A simple model of fission gas intra-granular behaviour in UO$_2$

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Kinetics of fission gas

Fission reactions

Generation of fission gases (Xe, Kr)

Intra-granular gas (single atoms)

Diffusion

Inter-granular gas (single atoms)

Intra-granular gas (bubbles)

Trapping

Resolution

Diffusion

Inter-granular gas (bubbles)

Inter-granular swelling

Thermal gas release (to the free volume)

Interconnection

Athermal release

0.1 μ
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 variation of the gas concentration in a grain:

\[
\frac{\partial C_{s,\text{atom}}}{\partial t} = D_{s,\text{atom}} \nabla^2 C_{s,\text{atom}} - g C_{s,\text{atom}} + b C_{\text{bubble}} + \beta
\]

\[
\frac{\partial C_{\text{bubble}}}{\partial t} = g C_{s,\text{atom}} - b C_{\text{bubble}}
\]

Addition →

\[
\frac{\partial C_{\text{tot}}}{\partial t} = D_{s,\text{atom}} \nabla^2 C_{s,\text{atom}} + \beta
\]

Quasi-stationary approach: \[ C_{s,\text{atom}} = \frac{b}{g} C_{\text{bubble}} \]

Then, the diffusion equation is:

\[
\frac{\partial C_{\text{tot}}}{\partial t} = D_{\text{eff},s} \nabla^2 C_{\text{tot}} + \beta
\]

where \[ C_{\text{tot}} = C_{s,\text{atom}} + C_{\text{bubble}} \]

**Effective diffusion coefficient** (Speight, 1969) – at present empirical in TRANSURANUS

\[ D_{\text{eff},s} = \frac{b}{b + g} D_{s,\text{atom}} \]

\[ D_{\text{eff},s} \] is assumed to be uniform in a grain

**g** = trapping parameter \([s^{-1}]\)

**b** = resolution parameter \([s^{-1}]\)

**\beta** = gas generation rate \([\text{atoms/(m}^3\text{s)}]\)
Tendency to **underpredict** gas transport to the grain boundaries under **high temperature and transient conditions**
Diffusion – bubble random motion (1)

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

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

The concentration of gas in bubble phase is:

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

with

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

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

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

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

\( \left( \frac{\partial C_{\text{bubble}}}{\partial t} \right)_{r.m.} = D_{\text{bubble}} \nabla^2 C_{\text{bubble}} \)
Diffusion – bubble random motion (2)

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

\[ \frac{\partial C_{\text{atom}}}{\partial t} = D_{\text{atom}} \nabla^2 C_{\text{atom}} - g C_{\text{atom}} + b C_{\text{bubble}} + \beta \]

\[ \frac{\partial C_{\text{bubble}}}{\partial t} = g C_{\text{atom}} - b C_{\text{bubble}} + D_{\text{bubble}} \nabla^2 C_{\text{bubble}} \]

 Addition →

\[ \frac{\partial C_{\text{tot}}}{\partial t} = D_{\text{atom}} \nabla^2 C_{\text{atom}} + D_{\text{bubble}} \nabla^2 C_{\text{bubble}} + \beta \]

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

\[ \frac{\partial C_{\text{tot}}}{\partial t} = D_{\text{eff}} \nabla^2 C_{\text{tot}} + \beta \]

\[ D_{\text{eff}} = \left( \frac{b}{b+g} \right) D_{\text{atom}} + \left( \frac{g}{b+g} \right) D_{\text{bubble}} \]

No modifications in the form of the equation
Diffusion – bubble random motion (3)

Preliminary study of $D_{\text{eff}}$

$$D_{\text{eff}} = \frac{b}{b+g} D_{\text{s.atm}} + \frac{g}{b+g} D_{\text{bubble}}$$

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

\[
V_{\text{bubble}} = \frac{D_v Q_v \Delta T}{kT^2}
\]

Consider bubble directed motion towards the grain boundary starting at a distance \( r \) from the boundary. Approximating \( \Delta T = \frac{T}{r} \) (Evans, 1994), we obtain:

\[
V_{\text{bubble}} = \frac{D_v Q_v}{kT r}
\]

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

\[
t_{\text{directed}} = \frac{r^2 kT}{D_v Q_v}
= \frac{r^2}{2D_{\text{bubble,tr}}}
\]

\[
D_{\text{bubble,tr}} = \frac{Q_v}{2kT} D_v
\]

We obtain an “equivalent” diffusion coefficient to be adopted during transients.
Diffusion – power transients (2)

Preliminary study of $D_{\text{eff, tr}}$

\[
D_{\text{eff, tr}} = \frac{b}{b + g} D_{\text{s, atom}} + \frac{g}{b + g} D_{\text{bubble, tr}}
\]

Single atom diffusion (Speight)  
bubble diffusion

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

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:

\[
\frac{\Delta V}{V} = N_{\text{bubble}} \left(\frac{4}{3} \pi R_{\text{bubble}}^3\right)
\]

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

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:

\[
p = \frac{2\gamma}{R_{\text{bubble}}}
\]

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

\[
p \left(V_{\text{bubble}} - \frac{bm}{N_{AV}}\right) = kMT
\]

where \(V_{\text{bubble}}\) is the bubble volume \([m^3]\) and \(m\) is the number of gas atoms per bubble \([\text{atoms/bubble}]\)
Swelling – calculation scheme

Calculation at time step $t$:

\[ C_{\text{tot}}(t) = C_{\text{tot}}(t - dt) + dC_{\text{tot}}(t) \]

**Total concentration of intra-granular gas**

\[ \text{[atoms/m}^3\text{]} \]

\[ C_{\text{bubble}}(t) = \frac{g}{b + g} C_{\text{tot}}(t) \]

**Concentration of gas retained in the bubbles**

\[ \text{[atoms/m}^3\text{]} \]

\[ m(t) = \frac{C_{\text{bubble}}(t)}{N_{\text{bubble}}} \]

**Number of gas atoms per bubble**

\[ \text{[atoms/bubble]} \]

Equation for the bubble radius [m]:

\[ 8\pi \gamma \cdot R_{\text{bubble}}^3(t) - 3k \cdot m(t) \cdot T(t) \cdot R_{\text{bubble}}(t) - \frac{6\gamma b}{N_{AV}} \cdot m(t) = 0 \]

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

\[ \frac{\Delta V}{V}(t) = N_{\text{bubble}} \cdot \frac{4}{3} \pi \cdot R_{\text{bubble}}^3(t) \]
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


Thank you for your attention