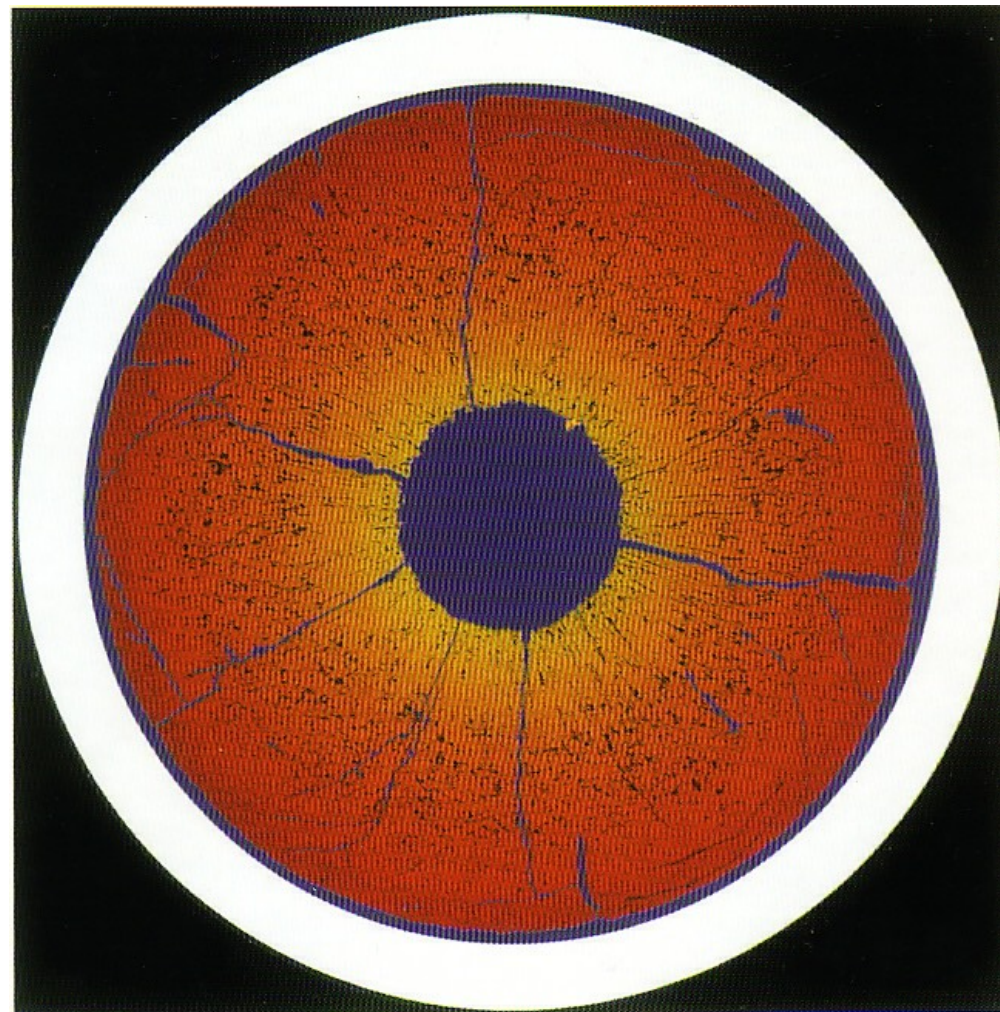# Fuels for Generation IV reactors

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Reactor Physics, KTH

Learning objectives

- After this lecture you will be able to select fuels for Generation-IV reactors based on the following criteria:

- Margin to melt

- High temperature stability

- Industrial scale reprocessability

Thermal conductivity

- Relation between linear power density χ, thermal conductivity k and temperature difference ΔT between fuel surface and centerline:

$$\frac{\chi}{4\pi} = \int_{T_s}^{T_c} k(T)dT$$

← Conductivity integral

- For a given linear power density, the temperature difference in a solid fuel pellet (without central hole) depends only on the thermal conductivity of the fuel - not on pellet radius!

- Fixing the linear power density, a higher thermal conductivity corresponds to a smaller temperature difference and hence a lower fuel temperature

Thermal conductivity: oxide fuels
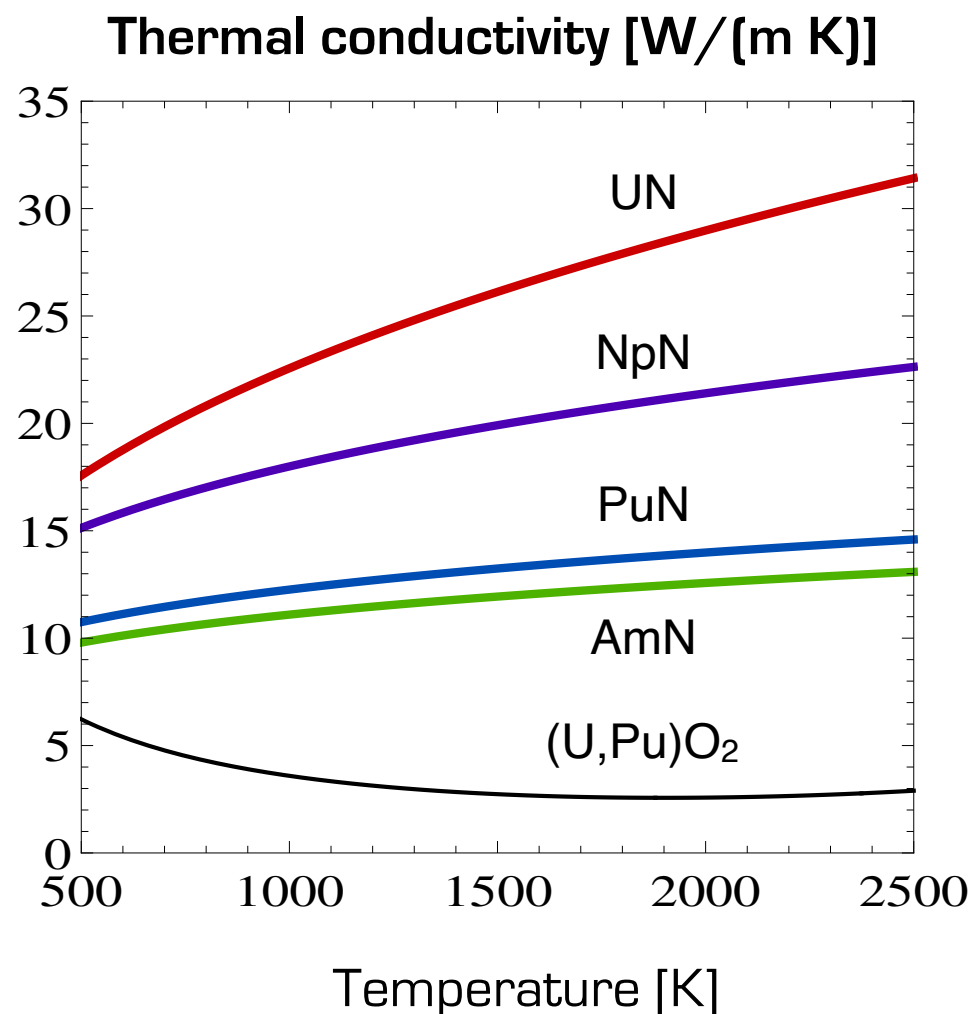
Thermal conductivity [W/m/K]
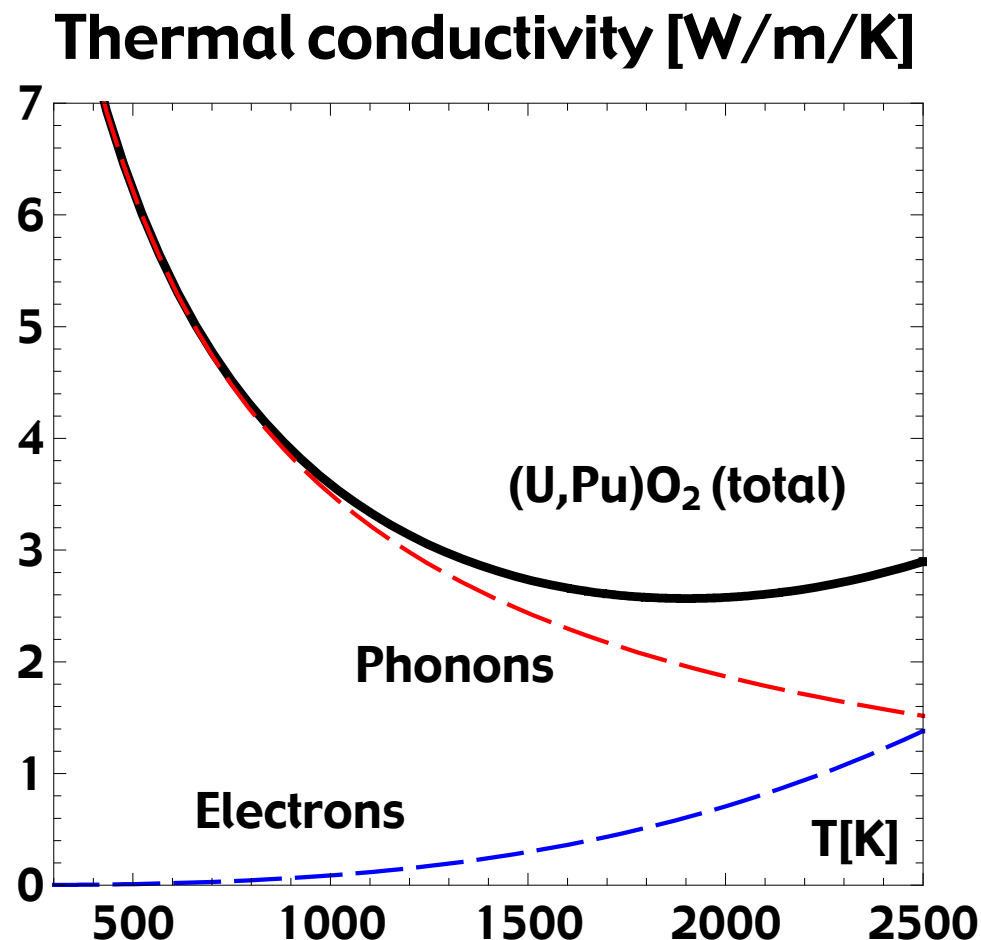
- Thermal conductivity is the sum of contributions from electrons and phonons (lattice vibrations)

- Phonon contribution peaks at temperatures below fuel surface temperature, and decreases with T.

- Electronic contribution increases with temperature.

- For (U,Pu)O_{2-x} fuels experimental data may be parametrised as function of sub-stoichiometry x as

$$\lambda_{TD}(T,x) = 1.158\left(\frac{1}{A(x)+B(x)T} + 6400\left(\frac{1000}{T}\right)^{5/2} exp\left(-\frac{16350}{T}\right)\right)$$

$$A(x) = 0.035 + 2.85x$$
$$B(x) = (0.286 - 0.715x)\times 10^{-3}$$

Thermal conductivity of nitride fuels

Thermal conductivity [W/(m K)]

- Chemical bond in nitride compounds of semi-metallic character

- Thermal conductivity of UN is comparable to that of metals and is dominated by electronic contribution

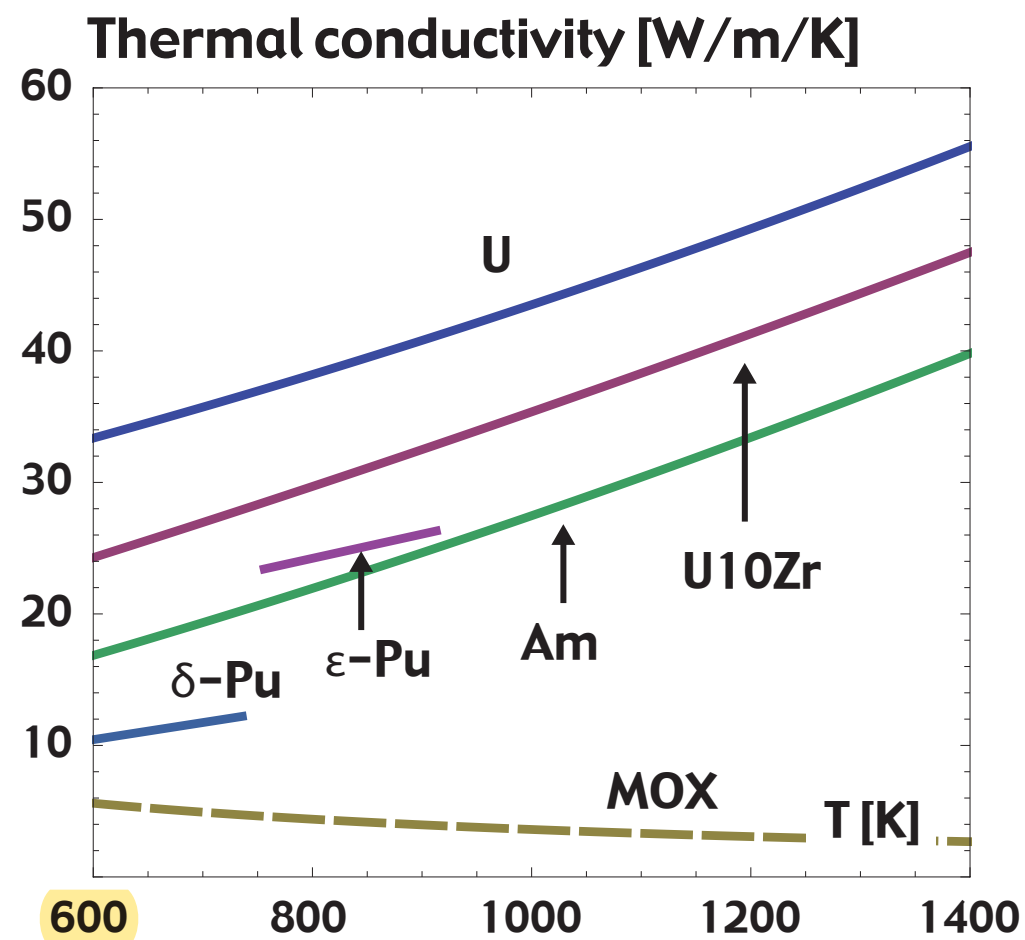- Conductivity reduces with increasing atomic number of the actinide

$$\lambda_{UN}(T) = 1.864 \times T^{0.361}$$

$$\lambda_{NpN}(T) = 7.89 + 0.0127 \times T - 4.32 \times 10^{-6}T^2$$

$$\lambda_{PuN}(T) = 8.18 + 0.0522 \times T - 9.44 \times 10^{-7}T^2$$

$$\lambda_{AmN}(T) = 8.99 + 0.00147 \times T - 2.54 \times 10^{-8}T^2$$

Metallic fuel thermal conductivity

Thermal conductivity [W/m/K]

- Thermal conductivity of metallic alloys fuels is similar to that of nitride fuels

- Conductivity of plutonium is lower than that of uranium

- Melting temperature is much lower (≈1100°C)

- Margin to melt is smaller

Learning activity

- Calculate the temperature difference between the surface and centerline of a fuel pellet, assuming a linear power of 36 kW/m and the approximation of a constant thermal conductivity equal to

- 3 W/m/K, corresponding to fresh $(U,Pu)O_2$ fuel, 100% dense

- 20 W/m/K, corresponding to fresh $(U,Pu)N$ fuel, 100% dense

$$\frac{\chi}{4\pi} = \int_{T_s}^{T_c} k(T)dT$$

- Real ceramic fuel pellets contain some porosity. How does this affect the effective thermal conductivity?

Correction for porosity

- Porosity reduces thermal conductivity, sometimes dramatically so.

$$\lambda_P = (1 - P_c)^a (1 - P_o)^b \lambda_{TD}$$

- P: porosity of the fuel

- P_c: closed porosity

- P_o: open porosity

- $a \approx 3/2$, exact for randomly distributed spherical pores

- $b < 7$, highly dependent on shape of open porosity.

Fuel temperature and performance

A small fuel temperature gradient corresponds to

- Smaller segregation, as a consequence of lower diffusion rates

- Lower fission gas release into the fuel pin plenum

- Lower pressure on fuel pin cladding at high burnup

However

- At some critical burnup, fission gas bubbles form inside fuel pellets, causing pressure leading to swelling of the pellet.

- Fuel design requires compromise between desire for low fission gas release and low swelling rates

High temperature stability

- By "instability" we mean loss of mechanical integrity under a phase change.

- Examples:

  - Melting

  - Vapourisation

  - Dissociation

- Actinide compounds may vapourise or dissociate at temperatures below their melting point!

Reprocessing

- Generations IV reactor fuels must be reprocessable, i.e. it must be possible to separate fission product from spent fuel, and recover actinides in a form suitable for new fuel fabrication.

- The losses of actinides to the fission product stream must be small, in order to achieve significant reductions (> factor of 10) of long term radiotoxic inventories

- Chemical reagents should to be cheap and radiation resistant

- Many chemical processes/reagents are known that can separate U, Pu, Am & Cm with small losses on laboratory scale (grams to kilograms)

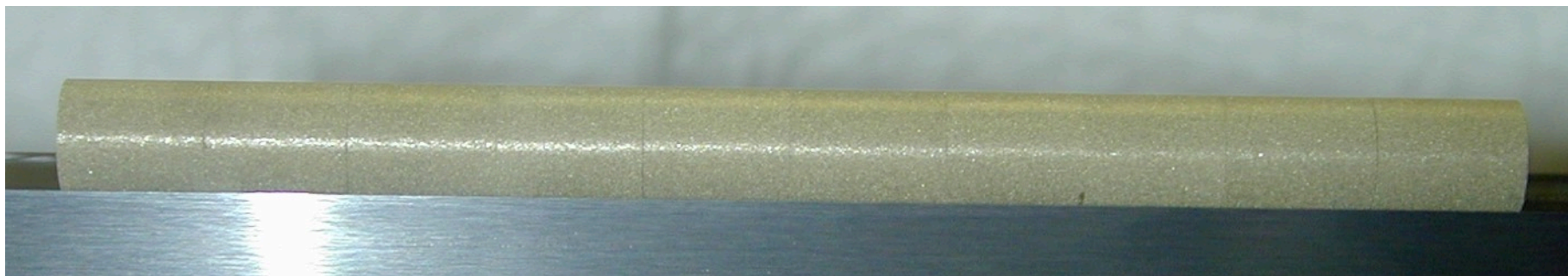- Not all processes can be scaled to industrial levels (tons)
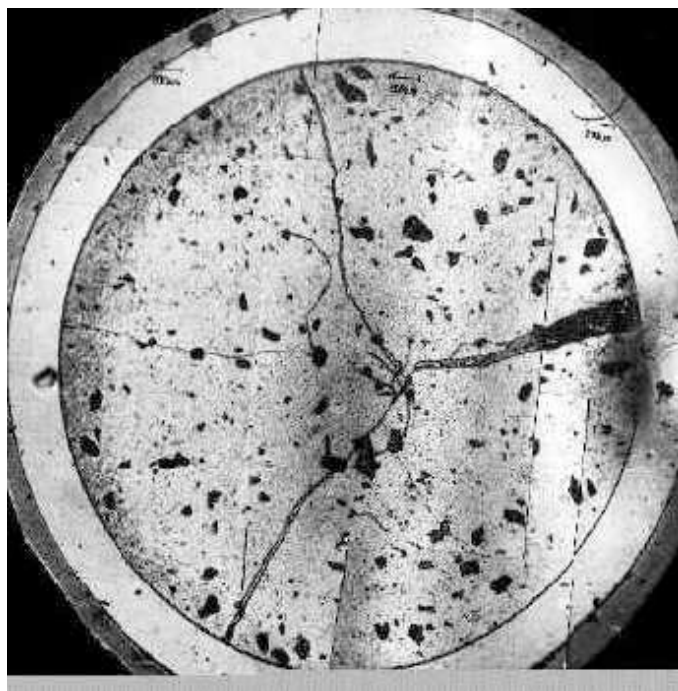
Break!

Potential fuel compositions for Gen-IV reactors

- The following fuel compositions have been used as driver fuels in fast neutron reactors

- Oxide fuels (USA, France, UK, Germany, Russia, Japan)

- Metal alloy fuels (EBR-II, USA, DFR, UK)

- Nitride fuels (BR-10, Russia)

- Carbide fuels (BR-10, Russia, FBTR, India)

Carbide fuels

- Carbide fuels pros & cons:

  + High conversion ratio (thanks to high density of actinides)

  + High thermal conductivity (small ΔT over fuel)

  + Proven burnup of 16% in FBTR

  − Pyrophoricity (must be fabricated in vacuum)

  − Volatility of AmC (vapourises at T > 1500 K in vacuum).

  − Reprocessing issues

  Cannot be used for recycle of Americium!

Metal alloy fuel design

Na-bond →

Clad →

Fresh fuel slug
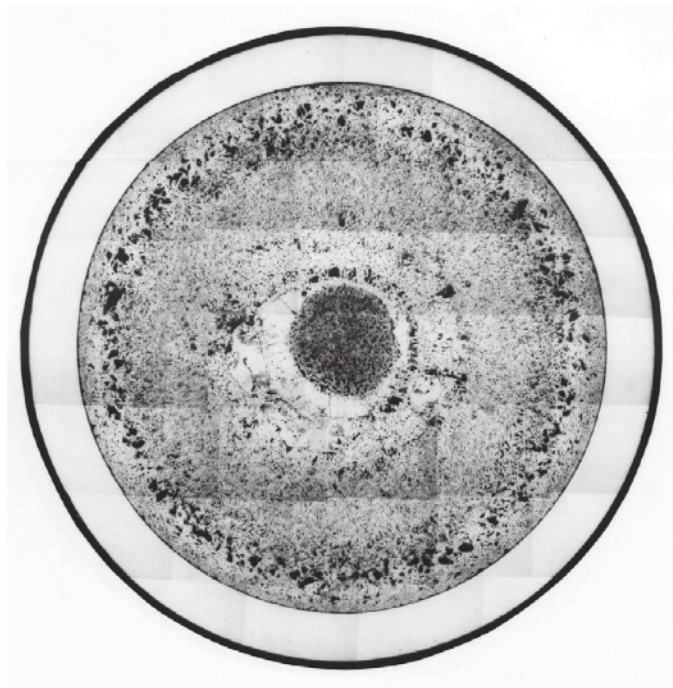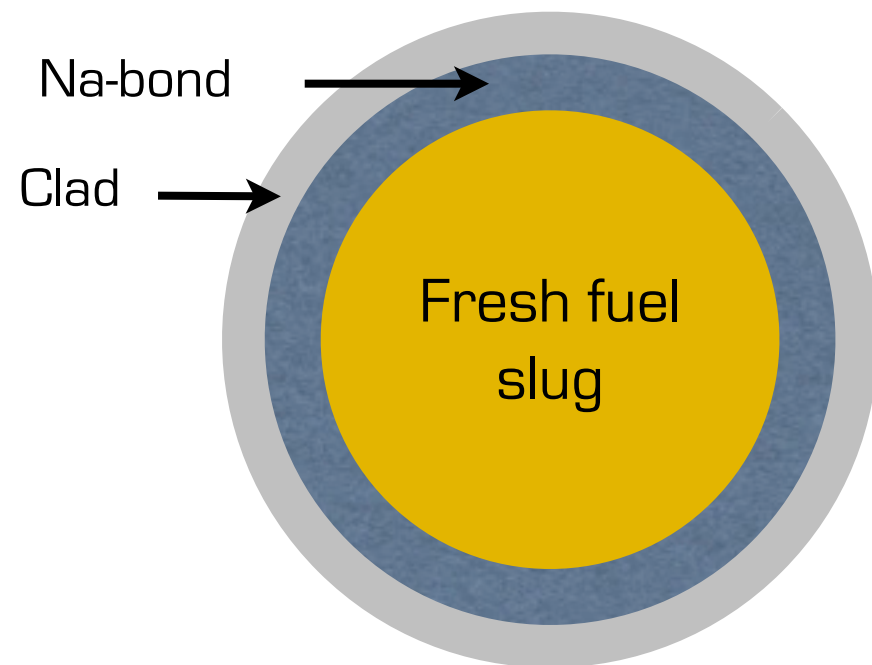
U-29Pu-4Am-2Np-30Zr @ 6% burnup

- Fuel is fabricated with 75% smear density.

- Initial swelling rate: >15% per percent burnup, due to pressure from fission gas bubbles!

- > 90% of fission gases are released when reaching ≈ 30% swelling. Plenum size is increased to reduce gas pressure induced clad stresses.

- No fuel clad mechanical interaction (FCMI) until solid fission products fill pores (> 18% burnup)

- Redistribution of alloy components take place due to thermal gradient in fuel.

- Effective thermal conductivity ≈ 20 W/(m*K)

- Linear rating: < 35 kW/m

Metal fuel melting temperature

- Plutonium metal has very low melting temperature

- Zirconium alloying required to raise melting temperature

Metal	T_{melt} [K]
U	1405
Np	910
Pu	912
Am	1449
Zr	2128

Metal alloy fuels

Metal alloy fuels pros & cons:

+ Highest conversion ratio - thanks to high density of actinides.

+ High thermal conductivity

+ Proven burnup of 18 % in EBR-II

+ Dust free fabrication

+ High axial expansion coefficient (substitutes for Doppler).

− Low melting temperature (Pu must be alloyed with Zr)

− Volatility of Am (rapid cooling of molten alloy during fabrication)

− Large swelling rate requires large sodium bonded pellet clad gap

− Sodium bond requires pyrometallurgical reprocessing

Nitride fuels

Nitride fuels pros & cons:

+ high conversion ratio, thanks to high density of actinides.

+ High thermal conductivity

+ High solubility rate of PuN in nitric acid

+ Proven burn-up of 20 %

− Low operational temperature results in small creep rate: pellet-clad mechanical interaction (PCMI) to be avoided

− Production of ^{14}C by $^{14}N(n,p)$ reaction. Enrichment in ^{15}N necessary, which may lead to significant cost penalty for Gen-IV fuels

− To avoid dissociation of AmN, T < 1800 Kelvin during fabrication
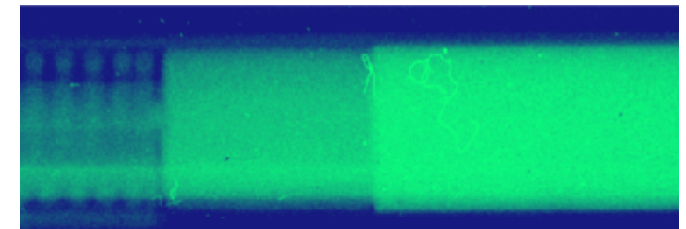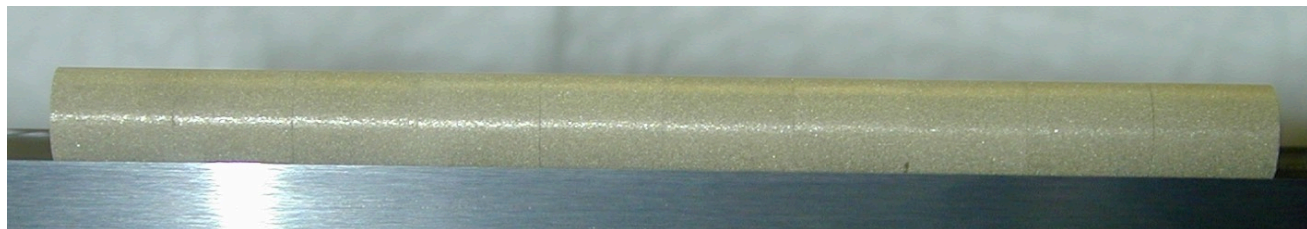
Nitride fuel fabrication: Carbothermic nitridation
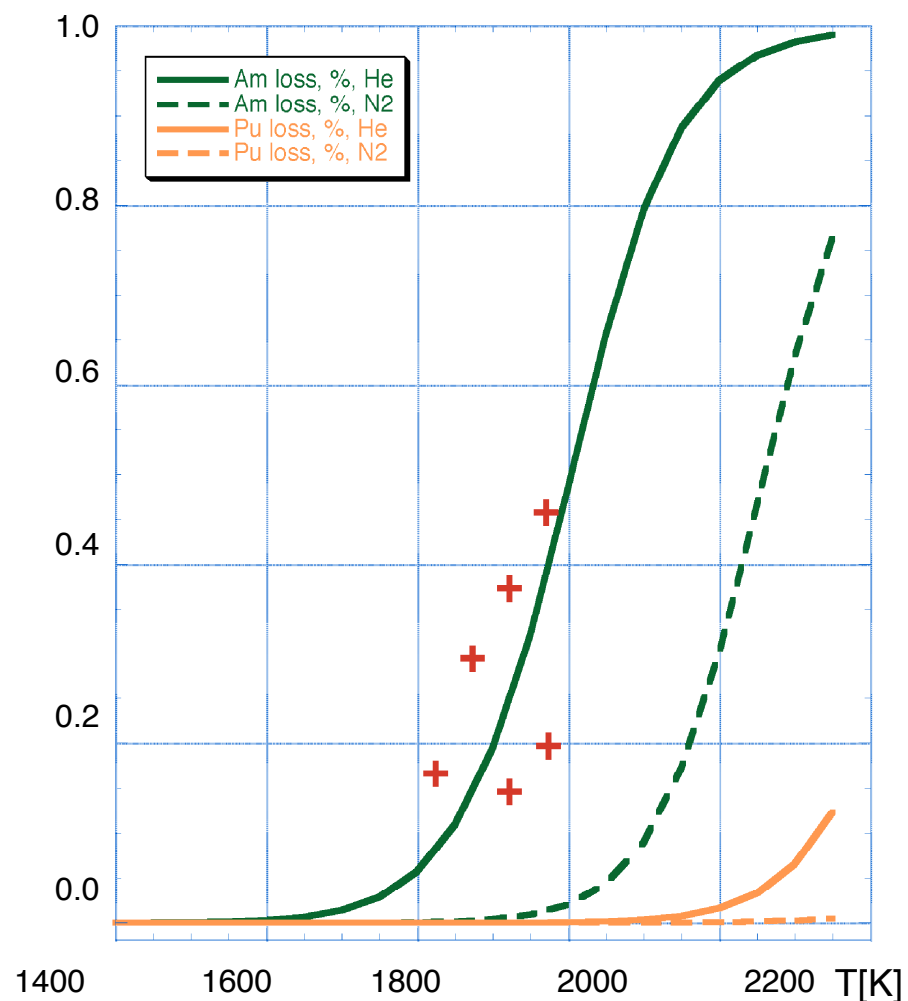
- (Pu,Am,Zr)N powder is fabricated by carbothermic nitridation of oxides:

- $2MO_2 + 4C + N_2 \rightarrow 2MN + 4CO$

- After pressing, pellets with ≈ 40% porosity are obtained.

- Porosity is reduced by sintering at high T.

- Sintering under argon, AmN is lost if T > 1600 K

- Sintering under nitrogen, T_{lim} = 1800 K.

Nitride fuel fabrication: Ammonolysis of fluorides and field assisted sintering techniques.

Uranium-ammonium-fluoride

UN

SPS

(U, Np, Pu, Am)N
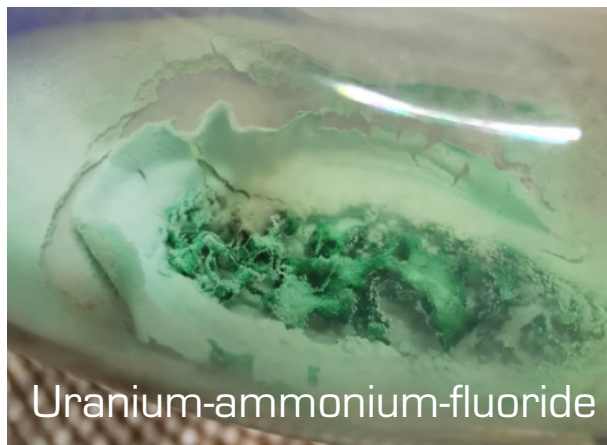
- UN powders fabricated at KTH by ammonolysis, i.e. exposure of UF_6 to ammonia.

- UN pellets fabricated at KTH using spark plasma sintering (SPS) technique: Current assisted (1000 A, 10 V) hot pressing.

- UN pellets with zero porosity obtained when holding for 3 minutes at 1650°C.

- Pellets with 95% density obtained at 1450°C.

- High-voltage electric pulse consolidation (HVEPC) applied by VNIINM in Russia for manufacture of (U, Np, Pu, Am)N pellets (40 kJ, 6 kV).

Nitride fuel performance: swelling

Volumetric swelling of UN [%]
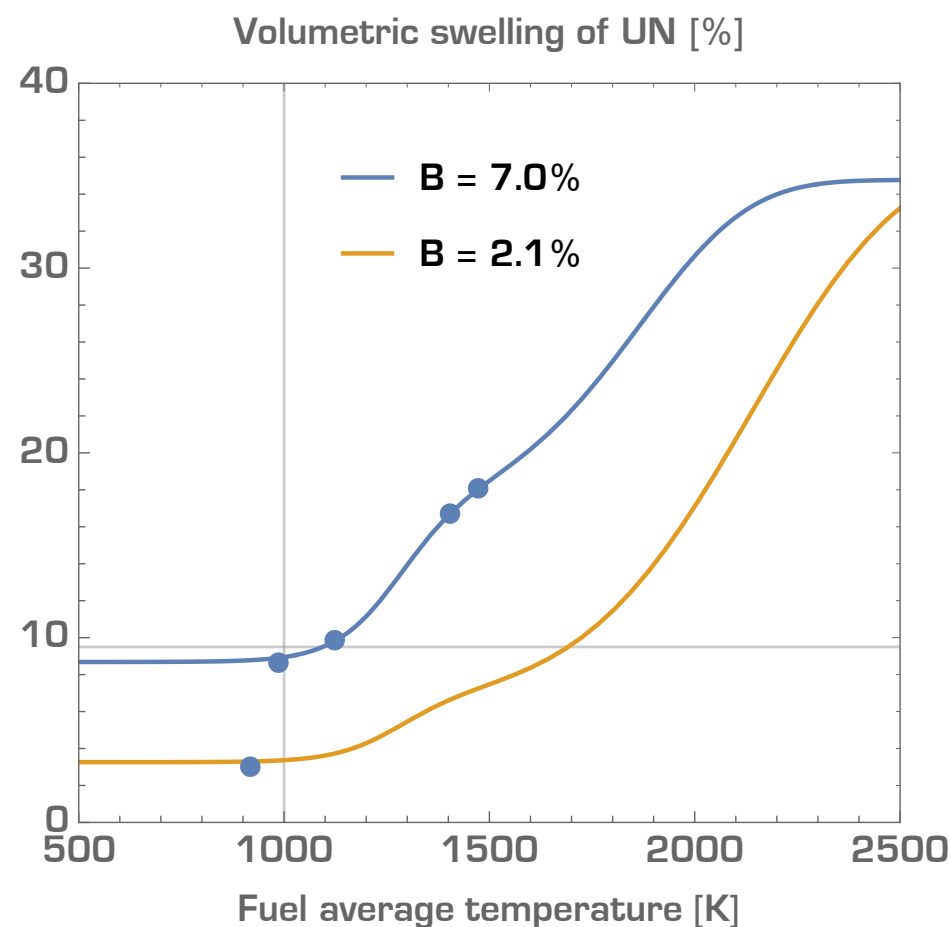
- Nitride fuels are dense, hence have less space in the lattice for accommodating fission products

- Swelling is highly temperature dependent

- Minimum swelling rate at low T: 1.5% per percent burn-up

- Once PCMI occurs (Pellet-Clad-Mechanical-Interaction) the fuel is so hard that cladding will break.

- Fuel rod to be designed with sufficiently large pellet-clad gap to accomodate swelling

Nitride fuel performance: gas release

Fission gas release [%] vs Temperature[K]

- 13–19% burnup (exp)
- 3.7–9.1% burnup (exp)
- 15% burnup (model)
- 7% burnup (model)

Fission gas release [%] vs Burn–up [FIMA]

- 1800 K
- 1600 K
- 1400 K
- 1200 K
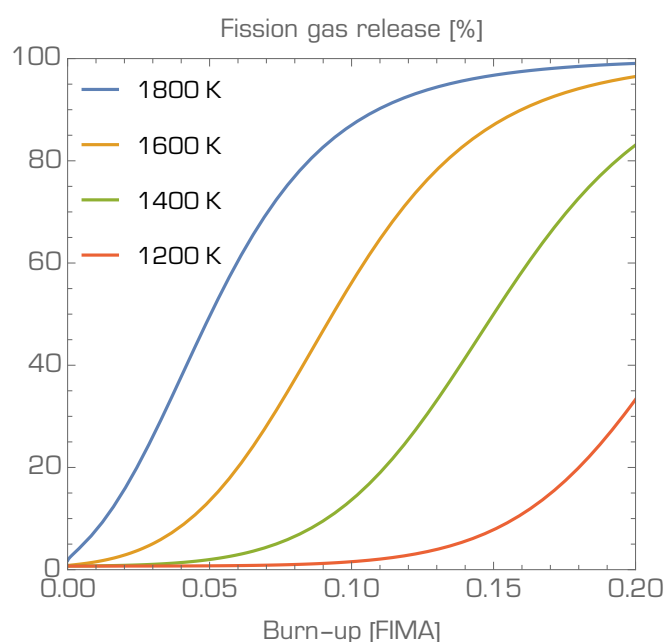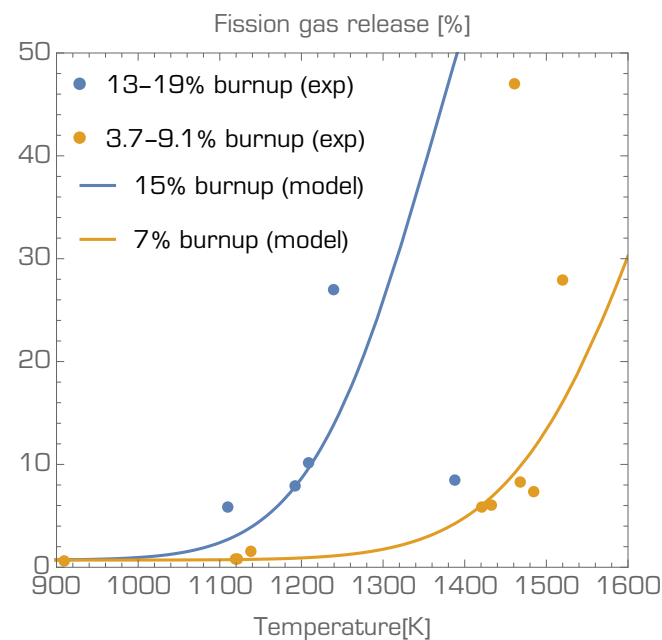
- Nitride fuels feature low fission gas (Xe & Kr) release .

- Even at very high power density, fission gas release remains below 50%.

- Fuel rod gas plenum size may be reduced.

- Experimental data can be represented by equation based on a calculated fission gas migration barriers [Wallenius 2022].
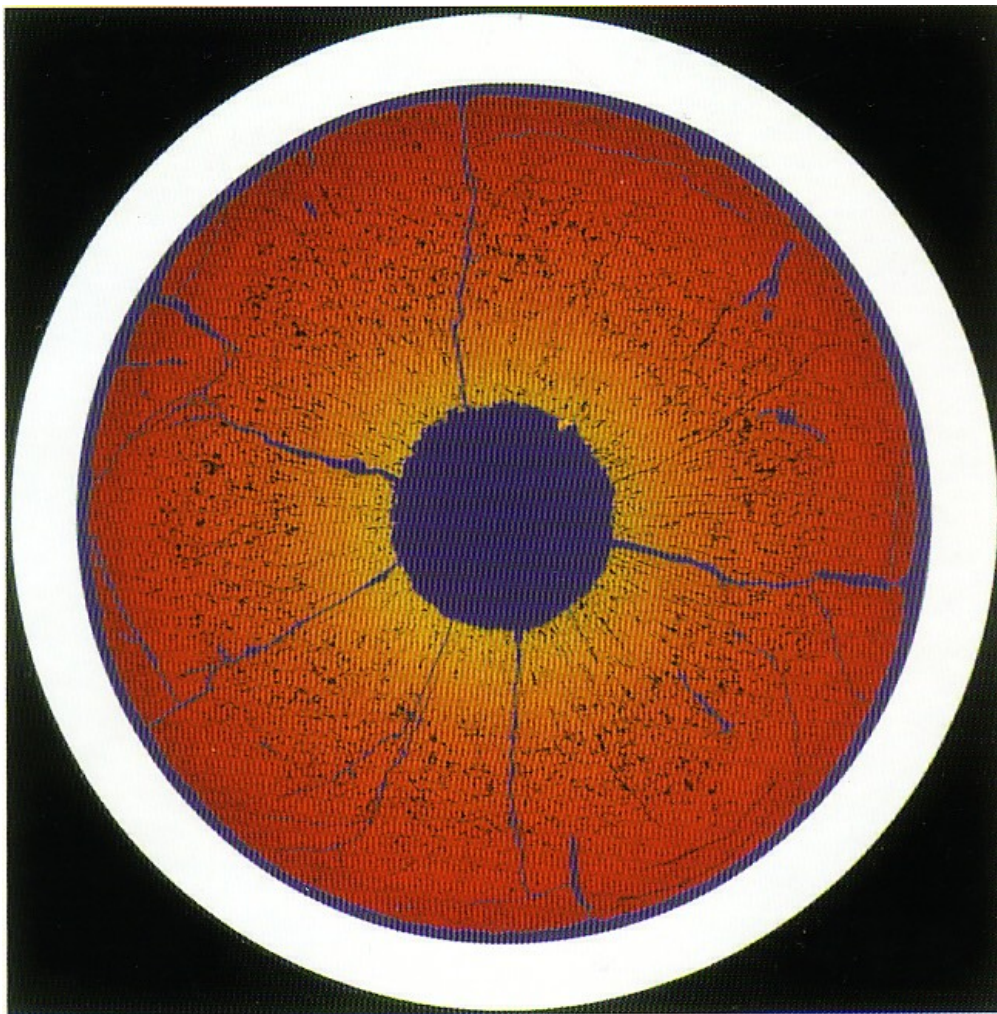
$$R(B, T) = R_{at} + \frac{1 - R_{at}}{1 + D_{FG}\,(1 - \sqrt{B})\,\exp\left[\frac{E_{mig}^{FG}}{k_B T}(1 - \sqrt{B})\right]}$$

Nitride fuel melting temperature

- Nitride fuels dissociate into metal and gas under normal pressure

- Under in-pile conditions, melting occurs once internal nitrogen gas pressure has built up.

| Compound	T_{melt} [K]	P_{N2}[bar]
UN	3120±30	> 2.5
NpN	3100±30	> 10
PuN	3100	> 50
AmN	?	?

Oxide fuel: restructuring

- Fresh oxide fuel has 5-10% porosity

- Low thermal conductivity leads to large radial temperature gradient.

- T > 1400 K: grain growth

- T > 2200 K: pore migration along ΔT

- Central hole forms in FR oxide fuels

- Swelling rate: 0.7% per percent burn-up.

- Gas release is high.

Oxide fuel melting temperature

Oxide fuels combine very high melting temperatures with the best high temperature stability of the fuels considered for Gen-IV applications.

Compound	T_{melt} [K]
UO_2	3130±20
PuO_2	3017±28
AmO_2	2773±16

Impact of irradiation on thermal conductivity

Thermal conductivity [W/m/K]

UOX (100% TD, O/M=2.0)

UOX (10% burnup)

T [K]
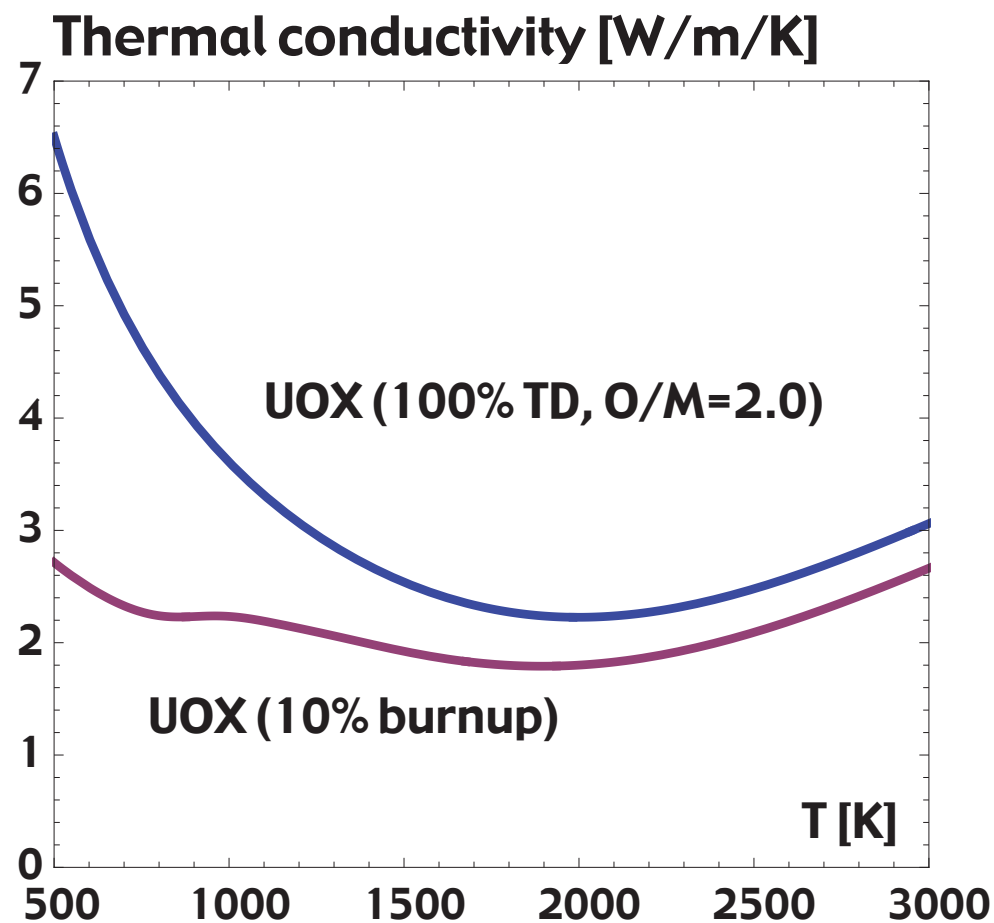
- Oxide fuel conductivity is dominated by crystal lattice contribution at low T.

- Fission product accumulation will perturb phonon propagation

- Thermal conductivity in the low T region is dramatically reduced by irradiation

Oxide fuels

Oxide fuels pros & cons:

+ Proven burnup > 25 % (large body of irradiation experience)

+ High operational temperature yields high creep rate, negligible PCMI

+ High melting temperature

+ Good stability at high T for Am_2O_3 and Cm_2O_3

− Potential for chemical interaction with clad (internal clad corrosion)

− Poor thermal conductivity, leading to redistribution

Summary

- Oxide fuels are relatively easy to fabricate and perform well in-pile. Their low thermal conductivity however leads to a low margin to failure. Hence, the cost penalty for increasing the americium fraction in driver fuel is high.

- Americium carbide is highly volatile under vacuum. Carbides must be fabricated under vacuum not to ignite. Hence not suitable for Generation IV reactor driver fuels.

- Metallic fuels swell rapidly, hence a large fuel-clad gap must be implemented and filled with sodium. Therefore, metal fuels cannot be reprocessed with hydro-metallurgical methods.

- Nitride fuels may be fabricated using field assisted sintering techniques. Nitrides are activated under irradiation. In order to avoid C-14 production, N-15 must be used for fabrication, at a cost penalty.