FAIRFUELS Workshop 2011

KTH University, Stockholm, Sweden, February 9-10, 2011

A simple model of fission gas intra-granular behaviour in UO_2

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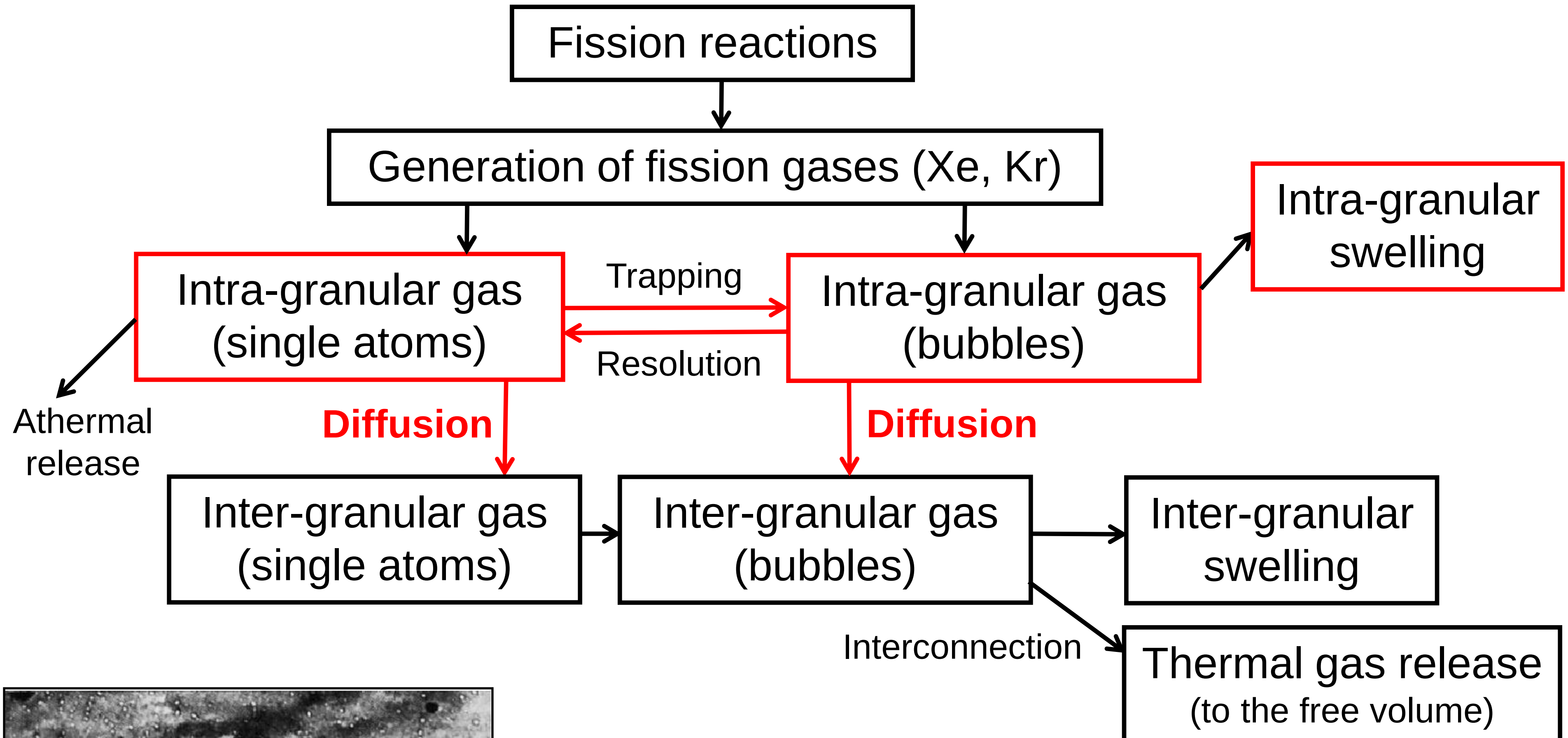
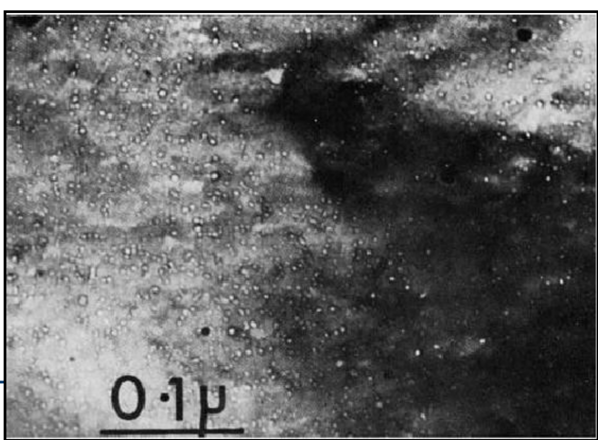
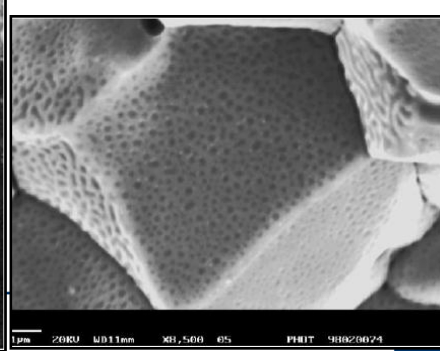
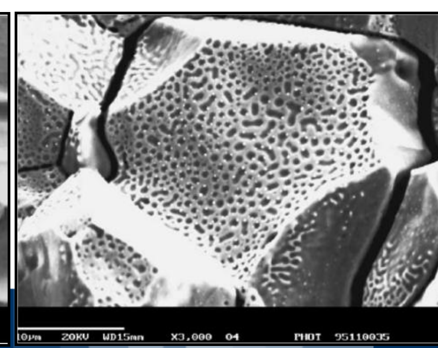
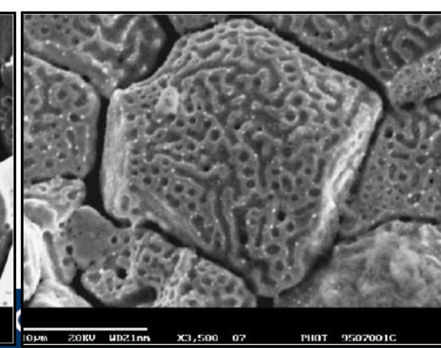
Kinetics of fission gas











Aim of the work

To develop a simple model of the intra-granular behaviour of fission gas in UO_2 fuel, suitable for implementation in an integral fuel performance code (TRANSURANUS).

Considered phenomena:

- Diffusion of fission gas from within the fuel grains to the grain boundaries (intra-granular diffusion)
- Swelling due to growth of fission gas bubbles inside the fuel grains (intra-granular swelling)

Model basic requirements:

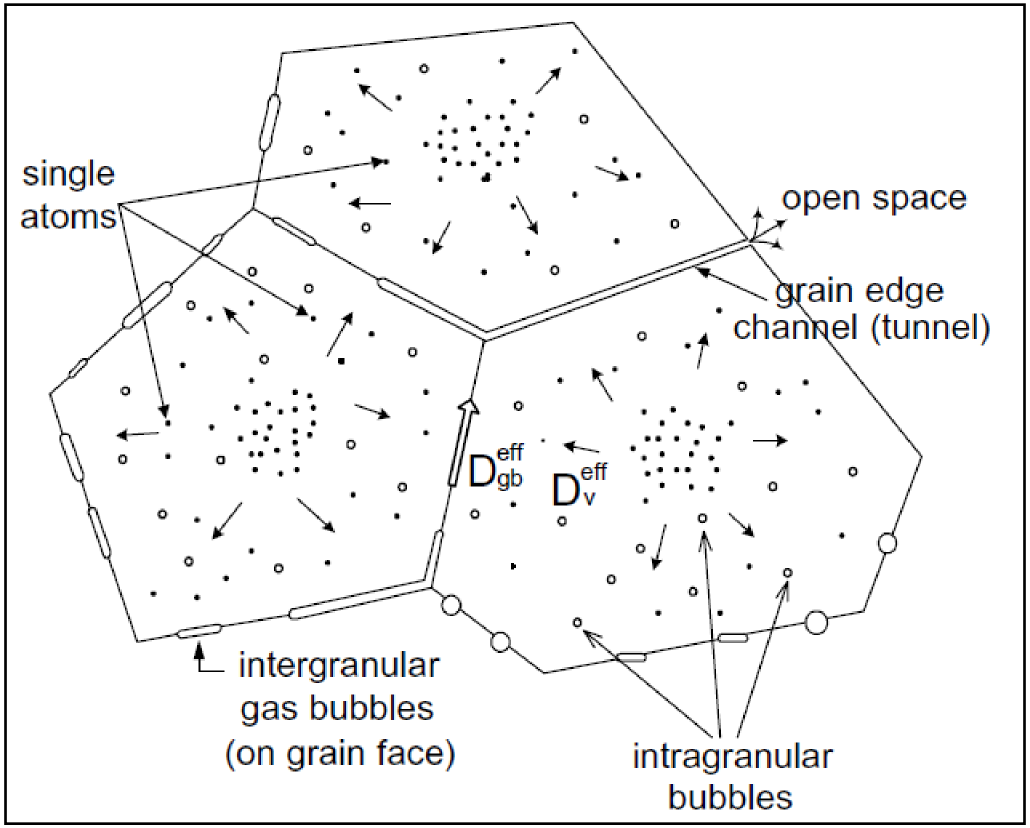
1. To be physically based
2. To be simple (employment in a fuel performance code)
3. To provide a reasonably accurate description of both normal and transient operation reactor conditions

Intra-granular diffusion – introduction

Current treatment of the intra-granular diffusion problem in TRANSURANUS

Premises:

- Intra-granular gas exists in two different phases: single atoms and bubbles
- ***Bubbles are immobile*** → gas transport is solely due to diffusion of single atoms
- Bubbles trap migrating gas atoms
- Gas atoms are knocked back from bubbles by the passage of fission fragments (resolution)



Diffusion – formulation of Speight (1)

Rate equations describing the <u>variation of the gas concentration in a grain</u>:

$$\frac{\partial C_{s.atom}}{\partial t} = D_{s.atom}\nabla^2 C_{s.atom} - gC_{s.atom} + bC_{bubble} + \beta$$

$$\frac{\partial C_{bubble}}{\partial t} = gC_{s.atom} - bC_{bubble}$$

g = trapping parameter [s^{-1}]
b = resolution parameter [s^{-1}]
β = gas generation rate [atoms/(m^3s)]

Addition →

$$\frac{\partial C_{tot}}{\partial t} = D_{s.atom}\nabla^2 C_{s.atom} + \beta$$

<u>Quasi-stationary approach</u>: $C_{s.atom} = \frac{b}{g} C_{bubble}$

Then, the **<u>diffusion equation</u>** is:

Average <u>fraction of gas</u> in single atom phase

$$\boxed{\frac{\partial C_{tot}}{\partial t} = D_{eff,s}\nabla^2 C_{tot} + \beta}$$

$$\boxed{D_{eff,s} = \frac{b}{b+g} D_{s.atom}}$$

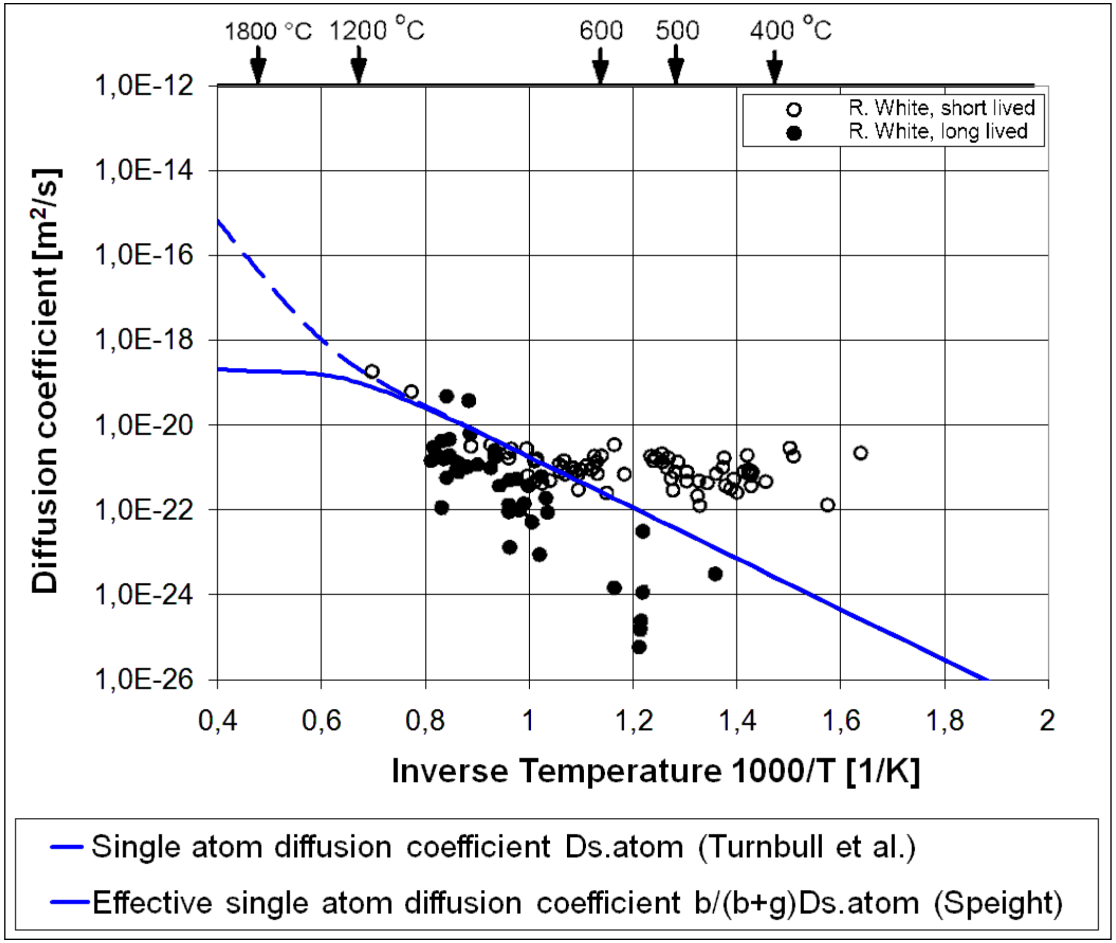
Effective **diffusion coefficient** (**Speight, 1969**) – <u>at present empirical in TRANSURANUS</u>

where $\boxed{C_{tot} = C_{s.atom} + C_{bubble}}$

$D_{eff,s}$ is assumed to be uniform in a grain

Diffusion – formulation of Speight (2)

Preliminary study of $D_{eff,s}$

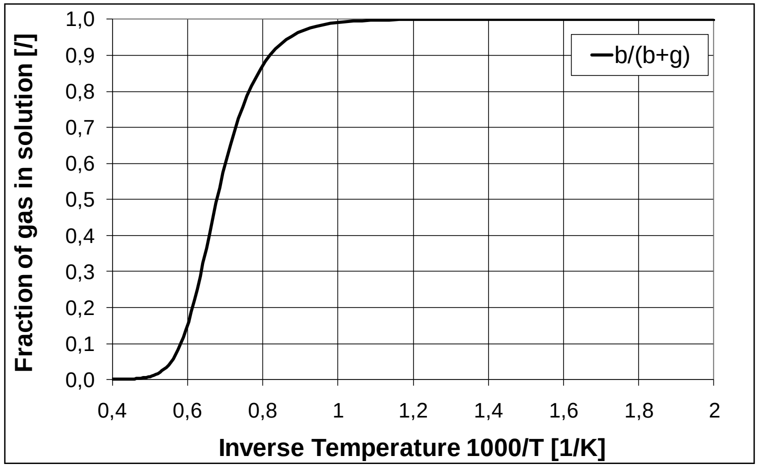


$$D_{eff,s} = \frac{b}{b+g} D_{s.atom}$$

$$b = 3.03\, F\, \pi\, l_f \left(R_{bubble} + Z_0\right)$$

$$g = 4\pi\, D_{s.atom}\; R_{bubble}\; N_{bubble}$$

average



Tendency to underpredict gas transport to the grain boundaries under high temperature and transient conditions

⟺ **Bubble mobility ?**

Diffusion – bubble random motion (1)

By applying the second Fick's law to the concentration of bubbles:

(1) $$\left(\frac{\partial N_{bubble}}{\partial t}\right)_{r.m.} = D_{bubble} \nabla^2 N_{bubble}$$

The concentration of gas in bubble phase is:

(2) $$C_{bubble} = m \cdot N_{bubble}$$

with

(3) $$\frac{\partial m}{\partial t} = 0$$ (quasi-stationary approach)

N_{bubble} = concentration of bubbles [bubbles/m^3]
m = number of gas atoms per bubble [atoms/bubbles]
C_{bubble} = concentration of gas in bubbles [atoms/m^3]
D_{bubble} = bubble diffusion coefficient [m^2/s]

Combining Eqs. (1), (2) and (3), we obtain:

$$\left(\frac{\partial C_{bubble}}{\partial t}\right)_{r.m.} = m\, D_{bubble} \nabla^2 N_{bubble}$$

Hypothesis: m is uniform in a grain (all bubbles contain the same number of atoms) →

$$\left(\frac{\partial C_{bubble}}{\partial t}\right)_{r.m.} = D_{bubble} \nabla^2 C_{bubble}$$

Diffusion – bubble random motion (2)

Rate equations describing the variation of the gas concentration in a grain in presence of bubble random motion:

$$\frac{\partial C_{s.atom}}{\partial t} = D_{s.atom} \nabla^2 C_{s.atom} - g C_{s.atom} + b C_{bubble} + \beta$$

$$\frac{\partial C_{bubble}}{\partial t} = g C_{s.atom} - b C_{bubble} + D_{bubble} \nabla^2 C_{bubble}$$

Bubble diffusion term

Addition →

$$\frac{\partial C_{tot}}{\partial t} = D_{s.atom} \nabla^2 C_{s.atom} + D_{bubble} \nabla^2 C_{bubble} + \beta$$

Again, by applying the quasi-stationary approach the **diffusion equation** takes the form:

$$\boxed{\frac{\partial C_{tot}}{\partial t} = D_{eff} \nabla^2 C_{tot} + \beta}$$

Average fraction of gas as single atoms

$$\boxed{D_{eff} = \frac{b}{b+g} D_{s.atom} + \frac{g}{b+g} D_{bubble}}$$

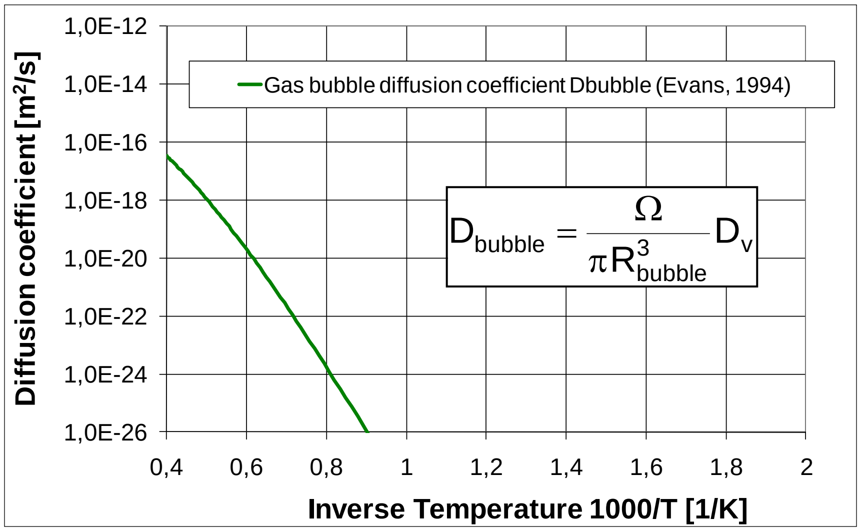
Average fraction of gas in bubbles

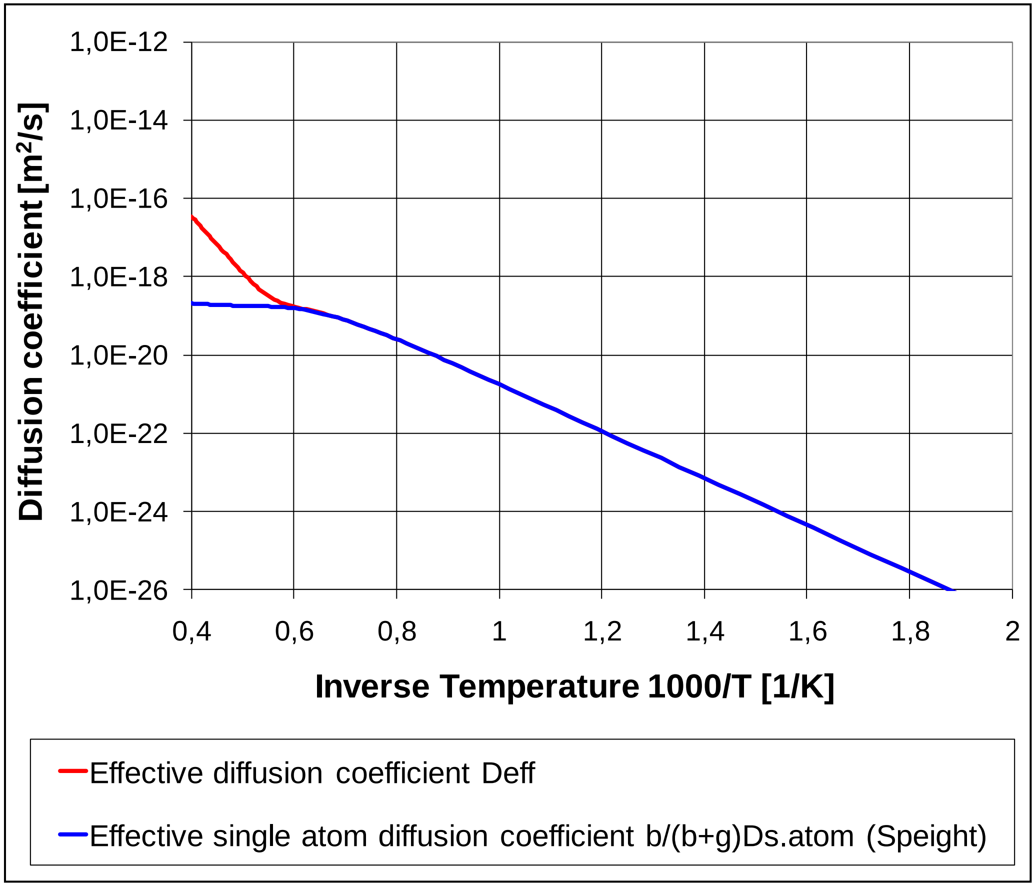
No modifications in the form of the equation

Diffusion – bubble random motion (3)

Preliminary study of D_{eff}

$$D_{eff} = \underbrace{\frac{b}{b+g} D_{s.atom}}_{\text{Single atom diffusion (Speight)}} + \underbrace{\frac{g}{b+g} D_{bubble}}_{\text{bubble diffusion}}$$





The contribution of bubble motion to the effective diffusion coefficient is significant above ~1500 °C (consistent with observations of bubble motion by Baker, 1977)

Diffusion – power transients (1)

Enhanced gas transport to the grain boundaries is observed during transients (bubble directed motion in a temperature/vacancy gradient).

Empirical approaches have been used to consider an additional release of gas during transients (Koo et al, 1999).

Bubble velocity up to a temperature/vacancy gradient (Nichols, 1969; Evans, 1994):

$$\boxed{v_{bubble} = \frac{D_v Q_v \Delta T}{kT^2}}$$

Consider bubble directed motion towards the grain boundary starting at a distance **r** from the boundary. Approximating ΔT = T/r (Evans, 1994), we obtain:

$$\boxed{v_{bubble} = \frac{D_v Q_v}{kT\,r}}$$

Attempt to represent the effect of directed diffusion by means of the random diffusion equation:

$$\boxed{t_{directed} = \frac{r^2 kT}{D_v Q_v}} \;=\; \boxed{t_{r.m.s.} = \frac{r^2}{2D_{bubble,tr}}}$$

$$\rightarrow \boxed{D_{bubble,tr} = \frac{Q_v}{2kT} D_v} \quad \text{(Evans, 1994)}$$

We obtain an "equivalent" diffusion coefficient to be adopted during transients.

v_{bubble} = bubble velocity [m/s]
D_v = volume self-diffusion coefficient [m^2/s]
Q_v = heat of transport of vacancies [eV]
k = Boltzmann constant [eV/K]
T = temperature [K]
r = starting distance from the grain boundary [m]

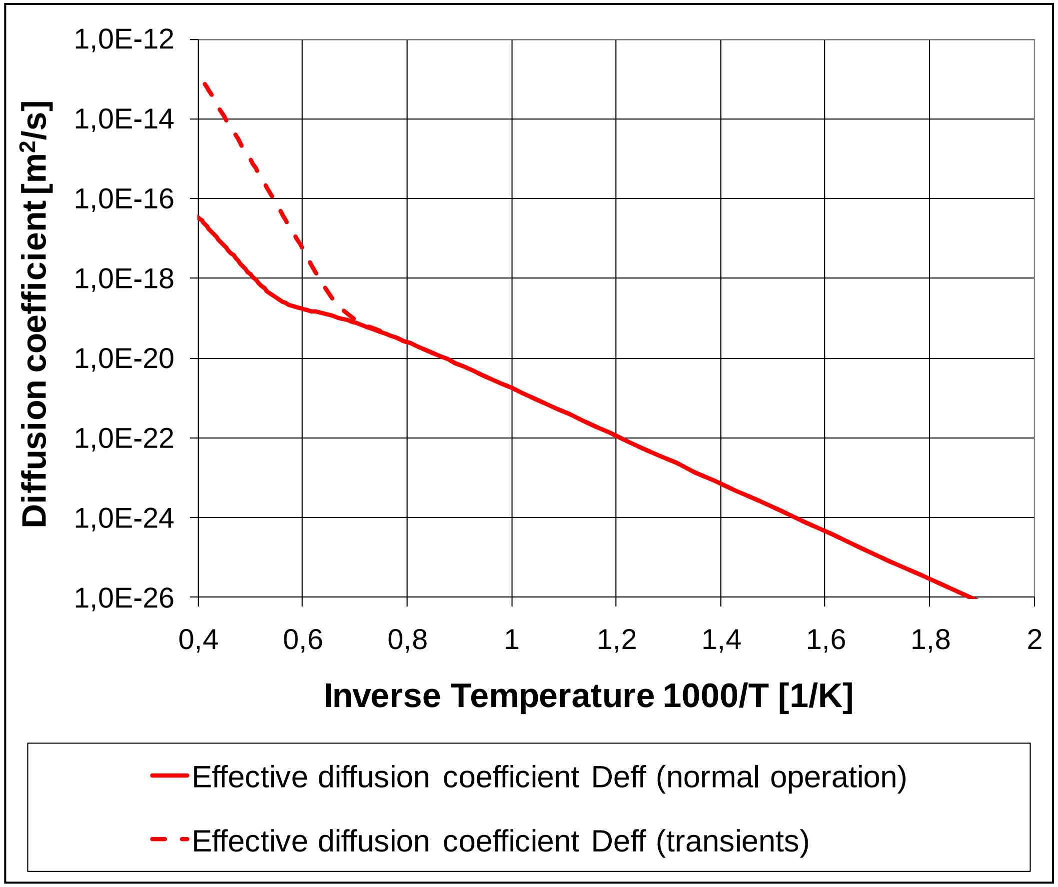
Diffusion – power transients (2)

Preliminary study of $D_{eff,tr}$

$$D_{eff,tr} = \underbrace{\frac{b}{b+g} D_{s.atom}}_{\text{Single atom diffusion (Speight)}} + \underbrace{\frac{g}{b+g} D_{bubble,tr}}_{\text{bubble diffusion}}$$

$$\frac{\partial C_{tot}}{\partial t} = D_{eff,tr} \nabla^2 C_{tot} + \beta$$

In this simplified approach, the enhanced gas transport during power transients is simulated by modification of the effective diffusion coefficient (bubble diffusion term)



The concept of representing the effect of directed bubble motion by an "equivalent" random diffusion equation is intended as a practical approach (easy implementation) for a first estimation of the enhanced gas transport during power transients

Swelling – model assumptions

I. All bubbles have the same size and the same (spherical) shape. It follows that the fractional volume of intra-granular swelling is given by:

$$\boxed{\frac{\Delta V}{V} = N_{bubble}\left(\frac{4}{3}\pi R_{bubble}^3\right)}$$

II. The concentration of gas bubbles N_{bubble} remains constant in time ($N_{bubble}=5\cdot10^{23}$ bubbles/m^3)

III. The bubbles are in mechanical equilibrium with the bulk solid. If the effect of hydrostatic stress is neglected (nm-size bubbles), the gas pressure p in the bubble is calculated as:

$$\boxed{p = \frac{2\gamma}{R_{bubble}}}$$

IV. The gas in the bubbles obeys the van der Waals equation of state:

$$\boxed{p\left(V_{bubble} - \frac{bm}{N_{AV}}\right) = kmT}$$

where V_{bubble} is the bubble volume [m^3] and m is the number of gas atoms per bubble [atoms/bubble]

Swelling – calculation scheme

Calculation at time step t:

Diffusion equation

$$\boxed{C_{tot}(t) = C_{tot}(t-dt) + dC_{tot}(t)}$$

Total concentration of intra-granular gas [atoms/m³]

$$\boxed{C_{bubble}(t) = \frac{g}{b+g} C_{tot}(t)}$$

Concentration of gas retained in the bubbles [atoms/m³]

$$\boxed{m(t) = \frac{C_{bubble}(t)}{N_{bubble}}}$$

Number of gas atoms per bubble [atoms/bubble]

Equation for the bubble radius [m]:

$$\boxed{8\pi\gamma \cdot R^3_{bubble}(t) - 3k \cdot m(t) \cdot T(t) \cdot R_{bubble}(t) - \frac{6\gamma b}{N_{AV}} \cdot m(t) = 0}$$

Intra-granular swelling (increment of fractional increase in fuel volume) [/]:

$$\boxed{\frac{\Delta V}{V}(t) = N_{bubble} \cdot \frac{4}{3}\pi \cdot R^3_{bubble}(t)}$$

Conclusions

- A simple but physically based model of fission gas intra-granular behaviour is proposed, which is intended for adoption in fuel performance codes (TRANSURANUS).
- For the treatment of the fission gas intra-granular diffusion, the concept of an effective diffusion coefficient (Speight) is maintained.
- A simple extension of the Speight´s formulation is proposed, which takes into account the contribution of bubble random motion to the total gas diffusion.
- A specific expression for the bubble diffusion coefficient is proposed for the simulation of power transients (bubble directed motion).
- The contribution of bubble motion may explain the tendency of the Speight´s formulation to underpredict gas transport to the grain boundaries under high temperature and transient conditions.
- The incorporation in the TRANSURANUS code of the above concepts, as well as of a simple model for the intra-granular swelling, is underway.

Main references

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Thank you for your attention