Nuclear Fuel Performance

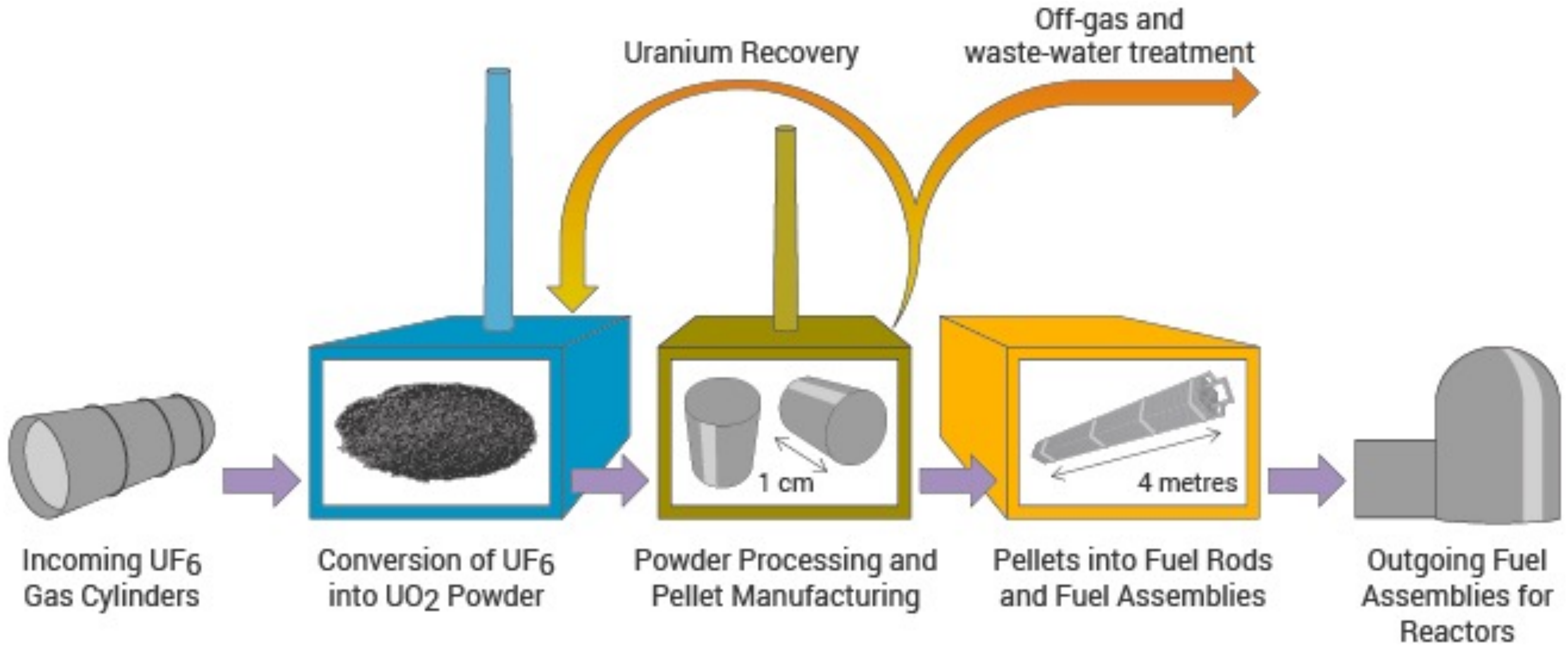
NE 533 Spring 2022

Last Time

- Finished fuel/reactor type overview
- 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
- LWRs have a certain way of meeting these requirements, but there are other options
- Various fuel geometries have been used
- Cladding is often required between the fuel and the coolant

FUEL FABRICATION

Fabrication Process



Uranium deposit types

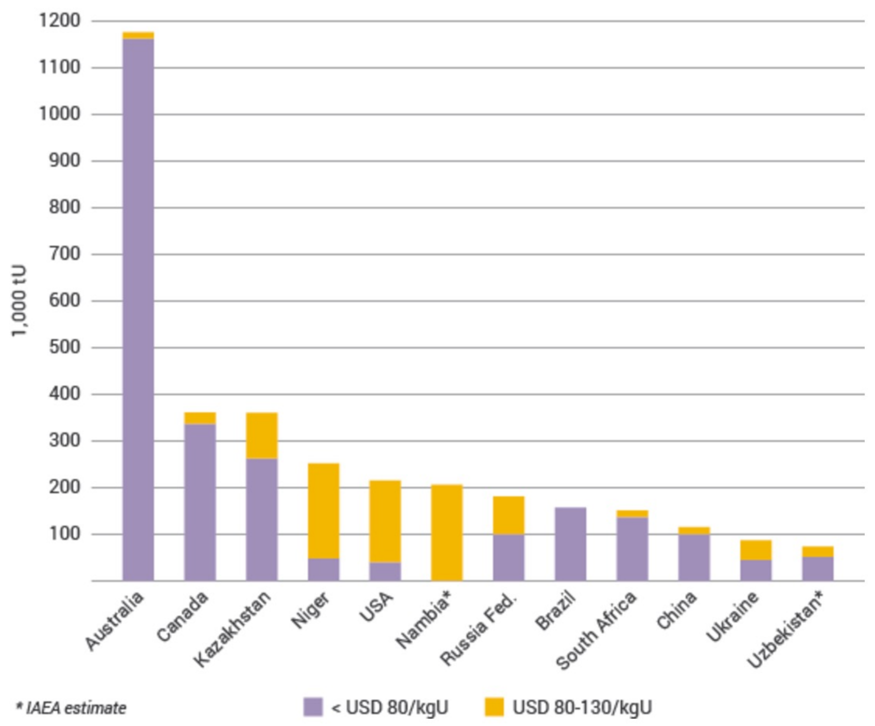
- There are mainly three types of uranium deposits
  - Sedimentary
    - Often found in sandstone; common in Canada and western US
  - Igneous/hydrothermal
    - Vein-type uranium ores from geothermal activity; Greenland and Namibia
  - Breccial
    - found in rocks that have been broken due to tectonic fracturing, or weathering; common in India, Australia and the US
- Less common means of uranium mining include seawater recovery, where U concentrations is 3.3 micrograms per liter

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

Reasonably Assured Resources of Uranium in 2009



Known Recoverable Resources of Uranium 2015

	tonnes U	percentage of world
Australia	1,664,100	29%
Kazakhstan	745,300	13%
Canada	509,000	9%
Russian Fed	507,800	9%
South Africa	322,400	6%
Niger	291,500	5%
Brazil	276,800	5%
China	272,500	5%
Namibia	267,000	5%
Mongolia	141,500	2%
Uzbekistan	130,100	2%
Ukraine	115,800	2%
Botswana	73,500	1%
USA	62,900	1%
Tanzania	58,100	1%
Jordan	47,700	1%
Other	232,400	4%
World total	5,718,400	

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 is U_3O_8

Conversion

- Uranium enrichment requires uranium as uranium hexafluoride, 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

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:

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

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

- V is the value function:
- The feed to product ratio is given by the expression
- The tails to product ratio is given by the expression
- The same amount of separative work will require different amounts of energy depending on the efficiency of the separation technology

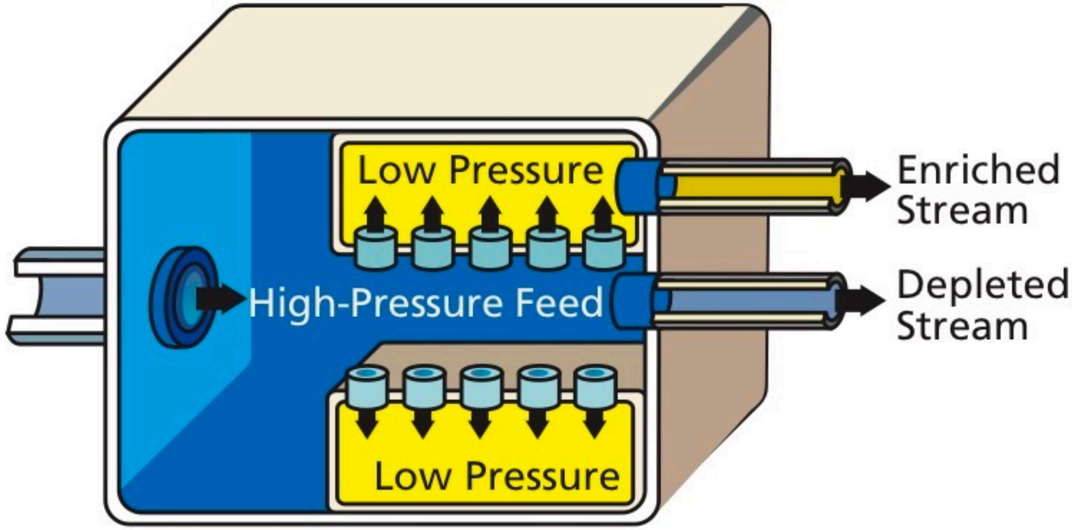
$$\frac{F}{P} = \frac{x_p - x_t}{x_f - x_t}$$

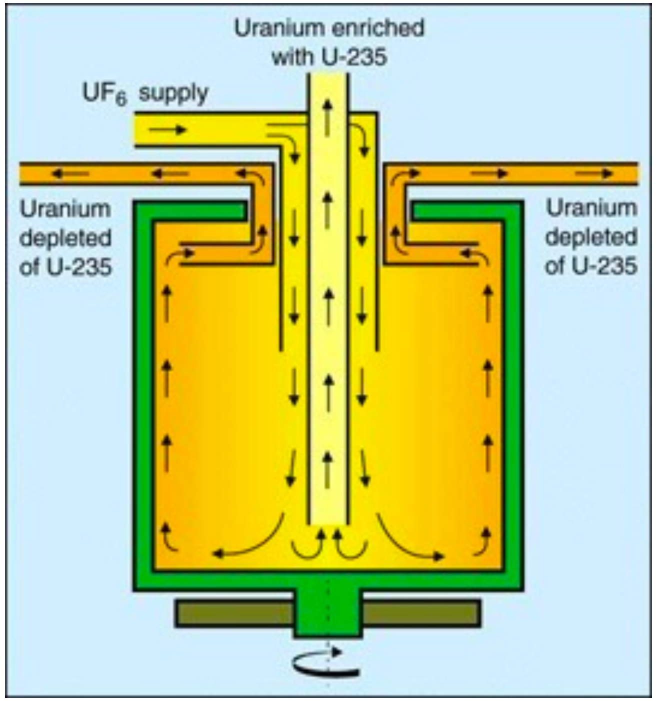
$$\frac{T}{P} = \frac{x_p - x_f}{x_f - x_t}$$

Enrichment

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

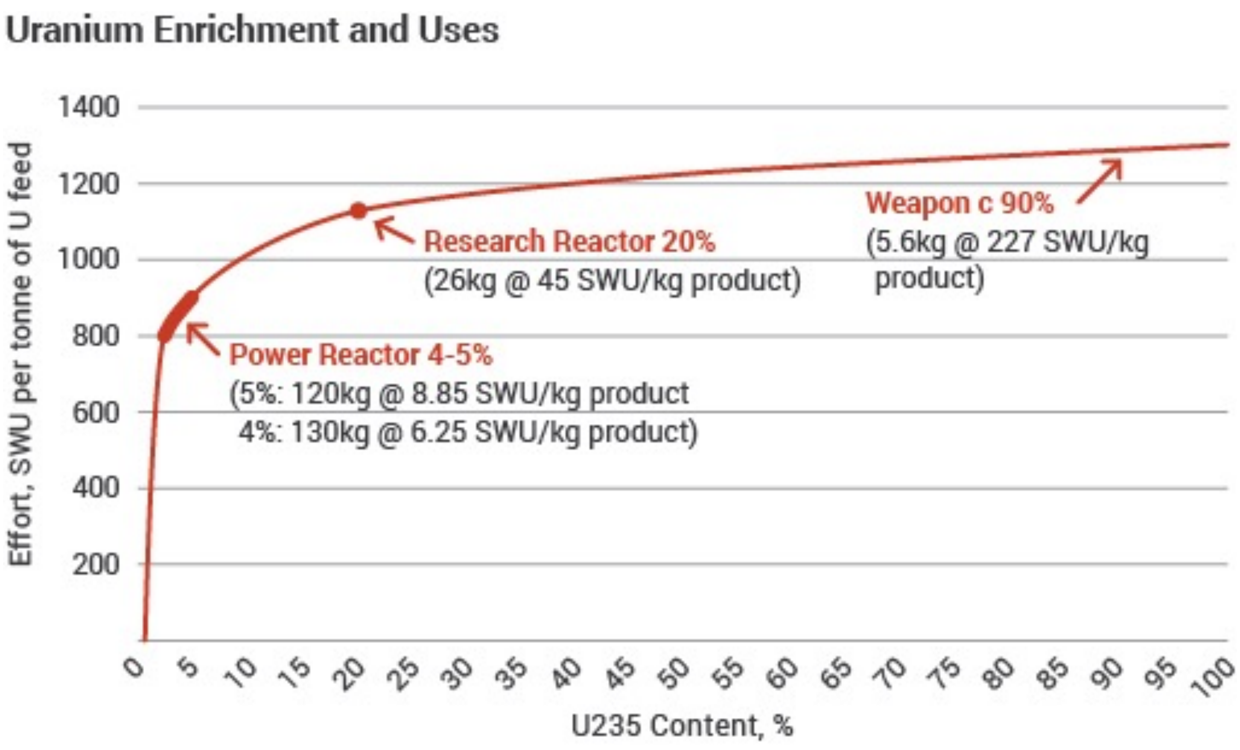
– Centrifuge
  - gas is placed in a gas centrifuge cylinder and rotated at a high speed
  - strong centrifugal force, heavier gas molecules move towards the outside of the cylinder





Enrichment

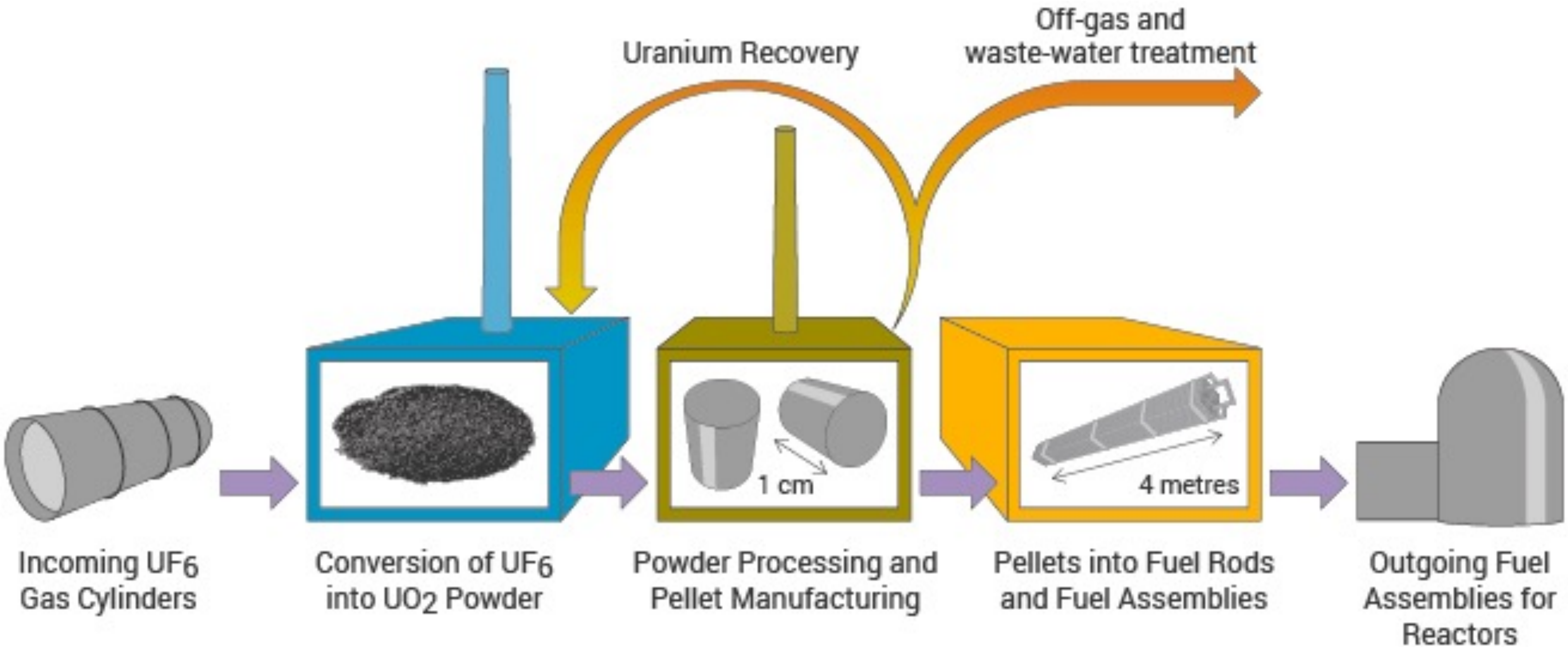
- One ton of natural uranium feedstock might end up: as 120-130 kg of uranium for power reactor fuel, as 26 kg of typical research reactor fuel, or conceivably as 5.6 kg of weapons-grade material
- The curve flattens out so much because the mass of material being enriched progressively diminishes, so 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

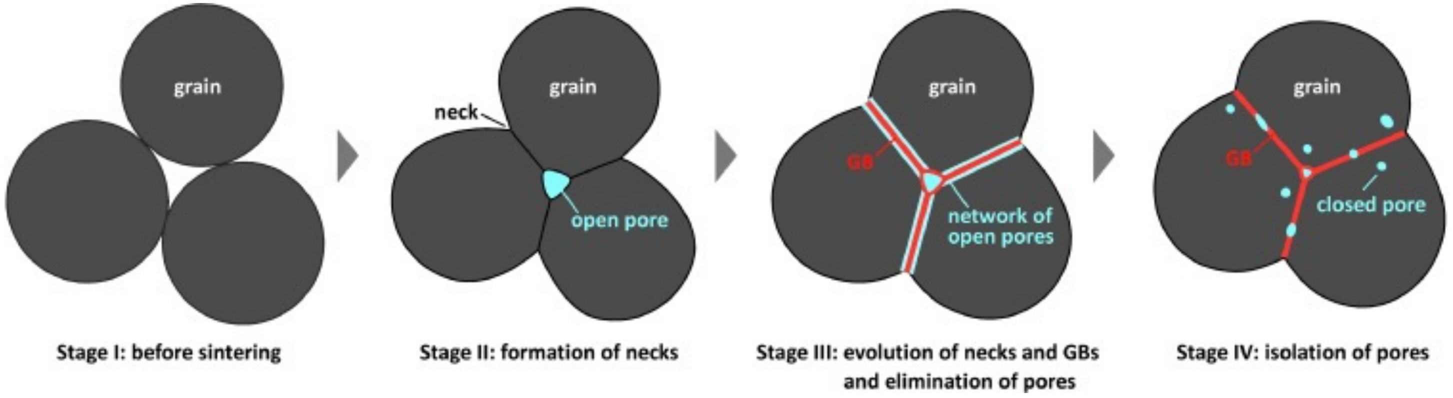
- Uranium typically arrives at a fuel manufacturing plant as uranium hexafluoride (UF_6) and needs to be converted to uranium dioxide (UO_2) prior to pellet fabrication
- An example conversion process injects UF_6 into water to form a UO_2F_2 particulate slurry, ammonia (NH3) is added to this mixture and the UO_2F_2 reacts to produce ammonium diuranate (ADU, $(NH_3)2U_2O_7$), 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

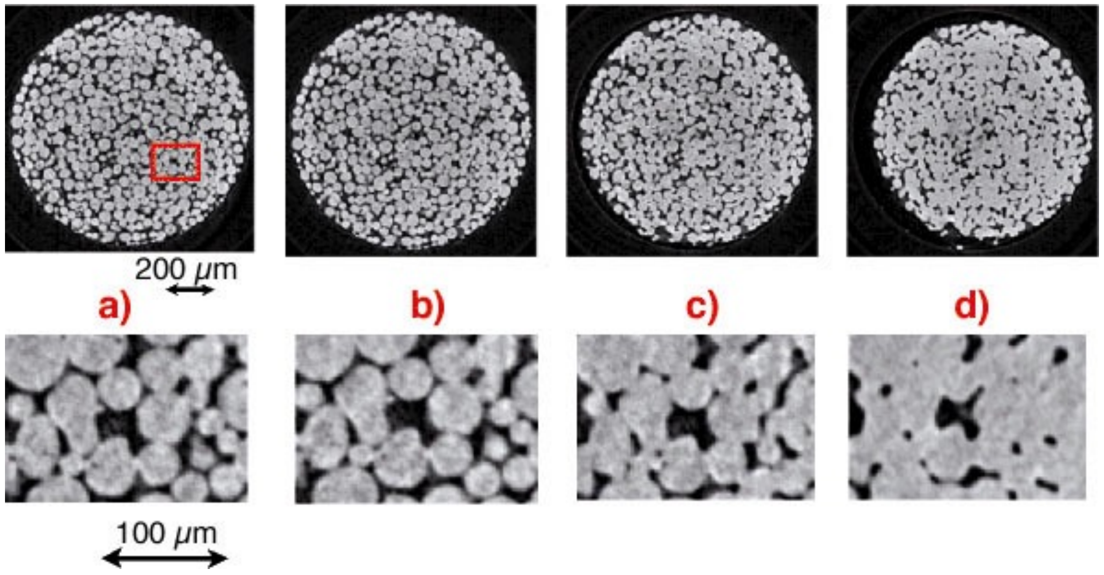
Powder/Pellet Processing

- 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

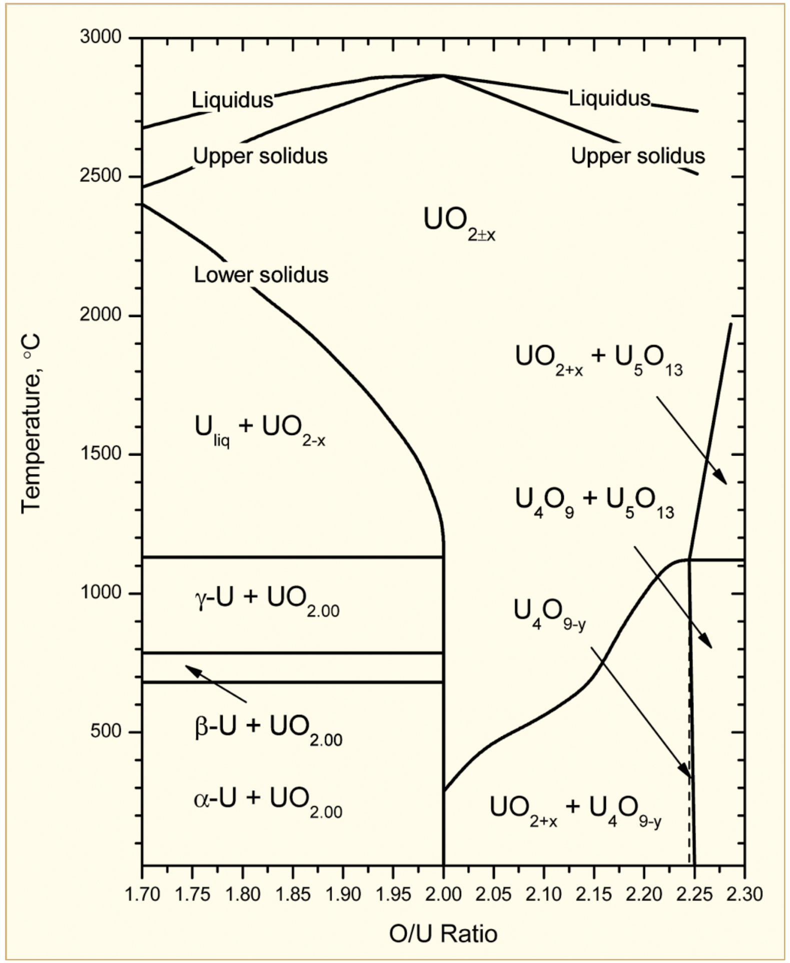
- During sintering, atoms in the materials diffuse across the boundaries of the particles, fusing the particles together and creating one solid piece
- The final fuel pellets are nearly fully dense with a uniform microstructure: grain size ~ 10 µm; pore size ~ 3 µm; density ~ 95 - 99%
- A single pellet in a typical reactor yields about the same amount of energy as one ton of coal





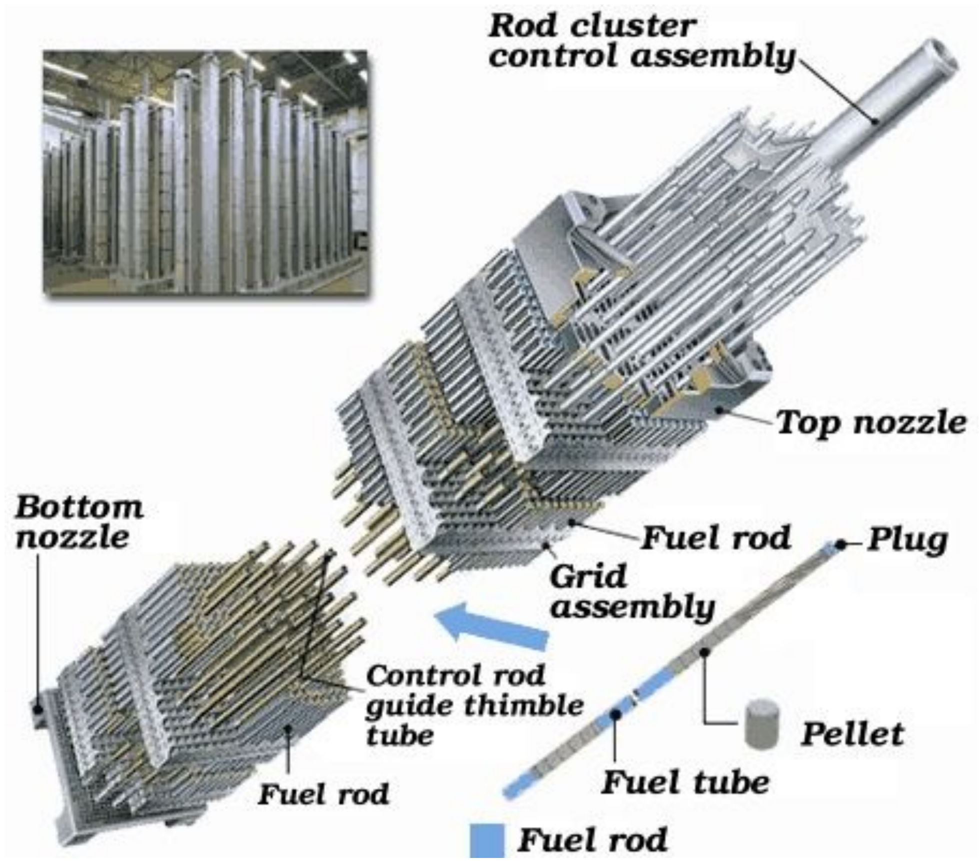
Fuel strictly manufactured to be UO_2

- 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

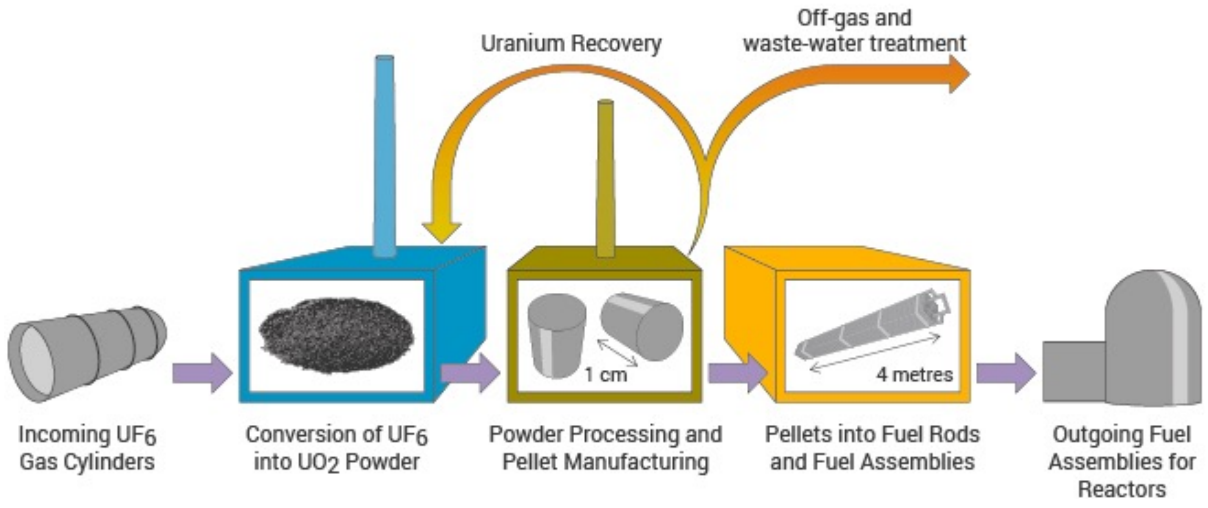
- Uranium is mined and converted into fuel in a number of countries
- USA, Russia, Kazakhstan and France are leaders
- There is a growing need for HALEU
  - High assay low enriched uranium
  - Uranium with 19.7% enrichment

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

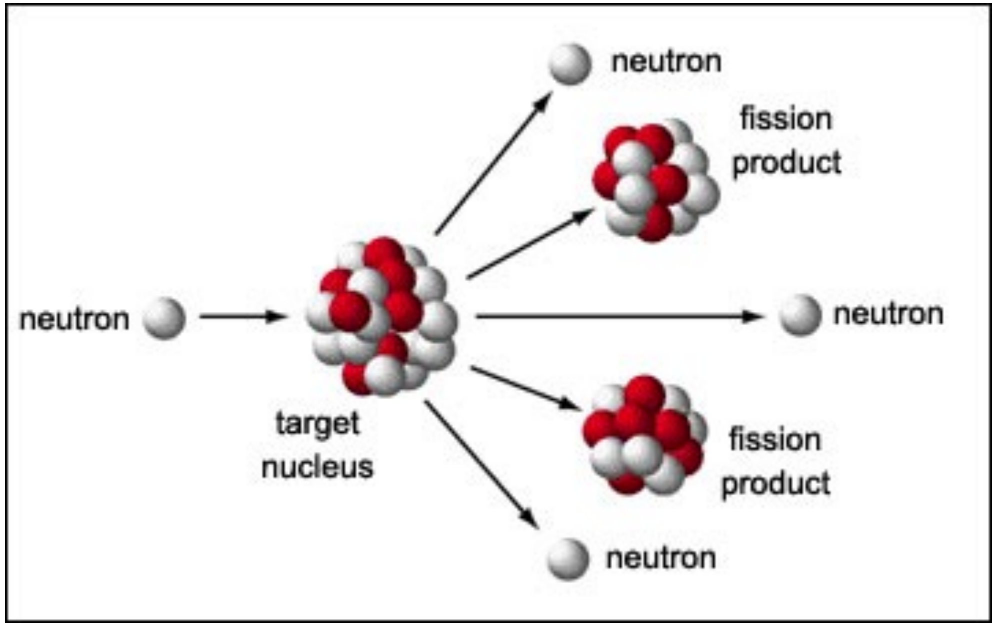
- Mining -> Processing -> Conversion -> Enrichment -> Powder -> Compaction/Sintering -> Rod/Assembly
- U_3O_8 must be converted to UF_6 for enrichment, which is then converted to UO_2 powder for pellet manufacture
- For different fuel types, enriched UF_6 follows a different path



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

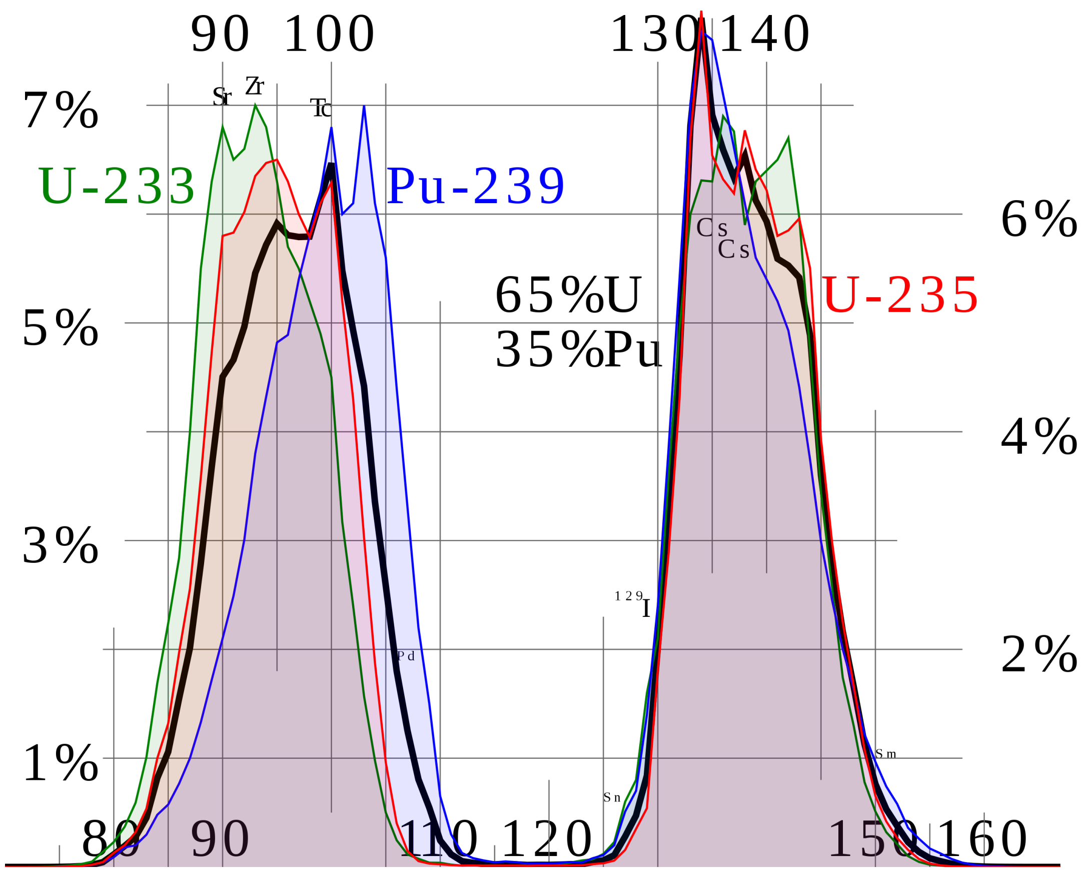
- Energy release is effectively agnostic with regards to the fissioning species
- Comparing U-235 with Pu-239 on the right
  - Pu releases about 9 MeV more usable energy per fission
  - Less than a 5% difference
- Partition of energy is largely identical as well

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

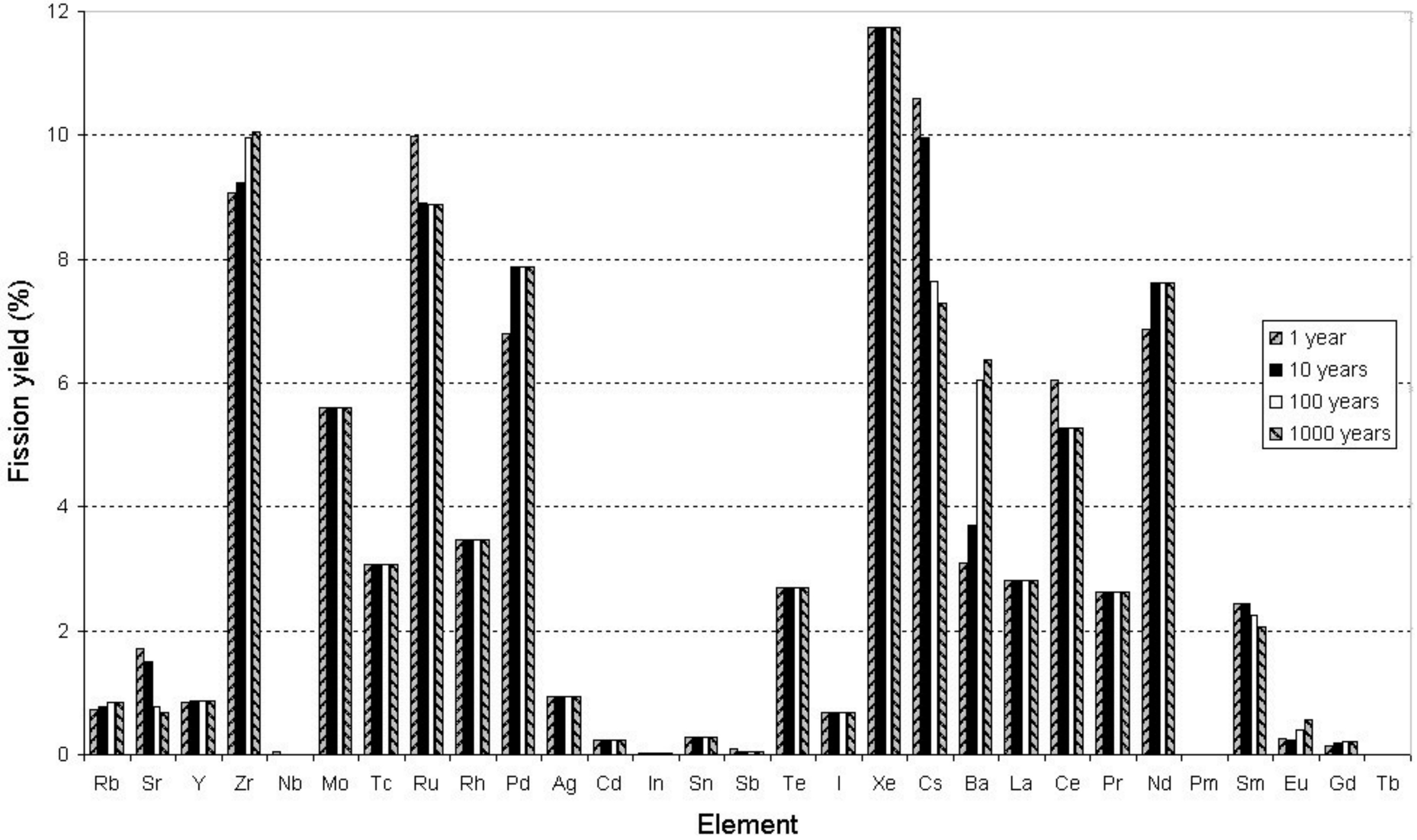
Note 1: Anti-neutrino energy is not absorbed in the reactor and does not contribute to the total available yield.

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 = UO_2 density x 1/molar mass x Avogadro's number x atom fraction x enrichment

Calculating heat generation rate for a given fuel

- Given a density of UO_2 (10.97 g/cm^3)
- Enrichment of 3%
- Molar mass of 3% enriched UO2
  - 235*0.03 + 238*0.97 + 2*16 = 269.9 g/mol
- Atom density of U-235 = 10.97 x 1/269.9 x 6.022E23 x 1/1 x 0.03
  - 7.34E20 atoms/cm^3
- Given a flux of 5E13 neutrons/cm^2/s
- $Q = E_f \times N_f \times \sigma_f \times \phi$
  - 200E6 eV x 1.602E-19 J/eV x 7.34E20 atoms/cc x 587E-24 cm^2 x 5E13 n/cm^2/s
- Q = 690 J/s/cm^3 = 690 W/cm^3

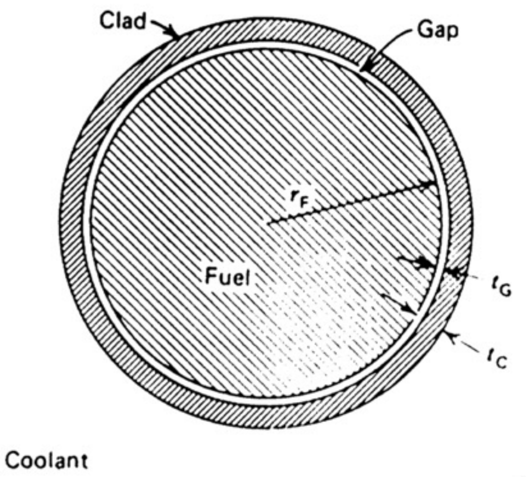
Some notes

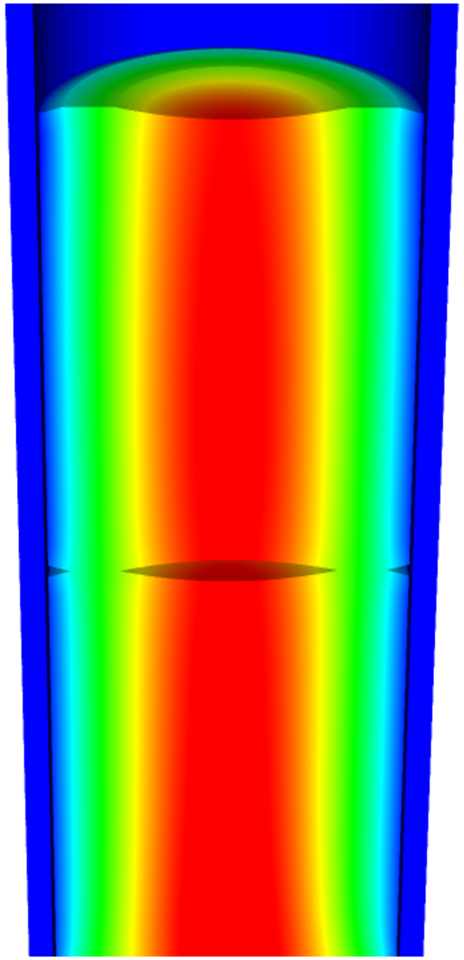
- Fast neutron cross section ~100x less than thermal neutron cross section
- Fuels for fast neutron spectrum typically have high enrichments, 19.7% U-235
- Historical research reactor fuels, such as UMo and USi, have had an enrichment of 90+%
- Neutron flux will vary depending on the reactor
  - HFIR has a peak neutron flux of 3E15 n/cm^2/s
  - PULSTAR has a peak neutron flux of 1E13 n/cm^2/s
- Significant variability in heat generation depending on fuel type and reactor conditions

HEAT TRANSPORT

Heat transport route

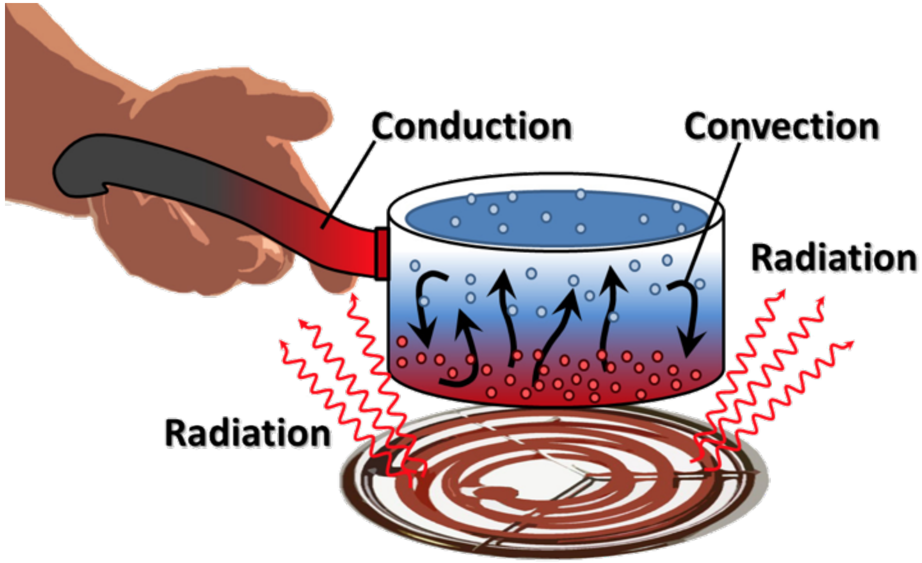
- Heat is produced in the fuel, transports through the cladding and gap, and into the coolant
- Important quantities include
  - Volumetric heat generation rate Q (W/cm^3)
  - Fuel Centerline temperature T_0
  - Surface temperature of the fuel T_F
  - Inner cladding temperature T_{CI}
  - Outer cladding temperature T_{CO}
  - Coolant temperature T_{cool}
  - Fuel pellet radius r_F
  - Gap thickness t_G
  - Cladding thickness t_c





Heat can be transported in three ways

- Convection
  - Heat transfer through mass movement of liquid or gas
- Radiation
  - Heat transfer by means of photons in electromagnetic waves
- Conduction
  - Heat transfer by molecular or atomic motion



Heat transfer mode in fuel systems?

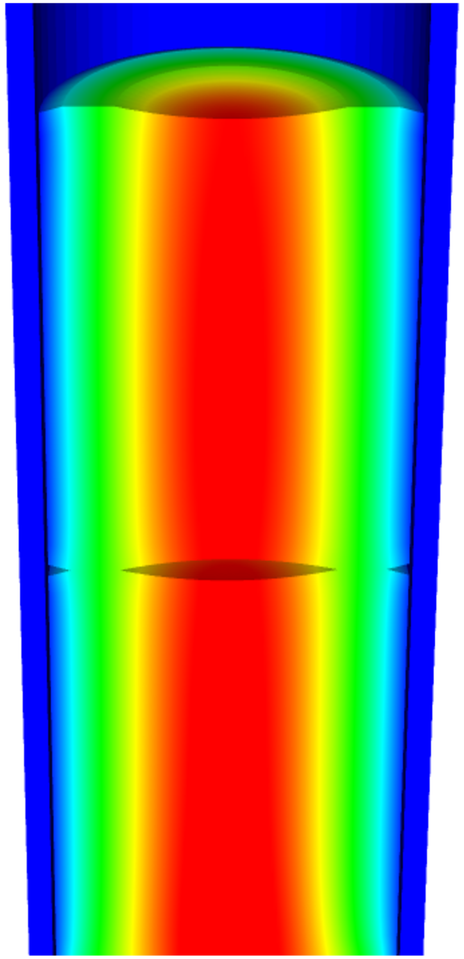
- How is heat transported through the fuel?

  Conduction
- How is the heat transported through the gap?

  Mostly conduction, some convection
- How is heat transported through the cladding?

  Conduction
- How is heat transported to the coolant?

  Convection



Heat conduction equation

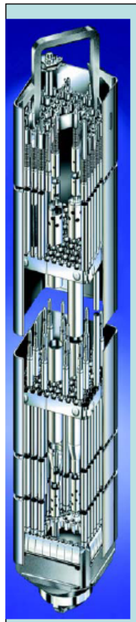
- ρ is the density, c_p is the specific heat, T is the temperature, t is the time, and k is the thermal conductivity
- It is a partial differential equation in time and space
- We are solving for the T as a function of space and time
  - T(**x**, t), where **x** is a vector defining the position in space
- What do we need to know to solve this equation?

$$\rho c_p \frac{\partial T}{\partial t} - \nabla \cdot (k \nabla T)$$

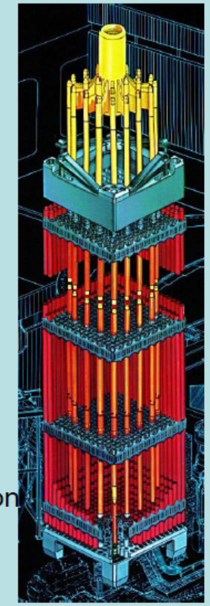
- The geometry of our problem
- The initial condition of T
- The boundary conditions of T
- Is each parameter is a function of T
- If they aren't a function of T, do they vary in space and time for some other reason?

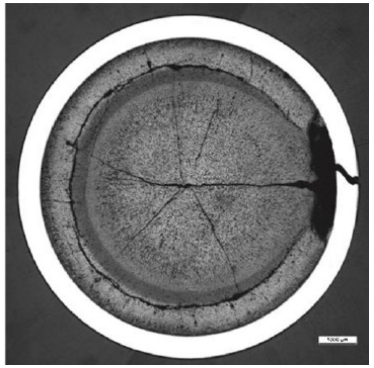
What is our geometry for the problem?

- Reactor geometry depends on reactor type
- The ideal geometry of each fuel rod is axisymmetric, but in reality it is 3D
- Fuel pellet defects cause 3D geometry
- The stacked pellets may not be stacked perfectly, causing their center axis to not be aligned, also causing 3D geometry



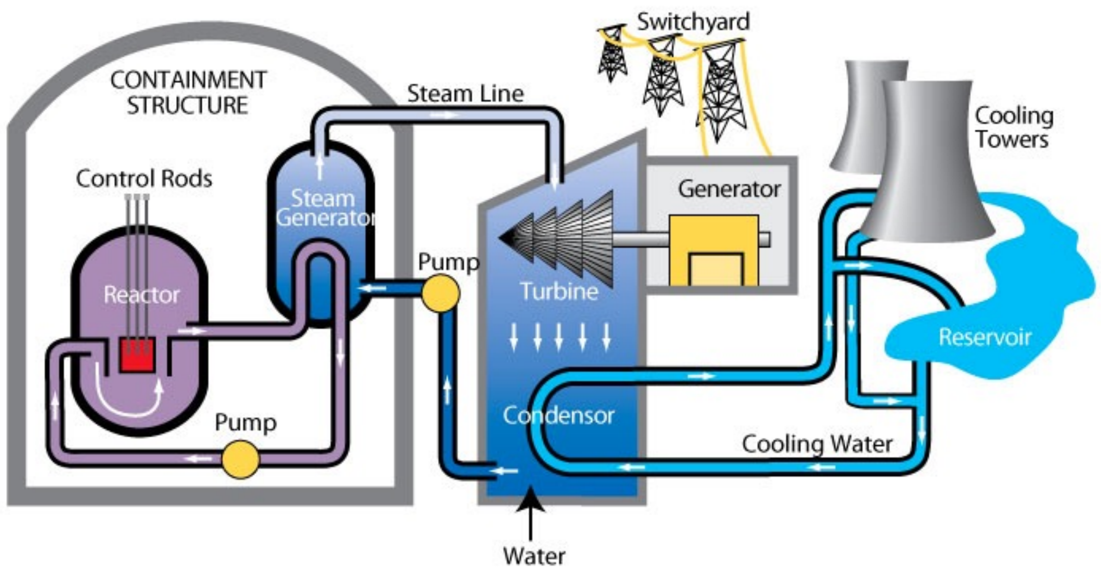
	BWR	PWR
Lattice	10x10	14x14 – 18x18
Lattice size	~5.3”	~9”
Height	120”-150”	144”-168”
Fuel	UO_2/MOx	UO_2/MOx
Fuel rods	~92	176-300
Part length rods	~14	0
Non-fueled rods	~2	20-25
Control	Ext. control rod	Int. control cluster
Cladding	Zr2 for PCI, nodular corrosion	Zr4/Zirlo/M5 for uniform corrosion & hydrogen
Channels	Yes	No
Fuel mass	~180 kgU	~600 kgU





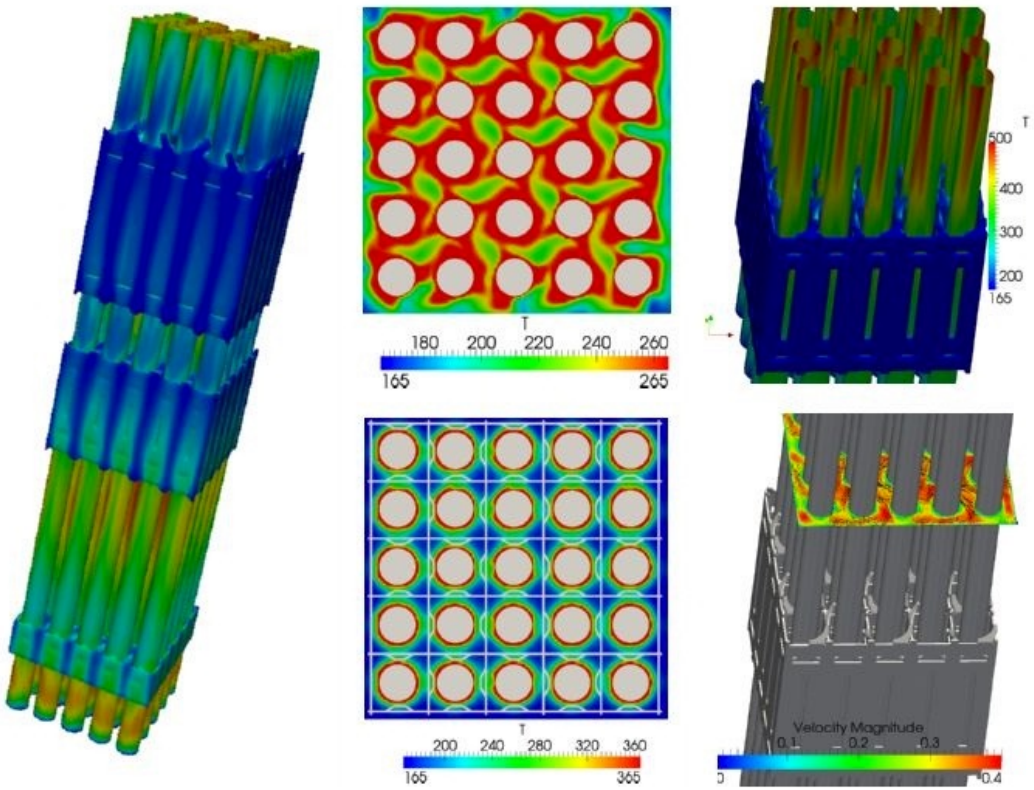
The initial condition of T

- The initial condition of T is set by the state of the reactor directly before startup
- What is the initial temperature profile of the fuel?
- The initial temperature is uniform throughout the fuel
- It is equal to the initial coolant temperature
- $T(\mathbf{x}, 0) = T_{cool}(0)$



Boundary conditions?

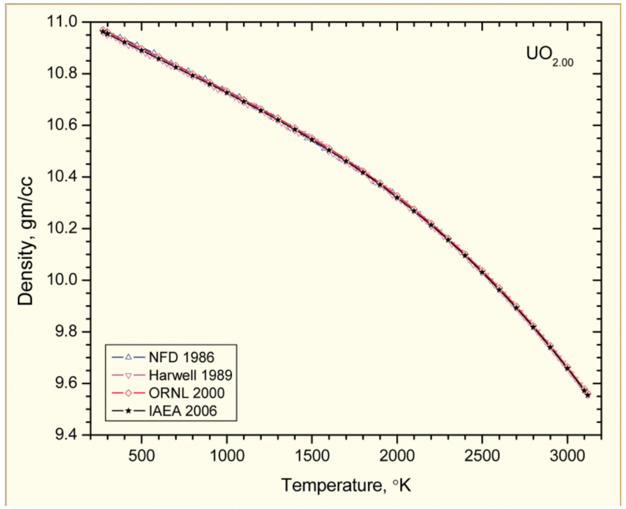
- The boundary conditions on T is set by the coolant flow
- The temperature of the coolant T_{cool} is complicated
  - It varies along the length of the fuel rod (axially)
  - It various around the circumference of the fuel rod

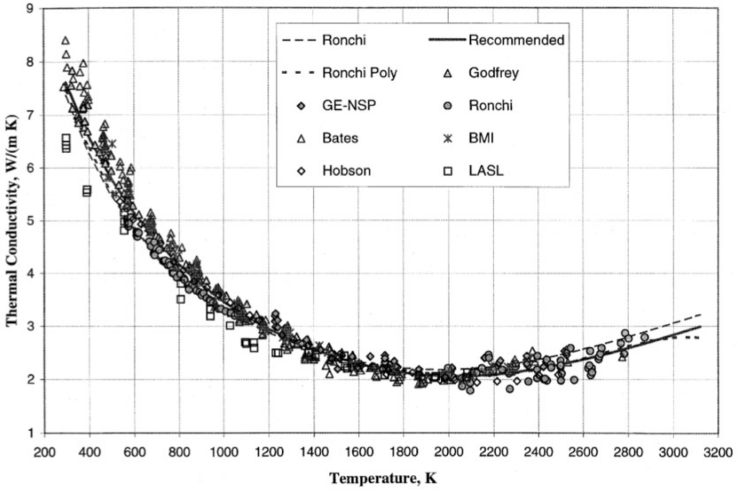


Fuel properties

- Density varies as a function of T (thermal expansion)
  - Also varies as a function of composition (thus as a function of burnup/time)
- Thermal conductivity also varies with temperature

$$k_0 = \frac{100}{7.5408_{17.629t} + 3.6142t^2} + \frac{6400}{t^{5/2}} exp\left(\frac{-16.35}{t}\right)$$

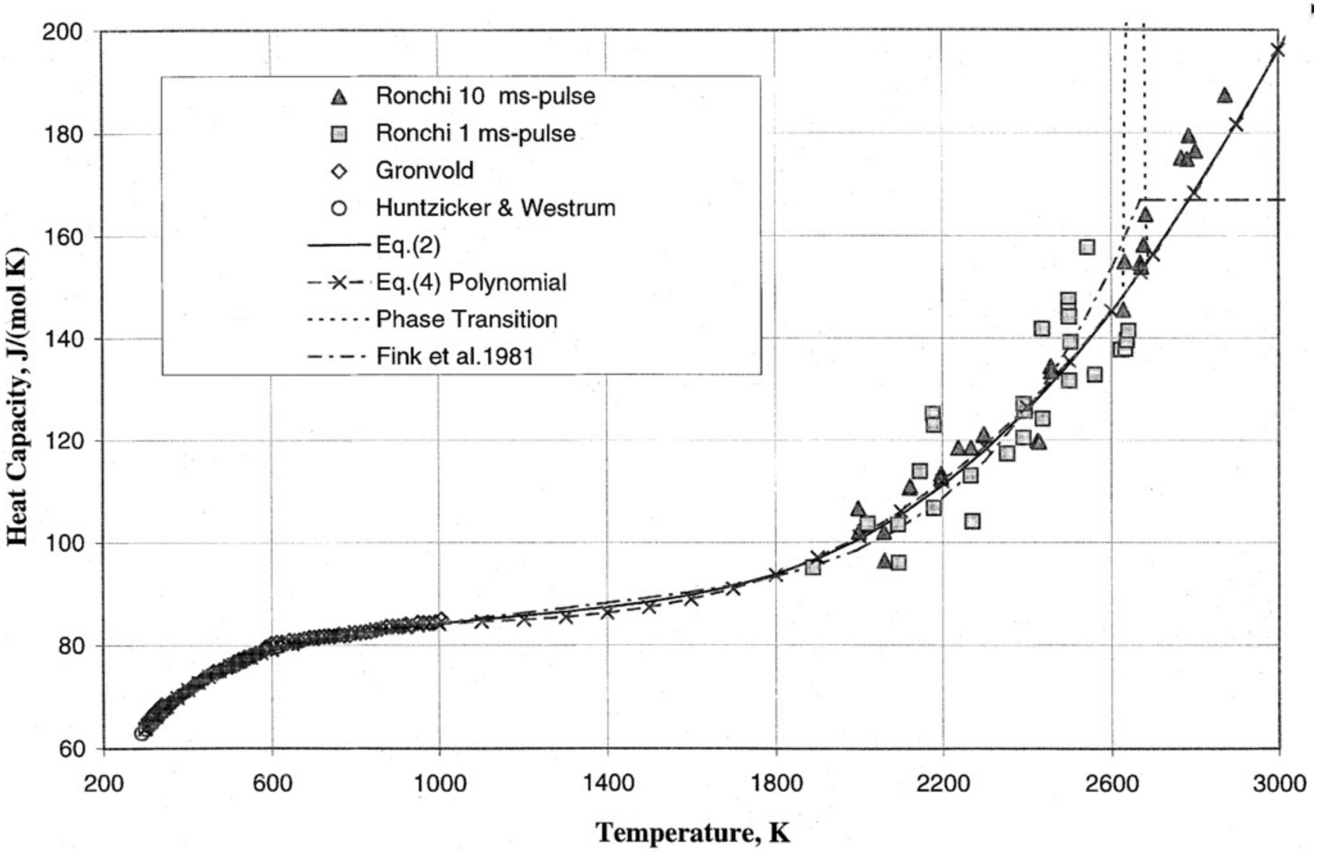




The heat capacity is a function of temperature

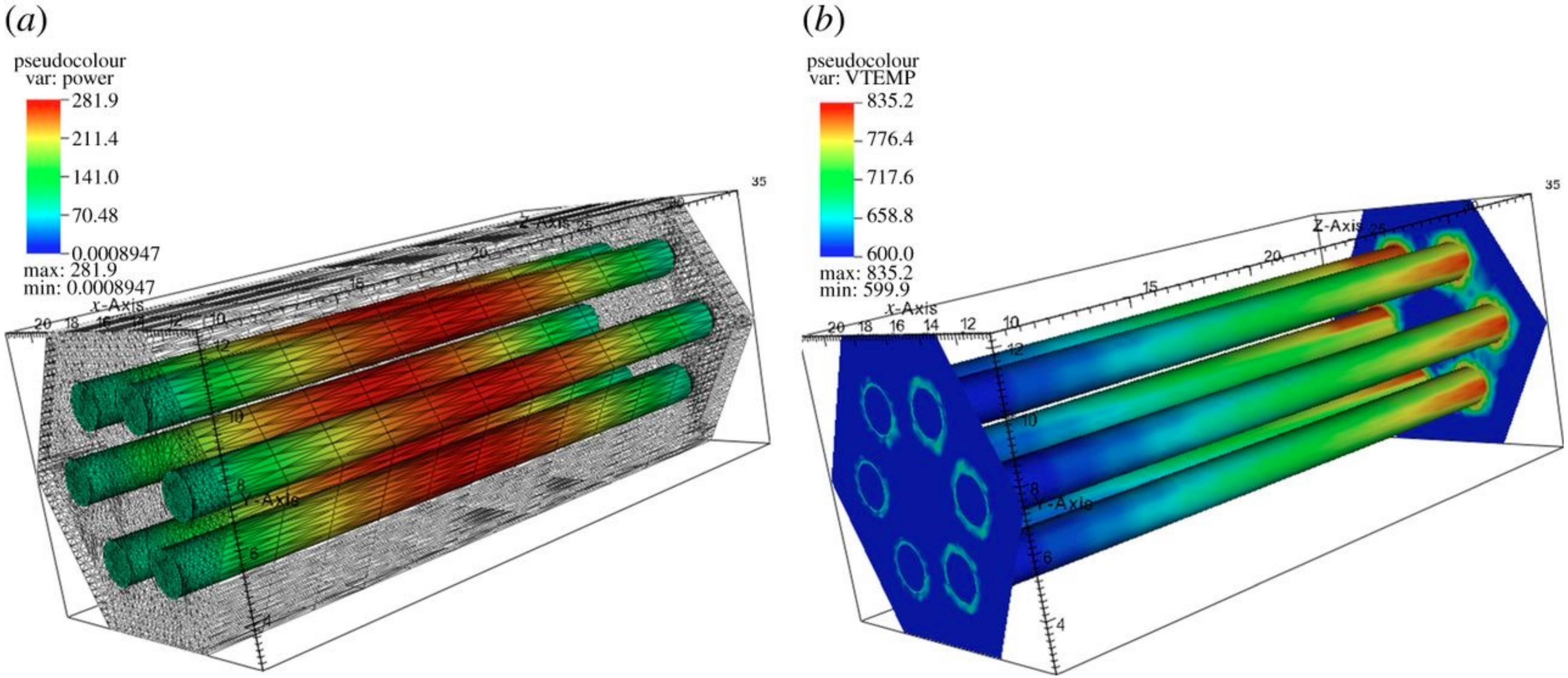
$$C_P = \frac{C_1\theta^2 e^{\theta/T}}{T^2(e^{\theta/T}-1)^2} + 2C_2T + \frac{C_3E_a e^{-E_a/T}}{T^2}$$

θ = 548.68,
$C_2 = 2.285 \times 10^{-3}$
$C_3 = 2.360 \times 10^{7}$
E_a = 18531.7



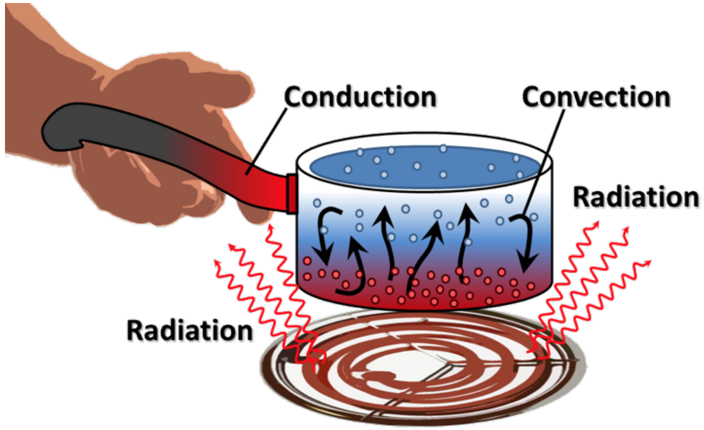
The heat generation rate is a function of the thermal neutron flux, which varies in time and space

$$Q = E_f N_f \sigma_f \varphi_{th}$$



Summary

- General heat transport
- Heat is produced in the fuel, transports through the cladding and gap, and into the coolant
- The geometry of our problem
- The initial condition of T
- The boundary conditions of T
- Is each parameter is a function of T
  - Thermal conductivity, heat capacity
- Function of space/time?
  - Heat generation, dependent upon flux



$$\rho c_p \frac{\partial T}{\partial t} - \nabla \cdot (k \nabla T)$$