

Supplementary Materials

Impacts of Am Aggregation on the Bulk Properties of Mixed Oxides (U, Am)O₂ from First Principles

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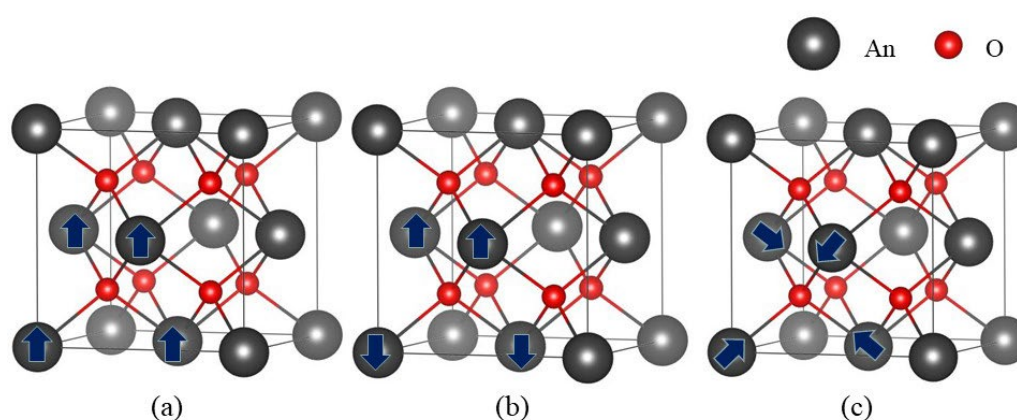


Figure 1S. The crystal structure of AnO₂: (a) the 1k-ferromagnetic (FM) order, (b) longitudinal 1k-collinear antiferromagnetic (AFM) order and (c) transverse 3k-noncollinear antiferromagnetic (AFM) order. 1k = [001] and 3k = [111].

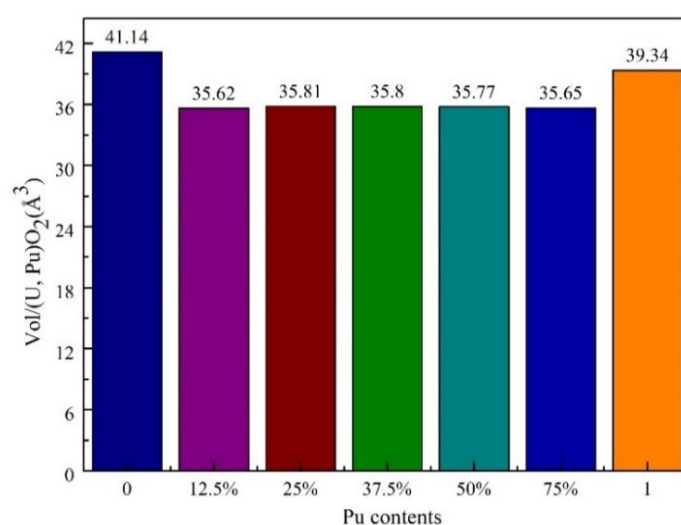


Figure 2S. Variation of the systems volume of mixed oxides (U, Pu)O₂ with different Pu aggregation contents.

Table S1. Magnetic stability including the lattice parameter and band gap of the (U, Pu)O₂ defect model for various Pu aggregation contents using PBEsol + *U*.

Pu Content	AFM			FM			$E_{\text{FM}} - E_{\text{AFM}}/\text{atom}$ (eV)
	a_0 (Å)	c/a	G (eV)	a_0 (Å)	c/a	G (eV)	
UO ₂	5.480	1.0	2.0	5.469	1.0	1.8	0.12
12.5%	5.223	1.0	1.1	5.222	1.0	0.9	0.00
25%	5.233	1.0	1.1	5.232	1.0	0.9	0.00
37.5%	5.232	0.999	1.1	5.232	0.998	0.9	0.00
50%	5.230	0.998	1.0	5.231	0.998	0.8	0.02
75%	5.224	0.998	0.0	5.224	0.999	0.0	0.00
PuO ₂	5.399	1.0	2.0	5.409	1.0	1.9	0.39

Table S2. Lattice parameter, band gap, magnetic moment of UO₂, PuO₂ and (U, Pu)O₂ using PBEsol + *U*.

Pu Content	Functional	a_0 (Å)		Gap (eV)	μ_{mag} (μ_B)	$E_{\text{FM}} - E_{\text{AFM}}/\text{atom}$ (eV)
		AFM	FM			
UO ₂	PBEsol + <i>U</i>	5.480	5.469	2.0	2.0	+0.12
	PBE + <i>U</i> [1]	5.543	5.547	2.5		
	Experiment [2]	5.470		2.1		
(U _{0.75} Pu _{0.25})O ₂	PBEsol + <i>U</i>	5.223	5.222	1.1	5.4	0.00
	PBE + <i>U</i> [1]	5.520	5.520	1.2		
(U _{0.5} Pu _{0.5})O ₂	PBEsol + <i>U</i>	5.217	5.217	1.1	5.4	0.00
	PBE + <i>U</i> [1]	5.497	5.495	1.2		
(U _{0.25} Pu _{0.75})O ₂	PBEsol + <i>U</i>	5.212	5.212	0.0	5.3	0.00
	PBE + <i>U</i> [1]	5.399	5.409	1.9	4.1	+0.39
PuO ₂	PBEsol + <i>U</i>	5.399	5.409	1.9	4.1	+0.39
	PBE + <i>U</i> [1]	5.453	5.452	2.0		
	Experiment [2]	5.398		1.8		

Table S3. Energy of formation of UO₂, PuO₂ and (U, Pu)O₂ using PBEsol + *U*.

Compound	E_f (eV)				
	Experiment	CALPHAD	PBE + <i>U</i> [1]	vdW-optPBE + <i>U</i> [1]	PBEsol + <i>U</i>
UO ₂	−11.24 [3]	−11.23 [4]	−10.86	−11.27	−11.78
(U _{0.75} Pu _{0.25})O ₂			−10.69	−11.18	−14.65
(U _{0.5} Pu _{0.5})O ₂			−10.51	−11.09	−14.19
(U _{0.25} Pu _{0.75})O ₂			−10.33	−10.99	−13.73
PuO ₂	−10.94 [5]	−10.99 [5]	−10.10	−10.90	−11.34

Table S4. Elastic constants and bulk modulus of (U, Pu)O₂ using PBEsol + *U*.

	Functional	UO ₂	(U _{0.75} Pu _{0.25})O ₂	(U _{0.5} Pu _{0.5})O ₂	(U _{0.25} Pu _{0.75})O ₂	PuO ₂
C_{11} (GPa)	PBEsol + <i>U</i>	383	378	357	349	387
	PBE + <i>U</i> [1]	364	375	365	368	375
	LDA + <i>U</i> [5]	401				
	Experiment [3]	389				
C_{12} (GPa)	PBEsol + <i>U</i>	126	101	99	98	126
	PBE + <i>U</i> [1]	112	108	116	115	111
	LDA + <i>U</i> [5]	132				
	Experiment [3]	119				
C_{44} (GPa)	PBEsol + <i>U</i>	72	51	44	30	71
	PBE + <i>U</i> [1]	58	62	66	68	70
	LDA + <i>U</i> [5]	94				
	Experiment [3]	60				

B_0 (GPa)	PBEsol + U	212	193	185	182	199
	PBE + U [1]	196	197	199	199	199
	LDA + U [5]	222	235			208
	Experiment [3]	207	156.9–221			178

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