Simulation of grain morphology evolution during sintering

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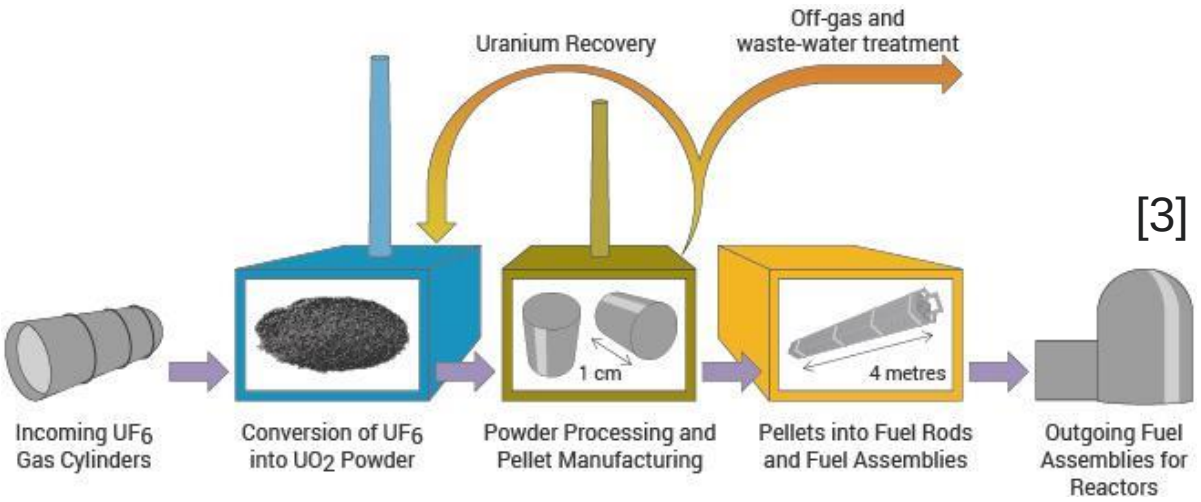
Gaelle Raveu, Maelys Gauthé – Framatome

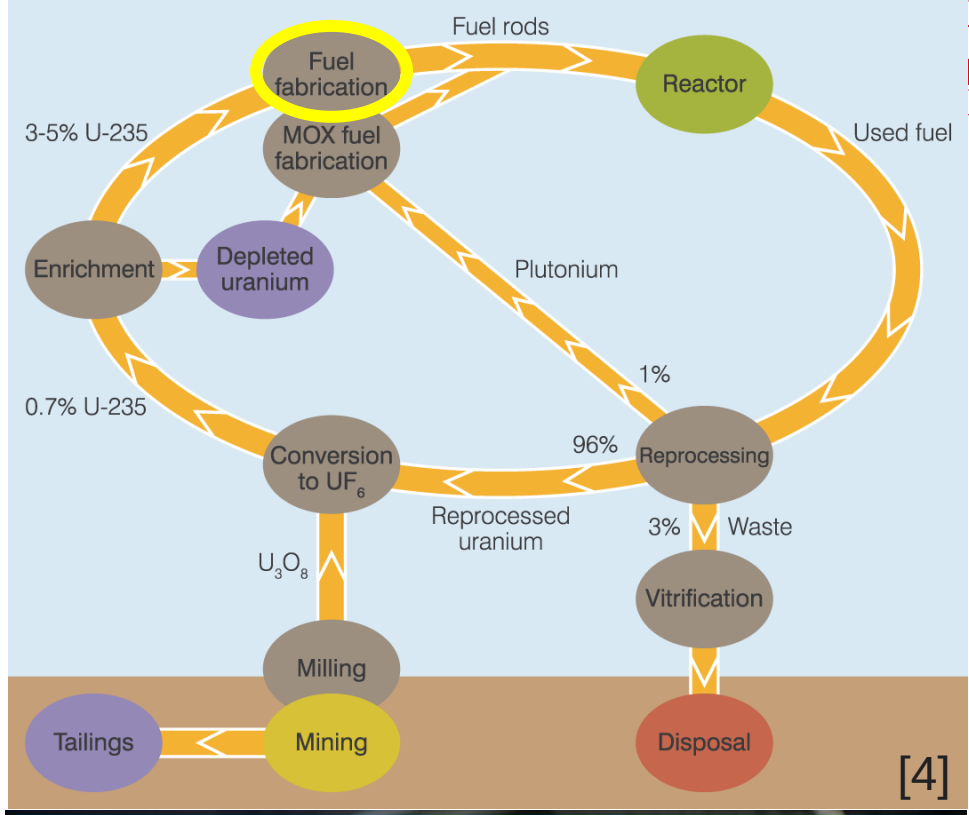
1. Introduction

Context : Nuclear Fuel

- In France 65% of the electricity come from the nuclear industry.
- 3rd rank in term of production in the world (380.5 TW/h or 13.5%) after the USA and China
- 2nd in installed power (63 GW or 16.8%) after the USA
- 57 Pressurized Water Reactors across the country [1,2]
- The fuel is constituted of cylinders pellets of sintered UO_2 and $(U, Pu)O_2$ (about ~13mm in height, ~8mm width and ~7.5g)
- Pellets are put in 4m long fuel rods organized in assemblies



[3]



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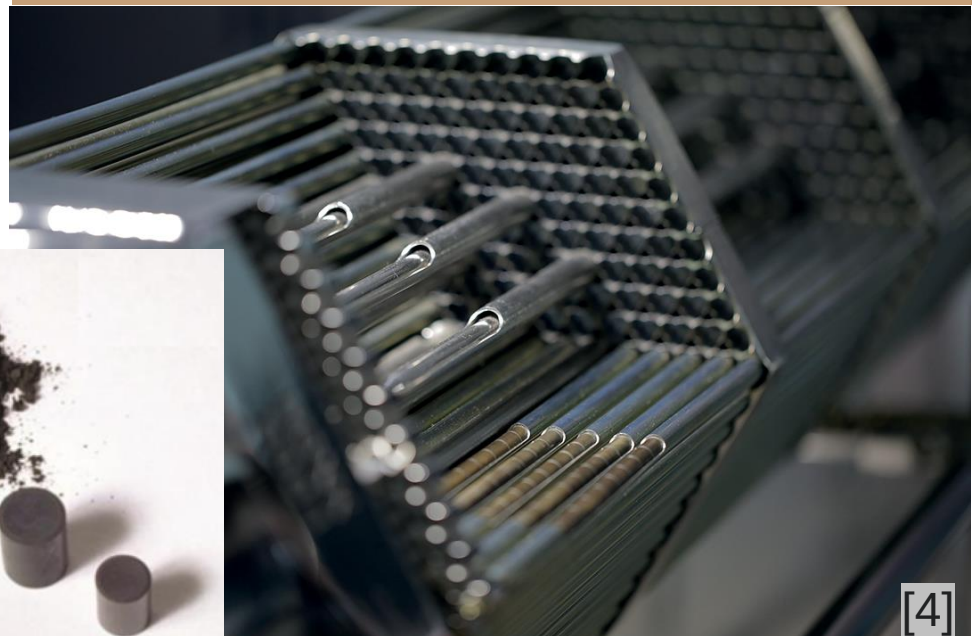
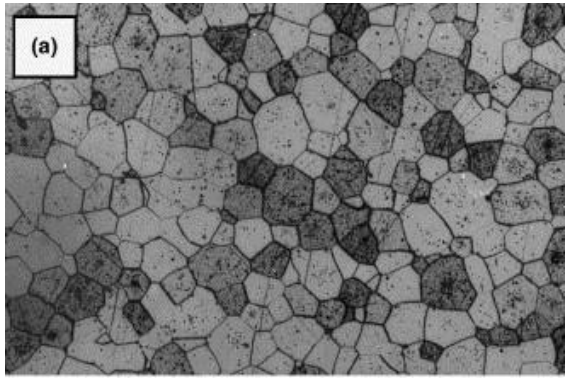


[4]

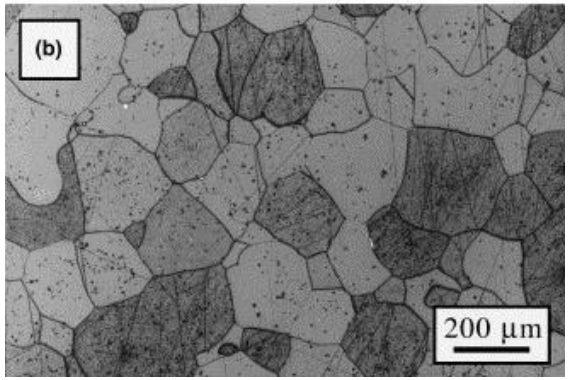
[1] 2025 Statistical Review of World Energy, Energy Institute
[2] 2025 World Statistic International Atomic Energy Agency
[3] Framatome Romans R&D presentation
[4] World nuclear association https://world-nuclear.org/information-library

Accident Tolerant Fuels

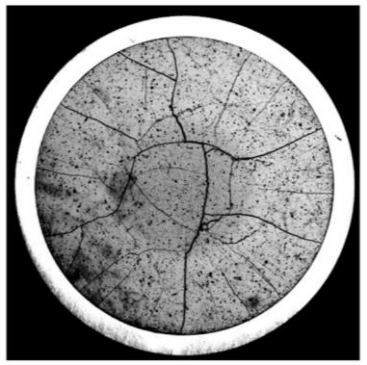
- Current developments in nuclear fuels are aimed at **Accident Tolerant Fuel** technologies, to enhance safety and performance.
- One of them is **Chromia-doped** fuel pellets, to retain fission gases better and improve pellet-cladding interaction.



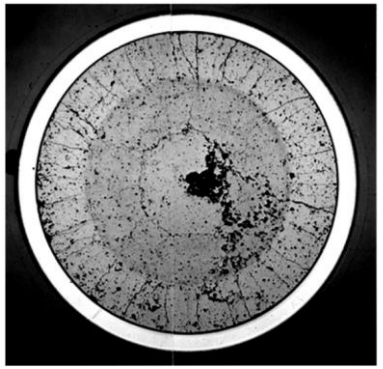
UO_2+0.1 wt% Cr_2O_3



UO_2+0.25 wt% Cr_2O_3



Pure UO_2



Cr_2O_3 doped UO_2

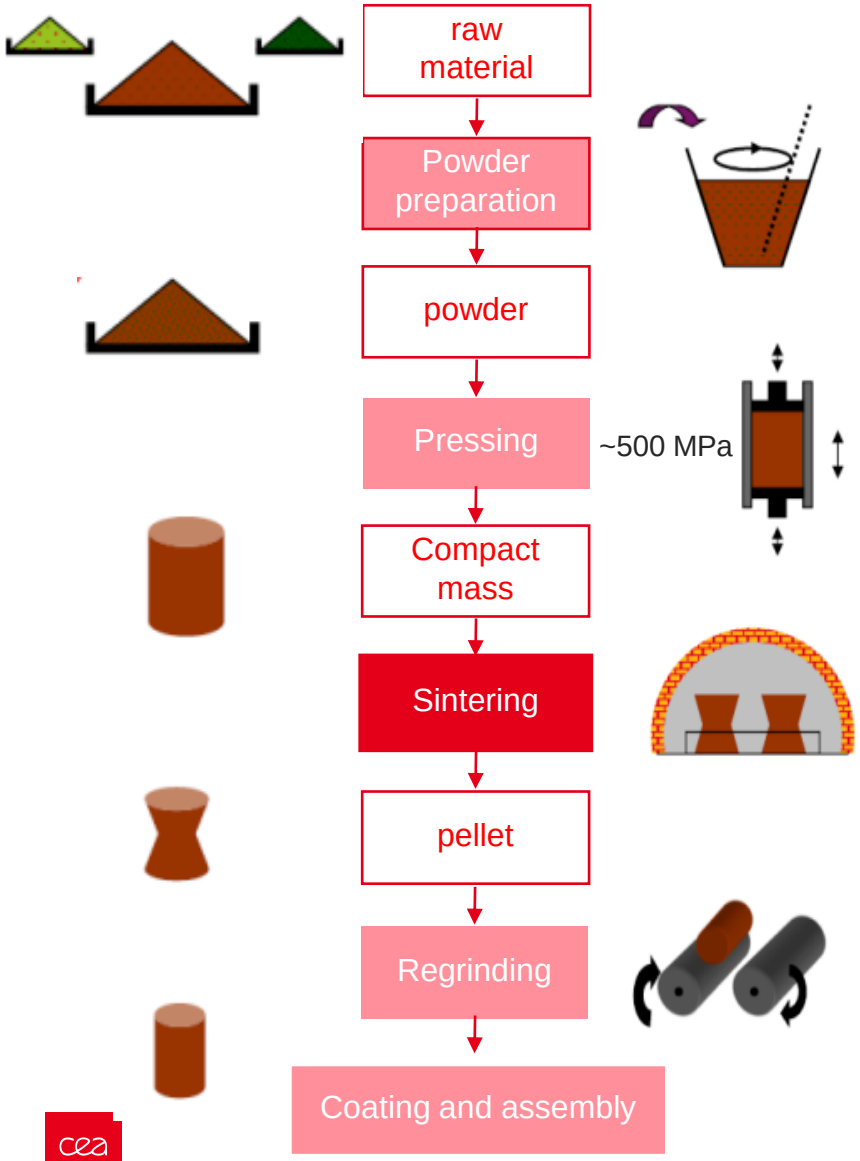
Bigger grains, which improves fission gas retention [1]

More radial cracks in the periphery of doped UO_2 → relieve of stress [2]

[1] L. Bourgeois and al., Factors governing microstructure development of Cr_2O_3-doped UO_2 during sintering, Journal of Nuclear Materials, 2001

[2] Nonon et al., PCI Behaviour of Chromium Oxide-Doped Fuel, OECD, Nuclear. Energy Agency, 2005

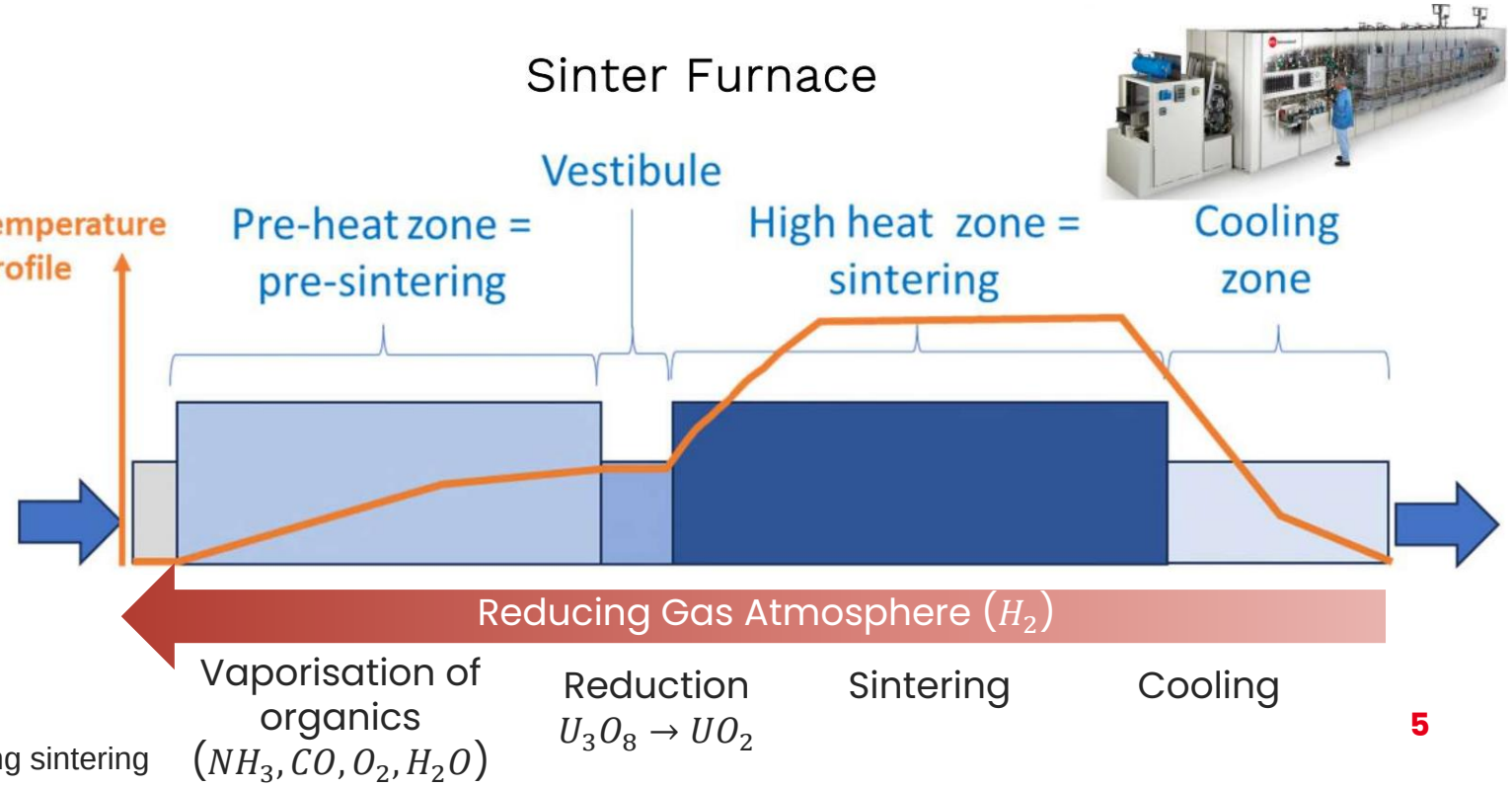
Context : Manufacturing of fuel pellets



Pellet manufacturing is one of the last steps of the fuel production.

The entire heating cycle takes around 24 hours, in a furnace at around 1700°C with reducing atmosphere (H_2)

Framatome Romans R&D presentation



CEA simulation and multiscale approach to sintering

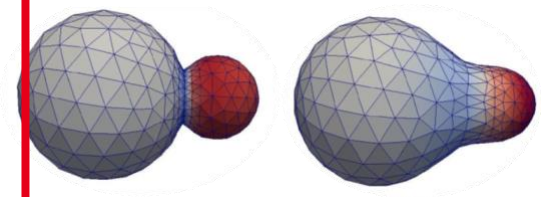
Numerical

Microscopic | Mesoscopic | Macroscopic | Furnace…

Crystallites/grains

- Modelling grain growth
- Material diffusion

Thesis S. Martin (2014)

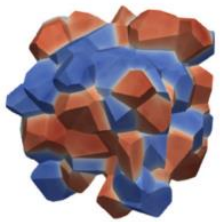


Salammbo

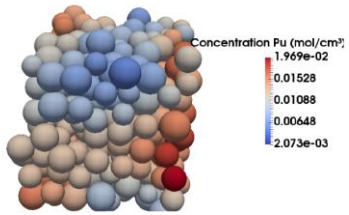
RVE

- Homogenization
- Microstructural changes

Thesis A. Ouhammou (2025)



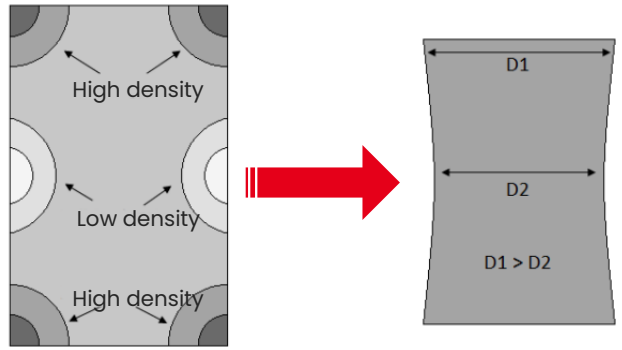
Thesis N. Dempowo (2022)



Model feeding

Pellet

- Defect formation during sintering
- Impact of heterogeneities
- Oxygen transport



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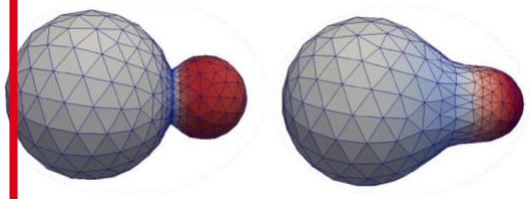

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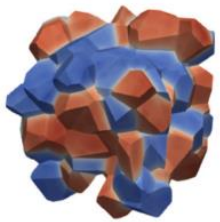

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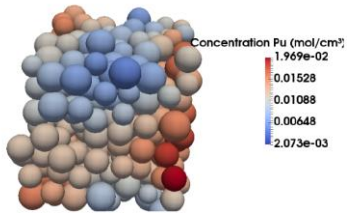

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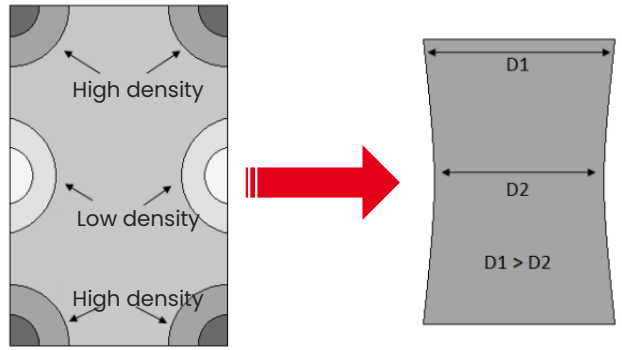
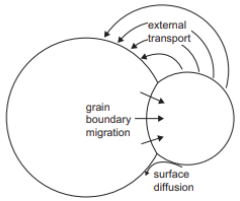
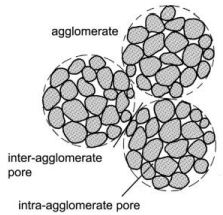


Ph.D. topic : simulation from 2 crystallites to RVE



2 grains → Multi-grain → RVE



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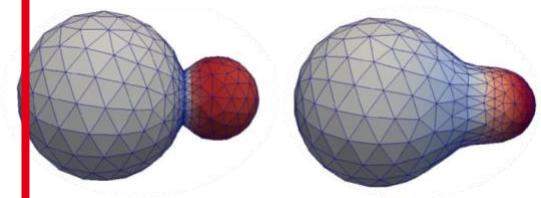

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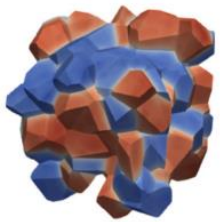

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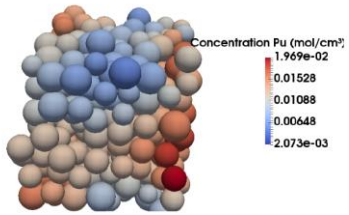

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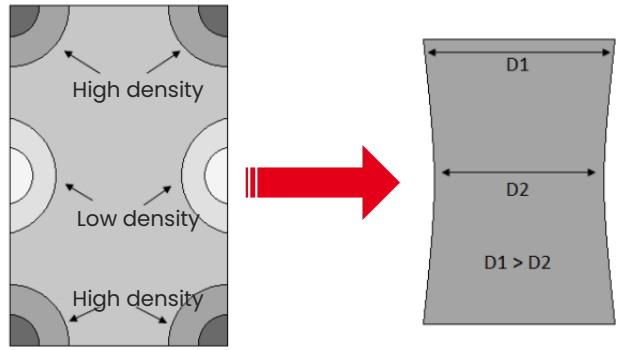
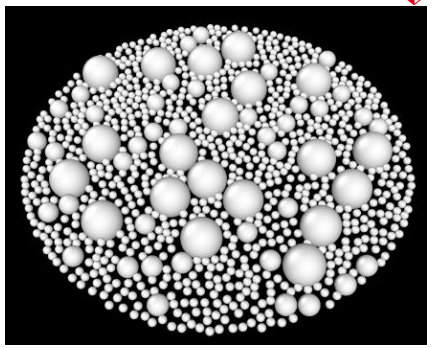


***Ph.D. topic :* simulation from 2 crystallites to RVE**

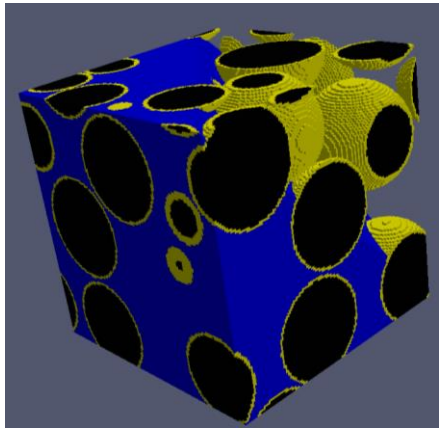
Presentation topic :
working on two interface evolution methods: phase field and adaptive meshes

2. Models

Hypothesis



Merope: A microstructure generator for simulation of heterogeneous materials, M. Josien, 2024

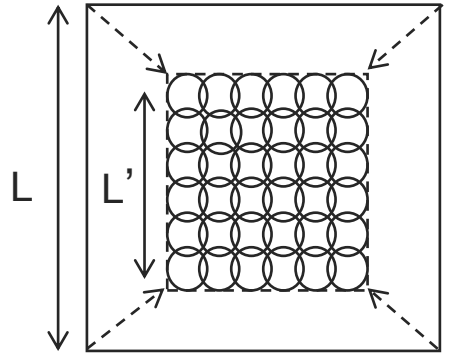


1. Initial time :
   - Temperature : 800–900 degrees
   - Chromium : already solubilized in some grains
   - Reduction to UO_2 at low temperature completed
   - Generation of an initial microstructure using a set of spheres

2. Overall evolution in the RVE :
   - O/M and temperature evolve uniformly across the domain
   - Effect of the dopant, modification of some/every diffusion coefficients

Considerations about the study domain (RVE) :

- Aimed to solve several thousand of Degrees of Freedom
- Evolving boundary conditions due to shrinkage



Multi-physics problem

Mechanical problem :

- Static equilibrium :

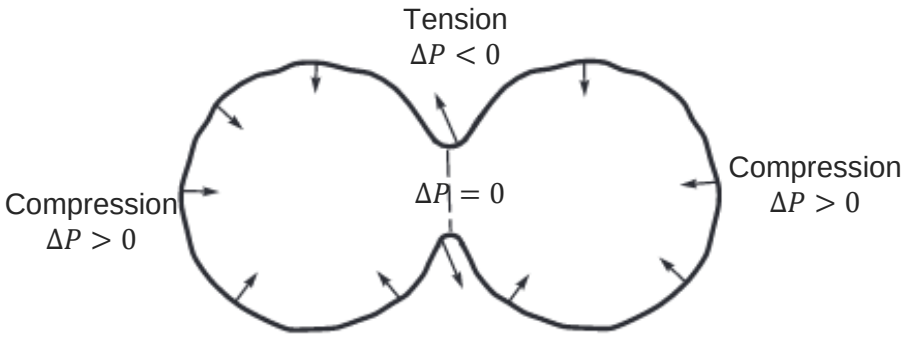
$$div(\bar{\bar{\sigma}}) = 0$$

- Infinitesimal strain :

$$\bar{\bar{\varepsilon}} = \frac{1}{2}(\nabla\vec{u} + {}^{t}\nabla\vec{u})$$

- Laplace-Young's law :

$$\Delta P = P_{int} - P_{ext} = \gamma\left(\frac{1}{R_1} + \frac{1}{R_2}\right)$$



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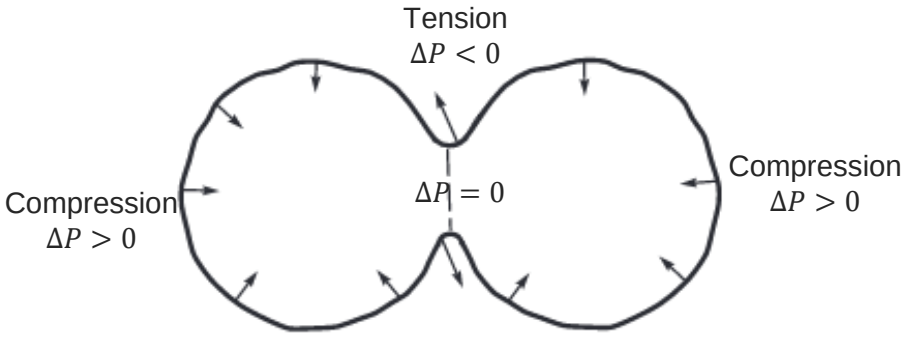

Chemical problem :

- Fick's law :

$$\vec{J} = -\frac{D_i}{RT\Omega}\nabla\mu_i$$

- Multi-species transport :

$$\frac{\partial[C_r]}{\partial t} = \nabla\cdot\left(\frac{D_{C_r,C_r}}{RT}[C_r]\nabla\mu_{C_r} + \frac{D_{U,C_r}}{RT}[U]\nabla\mu_U\right)$$

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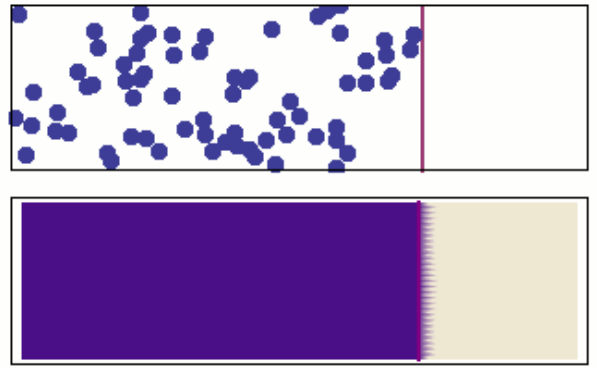

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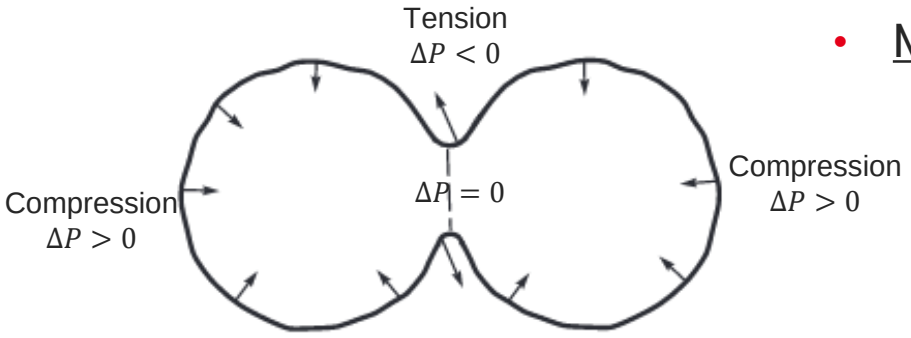

- Multiphysics coupling:

$$\mu_i = \left(\frac{\partial G}{\partial n_i}\right)_{T,P,i\neq j}$$

$$= \mu_{i,0}(T) + \mu_{i,C}(T) + \Omega\Delta P$$

$$= \mu_i^0 + RT\ln\left(\frac{C_i}{C_{i,0}}\right) - \Omega\frac{tr(\bar{\bar{\sigma}})}{3} - \Omega\frac{\bar{\bar{\sigma}}:\bar{\bar{\varepsilon}}}{2}$$

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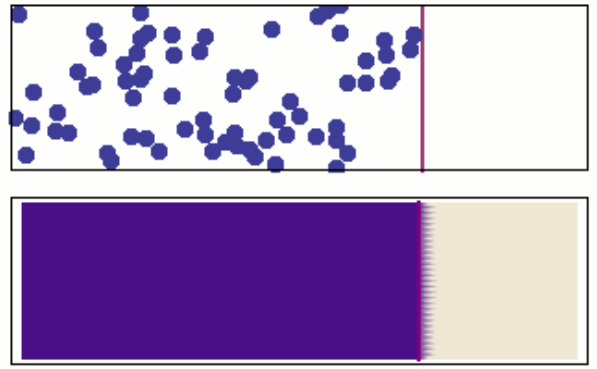


Interface evolution

FE Adaptive meshes [1]

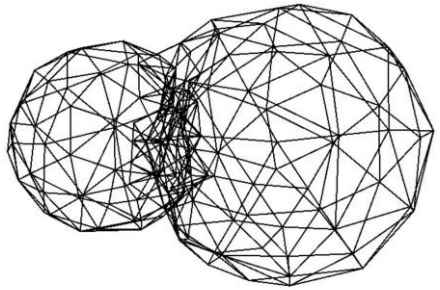
- Based directly on constitutive laws
- Lagrangian, dynamic evolution of the mesh of the surface of the grains
- Continuity of the flux :

$$J_{m,k} = -\frac{D_m}{3RT}\sum_{i=1}^{3}\frac{\partial \sigma_i}{\partial x_k} - \frac{D_m}{2RT}\sum_{i=1}^{6}\sum_{j=1}^{6} {c^{-1}}_{ij}\left(\sigma_i \frac{\partial \sigma_j}{\partial x_k} + \sigma_j \frac{\partial \sigma_i}{\partial x_k}\right)$$

$$\vec{\phi} = \Omega \iint_S \vec{J_v}\cdot\vec{n} + div_s\left(\vec{J_{int}} + \delta\vec{T}_{\vec{J_v}}\right) dS$$

$$\vec{\phi}_S \approx \Omega\left(A_S\vec{J_v}\cdot\vec{n} + \sum_{a=1}^{3} l_a\left(\vec{J_{int}} + \delta\vec{T}_{\vec{J_v}}\right)\cdot\vec{n}_a\right)$$

[1] J. Léchelle and al. «A sub-granular scale model for solid state free sintering: results on the evolution of two grains», Journal of chemical technology and metallurgy, 2014



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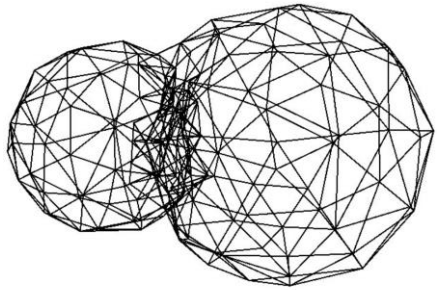


Phase-field modelling [2]

- Eulerian, evolution of the fields on a fixed mesh
- The interfacial energy between grains does not depend on the thickness of the continuous interfaces
- Total Grand Potential energy :

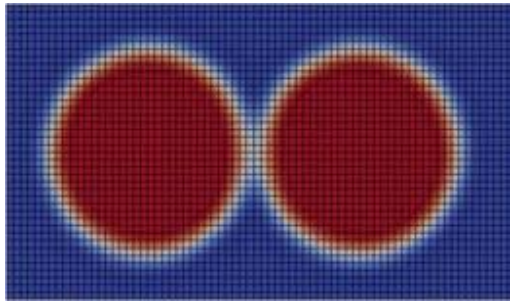
$$\upsilon(\phi, \vec{\eta}) = \int_V \omega_{well}(\phi, \vec{\eta}) + \omega_{grad}(\nabla\phi, \nabla\vec{\eta}) + \omega_{chem}(\phi, \mu)\, dV$$

- Allen-Cahn (grains and porosity phase) :

$$\frac{\partial \eta_i}{\partial t} = -L_i \frac{\partial \upsilon}{\partial \eta_i} \qquad \& \qquad \frac{\partial \phi}{\partial t} = -L_\phi \frac{\partial \upsilon}{\partial \phi}$$

- Cahn-Hilliard (diffusion) :

$$\frac{\partial \mu_I}{\partial t} = \frac{1}{\chi_{IJ}}\left[\nabla \cdot \left(M_{IJ}\nabla\mu_I\right) - \frac{\partial c_I}{\partial \phi}\frac{\partial \phi}{\partial t}\right]$$



[2] I. Greenquist and al. «Development of a microstructural grand potential-based sintering model», Computational Materials Science, 2020

CEA Software algorithm

SALAMMBO : FE Adaptive meshes [1]

Gmsh

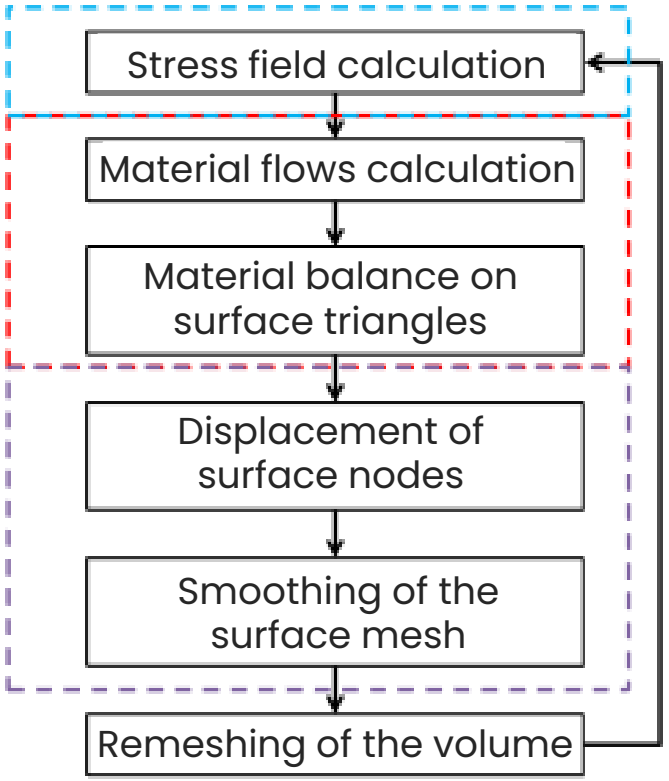
Mechanical section
3D Finite Element solving

Thermodynamic section
2D Finite Volume solving

Interfaces evolution
Lagrangian approch

Diffpack

CGAL



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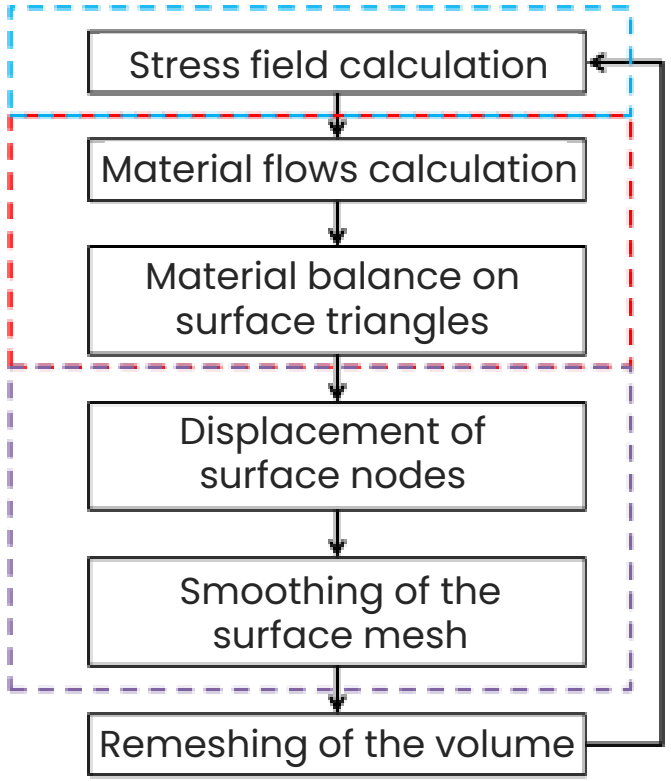

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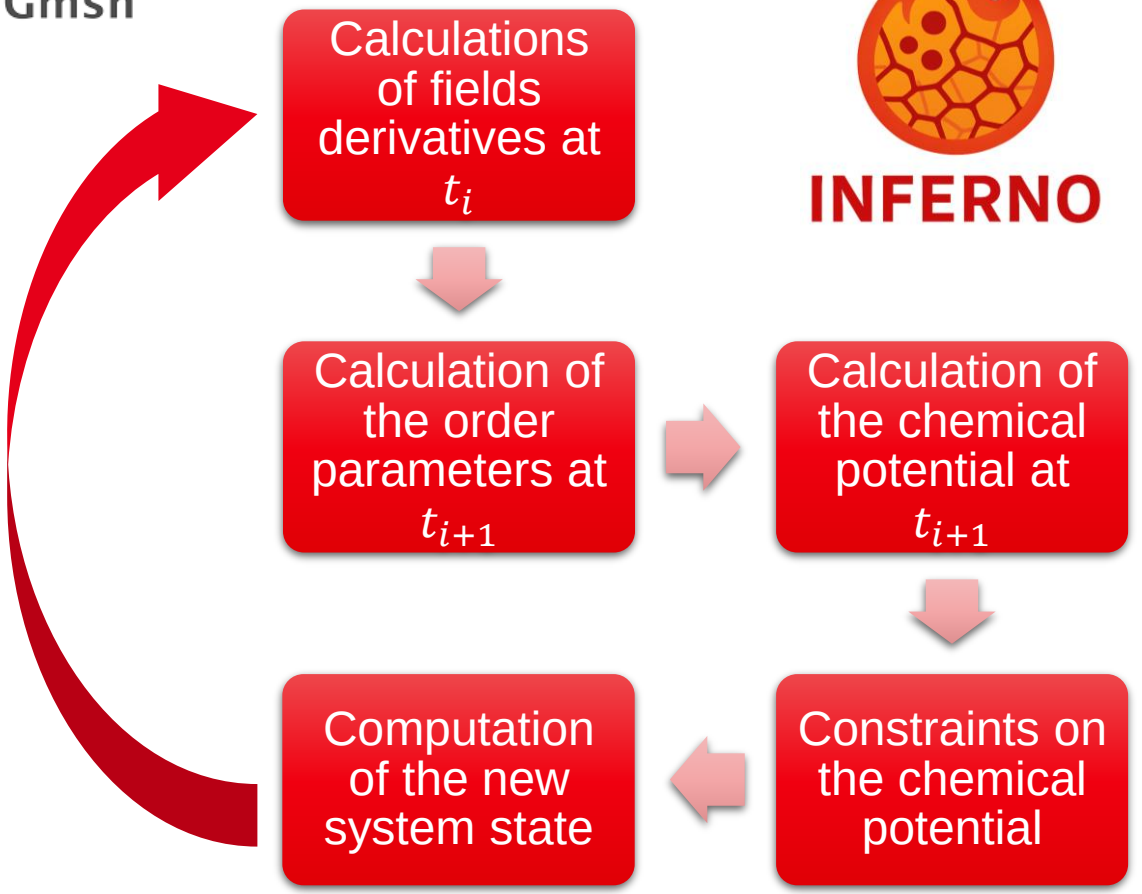
[1] J. Léchelle and al. «A sub-granular scale model for solid state free sintering: results on the evolution of two grains», Journal of chemical technology and metallurgy, 2014

INFERNO : Phase-field modelling [3]

INFERNO

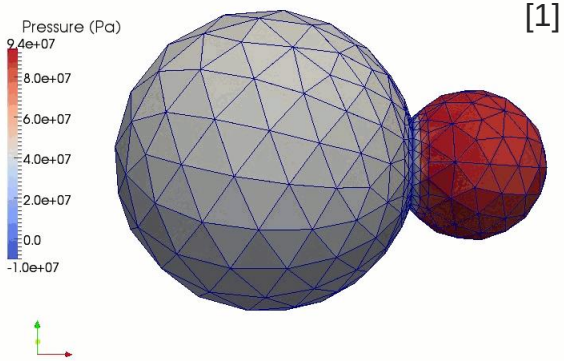


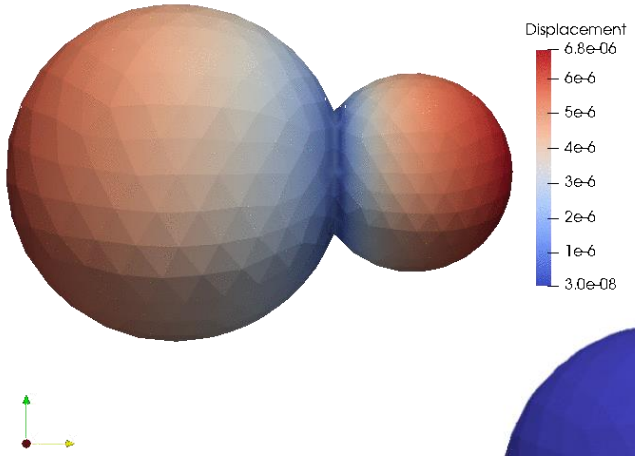
[3] L. Messina, “Phase-field modeling and benchmark of microstructure evolution in polycrystalline UO2 with the INFERNO code”, presented at the NuFuel conference, Delft, the Netherlands, Sept 18, 2025.

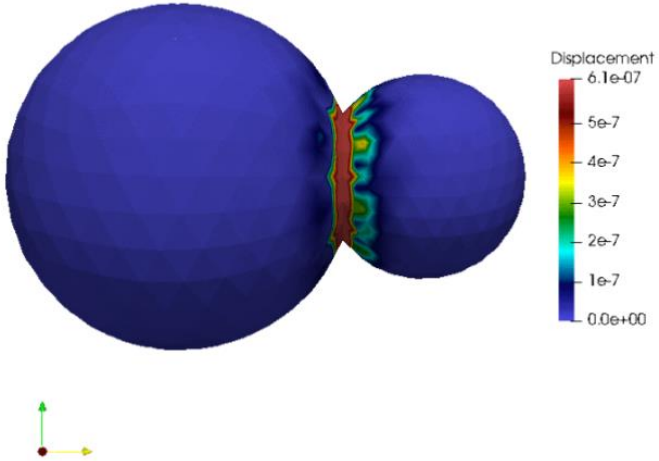
Results : Salammbo

Validation on CeO_2

[1]







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[2] Ph.D. G. I. Nkou Bouala, 2016, «First step of sintering of lanthanides and actinides oxides : an in situ study by ESEM at high temperature»

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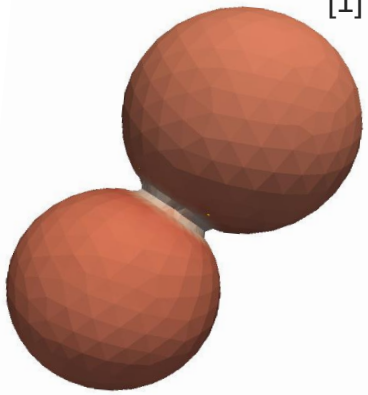

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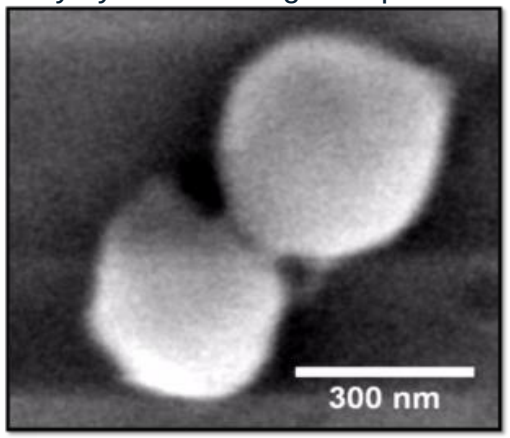

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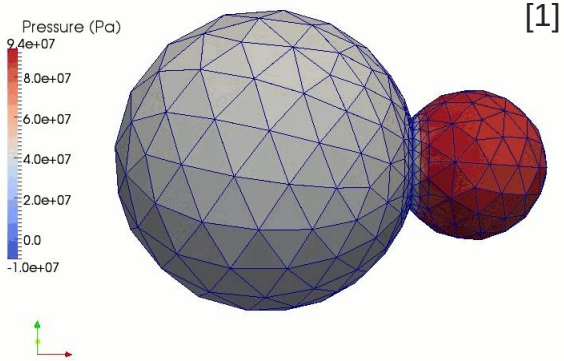

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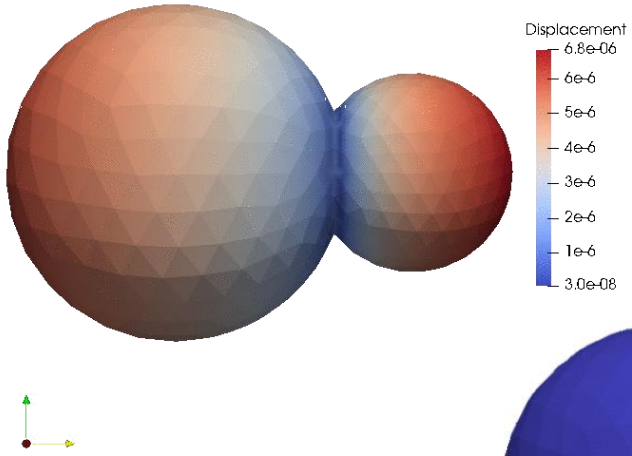

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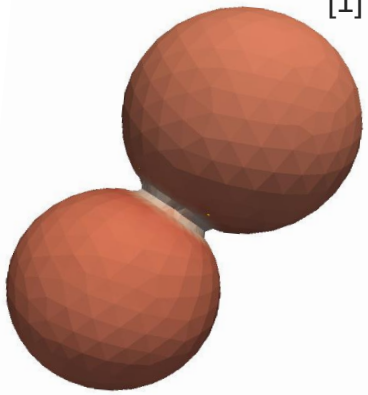

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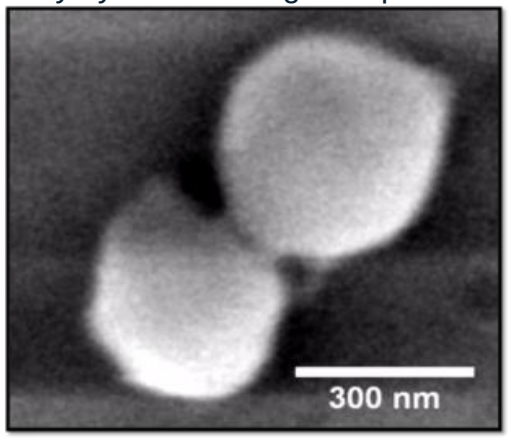
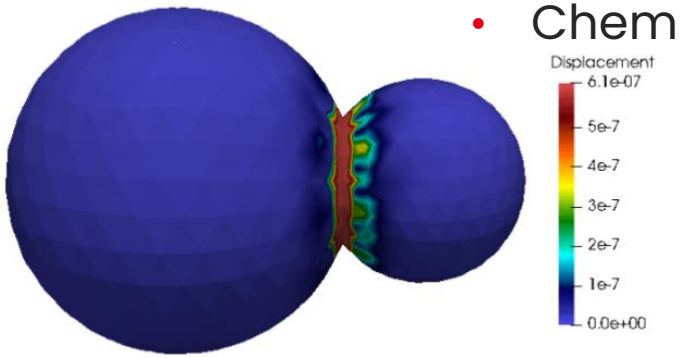
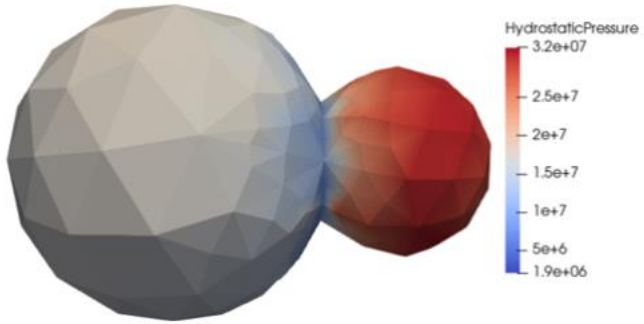
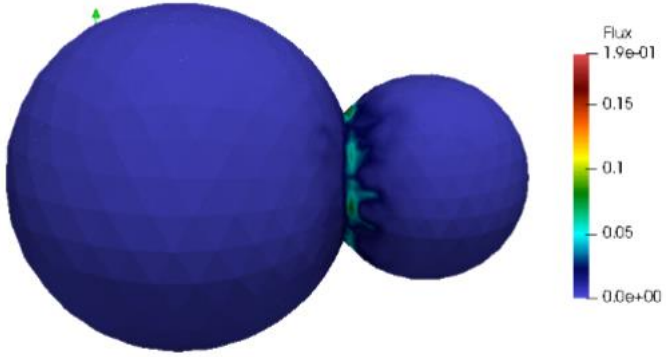


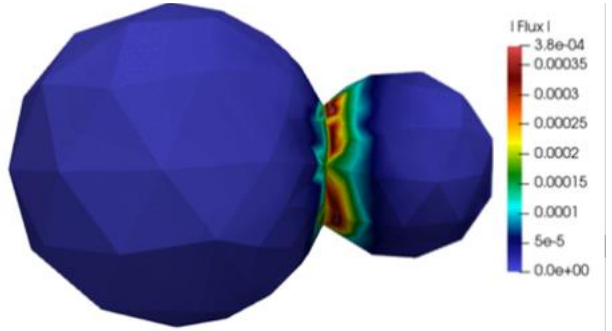
Extension to chromia doped

- Average dihedral angle at 70°
- Increase of the neck
- Chemical homogenization









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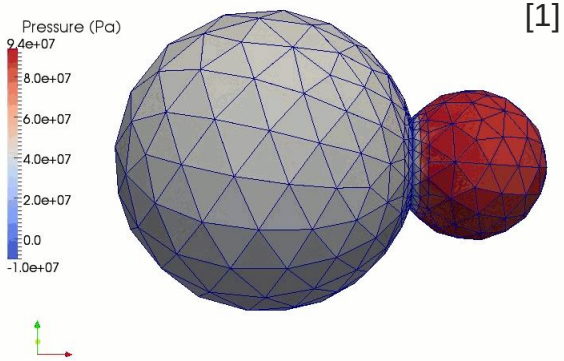

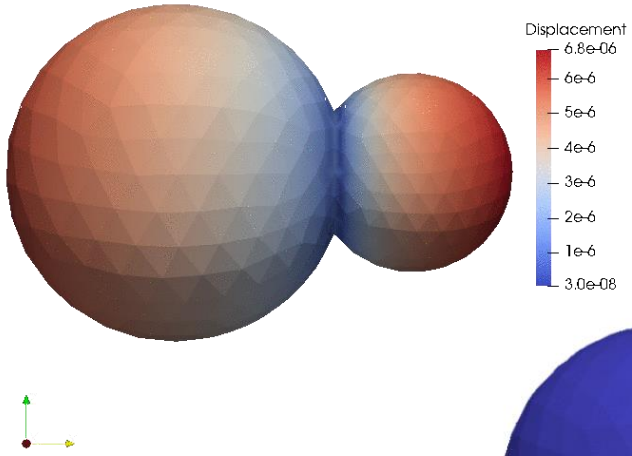

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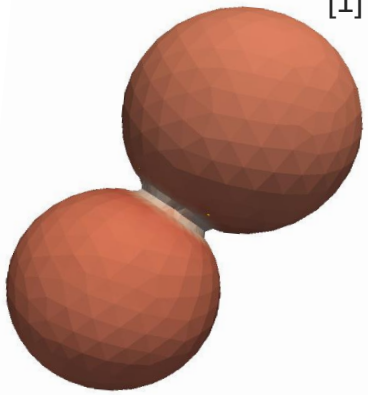

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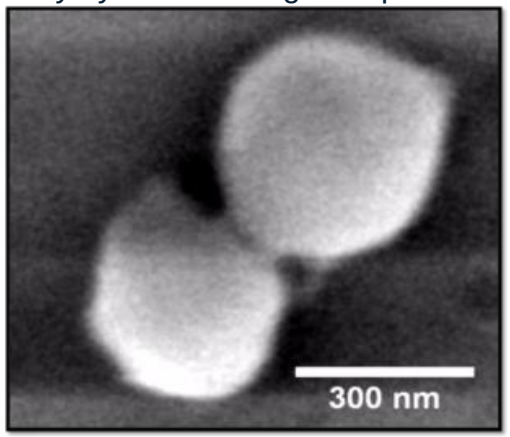

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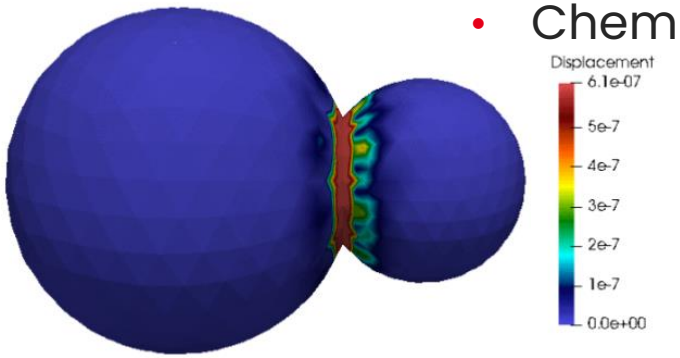
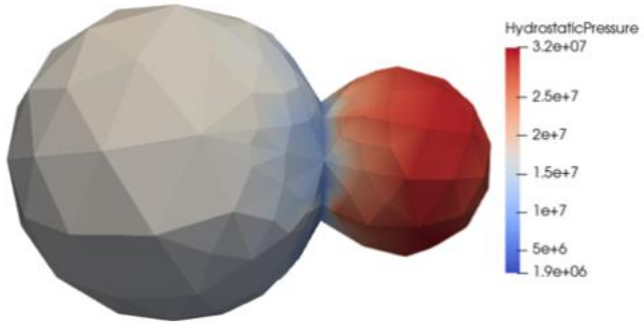
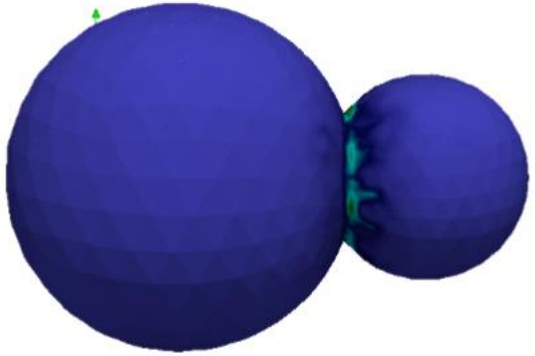
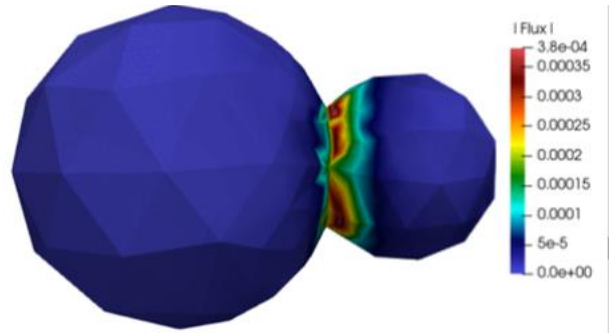




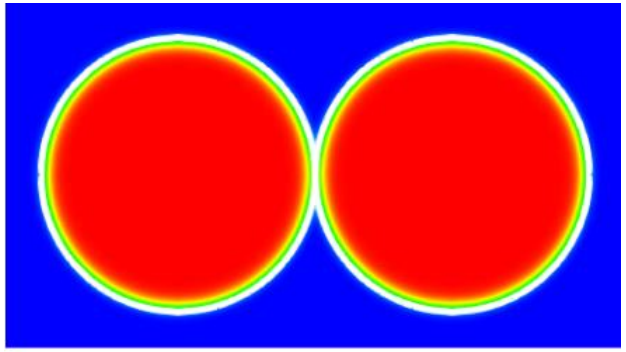


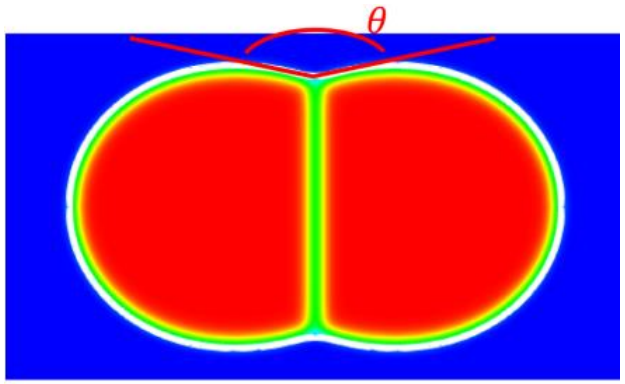


Issue with multiple grains → phase field approach

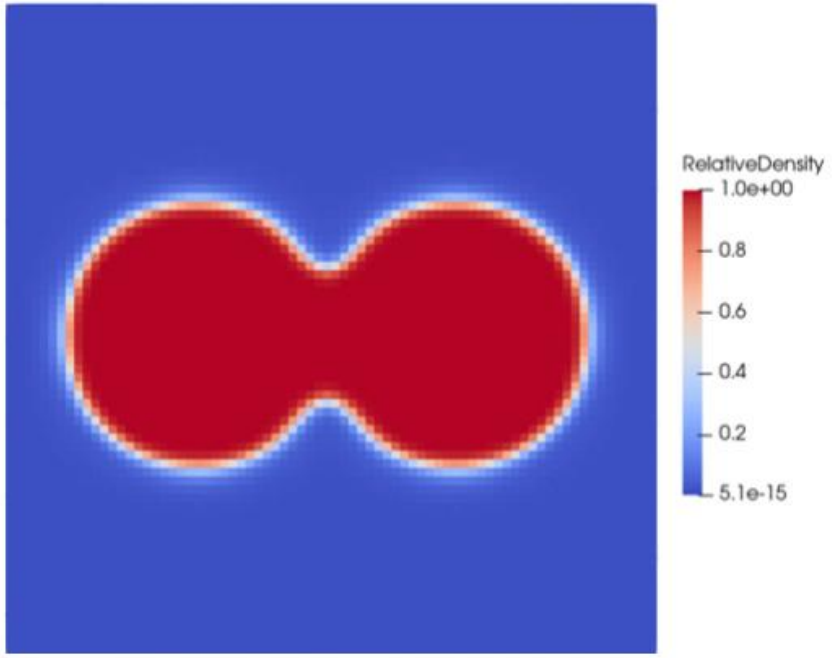
Verification : Inferno

- Implement Greenquist model, developed also for UO_2 and doped (chromium)
- Compare the first results





I. Greenquist and al. «Development of a microstructural grand potential-based sintering model», Computational Materials Science, 2020



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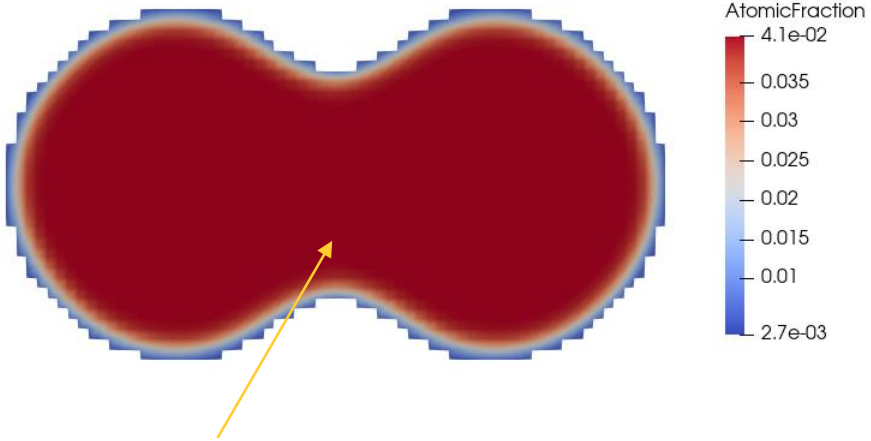
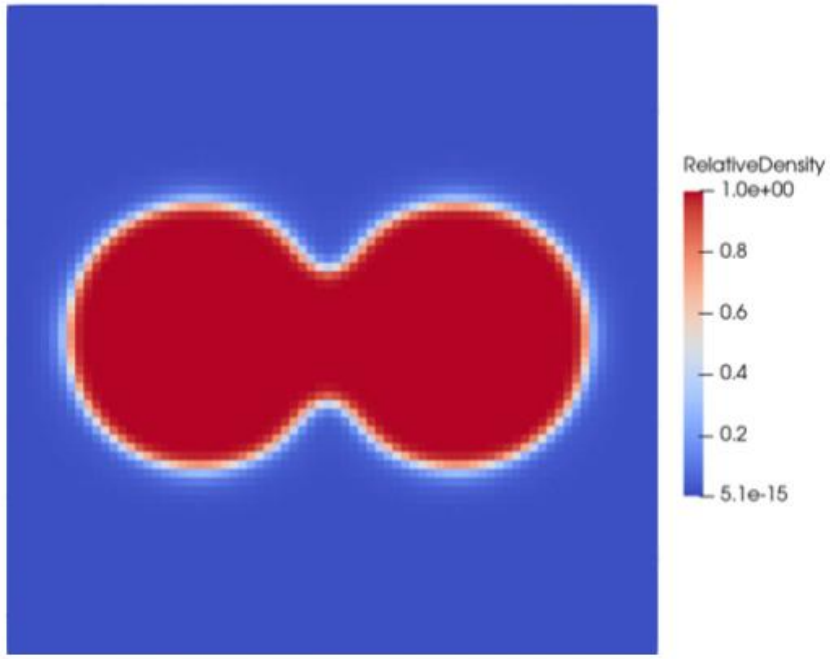


The concentration in grain must be heterogenous according to **[Greenquist et al., 2020]**
Must improve the concentration equilibrium computation



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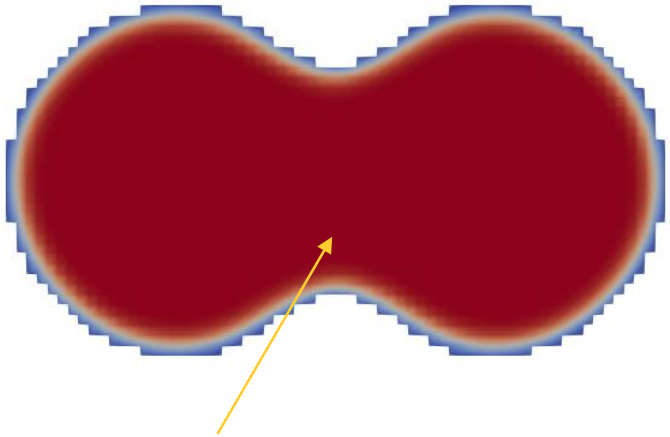

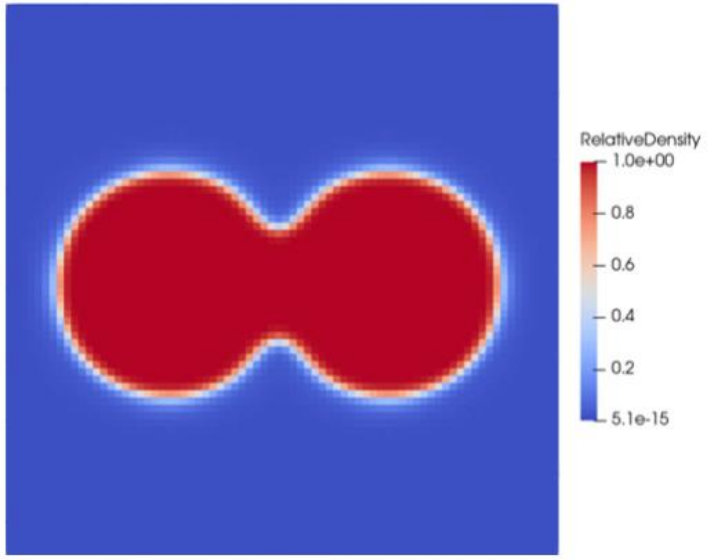

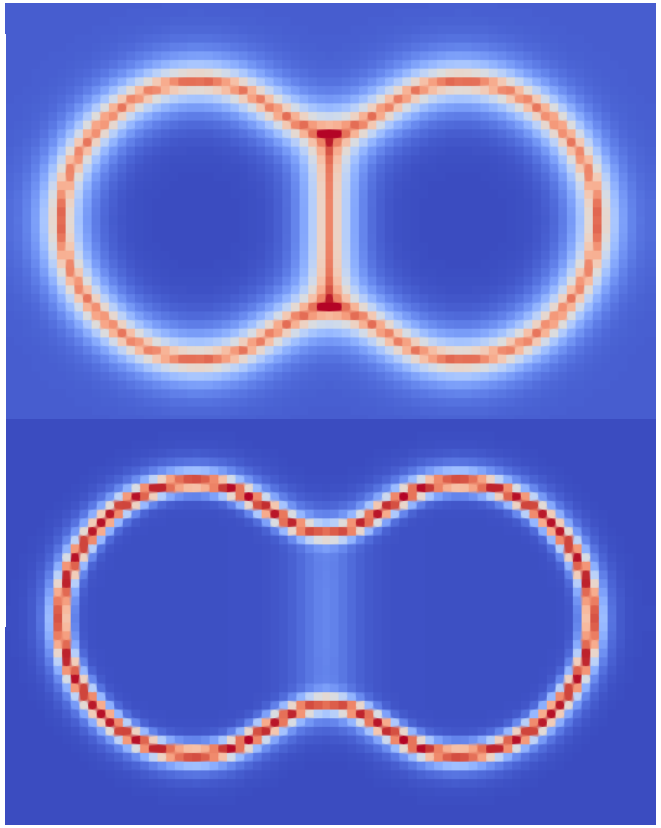

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- Issue with the equilibrium concentrations
- Extend Greenquist method to Uranium diffusion instead of vacancy in order to coupled phase -field with mechanical solver

3. Conclusion & Outlook

Conclusion

- Development of multi-species sintering model at RVE scale to study the chromia doped fuel
- Work on Salammbo (adaptive meshes):
  - Extend model for multi-species sintering
  - Further works will be focused on phase-field model due to CPU time and multi-grain issues
- First implementation of Greenquist model and verification

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Outlook

- Extend Greenquist method to Uranium diffusion instead of vacancy
- Benchmark between Salammbo and Inferno
- Coupled phase field method with mechanical solver, compared with multi-grains systems
- Extension to Uranium and doped sintering for phase-field model

“Thanks for your attention

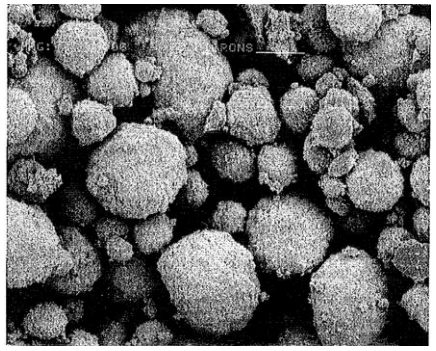
Evan Pereira

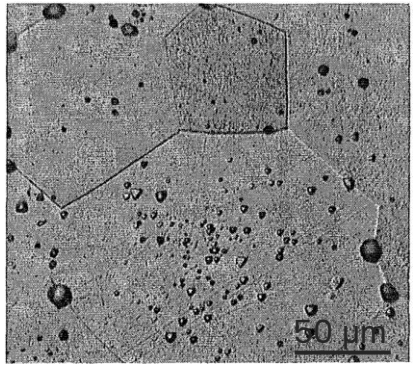
Evan.PEREIRA@cea.fr

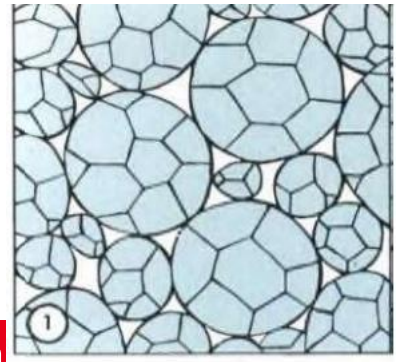
Context : Sintering of the fuel pellet

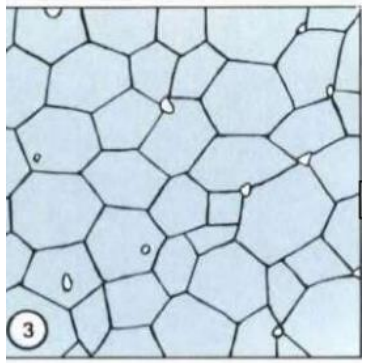
Micro-scale behavior :

- Mass transport, primarily driven by solid-state diffusion at grains boundaries and surface
- ↗ grain size (from 1 to 10 µm up to 5 and 25 µm)
- ↘ porosity and number of grains



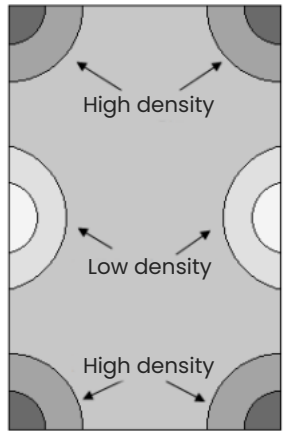


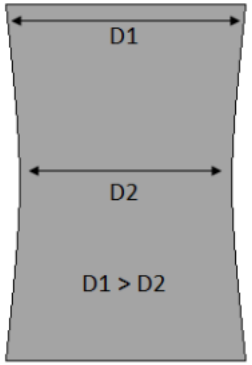




Macro-scale behavior :

- Target O/M stoichiometry close to 2
- ↘ volume due to shrinkage (may require adjustment)
- ↗ density from ~55% to ~95%





Residual closed pores are important to trap fission gases within the microstructure.