Supporting Information

High-Pressure Tuning of Electronic Structure, Stability and Mechanical Properties of Plutonium Oxides: A DFT+U Study

Xiaolin Zhang[1], Minzhuo Xiong[1], Jinxing Cheng [2,], Qingbo Wang[2], Youpeng Wu[2], Ziyu Hu[1,*]*

[1] College of Mathematics and Physics, Beijing University of Chemical Technology, Beijing, 100029, People's Republic of China

[2]Beijing Institute of High Technology, Beijing 100094, China

**Correspondence and requests for materials should be addressed to Z. Y. Hu (huziyu@mail.buct.edu.cn) and J.X. Cheng(chengjx@tsinghua.org.cn)*

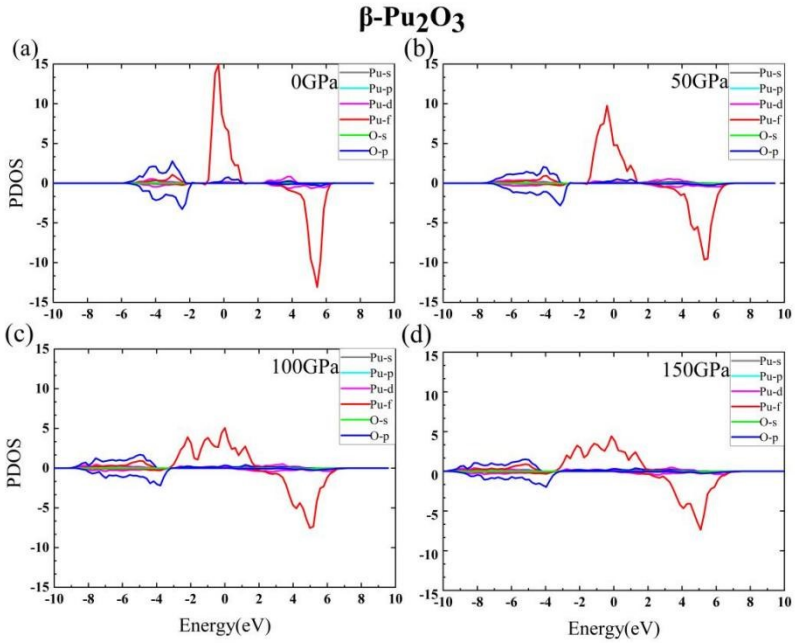

FIG. S1. The figure shows the total partial density of states (PDOS)of β-Pu_2O_3 under different pressures, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.

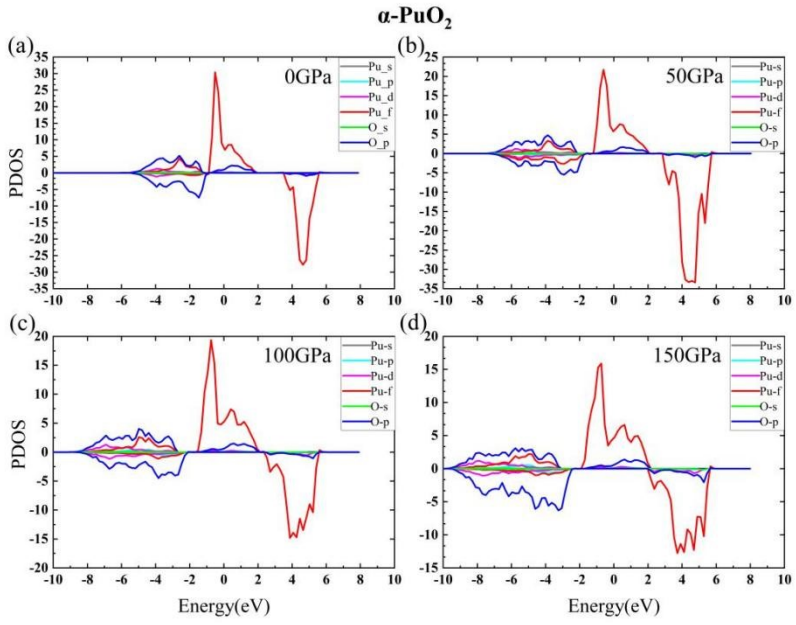

FIG. S2. The figure shows the total partial density of states (PDOS)of α-PuO_2 under different pressures, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.

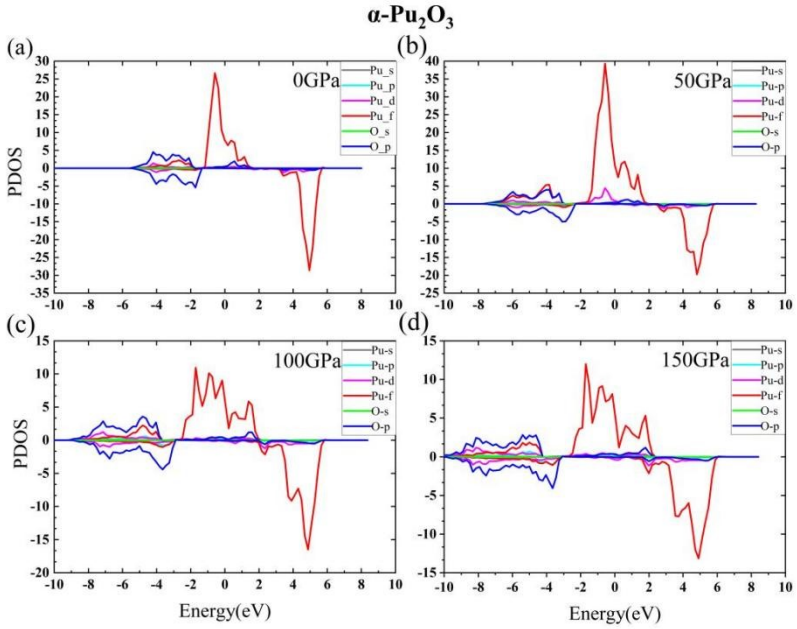

FIG. S3. The figure shows the total partial density of states (PDOS)of α-Pu_2O_3 under different pressures, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.

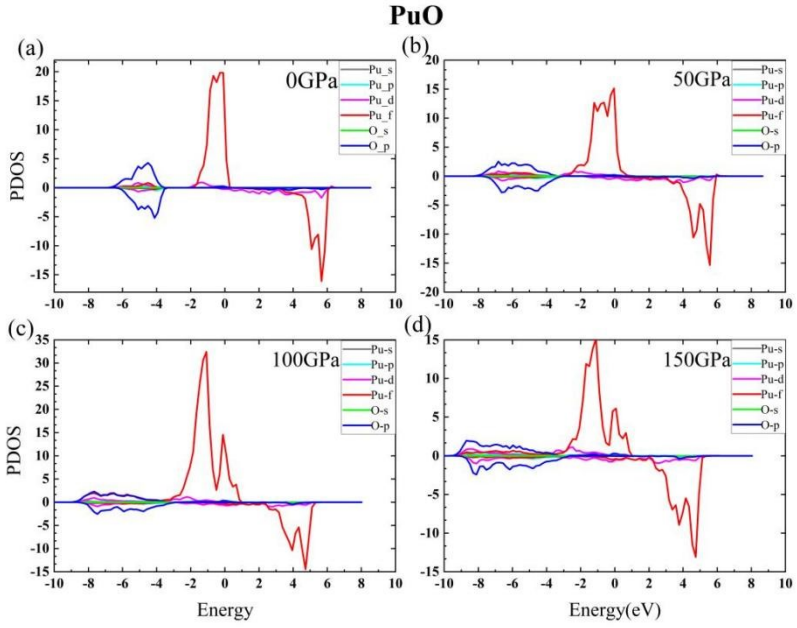

FIG. S4. The figure shows the total partial density of states (PDOS)of PuO under different pressures, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.

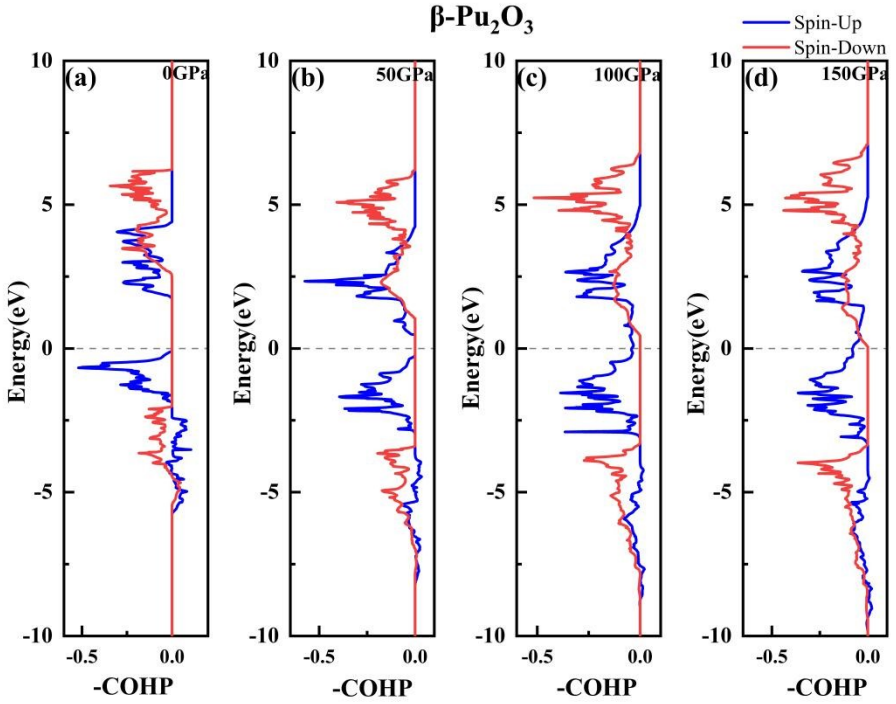

FIG. S5. Crystal orbital Hamiltonian populations of β-Pu_2O_3 under different pressures are shown, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.The red line is represented by electrons that spin downwards and the blue line is represented by electrons that spin upwards.

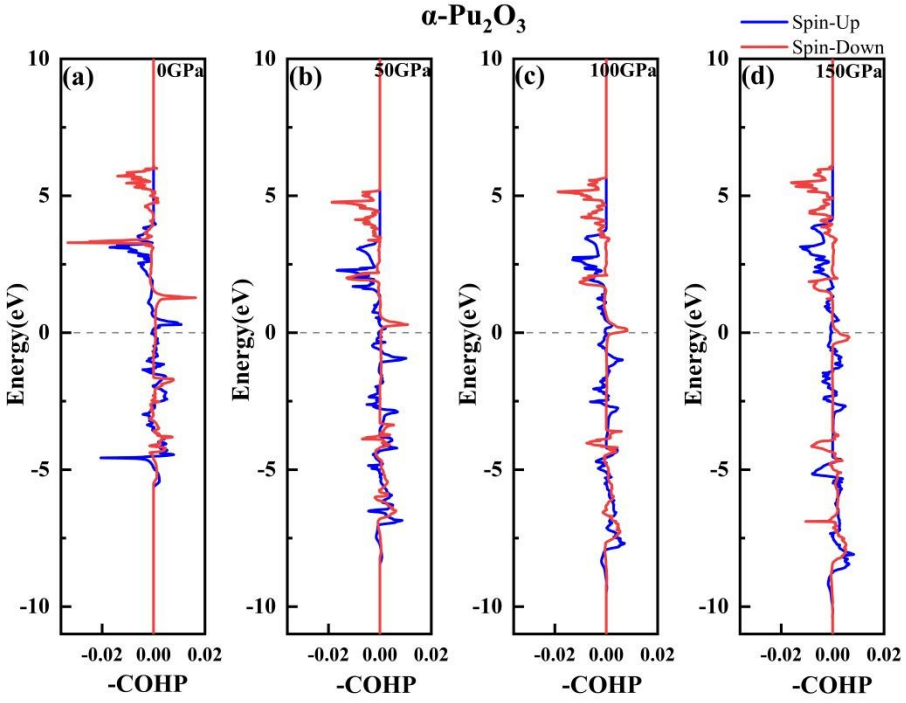

FIG. S6. Crystal orbital Hamiltonian populations of α-Pu_2O_3 under different pressures are shown, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.The red line is represented by electrons that spin

downwards and the blue line is represented by electrons that spin upwards.

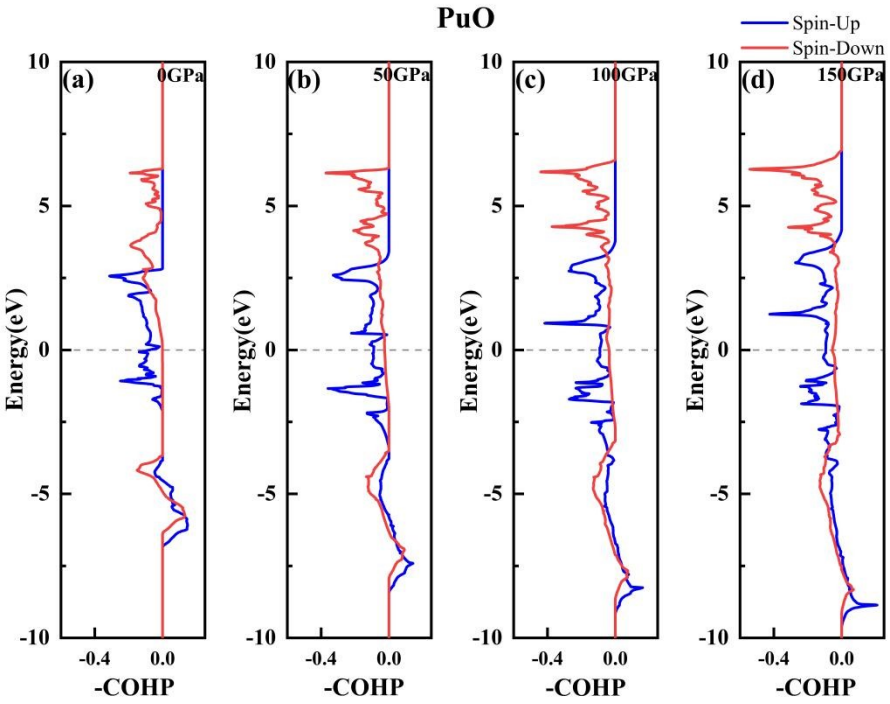

FIG. S7. Crystal orbital Hamiltonian populations of PuO under different pressures are shown, (a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.The red line is represented by electrons that spin downwards and the blue line is represented by electrons that spin upwards.

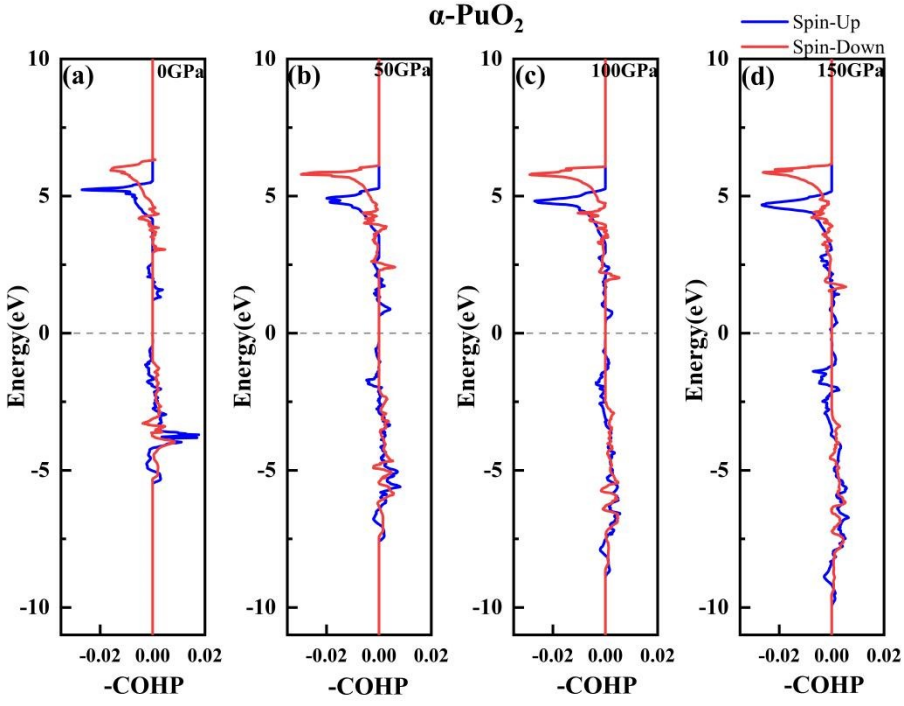

FIG. S8. Crystal orbital Hamiltonian populations of α-PuO_2 under different pressures are shown,

(a)0GPa, (b)50GPa, (c)100GPa, (d)150GPa.The red line is represented by electrons that spin downwards and the blue line is represented by electrons that spin upwards.

Table.S1. Lattice parameters of β-Pu_2O_3, α-Pu_2O_3, PuO and α-PuO_2 under 150GPa high pressures, include Chemical formula, Crystal system, lattice constant and Pu-O bond length.

Plutonium Oxide	Chemical formula	Crystal system	Pu Coordination Number	Lattice constant (Å)	Pu-O bond length (Å)
β-Pu_2O_3	Pu_2O_3	*P3m1*	CN=4	a=3.57, c=4.49	2.643
α-Pu_2O_3	Pu_4O_6	*Pn3m*	CN=6	a=4.97	2.364
PuO	Pu_4O_4	*Fm3m*	CN=8	a=5.38	2.550
α-PuO_2	Pu_4O_8	*Fm3m*	CN=8	a=4.81	2.357

Lists of structural coordination

β -Pu2O3.cif

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# CRYSTAL DATA
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data_VESTA_phase_1

_chemical_name_common                  'Pu2 O3                                '
_cell_length_a                         3.881091
_cell_length_b                         3.881091
_cell_length_c                         5.855335
_cell_angle_alpha                      90.000000
_cell_angle_beta                       90.000000
_cell_angle_gamma                      120.000000
_cell_volume                           76.381842
_space_group_name_H-M_alt              'P 1'
_space_group_IT_number                 1

loop_
_space_group_symop_operation_xyz
   'x, y, z'

loop_
   _atom_site_label
   _atom_site_occupancy
   _atom_site_fract_x
   _atom_site_fract_y
   _atom_site_fract_z
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    _atom_site_adp_type
    _atom_site_U_iso_or_equiv
    _atom_site_type_symbol
    Pu1        1.0    0.333333    0.666667    0.239301    Uiso    ? Pu
    Pu2        1.0    0.666667    0.333333    0.760699    Uiso    ? Pu
    O1         1.0   -0.000000   -0.000000   -0.000000    Uiso    ? O
    O2         1.0    0.333333    0.666667    0.646986    Uiso    ? O
    O3         1.0    0.666667    0.333333    0.353014    Uiso    ? O
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α -Pu2O3.cif

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 CRYSTAL DATA
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data_VESTA_phase_1

_chemical_name_common                  'Pu4 O6                                '
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_cell_length_c                         5.459193
_cell_angle_alpha                      90.000000
_cell_angle_beta                       90.000000
_cell_angle_gamma                      90.000000
_cell_volume                           162.699193
_space_group_name_H-M_alt              'P 1'
_space_group_IT_number                 1

loop_
_space_group_symop_operation_xyz
   'x, y, z'

loop_
    _atom_site_label
    _atom_site_occupancy
    _atom_site_fract_x
    _atom_site_fract_y
    _atom_site_fract_z
    _atom_site_adp_type
    _atom_site_U_iso_or_equiv
    _atom_site_type_symbol
    Pu1        1.0    0.750000    0.750000    0.250000    Uiso    ? Pu
    Pu2        1.0    0.250000    0.750000    0.750000    Uiso    ? Pu
    Pu3        1.0    0.750000    0.250000    0.750000    Uiso    ? Pu
    Pu4        1.0    0.250000    0.250000    0.250000    Uiso    ? Pu
```

O1	1.0	0.500000	0.000000	0.000000	Uiso	? O
O2	1.0	-0.000000	-0.000000	0.500000	Uiso	? O
O3	1.0	0.500000	0.000000	0.500000	Uiso	? O
O4	1.0	0.000000	0.500000	0.500000	Uiso	? O
O5	1.0	0.000000	0.500000	0.000000	Uiso	? O
O6	1.0	0.500000	0.500000	-0.000000	Uiso	? O

α -PuO2.cif

CRYSTAL DATA

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data_VESTA_phase_1

_chemical_name_common 'Pu4 O8 '

_cell_length_a 5.442401

_cell_length_b 5.442401

_cell_length_c 5.442401

_cell_angle_alpha 90.000000

_cell_angle_beta 90.000000

_cell_angle_gamma 90.000000

_cell_volume 161.202435

_space_group_name_H-M_alt 'P 1'

_space_group_IT_number 1

loop_

_space_group_symop_operation_xyz

'x, y, z'

loop_

_atom_site_label

_atom_site_occupancy

_atom_site_fract_x

_atom_site_fract_y

_atom_site_fract_z

_atom_site_adp_type

_atom_site_U_iso_or_equiv

_atom_site_type_symbol

Pu1	1.0	0.000000	-0.000000	0.000000	Uiso	? Pu
Pu2	1.0	0.000000	0.500000	0.500000	Uiso	? Pu
Pu3	1.0	0.500000	-0.000000	0.500000	Uiso	? Pu
Pu4	1.0	0.500000	0.500000	0.000000	Uiso	? Pu
O1	1.0	0.250000	0.250000	0.750000	Uiso	? O
O2	1.0	0.250000	0.750000	0.750000	Uiso	? O
O3	1.0	0.250000	0.750000	0.250000	Uiso	? O

O4	1.0	0.250000	0.250000	0.250000	Uiso	? O
O5	1.0	0.750000	0.250000	0.250000	Uiso	? O
O6	1.0	0.750000	0.750000	0.250000	Uiso	? O
O7	1.0	0.750000	0.750000	0.750000	Uiso	? O
O8	1.0	0.750000	0.250000	0.750000	Uiso	? O

PuO.cif

CRYSTAL DATA

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_cell_length_b	5.099202
_cell_length_c	5.099202
_cell_angle_alpha	90.000000
_cell_angle_beta	90.000000
_cell_angle_gamma	90.000000
_cell_volume	132.588717
_space_group_name_H-M_alt	'P 1'
_space_group_IT_number	1

loop_

_space_group_symop_operation_xyz

'x, y, z'

loop_

_atom_site_label

_atom_site_occupancy

_atom_site_fract_x

_atom_site_fract_y

_atom_site_fract_z

_atom_site_adp_type

_atom_site_U_iso_or_equiv

_atom_site_type_symbol

Pu1	1.0	0.500000	0.000000	0.000000	Uiso	? Pu
Pu2	1.0	0.500000	0.500000	0.500000	Uiso	? Pu
Pu3	1.0	-0.000000	-0.000000	0.500000	Uiso	? Pu
Pu4	1.0	0.000000	0.500000	-0.000000	Uiso	? Pu
O1	1.0	0.000000	0.000000	0.000000	Uiso	? O
O2	1.0	0.000000	0.500000	0.500000	Uiso	? O
O3	1.0	0.500000	-0.000000	0.500000	Uiso	? O

O4 1.0 0.500000 0.500000 0.000000 Uiso ? O
0.0000000000000000 0.5000000000000000 0.5000000000000000
0.5000000000000000 -0.0000000000000000 0.5000000000000000
0.5000000000000000 0.5000000000000000 0.0000000000000000