Radiation Effects in Americium Ceramic Compounds

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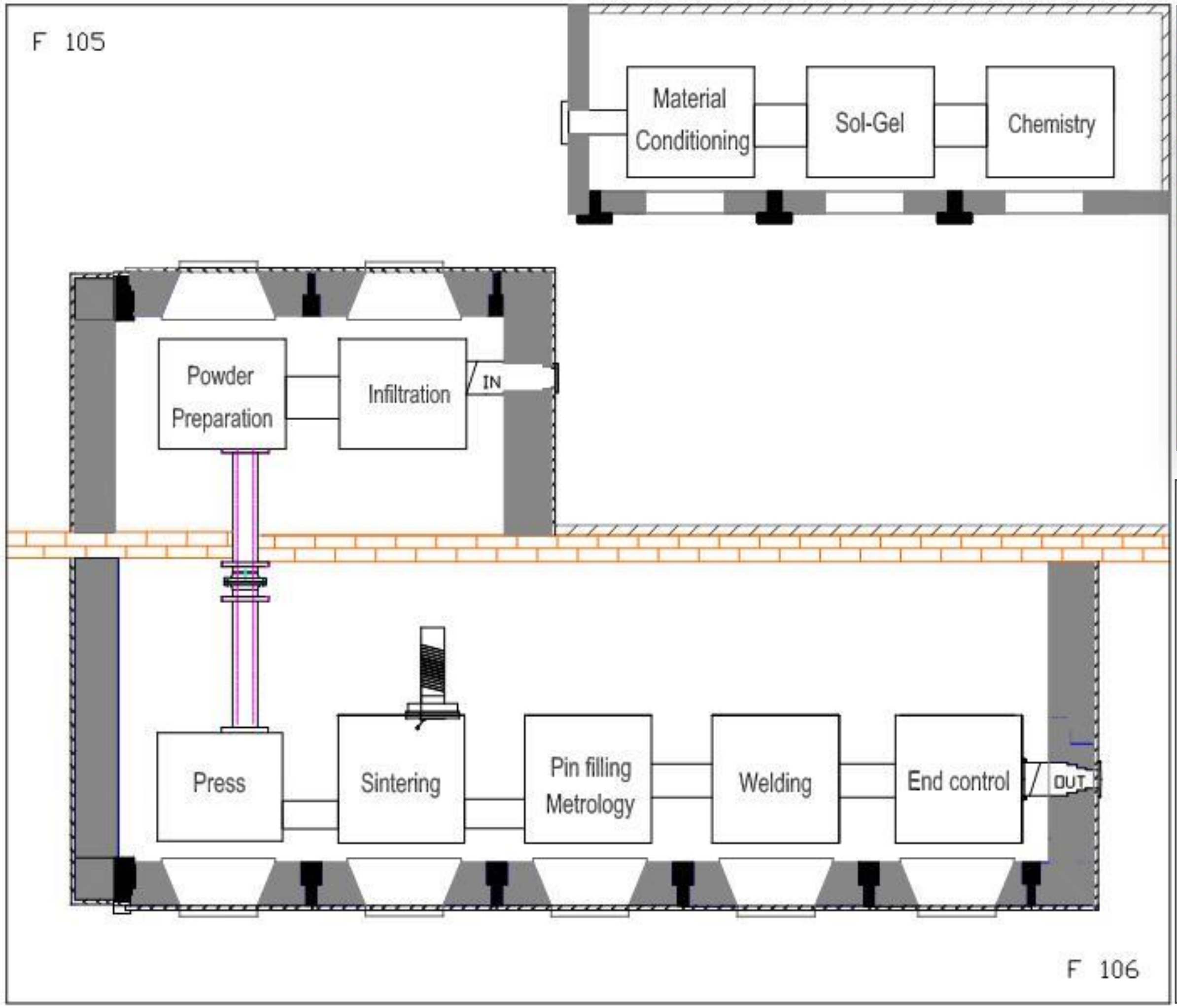
The JRC Minor Actinide Laboratory

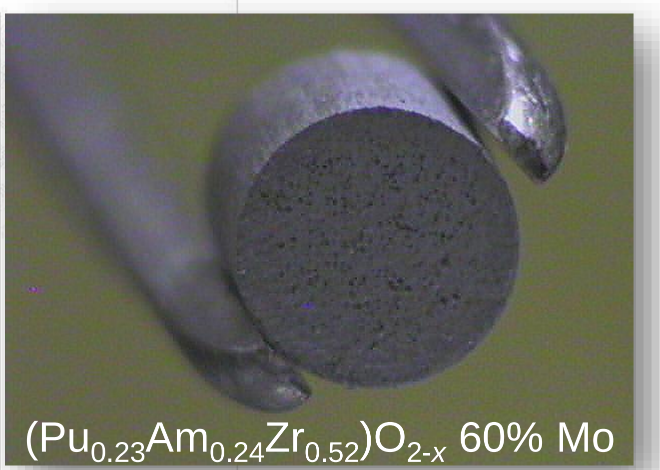
Isotope	Limiting mass (g) [a]	Criterion
^{231}Pa	10	Shielding
^{237}Np	–[b]	
^{241}Am	50	License [c]
^{242m}Am	0.1	Shielding
^{243}Am	65	License [c]
^{244}Cm	Isotope	Shielding & licence [c]

[a] to yield max 2 μSv/h at 1 metre

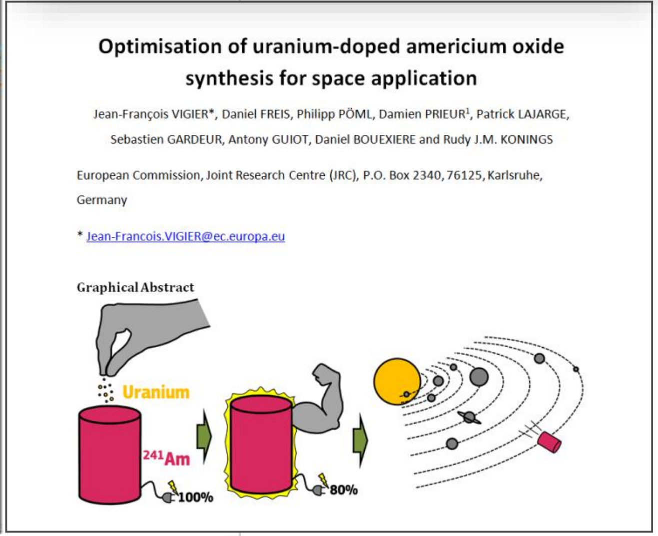
[b] no practical limit

[c] corresponding to the dose equivalent of 200 g Pu in the form of powder (oxide)





$(Pu_{0.23}Am_{0.24}Zr_{0.52})O_{2-x}$ 60% Mo



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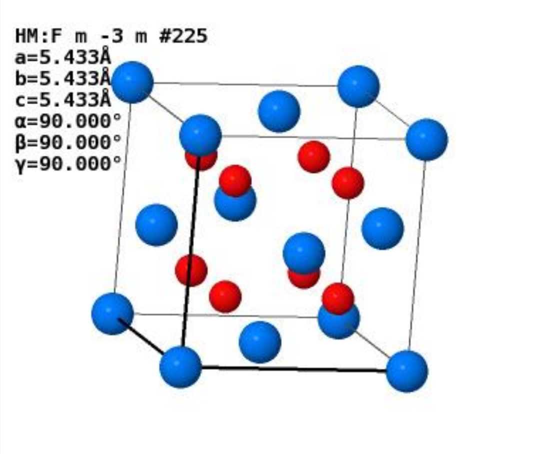
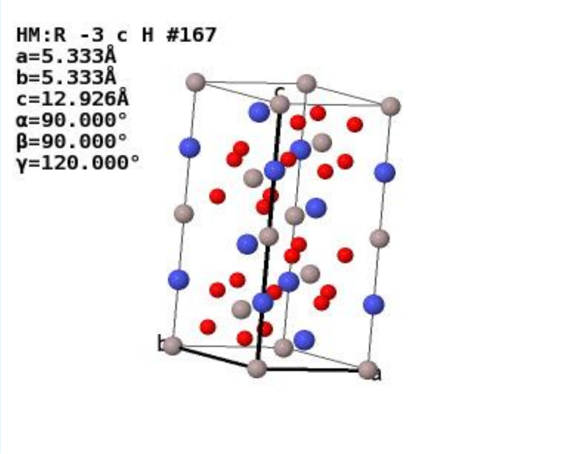
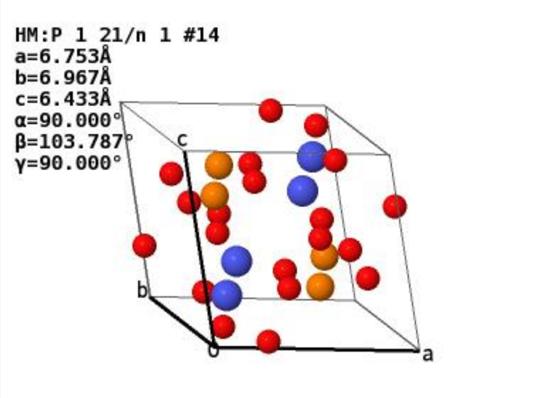
Am-ceramic compounds for radioisotope heater units and radioisotope thermoelectric generators



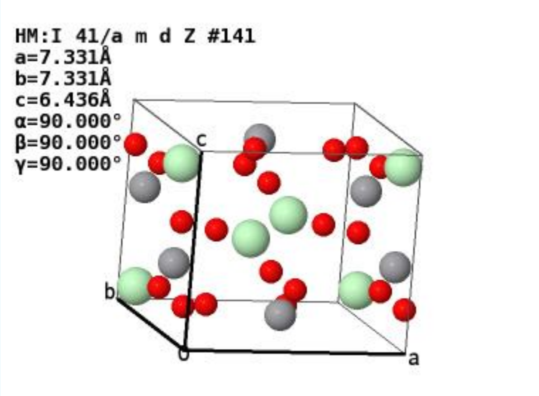
$Am_yU_{1-y}O_2$ (fluorite)



$AmAlO_3$, $AmVO_3$ (perovskites)



$AmPO_4$ (monazite)



$AmVO_4$ (zircon)

- Am-content
- amorphization behaviour
- swelling degree
- ^{237}Np-distribution
- natural analogue
- stability (HT/LT; air, inert atmosphere, vacuum, …)

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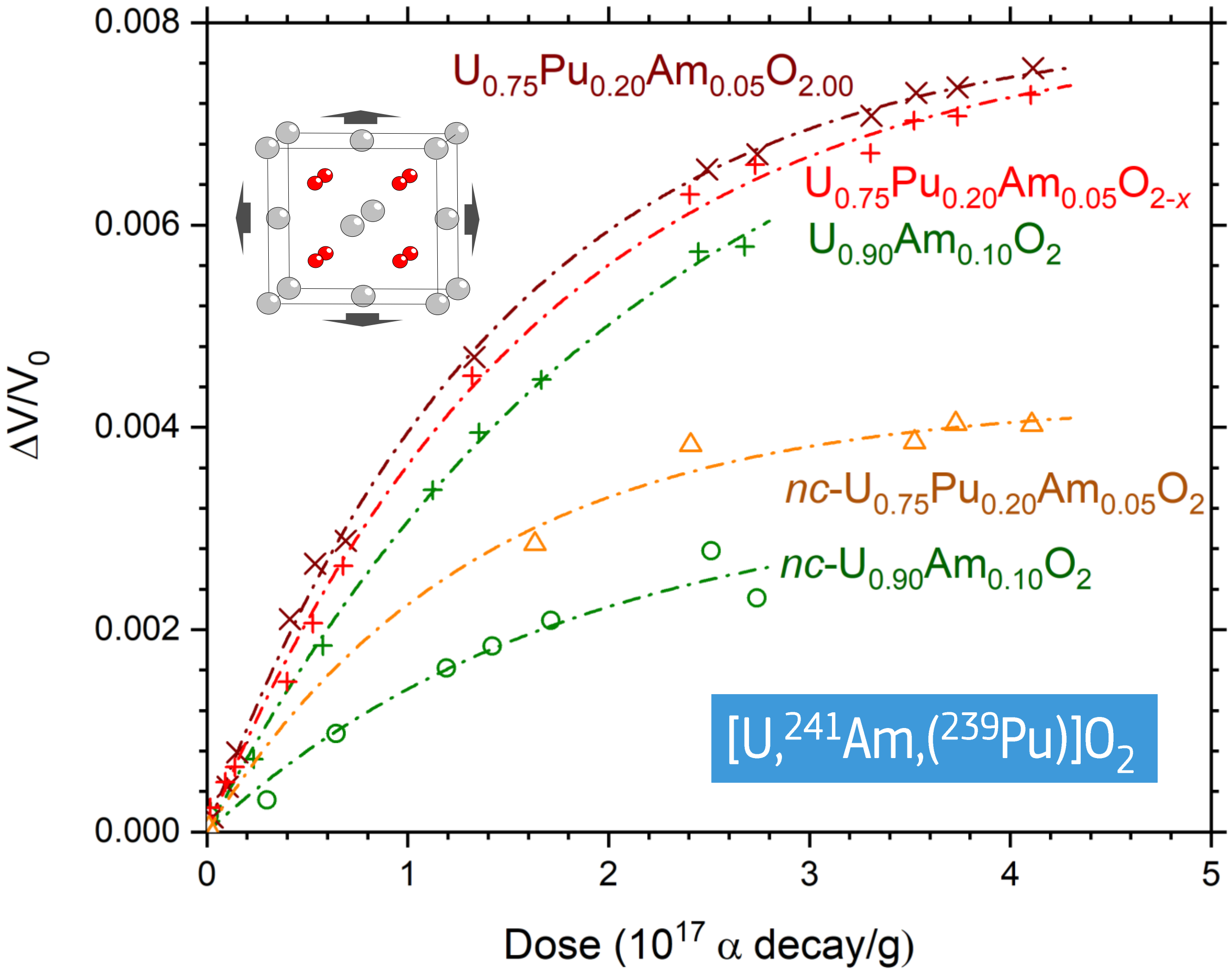
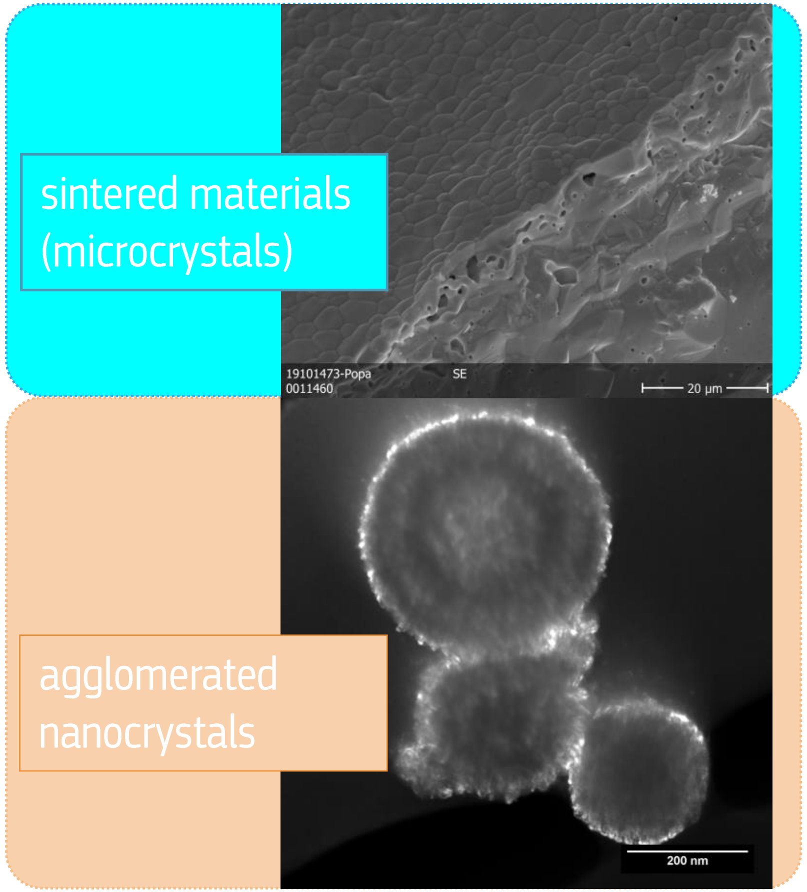
Swelling of the fluorite structure: influence of the particle size





shorter diffusion path of α particles to the grain boundaries

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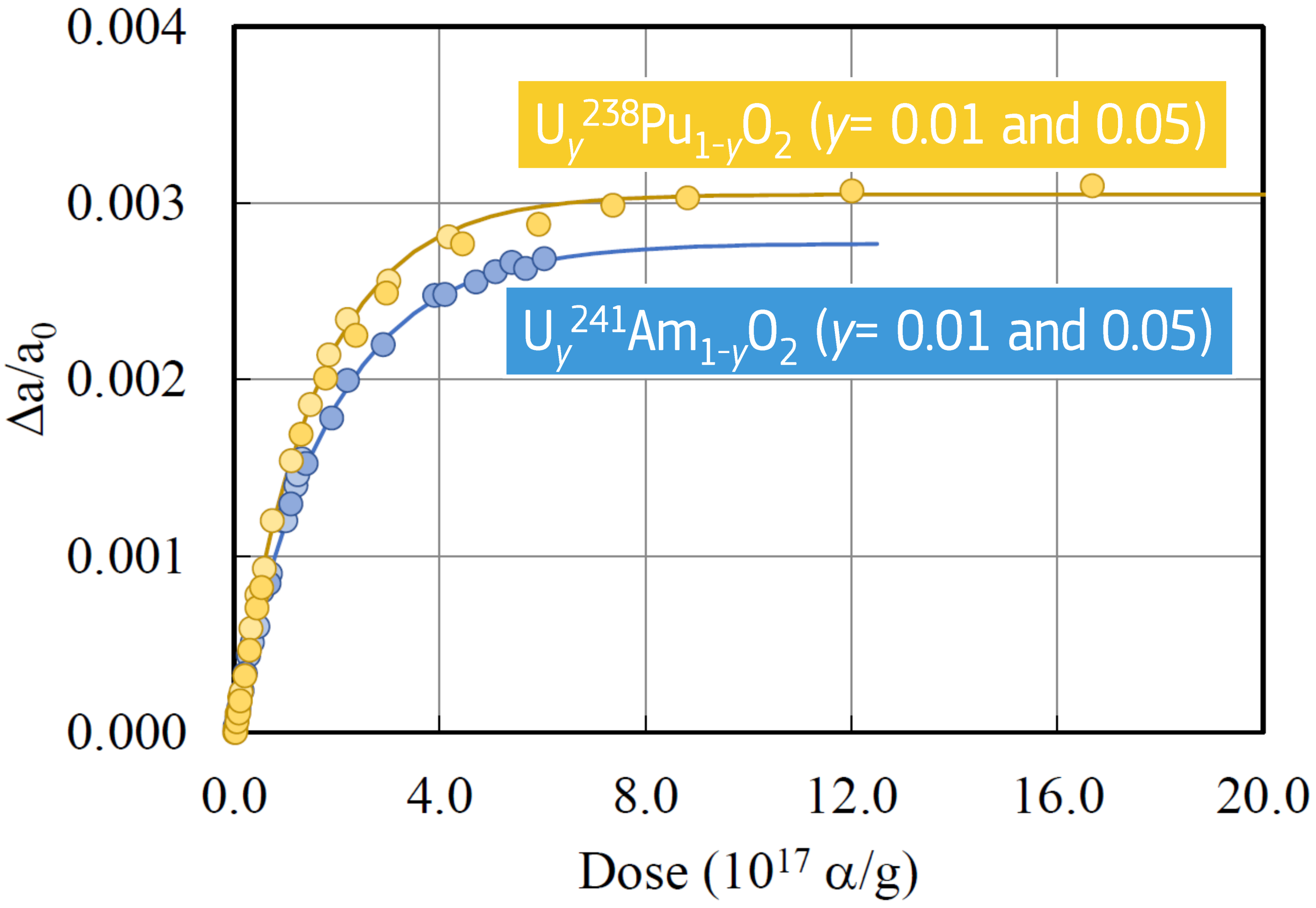
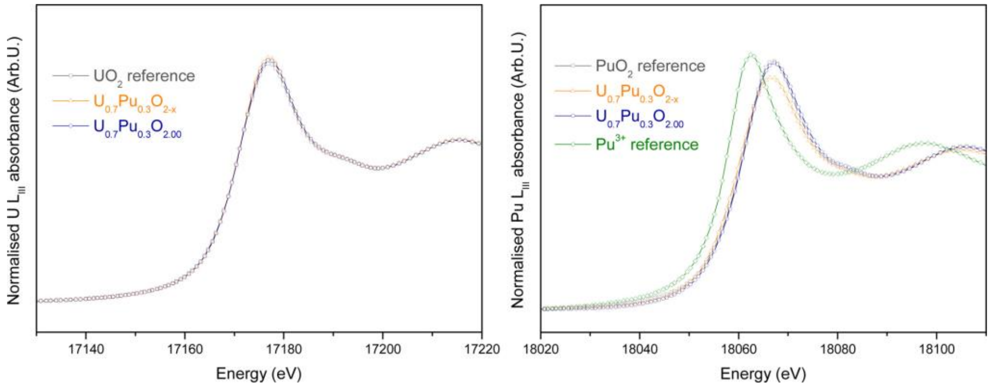
Swelling of the fluorite structure: influence of the nature of dopant

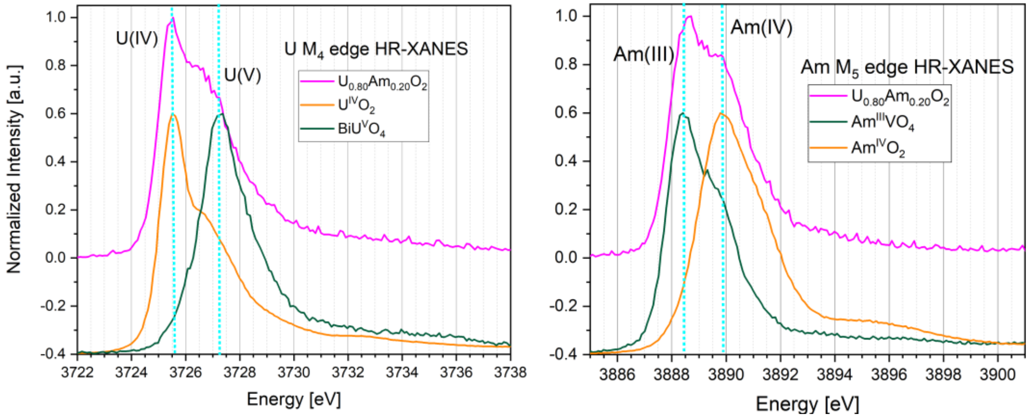


$(U^{4+},Pu^{4+})O_2$

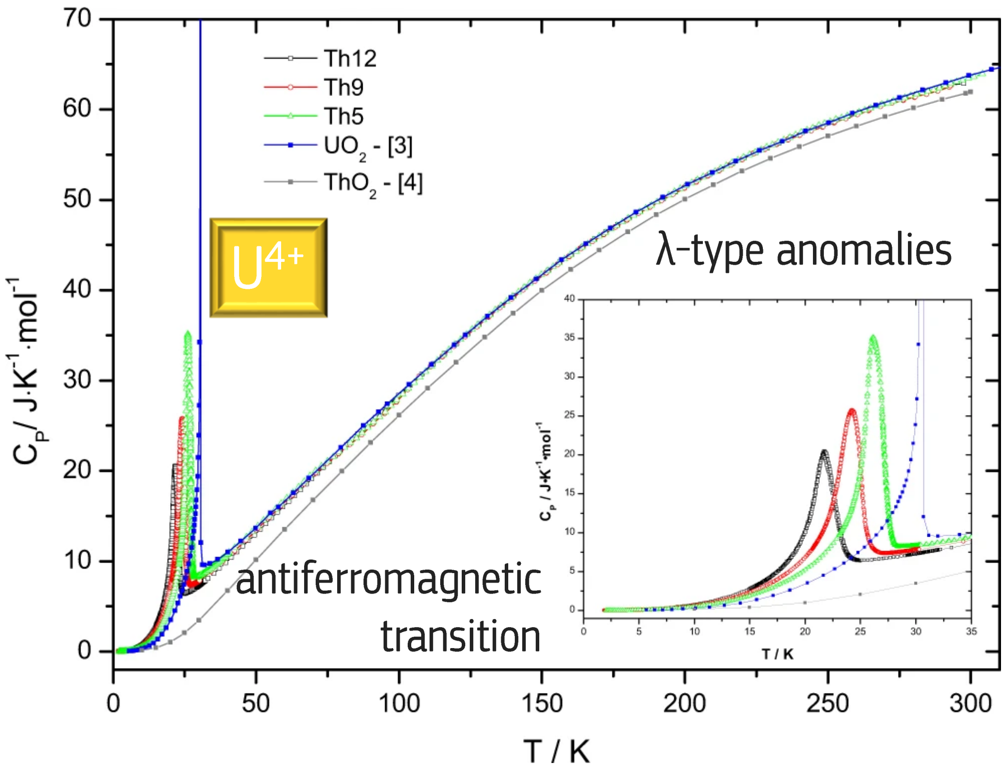
(Vigier *et al.*, *Inorg. Chem.* **54** (2015) 5358–5365)



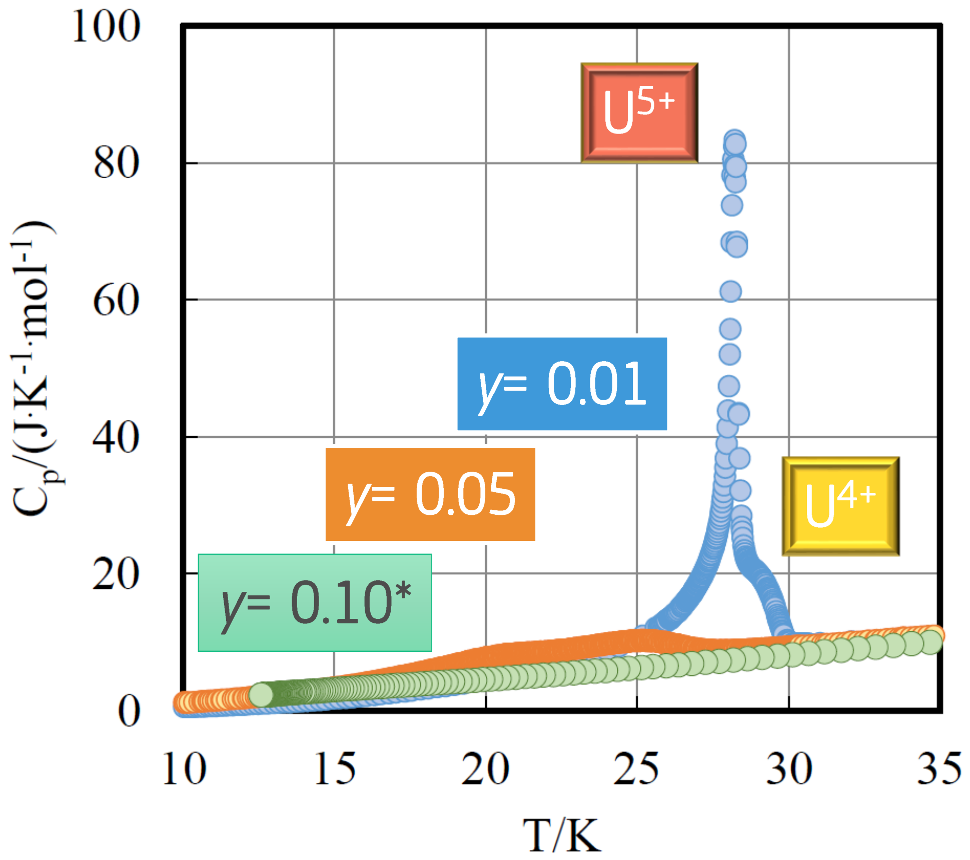
$(U^{4+},U^{5+},Am^{3+},Am^{4+})O_2$



Low temperature heat capacity of $U_{1-y}An_yO_2$ (An= Th, ^{241}Am)



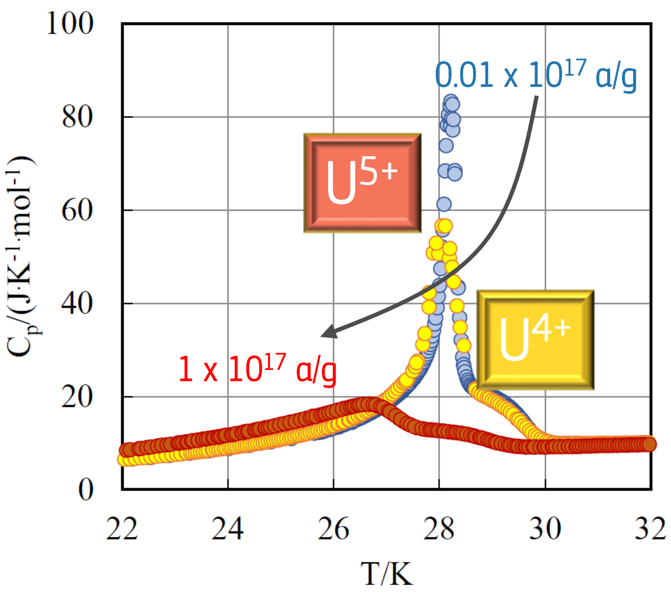
(Vălu *et al.*, *Sci. Rep.* **9** (2019) 5358–5365
[3] J. Huntzicker & E. Westrum, *J. Chem. Thermodyn.* **3** (1971) 61–67
[4] D. Osborne, & E. Westrum, *J. Chem. Phys.* **21** (1953) 1884–1887)



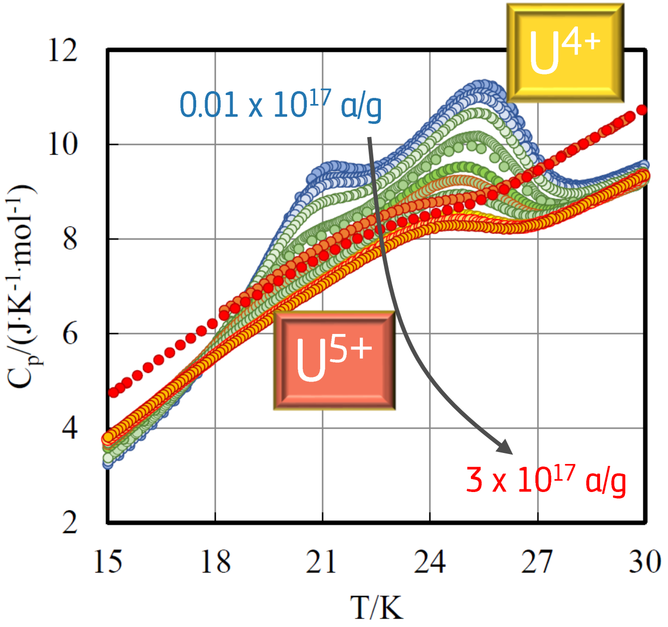
(J.-C. Griveau *et al*, *J. Appl. Phys.* (2022) submitted;
* from Vălu *et al.*, *J. Nucl. Mater.* **507** (2018) 126–134)

Low temperature heat capacity of $U_{1-y}An_yO_2$ (An= ^{241}Am, ^{238}Pu) – aging behaviour

$U_{0.99}{}^{241}Am_{0.01}O_2$

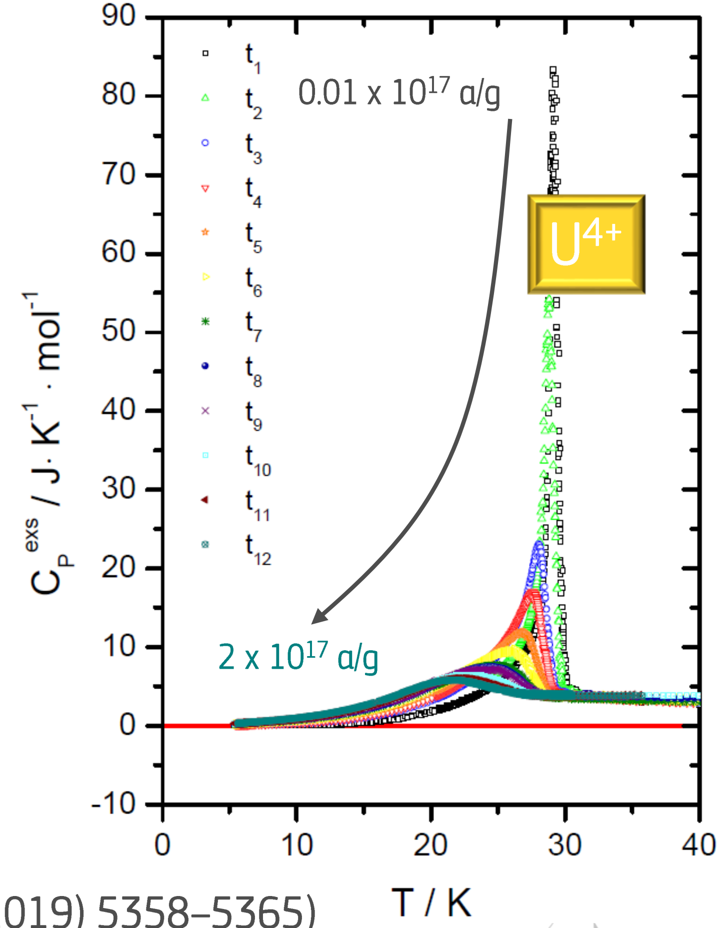


$U_{0.95}{}^{241}Am_{0.05}O_2$



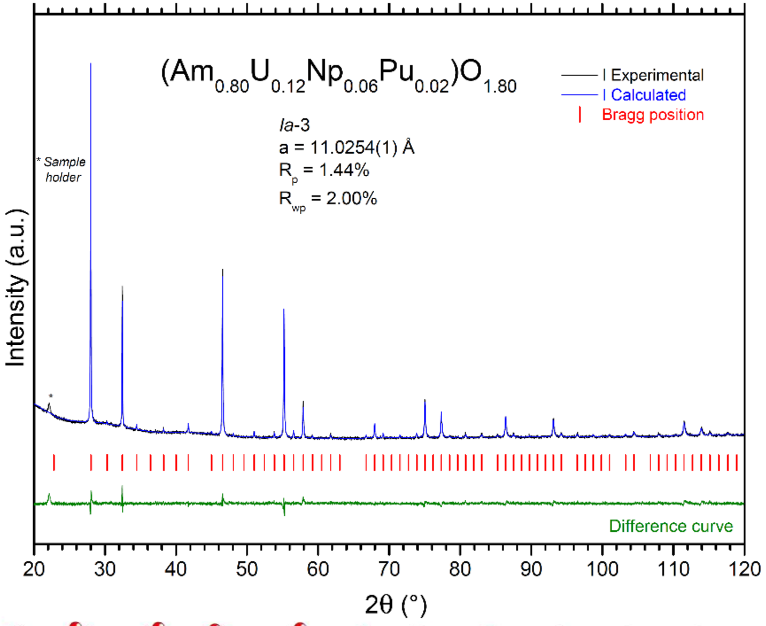
(J.-C. Griveau *et al.*, *J. Appl. Phys.* (2022) submitted)

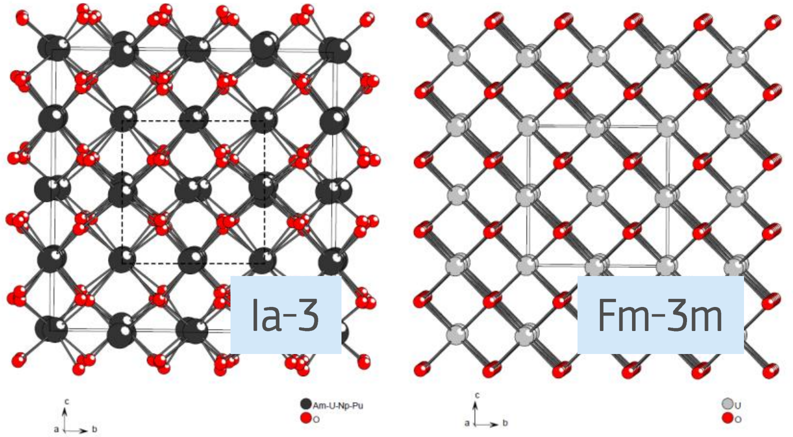
$U_{0.984}{}^{238}Pu_{0.016}O_2$



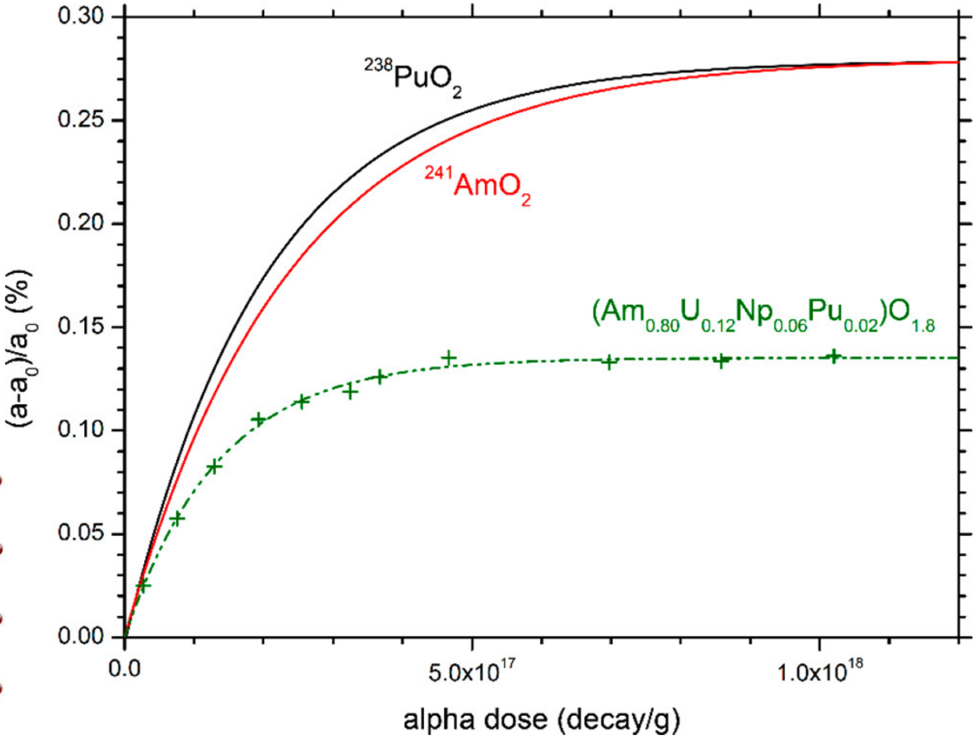
(Vălu *et al.*, *Sci. Rep.* **9** (2019) 5358–5365)

Self-irradiation behaviour of the fluorite and fluorite-related structures: influence of the nature of dopant

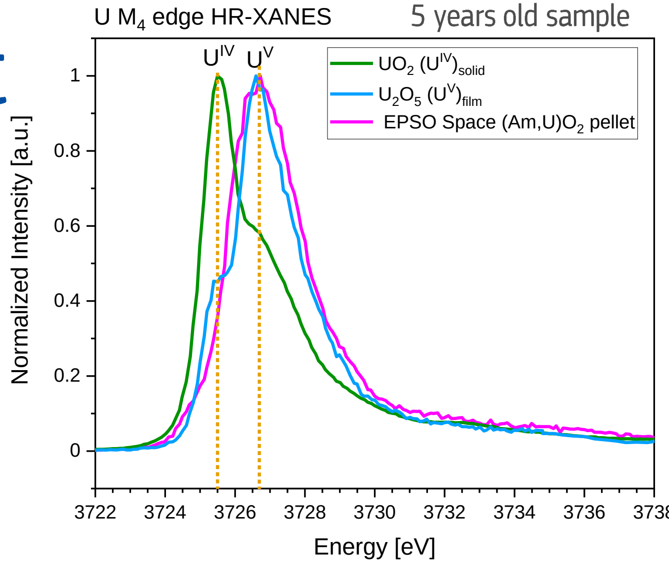


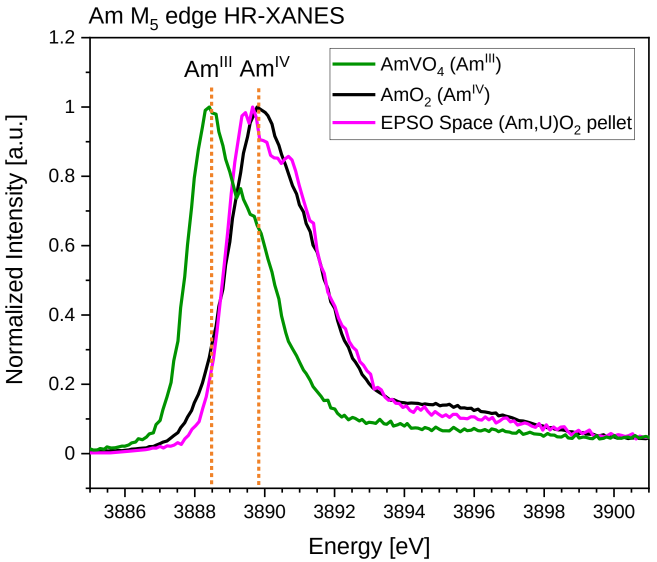


$Am_{0.80}U_{0.12}Np_{0.06}Pu_{0.02}O_{1.8}$



(Vigier *et al., Inorg. Chem.* **57** (2018) 4317-4327)





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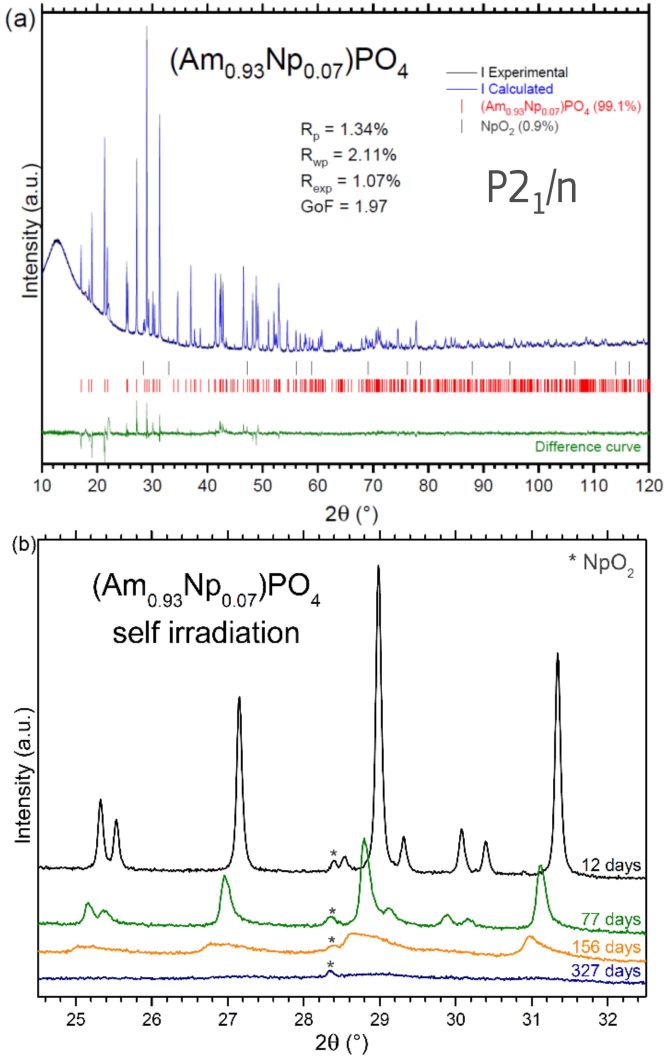
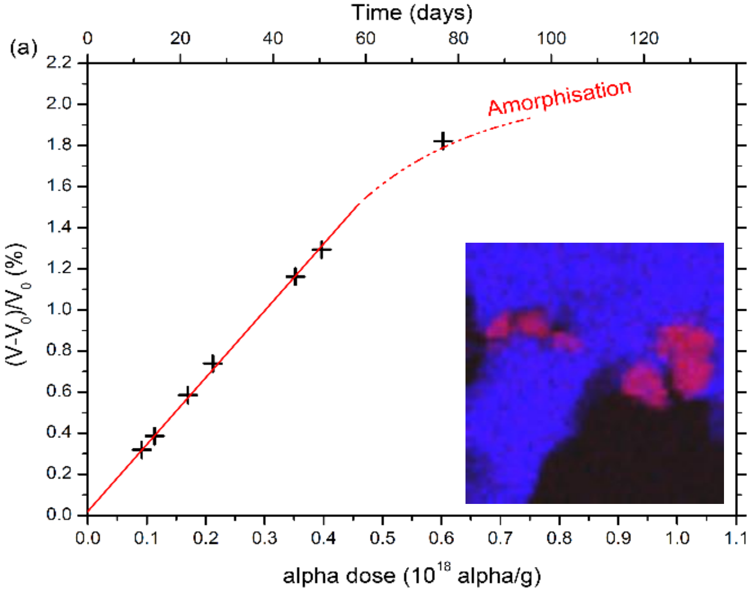
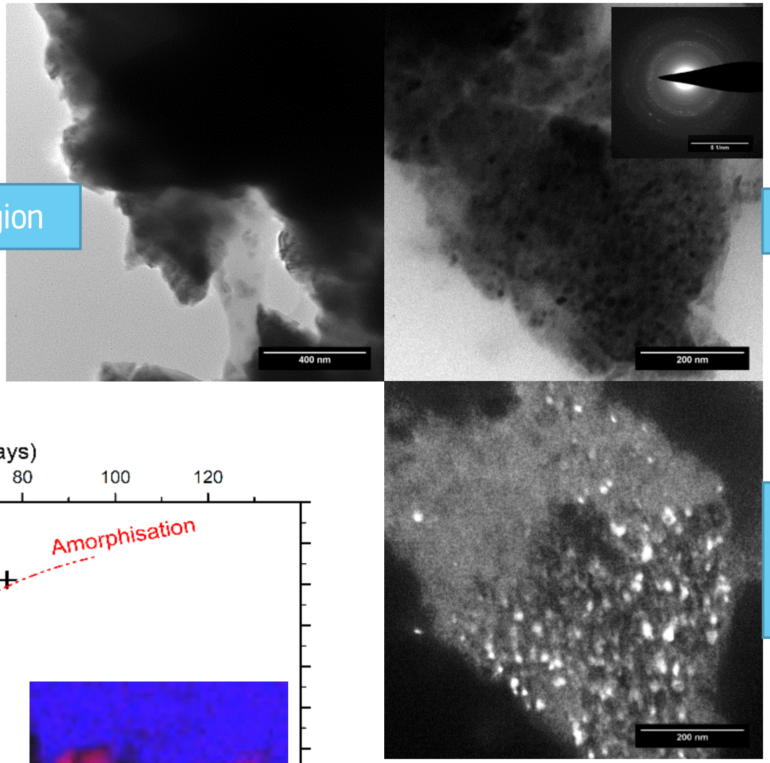
Self irradiation behaviour of $AmPO_4$





TEM, 320 days after synthesis



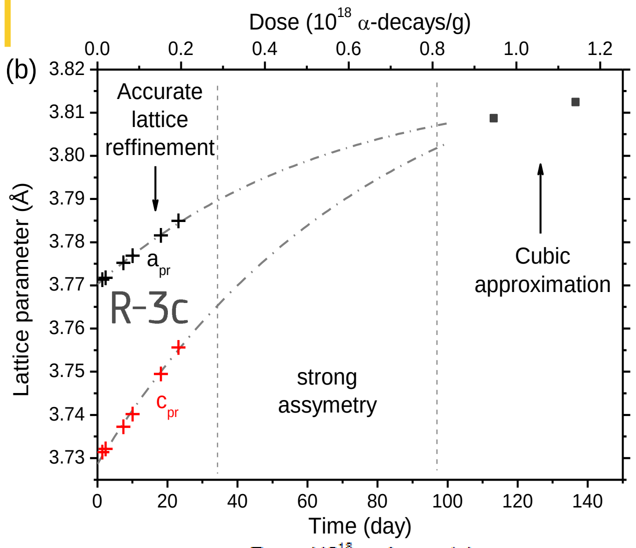
Perfectly amorphous region

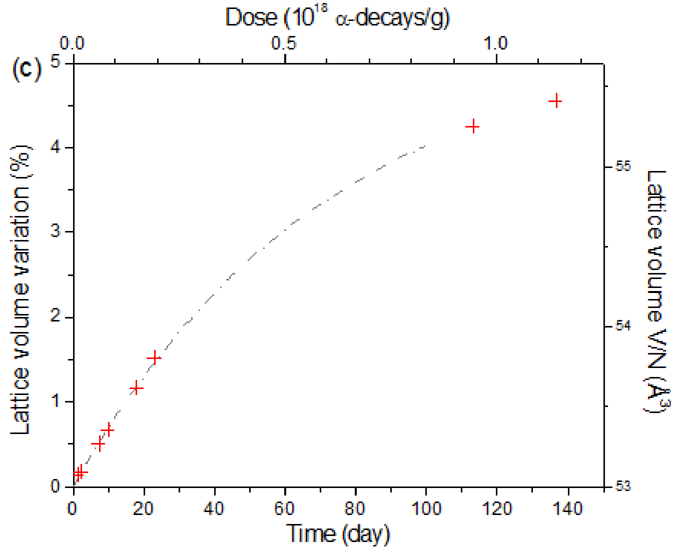
Polycrystalline particle

Dark field of the previous region proving its polycrystalline nature

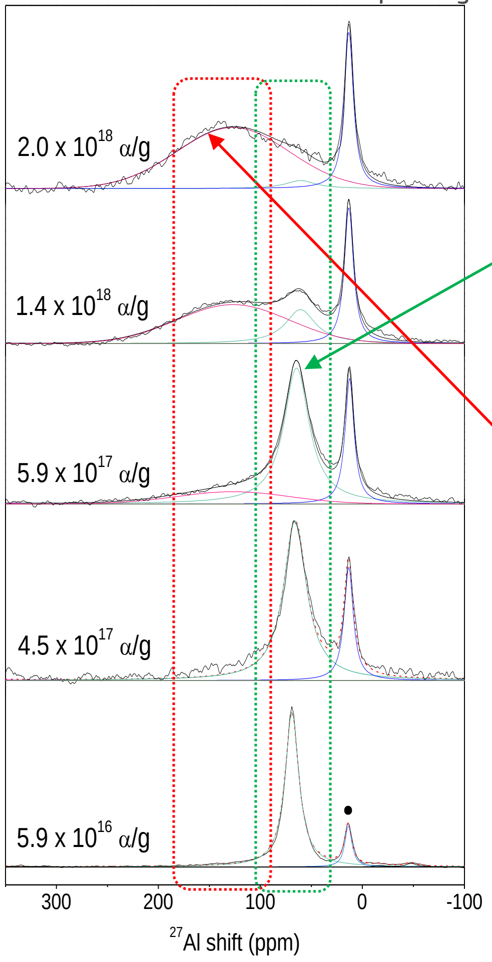
(K. Popa *et al.*, *Inorg. Chem.* **59** (2020) 6595-6602)

Self-irradiation behaviour of $AmAlO_3$

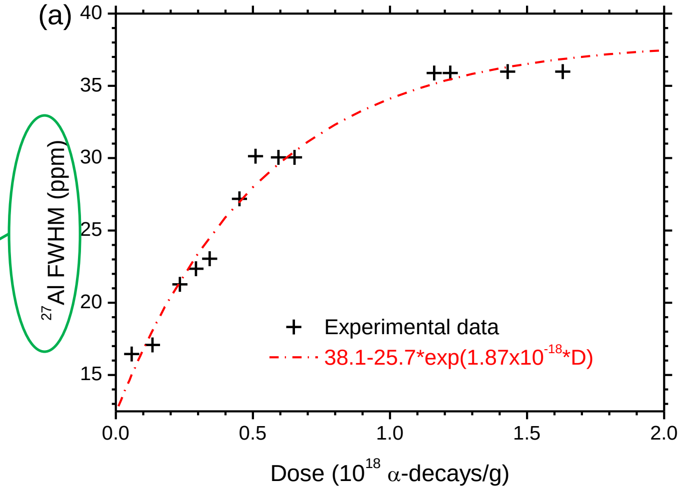


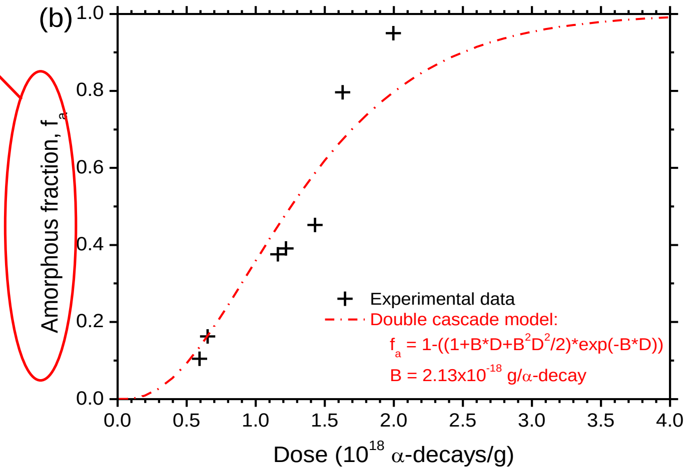


Variation of the ^{27}Al MAS-NMR spectra central transition with time and their corresponding fits



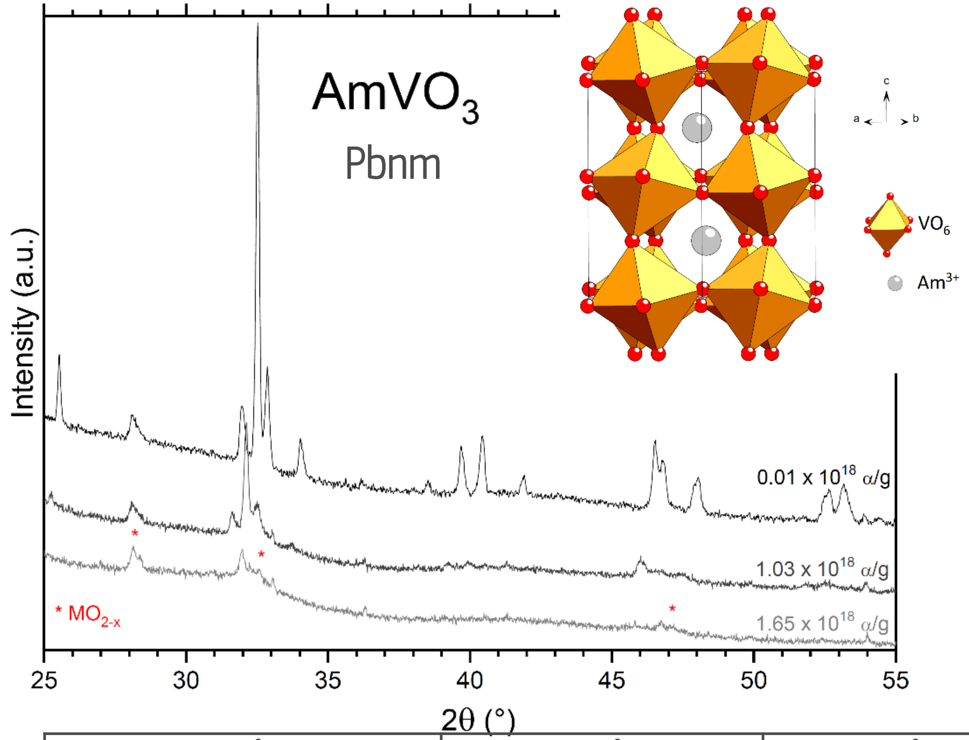
Variation of the ^{27}Al FWHM of crystalline phase (a) and the amorphous phase (b) content, with the alpha decay dose D

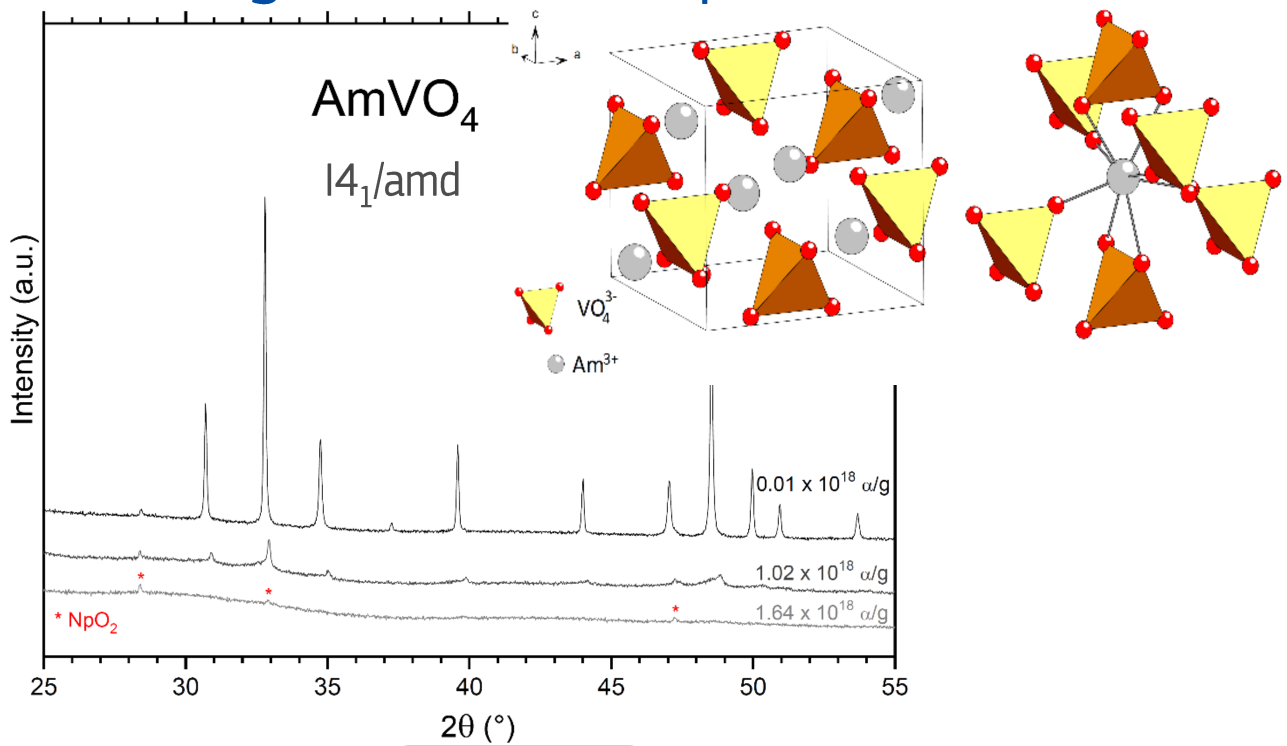




(J.-F. Vigier *et al.*, *Inorg. Chem.* **58** (2019) 9118-9126)

Self irradiation behaviour of $AmVO_3$ and $AmVO_4$





a, Å	b, Å	c, Å	V, Å³	Volume variation, %	Dose (α/g)	dpa
$AmVO_3$ - Pbnm - Perovskite						
5.444	5.590	7.755	235.982	-	0.01×10^{18}	0.0016
5.513	5.661	7.882	245.994	4.2	1.03×10^{18}	0.158
$AmVO_4$ - $I4_1/amd$ – Zircon						
7.294	7.294	6.428	341.952	-	0.01×10^{18}	0.0013
7.243	7.243	6.405	335.964	- 1.8	1.02×10^{18}	0.134

Comparison of the Am-containing ceramics

		$AmVO_3$	$AmVO_4$	$(Am,U)O_2$	$AmPO_4$	$AmAlO_3$
Structure		perovskite	zircon	fluorite	monazite	perovskite
^{241}Am-content (*wt.* %)		71	68	75	72	76
Volume variation (%)		+ 4.2	- 1.8	+ 0.4	+ 1.5	+ 5
Amorphization		fast	fast	no	slow	fast
	oxidising	RT - 280 °C	RT - 1500 °C	RT - 1500 °C	RT - 1500 °C	RT - 1500 °C
Stability	inert				RT - 1500 °C	
	reducing	RT - 1500 °C	RT - 280 °C	RT - 1500 °C		
^{237}Np-distribution		segregation	segregation	atomic scale	atomic scale + segregation	segregation

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References

J.-C. Griveau *et al.*, “Low-temperature heat capacity and magnetism in $(U_{1-y}Ln_y)O_2$ and $(U_{1-y}Am_y)O_2$ (y= 0.01 - 0.05) solid solutions: effects of substitution and self-irradiation”, *J. Appl. Phys.* (2022) submitted.

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Sebastien Gardeur	Patrick Lajarge

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