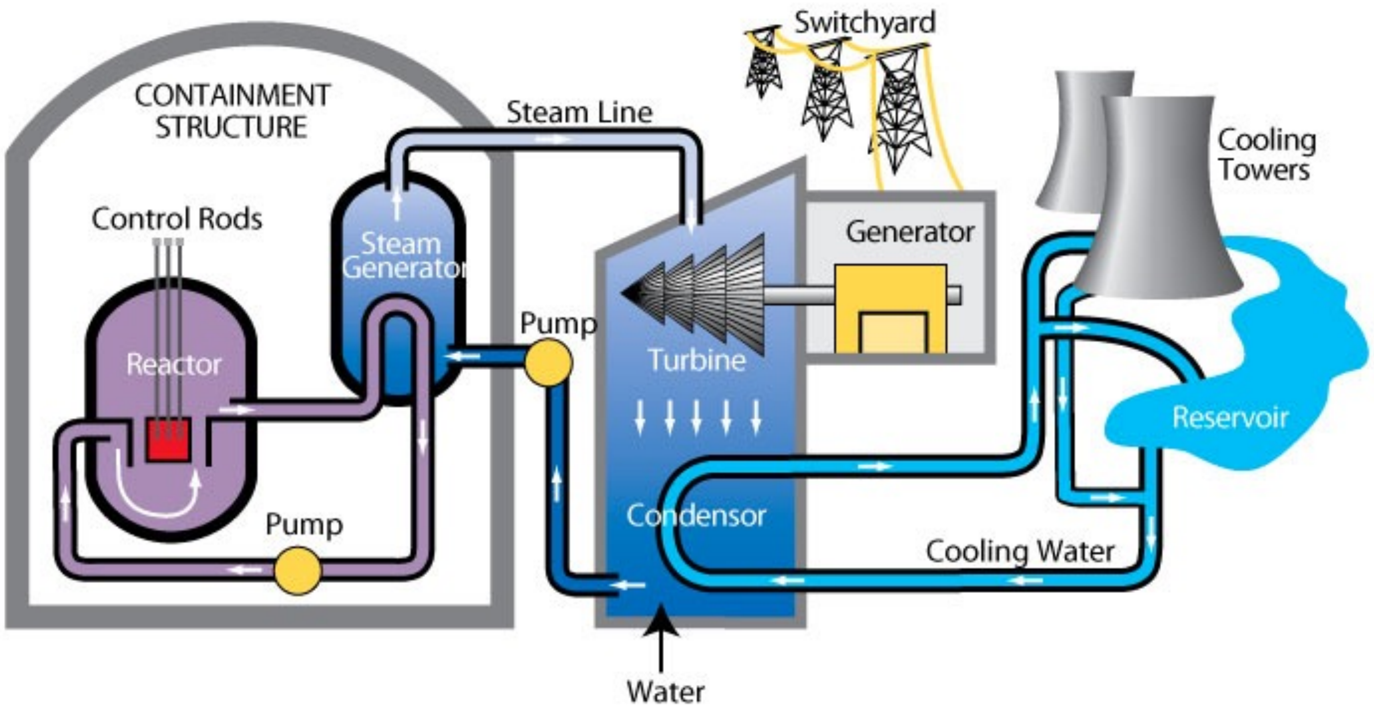
Last Time

- Intro to fuel performance as a three-pronged concept:
  - heat generation and delivery to coolant; fuel operation without problems; fuel behavior during accidents
- Uranium is combined with O, C, N, transition metals for a variety of fuel types
- UO2: ceramic, commercial reactor fuel, light water reactors
- Good characteristics:
  - high melting point, single phase, cladding compatibility, radiation resistance, stability in water
- Bad characteristics:
  - brittle, low thermal conductivity, limited power density, high burnup swelling

REACTOR SYSTEMS

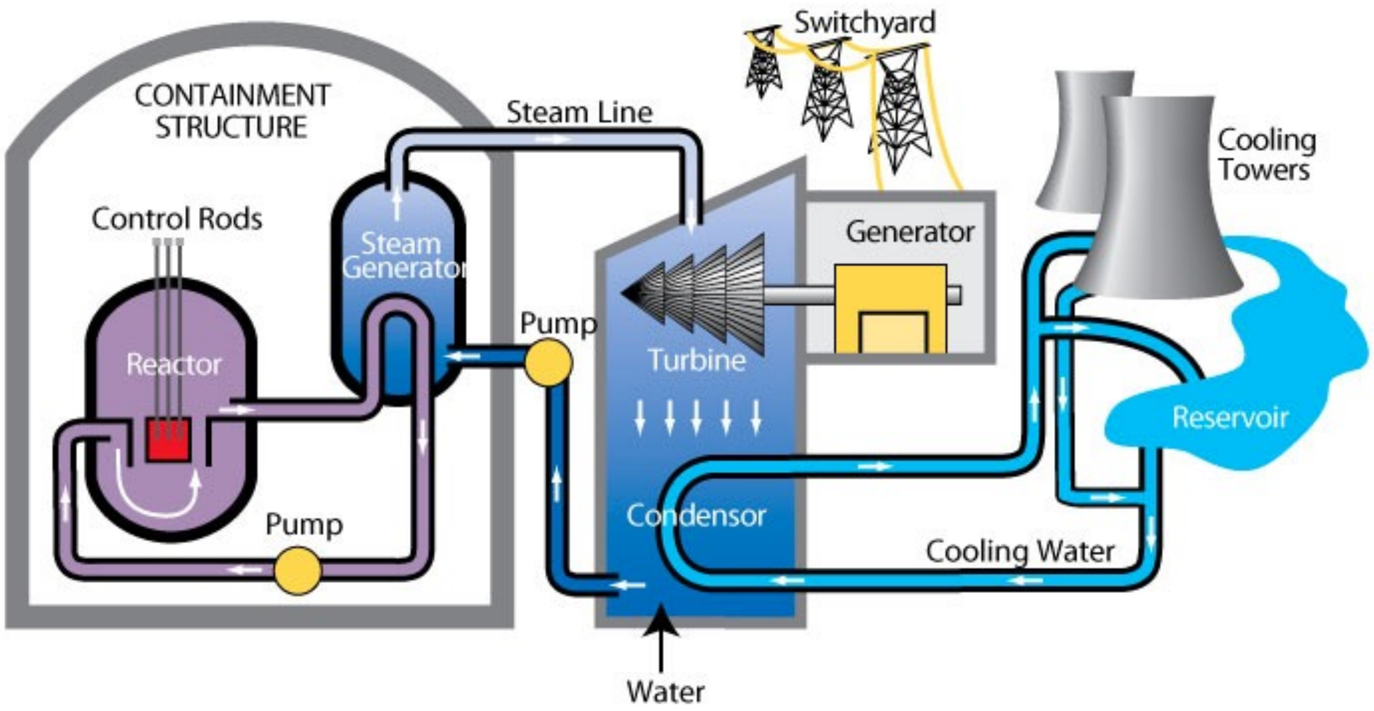
Heat removal systems

- Now we touch on how heat is removed from the fuel
- Primary mechanism to remove heat directly from fuel is a coolant
- Various coolant types, most common is water



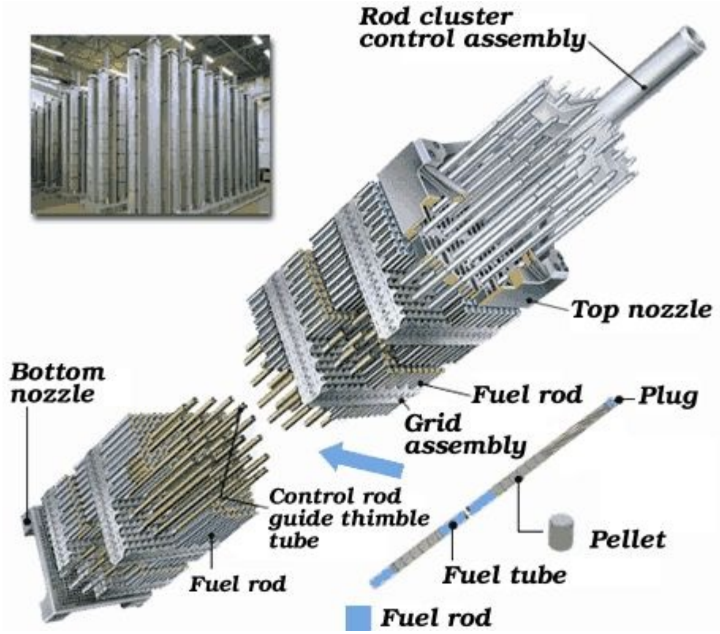
Heat removal systems

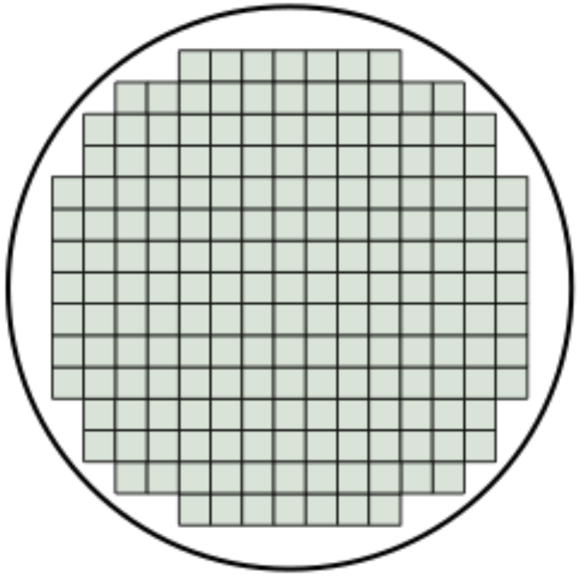
- Primary loop water runs through the core, transporting heat generated by the fuel, to a steam generator in a secondary water loop
- Steam drives a turbine, generating electricity
- A tertiary water loop helps to condense residual steam from the secondary loop via cooling towers and a water reservoir



Light Water Reactor Core Design

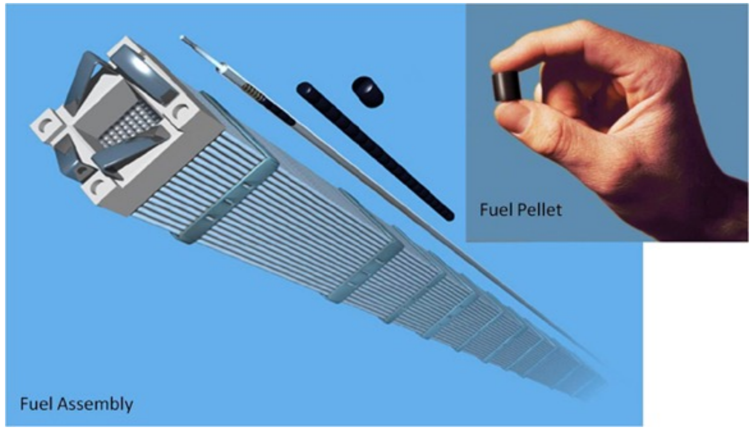
- An LWR core is comprised of fuel assemblies
- Each assembly contains a grid of fuel pins
  - In typical commercial LWR fuel designs, a 17x17 grid
  - Some pins are replaced by control rods
- Water flows from bottom to top

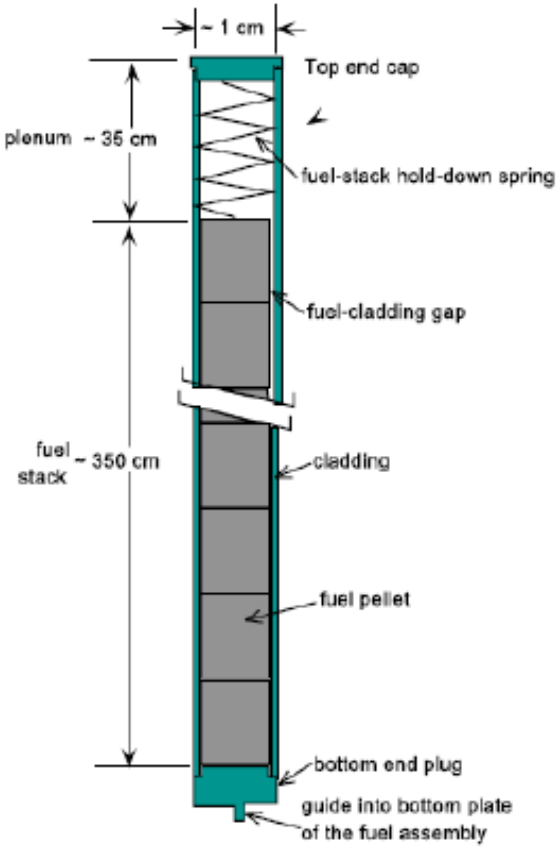



Westinghouse 4-loop PWR

LWR Fuel Pins

- LWR fuel pins are comprised of a hollow Zircaloy tube
  - This is the cladding
  - Zircaloy is a type of Zr alloy
- Inside the cladding are stacked UO2 pellets
- Each pellet is a cylinder about 1 cm in diameter and 1.5 cm in height



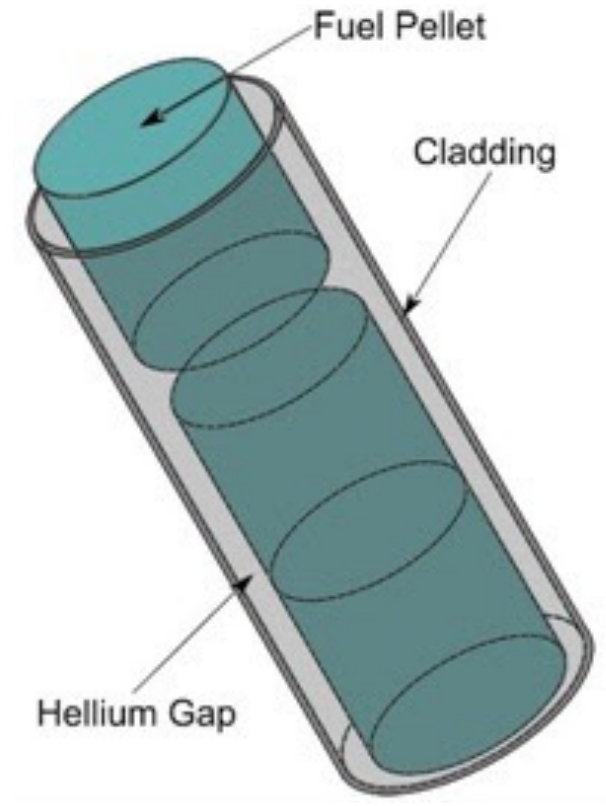


Most fuel designs employ some type of cladding

- The primary focus of the cladding is to separate the fuel from the coolant
  - Fuel contains radioactive fission products
  - Avoids corrosion of the fuel by the coolant
  - Keeps the fuel together, not blocking coolant flow
- The cladding should be thin and have a high thermal conductivity, so it doesn't trap any of the heat produced by the fuel
- Cladding should also be neutron transparent

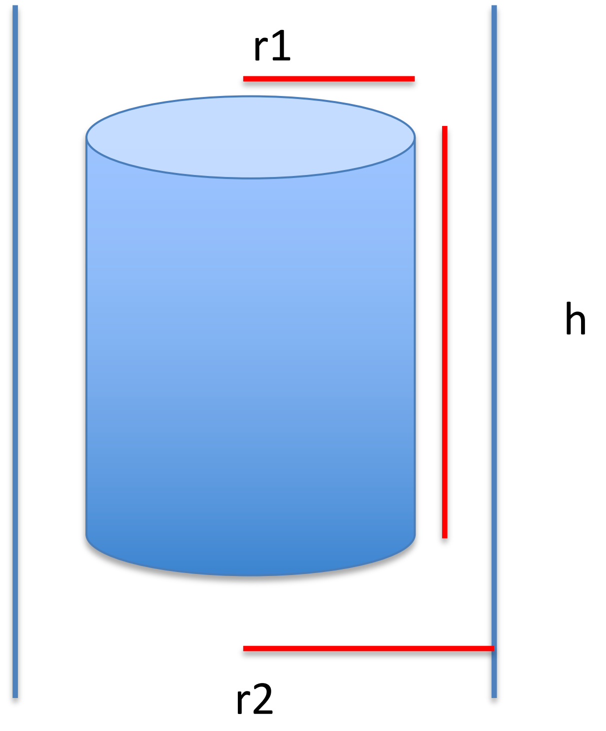
Fuel/Cladding Gap

- Fuel swells during reactor operation and the cladding creeps down around the fuel
- To avoid/limit both chemical and mechanical interaction, the pellet radius is smaller that the inner radius of the cladding
- In LWRs, the gap is filled with He gas, significantly impacting the heat transport
- In metal fuels, the gap is filled with liquid sodium, so there is little impact on the heat transport



Smear Density

- Smear density is the ratio of fuel volume to total internal volume of the fuel element
- Cylinder volume = $\pi r^2 h$
- Smear density = $\pi r_1^2 h / \pi r_2^2 h$
- Smear density = $r_1^2 / \, r_2^2$
- Typical smear density > 90%



Cladding material selection

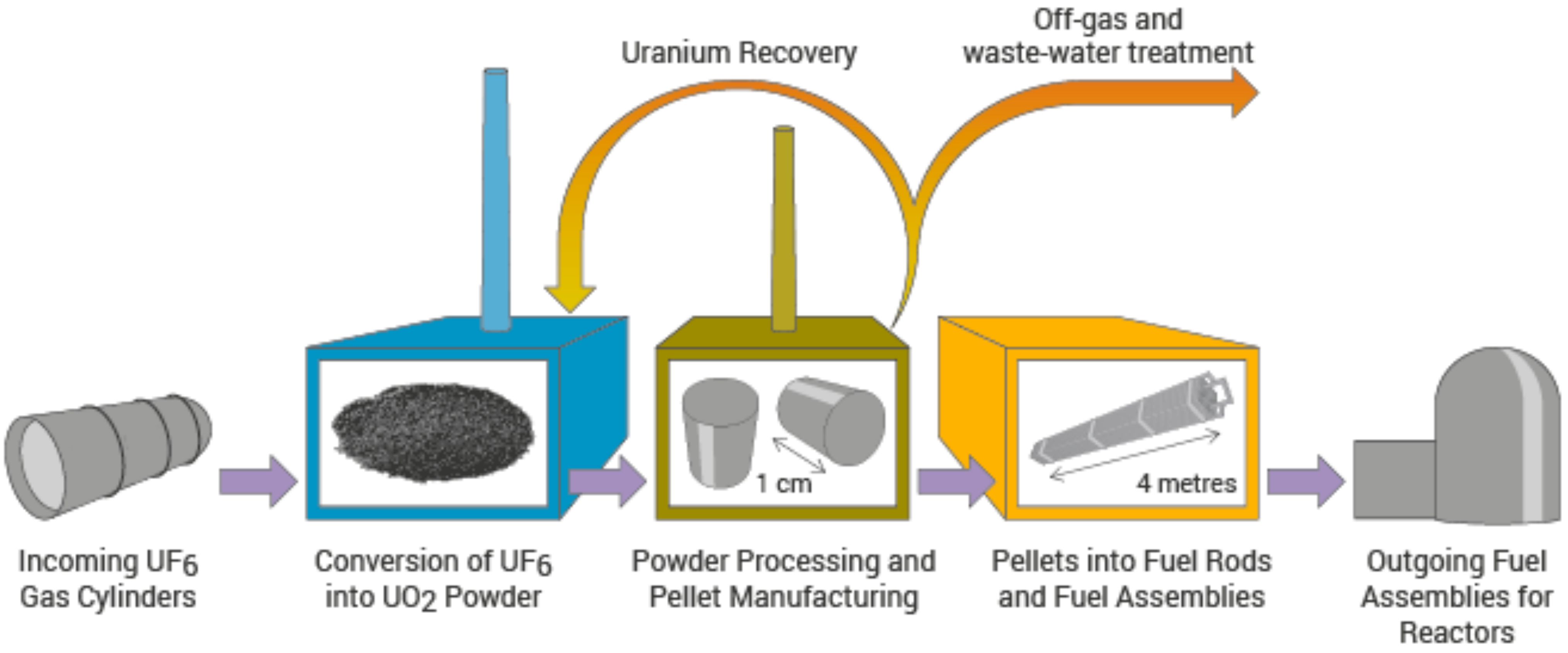
- Cladding must be compatible with the coolant, reasonably compatible with the fuel, have good thermal conductivity and reasonable radiation resistance
- Zirconium is used because of its
  - Low neutron cross section
  - Corrosion resistance in 300 C water
  - Resistance to void swelling
  - Adequate mechanical properties
  - Good thermal conductivity
  - Affordable cost
  - Available in large quantities
- Other cladding materials in use include
  - Stainless steel
  - Silicon Carbide
  - Ferritic-Martensitic steels like Fe-Cr and Fe-Cr-Al
  - Oxide dispersion strengthened (ODS) ferritic steels

Reactor Systems Wrap-up

- All reactors have basic requirements they must meet
  - An approach to remove the heat from the fuel, a method to convert heat to electricity, an approach to prevent radioactive products from leaving the fuel, a method to cycle the fuel, containment in case something goes very wrong
- Typically, the “fuel system” is thought to consist of the fuel itself, the gap, the cladding
  - primary coolant can also be included, or at least impacts of the coolant
- Cladding is the primary containment, and its integrity maintains safe operation
- LWRs have fuel pellets, a He gap (bond), and Zircaloy cladding

FUEL FABRICATION

Fabrication Process



Global Uranium Resources

Table 1: Typical natural uranium concentrations

Very high-grade ore (Canada) – 20% U	200,000 ppm U
High-grade ore – 2% U	20,000 ppm U
Low-grade ore – 0.1% U	1000 ppm U
Very low-grade ore* (Namibia) – 0.01% U	100 ppm U
Granite	3-5 ppm U
Sedimentary rock	2-3 ppm U
Earth's continental crust (av)	2.8 ppm U
Seawater	0.003 ppm U

Table 2: Uranium resources by country in 2021

	tonnes U	percentage of world
Australia	1,684,100	28%
Kazakhstan	815,200	13%
Canada	588,500	10%
Russia	480,900	8%
Namibia	470,100	8%
South Africa	320,900	5%
Niger	311,100	5%
Brazil	276,800	5%
China	223,900	4%
Mongolia	144,600	2%
Uzbekistan	131,300	2%
Ukraine	107,200	2%
Botswana	87,200	1%
USA	59,400	1%
Tanzania	58,200	1%
Jordan	52,500	1%
Other	266,600	5%
World total	6,078,500	

Table 3: Historical uranium production, 1945-2022

	Cumulative production (tU)
Canada	554,475
United States	378,038
USSR*	377,613
Kazakhstan	349,789
Australia	240,579
Germany	219,685
South Africa	165,692
Namibia	158,856
Niger	156,797
Czech Republic	112,055
Russia	90,725
Uzbekistan	76,808
France	76,021
China	53,712
Ukraine	24,670
Others	149,299
Total	3,184,812

world-nuclear.org

Uranium mining/processing

- Uranium ores are normally processed by grinding the ore materials to a uniform particle size and then treating the ore to extract the uranium by chemical leaching
- The milling process commonly yields dry powder-form material consisting of “yellowcake”, which was historically referred to as U_3O_8
- Most modern uranium recovery facilities produce a yellowish compound comprised mostly of uranyl peroxide dihydrate

Conversion

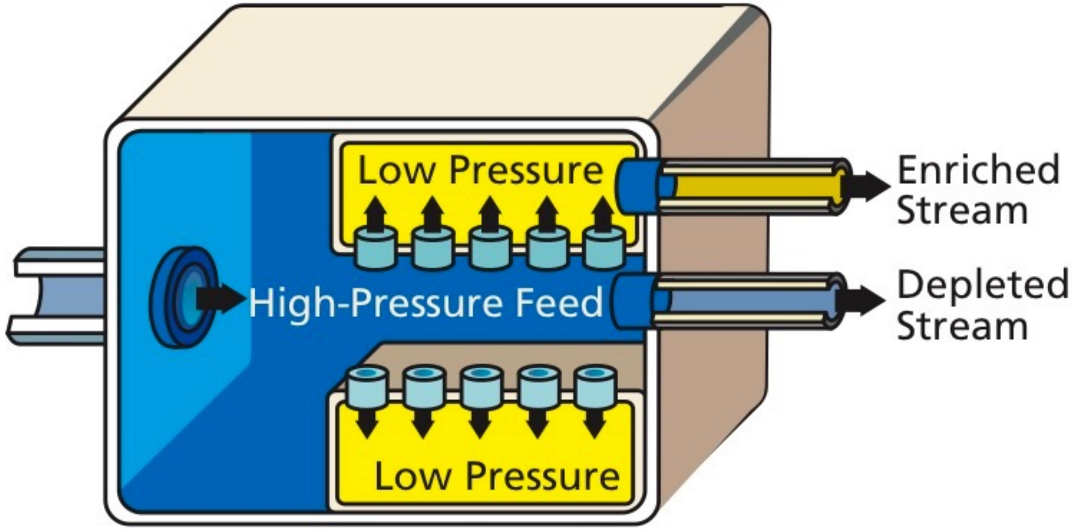
- Uranium enrichment requires uranium as uranium hexafluoride (gaseous state), which is obtained from converting uranium oxide to UF_6
- Uranium oxide can be reduced by hydrogen to produce UO2
  - $U_3O_8 + 2H_2 ===> 3UO_2 + 2H_2O$
- The oxide is then reacted with hydrogen fluoride to form uranium tetrafluoride (UF4)
  - $UO_2 + 4HF ===> UF_4 + 2H_2O$
- The tetrafluoride is then fed into a fluidized bed reactor with gaseous fluorine to produce uranium hexafluoride, UF6
  - $UF_4 + F_2 ===> UF_6$

Enrichment

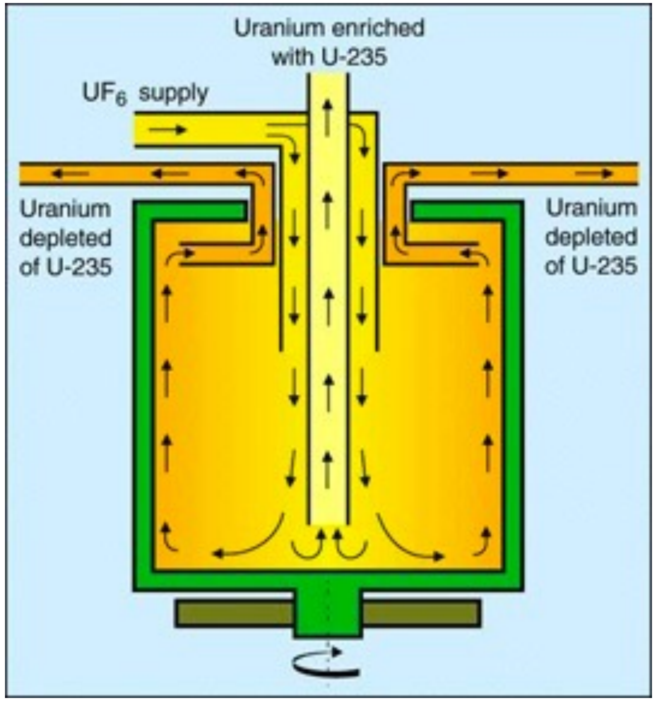
- Natural uranium only contains 0.7% U-235, and therefore must be enriched to obtain suitable fissile material for fuel (for most reactors)
- The difference in mass between U-235 and U-238 allows the isotopes to be separated and makes it possible to enrich the percentage of U-235
- The capacity of enrichment plants is measured in terms of 'separative work units' or SWU
- Two main enrichment processes
  - Gaseous diffusion: 2500 kWh per SWU
  - Centrifuge: 50 kWh per SWU

Enrichment

– Gaseous diffusion
  - Pushes UF6 through porous membrane
  - U235-F6 travels slightly faster
  - First Gen. technology, historical, but now outdated



– Centrifuge
  - gas is placed in a gas centrifuge cylinder and rotated at a high speed
  - strong centrifugal force, heavier gas molecules move towards the outside of the cylinder



SWUs

- The work W_{SWU} necessary to separate a mass F of feed of assay x_f into a mass P of product assay xp and tails of mass T and assay x_t is given by:

$$W_{\mathrm{SWU}} = P \cdot V(x_p) + T \cdot V(x_t) - F \cdot V(x_f)$$

- V is the value function:

$$V(x) = (2x - 1)\ln\left(\frac{x}{1-x}\right)$$

- The feed to product ratio (F/P) is given by the expression

$$\frac{F}{P} = \frac{x_p - x_t}{x_f - x_t}$$

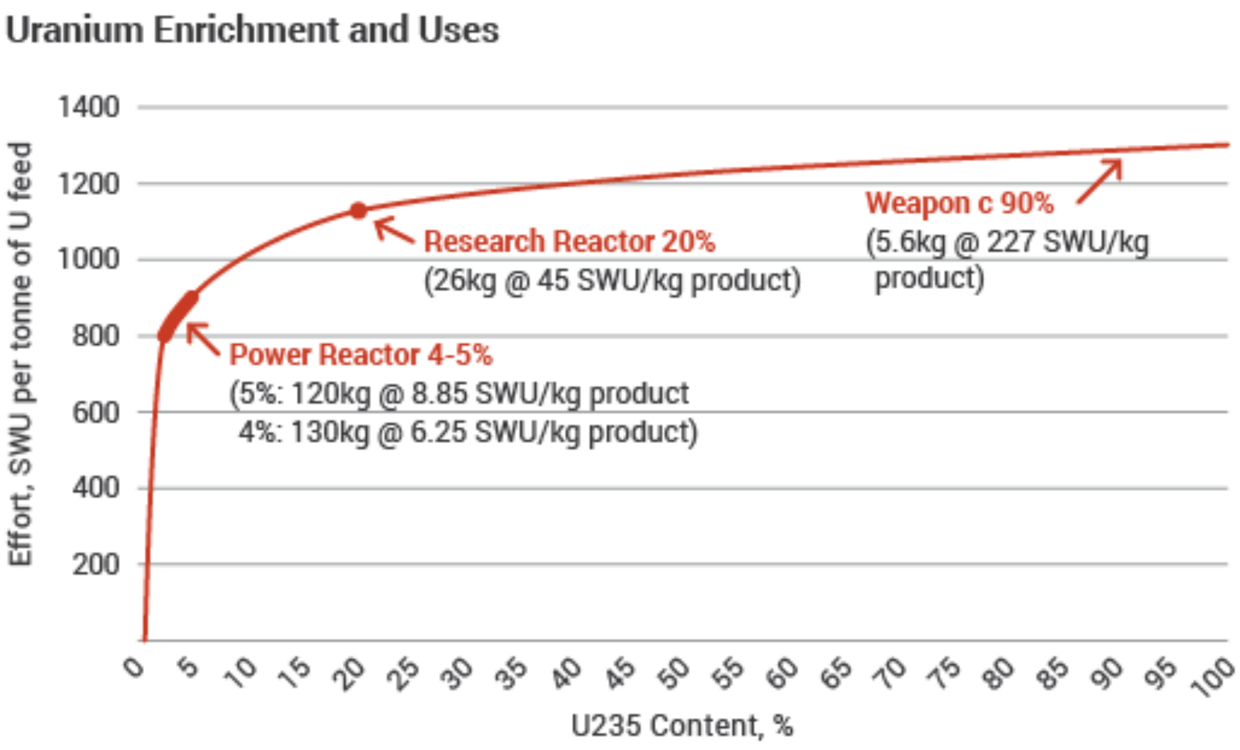
- The tails to product ratio (T/P) is given by the expression

$$\frac{T}{P} = \frac{x_p - x_f}{x_f - x_t}$$

- The same amount of separative work will require different amounts of energy depending on the efficiency of the separation technology

High Enriched Uranium

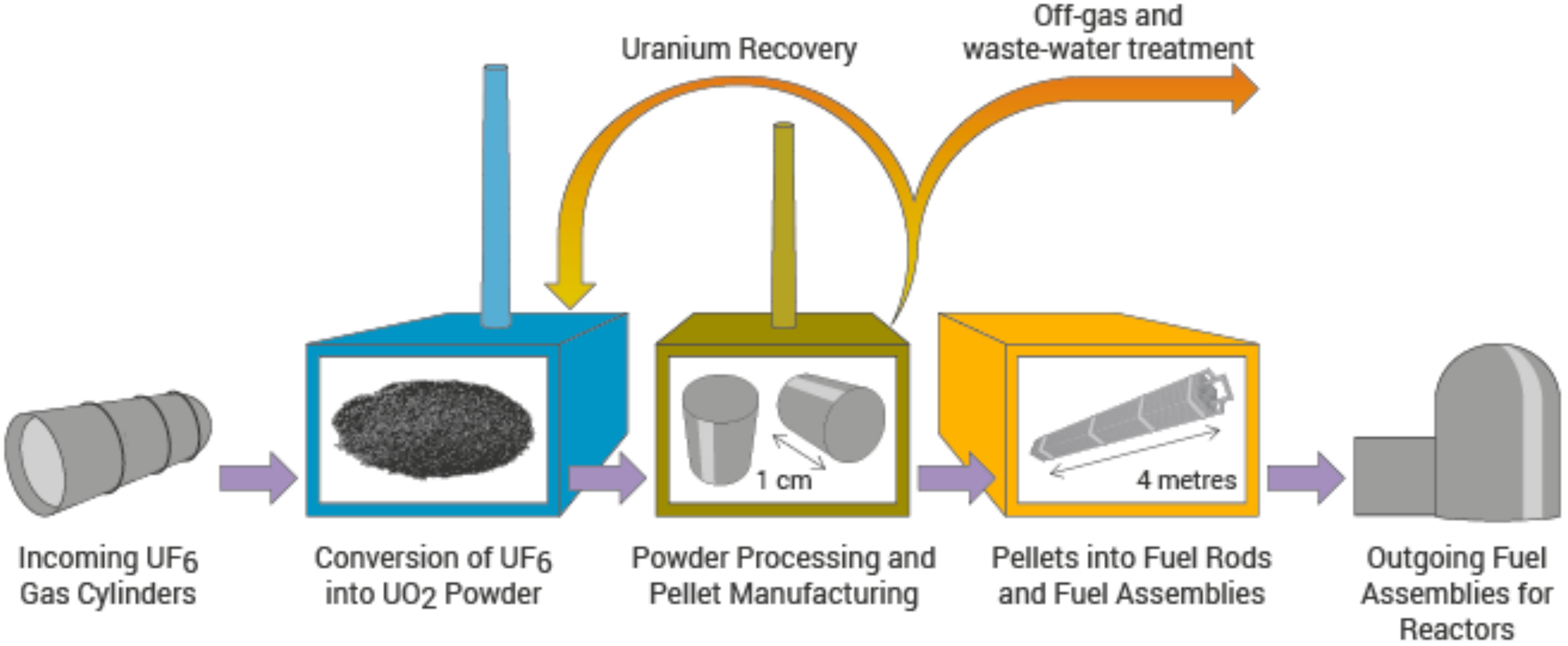
- One ton of natural uranium feedstock might end up: as 120-130 kg of uranium for power reactor fuel, as 26 kg of typical research reactor fuel, or conceivably as 5.6 kg of weapons-grade material
- The curve flattens out so much because the mass of material being enriched progressively diminishes, so requires less effort relative to what has already been applied to progress a lot further in percentage enrichment



Enrichment

- Enrichment accounts for almost half of the cost of nuclear fuel and about 5% of the total cost of the electricity generated
- It is also the main greenhouse gas impact from the nuclear fuel cycle where the electricity used for enrichment is typically generated from coal or natural gas
- However, it still only amounts to 0.1% of the carbon dioxide from equivalent coal-fired electricity generation if modern gas centrifuge plants are used

Fabrication Process



Powder Processing

- Uranium typically arrives at a fuel manufacturing plant as uranium hexafluoride (UF_6) and needs to be converted to uranium dioxide (UO_2) prior to pellet fabrication
- An example conversion process injects UF_6 into water to form a UO_2F_2 particulate slurry, ammonia (NH3) is added to this mixture and the UO_2F_2 reacts to produce ammonium diuranate (ADU, $(NH_4)2U_2O_7$), after which the slurry is filtered, dried and heated in a reducing atmosphere to pure UO_2
  - A reducing atmosphere is one in which oxidation is prevented by removal of oxygen and other oxidizing gases, and which may contain actively reducing gases such as hydrogen

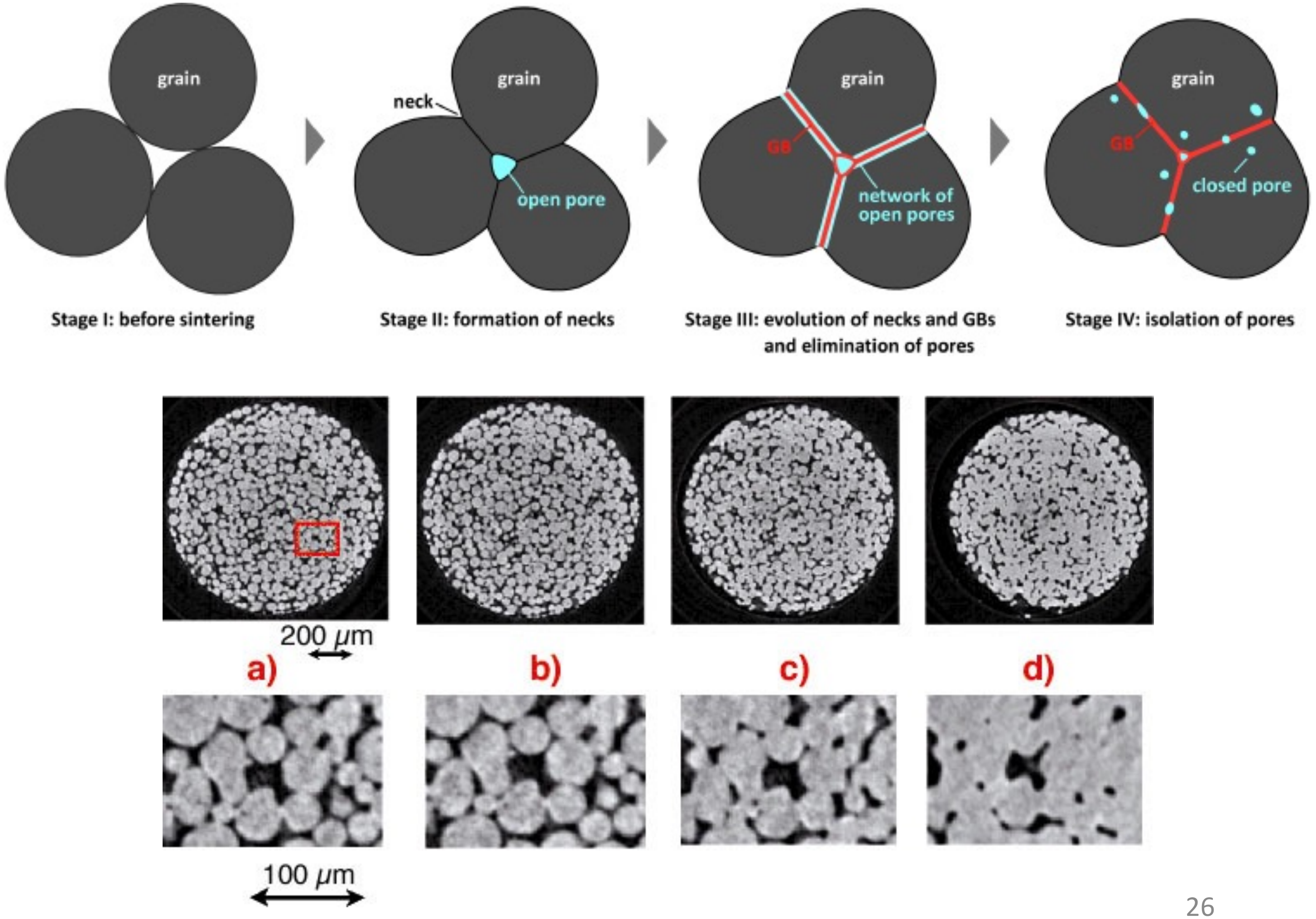
$$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF$$
$$2UO_2F_2 + 6NH_4OH \rightarrow (NH_4)_2U_2O_7 + 4NH_4F + 3H_2O$$
$$(NH_4)_2U_2O_7 + 2H_2 \rightarrow 2UO_2 + 2NH_3 + 3H_2O$$

Powder/Pellet Processing

- The UO_2 powder may need further processing or conditioning before it can be formed into pellets:
  - Homogenization: powders may need to be blended to ensure uniformity in terms of particle size distribution and specific surface area
  - Additives: U_3O_8 may be added to ensure satisfactory microstructure and density for the pellets and other fuel ingredients, such as lubricants, burnable absorbers (e.g., gadolinium) and pore-formers may also need to be added
- UO_2 powder is fed into dies and pressed biaxially into cylindrical pellet form using a load of several hundred MPa
- Pellets are then sintered in a heating furnace
  - Sintering is the process of compacting and forming a solid mass of material by heat or pressure

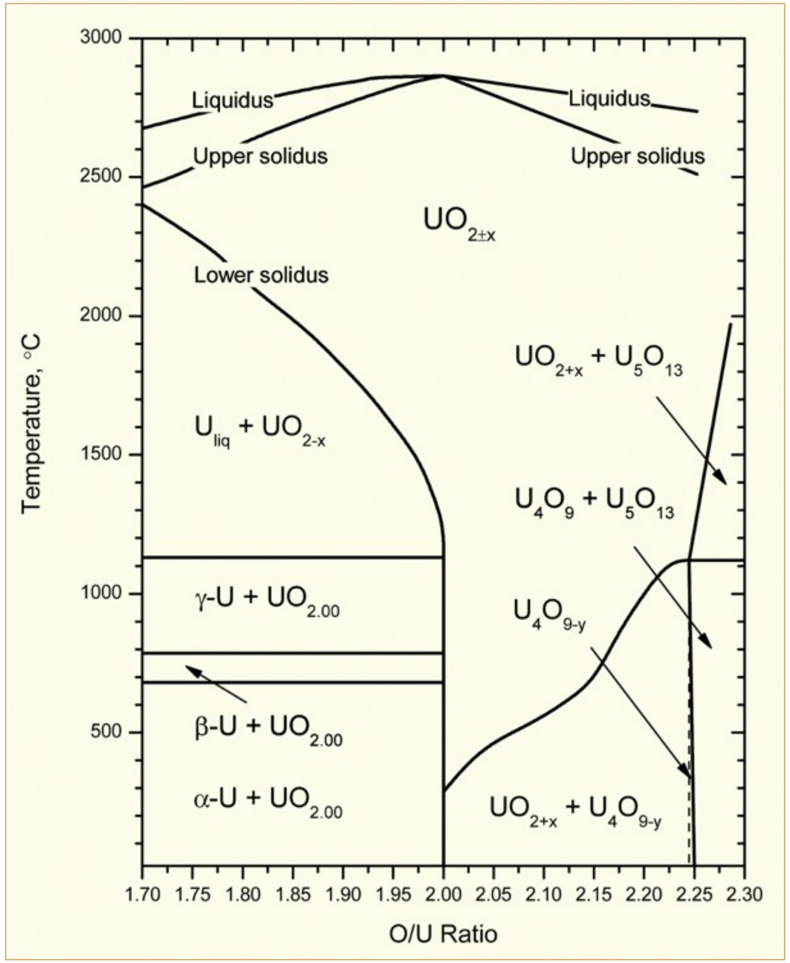
Sintering Process

- During sintering, atoms in the materials diffuse across the boundaries of the particles, fusing the particles together and creating one solid piece
- The final fuel pellets are nearly fully dense with a uniform microstructure: grain size ~ 10 μm; pore size ~ 3 μm; density ~ 95 - 99%
- The sintered microstructure (grain size and porosity) has a strong impact on the fuel evolution, and this processing stage can be tailored to meet microstructural needs



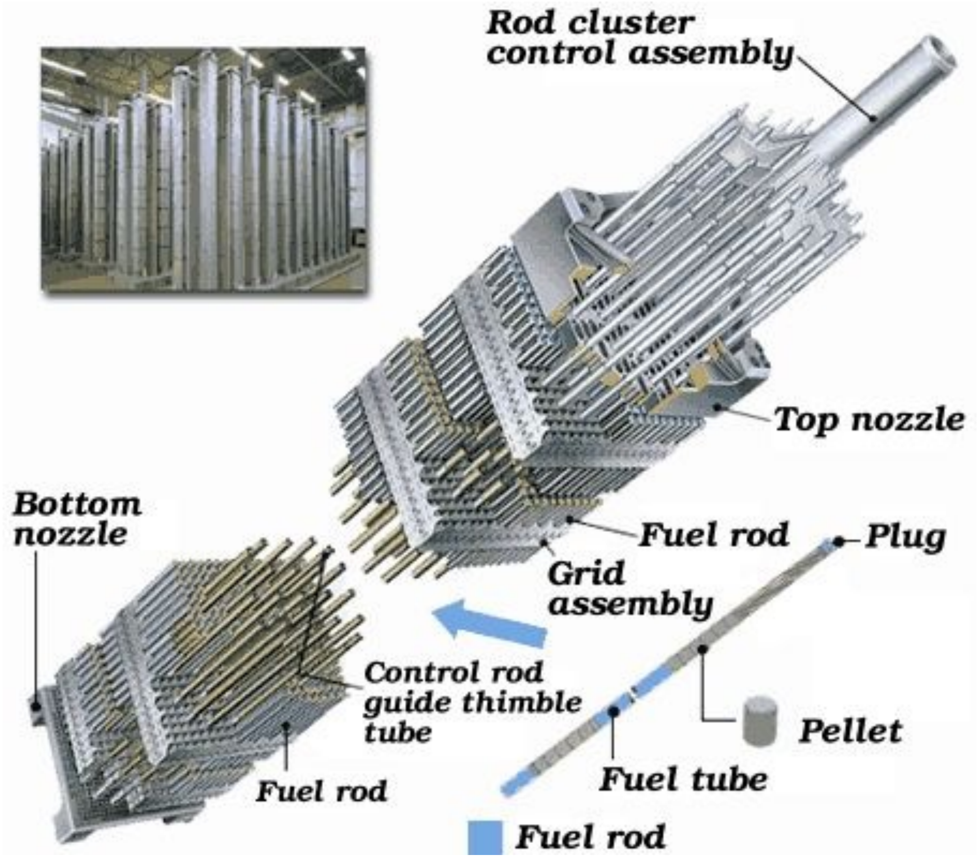
Fuel strictly manufactured to be UO_2

- Fuel fabricated to be nearly stoichiometric; i.e., $UO_{2.00\pm}$
  - Structure stable to T_{melt}
  - Maximum T_{melt}
- O/M ratio varies slightly during irradiation
- Large deviations from stoichiometry relevant to
  - Fabrication
  - Defected fuel behavior
  - Reprocessing
  - Accident conditions during dry storage or shipment of used nuclear fuel



Rods and Assemblies

- The fuel pellets are assembled in fuel rods and then put together in fuel assemblies
- Designs dictate that the pellet-filled rods have a precise physical arrangement in terms of their lattice pitch (spacing), and their relation to other features such as water (moderator) channels and control-rod channels
- Physical structures for holding the fuel rods are therefore engineered with extremely tight tolerances and are largely constructed of steel and zirconium alloys



Global Fuel Fabrication

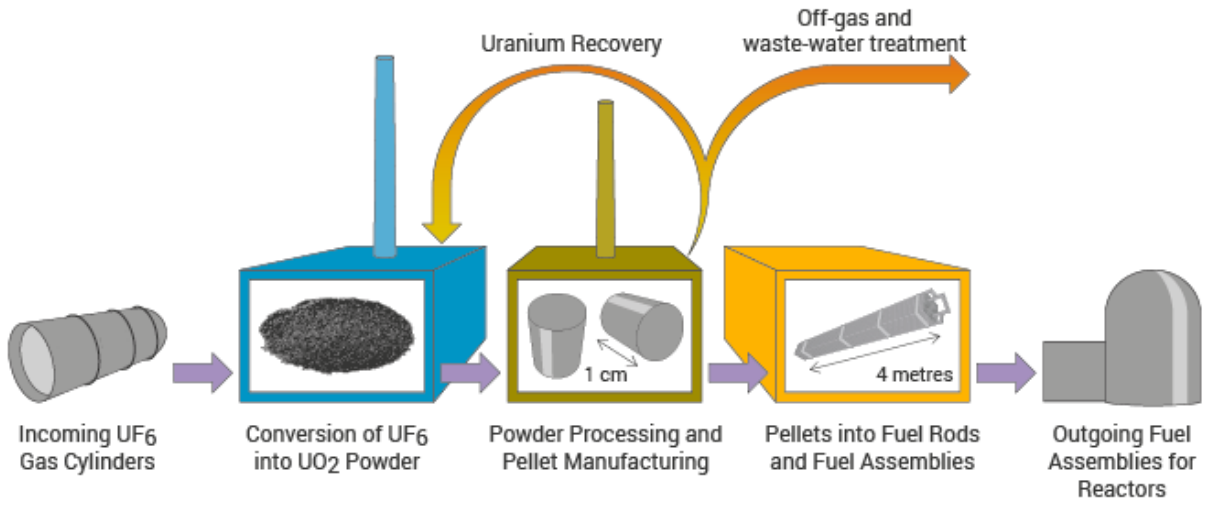
- LWR fuel pellets and rods/assemblies are manufactured in a number of countries
- USA, Russia, Kazakhstan and France are leaders
- There is a growing need for HALEU
  – High assay low enriched uranium
  – Uranium with 19.7% enrichment

Table 1: World LWR fuel fabrication capacity, tonnes/yr

	Fabricator	Location	Conversion	Pelletizing	Rod/assembly
Brazil	INB	Resende	160	160	240
China	CNNC	Yibin	400	400	450
		Baotou	200	200	200
France	AREVA NP-FBFC	Romans	1800	1400	1400
Germany	AREVA NP-ANF	Lingen	800	650	650
India	DAE Nuclear Fuel Complex	Hyderabad	48	48	48
Japan	NFI (PWR)	Kumatori	0	360	284
	NFI (BWR)	Tokai-Mura	0	250	250
	Mitsubishi Nuclear Fuel	Tokai-Mura	450	440	440
	Global NF-J	Kurihama	0	750	750
Kazakhstan	Ulba	Ust Kamenogorsk	2000	2000	0
Korea	KNFC	Daejeon	700	700	700
Russia	TVEL-MSZ*	Elektrostal	1500	1500	1560
	TVEL-NCCP	Novosibirsk	450	1200	1200
Spain	ENUSA	Juzbado	0	500	500
Sweden	Westinghouse AB	Västeras	600	600	600
UK	Westinghouse**	Springfields	950	600	860
USA	AREVA Inc	Richland	1200	1200	1200
	Global NF-A	Wilmington	1200	1000	1000
	Westinghouse	Columbia	1500	1500	1500
Total			13958	15418	13832

Fuel Fabrication Summary

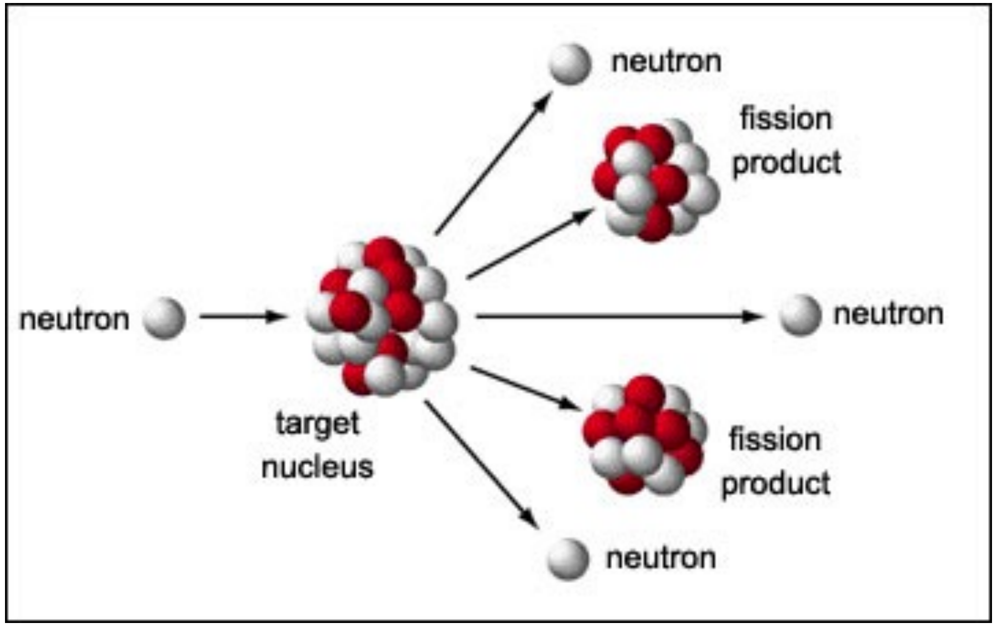
- Mining -> Processing -> Conversion -> Enrichment -> Powder -> Compaction/Sintering -> Rod/Assembly
- U_3O_8 must be converted to UF_6 for enrichment, which is then converted to UO_2 powder for pellet manufacture
- UO_2 microstructure from fabrication strongly impacts fuel performance



HEAT GENERATION

Fission basics

- Impinging neutron of a given energy
  - Neutron energy determines cross section which determines probability of fission event
- Neutron + Target Nucleus -> Two fission products, 2-3 neutrons
- Fission releases around 210 MeV of energy
  - 170 MeV to fission fragments
  - 2 MeV per neutron
  - 7 MeV gamma rays
  - Balance radioactive decay



Energy release with different nuclei

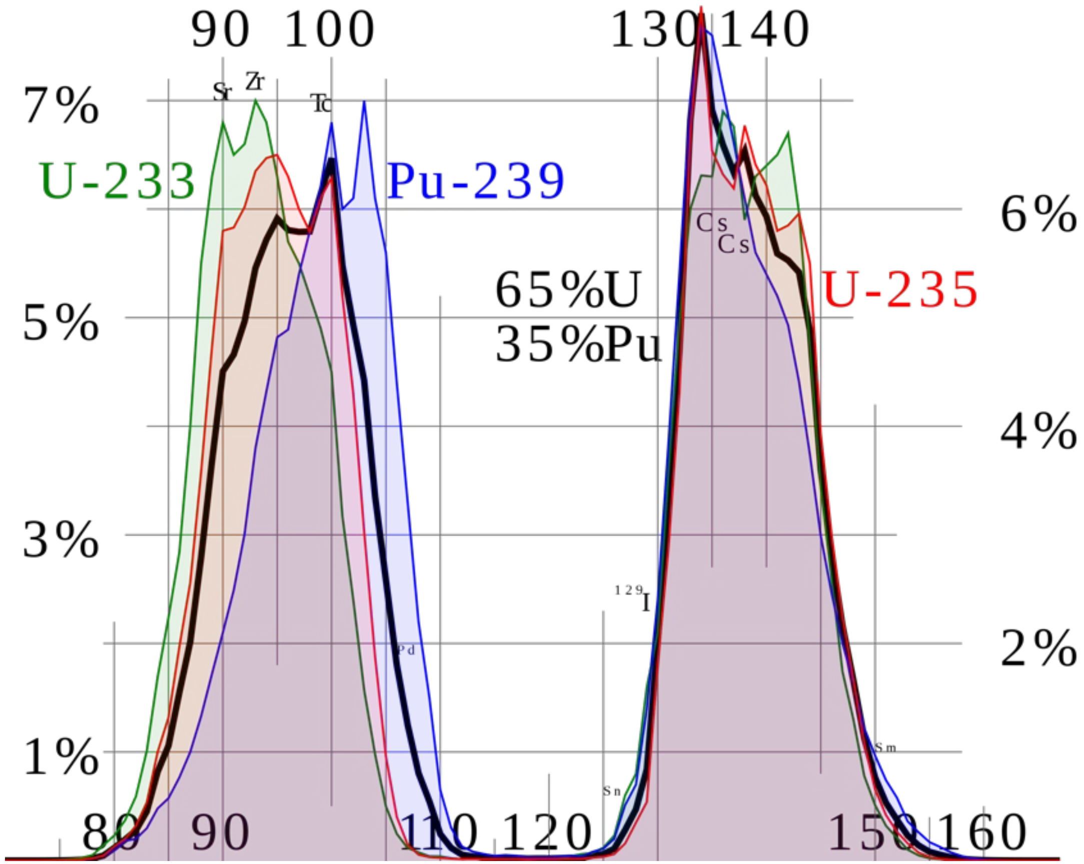
- Energy release is effectively agnostic with regards to the fissioning species
- Comparing U-235 with Pu-239 on the right
  - Pu releases about 9 MeV more usable energy per fission
  - Less than a 5% difference
- Partition of energy is largely identical as well

Source	Energy, MeV/f	
	^{235}U	^{239}Pu
Energy released instantaneously		
Kinetic energy of fission fragments	169.1	175.8
Kinetic energy of prompt neutrons	4.8	5.9
Energy of prompt γ-rays	7	7.8
Energy of γ-rays from $n\gamma$ capture	8.8	11.5
Energy from decay of fission products		
Energy of β^--particles	6.5	5.3
Energy of delayed γ-rays	6.3	5.2
Energy of anti-neutrinos[1]	8.8	7.1
Total available energy	202.5	211.5

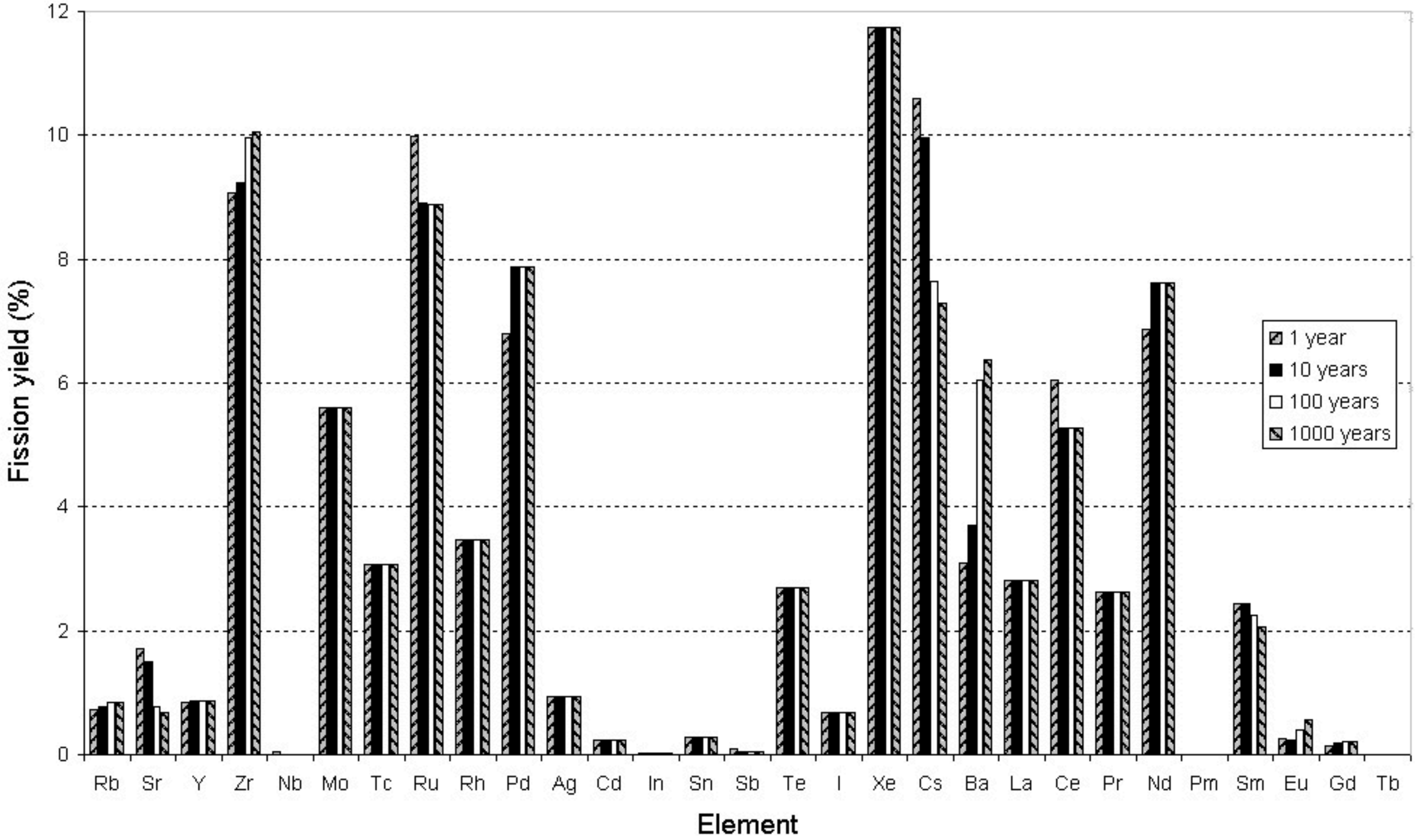
Note 1: Anti-neutrino energy is not absorbed in the reactor and does not contribute to the total available yield.

Fission product yield

- Regardless of fissioning isotope, fission product yields are effectively the same, in this double hump distribution
- One broad peak centered around A=95, the other around A=135
- Examples:
  - Mo (Z=42, A=96)
  - Cs (Z=55, A=133)



Fission Product Yields



Calculating heat generation rate for a given fuel

- We know about 200 MeV of energy is available due to a fission (210 MeV minus neutrinos)
- We know the fission cross section of the target nuclide (tabulated)
- We can calculate the fission atom density
- The heat generation rate, Q is given by:
  - $Q = E_f \times N_f \times \sigma_f \times \phi$
  - Where E_f is the fission energy, N_f is the fission atom density, σ_f is the fission cross section, and ϕ is the neutron flux
  - Units: J/fission x atoms/cm^3 x (fission/neutron)*(cm^2/atom) x (neutron/cm^2-s) = J/cm^3-s = W/cm^3

Calculating heat generation rate for a given fuel

- Cross sections:
  - ENDF database: Nuclear Data Sheets 148 (2018) 1–142
  - Thermal neutron (E=0.025 eV) U235 fission cross section: ~586.8 barns
    - 1 barn = 10^{-24} cm^2
- Fission atom density
  - Atom density of U-235 = UO2 density x 1/molar mass x Avogadro's number x atom fraction x enrichment

Example