Supplementary Materials for

**Impacts of Am** **aggregation on the bulk properties of mixed oxides (U, Am)O2 from first-principles**

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**Fig. S1.** Variation of the systems volume of mixed oxides (U, Pu)O2 with different Pu aggregation contents.



**Fig. S2.** Variation of the energy *E*tot 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*FM-*E*AFM/atom(eV) |
|  | *a*0 (Å) | *c*/*a* | *G* (eV) |  | *a*0 (Å) | *c*/*a* | *G* (eV) |  |
| UO2 | 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 |
| PuO2 | 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） | *G*（eV） | *μ*mag（*μB*） | *E*FM-*E*AFM/atom（eV） |
|  |  | AFM | FM | AFM | AFM |  |
| UO2 | PBEsol+*U* | 5.480 | 5.469 | 2.0 | 2.0 | +0.12 |
|  | PBE+*U* [2] | 5.543 | 5.547 | 2.5 |  |  |
|  | expt [3] | 5.470 |  | 2.1 |  |  |
| (U0.75Pu0.25)O2 | PBEsol+*U* | 5.223 | 5.222 | 1.1 | 5.4 | 0.00 |
|  | PBE+*U* [2] | 5.520 | 5.520 | 1.2 |  |  |
| (U0.5Pu0.5)O2 | PBEsol+*U* | 5.217 | 5.217 | 1.1 | 5.4 | 0.00 |
|  | PBE+*U* [2] | 5.497 | 5.495 | 1.2 |  |  |
| (U0.25Pu0.75)O2 | PBEsol+*U* | 5.212 | 5.212 | 0.0 | 5.3 | 0.00 |
| PuO2 | PBEsol+*U* | 5.399 | 5.409 | 1.9 | 4.1 | +0.39 |
|  | PBE+*U* [2] | 5.453 | 5.452 | 2.0 |  |  |
|  | expt [3] | 5.398 |  | 1.8 |  |  |

**Table S3**

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

|  |  |
| --- | --- |
|  | *Ef*（eV） |
| Compound | expt | CALPHAD | PBE+*U*[2] | vdW-optPBE+*U*[2] | PBEsol+*U* |
| UO2 | -11.24 [4] | -11.23[6] | -10.86 | -11.27 | -11.78 |
| (U0.75Pu0.25)O2 |  |  | -10.69 | -11.18 | -14.65 |
| (U0.5Pu0.5)O2 |  |  | -10.51 | -11.09 | -14.19 |
| (U0.25Pu0.75)O2 |  |  | -10.33 | -10.99 | -13.73 |
| PuO2 | -10.94 [5] | -10.99[7] | -10.10 | -10.90 | -11.34 |

**Table S4**

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

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | Functional | UO2 | (U0.75Pu0.25)O2 | (U0.5Pu0.5)O2 | (U0.25Pu0.75)O2 | PuO2 |
| *C*11（GPa） | PBEsol+*U* | 383 | 378 | 357 | 349 | 387 |
|  | PBE+*U* [2] | 364 | 375 | 365 | 368 | 375 |
|  | LDA+*U* [1] | 401 |  |  |  |  |
|  | expt [4] | 389 |  |  |  |  |
| *C*12（GPa） | PBEsol+*U* | 126 | 101 | 99 | 98 | 126 |
|  | PBE+*U* [2] | 112 | 108 | 116 | 115 | 111 |
|  | LDA+*U* [1] | 132 |  |  |  |  |
|  | expt [4] | 119 |  |  |  |  |
| *C*44（GPa） | PBEsol + *U* | 72 | 51 | 44 | 30 | 71 |
|  | PBE+*U* [2] | 58 | 62 | 66 | 68 | 70 |
|  | LDA+*U* [1] | 94 |  |  |  |  |
|  | expt [4] | 60 |  |  |  |  |
| *B*0（GPa） | PBEsol+*U* | 212 | 193 | 185 | 182 | 199 |
|  | PBE+*U* [2] | 196 | 197 | 199 | 199 | 199 |
|  | LDA+*U* [1] | 222 | 235 |  |  | 208 |
|  | expt [4] | 207 | 156.9-221 |  |  | 178 |

**Declaration of competing interest**

The authors declare that they have no known competing ﬁnancial interests or personal relationships that could have appeared to inﬂuence the work reported in this paper.

**Data availability**

No data was used for the research described in the article.

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