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

DE LA RECHERCHE À L'INDUSTRIE

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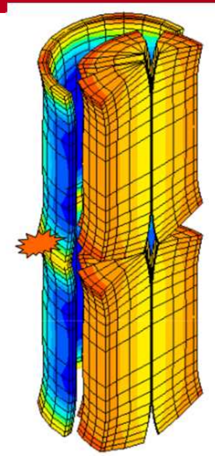
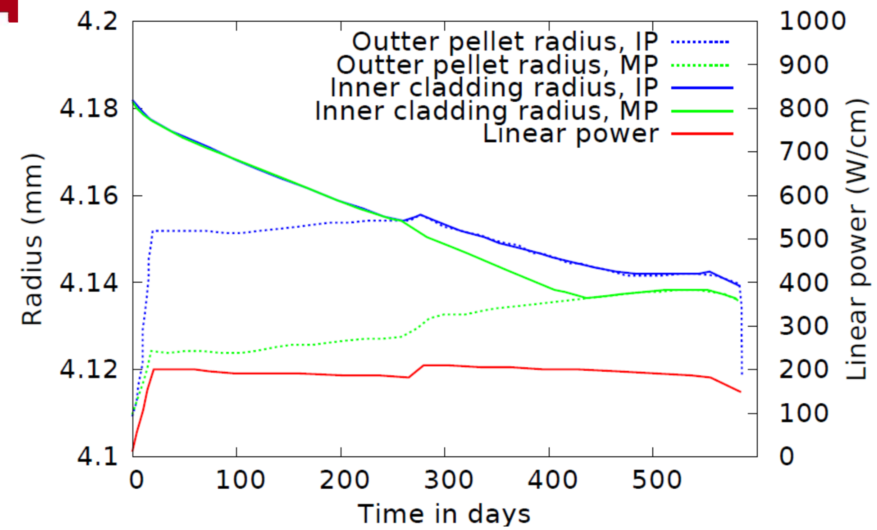
Outline of talk

- 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

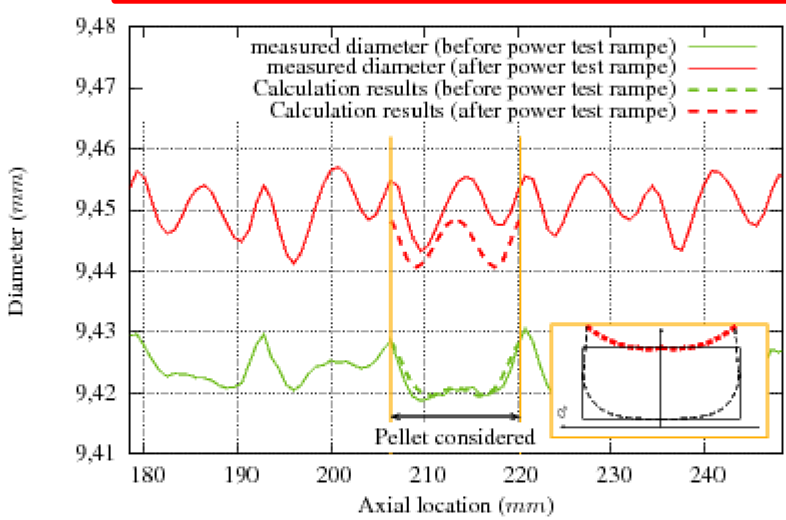
1.1 An essential modelling ingredient for fuel performance applications

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





Kinetics of formation of primary ridges & TM state of cladding are determined by radiation induced cree*p*



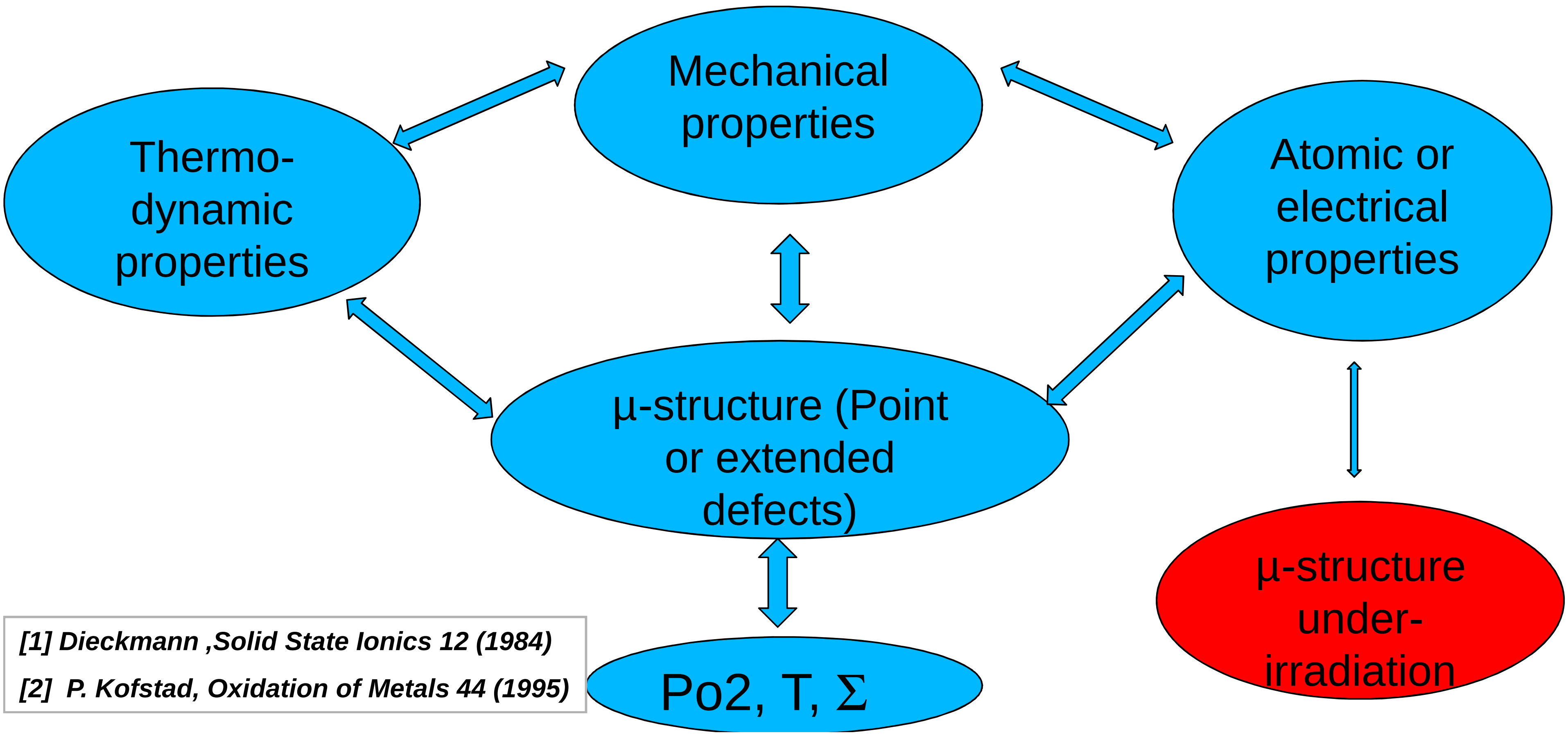
During power ramps, cladding stresses and strains controlled by thermal creep of pellet and cladding

[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…

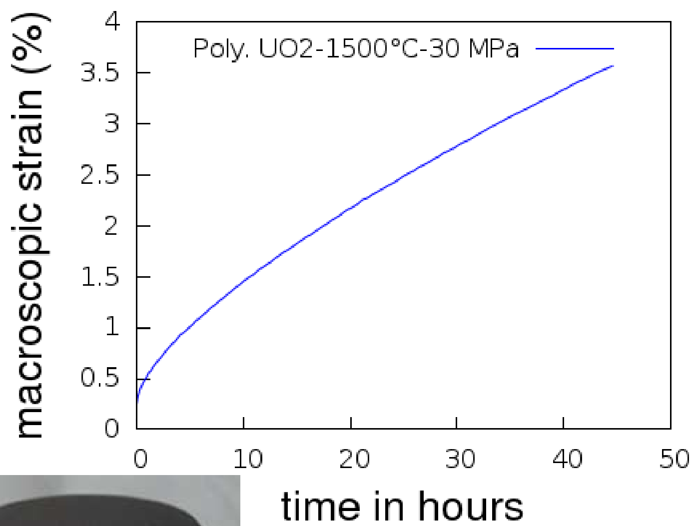


[1] Dieckmann ,Solid State Ionics 12 (1984)

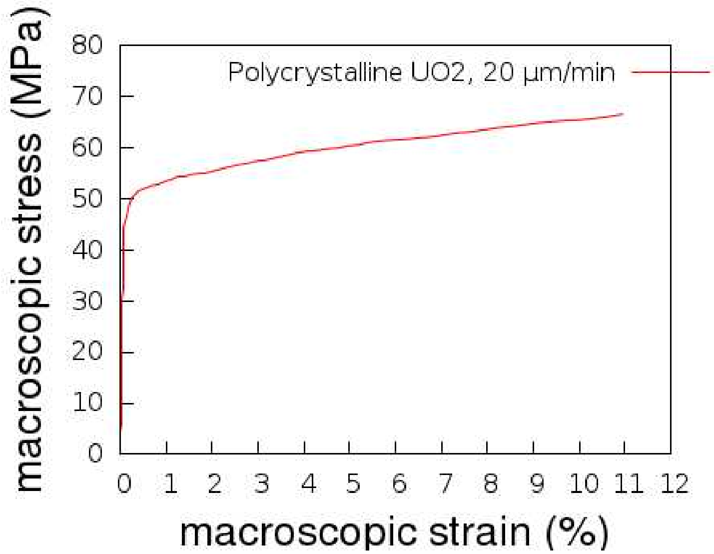
[2] P. Kofstad, Oxidation of Metals 44 (1995)

2.1 From macroscopic creep tests to mechanisms…

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



~10^{-7} s^{-1}



$2x10^{-5}$ s^{-1}

- Fuel behaviour applications:

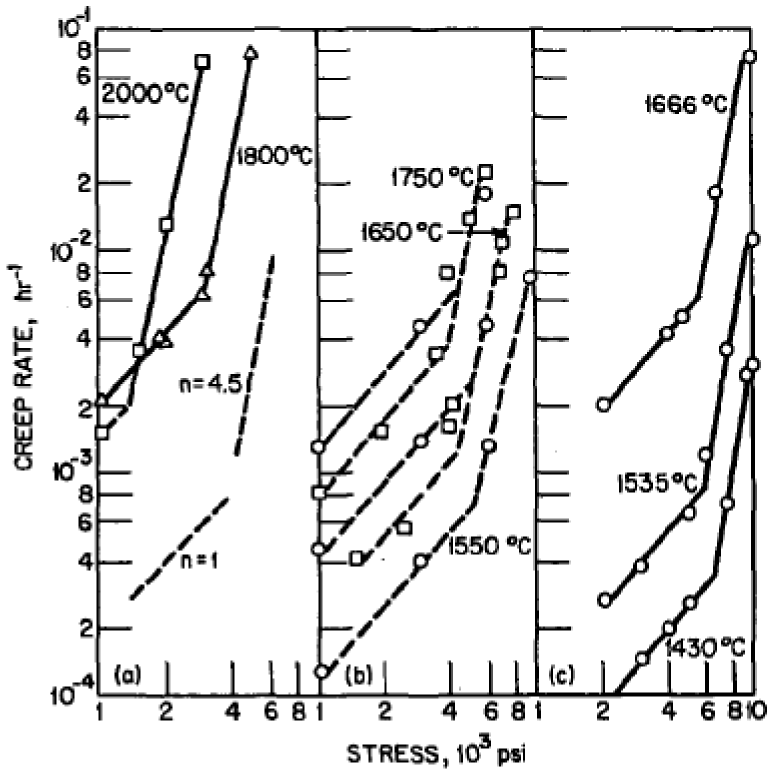
$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times Exp(-\frac{E}{kT})$$

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"

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

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



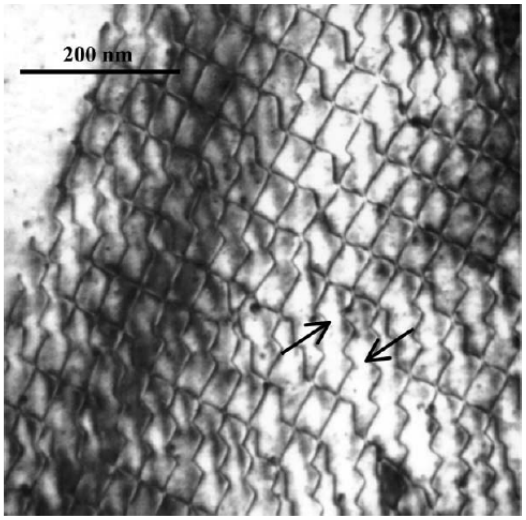
What physical meaning can be given to the activation energy?

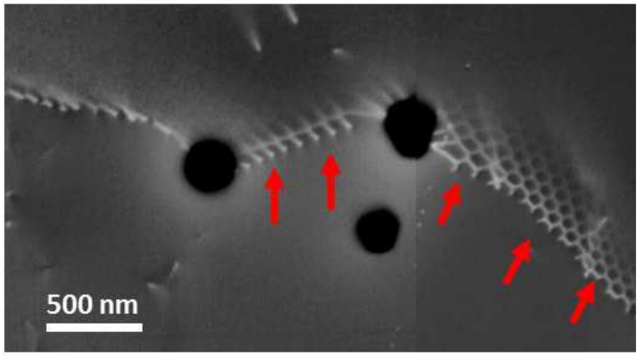
[1] M.S. Seltzer, JNM 34 (1970)
[2] Weertman JAP 26, XXXX
[3] C. Duguay, PhD, INSA Lyon (1998)
[4] C. Herring JAP 21, XXXX

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
  - ∝ 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]
  - ∝ D of slowest moving species

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





Ecci image: sub-grain boundary[5

[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 ⇒ strong dependence upon deviation from stoichiometry[1]
- More appropriate to interpret data f(po2)
  - po_2 is the true thermodynamic property[2,3,4]
  - Practical reasons: Dx ~ 2 10^{-3}, Δpo_2 : 4 orders of magnitude
  - Measured parameters are physically meaningful[2]
- One can expect the following

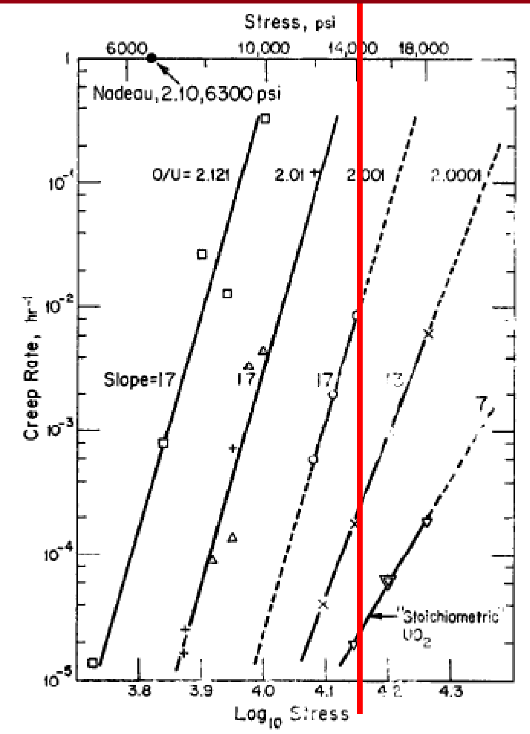


Fig. 2. Creep rate as a function of applied stress for UO_{2+x} single crystals tested in compression at 1100 °C. The compression data of Nadeau 20) for slip is included.

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

$$\dot{\epsilon} \sim K \times \sigma^n \times d^p \times f_U p_{O2}^{\alpha} Exp(-\frac{E_f}{kT}) \times D_{VU}^{o} Exp(-\frac{E_m}{kT})$$

α and Ef ⇒ characteristic of defect formation, Em and $D°_{VU}$ ⇒ characteristic of defect migration

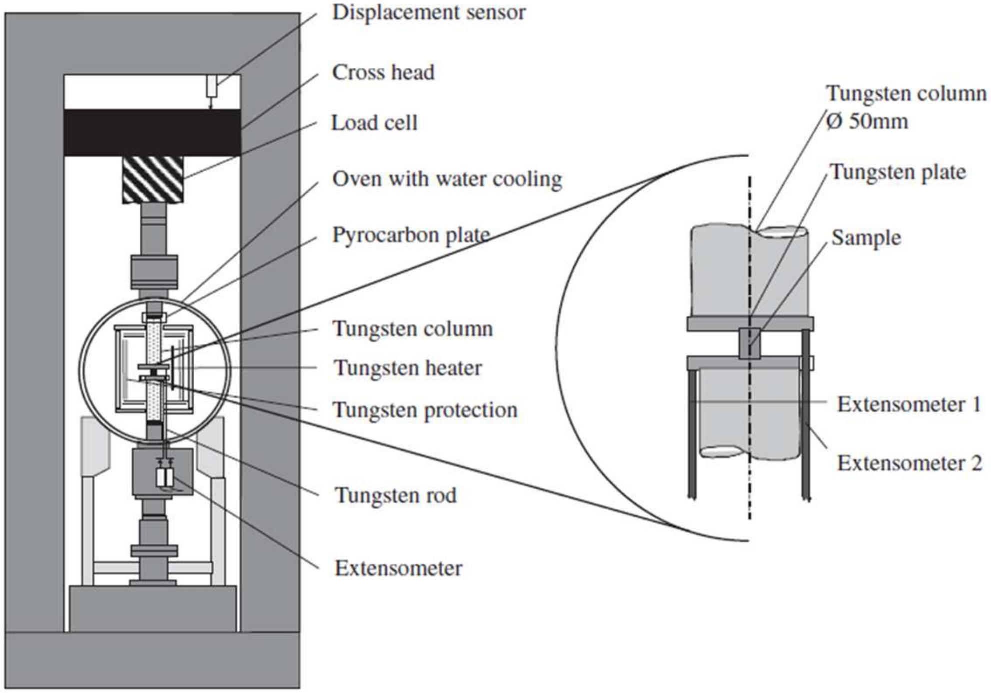
[1] Seltzer et al. J. Nucl. Mater (1972)

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

[3] Dorado et al. PRB 83, 2011

[4] Dorado et al. PRB 86, 2012

3.1 Creep tests under controlled atmosphere

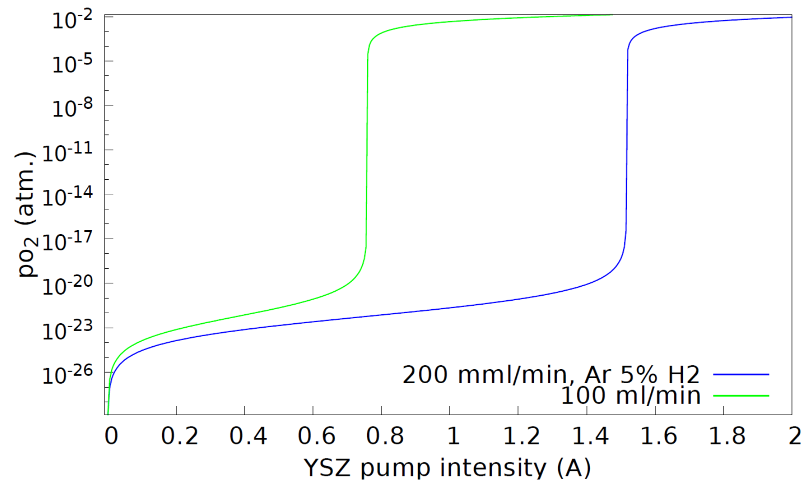


- Compression testing
  - 100 kN frame equipped with W ~1700℃ furnace & – *in situ* extensometers
  - Originally all tests carried out under Ar/H2
  - Roughly 50 l of "free volume"
- System equipped with oxygen and humidity probes, upstream and downstream

3.2 Atmosphere control

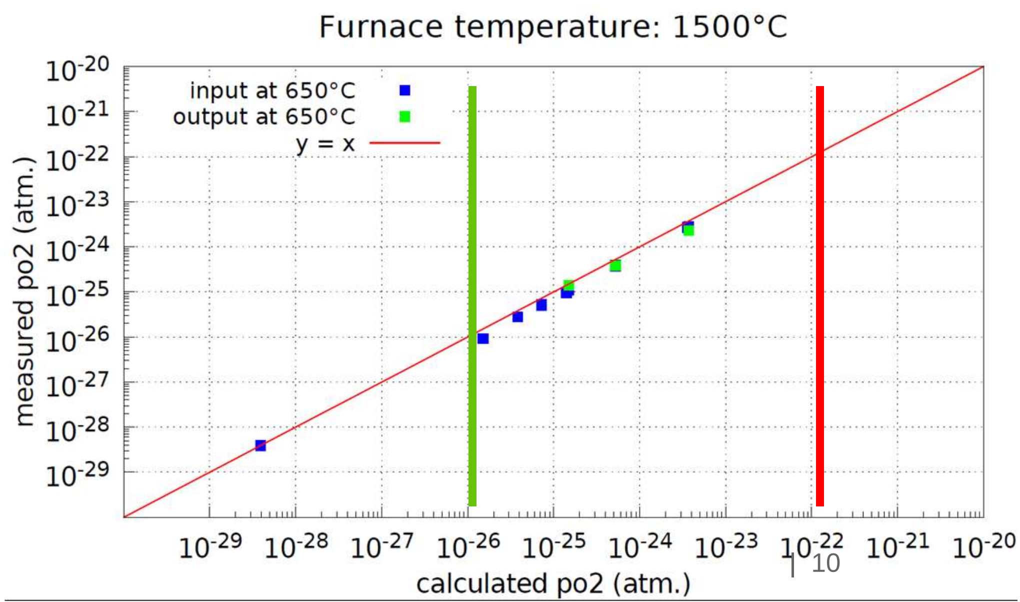
- Control based on buffering gas phase

$$H_2 + \frac{1}{2}O_2 \Leftrightarrow H_2O_{vap.}$$



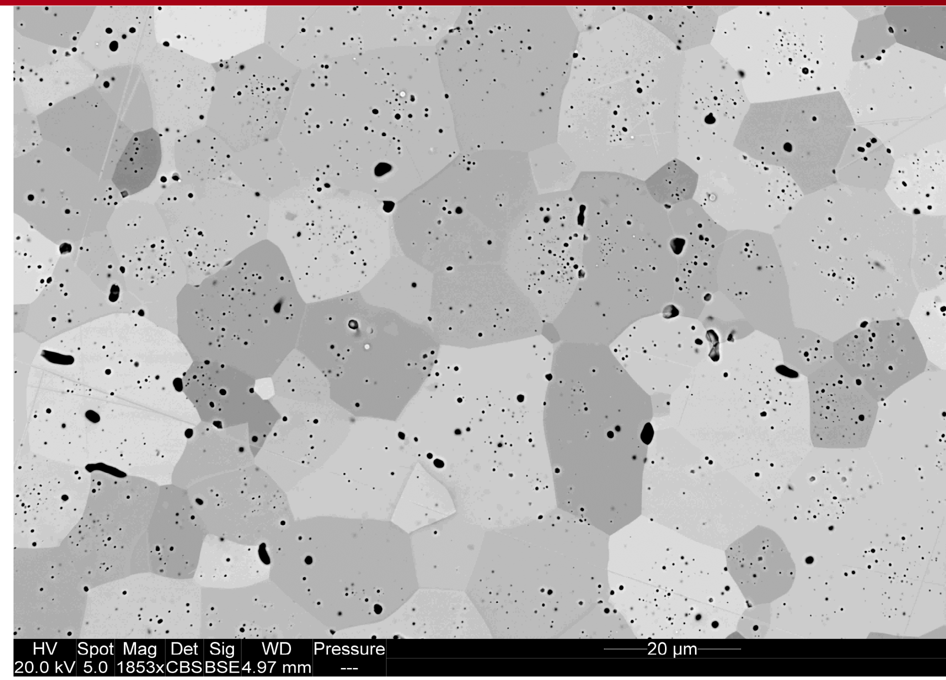
- Ar/H_2 carrier gas + humidification with YSZ oxygen pump
- Calculated values (know from Faraday's law [1]) ($n_{O2} \propto i$)

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



3.4 Materials used and tests run

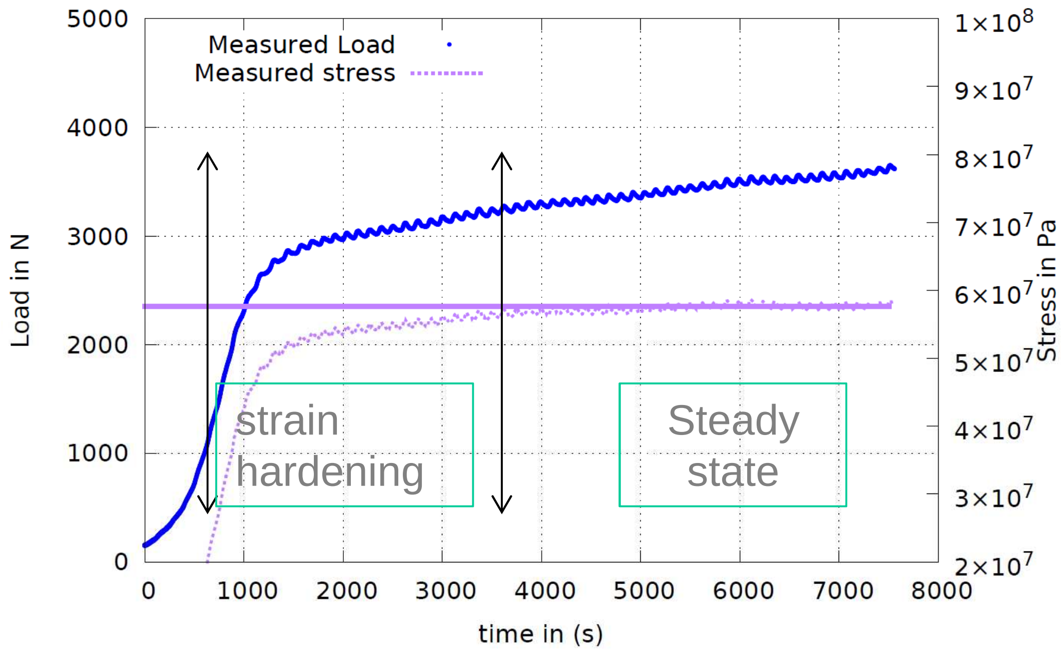
- All tests run at 1500℃ on polycrystalline samples
- 16.1 mm +/-0.2 mm height, 8.190 mm diameter +/-0.010 mm ⇒ limiting 2D effects
- Grain size ~ 14 µm – pore fraction 1.8 +/-0.1 % (high density)
- Final strain between 13 and 14%



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)

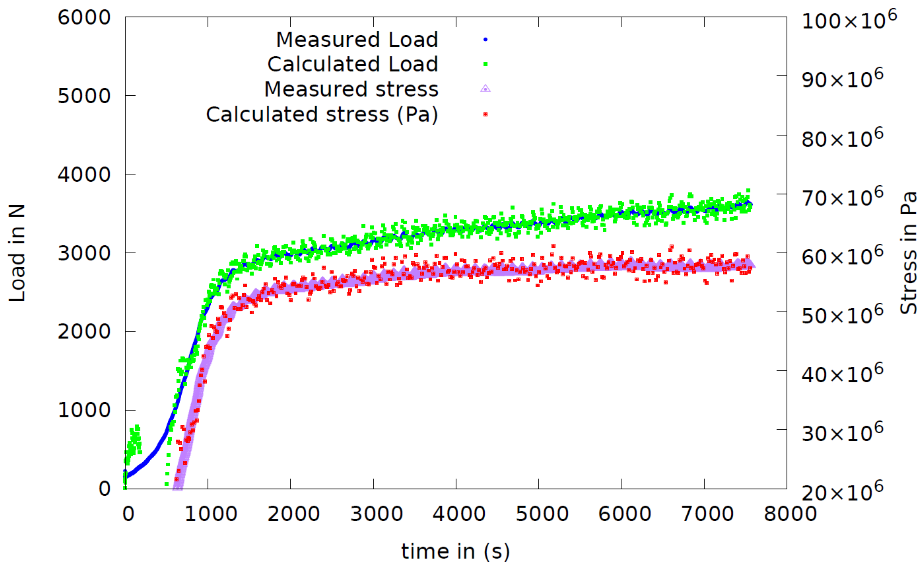
Uniaxial temptation is great

macroscopic hardening $\Leftrightarrow$ primary creep & increase in section

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

- Barrel-shaped pellet ⇒ 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 ⇒ primary creep[5]



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

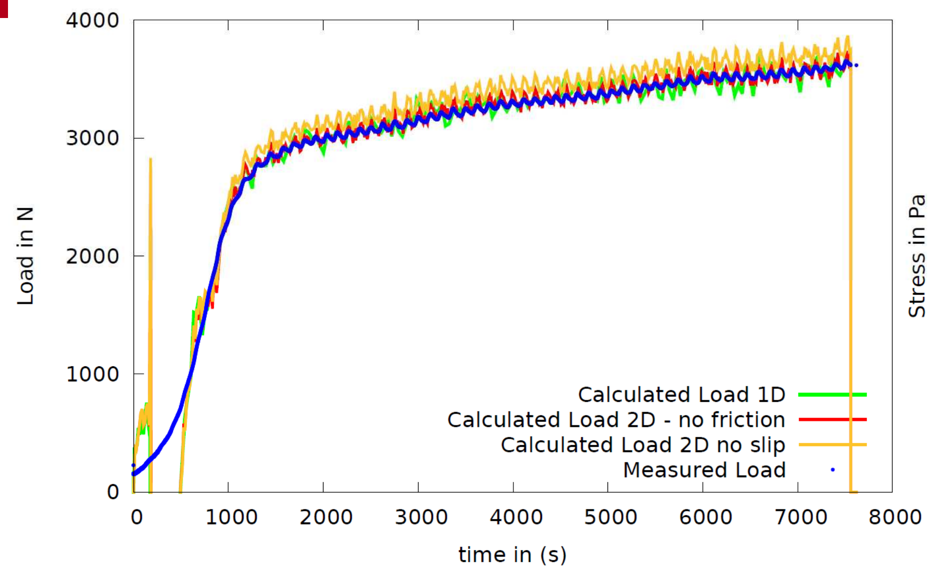
- 1D Model reproduces data
- Even fluctuations (contained in displacement load) are reproduced
- Correlated parameters

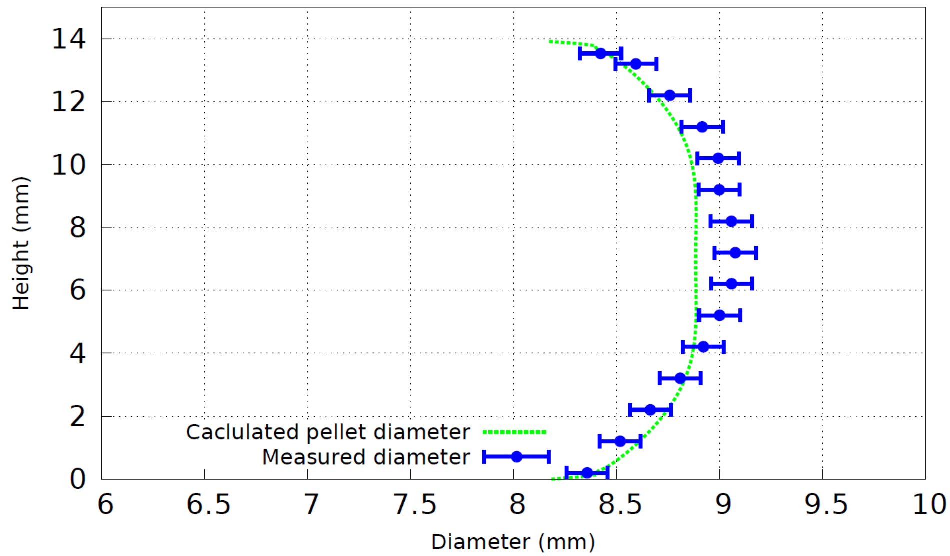
⇒ Confirms uniaxiality

[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
  - o assuming no friction
  - o assuming infinite friction
- Only slight change in parameters required
- 2D model reproduces profile ~
- Uniaxial approach justified



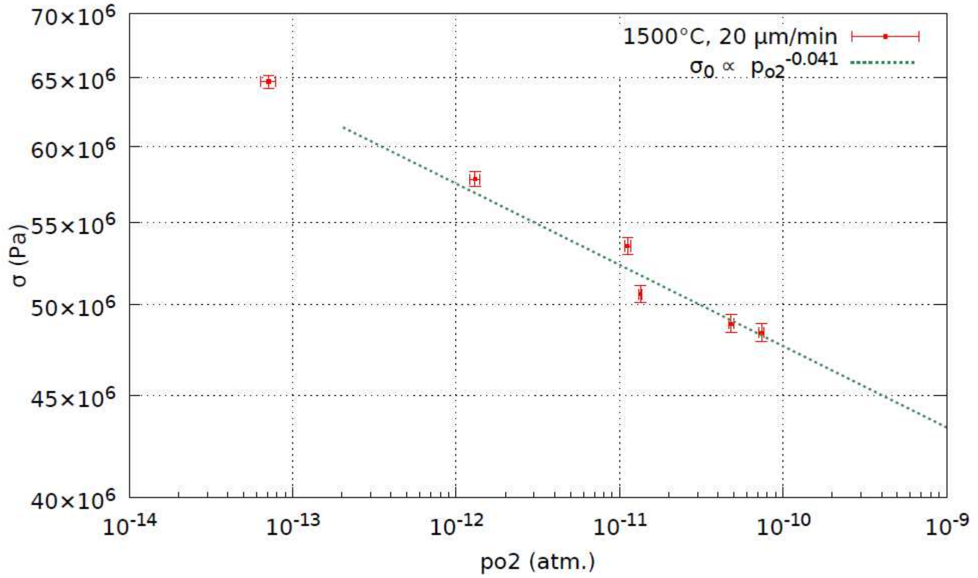


- 1D Model reproduces data
- Even fluctuations (contained in displacement load) are reproduced
- Correlated parameters

4.4 Result of 1D analysis

- If X<<σ, analysis is straightforward and
- Plotting $Log(\sigma) = f(Log(po2)) \Rightarrow -\alpha/n$

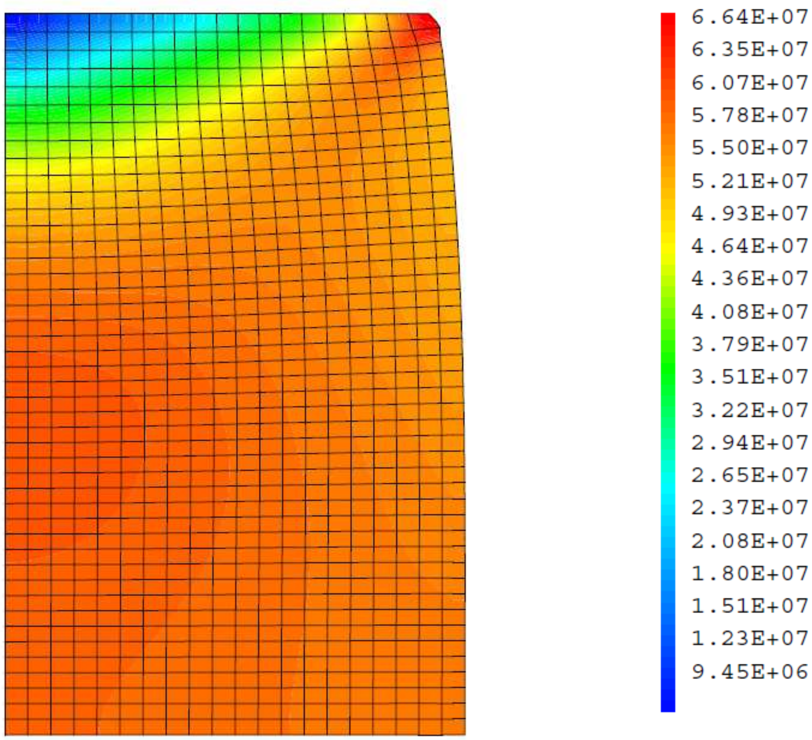
$$\dot{\varepsilon} \propto K {po_2}^{\alpha} \sigma^{n}$$

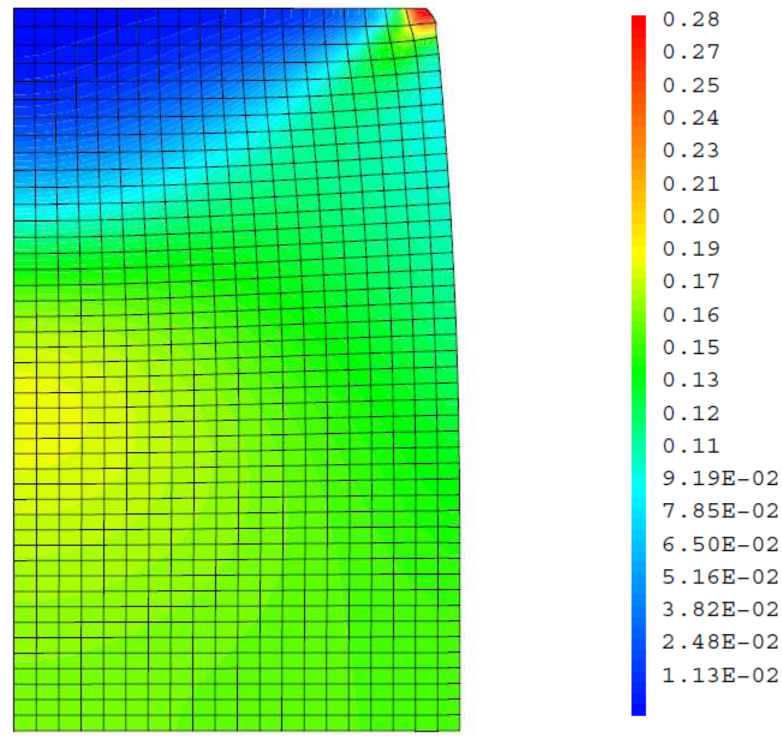


- Data points lie in ~ straight line
- $-\alpha/n$ ~ -0.041 ⇒ if n~ 4, α ~ 1/6
- Decreasing f(po2) ⇒ consistent with α > 0 and increasing [Vu] ~ $p_{o2}^{1/6}$
- n can be identified by changing conditions

4.5 2D Calculations and relationship to microstructure

- 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]

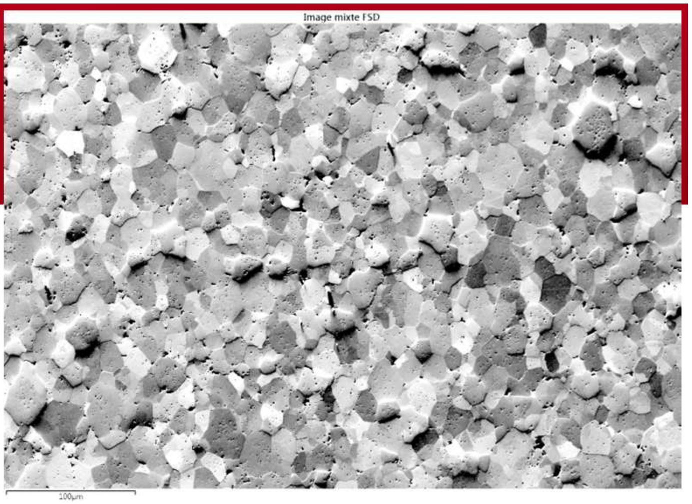
[1] Ben Saada et al., Material Characterization 133 (2017) *[2] Ndiaye, PhD, Université de Grenoble (2006)*

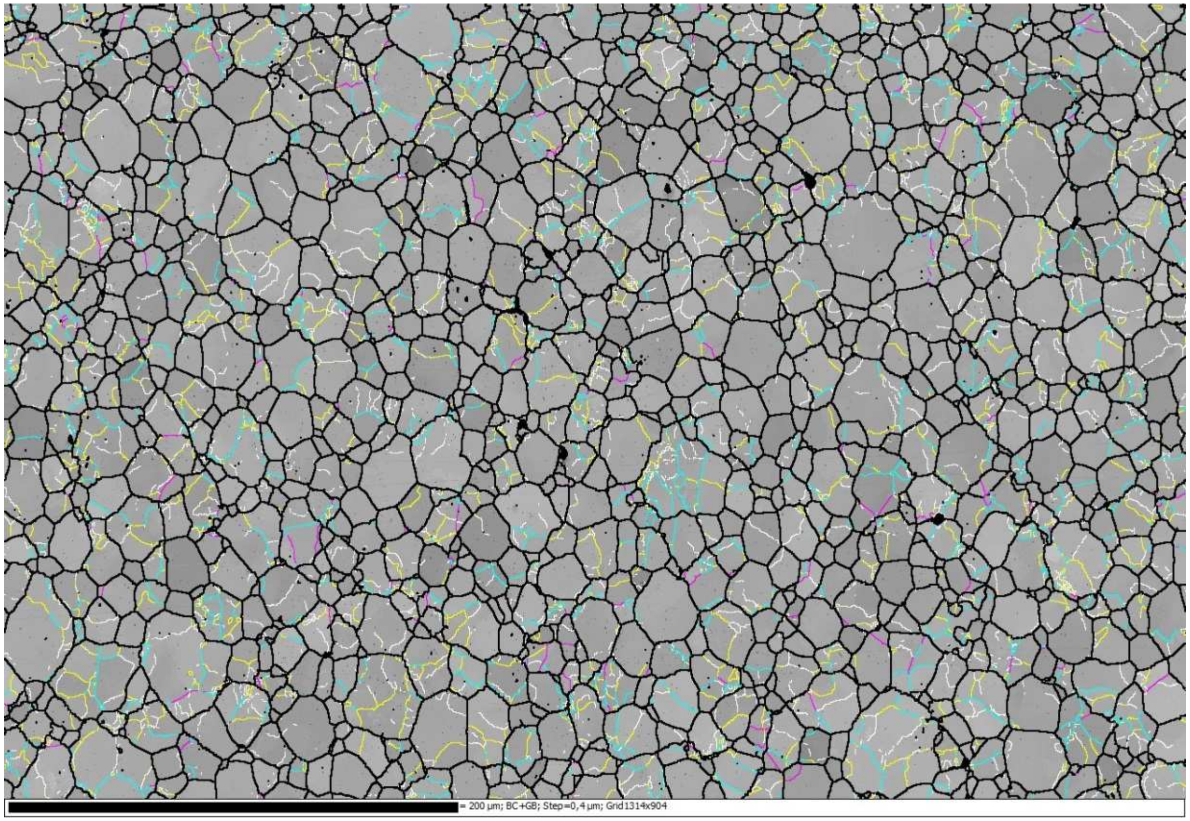
4.6 Preliminary characterization results

- P1, P2, P3 ($5\ 10^{-26}$, $5\ 10^{-25}$, $2\ 10^{-24}$): identical macroscopic strain
- P1: Damage localization
- Formation of pockets of voids, located “away from” the plane of symmetry
- With increasing po2
  - Volume fraction of pockets increases
  - Associated grain misorientation increases

4.7 P1: lowest po2 (site 1)

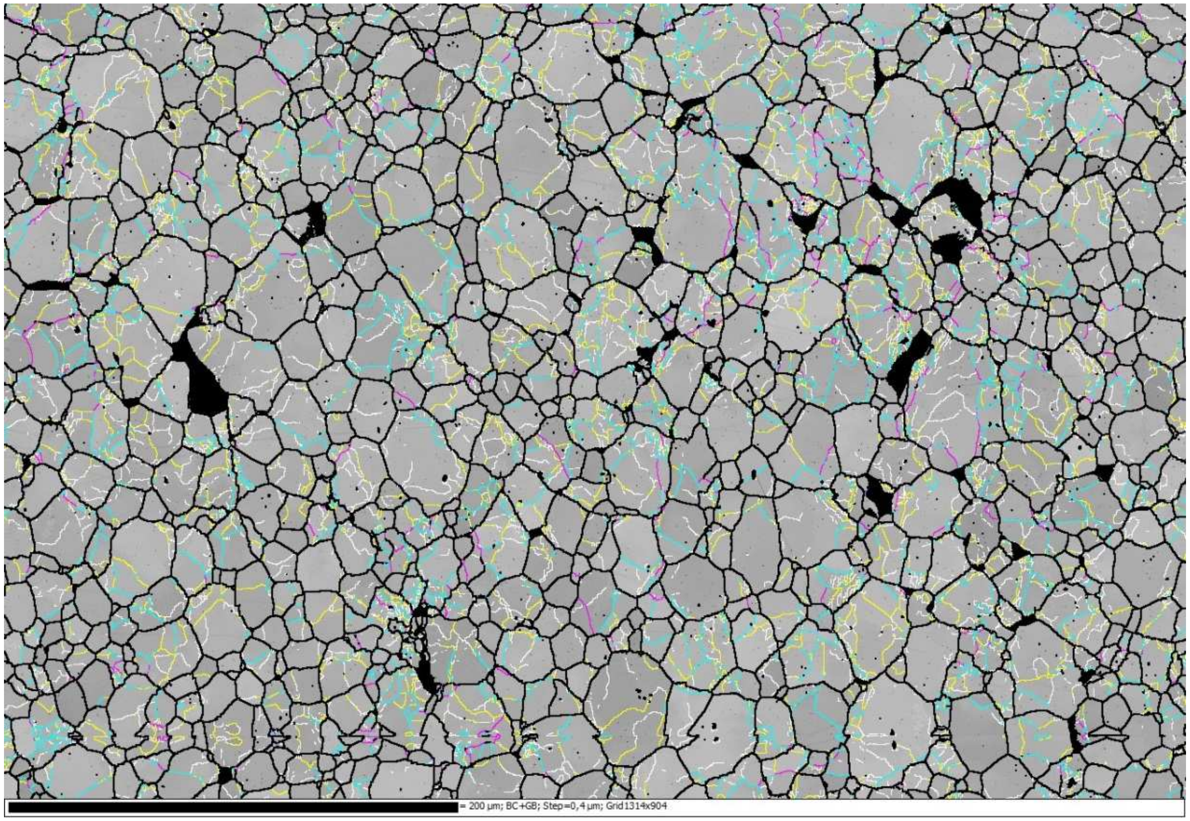
- EBSD characterisation
- Fragmentation of grains into regions with small misorientations (between 0.25° and 3°)
- Linear fraction of small angle boundaries > than in pellet tested under Ar/H2

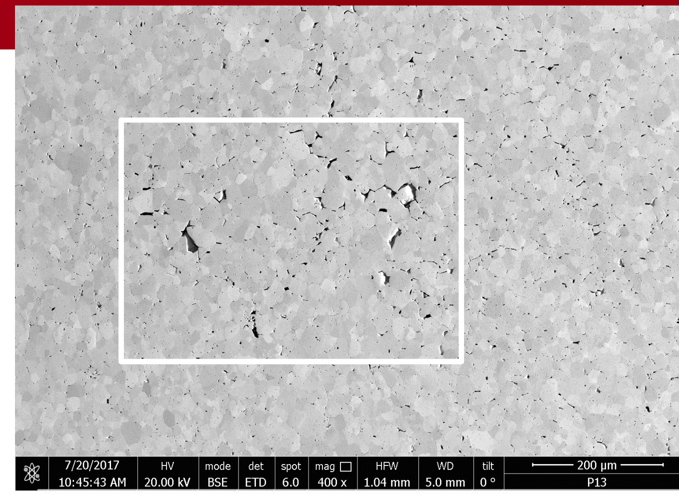


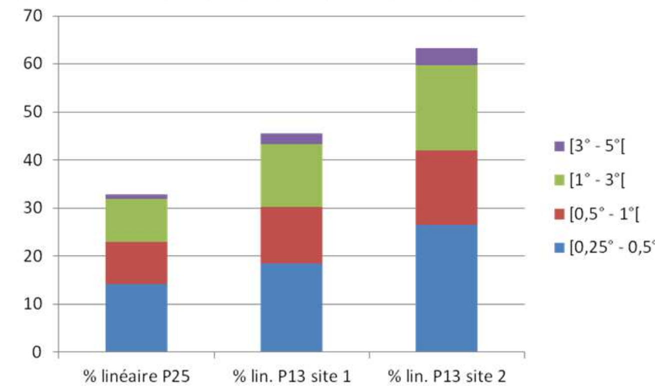


White: 0.25°/ Yellow: 0.5°/ Aqua : 1°/ Pink: 3°/ Bla ck: 5°(non indexed pixels)

4.8 P1: lowest po2 (site 2)



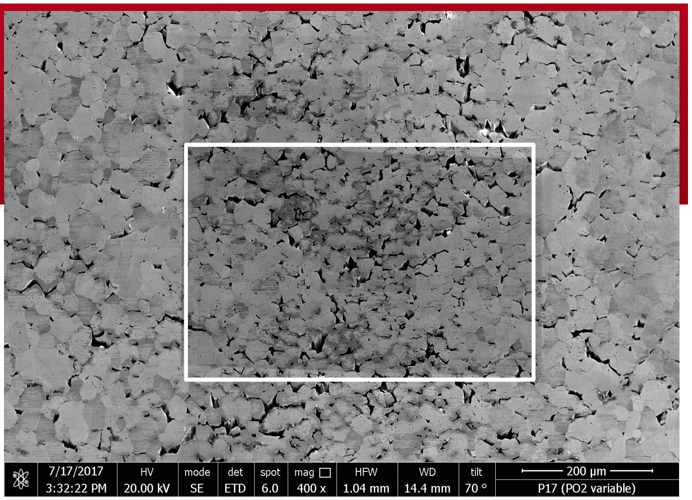


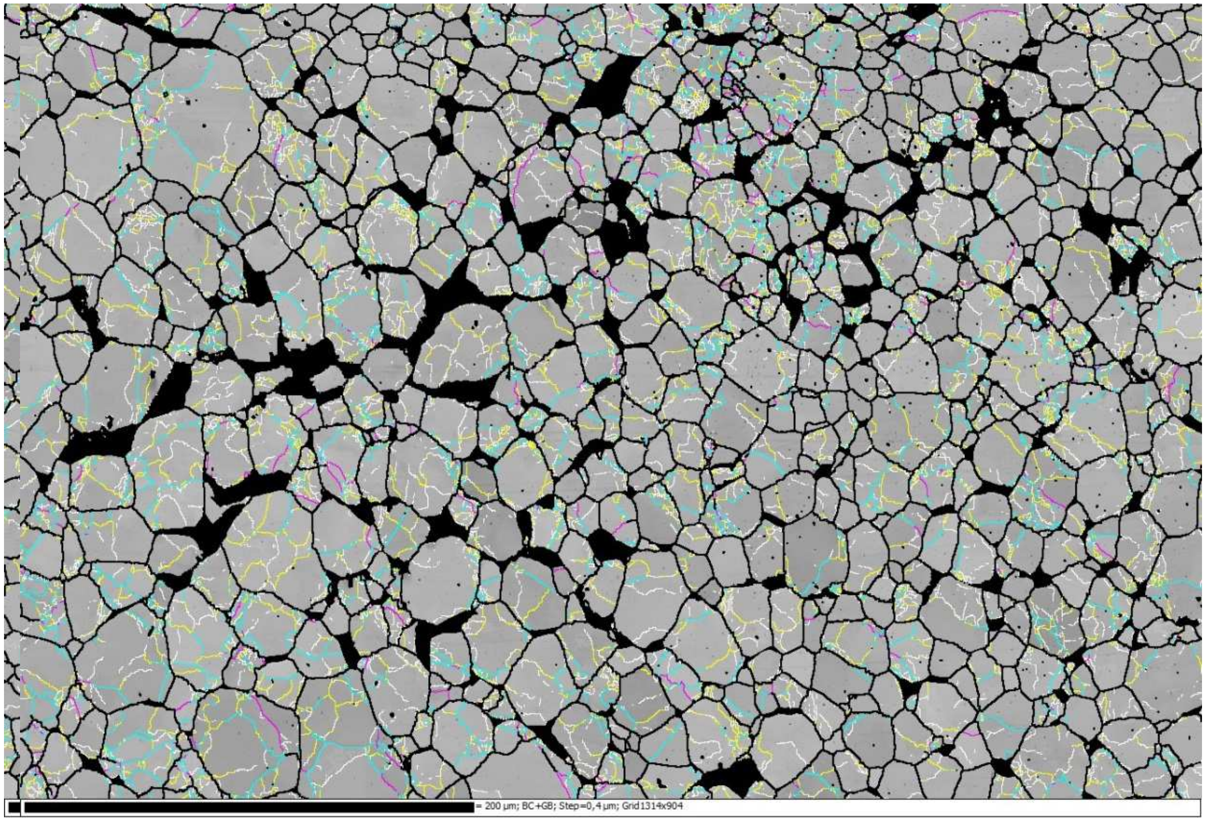


White: 0.25°/ Yellow: 0.5°/ Aqua : 1°/ Pink: 3°/ Black: 5°(non indexed pixels)

- 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)





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

5.1 Conclusions

- A set of compression creep experiments carried out at different oxygen pressures in high stress regime: consistent with restauration 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 po2 – consistent with cation diffusion assisted mechanism
- SEM Microstructural analysis reveals
  - Localisation of damage
  - Increased localisation and grain fragmentation with increased po2

5.2 Prospects

- Change conditions to different
  - Strain rates: identification of $\dot{\varepsilon} \propto K {po_2}^{\alpha} \sigma^{n}$
  - 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?

Acknowledgements

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INSPYRE

Investigations Supporting MOX Fuel Licensing in ESNII Prototype Reactors