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# Long-term storage behaviour of spent nuclear fuel simulated by accelerated radiation damage with $^{238}\text{Pu}$ -doped $\text{UO}_2$

E. De Bona <sup>a,b</sup>, M. Cologna <sup>a</sup>, T. Wiss <sup>a</sup>, R.J.M. Konings <sup>a</sup>, G. Baldinozzi <sup>b</sup>,

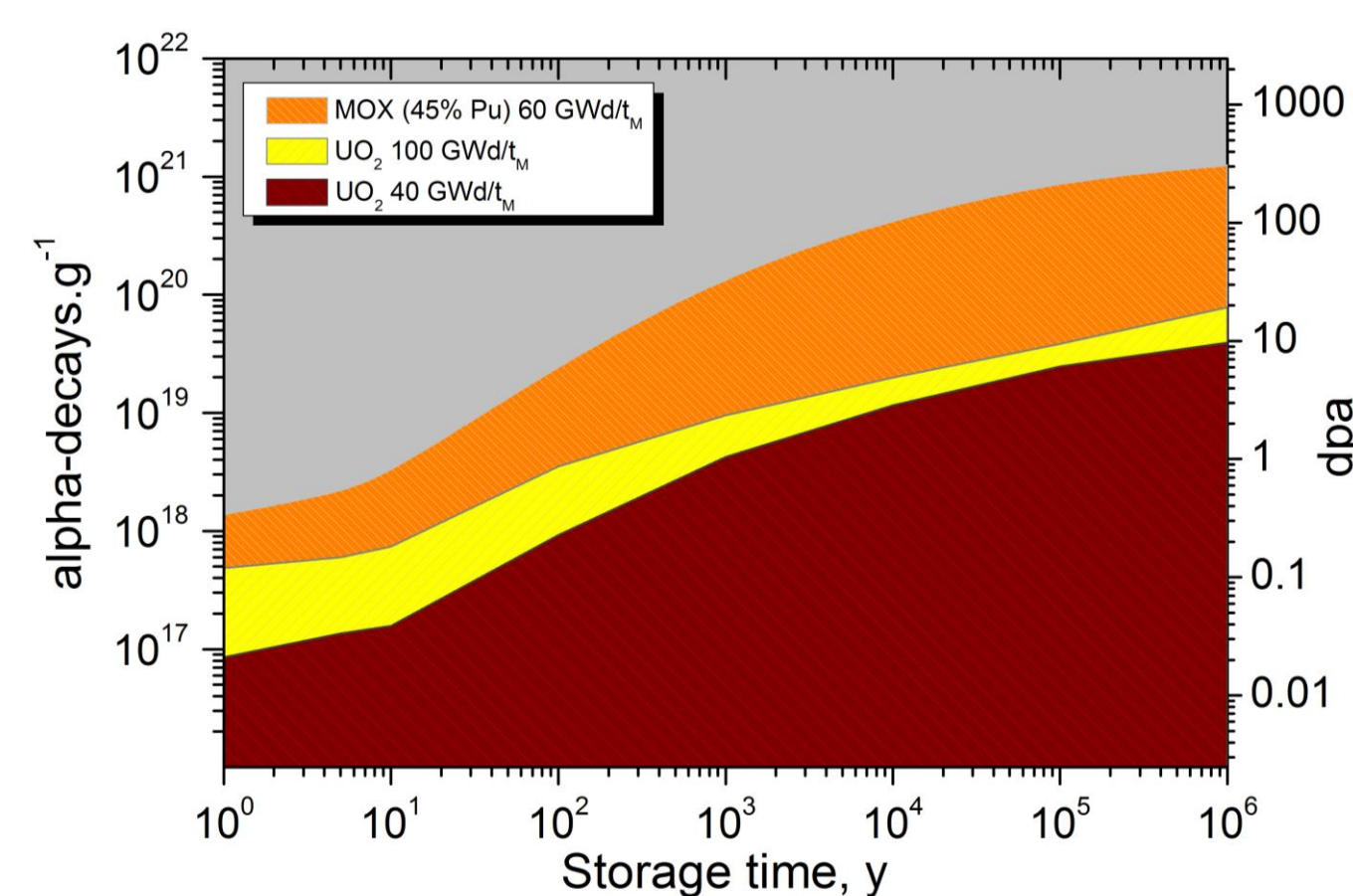
<sup>a</sup>JRC-Karlsruhe, Eggenstein-Leopoldshafen (Karlsruhe), 76344, Germany

<sup>b</sup>CentraleSupélec, Gif-sur-Yvette (Paris), F-91190, France

## Context

The radioactivity of the spent nuclear fuel is dominated by the  $\alpha$ -activity of the minor actinides generated during operational time and included within the  $\text{UO}_2$  matrix.

The accumulation of  $\alpha$ -damage and radiogenic He (shown in the graph) results in a modification of the microstructure and properties of the material, which has to be known to accurately predict the long term behaviour of the stored spent fuel.



## Experiment

- **Samples design:** bibliographic research of self-irradiation saturation values, choice of suitable compositions to reach saturation within PhD timeframe
- **Samples preparation:** powder synthesis, pressing and sintering, achievement of a  $\text{PuO}_2$  and  $\text{UO}_2$  solid solution
- **Fresh pellets characterization**
- **Periodic characterization:** 3 different storage temperatures (room T, wet storage T  $\sim 200^\circ\text{C}$ , liquid  $\text{N}_2$  T  $\sim -200^\circ\text{C}$ ), monitoring of the degradation of the thermophysical properties and microstructural evolution of LWR spent fuel due to self-irradiation and helium formation

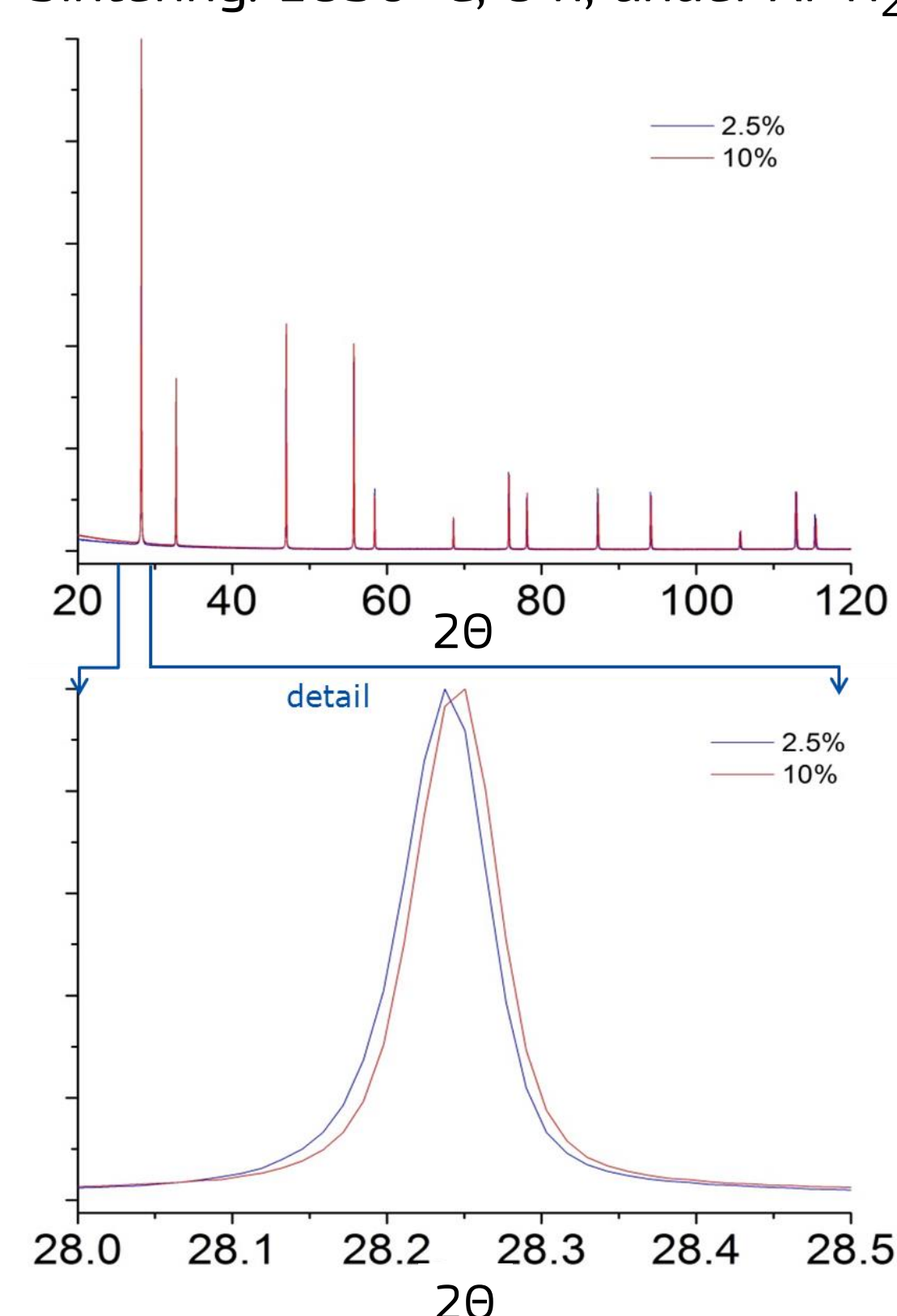
## Samples preparation

### Powder synthesis:

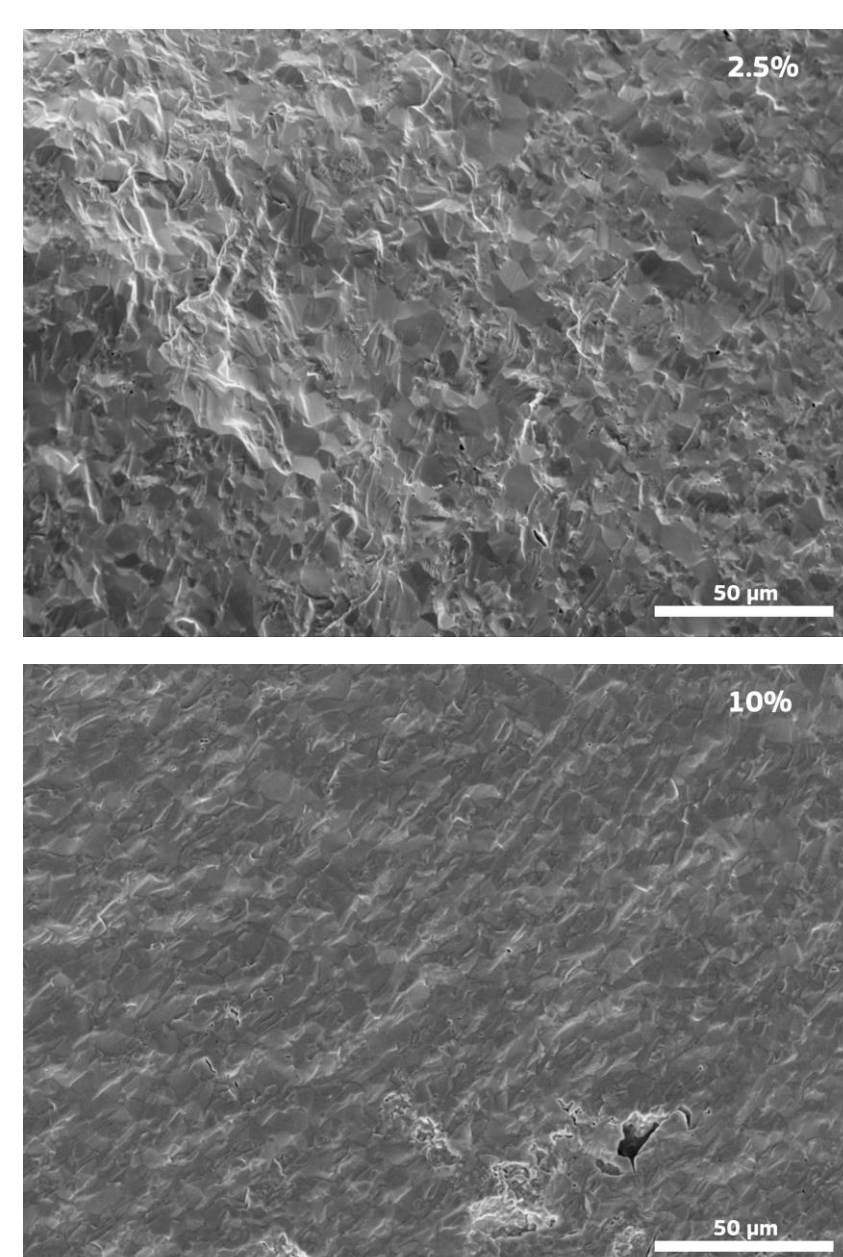
1. Dissolution of the Pu powder using  $\text{HNO}_3$  and HF
2. Separation in two batches and addition of the correct amount of U solution
3. Precipitation with ammonia addition
4. Filtration with paper filters
5. Pre-calcination ( $400^\circ\text{C}$ ) under air to burn the filters away
6. Calcination ( $700^\circ\text{C}$ ) under  $\text{Ar-H}_2$

### Powder pressing and sintering:

Pressing:  $\phi = 5\text{ mm}$ ;  $F = 14.5\text{ kN}$ ; resulting  $p = 738\text{ Mpa}$   
Sintering:  $1650^\circ\text{C}$ , 6 h, under  $\text{Ar-H}_2$



Two compositions were chosen:  
- **2.5wt Pu**, for the evolution of hardness and thermal conductivity in particular  
- **10wt Pu**, for those properties which degrade more slowly



Comparison between the XRD patterns of the two batches. Lattice parameter in good agreement with the Vegard Law for solid solutions.

## The European Commission's science and knowledge service

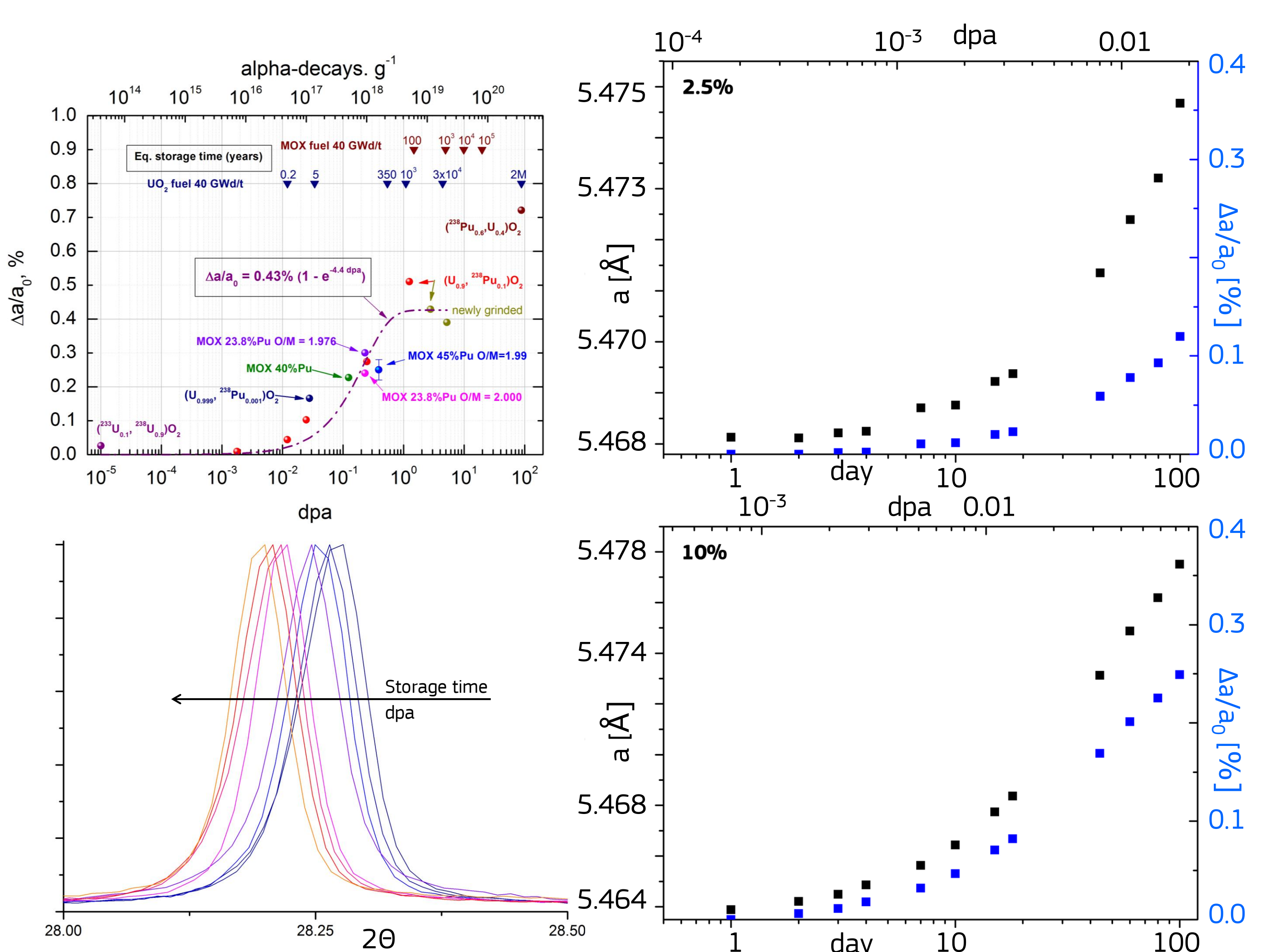
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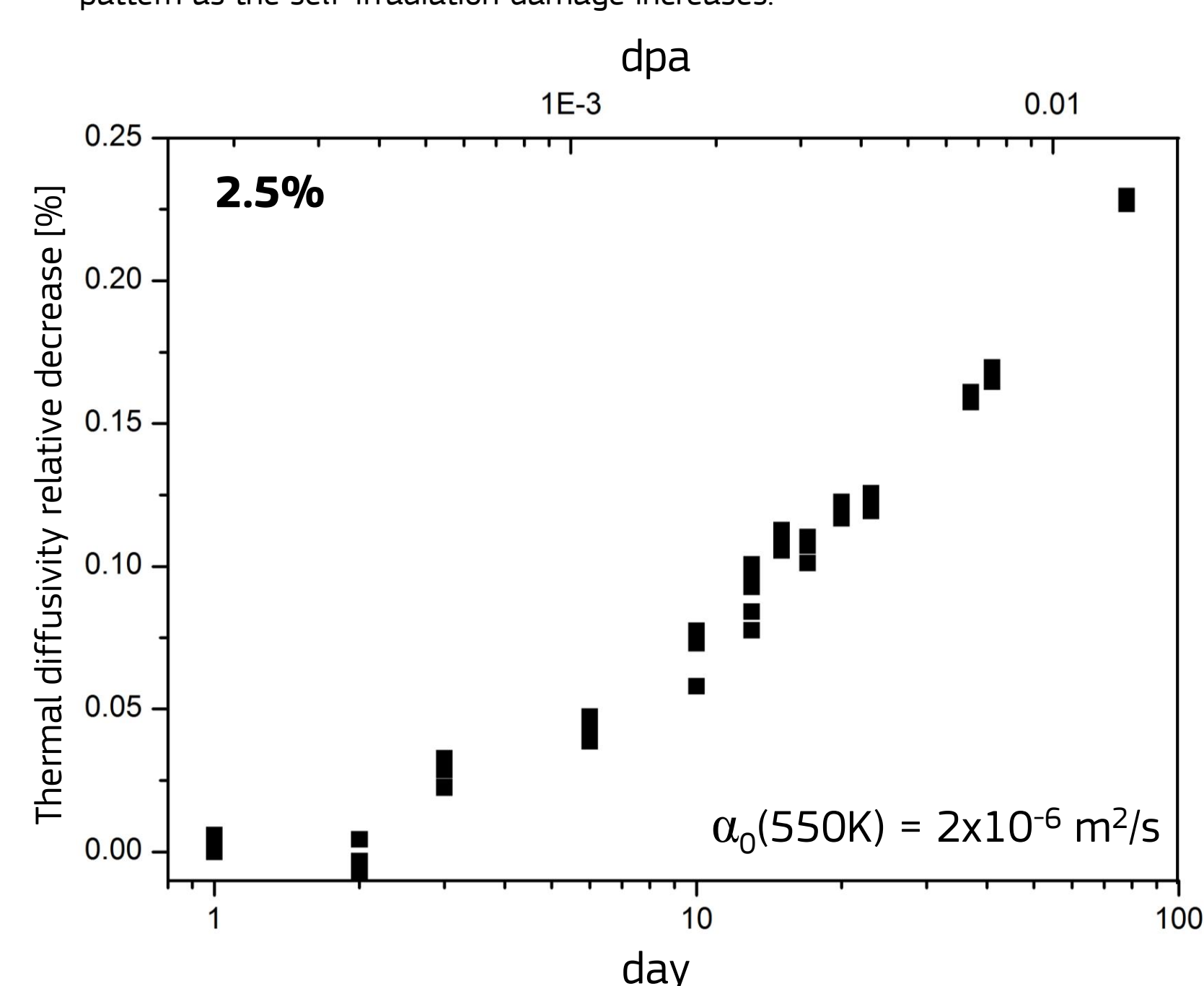
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## Periodic characterization – room T batch

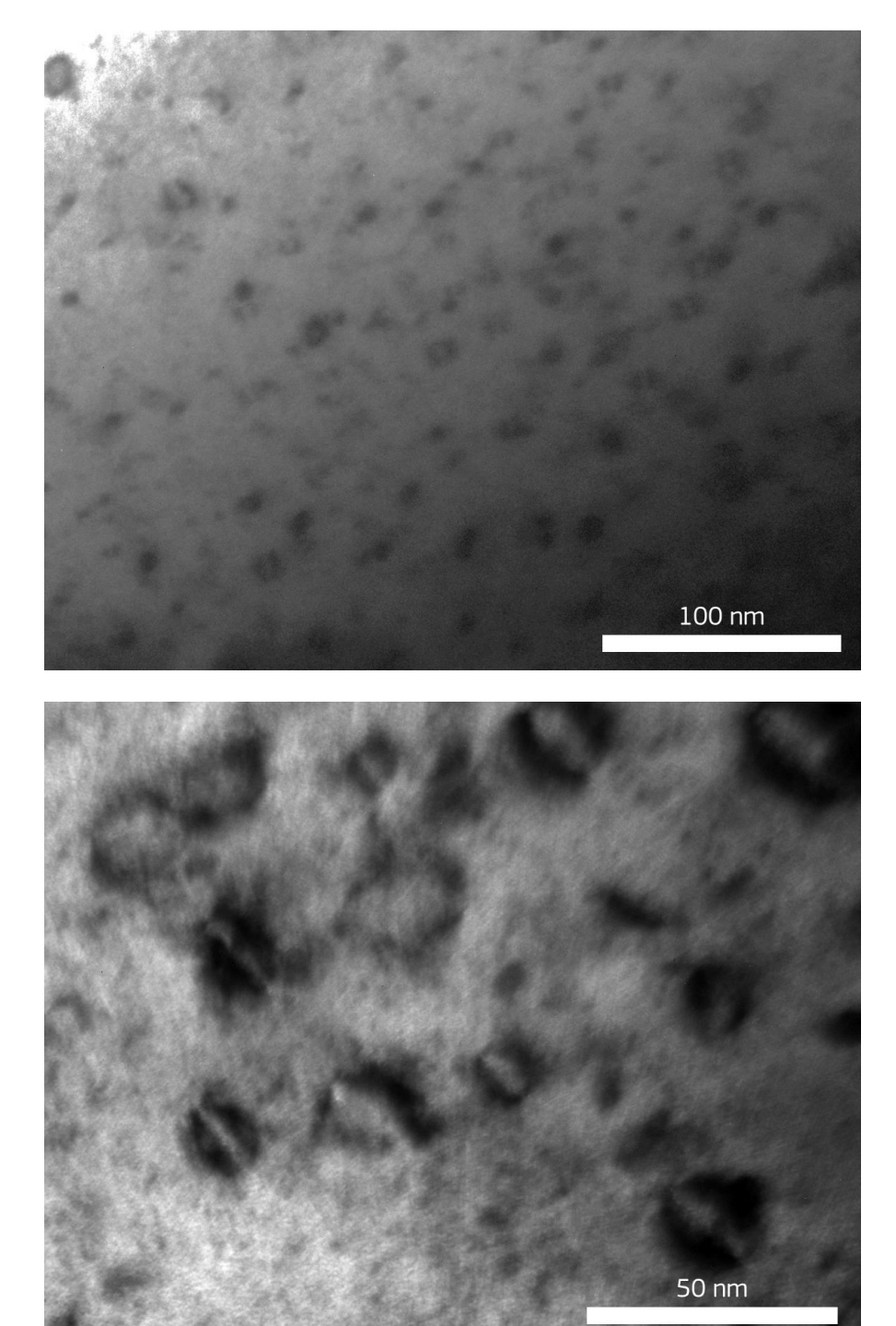
After annealing of defects (4h at  $1250^\circ\text{C}$  under  $\text{Ar-H}_2$ ), characterization started for the room T batch.



**First XRD results:** increase of the lattice parameter as function of the storage day and dpa. Shift of the first peak of the XRD pattern as the self-irradiation damage increases.



Left: **thermal diffusivity** degradation, measured at 550K by Laser Flash Analysis (LAF)  
Right: dislocation loops observed by **TEM** investigation on a 10%wt doped sample



## Conclusions

- **(U,Pu)O<sub>2</sub>** disks successfully sintered (geometrical density of 94-96%TD), composition measured by XRD in good agreement with the Vegard Law for solid solutions
- Started the characterization of the room T batch (XRD, LAF, TEM, but also RAMAN, EXAFS, NMR and DSC, not shown here)

## Future steps

- Periodic indentation and KEMS (Knudsen cell Effusion Mass Spectrometry, i.e. He thermal desorption) for room T batch
- Periodic characterization of samples stored at  $200^\circ\text{C}$  and  $-200^\circ\text{C}$

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Nuclear Safety and Security Directorate [Emanuele.DE-BONA@ec.europa.eu](mailto:Emanuele.DE-BONA@ec.europa.eu)