Synthesis and characterization of homogeneous (U,Am)O₂ and (U,Pu,Am)O₂ nanopowders

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Fig. S1 The autoclave (with Teflon insets) and the heating mantle used for the present experiment



Fig. S2 Pictures of the sintered disks: $U_{0.90}Am_{0.10}O_2$ (left) and $U_{0.80}Am_{0.20}O_2$ (right)



Fig. S3 Sample holders with embedded material for XRD and XAS measurements



Fig. S4 HR-XANES U M_4 edge for as-synthesised nanoparticles (lilac) and sintered (magenta) $U_{0.80}Am_{0.20}O_2$ samples in the post-edge (a) and whole energy (b) range along with the U(IV) O_2 (orange) and the BiU(V) O_4 (green) references.



Fig. S5. Variation of crystal size (top) and strain (bottom) of nanocrystalline materials as a function on alpha dose

Table S1. Characterisation of sintered disks out of geometrical measurements (calculated for a theoretical density of 11 g/cm^3)

Composition	Disk/ Pellet	h, mm	d, mm	m, mg	ρ, g/cm³	% TD		Observations
						Geometrical	Hydrostatic	
U _{0.90} Am _{0.10} O _{2.00}	1	1.765	3.424	189	10.382	94.64		
	2	2.230	3.611	240	10.509	95.80	98.80	
	3	1.668	3.640	166	9.564	87.18		
U _{0.80} Am _{0.20} O _{2.00}	1	1.628	3.73	179	10.062	91.72		a
	2	2.348	3.82	247	9.080	82.80		a
	3	2.055	3.675	222	10.185	92.94	96.04	
U _{0.75} Pu _{0.20} Am _{0.05} O _{2-x}	1	0.762	4.247	86	7.971	72.46		a
	2	1.223	4.244	150	8.674	78.86		a
	3	1.234	4.261	150	8.529	77.53		а
U _{0.75} Pu _{0.20} Am _{0.05} O _{2.00}	1	0.780	4.172	102	9.571	87.00		
	2	1.168	4.149	149	9.440	85.82		
	3	6.957	4.133	854	9.155	83.22		а

^a anisotropic radial shrinkage

Table S2. Uranium and plutonium isotopic compositions

	Mass fraction, wt%	Uncertainty, wt% [k = 2]	
²³³ U/U	< 0.001	-	
²³⁴ U/U	0.00530	0.00052	
²³⁵ U/U	0.7124	0.0022	
²³⁶ U/U	< 0.001	-	
²³⁸ U/U	99.2823	0.0072	
²³⁸ Pu/Pu	0.01398	0.00084	
²³⁹ Pu/Pu	90.713	0.018	
²⁴⁰ Pu/Pu	9.1170	0.0048	
²⁴¹ Pu/Pu	0.0824	0.0049	
²⁴² Pu/Pu	0.0732	0.0060	

<i>nc</i> -U _{0.90} Am _{0.10} O ₂		n	<i>c</i> -U _{0.80} Am	n _{0.20} O ₂	<i>nc</i> -U _{0.75} Pu _{0.20} Am _{0.05} O ₂			
Dose, α/g	CS, nm	strain	Dose, α/g	CS, nm	strain	Dose, α/g	CS, nm	strain
1.28E+15	12.0	0.00197	2.96E+15	10.0	0.00307	8.66E+14	12.9	0.00224
3.00E+16	12.2	0.00209	5.93E+16	10.4	0.00347	1.63E+17	13.0	0.00227
6.45E+16	13.4	0.00279	1.27E+17	10.7	0.00373	2.41E+17	12.3	0.00207
1.19E+17	13.9	0.00298	2.34E+17	10.8	0.00402	3.52E+17	12.5	0.00199
1.42E+17	12.9	0.00253	2.79E+17	10.4	0.00364	3.73E+17	12.5	0.00201
1.71E+17	14.3	0.00325	3.55E+17	10.4	0.0039	4.11E+17	12.8	0.00213
2.51E+17	15.9	0.00397	4.93E+17	11.4	0.00445			
2.74E+17	13.7	0.00307	5.38E+17	11.2	0.00429			

Table S3. Variation of crystal size and strain of nanocrystalline materials as a function on alpha dose