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High temperature creep of uranium dioxide: on the influence of equilibrium oxygen partial pressure

P. Garcia, A. Miard, J.-B. Parise, M. Ben Saada, X. Iltis, C. Introni, T. Helfer

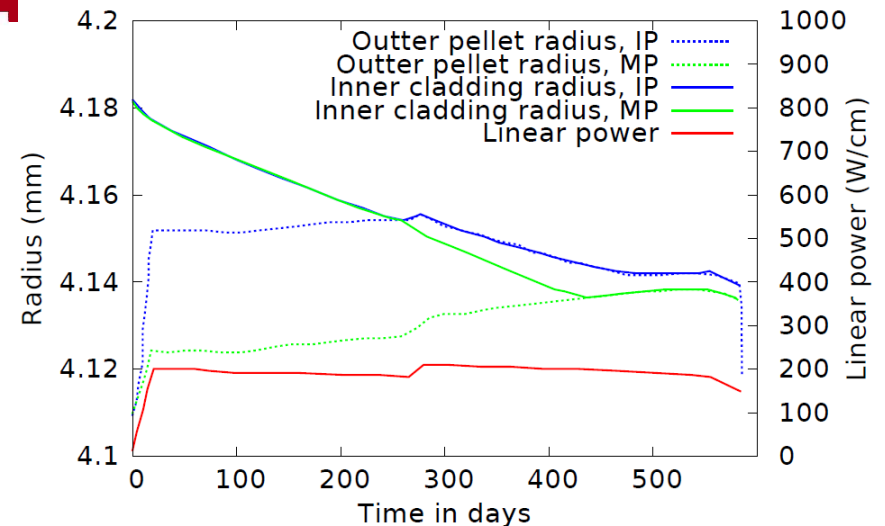
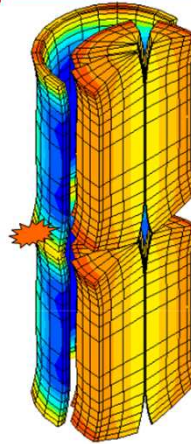
CEA Cadarache, DEN, DEC

MRS Spring meeting - Phoenix | 4 April 2018

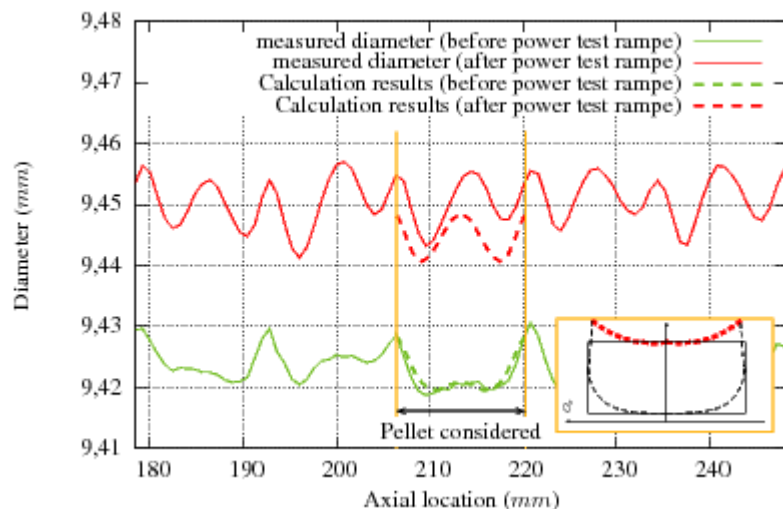
- 1. Context of study
- 2. From creep testing to understanding microstructural effects
- 3. Experimental details
- 4. Modelling and interpretation
 - 1 vs. 2D modelling
 - Microstructural changes
- 5. Conclusion and prospects

1.1 An essential modelling ingredient for fuel performance applications

- Pellet and cladding subject to range of TM effects^[1]
- Differential thermal expansion
 - pellet cracking \Rightarrow hourglass
 - Radiation induced Creep



Kinetics of formation of primary ridges & TM state of cladding are determined by radiation induced creep



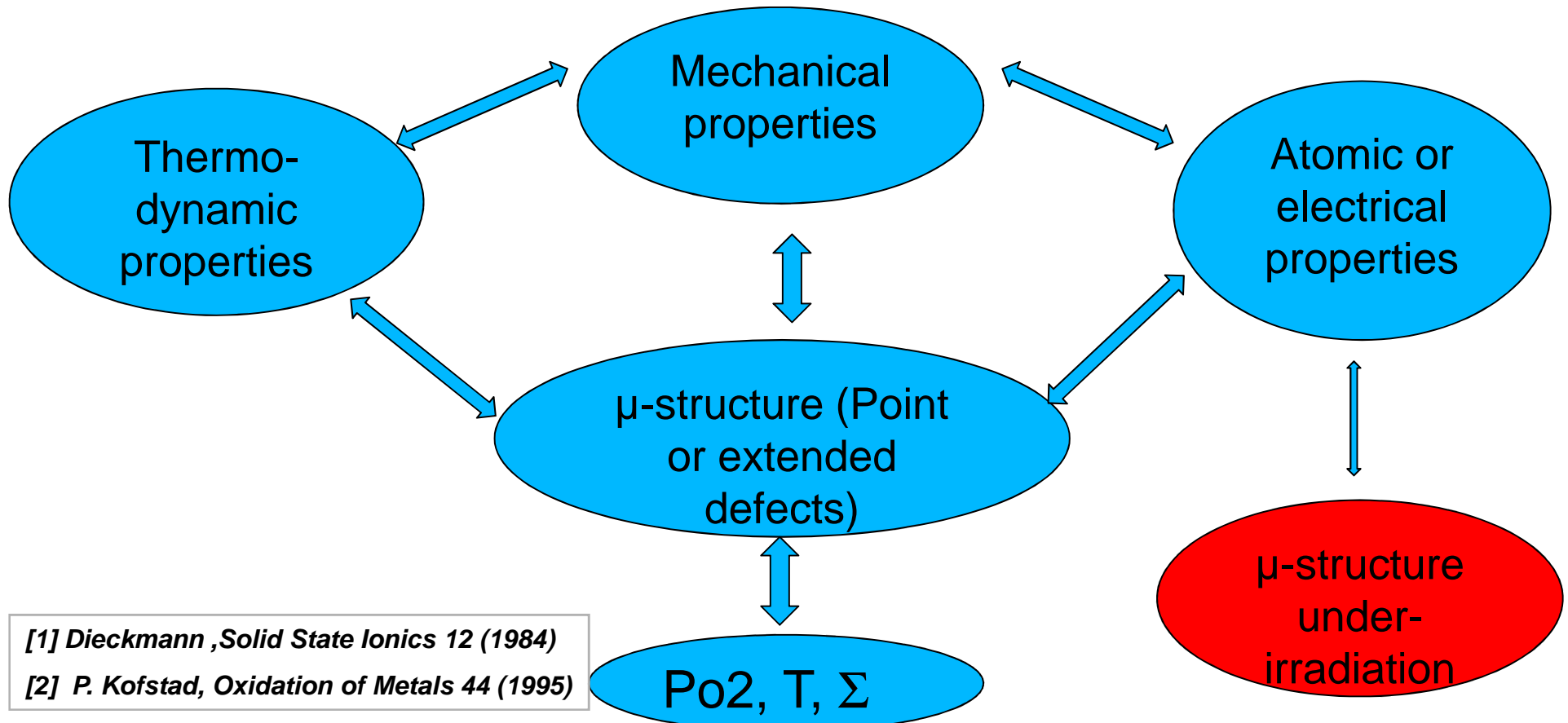
During power ramps, cladding stresses and strains controlled by thermal creep of pellet and cladding

[1] Helfer, Castelier, Garcia, Euromech 2005, Eindhoven

1.2 Material's property approach

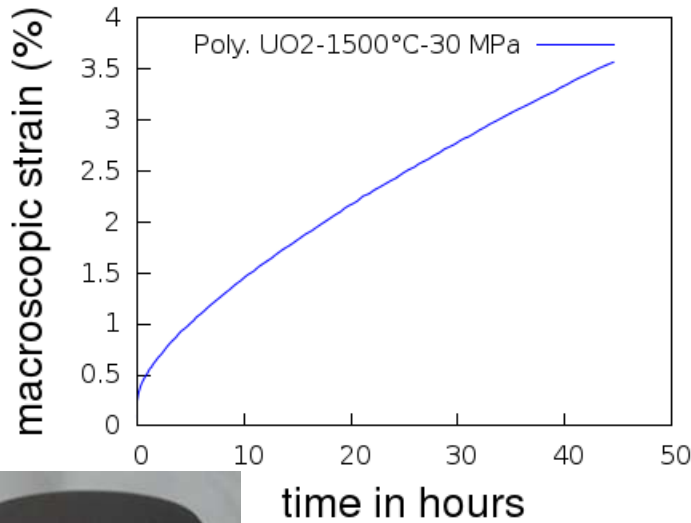
Quantify, understand and model the influence of microstructure upon oxide fuel properties^[1,2]

- Redox activity, dopants or additives, grain size, porosity, irradiation...

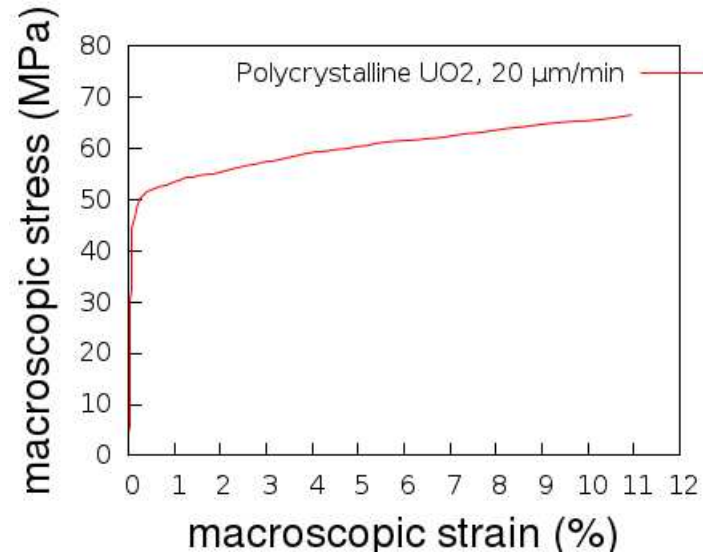


2.1 From macroscopic creep tests to mechanisms...

- Macroscopic compression/bending tests: constant load or strain rate



time in hours
 $\sim 10^{-7} \text{ s}^{-1}$



$2 \times 10^{-5} \text{ s}^{-1}$



- Fuel behaviour applications:

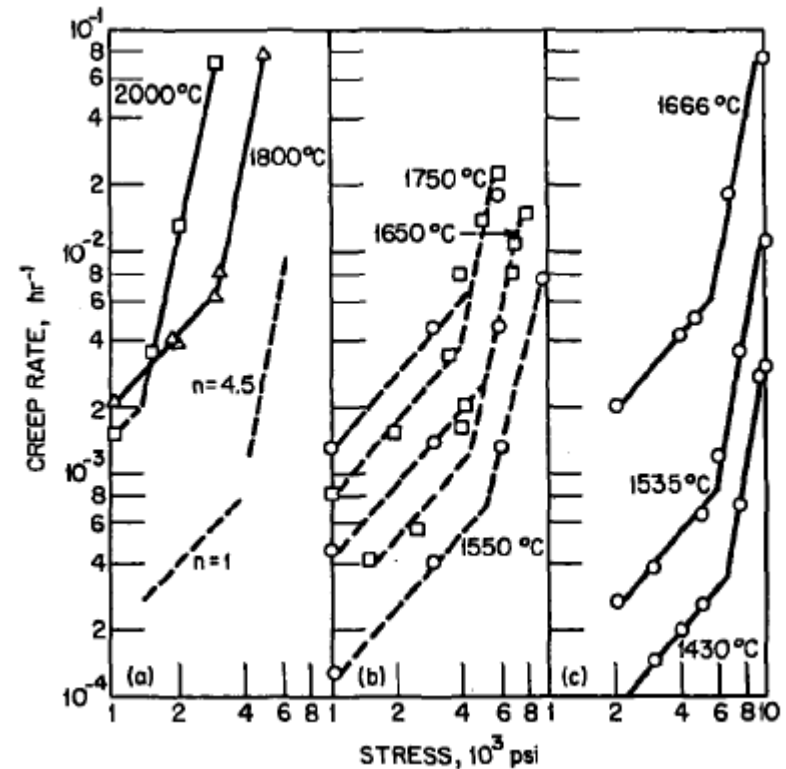
$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times \text{Exp}\left(-\frac{E}{kT}\right)$$

How well do these laws reproduce the behaviour of UO_2 ?
What do the coefficients actually represent?

2.2 Effect of stress and grain size in “nominally stoichiometric material”

$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times \text{Exp}\left(-\frac{E}{kT}\right)$$

- High $\sigma \Rightarrow$ high n values ($4.5^{[1,2]}$):
 - dislocation sources: glide and climb
 - dislocation movement inhibited by GB (hardening, $p \sim -2$)^[3]
- Low $\sigma \Rightarrow$ low n ($1^{[1,4]}$):
 - Diffusion of defects due to chemical potential gradient
 - strain rate inversely proportional to grain size ($p \sim -2$), volume diffusion control^[2]
- Transition at σ level dependent upon grain size



What physical meaning can be given to the activation energy?

[1] M.S. Seltzer, JNM 34 (1970)

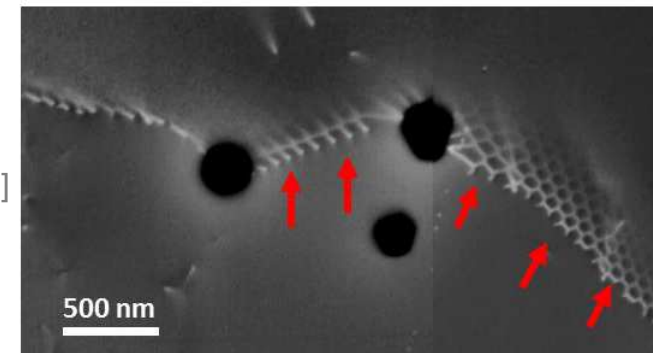
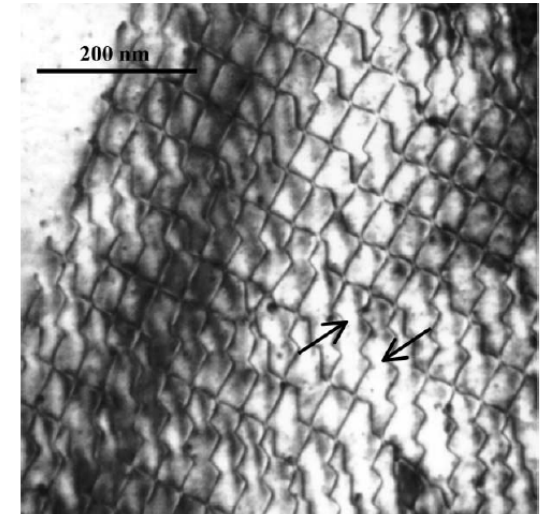
[2] Weertman JAP 26, XXXX

[3] C. Duguay, PhD, INSA Lyon (1998)

[4] C. Herring JAP 21, XXXX

2.3 Relationship between cation diffusion and creep?

- In pure metals, E is the self-diffusion activation energy (Weertman & Herring)
- Creep properties in binary or ternary oxides^[1]?
- At low stress:
 - Diffusional controlled deformation process and kinetics
 - $\propto D$ of slowest moving element, migrates with least abundant defect population^[2] (cation)
- At high stress:
 - Temperature activated glide & Dislocation climb (kinetics)
 - In UO_2 combination of both (recovery-creep)^[2,3,4]
 - $\propto D$ of slowest moving species



Ecci image: sub-grain boundary^[5]

$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times D_U$$

[1] J. Philibert *ssi.* 12 (1984)

[2] Garcia et al. *JNM* 494, 2017

[3] Dherbey et al. *Acta Mater.* 50 (2002)

[4] Alamo et al.

[5] Ben Saada PhD, Metz 2017 (Gey, Maloufi, Iltis)

2.4 Effect of non-stoichiometry and oxygen pressure

- If creep is self-diffusion controlled \Rightarrow strong dependence upon deviation from stoichiometry^[1]
- More appropriate to interpret data $f(p_{O_2})$
 - p_{O_2} is the true thermodynamic property^[2,3,4]
 - Practical reasons: $\Delta x \sim 2 \cdot 10^{-3}$, Δp_{O_2} : 4 orders of magnitude
 - Measured parameters are physically meaningful^[2]
- One can expect the following

$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times f_U[V_U] D_{VU}$$

$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times f_U p_{O_2}^\alpha \exp\left(-\frac{E_f}{kT}\right) \times D_{VU}^0 \exp\left(-\frac{E_m}{kT}\right)$$

α and $E_f \Rightarrow$ characteristic of defect formation, E_m and $D_{VU}^0 \Rightarrow$ characteristic of defect migration

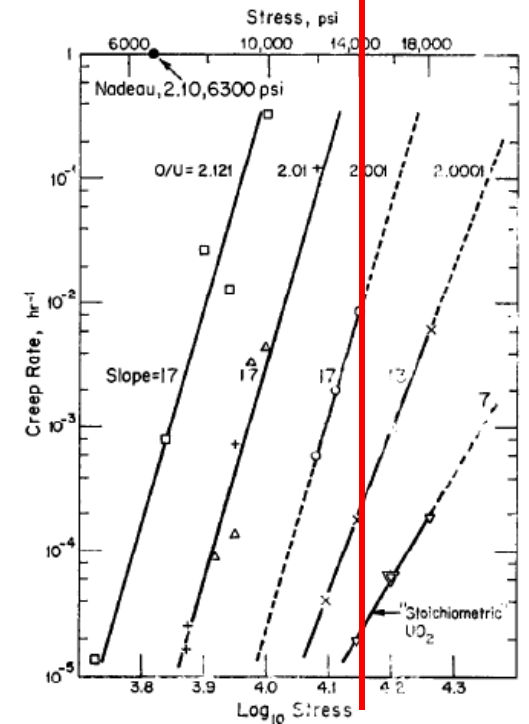


Fig. 2. Creep rate as a function of applied stress for UO_{2+x} single crystals tested in compression at 1100 °C. The compression data of Nadeau²⁰⁾ for slip is included.

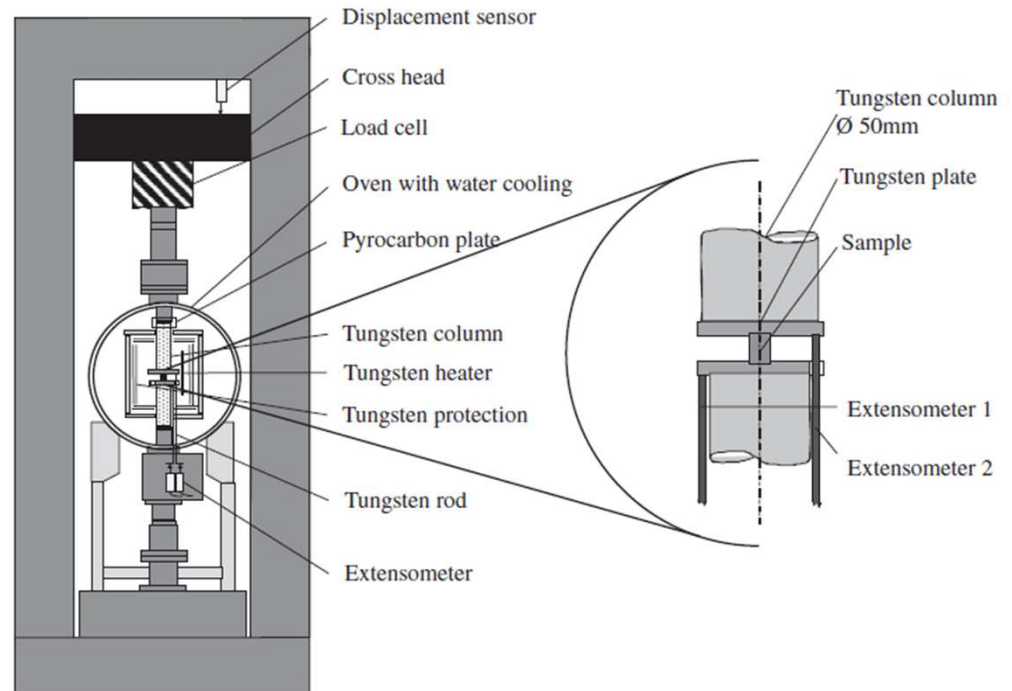
[1] Seltzer et al. J. Nucl. Mater (1972)

[3] Dorado et al. PRB 83, 2011

[2] J. Philibert ssi. 12 (1984)

[4] Dorado et al. PRB 86, 2012

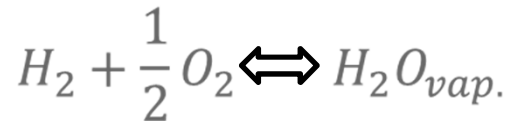
3.1 Creep tests under controlled atmosphere



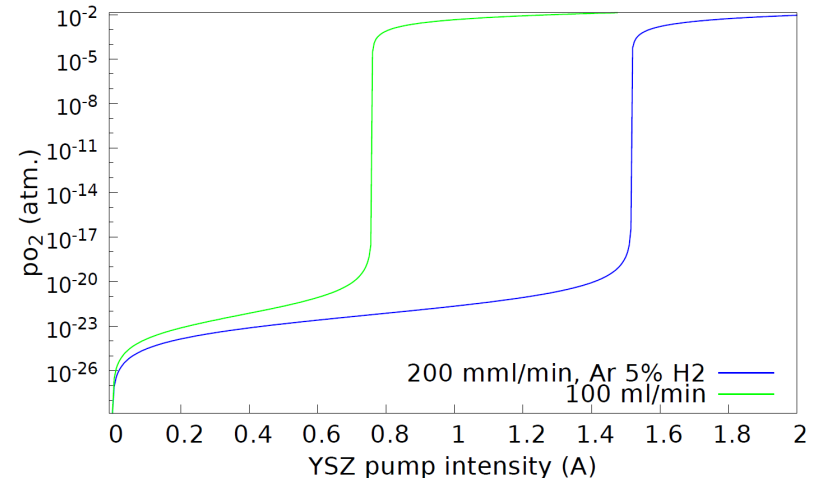
- Compression testing
 - 100 kN frame equipped with W ~1700°C furnace & – *in situ* extensometers
 - Originally all tests carried out under Ar/H₂
 - Roughly 50 l of “free volume”
- System equipped with oxygen and humidity probes, upstream and downstream

3.2 Atmosphere control

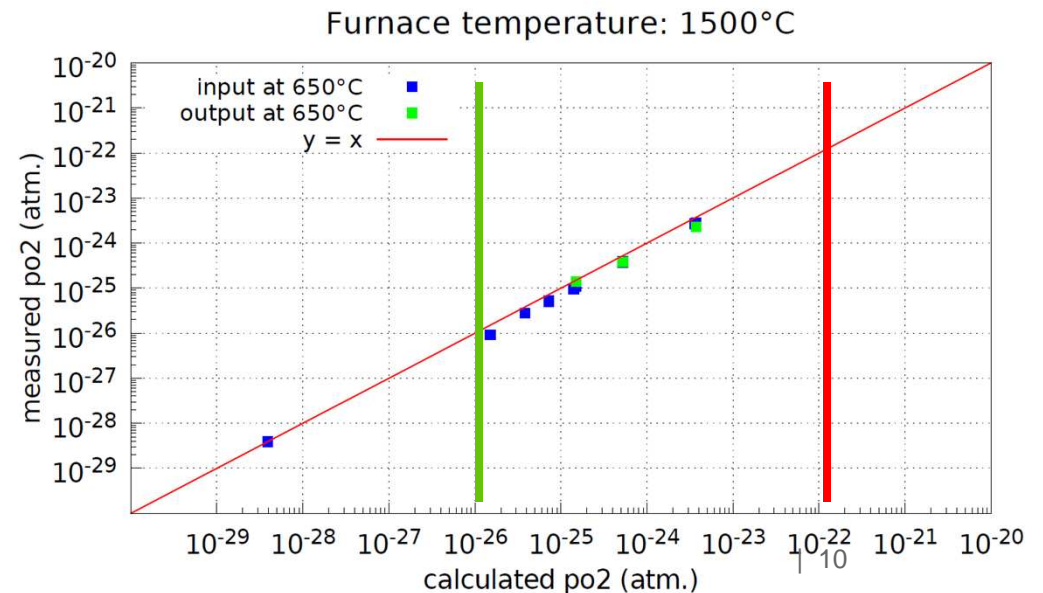
- Control based on buffering gas phase



- Ar/H₂ carrier gas + humidification with YSZ oxygen pump
- Calculated values (know from Faraday's law [1]) ($n_{O_2} \propto i$)

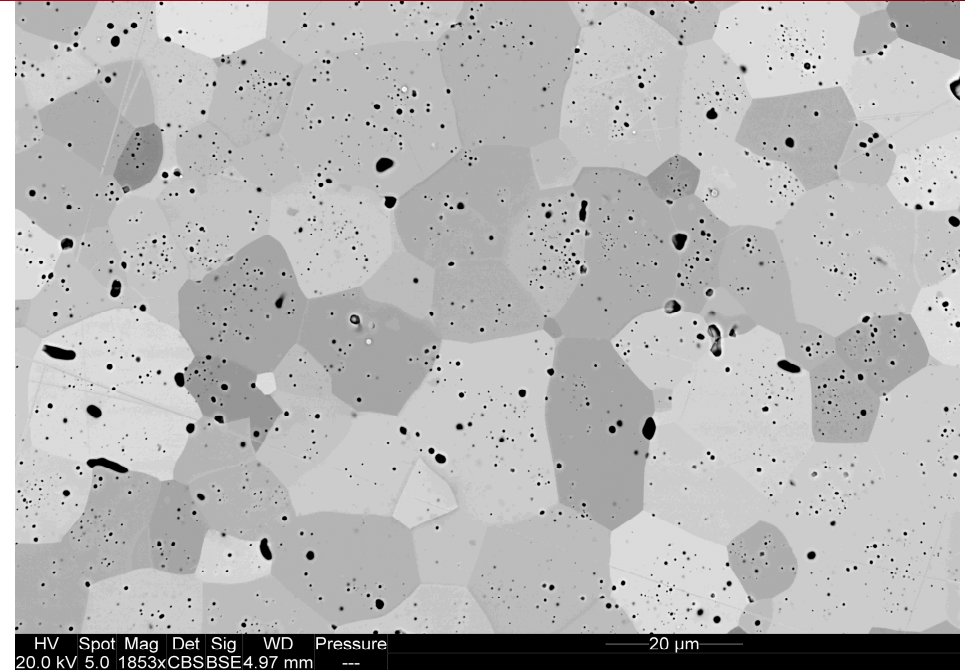


- Excellent agreement bet. th. and exp.
- Actual equilibrium of input and output \Leftrightarrow 12 hours
- Restricted range of accessible pressures
- Lower bound: determined by necessity for buffering to function
- Upper bound: furnace oxidation



cea 3.4 Materials used and tests run

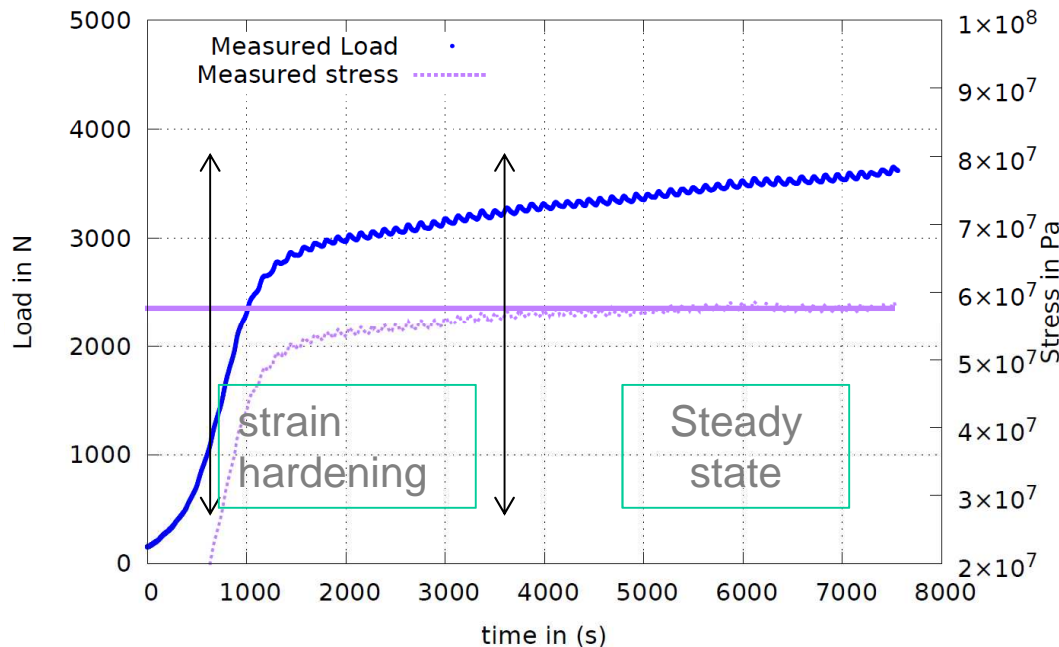
- All tests run at 1500°C on polycrystalline samples
- 16.1 mm +/-0.2 mm height, 8.190 mm diameter +/-0.010 mm ⇒ limiting 2D effects
- Grain size ~ 14 μm – pore fraction 1.8 +/-0.1 % (high density)
- Final strain between 13 and 14%



Test number	Crosshead speed (μm/min.)	Approximate linear strain rate (s ⁻¹)	Oxygen pressure (atm.)	Duration (mins)
1	20	2×10^{-5}	1.3×10^{-12}	126
2	20	2×10^{-5}	1.1×10^{-11}	123
3	20	2×10^{-5}	1.3×10^{-11}	129
4	20	2×10^{-5}	4.8×10^{-11}	127
5	20	2×10^{-5}	7.3×10^{-11}	130

4.1 Macroscopic experimental data available and preliminary analysis

- Experimental data available:
 - *In situ* pellet height $h(t)$, load $F(t)$
 - Post-test profile, height and density



- **Linear** crosshead displacement
- Initial phase: **combined response** of setup and pellet
- Fluctuations in measured load values
- Small fluctuations in displacements measured by extens.

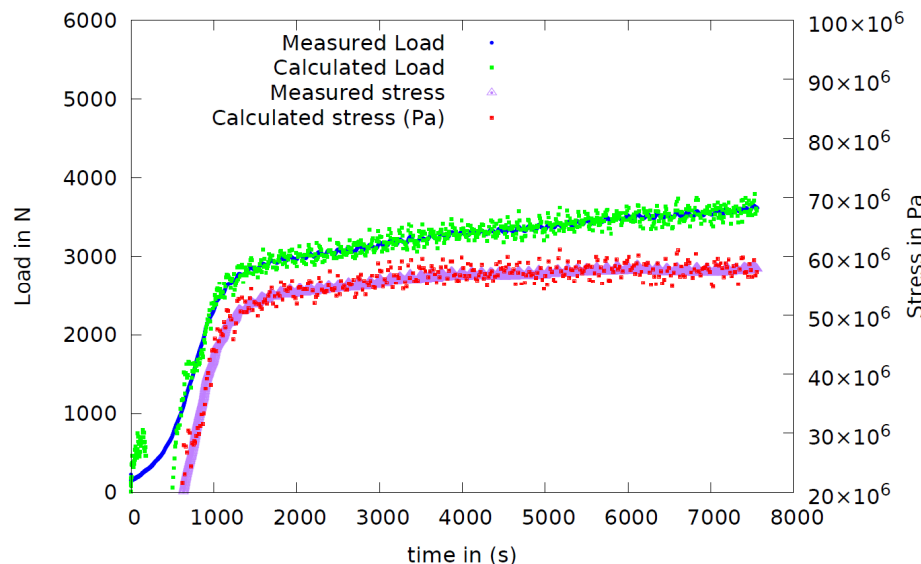
- Uniaxial hypothesis & constant V process $\Rightarrow \sigma_{eq}(t) \approx \sigma_z(t) \approx \frac{F(t)}{S(t)} \approx \frac{F(t)h(t)}{h_0 S_0}$
- Primary & secondary creep (Norton)

Uniaxial temptation is great
macroscopic hardening \Leftrightarrow primary creep & increase in section

4.2 How uniaxial are tests carried out at constant crosshead speed?

- Barrel-shaped pellet \Rightarrow friction with tungsten plates
- Salvo *et al.* have suggested pellet is clamped^[1]...

- 1 and 2D (axisymmetric) model using “mfront”^[2]:
behaviour law integrator
 - “mtest” in 1D^[3] environment
 - “licos@Cast3M”^[3,4]
 - Chaboche-type Kinematic hardening law \Rightarrow primary creep^[5]



$$\dot{\epsilon}_{vp} = \left(\frac{|\underline{\sigma} - \underline{X}|}{K} \right)^n$$



- 1D Model reproduces data
- Even fluctuations (contained in displacement load) are reproduced
- Correlated parameters

\Rightarrow Confirms uniaxiality

[1] Salvo *et al.* JNM 456 2015

[2] Helfer *et al.* Computers & mathematics with applications 70 (2015)

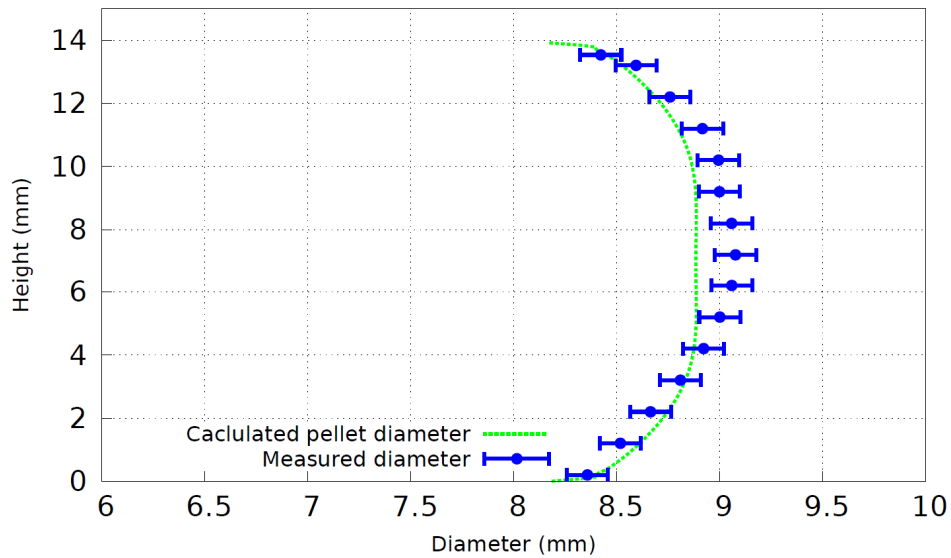
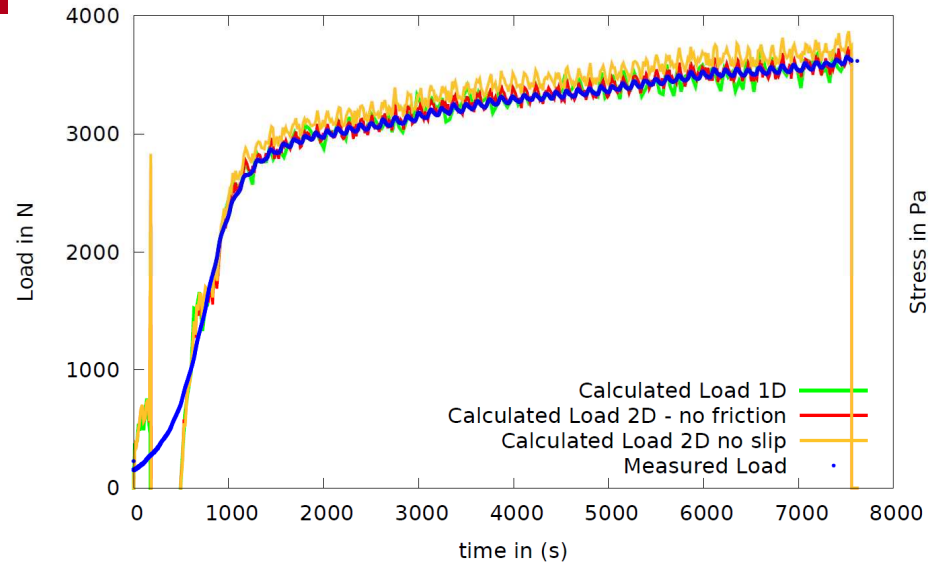
[3] Helfer *et al.*, J. Nuc. Eng & Des. 294 (2015)

[4] <http://www-cast3m.cea.fr/>

[5] Colin, Thèse Ecole des Mines, 2006

4.3 Comparison of 1 and 2 D approaches

- 1D determination of model param.
- 2D: identical parameters and load
 - assuming no friction
 - assuming infinite friction
- Only **slight change in parameters** required
- 2D model reproduces profile ~
- **Uniaxial approach justified**

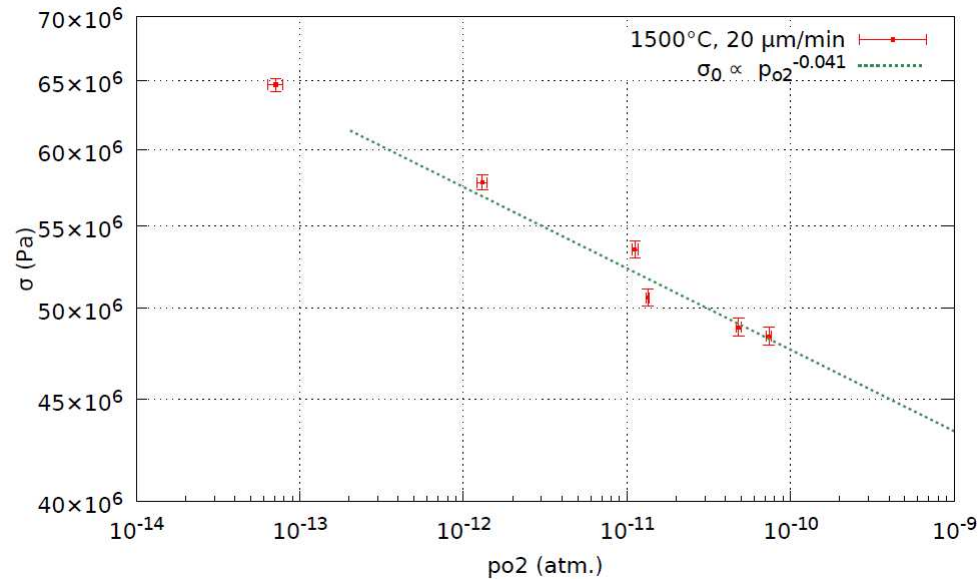


- 1D Model reproduces data
- Even fluctuations (contained in displacement load) are reproduced
- Correlated parameters

4.4 Result of 1D analysis

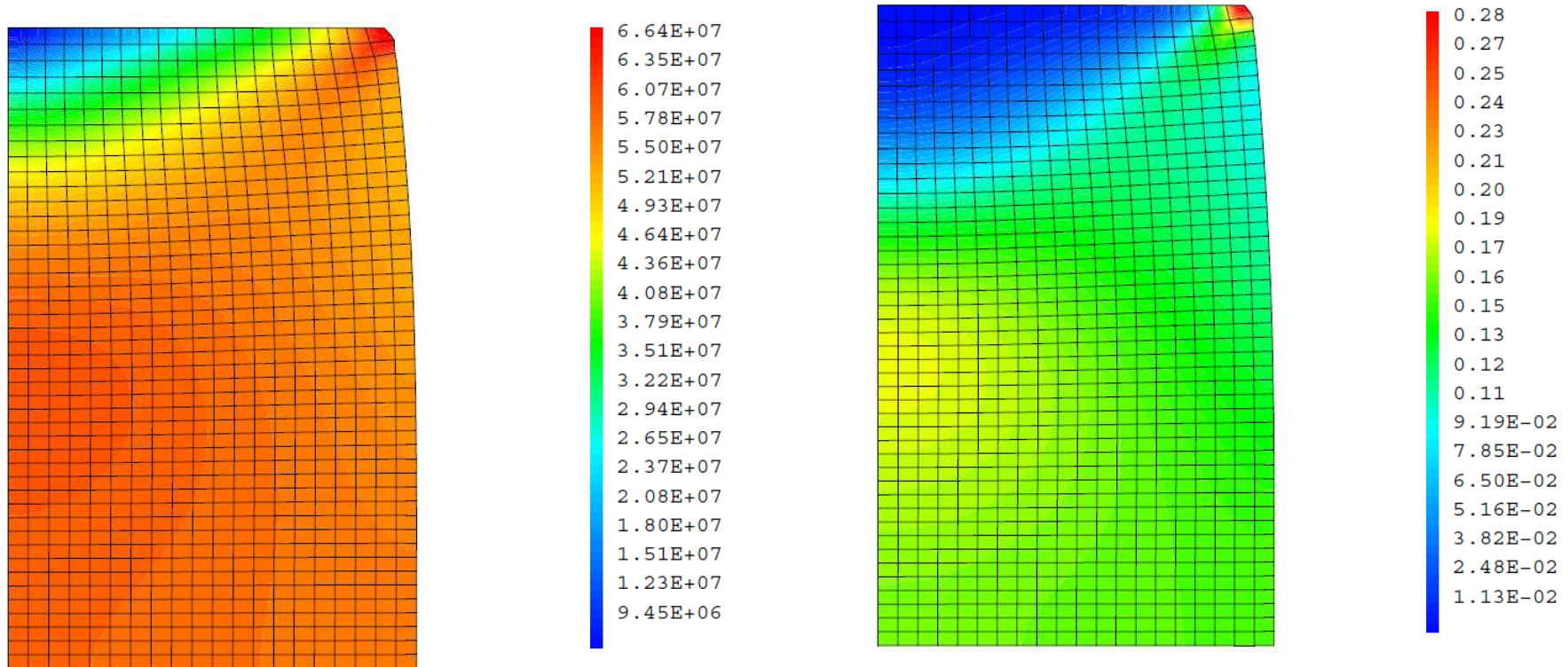
- If $X \ll \sigma$, analysis is straightforward and
- Plotting $\text{Log}(\sigma) = f(\text{Log}(p_{o_2})) \Rightarrow -\alpha/n$

$$\dot{\epsilon} \propto K p_{o_2}^{\alpha} \sigma^n$$



- Data points lie in ~ **straight line**
- $-\alpha/n \sim -0.041 \Rightarrow$ if $n \sim 4$, $\alpha \sim 1/6$
- Decreasing $f(p_{o_2}) \Rightarrow$ consistent with $\alpha > 0$ and **increasing $[Vu] \sim p_{o_2}^{1/6}$**
- n **can be identified** by changing conditions

■ Stress and strain inhomogeneity within the pellet



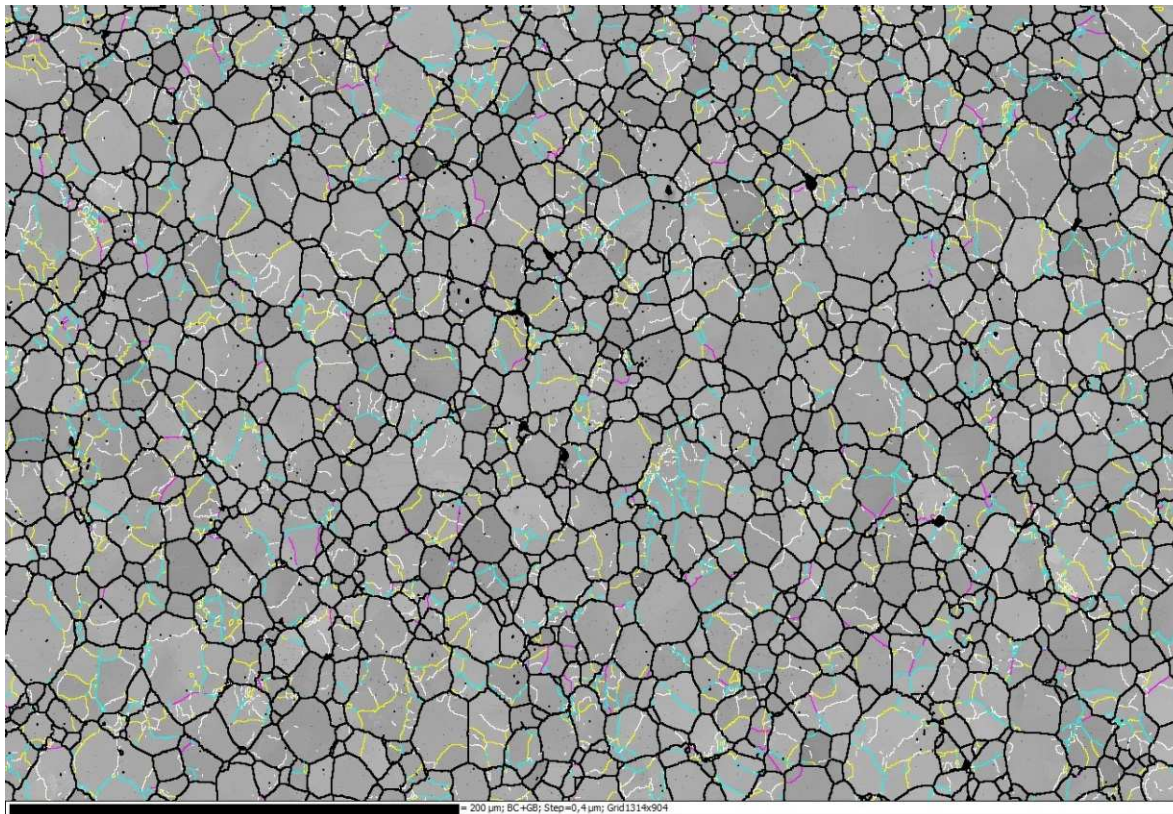
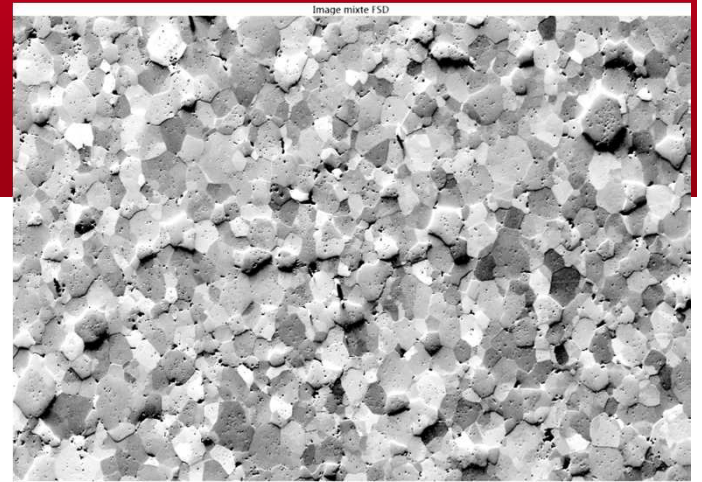
- Can one qualitatively correlate calculations to observable local microstructural changes?
 - Material reacts through process of grain fragmentation^[1]
 - Formation of cavities (void swelling) at high deformation values^[2]

cea 4.6 Preliminary characterization results

- P1, P2, P3 ($5 \cdot 10^{-26}$, $5 \cdot 10^{-25}$, $2 \cdot 10^{-24}$): **identical macroscopic** strain
- P1: Damage localization
- Formation of pockets of voids, located “away from” the plane of symmetry
- With increasing p_{o2}
 - **Volume fraction of pockets increases**
 - Associated grain **misorientation increases**

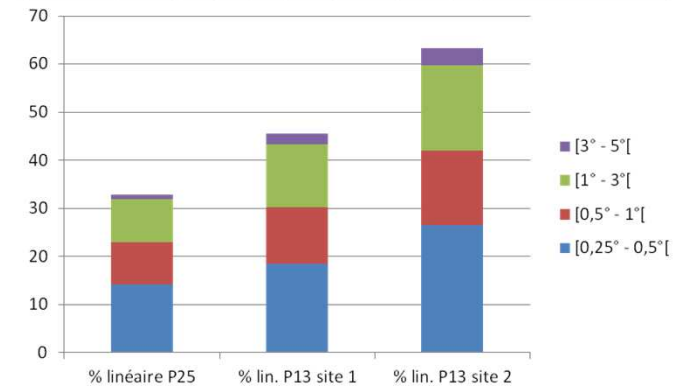
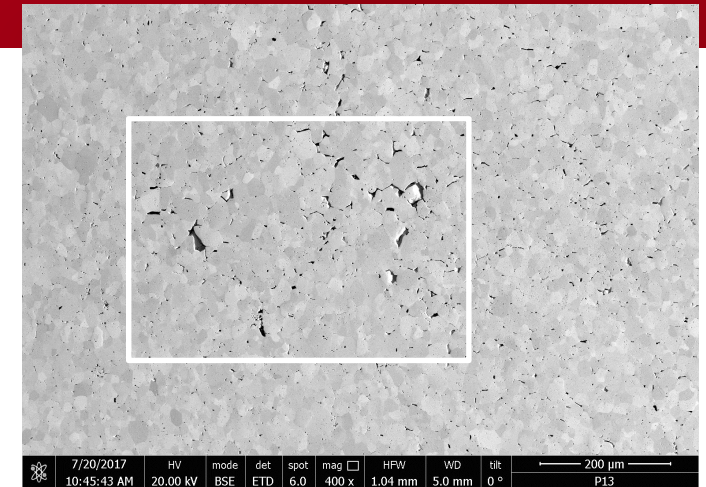
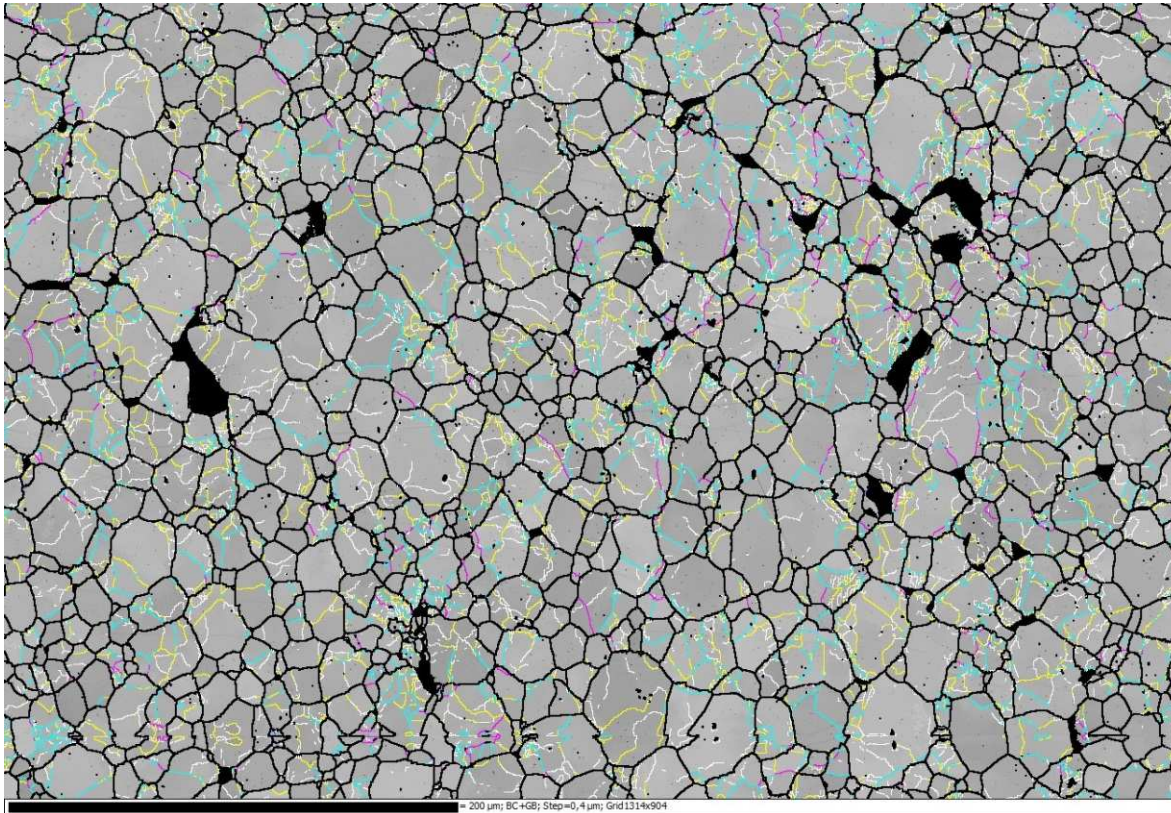
4.7 P1: lowest po2 (site 1)

- EBSD characterisation
- Fragmentation of grains into regions with small misorientations (between 0.25° and 3°)
- Linear fraction of small angle boundaries $>$ than in pellet tested under Ar/H₂



White: 0.25° / Yellow: 0.5° / Aqua : 1° / Pink: 3° / Black: 5° (non indexed pixels)

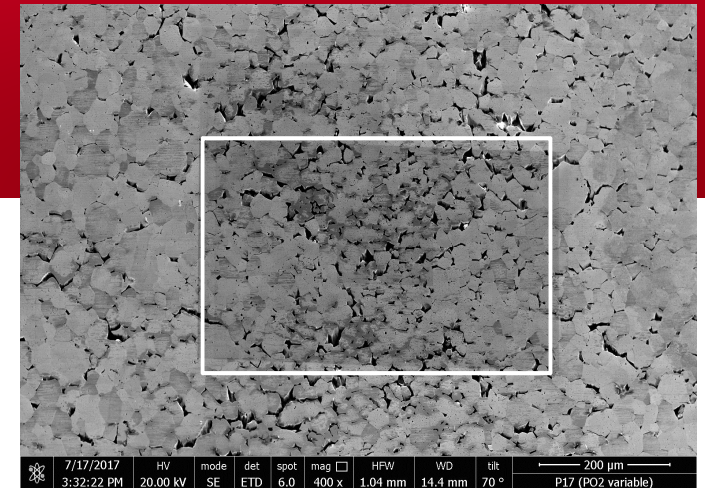
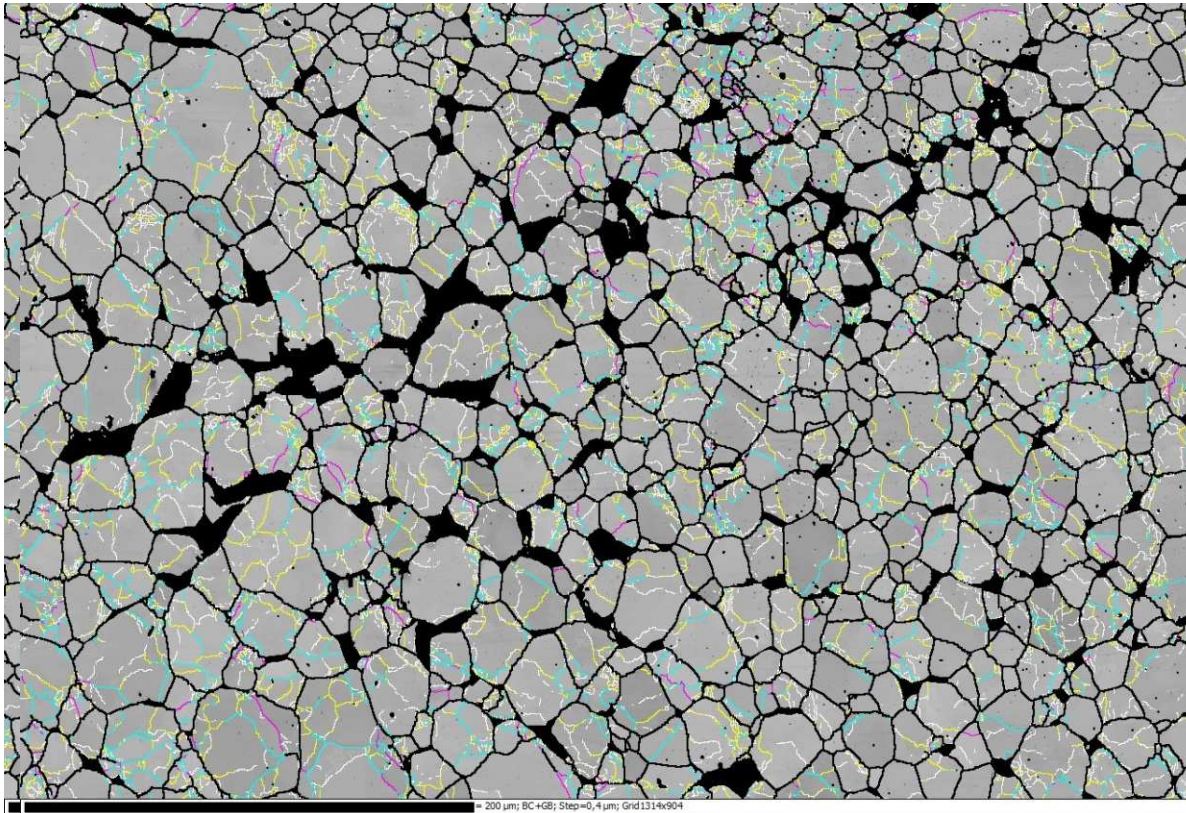
4.8 P1: lowest po2 (site 2)



White: 0.25° / Yellow: 0.5° / Aqua : 1° / Pink: 3° / Black: 5° (non indexed pixels)

- Void fraction increase related
 - to damage/strain localisation
 - Increasing incompatibility between grains of different orientations
- Confirms **macroscopic heterogeneity**

4.9 P3: highest po2 (site 2)



White: 0.25°/ Yellow: 0.5°/ Aqua : 1°/ Pink: 3°/ Black: 5°(non indexed pixels)

- **Damage localisation** and apparent incompatibilities between orientations **increase** with oxygen activity
- Possible sign that diffusion processes proceed at faster rates?

- A set of compression creep experiments carried out at different oxygen pressures in high stress regime: consistent with restoration creep
- Tests are reproducible
- Phenomenological analysis of primary and secondary creep – 4 parameter model satisfactory
- Backed up by 1 and 2 dimensional modelling
- uniaxial hypothesis is relevant for model identification
- Decrease of flow stress with p_{O_2} – consistent with cation diffusion assisted mechanism
- SEM Microstructural analysis reveals
 - Localisation of damage
 - Increased localisation and grain fragmentation with increased p_{O_2}

- Change conditions to different
 - Strain rates: identification of $\dot{\epsilon} \propto K p o_2^\alpha \sigma^n$
 - Temperatures: true activation energy
- Study purely Nabarro-Herring creep
- Systematic 2D simulation: correlation to microstructural study
- Look at different materials: SPS, single crystals and (U,Ce) mixed oxide
- *Relationship between macroscopic/local strains - fragmentation – oxygen activity remains to be understood*
- Parallel may be drawn with response to irradiation?



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Investigations Supporting MOX Fuel Licensing
in ESNII Prototype Reactors