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Long-term storage behaviour of spent nuclear fuel simulated by accelerated radiation damage with ^{238}Pu -doped UO_2

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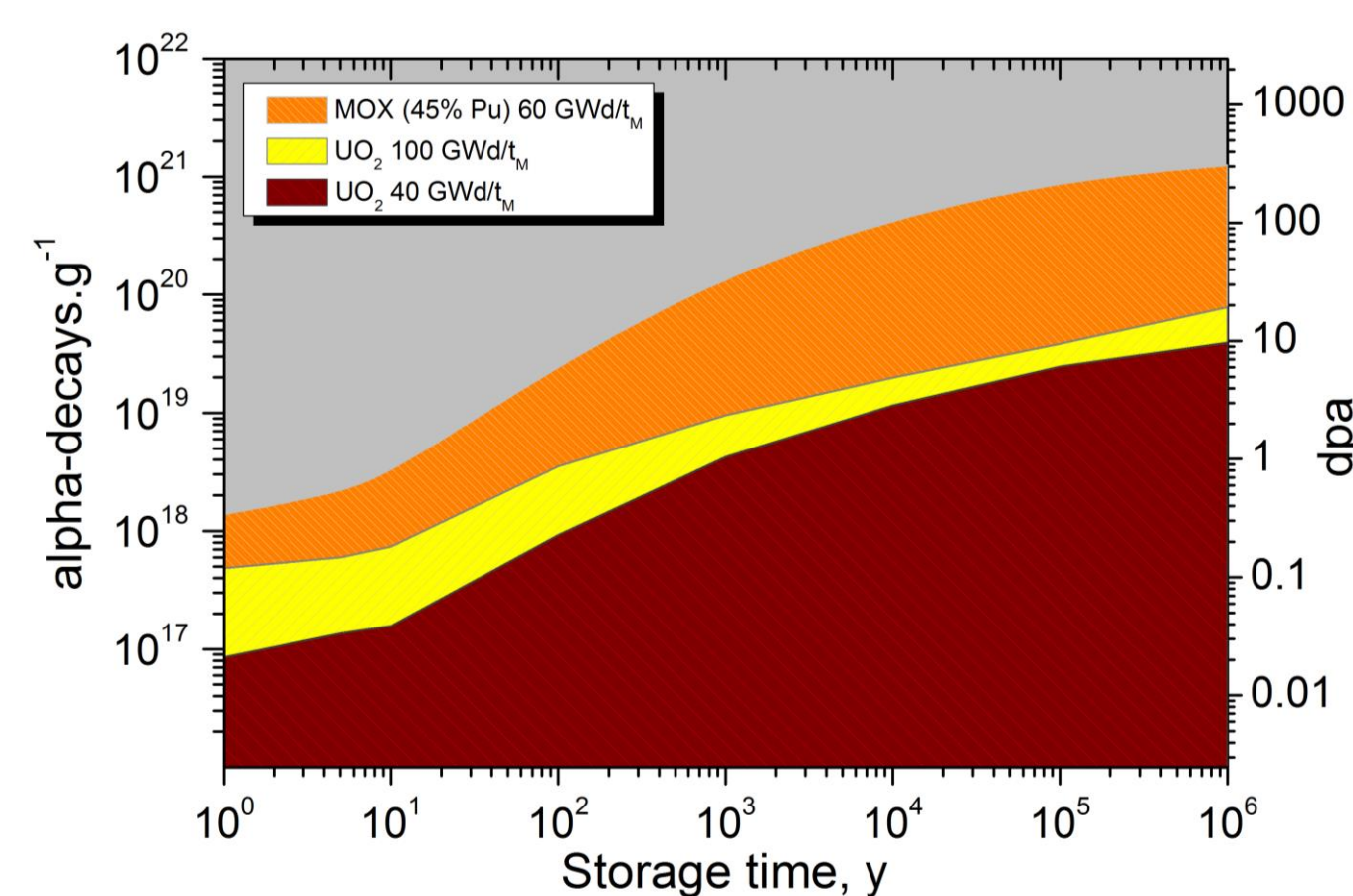
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Context

The radioactivity of the spent nuclear fuel is dominated by the α -activity of the minor actinides generated during operational time and included within the UO_2 matrix.

The accumulation of α -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 PuO_2 and UO_2 solid solution
- **Fresh pellets characterization**
- **Periodic characterization:** 3 different storage temperatures (room T, wet storage T $\sim 200^\circ\text{C}$, liquid 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

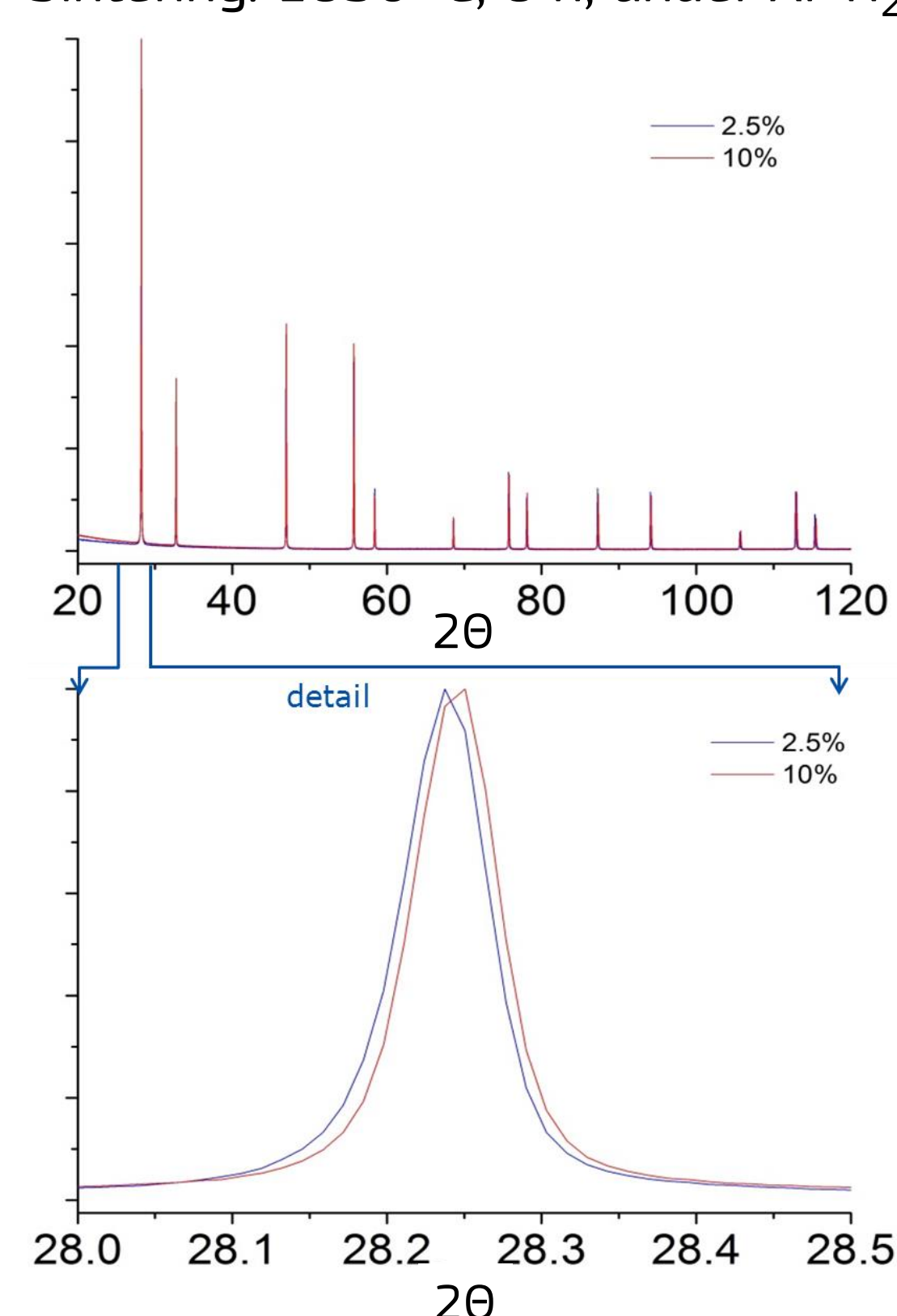
Samples preparation

Powder synthesis:

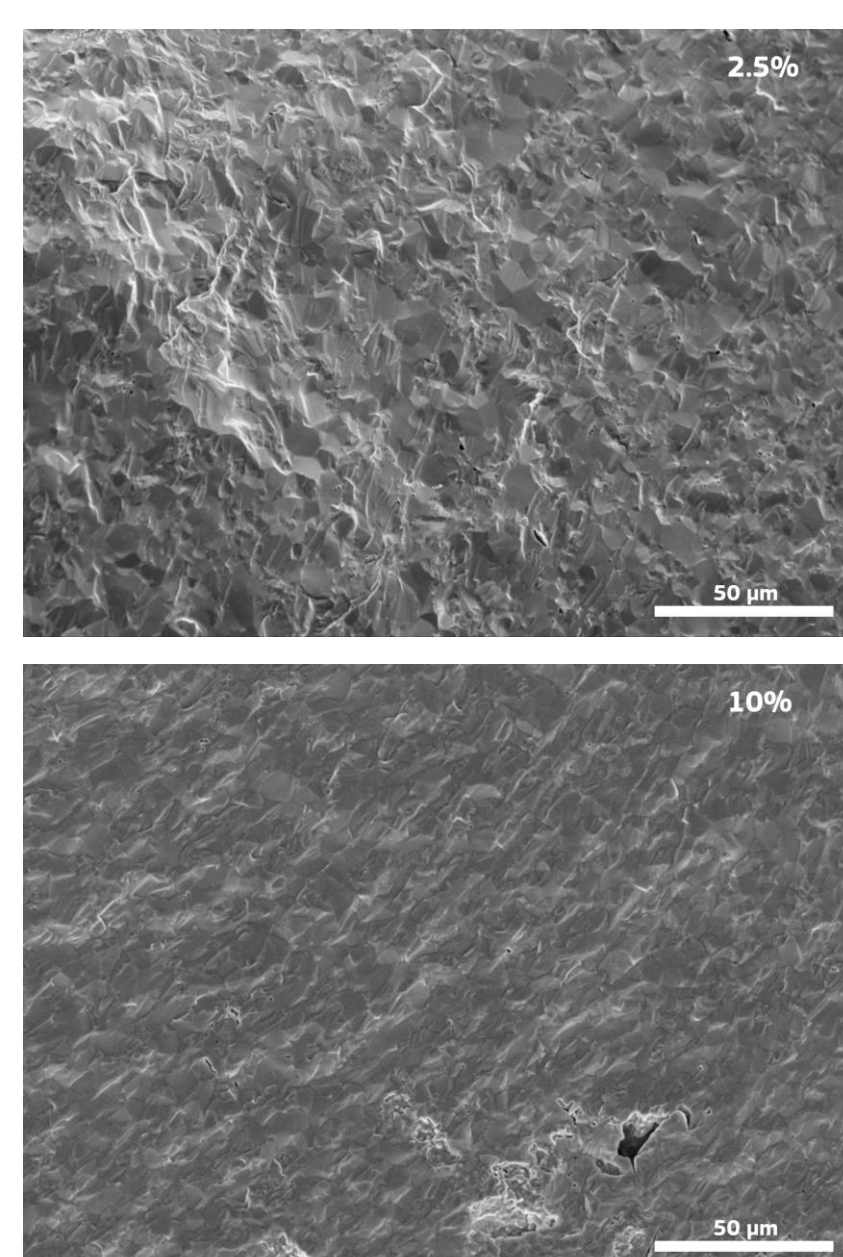
1. Dissolution of the Pu powder using HNO_3 and HF
2. Separation in two batches and addition of the correct amount of U solution
3. Precipitation with ammonia addition
4. Filtration with paper filters
5. Pre-calcination (400°C) under air to burn the filters away
6. Calcination (700°C) under Ar-H_2

Powder pressing and sintering:

Pressing: $\phi = 5\text{ mm}$; $F = 14.5\text{ kN}$; resulting $p = 738\text{ Mpa}$
Sintering: 1650°C , 6 h, under 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.

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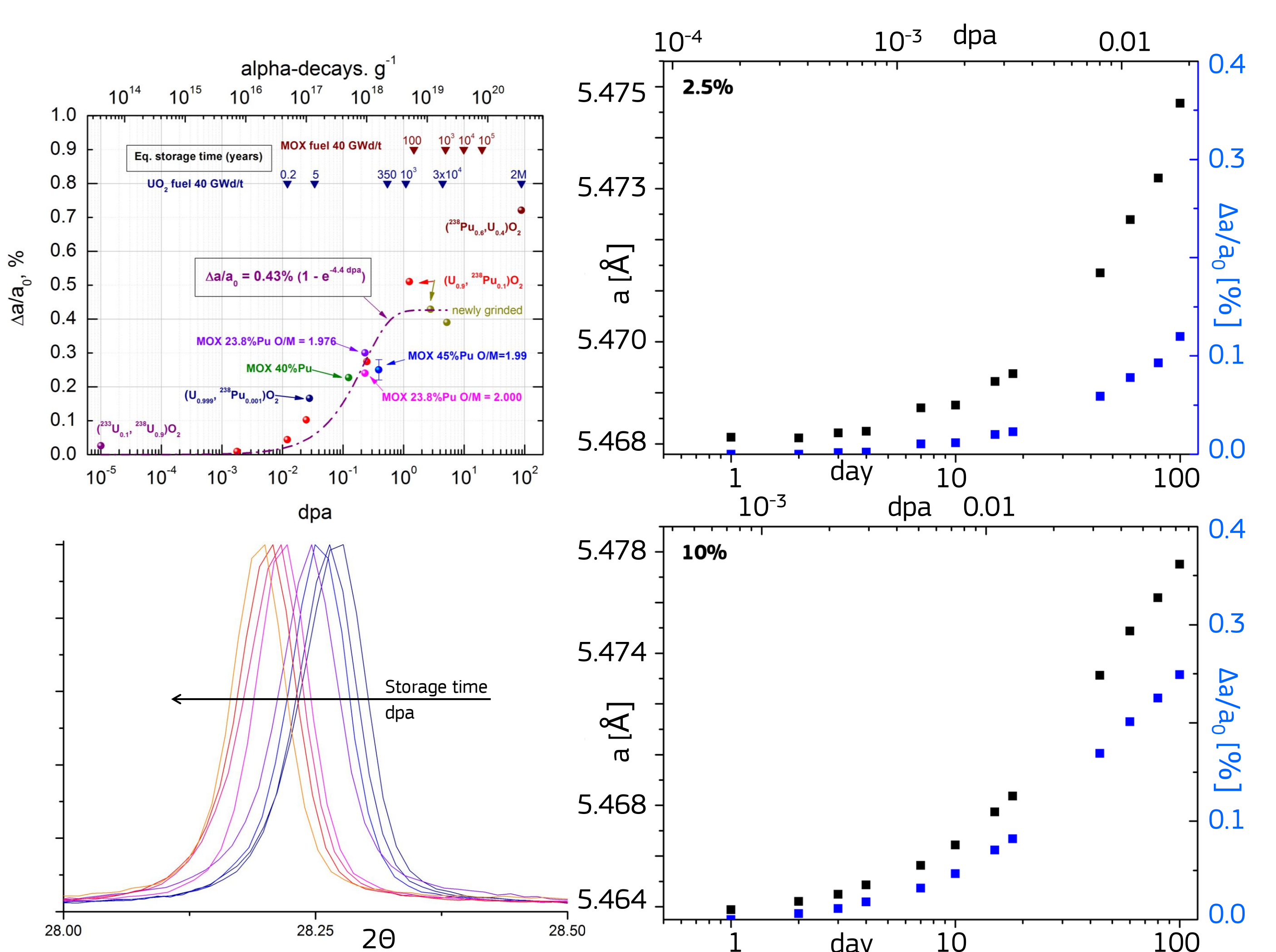
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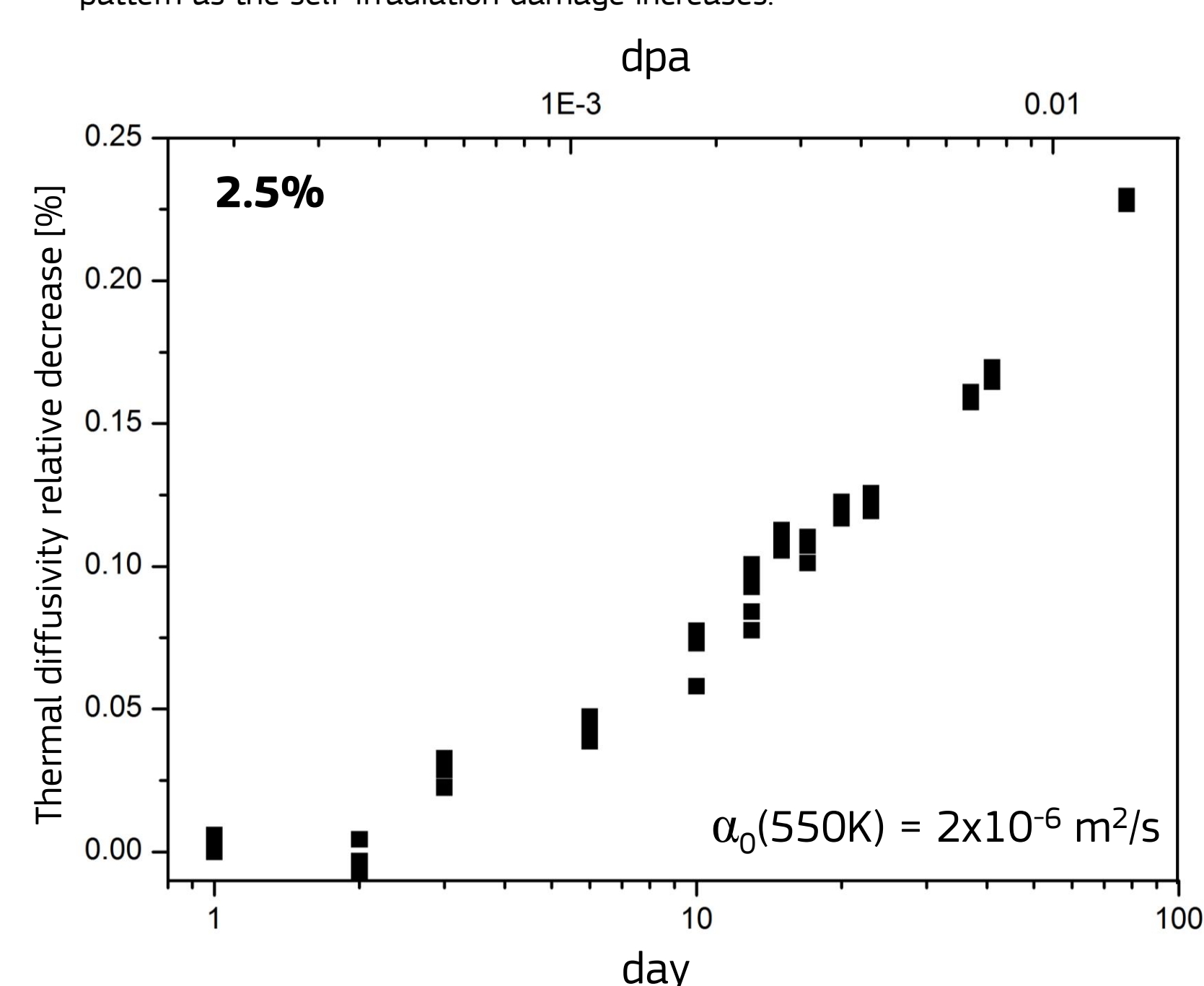
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Periodic characterization – room T batch

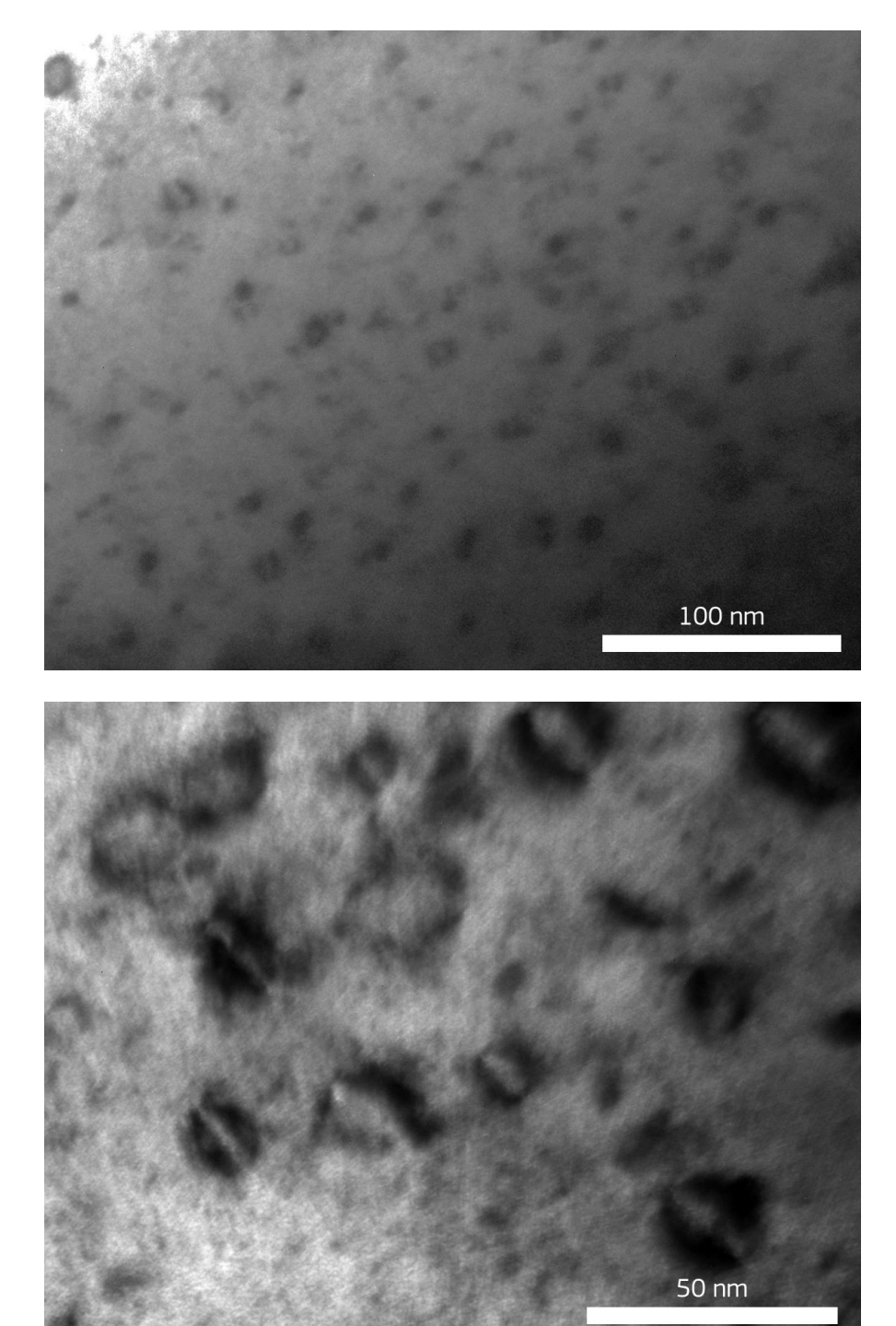
After annealing of defects (4h at 1250°C under Ar-H_2), characterization started for the room T batch.



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)O₂** 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°C and -200°C

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