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Fission gas release

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Fission gas release

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Joint ICTP-IAEA Workshop on

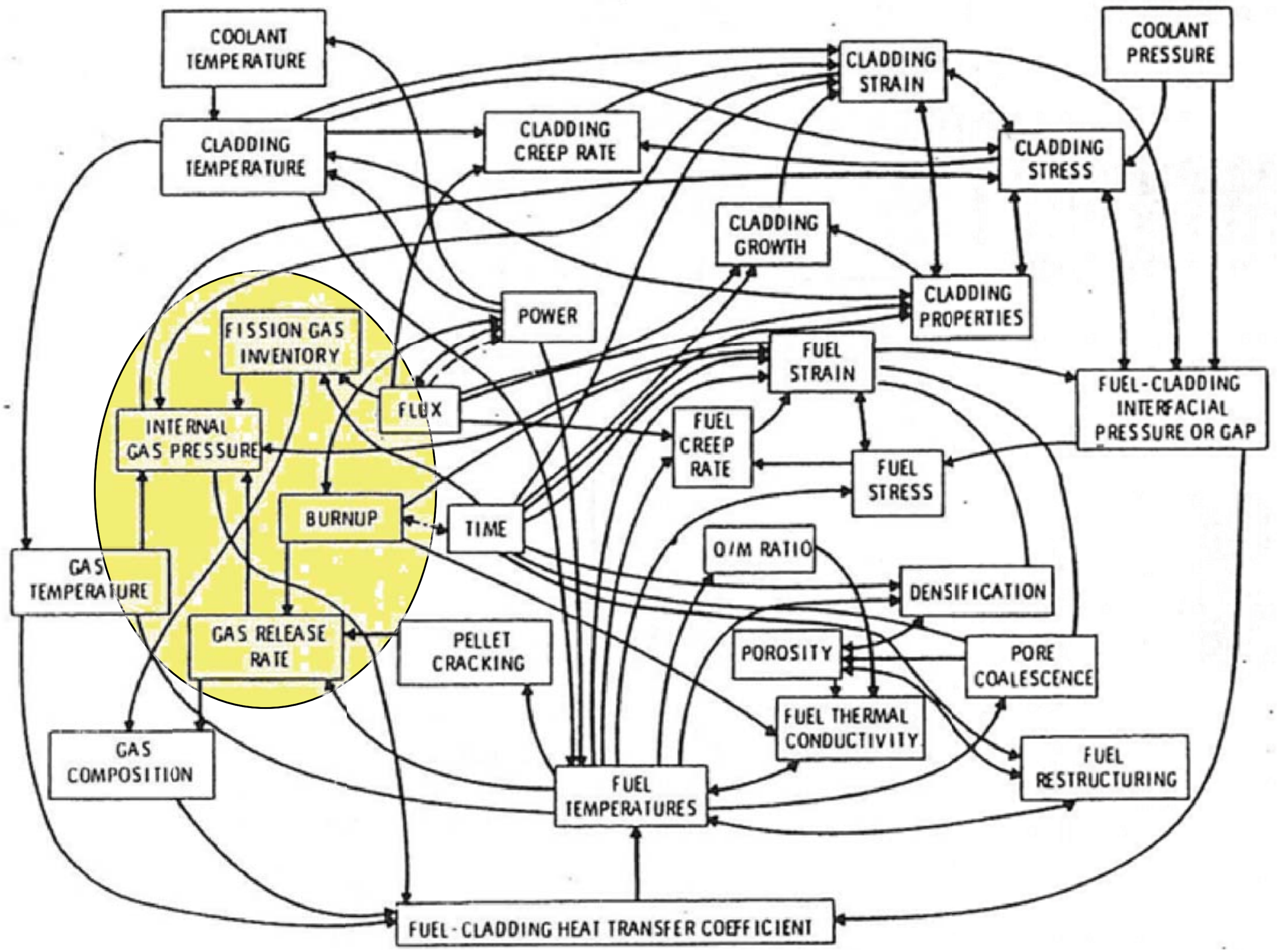
The Training in Basic Radiation Materials Science and its Application to Radiation Effects Studies and Development of Advanced Radiation-Resistant Materials

Trieste, April 12-23, 2010

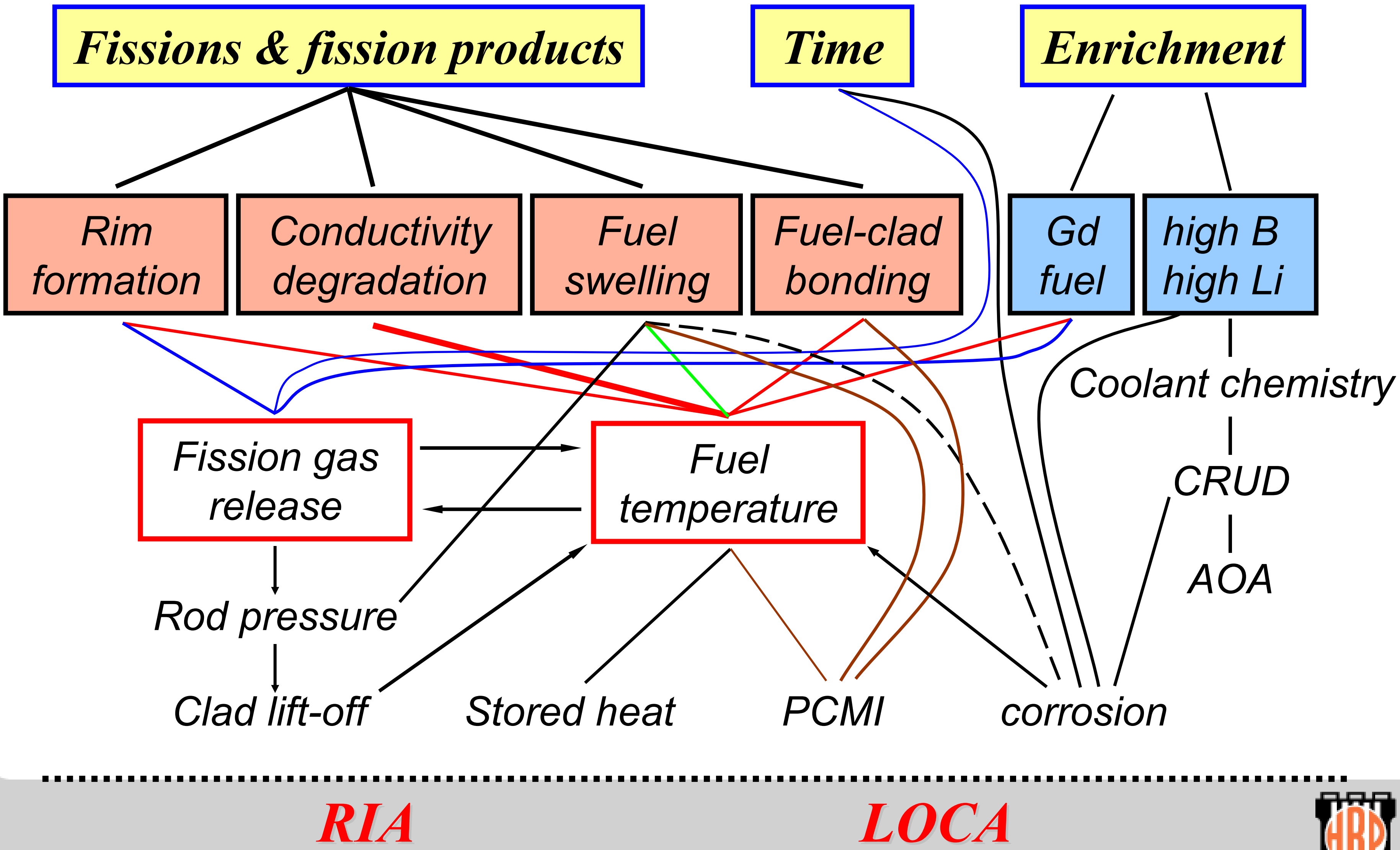
Overview

- Why fission gas release calculation?
  - safety criteria
  - interaction with other phenomena
- Basic mechanisms
- Experimental data

... but it may be more complicated

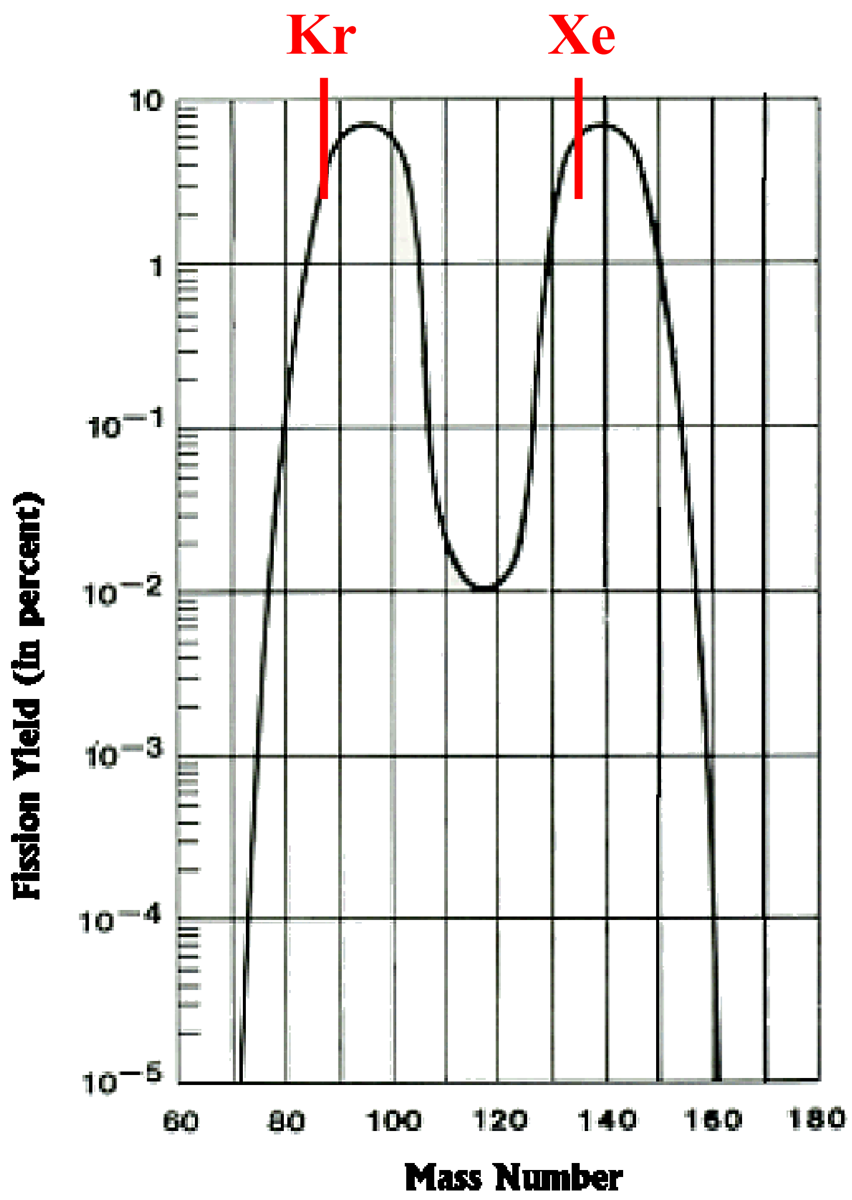


Fuel behaviour codes must address and untangle interactions that become more and more complex as fuel burnup increases



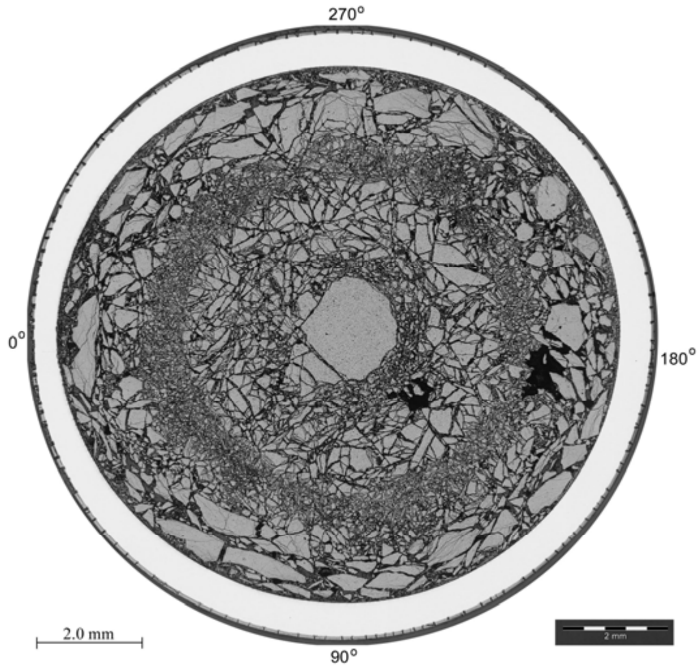
Fission products

- The fission products range in mass from around 72 to 161 (U-235) with peaks in the yield distribution close to the elements krypton and xenon
- The overall yield of these so-called fission gases is approximately 28 atoms per 100 fissions
- About 85% are Xe atoms, about 15% are Kr atoms (Kr yield is lower for Pu fiss.)
- Xe and Kr are noble gases with low solubility in the fuel matrix



There is no ideal place for fission gases

- If released, they can lead to
  - thermal runaway (gap cond.)
  - rod-overpressure
    → restricted by safety criteria
- If retained, they can lead to
  - large fission gas swelling and thus pellet-clad interaction
  - grain boundary embrittlement and porosity with high pressure, both contributing to fuel fragmentation under accident conditions (RIA, LOCA)



- Safe reactor operation therefore requires a thorough understanding of the behaviour of fission gas

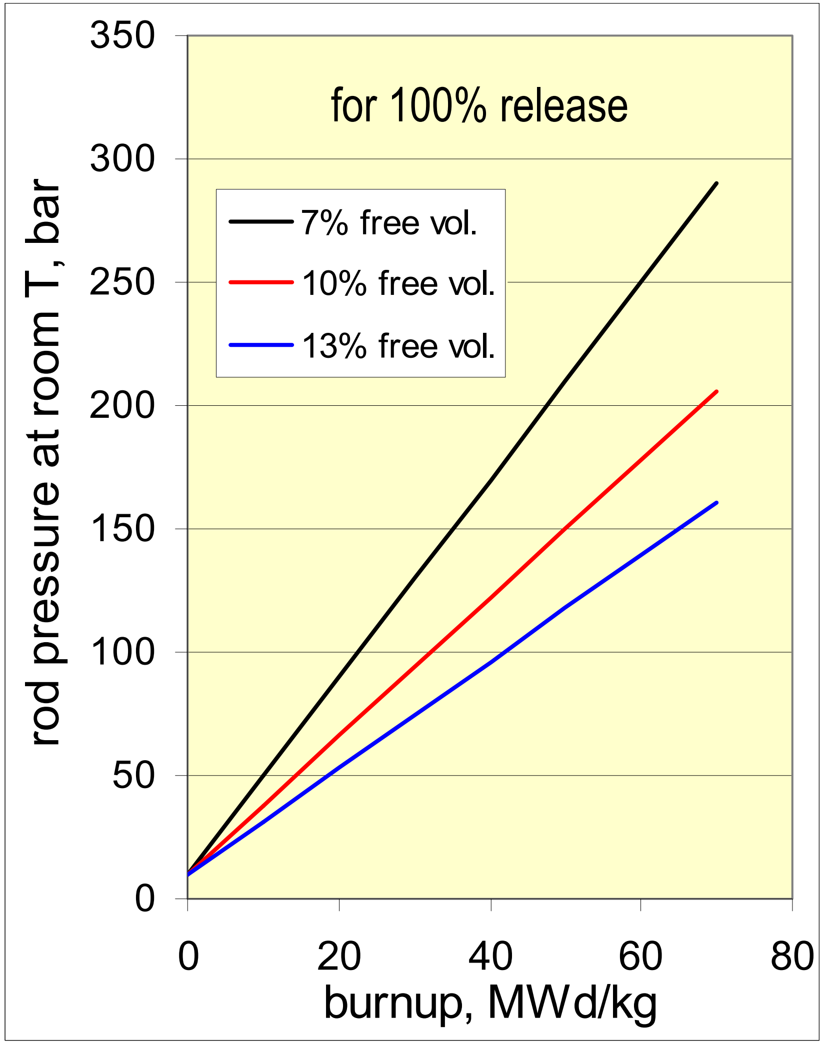
Fission gas release and safety criteria

- Fission gas generation is about 28 cm^3/MWd
- A fraction is released from the fuel matrix to the free rod volume, increasing the rod pressure
  - all gas released → rod pressure 150 - 200 bar at room T
- Rod pressure is limited by safety criteria and must therefore be calculated for the safety case
- Regulation differs from country to country and may prescribe:
  - rod pressure must not exceed system pressure
  - a pressure limit which is above system pressure (e. g. Belgium, 184 bar)
  - pressure may exceed system pressure, but must not lead to gap opening (no “lift-off” causing thermal feed-back)

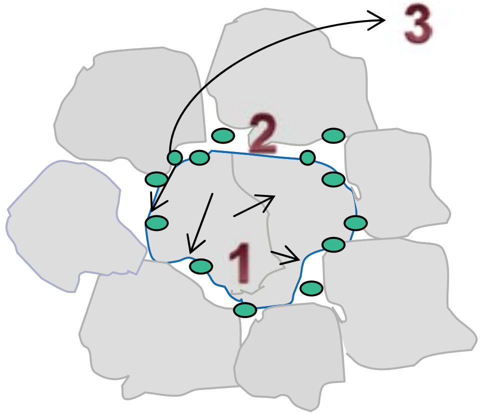
Rod pressure increase

For rod pressure calculation, fuel modelling codes have to take into account

- generated fission gas (easy)
- **released fission gas** (hard)
- change of free volume
  - fuel densification and swelling
  - clad creep and growth
- temperature of free volume
  - plenum
  - dishing, centre hole
  - pellet-cladding gap, chamfer
  - open fuel vol. (pores, cracks)



Fission gas release
- basic mechanisms -



- Fission gas atoms diffuse from within the fuel grain to the grain surface (temperature driven)
- The fission gas atoms accumulate at the grain surface in gas bubbles
- When the surface is saturated with bubbles, they interlink and the gas is released out of the fuel matrix
- FGR depends on temperature and burn-up (time)
- FGR is <1% for temperatures below ~1000-1200^{o}C and ~10-20% at ~1500 ^{o}C

Fission gas release
- phenomena involved -

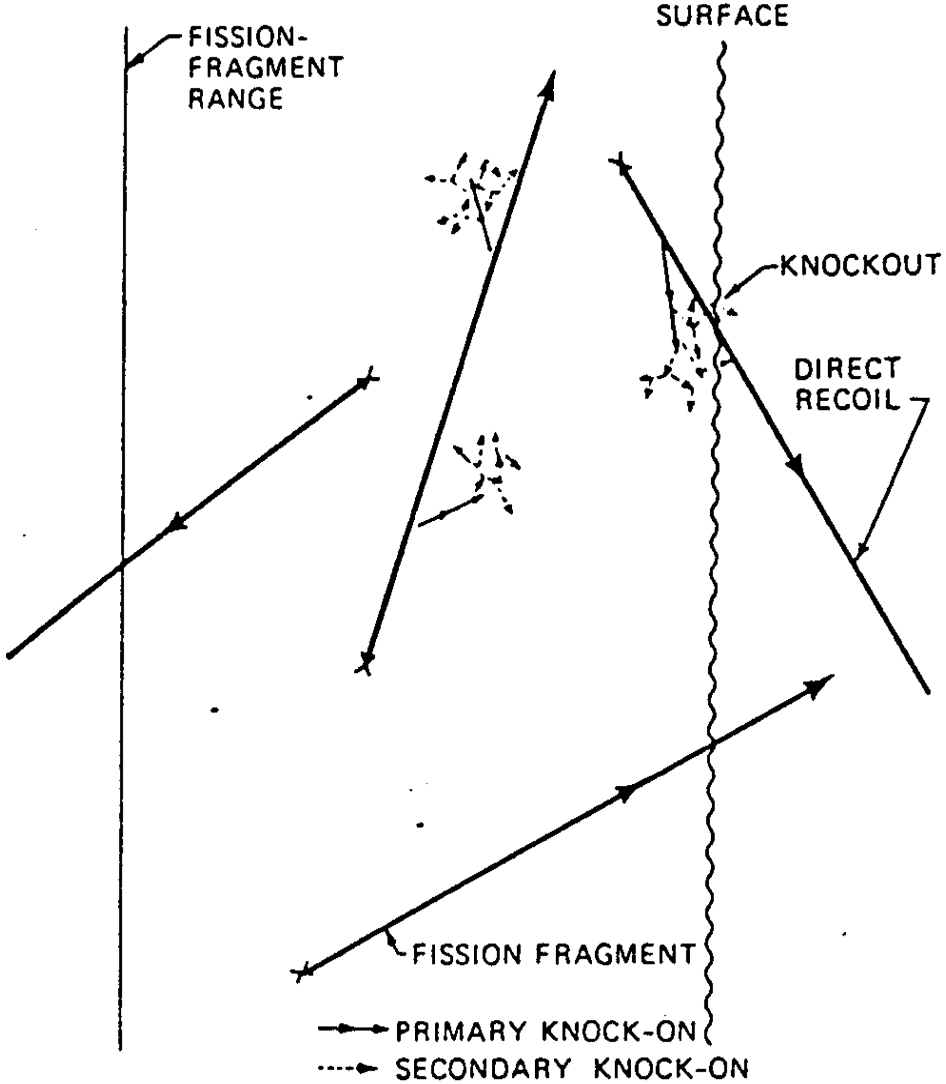
1. *Recoil*
2. *Knock-out & sputtering*
3. ***Lattice diffusion***
4. Trapping
5. Irradiation re-solution
6. Thermal re-solution
7. Densification
8. Thermal diffusion
9. Grain boundary diffusion
10. Grain boundary sweeping
11. Bubble migration
12. ***Bubble interconnection***
13. Sublimation or vaporisation

For a detailed discussion of these phenomena (and more) see

FISSION GAS RELEASE, SWELLING & GRAIN GROWTH IN UO_2

R. White, HPR-368, February 2008

(on Summer School CD)



$$R_{recoil} \cong Surface / volume$$

Recoil

- When a fission fragment is close enough to a free surface (< 6-9 µm), it can *directly* escape from the fuel due to its high kinetic energy (60-100 MeV)
- It only affects the geometric outer layer of the fuel

  $R_{recoil} \sim Fis.rate \cdot Surface/Volume$
- It is an athermal mechanism (independent of temperature and temperature gradient)
- Domain of application is for T<1000°C

Knock-out and sputtering

- Knock-out
  - The interaction of a fission fragment, a collision cascade or a fission spike with a stationary gas atom close enough to the surface can cause the latter to be ejected
- Sputtering
  - A fission fragment travelling through oxide looses energy by interactions with UO_2 at a rate ~1keV/Å
    - $\rightarrow$ high local heat pulse
    - $\rightarrow$ when it leaves the free UO_2 surface, the heated zone will evaporate or sputter
- Affect outer layer and open surfaces: $(S/V)_{total}$
- Athermal; independent of T and temp. gradient

$$Release/Birth \sim (Surface/Volume)_{total}$$

Lattice diffusion, diffusion coefficient

$$D = D_a + D_b + D_c$$

- *Thermal coefficient (T>1400°C)*

$$D_a = D_0 \exp\left(-\frac{\Delta H}{RT}\right)$$

- *Intermediate*

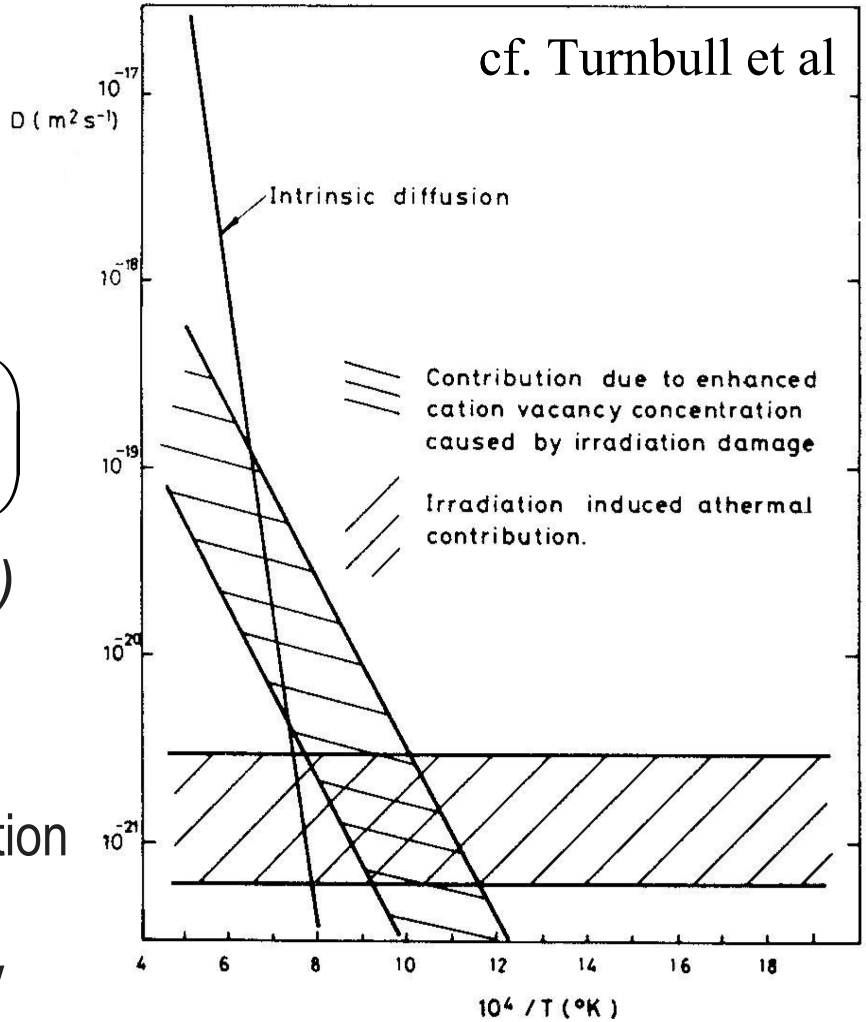
$$D_b = 6.64 \cdot 10^{-25} \sqrt{\dot{F}} \exp\left(-\frac{13800}{T}\right)$$

- *Athermal coefficient (T<1000°C)*

$$D_c = A \cdot \dot{F}$$

- The release rate is enhanced by deviation from stoichiometry (UO_{2+x}) and by impurities simulating non-stoichiometry

cf. Turnbull et al



Fission gas release
- basic diffusion model -

- Fission gas release was observed very early on and explained by diffusion out of the fuel grains (Booth)
- Assumptions for (simple) model:
  - atomic diffusion in hypothetical sphere
  - grain boundary = perfect sink
  - gas at grain boundary immediately released (?)
  - constant conditions of temperature and fission rate

$$\frac{\partial c}{\partial t} = D\Delta C + S$$

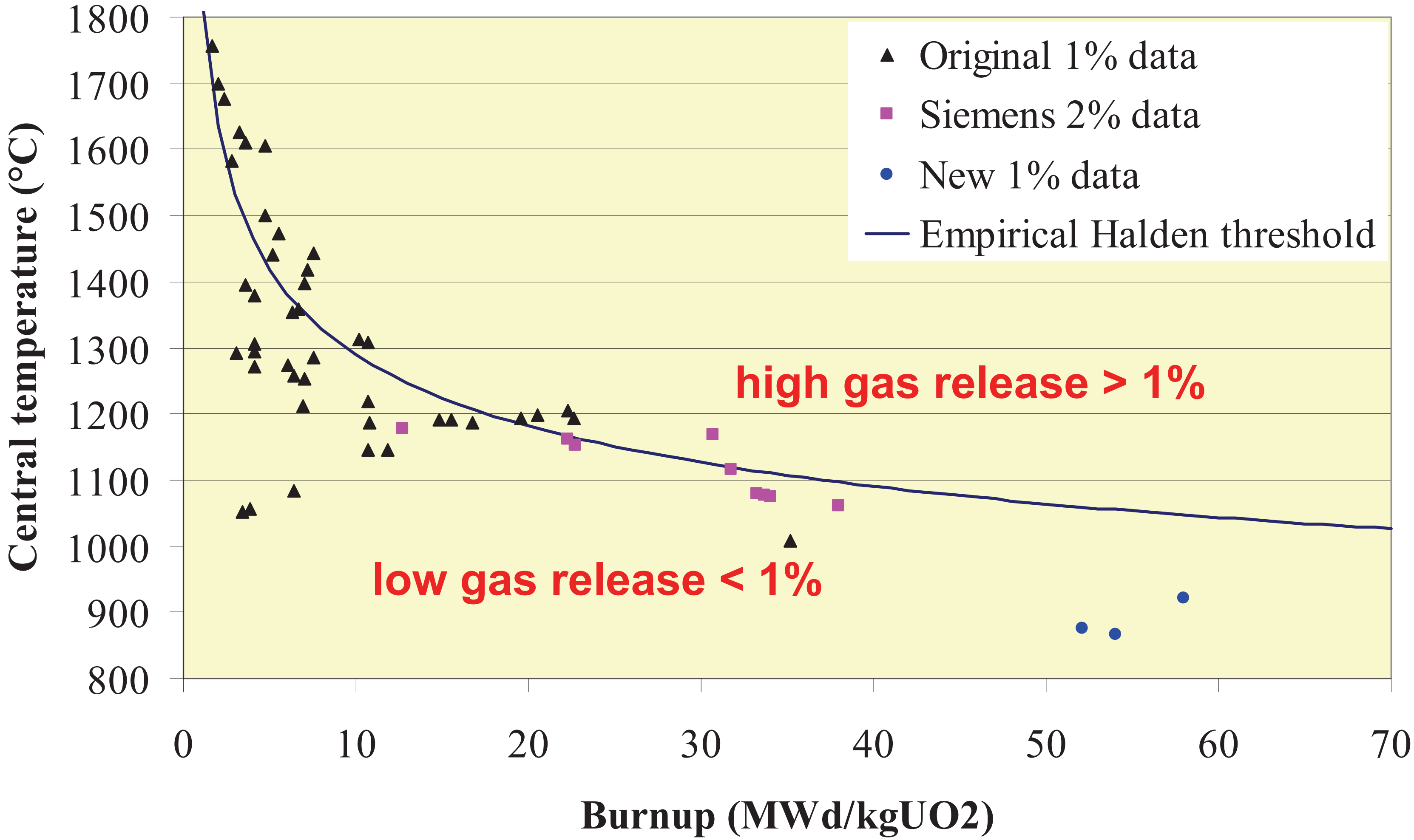
$$\begin{cases} C\left(R_{gr}, t\right) = 0 \\ \left.\dfrac{\partial c}{\partial r}\right|_{r=0} = 0 \end{cases}$$

$$C\left(R_{gr}, 0\right) = 0$$

  - Solution (Booth diffusion, release rate)

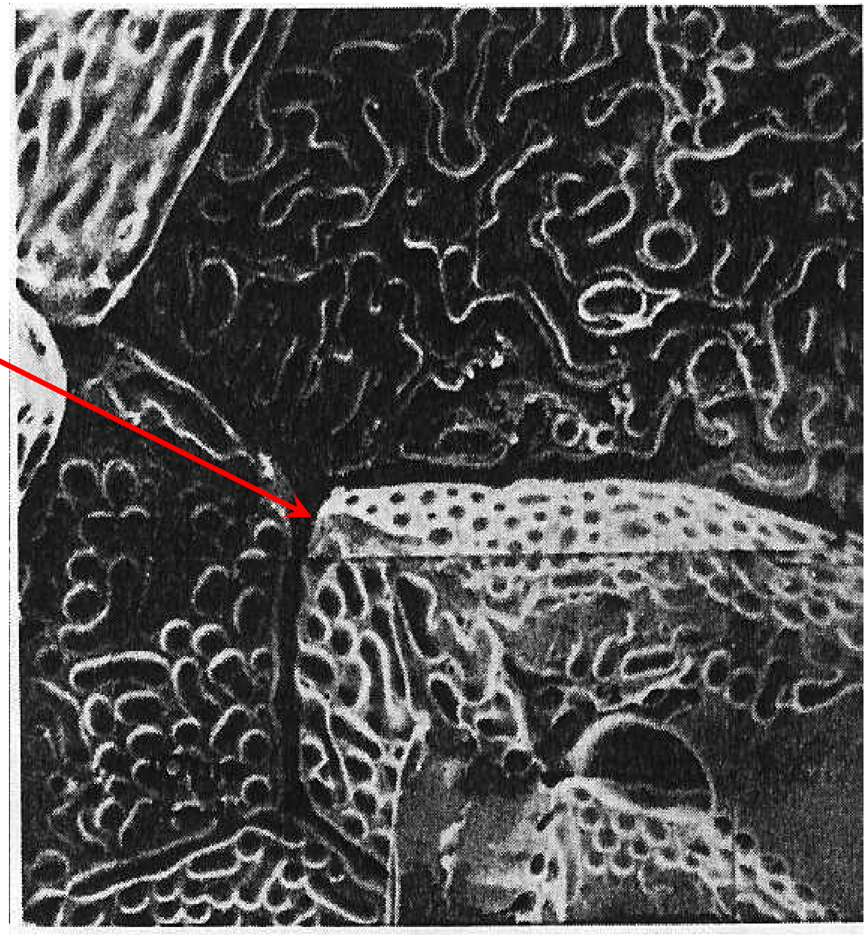
$$f(t) = 4\sqrt{\frac{Dt}{\pi R_{gr}^2}} - \frac{3}{2}\frac{Dt}{R_{gr}^2}$$

Observation: incubation threshold



Fission gas release
- Bubble interconnection -

- **Explains incubation** and onset of (stable) fission gas release during normal operation due to increase of open surface (*open tunnel network)*
- **Explains burst release** (micro-cracking) during abrupt power variations → requires precise knowledge of local stress



Classification of models

Empirical

- ☹ calibrated with specific data-base
- ☹ limited application range

- ☺ excellent performance
- ☺ inexpensive in use
- ☺ efficient tool for fuel design

Focus on speed, robustness, precision

Mechanistic

- ☹ many unknown parameters
- ☹ lack of detailed experimental data

- ☺ physical description of phenomena
- ☺ wide range of application
- ☺ possible to extend range

Focus on (physical) understanding

Fission Gas Release models

- Large number of models
  - Improvements
    - numerical techniques
    - new mechanisms
  - Various applications: conditions, reactor types, …
- Large uncertainties
  - (mechanistic) model parameters
    - diffusion coefficient
    - resolution, ...
  - input parameters:
    - temperatures
    - hydrostatic stress, ...

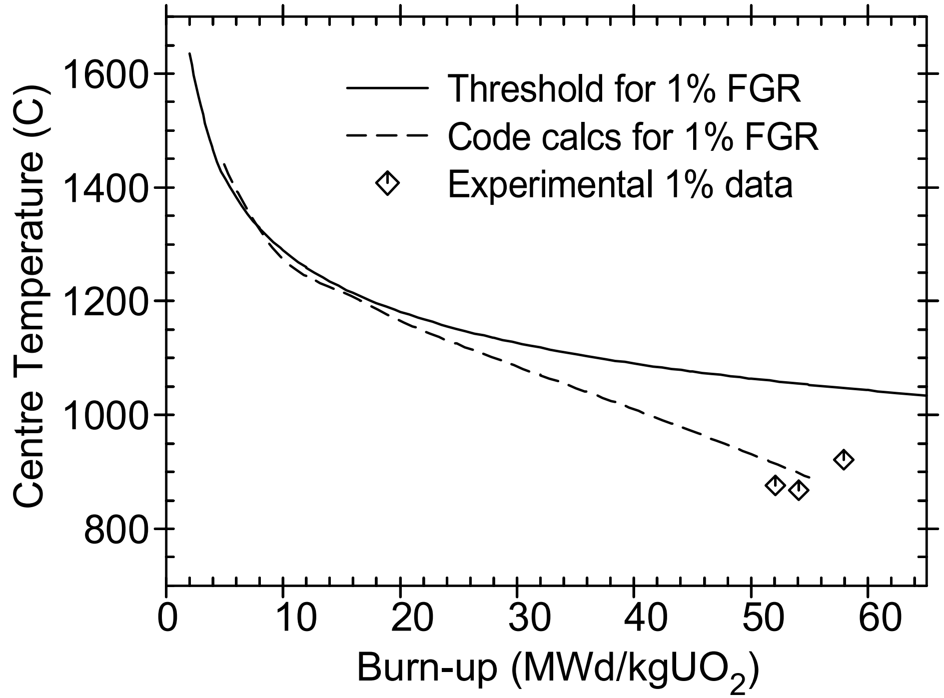
A simple model
(in use at Halden)

A simple, yet fairly successful model is in use at Halden which considers the basic influences and observations.

- Athermal release, always present
- Three-stage diffusion driven release process
  1. intergranular diffusion to the grain boundary, Booth model
  2. accumulation of gas atoms on the grain surface to a concentration of $5 \cdot 10^{15}$ cm^{-1} (not released to the free volume)
  3. reaching the limit concentration signifies bubble interlinkage, and from then on fission gas atoms arriving at the grain surface are assumed to be released to the free volume
- Special procedure to apply the Booth solutions (which are for constant conditions) to varying powers and temperatures

Comparison with FG release data

- An important result from the Halden Programme is the 1% FGR threshold established for UO_2 up to ~30 MWd/kgUO_2
- The simple model is able to reproduce the observations in the low burnup range using a three-term diffusion coefficient and fission gas accumulation on grain boundaries
- For higher burnup, a burnup dependent diffusion coefficient is required to maintain good agree-ment with the experimental data



Comparison of simple fission gas release model with empirically derived release threshold and experimental data at high burnup

Experimental

Measurements:

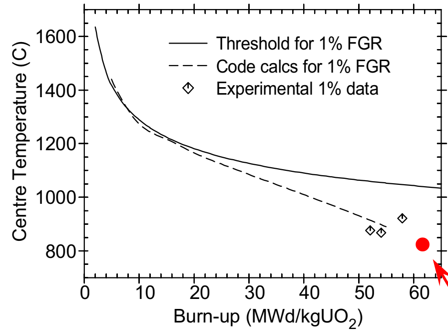
(see introductory lecture)

- Rod pressure
- Gamma spectroscopy
- Rod puncturing and gas analysis

Observations

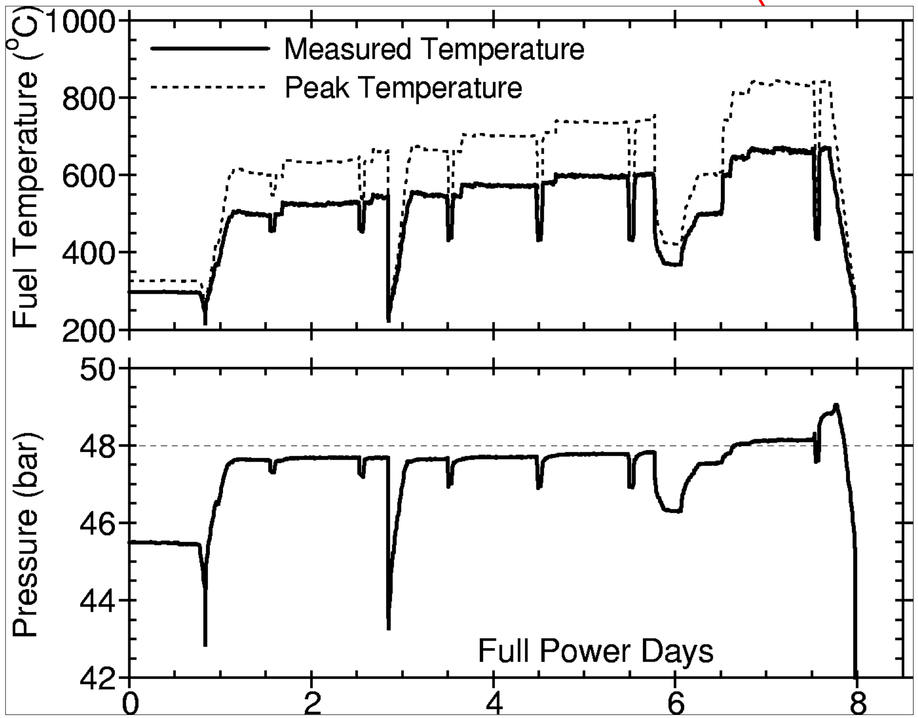
- Release onset
- Interlinkage (→ release onset)
- Release kinetics
- Grain size effect
- Gas mixing

Fission gas release onset



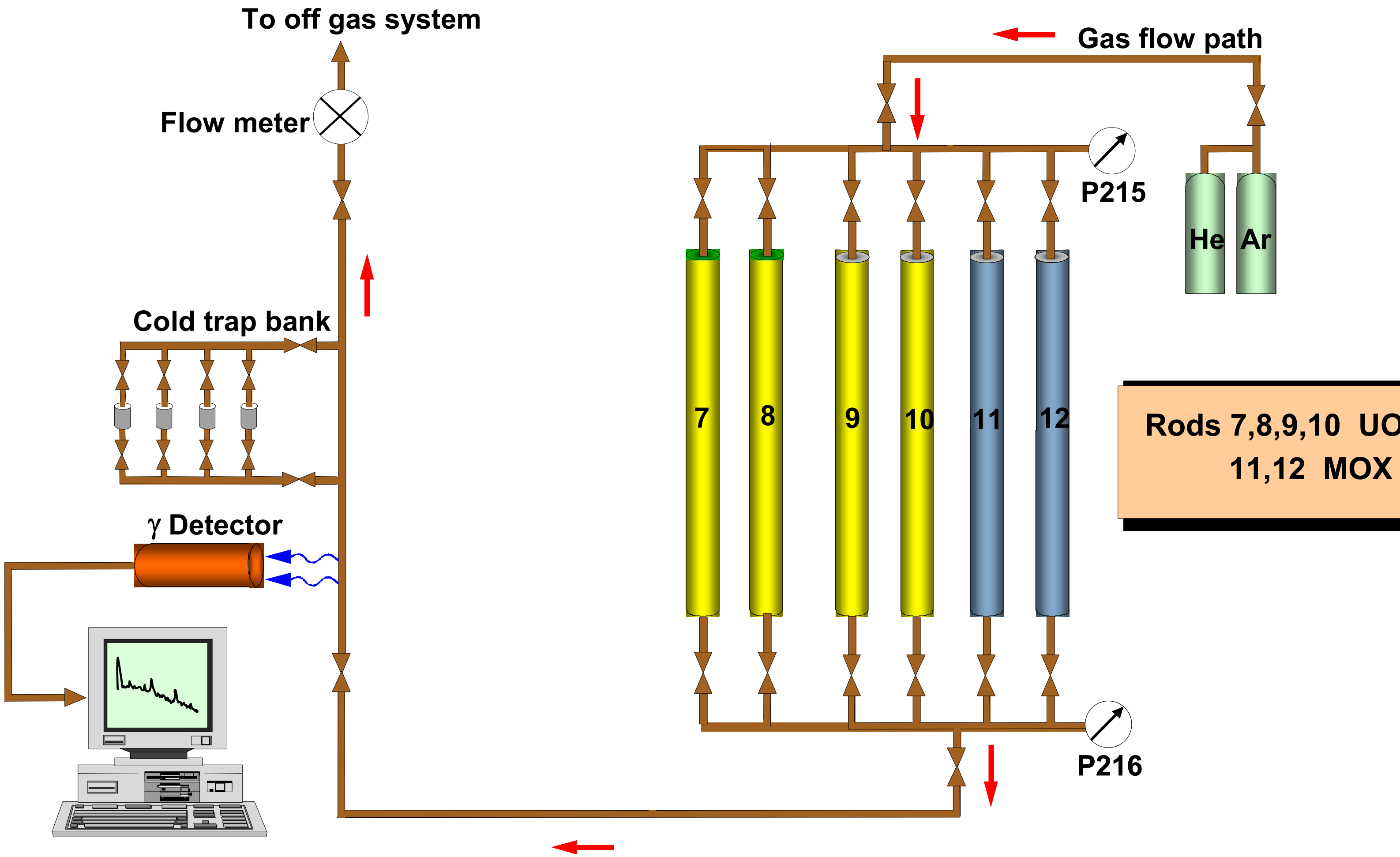
new point

- Irradiation of fresh and high burnup fuel (segments from LWRs)
- Instrumentation:
  - fuel thermocouple
  - rod pressure sensor
- Stepwise power / temp. increase to establish onset of fission gas release
- Simultaneous measurement of fuel temperature (most important parameter) and pressure



Temperature history and measured rod pressure (fission gas release)

Gas flow system
(radioactive fission product analysis)



Rods 7,8,9,10 UO_2
11,12 MOX

Gas flow analysis

Evaluation method

Isotopic release rate from γ-spectrometry

Recoil release to birth rate

$$\frac{R}{B} = \frac{S}{V}\sqrt{D}\sqrt{\frac{\alpha}{\lambda}} + \frac{R}{B}_{recoil}$$

Isotopic birth rate from fission yield calculations

Surface area to volume ratio * square root of diffusion coefficient

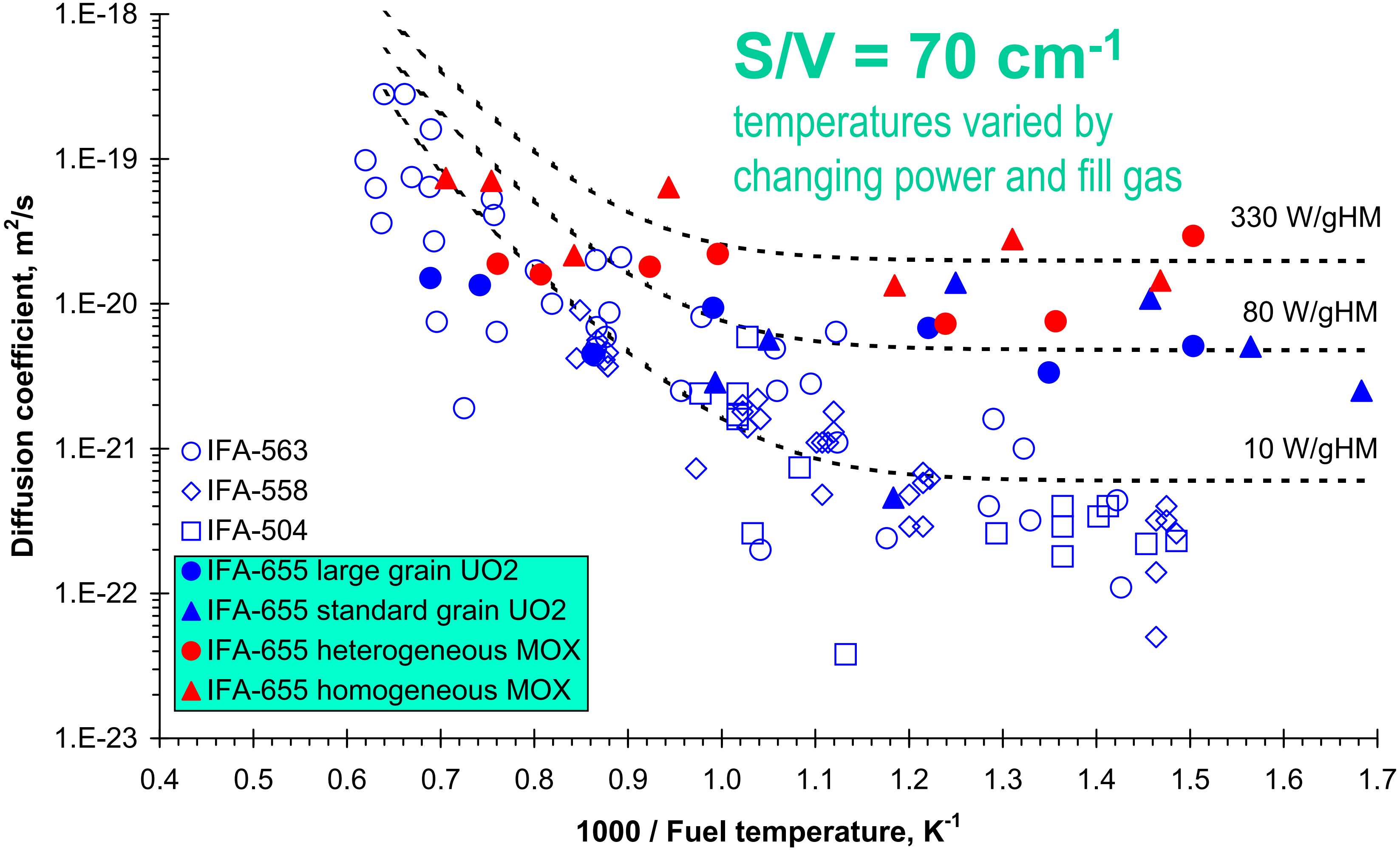
Square root of precursor enhancement factor / decay constant

In start-up diffusivity analysis S/V fixed

In first cycle analysis D fixed

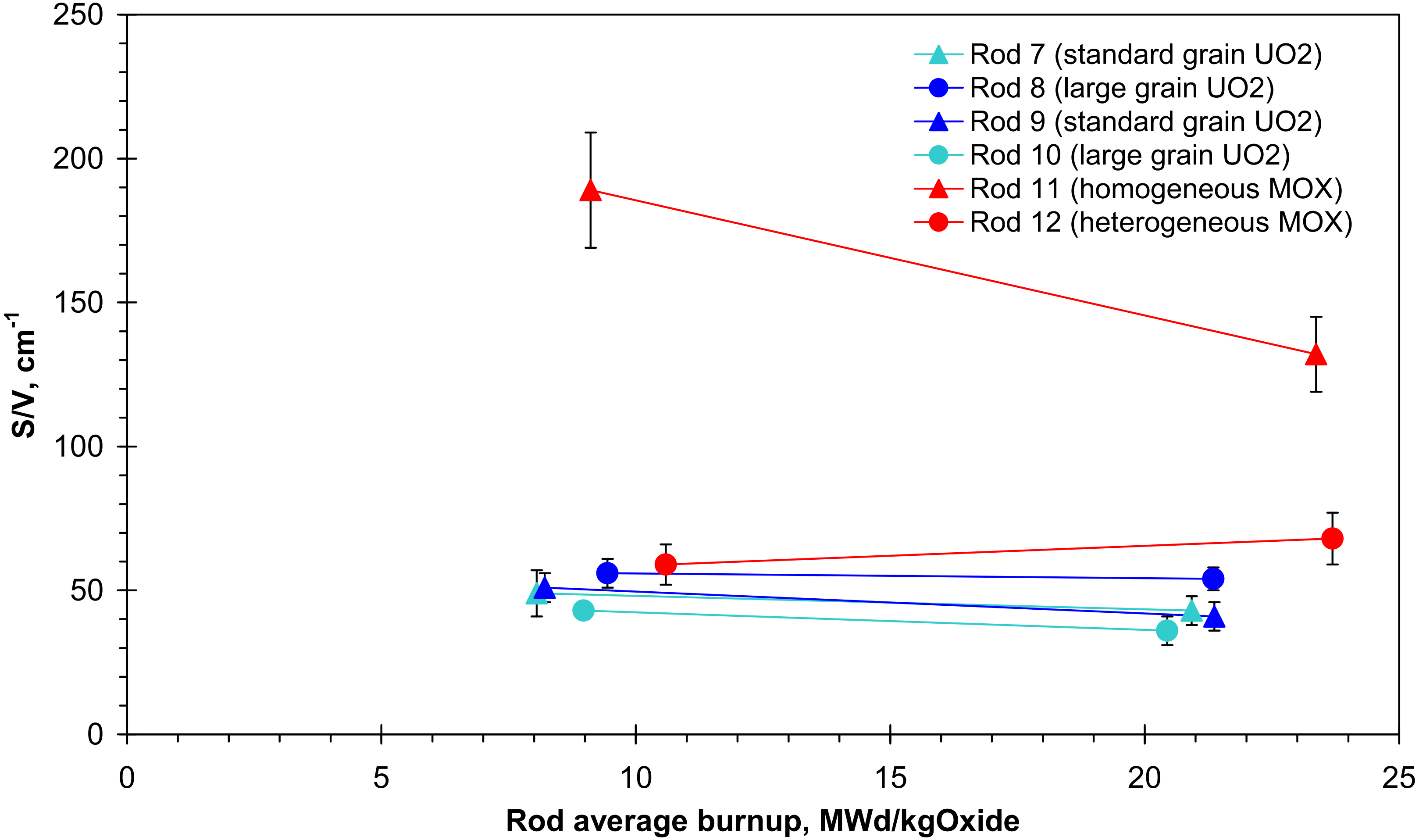
Gas flow analysis

Assume S/V and derive diffusion coefficient

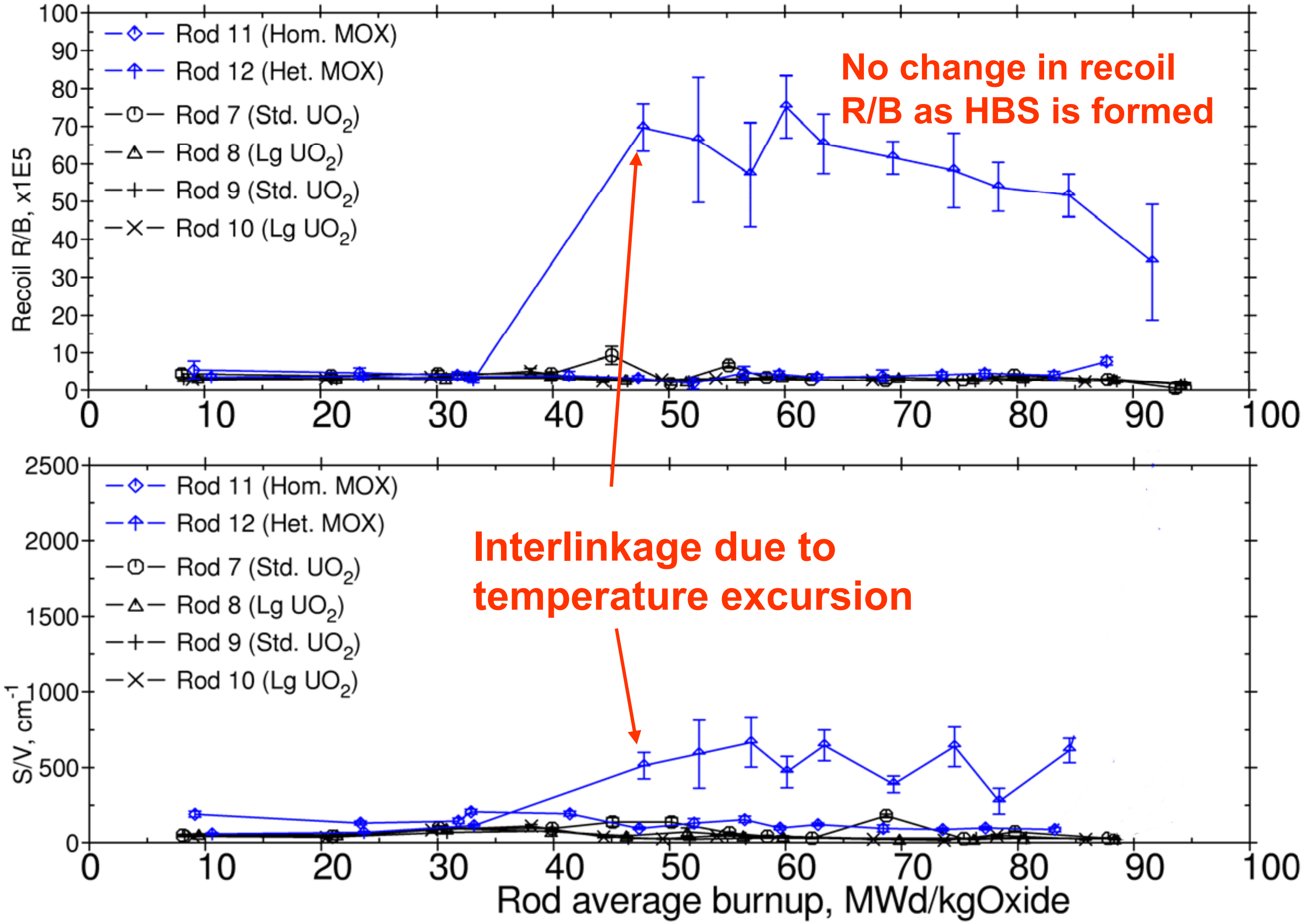


Gas flow analysis

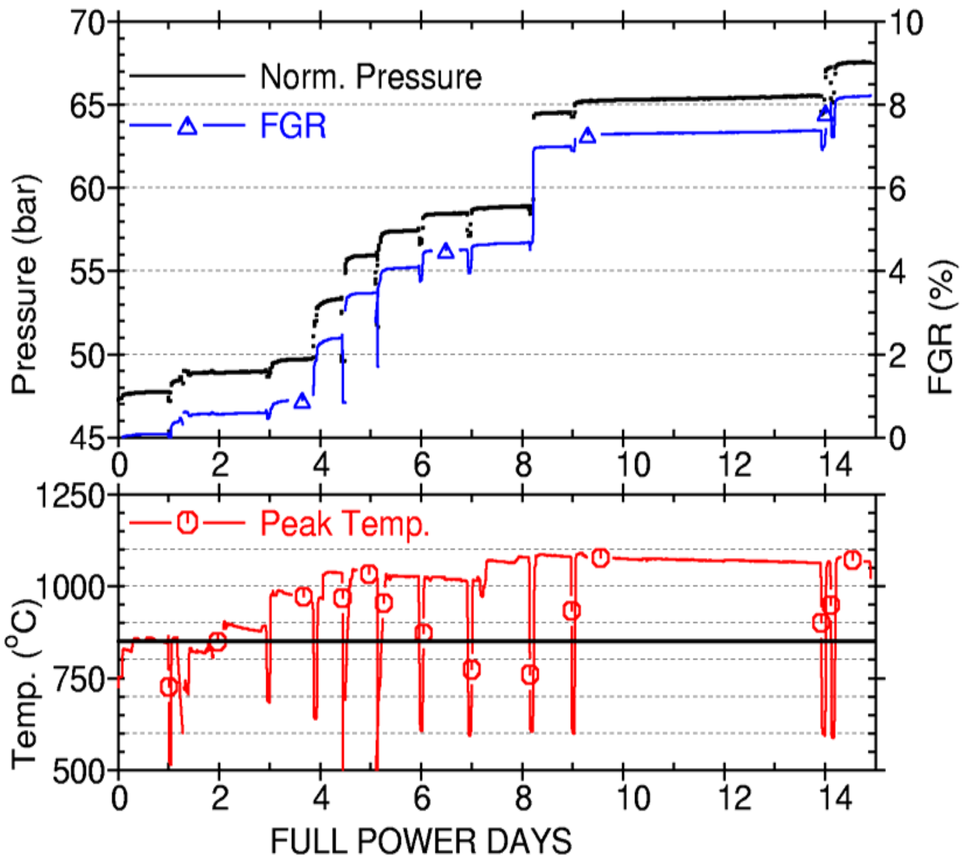
Assume diffusion coefficient and derive S/V



Through-life gas flow measurements



Fission gas release kinetics



Temperature and FGR history

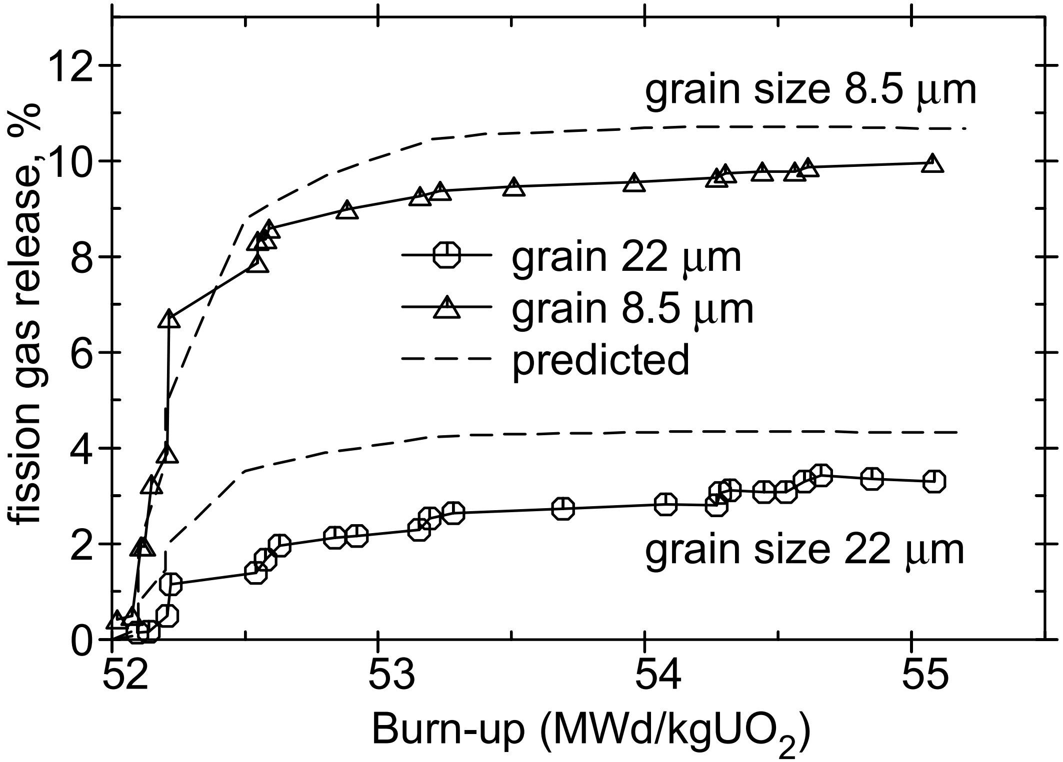
- Steady state power (temperature) for long-term kinetics
- For high burnup fuel, power dips are necessary in order to obtain communication with the plenum (tight fuel column)
- Envelope of release curve indicates diffusion controlled release

Influence of grain size on gas release (I)

- According to diffusion models, an increased grain size will in general result in reduced fission gas release
- The onset of fission gas release occurs at about the same time since the inner (grain) surface varies with $1/R_{grain}$ and thus compensates for fewer atoms arriving at the grain surface
- Larger grains are produced by using additives which may influence (increase) the diffusion coefficient
- Concerning gas release properties, the effect of increased diffusion length (lower FGR) therefore competes with the effect of increased diffusion coefficient (higher FGR)

Influence of grains size on gas release (II)

- Grain size increase is less effective at
  - higher power and FGR >10%
  - high burnup
- Satisfactory prediction with diffusion-based FGR model (Turnbull)

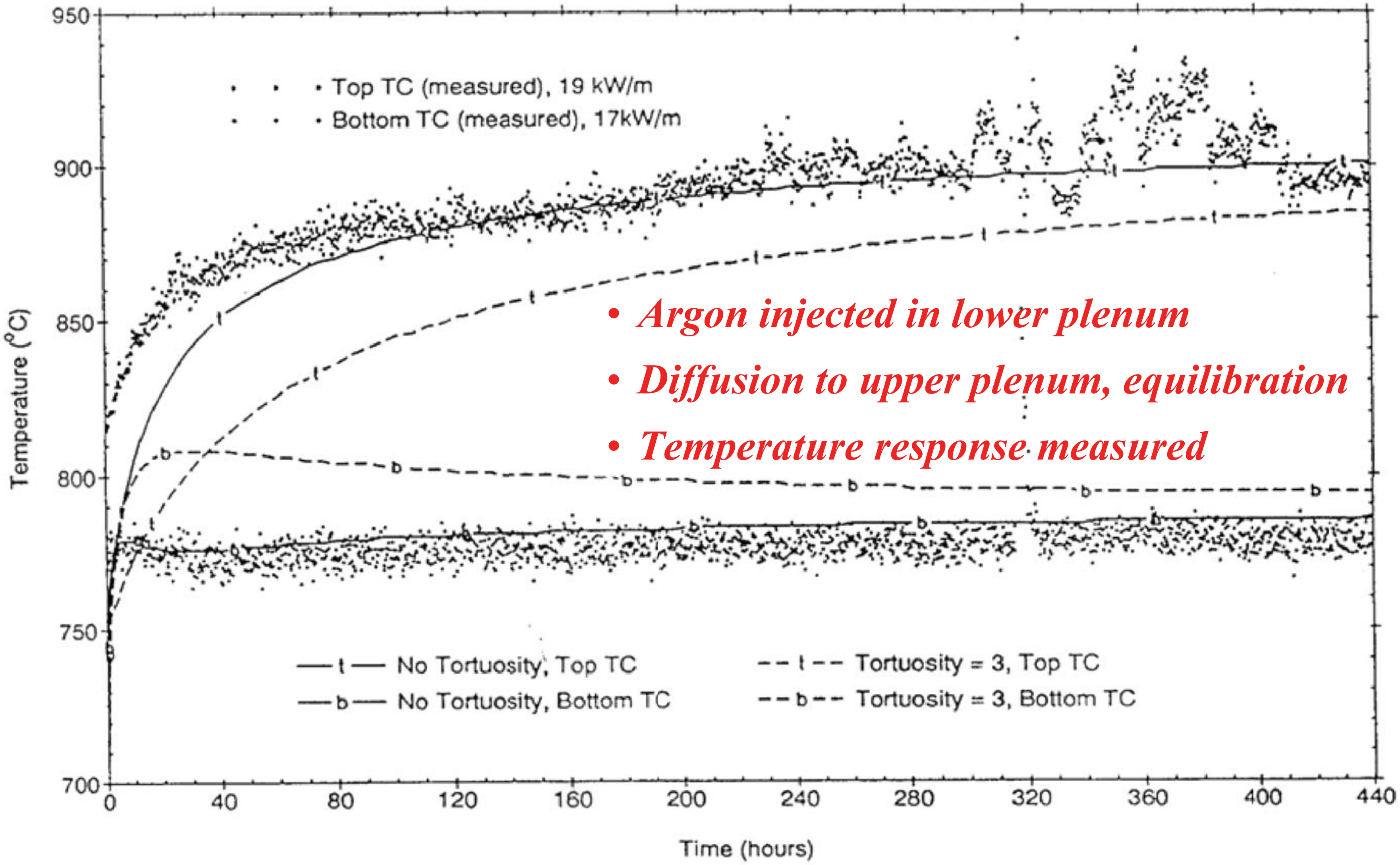


Measured and predicted FGR for UO_2 fuel with different grain sizes

Gas mixing in a fuel rod (I)

- Released fission gas has to move (diffuse) to the rod plenum
- A temporary strong dilution of the fill gas in the fuel column may result …
- ... and cause increased fuel temperatures and positive feedback on fission gas release.
- Gas mixing behaviour and feedback has been investigated in different ways:
  - Injection of argon at one end and tracing the equilibration
  - Cause FGR and monitor the temperature response

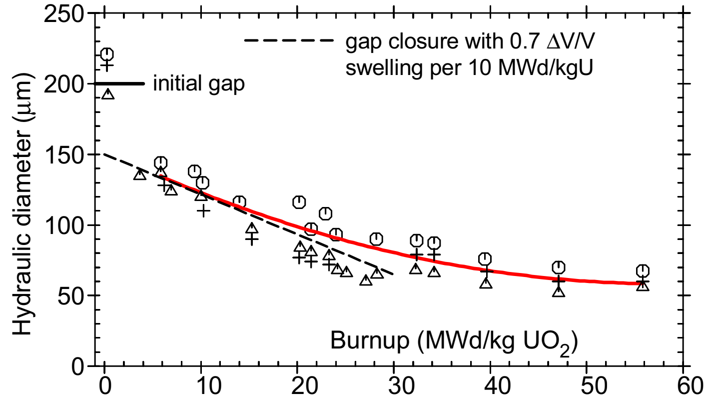
Gas mixing in a fuel rod (II)

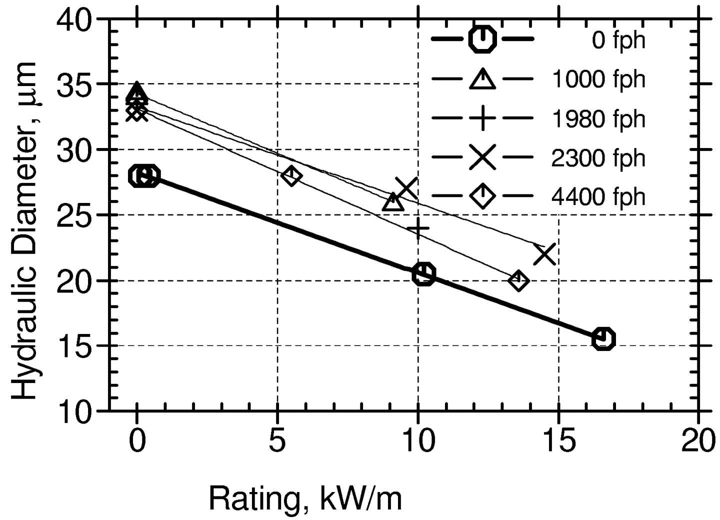


Related: gas flow through the fuel column
- hydraulic diameter measurements -

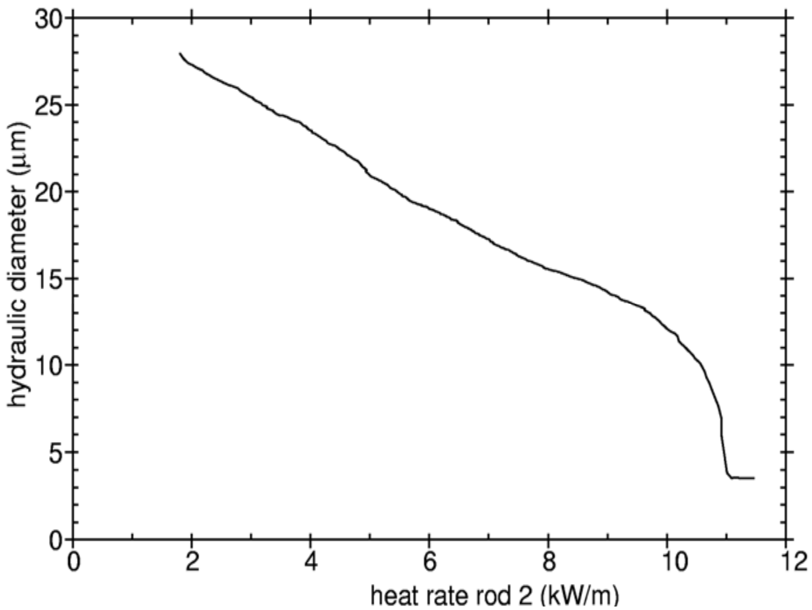
$$D_H = \sqrt[4]{\frac{0.021 \cdot \eta \cdot L \cdot R \cdot T \cdot \dot{n}}{\pi \cdot D \cdot \left(p_1^2 - p_2^2\right)}}$$

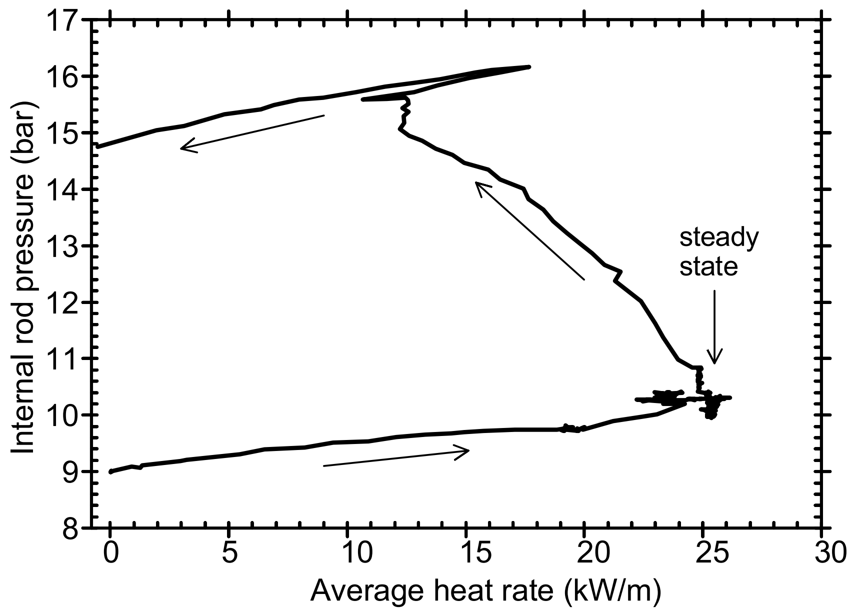
- D = mean fuel diameter
- D_H = hydraulic diameter
- p_1 = gas pressure – supply side (high pressure = rod pressure)
- p_2 = gas pressure – return side (low pressure = rig pressure)
- Ha = Hagen number
- η = dynamic viscosity
- L = flow channel length
- R = universal gas constant
- T = gas temperature





Related: gas flow through the fuel column
- delayed fission gas release -





When the accommodation power level is approached, the hydraulic diameter decreases at a higher rate and the fuel column becomes very tight.

The fuel column becomes permeable after a limited power reduction. The release of the inner overpressure to the fuel rod plenum is detected then.

Delayed fission gas release

- Delayed fission gas release means that the rod pressure, as detected by a pressure transducer located in the rod plenum, increases when power is reduced and a path to the plenum is opened
- The phenomenon is regularly observed when fuel burnup exceeds 40-50 MWd/kg
- It cannot be diffusion driven since the pressure step is also observed after a reactor scram (the fuel is cooled down by several hundred degrees within a few seconds, and any diffusion is effectively stopped)

The END