Nuclear fuels for the future

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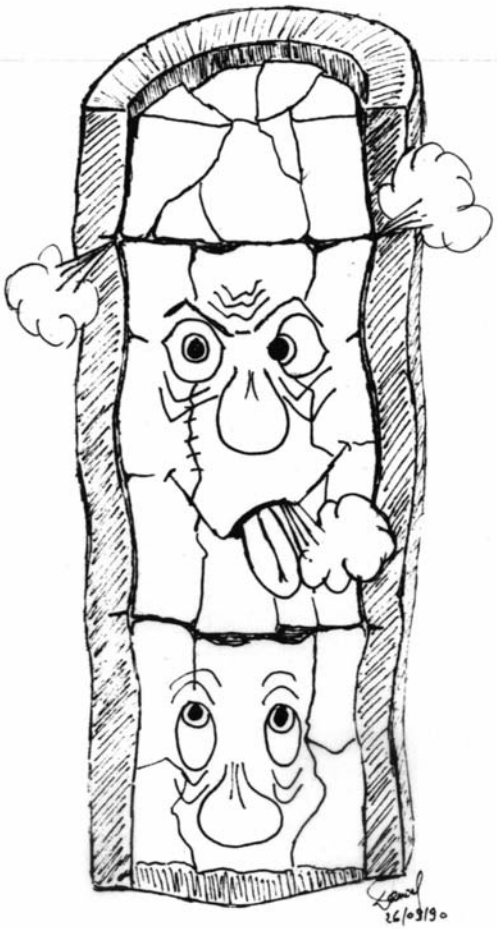
1. Core components and fuels of light-water reactors

1. Fuel elements of LWRs

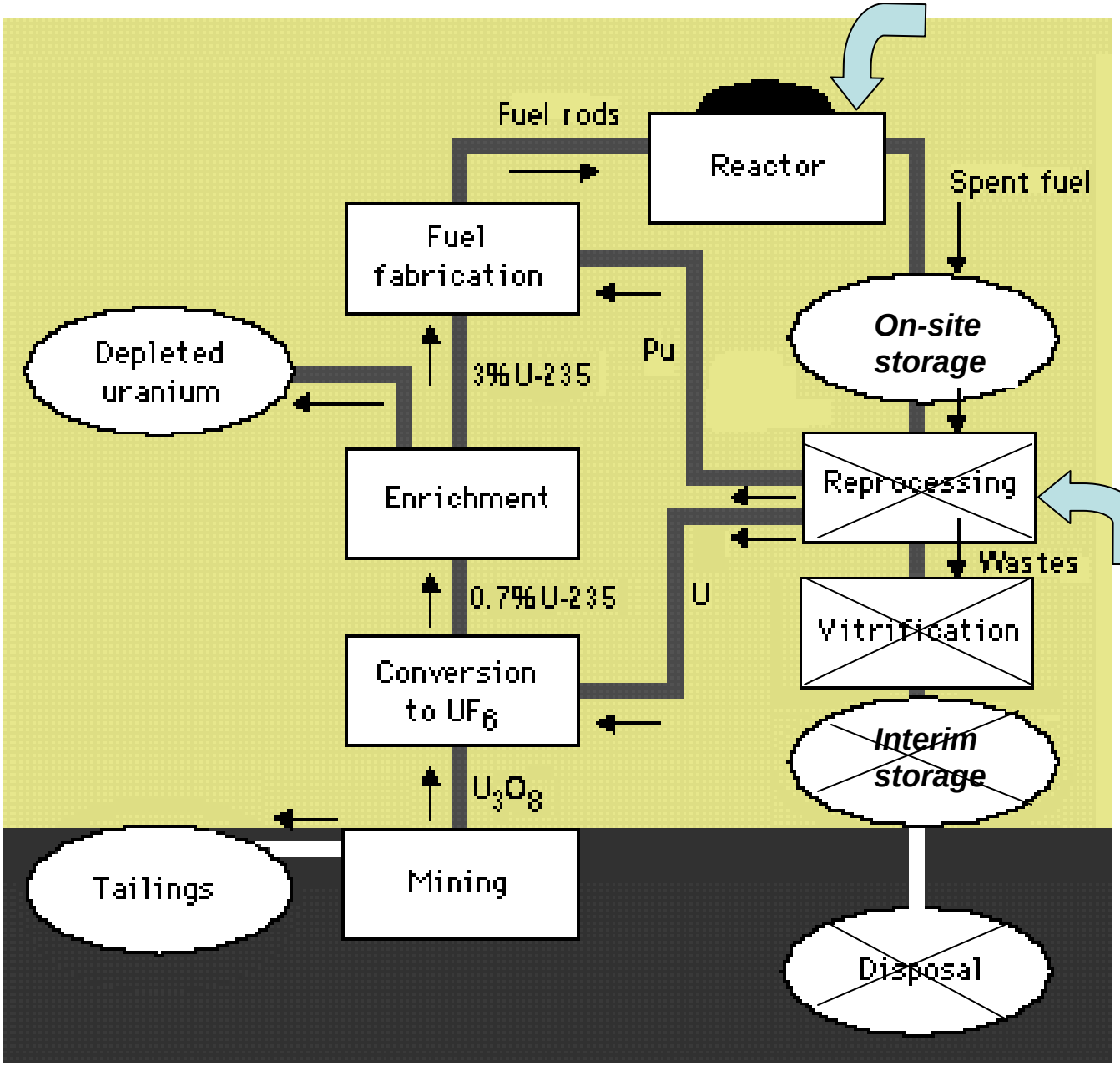
2. Materials problems with oxide fuel

3. Generation IV Reactor concepts

4. New reprocessing schemes



Standard Nuclear Fuel Cycle



Generations ("Gen") of Nuclear Reactors

- **Gen I – first nuclear electricity: EBR-I, Shippingport, Magnox, …..**
- **Gen II – Current fleet of *light-water reactors* (LWRs) - *pressurized-water* (PWR) or *boiling-water* (BWR); VVER; CANDU**
- **Gen III - LWRs of completely new design – passive safety, fewer valves, shorter piping: ABWR (GE-Toshiba), AP1000 (Westinghouse-AREVA)**
- **Gen IV – completely new designs or resuscitation of old reactor types – Sodium fast reactor; Very-high-temperature reactor**

1. Components of a 1000 MWe light-water reactor system (PWR & BWR)

Containment → ***pressure vessel*** → ***Reactor Core***

(40 m diam.)

(5 m diam. 14 m high)

(4 m diam., 4 m high)

240 ***fuel assemblies***

(20 x 20 cm, 4 m high)

↓

256 ***fuel elements***

(1 cm diam. 4 m long)

300 ***fuel pellets***

(8 mm diam. 1 cm height)

↓

Spent fuel load (U.S.)

$$18\ M\ \frac{pellets}{core} \times \frac{10\ g}{pellet} \times \frac{1\ core}{6\ yrs} = 30\ \frac{tons}{yr\text{-}reactor} \times 100\ reactors = 3000\ \frac{tons}{yr}$$

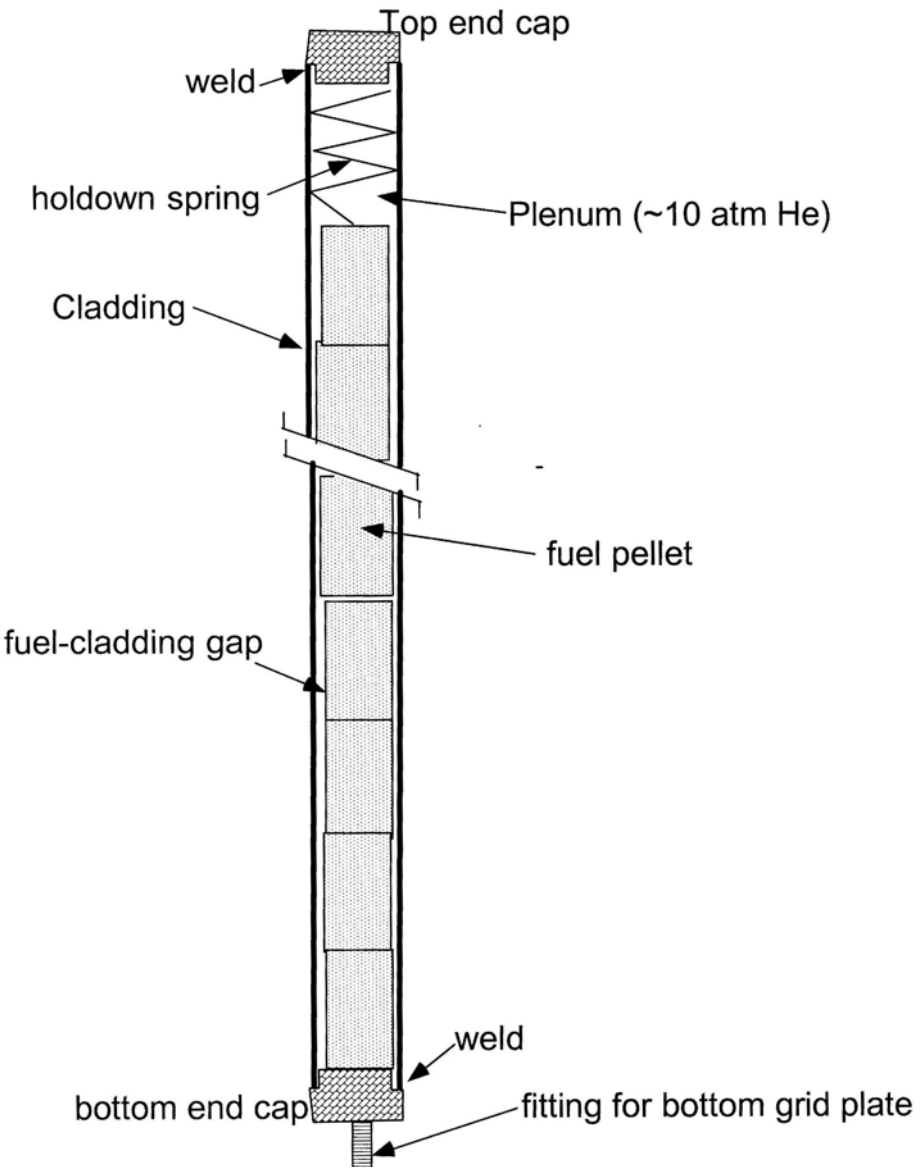
2. Light-water reactor fuel element

Length ~ 4 m
Diameter ~ 1 cm

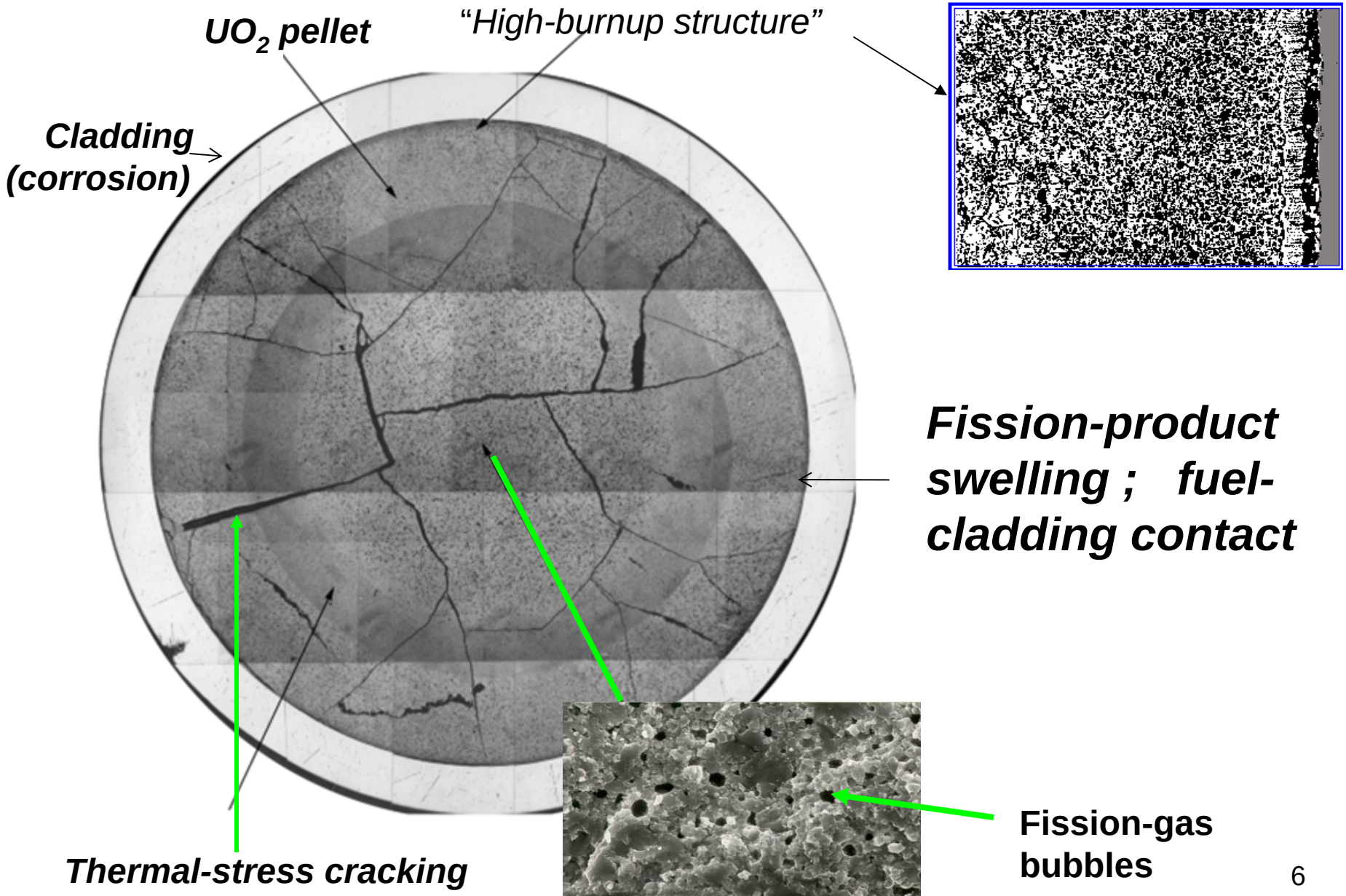
Cladding:
- Zr/1.5%Sn/0.5%FeCr (Zircaloy)
- Zr/Nb (Zirlo, M5)

Fuel:
- UO_2 (oxide)
- $(U,Pu)O_{2-x}$ (MOX)

Fuel-cladding gap (bond)
– helium at ~ 10 atm

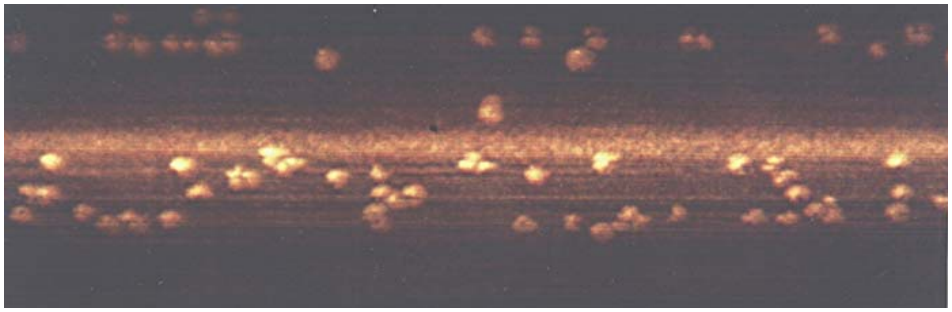


3. Problems with oxide fuel

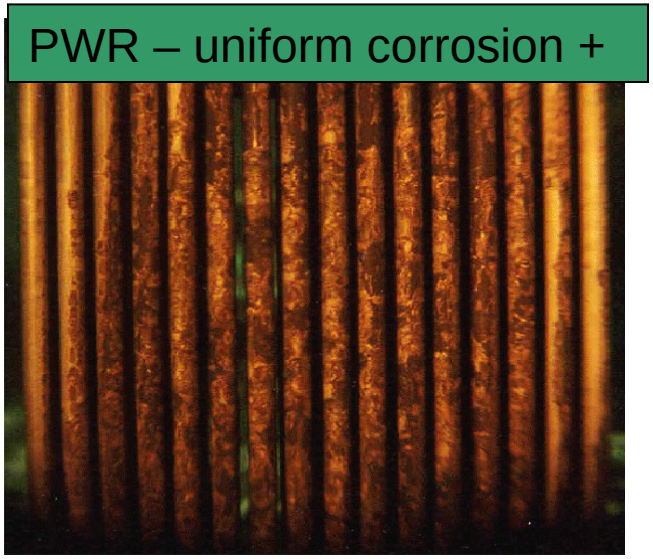


Cladding corrosion

BWR - Nodular corrosion



PWR – uniform corrosion +



$2H_2O + 4e^- \rightarrow xH_2\uparrow + 2O^{2-} + 2(2-x)H\downarrow$

$2O^{2-} + Zr \rightarrow ZrO_2 + 4e^-$

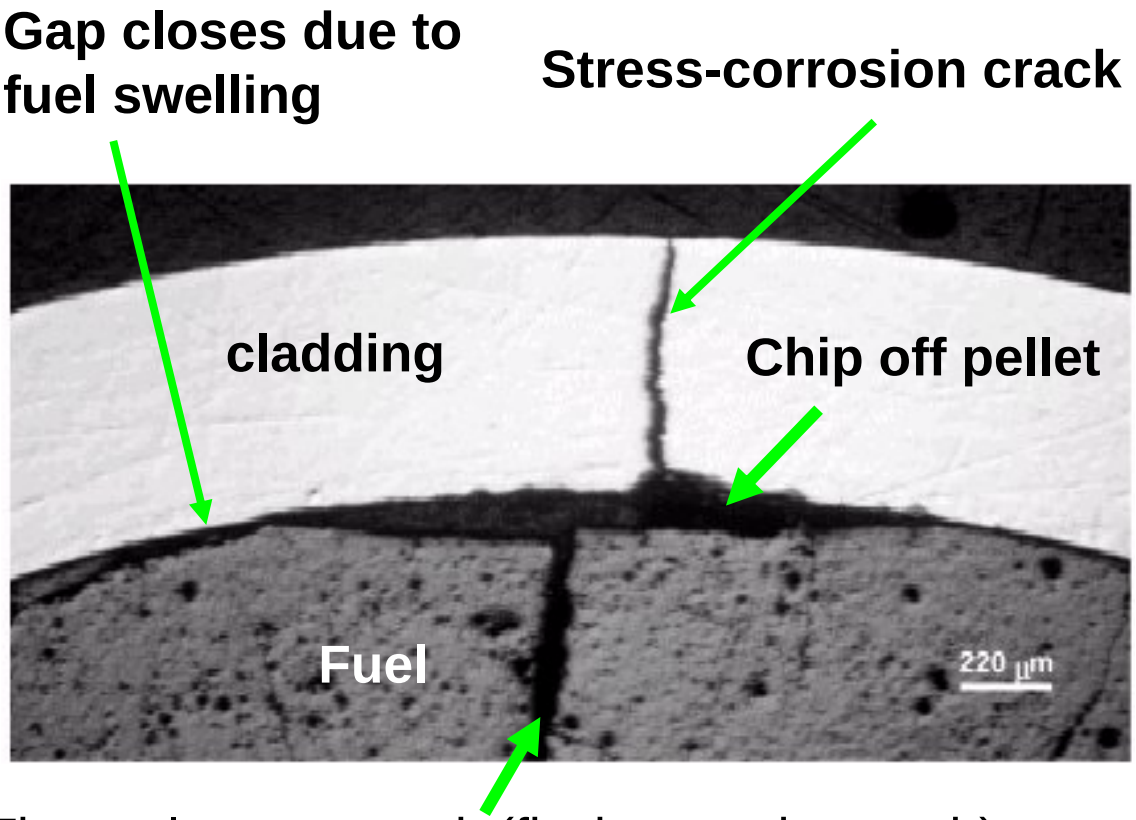
Oxide scale

cladding

Hydride embrittlement

UO_2 pellet

Pellet-cladding interaction



5. Generation IV reactor concepts

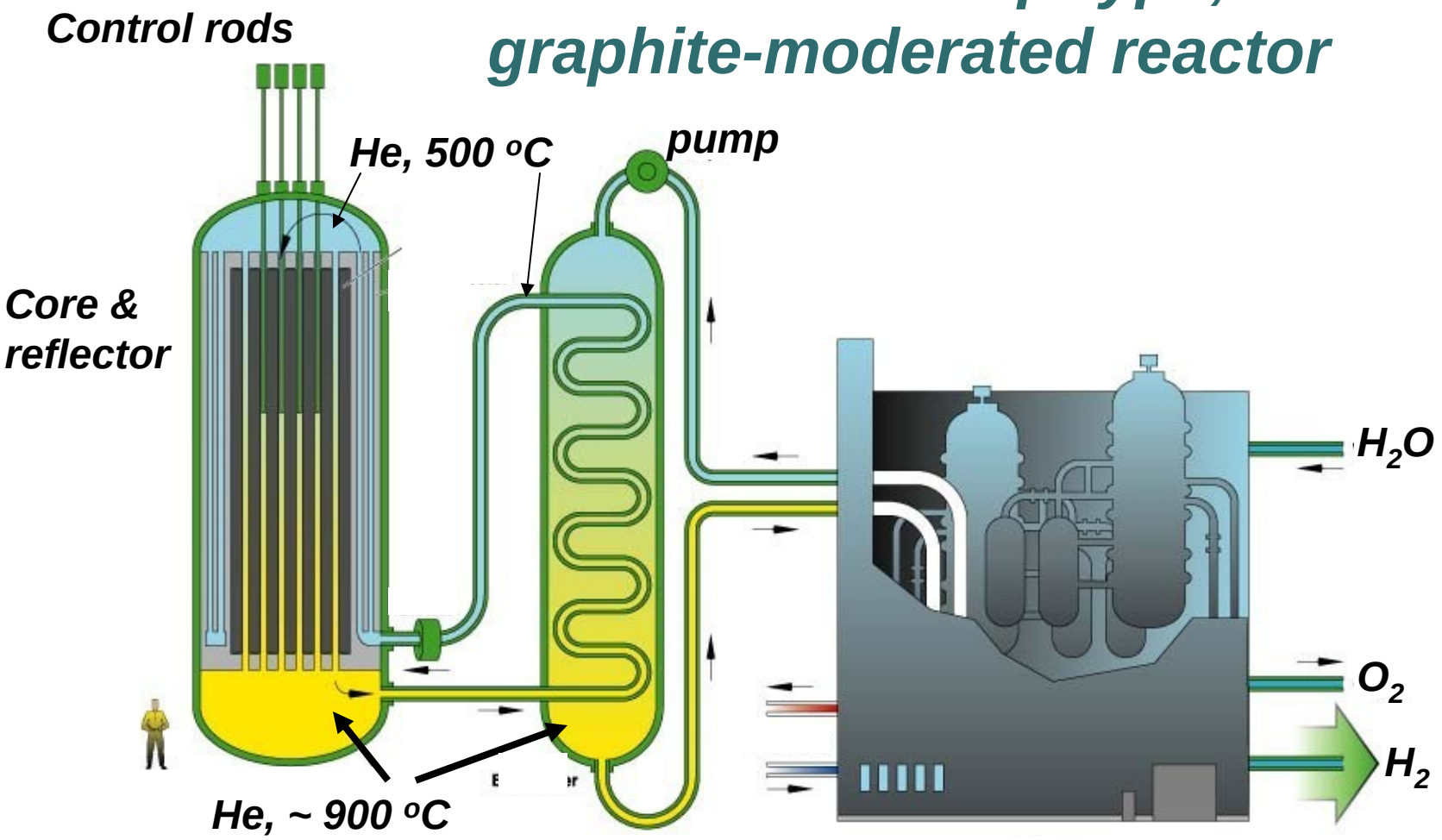
Reactor Coolant	Neutron energy	Purpose: electricity +:
Helium	Fast	H_2 production
Lead-bismuth	Fast	Encapsulated, 30-year life, small reactor ; BREST
Molten fluoride Salt	Thermal	Safety; H_2 production
→ Sodium*	Fast#	Burner of minor actinides from light-water reactors
Supercritical Water	Thermal	higher electrical efficiency than current LWRs
→ Helium*	Thermal&	Very-high temp. process heat (e.g. for H_2 production)

*current favorite

E_n ~ 1 MeV

& E_n ~ 0.025 eV

The VHTR – a loop-type, He-cooled, graphite-moderated reactor

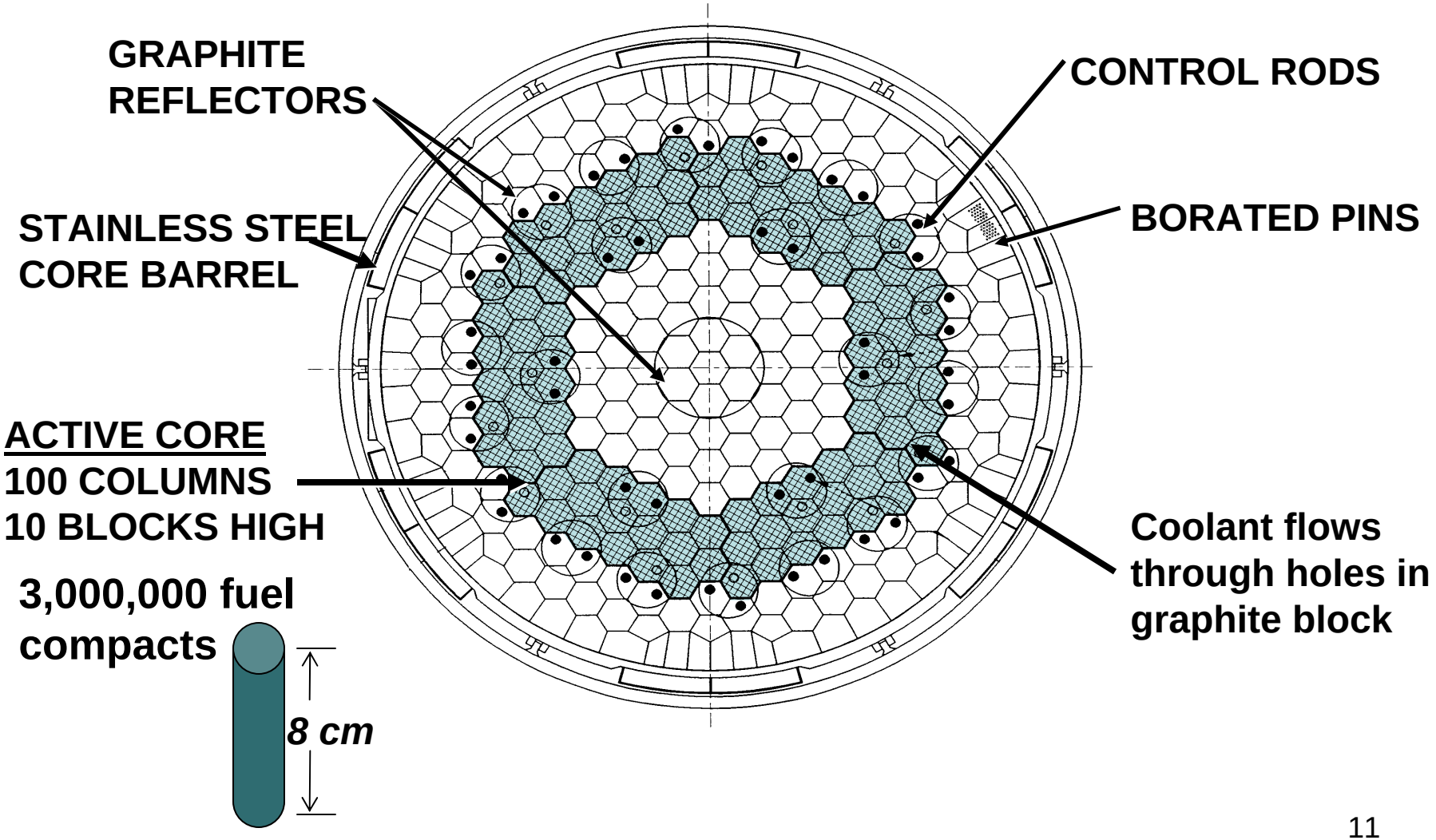


Reactor

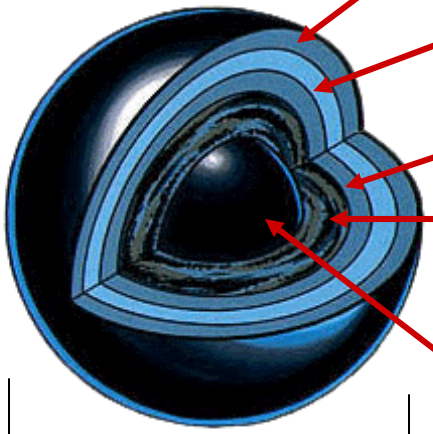
heat exchanger (He-He)

H_2 – production plant

Core configuration of the VHTR



TRISO fuel for the VHTR



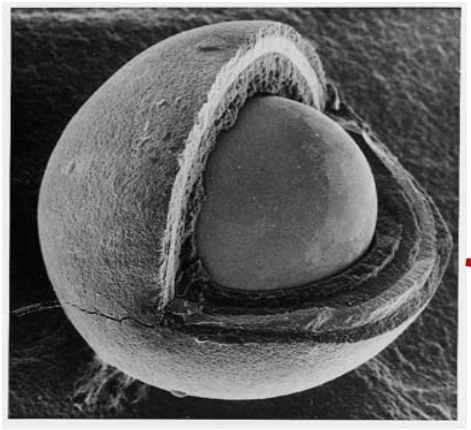
Outer pyro-carbon – structural strength

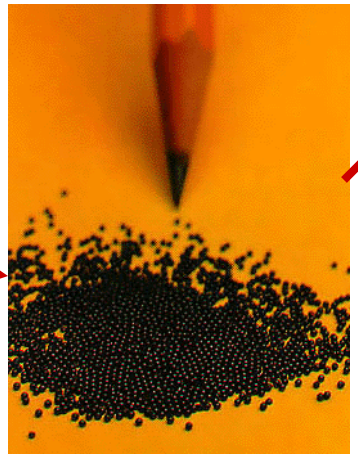
SiC – main fission-product barrier

inner pyro-carbon – retains fission gases

Porous carbon buffer – space for fission gases and CO

Kernel UO_2 + UC_2 (for uranium without oxygen)

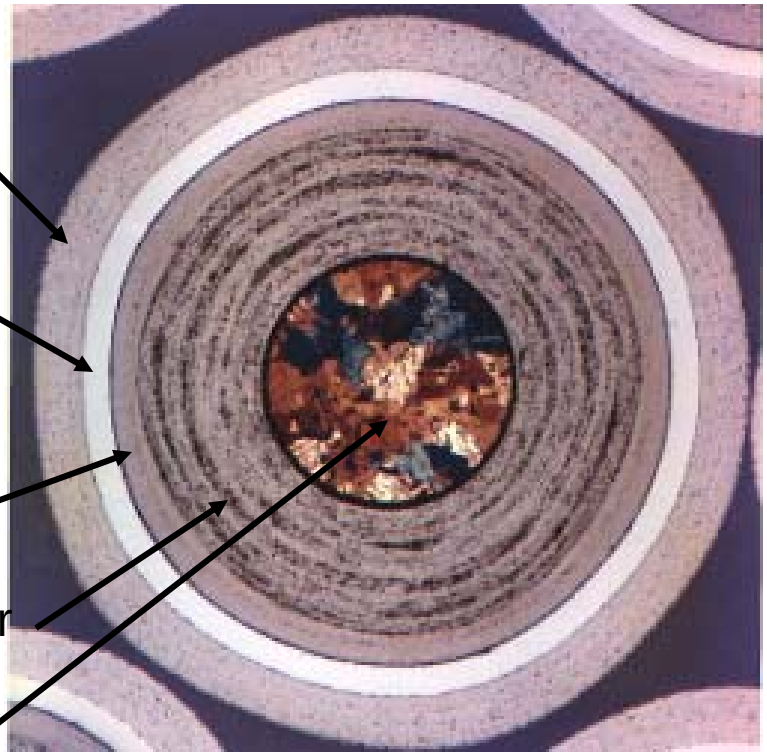


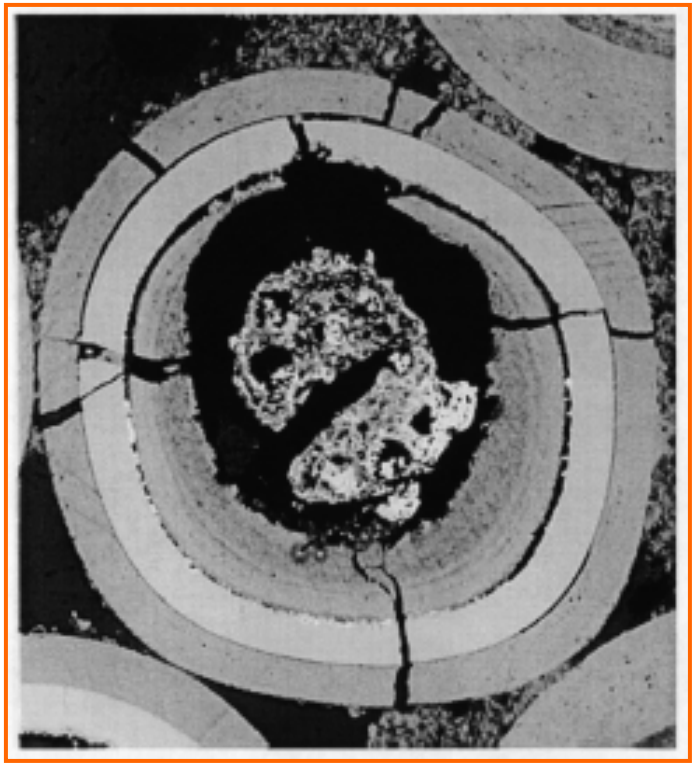


Final fuel forms

As-fabricated

Irradiated

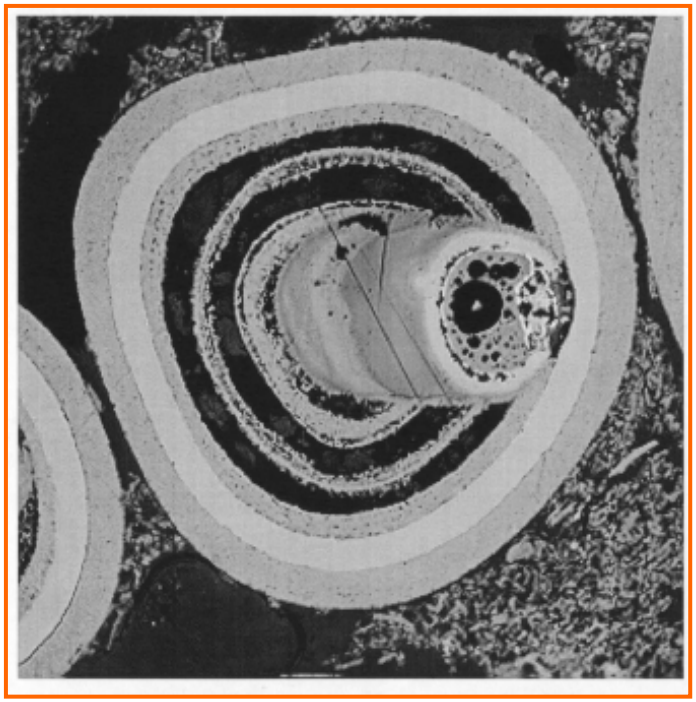


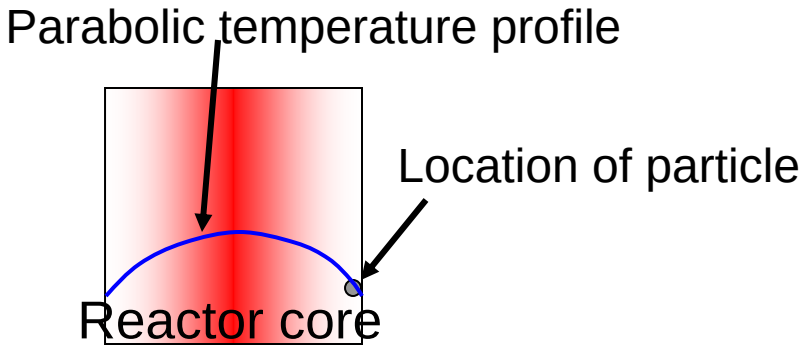


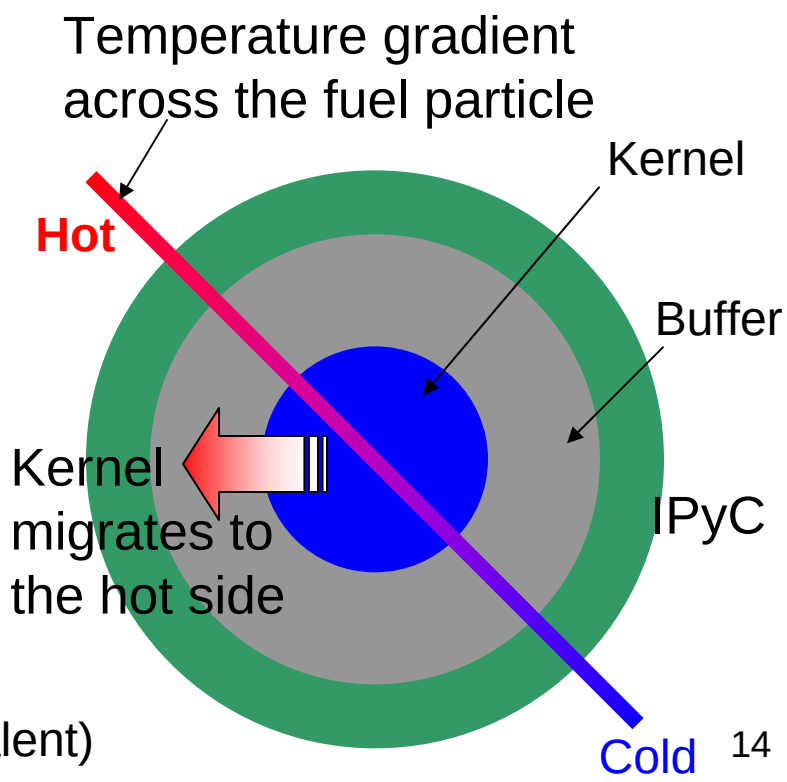
- **Xe and Kr released from fuel kernel**
- **oxygen liberated from $(U,Pu)O_{2-x}$**
- **$2C + O_2 \rightarrow 2CO$ reaction of carbon in buffer layer**

Probably failed by overpressure due to fission gases and CO

Kernel Migration





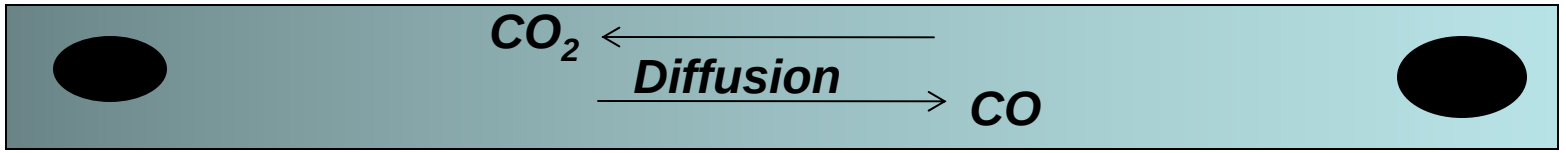


CO produced from carbon in buffer and oxygen released from fuel: $2C + O_2 \rightarrow 2CO$

Why O released from fuel? U in UO_2 binds two O; fission products on average bind only ~ 1.67 O (most soluble fps are trivalent or divalent)

Mechanism

hot ***cold***



Low P_{CO2} High P_{CO2}

small K
CO_2 decomposes, Produces CO and consumes C

$$2CO = CO_2 + C$$

$$K = P_{CO2}/(P_{CO})^2$$

K ↑ as T↓

Large K
CO decomposes, Produces CO_2 and produces C

∴ kernel moves into space made by gasification of C – i.e. to hot side

Solution?

<u>Add coat of ZrC around kernel</u>

$ZrC + O_2 = ZrO_2 + C$

- ***K large, p_{O2} reduced***
- ***If p_{O2} decreased, p_{CO} is reduced***
- ***eliminates both the CO overpressure and kernel migration***

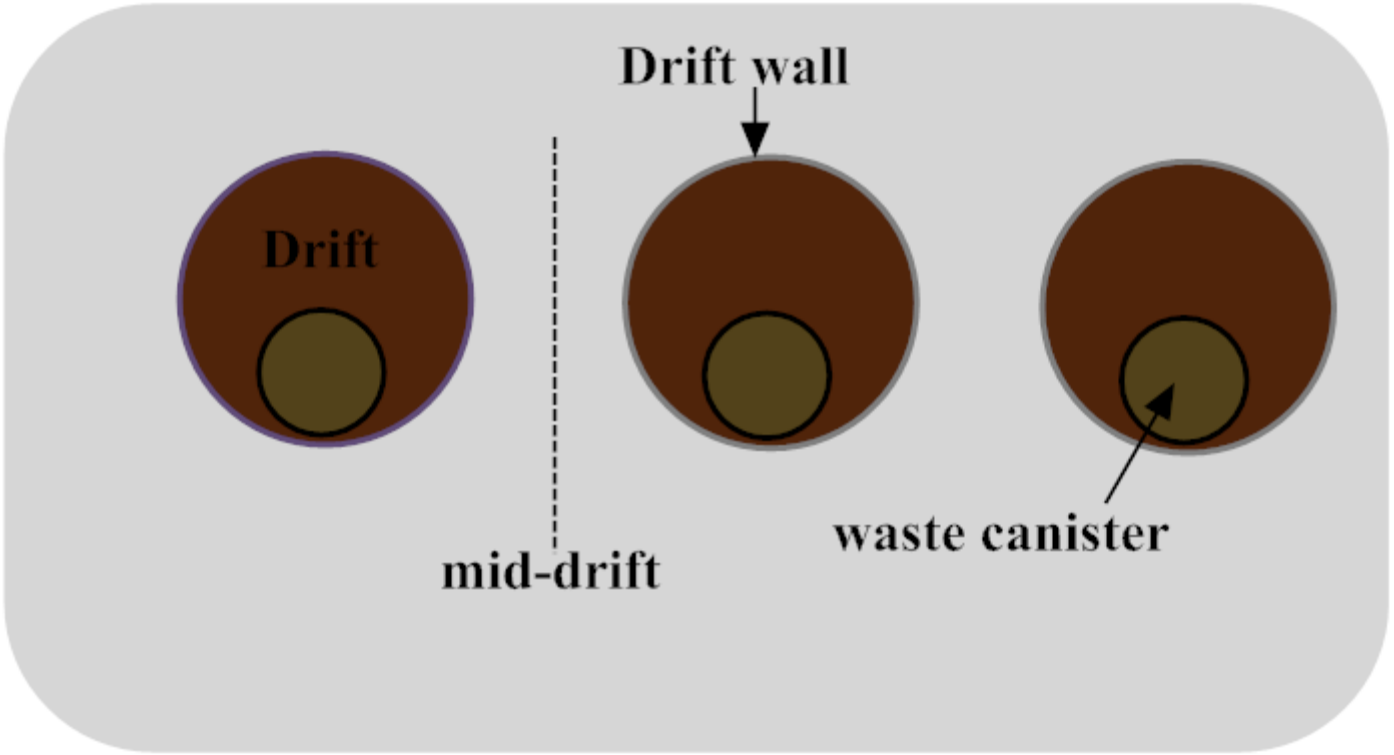
<u>Use $(U,Pu)O_2$ + (U,Pu)C fuel in kernel</u>

-Fission in carbide releases C, not O

- O released from oxide consumed by (U,Pu)C

$$2O + (U,Pu)C \longrightarrow (U,Pu)O_2 + C$$

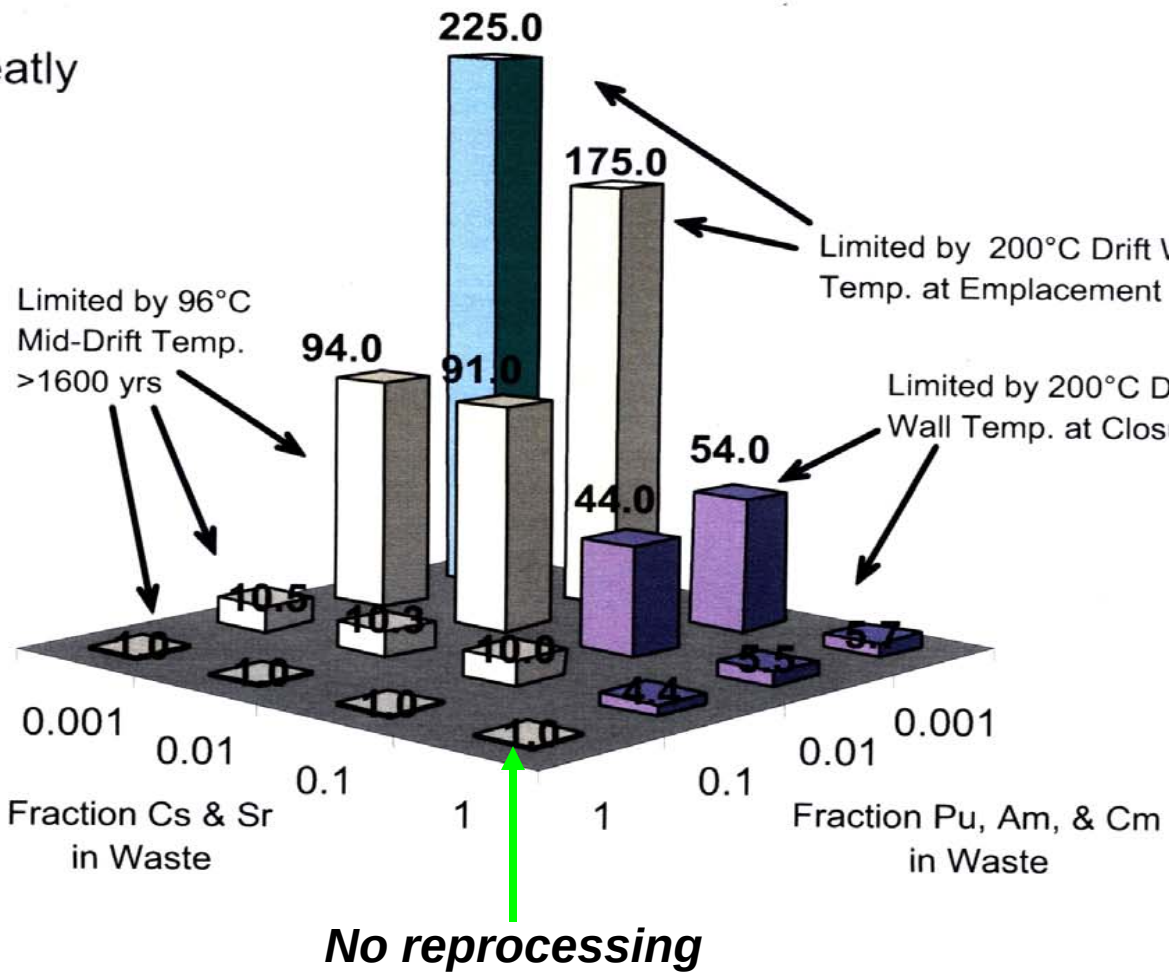
Geologic Repository



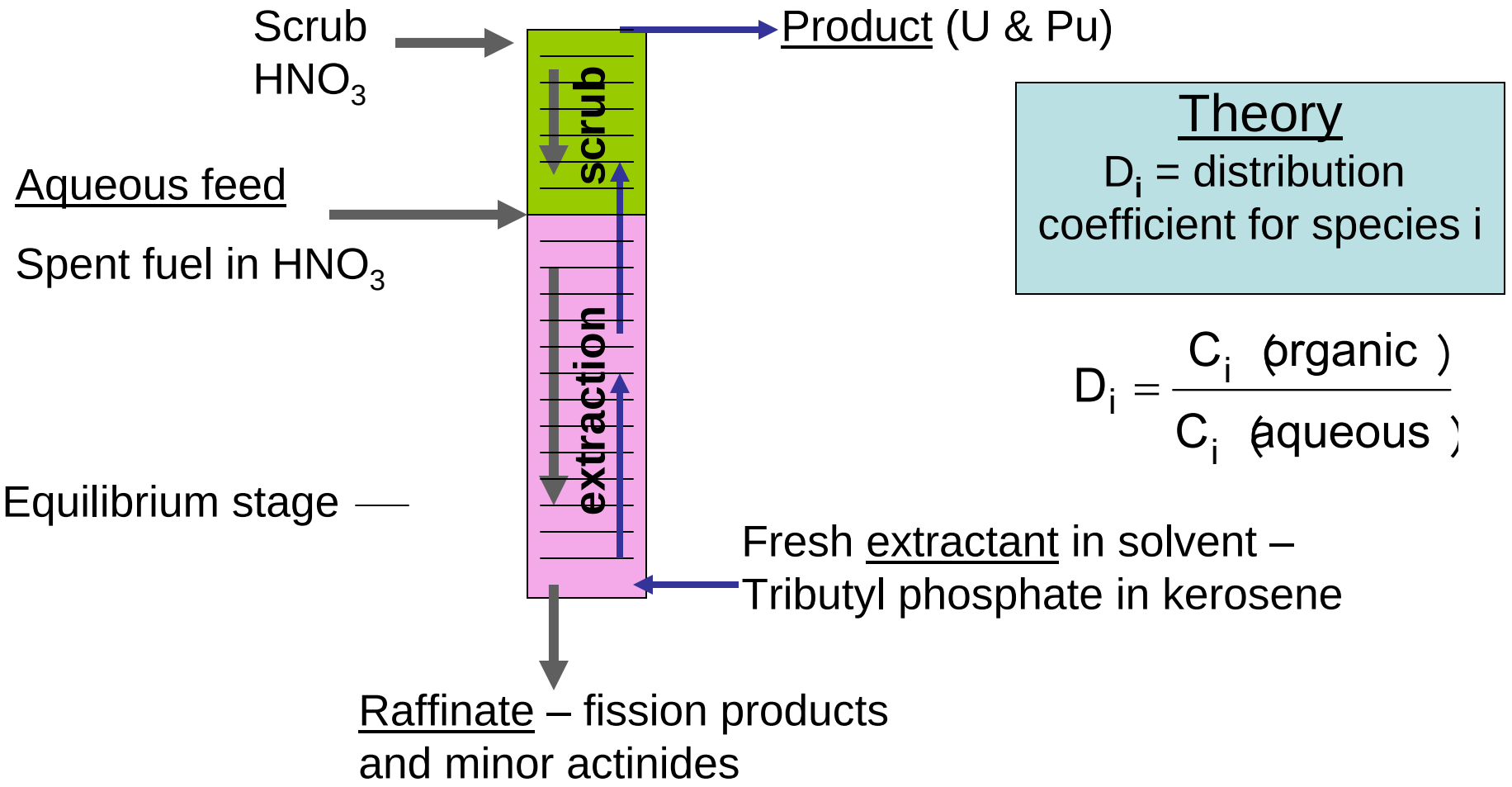
Why reprocessing is necessary

- Removal of Cs, Sr and TRU greatly reduces the heat load on the repository
- Assumptions:
  - Burnup = 50 GWd/MT
  - Separation in 25 years
  - Emplacement in 25 years
  - Closure in 100 years

Numbers on top of the columns represent the ratio of the quantity of reprocessed waste to the quantity of spent fuel that can be placed in a drift in repository



PUREX Solvent Extraction Process ~ 1950



Theory

D_i = distribution coefficient for species i

$$D_i = \frac{C_i\ (\text{organic})}{C_i\ (\text{aqueous})}$$

New reprocessing requirements

- ***Pure Pu must not be separated***

- ***Cs & Sr must be separated from other fission products***

- ***Minor actinides to be separated (Np, Am, Cm)***

- ***Tc to be separated***

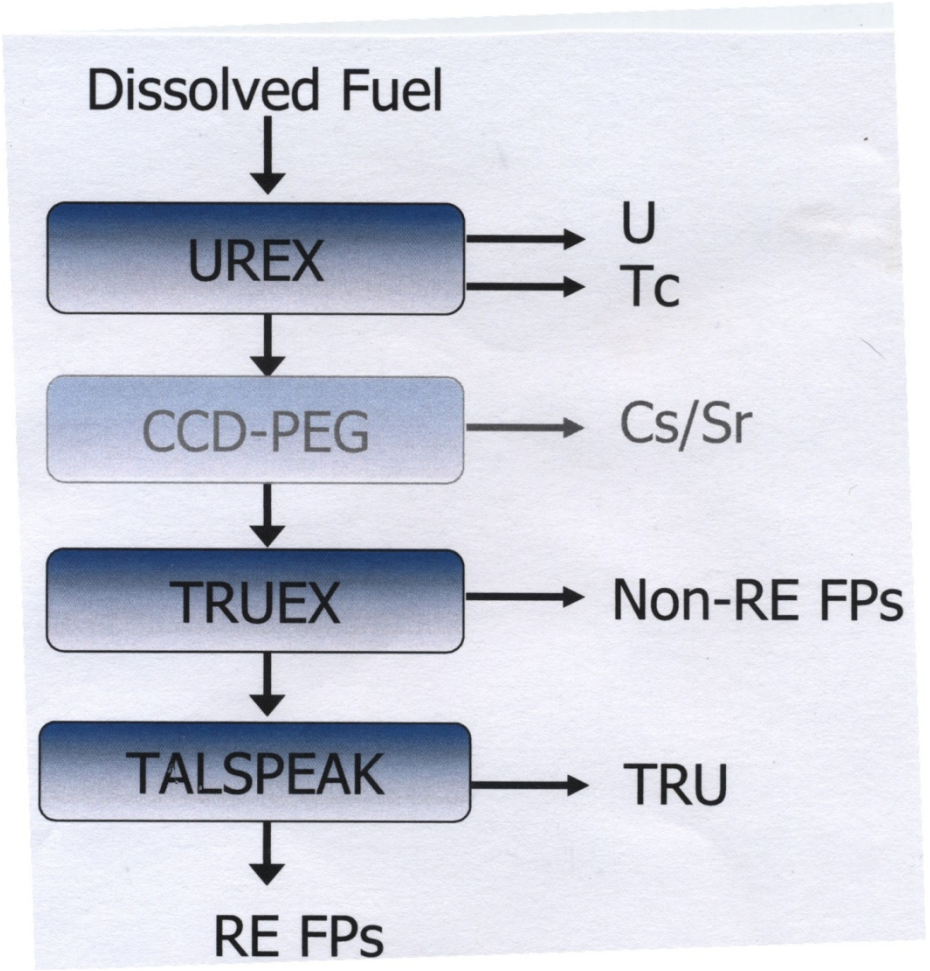
The UREX1a reprocessing suite

UREX = URanium EXtraction

CCD-PEG* = shorthand for extractant used

TRUEX = TRansUranium EXtraction

TALSPEAK = Trivalent Actinide/Lanthanide Separation by Phosphorus-reagent Extraction by Aqueous Komplexes

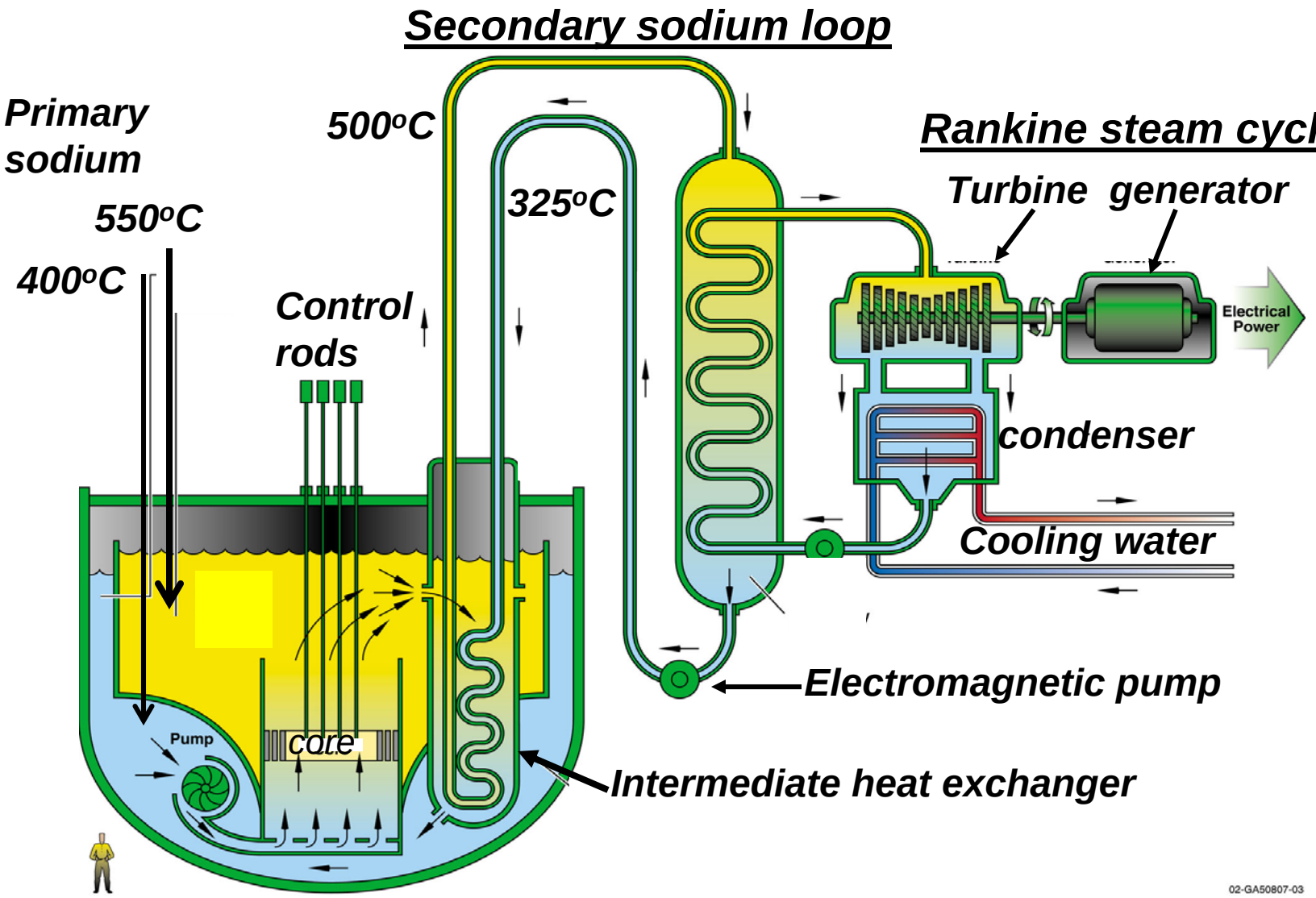


*** Developed in Czech Republic, Russia**

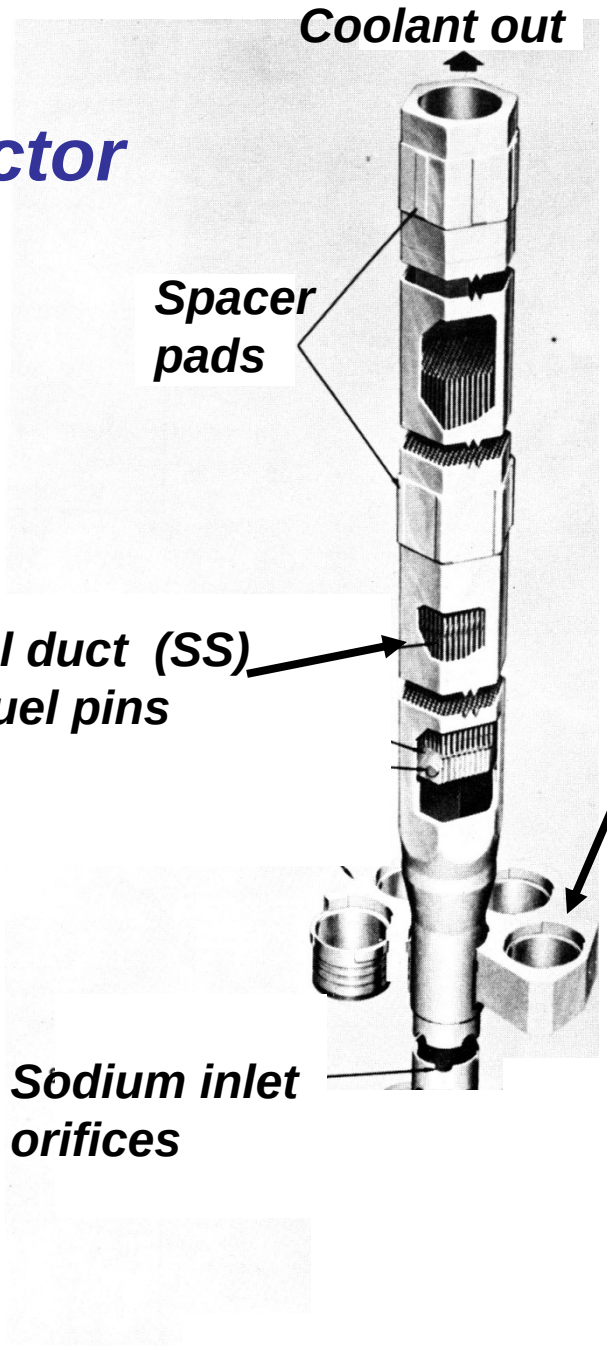
REMAINING PROBLEMS

- Stability of exotic organic extractants in the high radiation field of the fission products
- Sensitivity of distribution coefficients to extraction conditions (pH, concentration of extractants, temp., oxygen potential)
- Inherent difficulty of some separations (esp. TRU from RE fp – both are trivalent)
- Conversion of products to solid wasteforms suitable for disposal in repository
- Lack of even pilot-plant experience with TRUEX and TALSPEAK

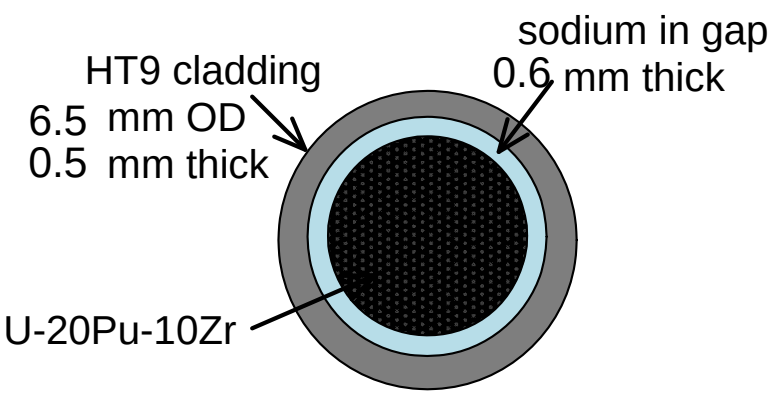
Sodium Fast Reactor (SFR)

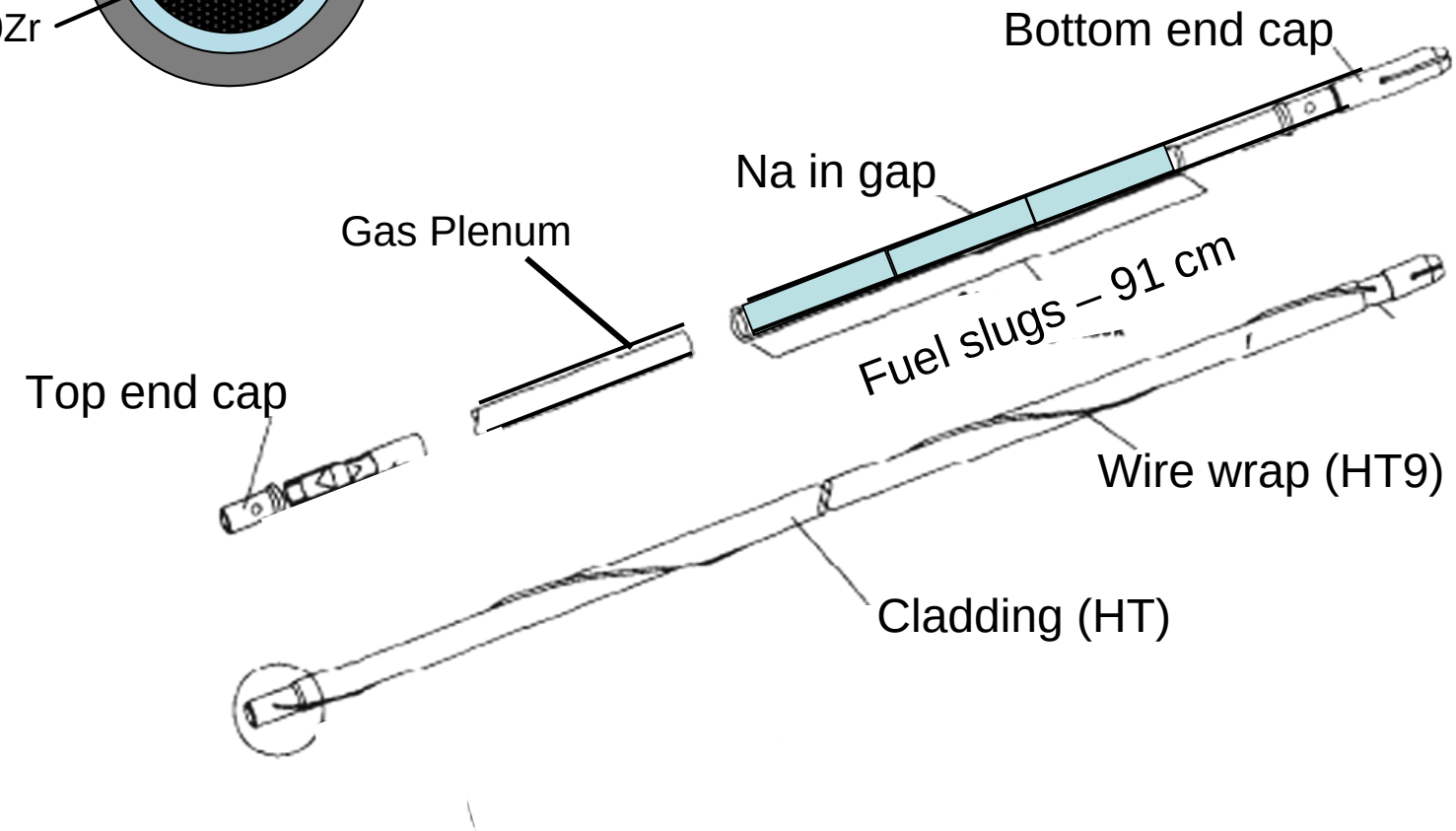


Sodium fast-reactor fuel assembly



Metal fuel for SFR
U-20%Pu-10%Zr (+ MA)





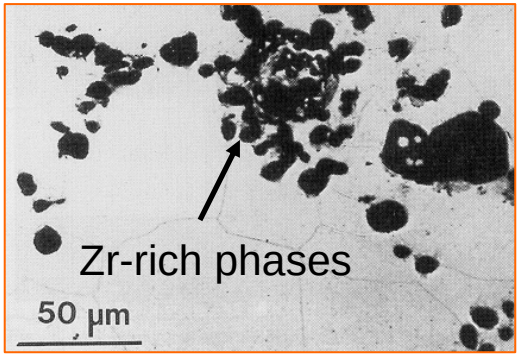
Metal Fuel Performance Phenomena

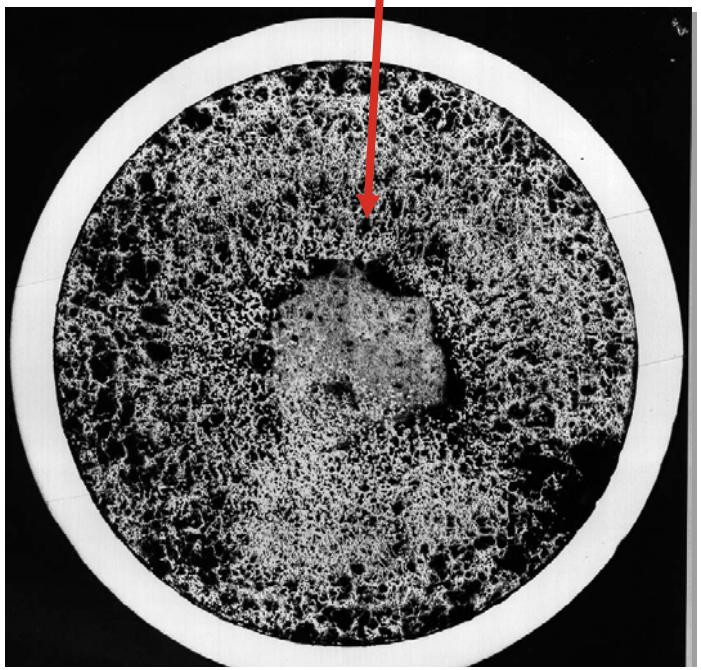
- Irradiation growth: ~ 3% at 14% burnup of metal atoms
- Fuel swelling and fuel-cladding mechanical interaction (FCMI)
- Gas release
- Fuel-cladding chemical interaction (FCCI)
- Fuel constituent redistribution

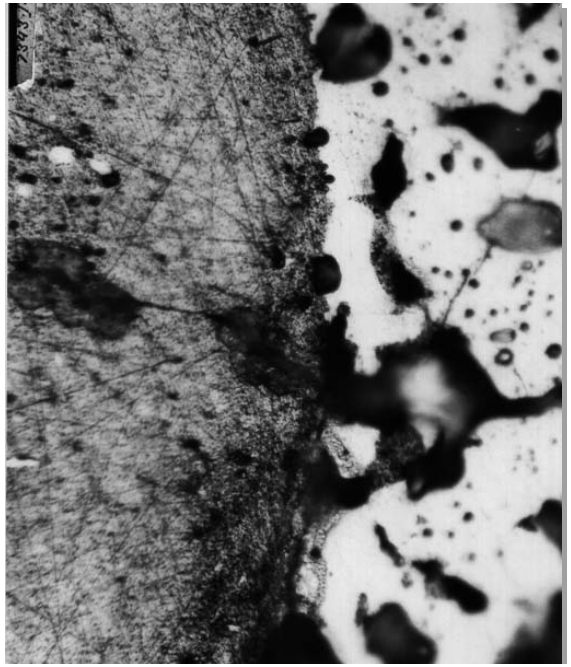
Low- Melting Phase

La, Ce, Pr, Nd, Pu react with SS cladding

as-fabricated

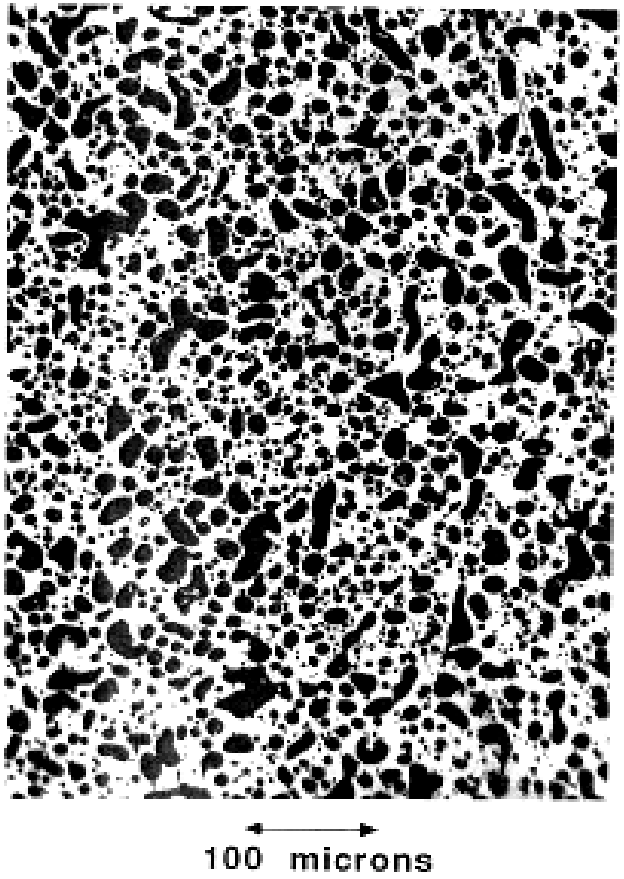




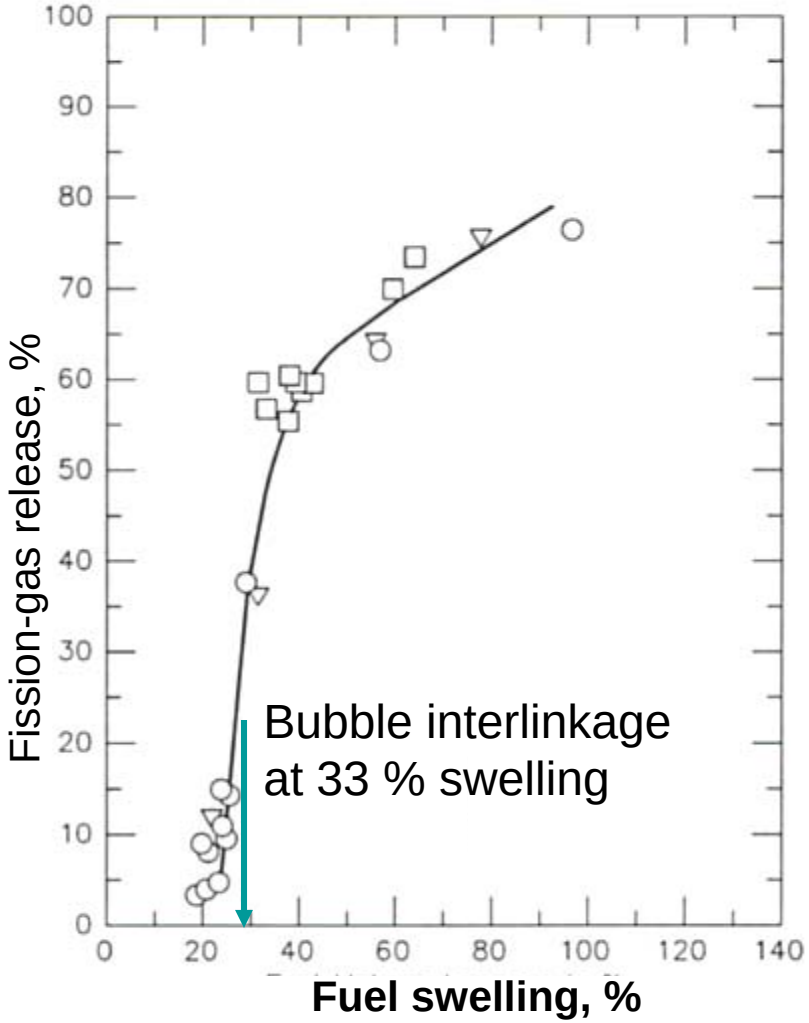


Swelling & Gas Release in metal fuel

Gas bubbles



2% FIMA (**f**issions per **i**nitial **m**etal **a**tom)

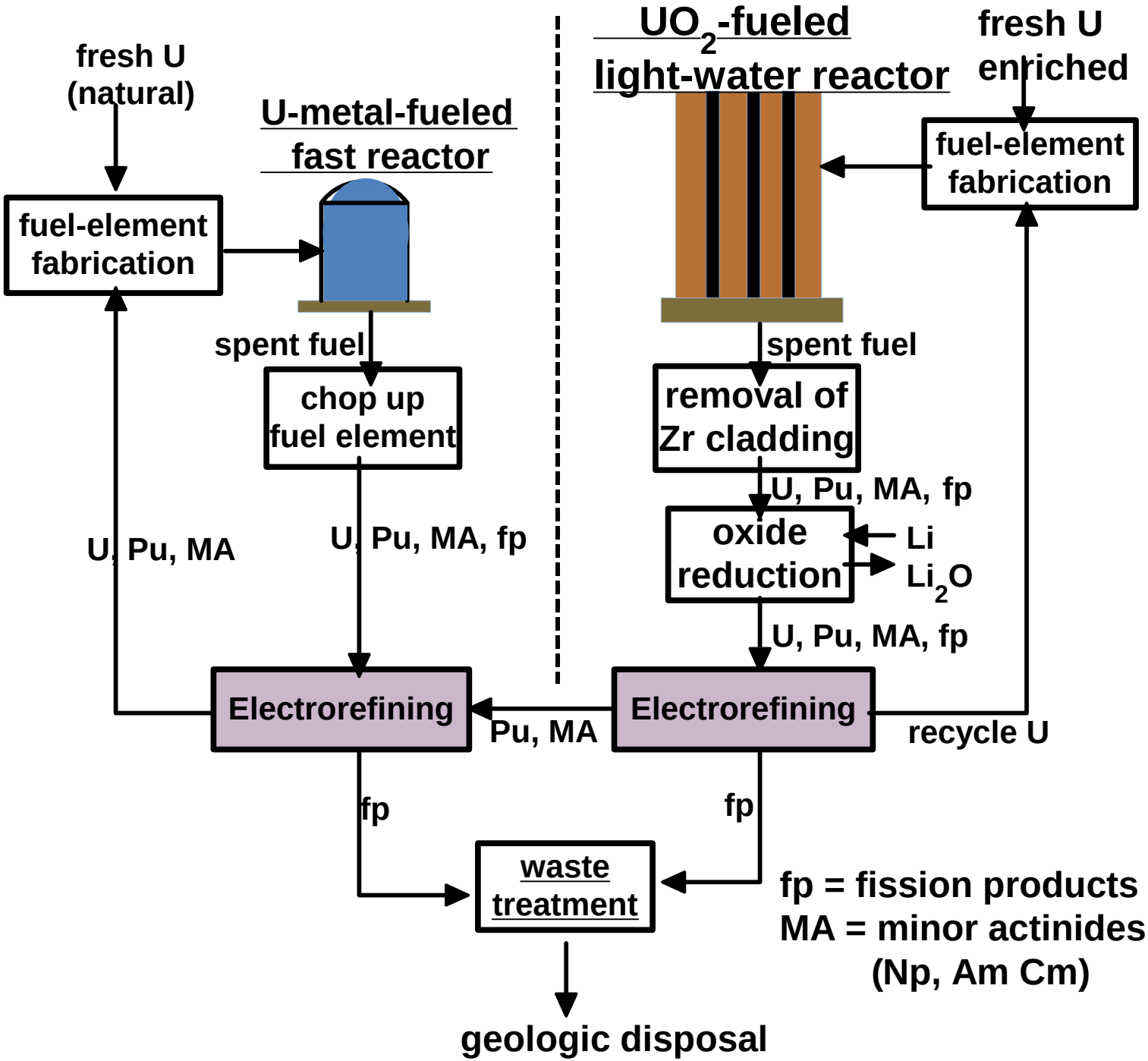


Pyroprocessing

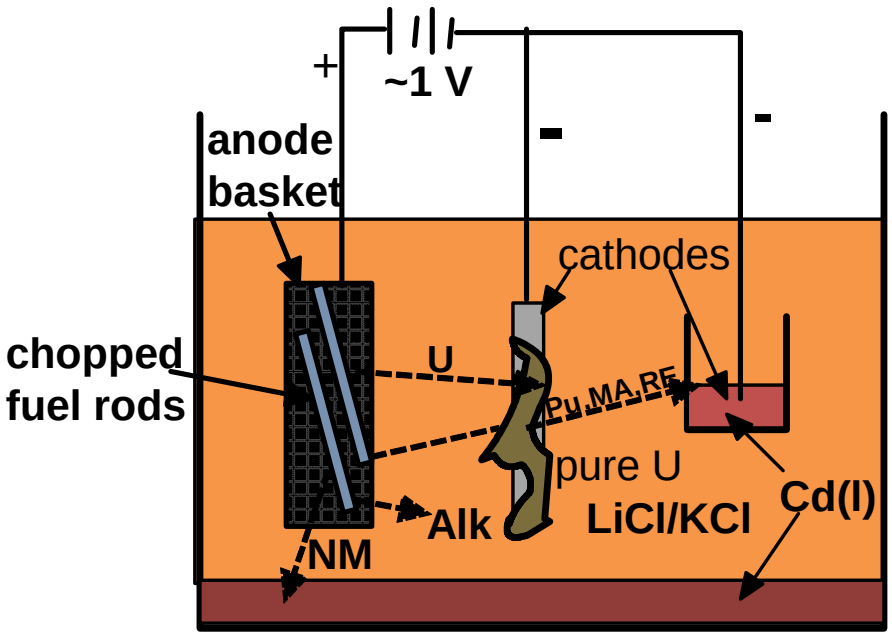
• What is it? A high-temperature, nonaqueous electrochemical method of reprocessing spent nuclear fuel to separate the valuable constituents from the radioactive waste

• Motivations:

- nonproliferation (plutonium)
- recovery of uranium
- sequestering of radioactive wastes
- destruction of “minor actinides” (Np, Am, Cm) and Pu


fresh U
(natural)
U-metal-fueled
fast reactor
UO2-fueled
light-water reactor
fresh U
enriched
fuel-element
fabrication
fuel-element
fabrication
spent fuel
spent fuel
chop up
fuel element
removal of
Zr cladding
U, Pu, MA, fp
oxide
reduction
Li
Li2O
U, Pu, MA
U, Pu, MA, fp
U, Pu, MA, fp
Electrorefining
Pu, MA
Electrorefining
recycle U
fp
fp
waste
treatment
fp = fission products
MA = minor actinides
(Np, Am Cm)
geologic disposal

Electrorefiner



- spent fuel dissolved/reacted in SS anode basket
- high-purity U transferred to SS cathode
- Pu and the minor actinides transferred of Cd(liq) cathode
- Cd puddle collects noble-metal fission products

fission products: ***NM = Rh, Ru, Pd;*** ***Alk = Na, Sr, Ba;*** ***RE = La,...Y***

Stable (in salt phase)	**Electro-transportable**	**stable** (in Cd pool)
Cs, Ba, Sr	U, Pu, MA, RE	Mo, Pd, Rh, Ru

Electrotransport rate = $i/3\mathfrak{F}$; $i \propto V \times \chi_{M^{3+}}$

DOE's NGNP program

NGNP = Next Generation Nuclear Plant (Gen IV)

- ***six reactor types; two are favored:***

➢ VHTR: 5-layer fuel spheres; He cooled, graphite moderated. 900^{o}C outlet temp. For power and/or H_2 production.

➢ Aqueous reprocessing of spent fuel

➢ SFR: metal fuel rods; sodium-cooled; unmoderated. For power and burning of minor-actinide elements from current fleet of LWRs.

➢ Pyrometallurgical reprocessing of spent fuel

- ***Neither are new designs***
- ***Demonstration plant circa 2020?***