Supporting Information

The Energetics of Carbonated PuO_2 Surfaces Affects Nanoparticle Morphology: A DFT+U Study

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Section S1: Bulk Structures and Magnetisation - PuO_2 and Pu_2O_3

Table S1 *- The final minimised structure of the bulk 12 atom unit cell. The unit cell is given in angstrom (Å), the ionic positions are given as fractional coordinates and the initial and final magnetic vectors are presented. Oxygen atoms always have initial and final magnetics vectors of 0 μ_B. The experimental structure was from D. Taylor, Trans. J. Br. Ceram. Soc., 1984,* ***83****, 32–37*

Unit Cell Vectors			
	X	Y	Z
	5.41746	0.00000	0.00000
	0.00000	5.41746	0.00000
	0.00000	0.00000	5.41746

Atom	Coordinates (Direct)			Initial Magnetic Vector (μ_B)			Final Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z	X	Y	Z
Pu A	0.00000	0.00000	0.00000	1.0	1.0	1.0	2.2	2.2	2.2
Pu B	0.50000	0.50000	0.00000	-1.0	-1.0	1.0	-2.2	-2.2	2.2
Pu C	0.50000	0.00000	0.50000	-1.0	1.0	-1.0	-2.2	2.2	-2.2
Pu D	0.00000	0.50000	0.50000	1.0	-1.0	-1.0	2.2	-2.2	-2.2
O	0.25000	0.25000	0.25000						
O	0.25000	0.25000	0.75000						
O	0.25000	0.75000	0.25000						
O	0.25000	0.75000	0.75000						
O	0.75000	0.75000	0.75000						
O	0.75000	0.75000	0.25000						
O	0.75000	0.25000	0.75000						
O	0.75000	0.25000	0.25000						

Table S2 *- The final structure and magnetisation of bulk Pu_2O_3. The unit cell is given in angstrom (Å), the ionic positions are given as fractional coordinates and the initial and final magnetic vectors are presented. Oxygen atoms always have initial and final magnetics vectors of 0 μ_B. The experimental structure was obtained from T. D. Chikalla, C. E. McNeilly and R. E. Skavdahl, J. Nucl. Mater., 1964,* ***12****, 131–141.*

Unit Cell Vectors			
	11.08920	0.00000	0.00000
	0.00000	11.08929	0.00000
	0.00000	0.00000	11.08903

Atom	Coordinates (Direct)			Initial Magnetic Vector (μ_B)			Final Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z	X	Y	Z
Pu A	0.00004	0.00000	0.99991	1.00	1.00	1.00	2.77	2.79	2.78
Pu A	0.00005	0.00005	0.49998	1.00	1.00	1.00	-2.81	2.77	2.76
Pu A	0.00001	0.49997	0.00018	1.00	1.00	1.00	2.79	2.79	-2.76
Pu A	0.50003	0.99993	0.99990	1.00	1.00	1.00	2.76	-2.79	2.80
Pu A	0.49999	0.49998	0.00007	1.00	1.00	1.00	-2.74	2.82	2.78
Pu A	0.49994	0.99996	0.49996	1.00	1.00	1.00	2.79	2.76	-2.80
Pu A	0.99982	0.50004	0.50011	1.00	1.00	1.00	2.79	-2.80	2.76
Pu A	0.49996	0.50002	0.49995	1.00	1.00	1.00	2.80	2.77	2.78
Pu B	0.27814	0.00006	0.25003	-1.00	-1.00	1.00	-4.80	0.13	-0.19
Pu B	0.74996	0.27818	0.50010	-1.00	-1.00	1.00	0.22	-4.79	0.34
Pu B	0.25007	0.77814	0.00004	-1.00	-1.00	1.00	0.27	-4.78	0.42
Pu B	0.74996	0.72185	0.00008	-1.00	-1.00	1.00	-0.22	-4.78	0.41
Pu B	0.50002	0.75007	0.27811	-1.00	-1.00	1.00	-0.14	-0.13	4.80
Pu B	0.00007	0.25004	0.77815	-1.00	-1.00	1.00	-0.31	-0.28	4.79
Pu B	0.99997	0.75004	0.72191	-1.00	-1.00	1.00	0.11	0.08	4.81
Pu B	0.27812	0.49998	0.75000	-1.00	-1.00	1.00	-4.80	-0.17	-0.11
Pu C	0.77821	0.00012	0.24991	-1.00	1.00	-1.00	-4.81	0.06	-0.08
Pu C	0.72185	0.99989	0.74986	-1.00	1.00	-1.00	-4.80	0.13	0.04
Pu C	0.24995	0.72186	0.49995	-1.00	1.00	-1.00	-0.26	4.79	-0.31
Pu C	0.75001	0.22186	0.99986	-1.00	1.00	-1.00	-0.16	4.80	-0.26
Pu C	0.25007	0.27813	0.00004	-1.00	1.00	-1.00	0.14	4.80	-0.23
Pu C	0.50000	0.24993	0.72179	-1.00	1.00	-1.00	0.04	0.16	-4.80
Pu C	0.00004	0.75001	0.22186	-1.00	1.00	-1.00	-0.11	-0.05	-4.80
Pu C	0.99993	0.25006	0.27817	-1.00	1.00	-1.00	-0.18	0.15	-4.80
Pu D	0.72184	0.49997	0.25003	1.00	-1.00	-1.00	4.81	-0.09	-0.02
Pu D	0.22189	0.99994	0.74999	1.00	-1.00	-1.00	4.81	0.04	0.09
Pu D	0.24993	0.22191	0.50000	1.00	-1.00	-1.00	-0.02	-4.81	-0.05
Pu D	0.50002	0.24989	0.22183	1.00	-1.00	-1.00	0.14	0.01	-4.80
Pu D	0.22183	0.49996	0.25009	1.00	-1.00	-1.00	4.79	-0.28	0.23
Pu D	0.75003	0.77818	0.49994	1.00	-1.00	-1.00	0.36	-4.78	-0.37
Pu D	0.50003	0.74997	0.77811	1.00	-1.00	-1.00	0.44	-0.27	-4.78
Pu D	0.77811	0.50000	0.75014	1.00	-1.00	-1.00	4.80	0.09	-0.16
O	0.14061	0.12761	0.89722						
O	0.10271	0.14038	0.62759						
O	0.89720	0.64033	0.87248						
O	0.39724	0.85973	0.12760						

O	0.10277	0.85943	0.87242
O	0.62761	0.10278	0.14028
O	0.87246	0.89726	0.64027
O	0.12762	0.39727	0.85975
O	0.87238	0.10274	0.85928
O	0.14055	0.62764	0.10280
O	0.64040	0.87226	0.89719
O	0.85949	0.12775	0.39715
O	0.85958	0.87244	0.10273
O	0.89730	0.85970	0.37237
O	0.10287	0.35951	0.12777
O	0.60286	0.14032	0.87230
O	0.89729	0.14048	0.12750
O	0.37251	0.89722	0.85943
O	0.12759	0.10282	0.35951
O	0.87245	0.60276	0.14062
O	0.12769	0.89723	0.14042
O	0.85979	0.37239	0.89737
O	0.35973	0.12760	0.10279
O	0.14047	0.87234	0.60282
O	0.60265	0.64049	0.12762
O	0.39719	0.14048	0.37239
O	0.89716	0.35954	0.62774
O	0.60276	0.35945	0.37239
O	0.12751	0.60282	0.64043
O	0.37237	0.39716	0.14042
O	0.62768	0.89727	0.35950
O	0.37234	0.60282	0.35933
O	0.64034	0.12754	0.60271
O	0.14018	0.37241	0.39737
O	0.35949	0.62754	0.89715
O	0.35955	0.37243	0.60271
O	0.39730	0.35946	0.87234
O	0.60275	0.85949	0.62758

Section S2: Surface Structure and Magnetisation - Stoichiometric and Reduced Slabs

Table S3 - *The simulation cell dimensions for the {100}, {110} and {111} slabs.*

Slab								
100			110			111		
X	Y	Z	X	Y	Z	X	Y	Z
Unit Cell			Unit Cell			Unit Cell		
0.00000	7.66144	0.00000	10.83492	0.00000	0.00000	7.66144	0.00000	0.00000
0.00000	0.00000	7.66144	0.00000	7.66144	0.00000	3.83072	6.63501	0.00000
40.00000	0.00000	0.00000	0.00000	0.00000	33.40753	0.00000	0.00000	34.07497

Table S4 *- The initial magnetisation of the plutonium atoms in the {100}, {110} and {111} PuO_2 slabs. Oxygen atoms always have initial and final magnetics vectors of 0 μ_B.*

		Initial Magnetisation		
Slab	Plutonium Atom	X	Y	Z
100	Pu A	2.20	3.10	0.00
	Pu B	2.20	-3.10	0.00
	Pu C	-2.20	0.00	-3.10
	Pu D	2.20	0.00	-3.10
110	Pu A	-1.90	-3.28	0.00
	Pu B	1.90	0.00	3.28
	Pu C	1.90	0.00	-3.28
	Pu D	-1.90	3.28	0.00
111	Pu A	-3.10	-1.79	-1.26
	Pu B	0.00	3.58	-1.26
	Pu C	3.10	-1.79	-1.26
	Pu D	0.00	0.00	3.79

Table S5 - *The final minimised structure of the stoichiometric 3K AFM {100} configuration using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00029	0.99992	0.08299	3.74	0.78	0.02
Pu A	0.50029	0.49992	0.08299	3.74	0.78	0.02
Pu A	0.00008	0.00002	0.21925	2.18	3.11	0.08
Pu A	0.50008	0.50002	0.21925	2.18	3.11	0.08
Pu A	0.99992	0.99998	0.35478	2.18	3.11	0.08
Pu A	0.49992	0.49998	0.35478	2.18	3.11	0.08
Pu A	0.99971	0.00008	0.49104	3.74	0.78	0.02
Pu A	0.49971	0.50008	0.49104	3.74	0.78	0.02
Pu B	0.99972	0.49992	0.08299	3.74	-0.77	0.02
Pu B	0.49972	0.99992	0.08299	3.74	-0.77	0.02
Pu B	0.99992	0.50002	0.21925	2.18	-3.11	0.08
Pu B	0.49992	0.00002	0.21925	2.18	-3.11	0.08
Pu B	0.00008	0.49998	0.35478	2.18	-3.11	0.08
Pu B	0.50008	0.99998	0.35478	2.18	-3.11	0.08
Pu B	0.00028	0.50008	0.49104	3.74	-0.77	0.02
Pu B	0.50028	0.00008	0.49104	3.74	-0.77	0.02
Pu C	0.25000	0.75008	0.15138	-2.30	0.00	-3.03
Pu C	0.75000	0.25008	0.15138	-2.30	0.00	-3.03
Pu C	0.25000	0.75000	0.28701	-2.22	0.00	-3.09
Pu C	0.75000	0.25000	0.28701	-2.22	0.00	-3.09
Pu C	0.25000	0.74992	0.42265	-2.30	0.00	-3.03
Pu C	0.75000	0.24992	0.42265	-2.30	0.00	-3.03
Pu D	0.25000	0.24994	0.15140	2.25	0.00	-3.06
Pu D	0.75000	0.74994	0.15140	2.25	0.00	-3.06
Pu D	0.25000	0.25000	0.28701	2.07	0.00	-3.19
Pu D	0.75000	0.75000	0.28701	2.07	0.00	-3.19
Pu D	0.25000	0.25006	0.42263	2.25	0.00	-3.06
Pu D	0.75000	0.75006	0.42263	2.25	0.00	-3.06
O	0.25000	0.50000	0.05720			
O	0.75000	0.00000	0.05720			
O	0.25000	0.99982	0.05697			
O	0.75000	0.49982	0.05697			
O	0.25000	0.49987	0.12193			
O	0.75000	0.99987	0.12193			
O	0.49998	0.74992	0.11201			
O	0.99998	0.24992	0.11201			
O	0.50002	0.24992	0.11201			
O	0.00002	0.74992	0.11201			
O	0.25000	0.99998	0.12145			
O	0.75000	0.49998	0.12145			
O	0.25000	0.50007	0.18735			
O	0.75000	0.00007	0.18735			
O	0.00001	0.25002	0.18341			
O	0.50001	0.75002	0.18341			
O	0.99999	0.75002	0.18341			

O	0.49999	0.25002	0.18341
O	0.25000	0.99995	0.18687
O	0.75000	0.49995	0.18687
O	0.25000	0.50004	0.25385
O	0.75000	0.00004	0.25385
O	0.49999	0.75004	0.25262
O	0.99999	0.25004	0.25262
O	0.50001	0.25004	0.25262
O	0.00001	0.75004	0.25262
O	0.25000	0.00003	0.25340
O	0.75000	0.50003	0.25340
O	0.25000	0.49997	0.32063
O	0.75000	0.99997	0.32063
O	0.99999	0.24996	0.32140
O	0.49999	0.74996	0.32140
O	0.00001	0.74996	0.32140
O	0.50001	0.24996	0.32140
O	0.25000	0.99996	0.32018
O	0.75000	0.49996	0.32018
O	0.25000	0.50005	0.38716
O	0.75000	0.00005	0.38716
O	0.50001	0.74998	0.39062
O	0.00001	0.24998	0.39062
O	0.49999	0.24998	0.39062
O	0.99999	0.74998	0.39062
O	0.25000	0.99993	0.38668
O	0.75000	0.49993	0.38668
O	0.25000	0.50002	0.45258
O	0.75000	0.00002	0.45258
O	0.99998	0.25008	0.46202
O	0.49998	0.75008	0.46202
O	0.00002	0.75008	0.46202
O	0.50002	0.25008	0.46202
O	0.25000	0.00013	0.45210
O	0.75000	0.50013	0.45210
O	0.25000	0.50018	0.51706
O	0.75000	0.00018	0.51706
O	0.25000	0.00000	0.51683
O	0.75000	0.50000	0.51683

Table S6 - *The final minimised structure of the Reduced {100} using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00105	0.99977	0.08434	3.84	-0.13	-0.01
Pu A	0.47786	0.49988	0.08433	4.20	-2.31	0.25
Pu A	0.99824	0.00006	0.21934	2.18	3.11	0.08
Pu A	0.50033	0.50005	0.21919	2.16	3.13	0.08
Pu A	0.99821	0.00002	0.35470	2.19	3.11	0.08
Pu A	0.50027	0.50004	0.35485	2.18	3.11	0.08
Pu A	0.00180	-0.00009	0.48964	3.84	0.21	-0.03
Pu A	0.47772	0.50033	0.48974	3.84	2.52	-1.39
Pu B	0.02228	0.49981	0.08436	4.20	2.23	-0.66
Pu B	0.49759	-0.00007	0.08437	3.76	-0.78	0.03
Pu B	0.99970	0.50006	0.21919	2.16	-3.13	0.08
Pu B	0.50176	0.00006	0.21934	2.18	-3.11	0.08
Pu B	-0.00021	0.50003	0.35485	2.18	-3.11	0.08
Pu B	0.50185	0.00002	0.35470	2.19	-3.11	0.08
Pu B	0.02194	0.50025	0.48976	4.10	-2.49	-0.39
Pu B	0.49835	-0.00001	0.48961	3.83	-0.34	-0.03
Pu C	0.25009	0.75284	0.15234	-2.28	0.01	-3.04
Pu C	0.75005	0.25275	0.15055	-2.22	-0.01	-3.09
Pu C	0.25002	0.75027	0.28702	-2.23	0.00	-3.08
Pu C	0.75002	0.25038	0.28702	-2.23	0.00	-3.08
Pu C	0.25002	0.75256	0.42169	-2.21	0.00	-3.10
Pu C	0.74999	0.25263	0.42347	-2.31	0.00	-3.02
Pu D	0.25002	0.24731	0.15236	2.25	0.00	-3.07
Pu D	0.75003	0.74728	0.15058	2.20	0.00	-3.11
Pu D	0.25002	0.24978	0.28702	2.08	0.00	-3.18
Pu D	0.75002	0.74968	0.28702	2.08	0.00	-3.18
Pu D	0.25005	0.24772	0.42168	2.18	0.00	-3.13
Pu D	0.75002	0.74770	0.42342	2.22	0.00	-3.09
O	0.25003	0.49918	0.05271			
O	0.74973	0.00007	0.05710			
O	0.24962	0.99932	0.05766			
O	0.25010	0.49997	0.12144			
O	0.75016	0.99981	0.12136			
O	0.49701	0.75494	0.11398			
O	0.00310	0.24464	0.11397			
O	0.49702	0.24500	0.11398			
O	0.00315	0.75492	0.11399			
O	0.25062	0.00003	0.12206			
O	0.75012	0.49994	0.11758			
O	0.25003	0.50014	0.18706			
O	0.74997	0.00010	0.18675			
O	0.99918	0.25045	0.18404			
O	0.50086	0.74968	0.18403			
O	0.99919	0.74970	0.18404			
O	0.50084	0.25045	0.18403			
O	0.24996	0.00002	0.18722			

O	0.75003	0.49997	0.18550
O	0.25002	0.50006	0.25363
O	0.75000	0.00007	0.25381
O	0.50046	0.74985	0.25278
O	0.99955	0.25030	0.25280
O	0.50048	0.25029	0.25279
O	0.99957	0.74985	0.25279
O	0.25002	0.00006	0.25342
O	0.75000	0.50006	0.25307
O	0.25002	0.49999	0.32077
O	0.75003	1.00000	0.32058
O	0.99957	0.25017	0.32126
O	0.50046	0.74982	0.32124
O	0.99959	0.74982	0.32125
O	0.50048	0.25017	0.32125
O	0.25002	0.99998	0.32027
O	0.75004	0.49999	0.32060
O	0.25005	0.50015	0.38731
O	0.75003	0.00014	0.38760
O	0.50084	0.74966	0.38995
O	0.99923	0.25048	0.39002
O	0.50082	0.25048	0.39000
O	0.99924	0.74966	0.38996
O	0.25004	0.00005	0.38653
O	0.75004	0.50006	0.38821
O	0.24986	0.49994	0.45279
O	0.75000	0.00026	0.45282
O	0.00255	0.24525	0.46013
O	0.49707	0.75551	0.45989
O	0.00304	0.75555	0.45993
O	0.49733	0.24513	0.46003
O	0.24989	0.00042	0.45184
O	0.74984	0.50033	0.45640
O	0.24997	0.49955	0.52145
O	0.74992	0.99841	0.51692
O	0.25014	-0.00078	0.51632

Table S7 - *The final minimised structure of the stoichiometric 3K AFM {110} configuration using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00029	0.99992	0.08299	-1.99	-3.32	-0.04
Pu A	0.50029	0.49992	0.08299	-1.40	-3.51	0.56
Pu A	0.00008	0.00002	0.21925	-2.19	-3.10	-0.01
Pu A	0.50008	0.50002	0.21925	-2.19	-3.11	0.02
Pu A	0.99992	0.99998	0.35478	-2.19	-3.10	-0.01
Pu A	0.49992	0.49998	0.35478	-2.21	-3.09	0.04
Pu A	0.99971	0.00008	0.49104	-1.90	-3.37	0.08
Pu A	0.49971	0.50008	0.49104	-1.80	-3.32	0.62
Pu B	0.99972	0.49992	0.08299	2.42	0.00	2.94
Pu B	0.49972	0.99992	0.08299	2.04	0.00	3.20
Pu B	0.99992	0.50002	0.21925	2.23	0.00	3.07
Pu B	0.49992	0.00002	0.21925	2.20	0.00	3.09
Pu B	0.00008	0.49998	0.35478	2.26	-0.01	3.04
Pu B	0.50008	0.99998	0.35478	2.10	0.00	3.17
Pu B	0.00028	0.50008	0.49104	2.10	0.00	-3.17
Pu B	0.50028	0.00008	0.49104	2.22	0.00	-3.08
Pu C	0.25000	0.75008	0.15138	2.21	0.00	-3.09
Pu C	0.75000	0.25008	0.15138	2.22	0.00	-3.08
Pu C	0.25000	0.75000	0.28701	2.22	0.00	-3.09
Pu C	0.75000	0.25000	0.28701	2.26	0.00	-3.05
Pu C	0.25000	0.74992	0.42265	-1.97	3.34	-0.04
Pu C	0.75000	0.24992	0.42265	-1.32	3.53	0.62
Pu D	0.25000	0.24994	0.15140	-2.19	3.10	0.00
Pu D	0.75000	0.74994	0.15140	-2.19	3.11	0.02
Pu D	0.25000	0.25000	0.28701	-2.20	3.10	-0.01
Pu D	0.75000	0.75000	0.28701	-2.21	3.09	0.04
Pu D	0.25000	0.25006	0.42263	-1.94	3.35	0.09
Pu D	0.75000	0.75006	0.42263	-1.79	3.32	0.62
O	0.25000	0.50000	0.05720			
O	0.75000	0.00000	0.05720			
O	0.25000	0.99982	0.05697			
O	0.75000	0.49982	0.05697			
O	0.25000	0.49987	0.12193			
O	0.75000	0.99987	0.12193			
O	0.49998	0.74992	0.11201			
O	0.99998	0.24992	0.11201			
O	0.50002	0.24992	0.11201			
O	0.00002	0.74992	0.11201			
O	0.25000	0.99998	0.12145			
O	0.75000	0.49998	0.12145			
O	0.25000	0.50007	0.18735			
O	0.75000	0.00007	0.18735			
O	0.00001	0.25002	0.18341			
O	0.50001	0.75002	0.18341			

O	0.99999	0.75002	0.18341
O	0.49999	0.25002	0.18341
O	0.25000	0.99995	0.18687
O	0.75000	0.49995	0.18687
O	0.25000	0.50004	0.25385
O	0.75000	0.00004	0.25385
O	0.49999	0.75004	0.25262
O	0.99999	0.25004	0.25262
O	0.50001	0.25004	0.25262
O	0.00001	0.75004	0.25262
O	0.25000	0.00003	0.25340
O	0.75000	0.50003	0.25340
O	0.25000	0.49997	0.32063
O	0.75000	0.99997	0.32063
O	0.99999	0.24996	0.32140
O	0.49999	0.74996	0.32140
O	0.00001	0.74996	0.32140
O	0.50001	0.24996	0.32140
O	0.25000	0.99996	0.32018
O	0.75000	0.49996	0.32018
O	0.25000	0.50005	0.38716
O	0.75000	0.00005	0.38716
O	0.50001	0.74998	0.39062
O	0.00001	0.24998	0.39062
O	0.49999	0.24998	0.39062
O	0.99999	0.74998	0.39062
O	0.25000	0.99993	0.38668
O	0.75000	0.49993	0.38668
O	0.25000	0.50002	0.45258
O	0.75000	0.00002	0.45258
O	0.99998	0.25008	0.46202
O	0.49998	0.75008	0.46202
O	0.00002	0.75008	0.46202
O	0.50002	0.25008	0.46202
O	0.25000	0.00013	0.45210
O	0.75000	0.50013	0.45210
O	0.25000	0.50018	0.51706
O	0.75000	0.00018	0.51706
O	0.25000	0.00000	0.51683
O	0.75000	0.50000	0.51683

Table S8 *- The final minimised structure of the Reduced {110} using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00102	0.49927	0.06458	-1.69	-3.47	0.16
Pu A	0.50860	0.51705	0.06198	-3.52	-2.72	-1.80
Pu A	0.99804	0.99860	0.17571	-2.22	-3.08	0.05
Pu A	0.49843	0.00017	0.17513	-2.23	-3.08	-0.06
Pu A	0.99694	0.50208	0.28817	-2.20	-3.10	0.01
Pu A	0.49939	0.50012	0.28954	-2.19	-3.10	-0.09
Pu A	0.00407	0.99900	0.39877	-1.77	-3.43	-0.18
Pu A	0.51201	0.98313	0.40183	1.14	-2.15	-4.14
Pu B	0.24994	0.75039	0.11362	2.30	0.01	3.03
Pu B	0.75472	0.74844	0.11563	2.29	-0.02	3.03
Pu B	0.24971	0.25053	0.23146	2.13	0.00	3.15
Pu B	0.74708	0.25005	0.23218	2.25	0.01	3.06
Pu B	0.25422	0.74996	0.35074	1.95	-0.01	3.26
Pu B	0.75350	0.74991	0.34882	2.35	0.00	2.98
Pu B	0.24174	0.25200	0.11844	2.77	-0.12	-3.93
Pu B	0.74847	0.24909	0.11537	2.31	-0.01	-3.03
Pu C	0.24825	0.75013	0.23215	2.26	0.01	-3.05
Pu C	0.74569	0.74953	0.23195	2.22	0.00	-3.08
Pu C	0.24251	0.25009	0.34384	2.59	0.00	-2.80
Pu C	0.74703	0.25002	0.35026	2.12	0.00	-3.15
Pu C	0.00109	0.99998	0.06462	-1.70	3.46	0.19
Pu C	0.50766	0.97846	0.06440	-0.83	2.27	-3.00
Pu D	0.99794	0.50116	0.17572	-2.23	3.07	0.05
Pu D	0.49832	0.50067	0.17416	-2.21	3.09	-0.05
Pu D	0.99695	0.99779	0.28812	-2.20	3.10	0.02
Pu D	0.49939	0.00015	0.28978	-2.19	3.11	-0.10
Pu D	0.00406	0.50074	0.39880	-1.78	3.43	-0.18
Pu D	0.51195	0.51700	0.40175	1.12	2.17	-4.14
O	0.11999	0.74938	0.05925			
O	0.63007	0.75766	0.05922			
O	0.37933	0.76327	0.05810			
O	0.88479	0.74875	0.06141			
O	0.12050	0.24989	0.05804			
O	0.58657	0.23642	0.05903			
O	0.87790	0.24933	0.06128			
O	0.11949	0.50921	0.11773			
O	0.62985	0.50107	0.11688			
O	0.12014	0.99199	0.11764			
O	0.62815	0.99548	0.11788			
O	0.37662	0.49828	0.11428			
O	0.87572	0.49705	0.11759			
O	0.37926	0.00736	0.11484			
O	0.87538	0.00086	0.11753			
O	0.11947	0.25021	0.17575			
O	0.62328	0.24994	0.17345			
O	0.37645	0.25058	0.17656			

O	0.87061	0.24949	0.17407
O	0.12397	0.75014	0.17542
O	0.62318	0.74931	0.17460
O	0.37305	0.75133	0.17249
O	0.87346	0.74954	0.17451
O	0.12354	0.00174	0.23210
O	0.62230	0.00198	0.23196
O	0.12349	0.49863	0.23215
O	0.62207	0.49767	0.23175
O	0.37373	0.99490	0.23228
O	0.87202	0.99885	0.23182
O	0.37383	0.50610	0.23195
O	0.87199	0.50058	0.23186
O	0.12306	0.75002	0.28871
O	0.62306	0.74999	0.28862
O	0.37480	0.75007	0.29241
O	0.87313	0.74978	0.28922
O	0.12206	0.24999	0.28924
O	0.62233	0.25001	0.29014
O	0.37239	0.25044	0.28789
O	0.87219	0.24992	0.28931
O	0.12369	0.50400	0.34548
O	0.62954	0.50448	0.34529
O	0.12378	0.99593	0.34548
O	0.62963	0.99553	0.34536
O	0.37399	0.48076	0.34915
O	0.87762	0.49749	0.34674
O	0.37414	0.01957	0.34917
O	0.87767	0.00230	0.34671
O	0.13372	0.24995	0.40199
O	0.60974	0.25009	0.40300
O	0.88842	0.24987	0.40367
O	0.12118	0.74988	0.40401
O	0.63510	0.75004	0.40480
O	0.37703	0.75017	0.40337
O	0.88877	0.74987	0.40237

Table S9 - *The final minimised structure of the stoichiometric 3K AFM {111} configuration using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.66692	0.66682	0.08245	-3.21	-1.85	-0.95
Pu A	0.33326	0.33326	0.17335	-3.09	-1.79	-1.30
Pu A	0.00000	1.00000	0.26522	-3.10	-1.79	-1.26
Pu A	0.66674	0.66674	0.35710	-3.09	-1.79	-1.30
Pu A	0.33308	0.33318	0.44800	-3.21	-1.85	-0.95
Pu B	0.66687	0.16635	0.08245	0.00	3.71	-0.95
Pu B	0.33324	0.83357	0.17334	0.00	3.57	-1.29
Pu B	0.00000	0.50000	0.26522	0.00	3.58	-1.26
Pu B	0.66676	0.16643	0.35710	0.00	3.57	-1.29
Pu B	0.33313	0.83365	0.44799	0.00	3.71	-0.95
Pu C	0.16640	0.66680	0.08245	3.21	-1.85	-0.95
Pu C	0.83357	0.33324	0.17335	3.09	-1.78	-1.30
Pu C	0.50000	1.00000	0.26522	3.10	-1.79	-1.26
Pu C	0.16643	0.66676	0.35710	3.09	-1.78	-1.30
Pu C	0.83360	0.33320	0.44799	3.21	-1.85	-0.95
Pu D	0.16674	0.16663	0.08229	0.04	-0.11	3.79
Pu D	0.83337	0.83336	0.17339	0.00	0.00	3.80
Pu D	0.50000	0.50000	0.26522	0.00	0.00	3.80
Pu D	0.16663	0.16664	0.35705	0.00	0.00	3.80
Pu D	0.83326	0.83337	0.44815	0.04	-0.11	3.79
O	0.00003	-0.00002	0.06007			
O	0.50005	0.49996	0.06043			
O	0.50013	-0.00003	0.06007			
O	0.00003	0.50007	0.06006			
O	0.33302	0.83413	0.10563			
O	0.83417	0.33298	0.10564			
O	0.83337	0.83330	0.10547			
O	0.33298	0.33299	0.10564			
O	0.66676	0.66673	0.14985			
O	0.16667	0.16668	0.15017			
O	0.16658	0.66675	0.14985			
O	0.66678	0.16656	0.14984			
O	0.99995	0.50024	0.19629			
O	0.50029	-0.00009	0.19628			
O	0.50004	0.50004	0.19627			
O	-0.00015	0.99992	0.19629			
O	0.33336	0.33330	0.24232			
O	0.83332	0.83332	0.24230			
O	0.83336	0.33332	0.24232			
O	0.33329	0.83341	0.24230			
O	0.66671	0.16659	0.28814			
O	0.16664	0.66668	0.28813			
O	0.16668	0.16668	0.28815			
O	0.66664	0.66670	0.28813			

O	0.00015	0.00008	0.33416
O	0.49996	0.49996	0.33418
O	0.49971	0.00009	0.33417
O	0.00005	0.49976	0.33415
O	0.33322	0.83344	0.38061
O	0.83342	0.33325	0.38059
O	0.83333	0.83332	0.38028
O	0.33324	0.33327	0.38060
O	0.66702	0.66701	0.42480
O	0.16663	0.16670	0.42498
O	0.16583	0.66702	0.42481
O	0.66698	0.16587	0.42482
O	0.99997	0.49993	0.47038
O	0.49987	0.00003	0.47038
O	0.49995	0.50004	0.47002
O	-0.00003	0.00002	0.47038

Table S10 - *The final minimised structure of the Reduced {111} using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

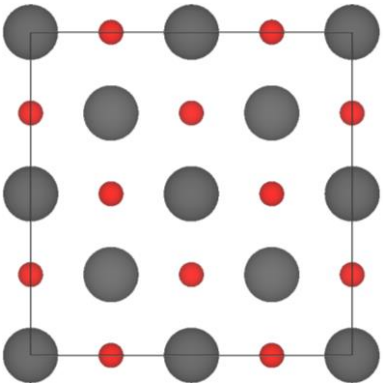
Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.66769	0.66738	0.08020	-3.26	-1.86	-0.75
Pu A	0.33502	0.33476	0.17556	-3.09	-1.78	-1.31
Pu A	0.99967	0.99898	0.26502	-3.09	-1.79	-1.29
Pu A	0.66574	0.66624	0.35782	-3.06	-1.76	-1.40
Pu A	0.32433	0.32458	0.44783	-2.98	-1.95	-3.22
Pu B	0.64887	0.17607	0.08245	-2.80	2.14	3.26
Pu B	0.33201	0.83388	0.17287	-0.01	3.62	-1.15
Pu B	0.99816	0.50240	0.26521	0.00	3.57	-1.29
Pu B	0.66585	0.16815	0.35757	-0.01	3.53	-1.39
Pu B	0.32422	0.85140	0.44792	0.20	3.56	-3.21
Pu C	0.17614	0.64866	0.08245	0.24	-3.51	3.26
Pu C	0.83440	0.33158	0.17293	3.13	-1.81	-1.15
Pu C	0.50272	0.99764	0.26502	3.09	-1.78	-1.29
Pu C	0.16700	0.66666	0.35814	3.08	-1.78	-1.33
Pu C	0.85506	0.32251	0.44603	1.00	-0.59	-3.64
Pu D	0.17771	0.17739	0.08443	0.83	0.49	3.69
Pu D	0.83410	0.83369	0.17209	-0.04	-0.01	3.80
Pu D	0.50080	0.50076	0.26561	0.04	0.02	3.80
Pu D	0.16910	0.16549	0.35478	-0.02	0.02	3.80
Pu D	0.83408	0.83330	0.45049	0.01	-0.34	3.77
O	0.51081	0.51055	0.05803			
O	0.48980	0.99577	0.05742			
O	0.99580	0.48967	0.05741			
O	0.30620	0.85558	0.10389			
O	0.85610	0.30549	0.10386			
O	0.83706	0.83692	0.10332			
O	0.33186	0.33155	0.10675			
O	0.66432	0.66355	0.14689			
O	0.17100	0.17063	0.15198			
O	0.17034	0.66202	0.15163			
O	0.66246	0.17012	0.15167			
O	0.99425	0.50394	0.19643			
O	0.50431	0.99375	0.19634			
O	0.50280	0.50257	0.19693			
O	0.00011	0.99948	0.19561			
O	0.33336	0.33302	0.24265			
O	0.83436	0.83392	0.24206			
O	0.83486	0.33164	0.24219			
O	0.33206	0.83441	0.24217			
O	0.66600	0.16816	0.28827			
O	0.16842	0.66559	0.28842			
O	0.16718	0.16676	0.28765			
O	0.66639	0.66583	0.28822			
O	0.99788	0.99711	0.33390			
O	0.50058	0.50022	0.33490			

O	0.50555	0.99689	0.33360
O	0.99729	0.50552	0.33400
O	0.33209	0.83785	0.37880
O	0.84137	0.32931	0.37871
O	0.82853	0.83586	0.38330
O	0.33228	0.33020	0.37869
O	0.66164	0.64498	0.42667
O	0.16334	0.16839	0.42357
O	0.17392	0.66320	0.42710
O	0.66197	0.19318	0.42663
O	0.98543	0.50408	0.47305
O	0.52106	0.98967	0.47248
O	0.98536	0.01067	0.47315

Section S3: Structural Configurations - Stoichiometric, Reduced and Adsorption Configurations

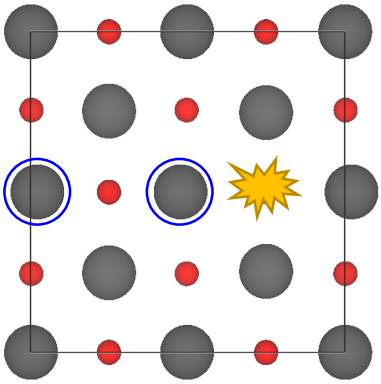
Figure S1 - *The top view images of the relaxed final configuration of a stoichiometric {100}, reduced {100} and CO_2 adsorbed on the {100} surface; Pu^{4+} in dark grey, O in red, Oxygen Vacancies are marked by a yellow star and Pu^{3+} is highlighted by a circle. Note we only drawn the top side of the slab as the bottom side is symmetric. Also note we show a side view for the {100}-Redu-CO_2 ⊥ (Configuration – IVa) for clarity.*

Stoichiometric {100)



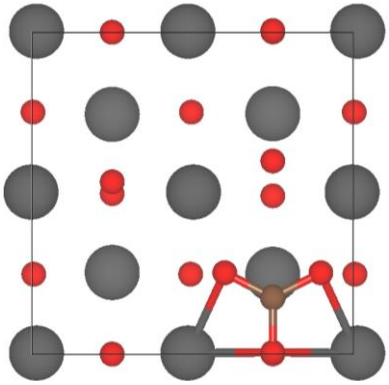
Surface Energy = 1.98eV

Reduced {100}

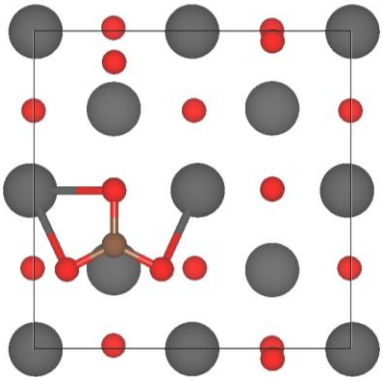


Surface Energy = 2.53eV

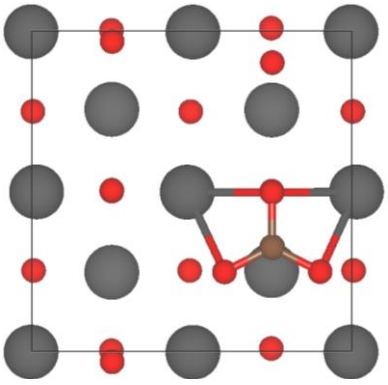
{100}-Stoich-CO_3 ||



Configuration – Ia E_{ads} = -2.69eV

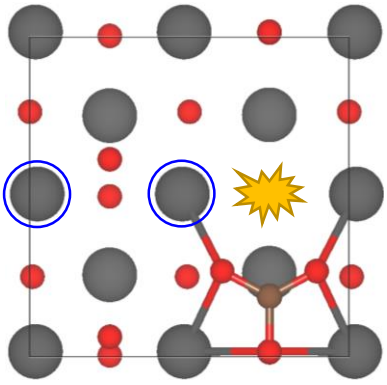


Configuration – Ib E_{ads} = -2.68eV

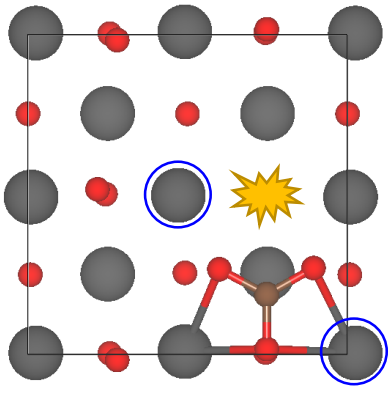


Configuration – Ic E_{ads} = -2.67eV

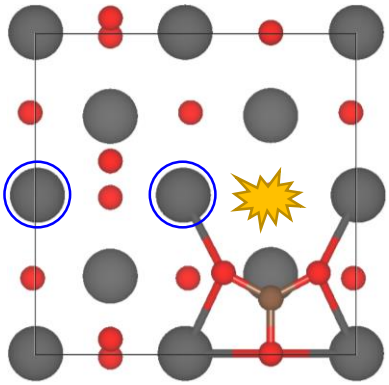
{100}-Redu-CO_3 ||



Configuration – IIa E_{ads} = -3.17eV

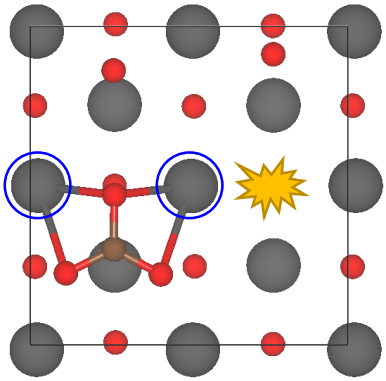


Configuration – IIb E_{ads} = -3.13eV



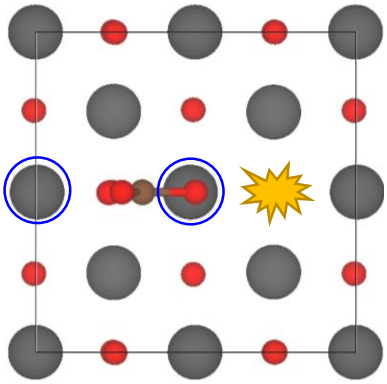
Configuration – IIc E_{ads} = -3.09eV

cont. {100}-Redu-CO_3 ∥



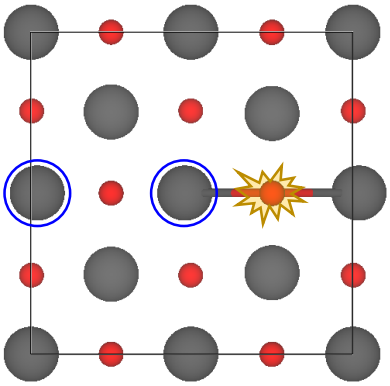
Configuration – IId E_{ads} = -2.98eV

{100}-Redu-CO_3 ⊥

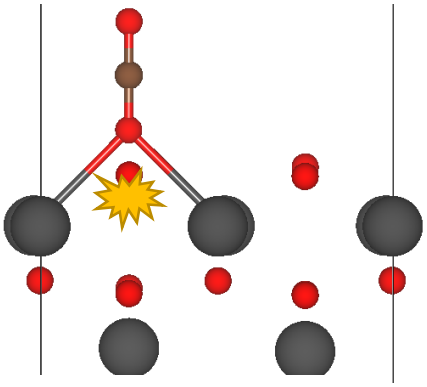


Configuration – IIIa E_{ads} = -1.17eV

{100}-Redu-CO_2 ⊥



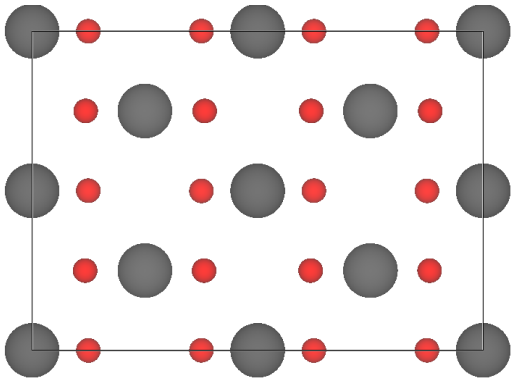
Configuration – IVa E_{ads} = -0.43eV



Configuration – IVa (Side View)

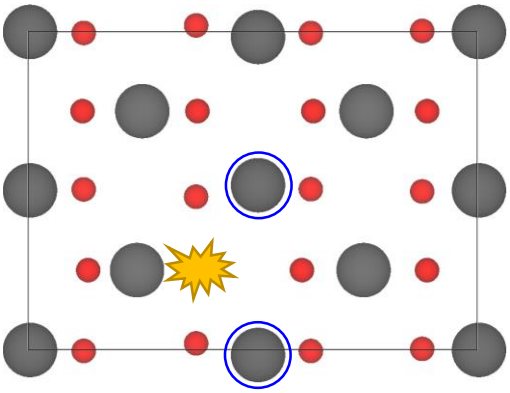
Figure S2 - *The top view images of the relaxed final configuration of a stoichiometric {110}, reduced {110} and CO_2 adsorbed on the {110} surface; Pu^{4+} in dark grey, O in red, Oxygen Vacancies are marked by a yellow star and Pu^{3+} is highlighted by a circle. Note we only drawn the top side of the slab as the bottom side is symmetric.*

Stoichiometric {110}



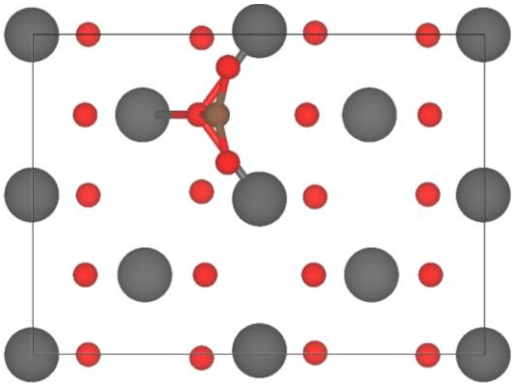
Surface Energy = 1.36eV

Reduced {110}

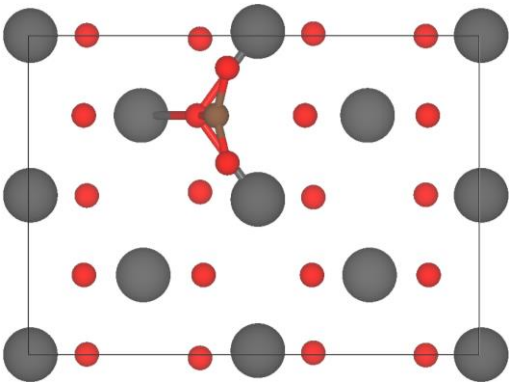


Surface Energy = 1.82eV

{110}-Stoich-CO_3 ∠

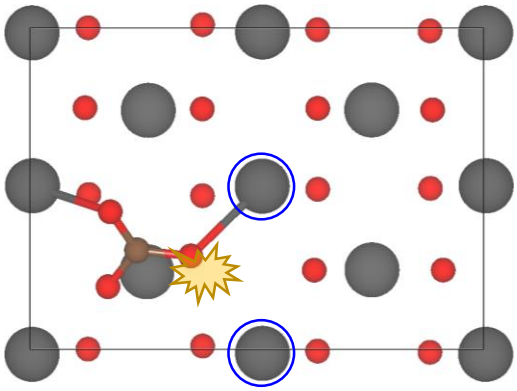


Configuration – Va E_{ads} *= -1.67*

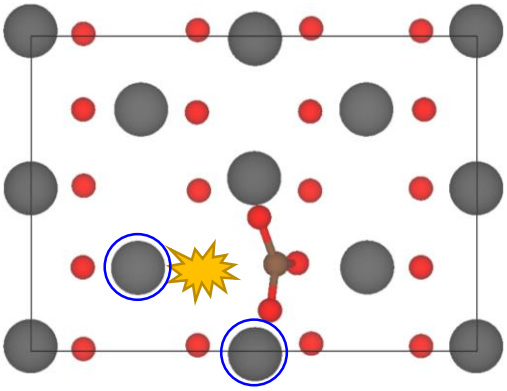


Configuration – Vb E_{ads} *= -1.67eV*

{110}-Redu-CO_3 ∠

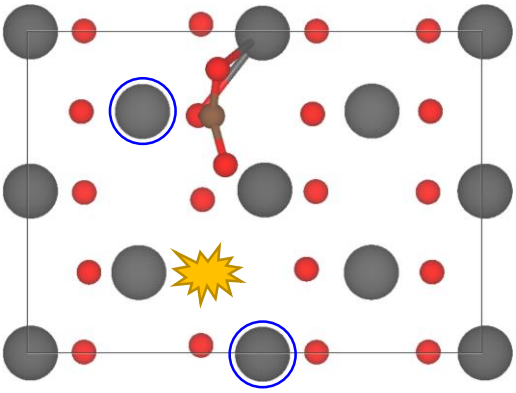


Configuration – Via E_{ads} *= -1.99eV*

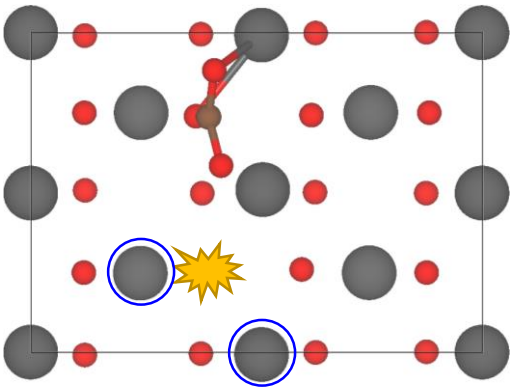


Configuration – VIb E_{ads} *= -1.41eV*

cont. {110}-Redu-CO_3 ∠

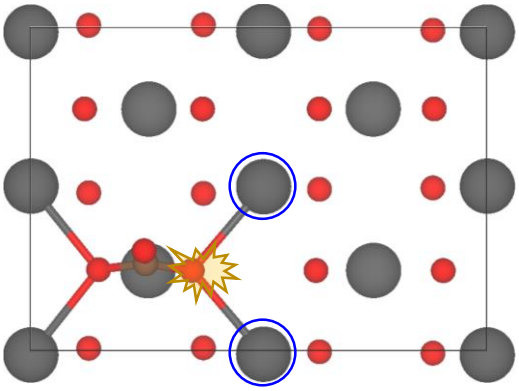


Configuration – VIc E_{ads} *= -1.40eV*



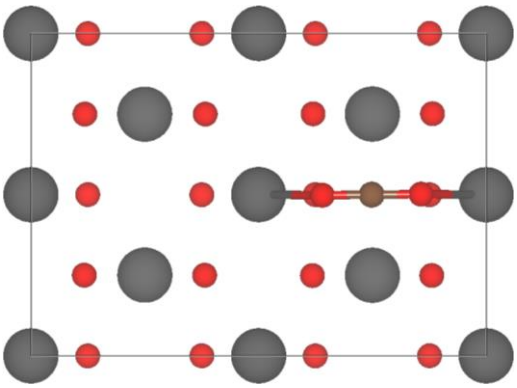
Configuration – VId E_{ads} *= -1.35eV*

{110}-Redu-CO_3 ⊥



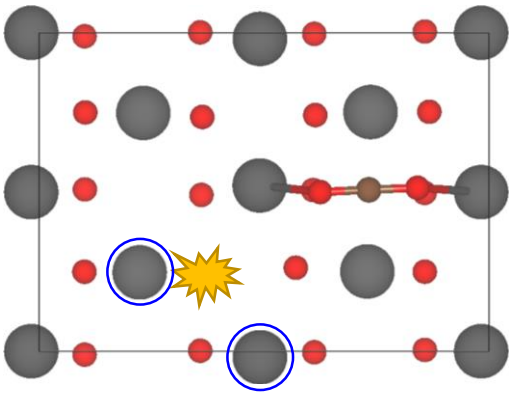
Configuration – VIIa E_{ads} *= -1.93eV*

{110}-Stoich-CO_2 ||



Configuration – VIIIa E_{ads} *= -0.35eV*

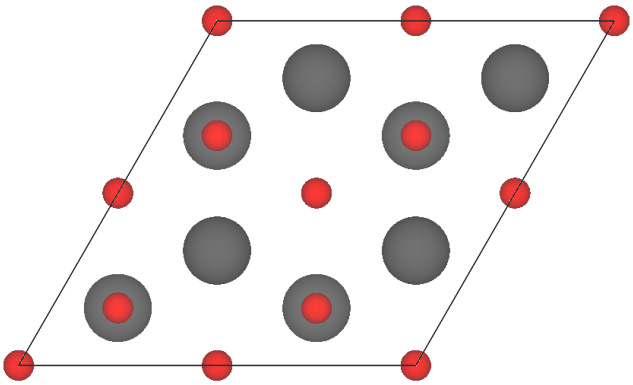
{110}-Redu-CO_2 ||



Configuration – IXa E_{ads} *= -0.32eV*

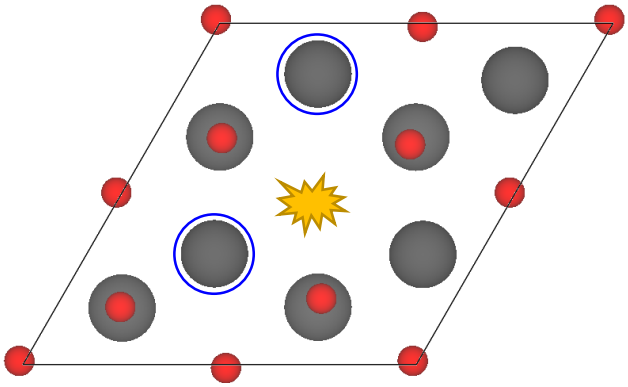
Figure S3 - *The top view images of the relaxed final configuration of a stoichiometric {111}, reduced {111} and CO_2 adsorbed on the {111} surface; Pu^{4+} in dark grey, O in red, Oxygen Vacancies are marked by a yellow star and Pu^{3+} is highlighted by a circle. Note we only drawn the top side of the slab as the bottom side is symmetric.*

Stoichiometric {111}



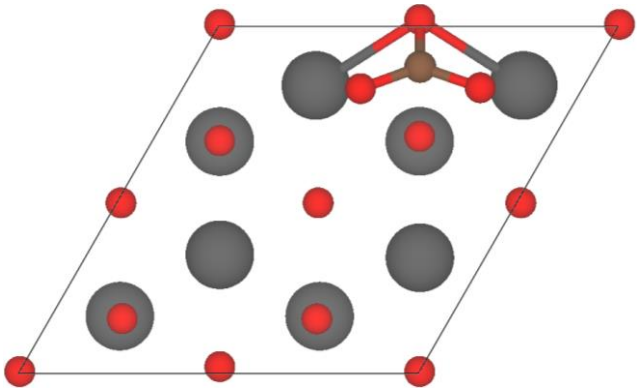
Surface Energy = 0.95eV

Reduced {111}



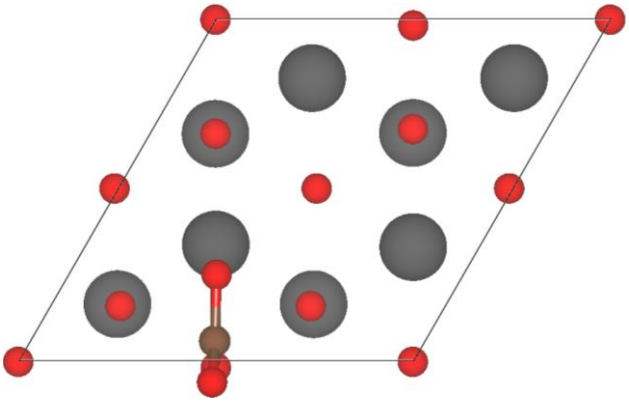
Surface Energy = 1.95eV

{111}-Stoich-CO_3 ∠



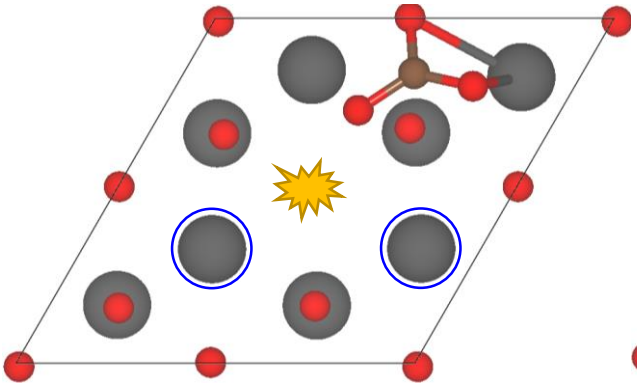
Configuration – Xa E_{ads} *= -0.82eV*

{111}-Stoich-CO_3 ⊥

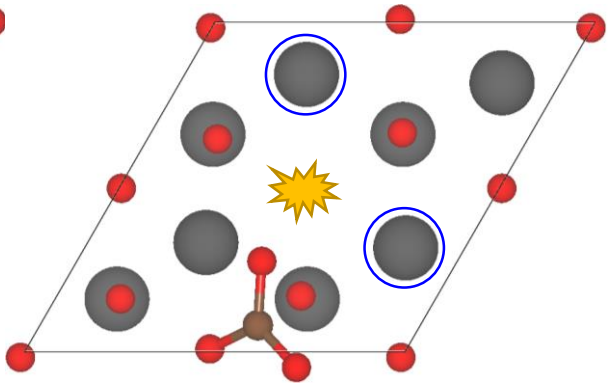


Configuration – XIa E_{ads} *= -0.45eV*

{111}-Redu-CO_3 ∠

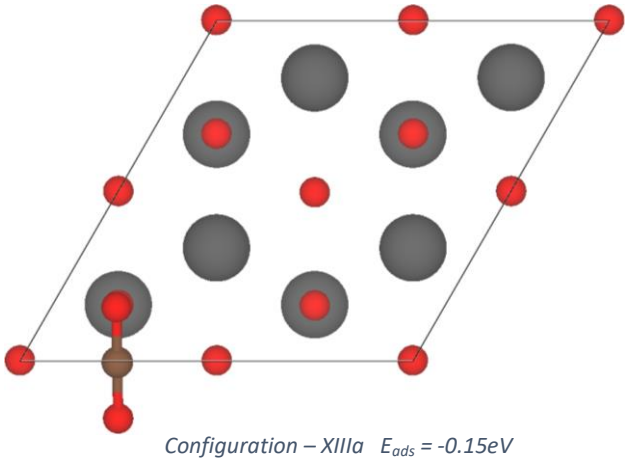


Configuration – XIIa E_{ads} *= -1.35eV*



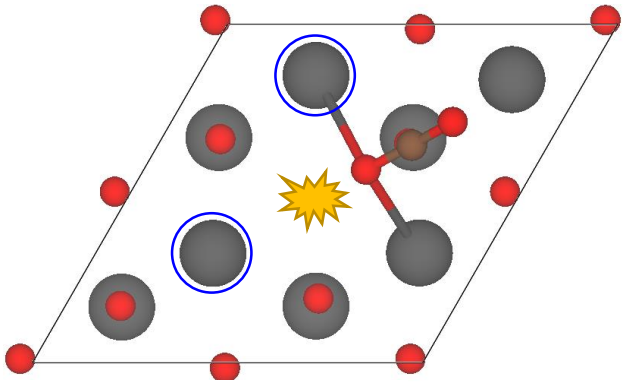
Configuration – XIIb E_{ads} *= -1.16eV*

{111}-Stoich-CO_2 ∠

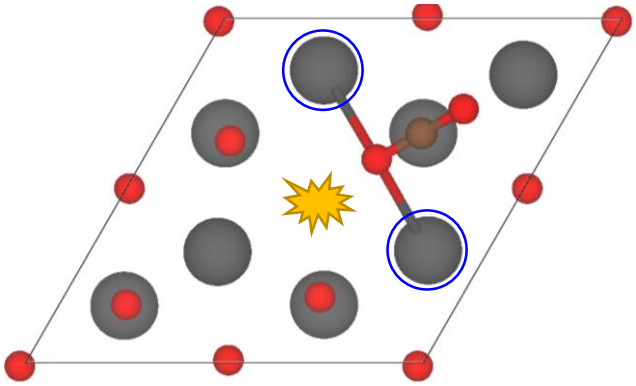


Configuration – XIIIa E_{ads} = -0.15eV

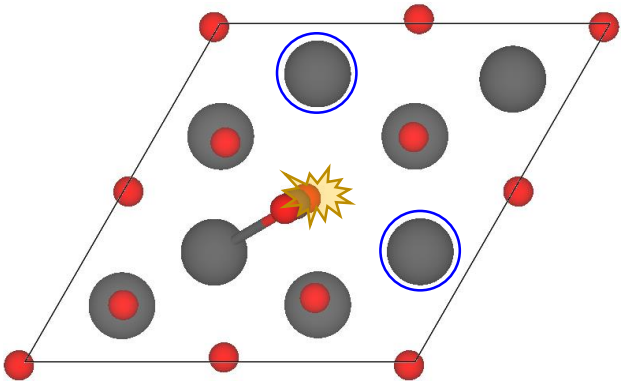
{111}-Redu-CO_2 ∠



Configuration – XIVa E_{ads} = -0.46eV



Configuration – XIVb E_{ads} = -0.35eV



Configuration – XIV E_{ads} = -0.33eV

Section S4: Surface Structure and Magnetisation - {100} Adsorption

Table S11 *- The final minimised structure of the {100}-Stoich-Flat-CO_3 ‖ configuration Ia using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.01451	0.00406	0.08488	2.23	3.06	0.49
Pu A	0.50283	0.50421	0.08200	2.67	-0.23	2.67
Pu A	0.00011	0.99908	0.21903	2.18	3.11	0.06
Pu A	0.50131	0.49961	0.21951	2.18	3.11	0.09
Pu A	0.00003	0.99910	0.35500	2.16	3.13	0.08
Pu A	0.50108	0.49965	0.35453	2.18	3.11	0.06
Pu A	0.01423	0.00426	0.48916	-2.09	3.17	0.43
Pu A	0.50297	0.50444	0.49202	2.53	0.27	-2.80
Pu B	0.99717	0.50424	0.08201	2.83	0.15	2.50
Pu B	0.48553	0.00403	0.08489	2.23	-3.06	0.49
Pu B	0.99876	0.49961	0.21951	2.18	-3.11	0.09
Pu B	0.49995	0.99908	0.21903	2.18	-3.11	0.06
Pu B	0.99889	0.49965	0.35453	2.19	-3.11	0.06
Pu B	0.49996	0.99910	0.35500	2.15	-3.13	0.08
Pu B	0.99699	0.50443	0.49202	2.55	-0.32	-2.77
Pu B	0.48561	0.00427	0.48917	2.10	3.16	-0.50
Pu C	0.25003	0.74574	0.15190	-2.24	0.00	-3.07
Pu C	0.75004	0.24895	0.15143	-2.16	0.00	-3.14
Pu C	0.25002	0.74957	0.28701	-2.23	0.00	-3.08
Pu C	0.75001	0.24879	0.28703	-2.23	0.00	-3.08
Pu C	0.25002	0.74573	0.42215	-2.30	0.02	-3.03
Pu C	0.74994	0.24882	0.42260	-2.10	-0.01	-3.18
Pu D	0.25003	0.25442	0.15220	2.13	0.00	-3.15
Pu D	0.75002	0.74778	0.15046	2.15	0.00	-3.14
Pu D	0.25002	0.24924	0.28701	2.08	0.00	-3.18
Pu D	0.75001	0.75008	0.28703	2.08	0.00	-3.19
Pu D	0.24995	0.25457	0.42186	2.22	0.02	-3.09
Pu D	0.75002	0.74786	0.42359	2.15	-0.02	-3.14
O	0.24995	0.53540	0.05779			
O	0.25003	0.99974	0.05677			
O	0.24994	0.49940	0.12207			
O	0.74995	0.00158	0.11698			
O	0.49650	0.75270	0.11396			
O	0.00595	0.24902	0.11472			
O	0.49410	0.24913	0.11472			
O	0.00349	0.75280	0.11395			
O	0.25004	0.00096	0.12100			
O	0.75019	0.49272	0.12139			
O	0.25004	0.50028	0.18770			
O	0.75005	0.99733	0.18533			
O	0.99832	0.24972	0.18453			
O	0.50176	0.74871	0.18381			
O	0.99830	0.74870	0.18382			
O	0.50175	0.24971	0.18453			

O	0.25003	0.99877	0.18658
O	0.75002	0.50033	0.18650
O	0.25003	0.49956	0.25386
O	0.75002	-0.00065	0.25329
O	0.50037	0.74995	0.25279
O	0.99966	0.24889	0.25285
O	0.50039	0.24889	0.25285
O	0.99968	0.74995	0.25279
O	0.25004	-0.00075	0.25318
O	0.75003	0.49957	0.25352
O	0.24999	0.49950	0.32049
O	0.75000	-0.00073	0.32116
O	0.99967	0.24882	0.32117
O	0.50033	0.74991	0.32123
O	0.99968	0.74990	0.32123
O	0.50034	0.24882	0.32117
O	0.25000	-0.00083	0.32050
O	0.74999	0.49950	0.32021
O	0.24998	0.50043	0.38658
O	0.74998	0.99756	0.38914
O	0.50174	0.74883	0.39020
O	0.99833	0.24979	0.38951
O	0.50160	0.24980	0.38950
O	0.99826	0.74883	0.39020
O	0.24998	0.99889	0.38711
O	0.74998	0.50048	0.38731
O	0.24996	0.49958	0.45201
O	0.75001	0.00177	0.45733
O	0.00573	0.24920	0.45931
O	0.49661	0.75290	0.46005
O	0.00345	0.75288	0.46005
O	0.49416	0.24919	0.45931
O	0.25001	0.00112	0.45292
O	0.75000	0.49273	0.45261
O	0.25001	0.53621	0.51620
O	0.24991	0.00069	0.51715
O	0.75000	0.60097	0.51270
O	0.74995	0.00625	0.52575
O	0.74990	0.60009	0.06127
O	0.74999	0.00561	0.04845
O	0.89959	0.25372	0.52486
O	0.89960	0.25309	0.04922
O	0.60033	0.25371	0.52486
O	0.60032	0.25305	0.04922
C	0.74997	0.17695	0.52646
C	0.74998	0.17637	0.04765

Table S12 - *The final minimised structure of the {100}-Stoich-Flat-CO_3 ‖ configuration Ib using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00408	0.00203	0.08201	2.41	-0.30	2.89
Pu A	0.51560	0.50180	0.08487	2.23	3.07	0.48
Pu A	0.00276	0.99895	0.21951	2.18	3.11	0.10
Pu A	0.50158	0.49832	0.21903	2.18	3.11	0.06
Pu A	0.00290	0.00112	0.35454	2.19	3.11	0.09
Pu A	0.50177	0.50175	0.35500	2.16	3.13	0.08
Pu A	0.00531	0.99745	0.49209	3.66	-0.51	0.86
Pu A	0.51667	0.49827	0.48914	2.10	-3.16	0.47
Pu B	0.98658	0.50180	0.08487	2.23	-3.07	0.48
Pu B	0.49842	0.00201	0.08198	2.42	0.30	2.88
Pu B	0.00141	0.49832	0.21903	2.18	-3.11	0.06
Pu B	0.50022	0.99896	0.21951	2.19	-3.11	0.10
Pu B	0.00174	0.50177	0.35500	2.16	-3.13	0.08
Pu B	0.50068	0.00113	0.35454	2.19	-3.11	0.09
Pu B	0.98795	0.49758	0.48917	-2.09	-3.16	-0.53
Pu B	0.49923	0.99822	0.49200	2.39	-0.36	2.90
Pu C	0.25134	0.74714	0.15143	-2.17	0.00	-3.13
Pu C	0.75134	0.24398	0.15189	-2.24	0.00	-3.07
Pu C	0.25167	0.74941	0.28702	-2.22	0.00	-3.09
Pu C	0.75167	0.25023	0.28703	-2.22	0.00	-3.09
Pu C	0.25175	0.75402	0.42363	-2.17	0.00	-3.12
Pu C	0.75171	0.24723	0.42184	-2.29	-0.02	-3.04
Pu D	0.25134	0.24599	0.15046	2.15	0.00	-3.14
Pu D	0.75133	0.75263	0.15220	2.13	0.00	-3.15
Pu D	0.25168	0.25071	0.28703	2.08	0.00	-3.18
Pu D	0.75167	0.74992	0.28700	2.09	0.00	-3.18
Pu D	0.25158	0.25304	0.42261	2.04	0.01	-3.22
Pu D	0.75177	0.75609	0.42213	2.26	-0.03	-3.06
O	0.75129	0.03287	0.05777			
O	0.25130	0.09778	0.06124			
O	0.75107	0.49665	0.05676			
O	0.25129	0.49949	0.11696			
O	0.75135	0.99746	0.12206			
O	0.50716	0.74698	0.11471			
O	0.99774	0.25069	0.11394			
O	0.50480	0.25056	0.11393			
O	0.99532	0.74689	0.11474			
O	0.25117	0.99076	0.12141			
O	0.75113	0.49901	0.12098			
O	0.25140	0.49585	0.18533			
O	0.75140	0.99876	0.18769			
O	0.00310	0.24740	0.18380			
O	0.49966	0.74845	0.18454			
O	0.00310	0.74848	0.18455			
O	0.49969	0.24746	0.18379			

O	0.25145	0.99882	0.18652
O	0.75146	0.49728	0.18657
O	0.25160	0.49959	0.25329
O	0.75158	0.99962	0.25386
O	0.50127	0.74865	0.25293
O	0.00199	0.24969	0.25272
O	0.50127	0.24967	0.25271
O	0.00200	0.74863	0.25294
O	0.25156	0.99954	0.25353
O	0.75156	0.49946	0.25317
O	0.25174	0.50056	0.32116
O	0.75177	0.00053	0.32047
O	0.00209	0.25148	0.32110
O	0.50139	0.75043	0.32131
O	0.00211	0.75044	0.32132
O	0.50141	0.25149	0.32110
O	0.25177	0.00058	0.32022
O	0.75176	0.50064	0.32049
O	0.25175	0.50420	0.38914
O	0.75188	0.00130	0.38652
O	0.50006	0.75258	0.39023
O	0.00344	0.25162	0.38948
O	0.50008	0.25160	0.38949
O	0.00359	0.75261	0.39025
O	0.25186	0.00108	0.38737
O	0.75178	0.50273	0.38709
O	0.25194	0.50025	0.45730
O	0.75089	0.00229	0.45188
O	0.99562	0.25285	0.45932
O	0.50520	0.74951	0.46005
O	0.99869	0.74904	0.46019
O	0.50743	0.25300	0.45928
O	0.25075	0.00952	0.45272
O	0.75193	0.50093	0.45289
O	0.75269	0.96767	0.51617
O	0.25300	0.90188	0.51278
O	0.75240	0.50280	0.51714
O	0.25295	0.49669	0.52572
O	0.25093	0.50351	0.04840
O	0.40321	0.24954	0.52493
O	0.40062	0.75091	0.04920
O	0.10410	0.24874	0.52485
O	0.10130	0.75095	0.04922
C	0.25333	0.32598	0.52649
C	0.25096	0.67424	0.04763

Table S13 - *The final minimised structure of the {100}-Stoich-Flat-CO_3 ‖ configuration Ic using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.99580	0.00359	0.08196	3.64	-1.06	-0.26
Pu A	0.48510	0.50227	0.08492	-2.26	3.04	-0.48
Pu A	0.99889	0.99931	0.21949	2.18	3.11	0.09
Pu A	0.50007	0.49872	0.21903	2.16	3.12	0.08
Pu A	0.99877	0.00077	0.35451	2.18	3.11	0.10
Pu A	0.49993	0.50139	0.35498	2.18	3.11	0.06
Pu A	0.99787	-0.00254	0.49199	2.22	-0.39	3.02
Pu A	0.48586	0.49685	0.48916	2.22	3.07	0.50
Pu B	0.01414	0.50327	0.08485	-2.24	-3.05	-0.53
Pu B	0.50208	0.00236	0.08203	2.28	-0.47	2.97
Pu B	0.00009	0.49870	0.21904	2.16	-3.13	0.08
Pu B	0.50115	0.99930	0.21949	2.18	-3.11	0.09
Pu B	0.00006	0.50137	0.35498	2.18	-3.11	0.06
Pu B	0.50132	0.00076	0.35451	2.18	-3.11	0.10
Pu B	0.01480	0.49761	0.48909	2.23	-3.06	0.49
Pu B	0.50367	-0.00343	0.49207	3.68	-0.76	0.54
Pu C	0.25015	0.75363	0.15219	-2.31	0.01	-3.02
Pu C	0.75015	0.24669	0.15041	-2.17	-0.03	-3.12
Pu C	0.25003	0.74987	0.28699	-2.22	0.00	-3.09
Pu C	0.75003	0.25069	0.28701	-2.22	0.00	-3.09
Pu C	0.24995	0.75533	0.42209	-2.25	0.00	-3.07
Pu C	0.74980	0.25217	0.42261	-2.15	0.01	-3.14
Pu D	0.25017	0.24472	0.15192	2.26	-0.02	-3.06
Pu D	0.75033	0.74746	0.15140	2.05	-0.01	-3.22
Pu D	0.25004	0.25017	0.28702	2.09	0.00	-3.18
Pu D	0.75004	0.74939	0.28700	2.08	0.00	-3.18
Pu D	0.24993	0.24665	0.42180	2.13	0.00	-3.15
Pu D	0.74992	0.75331	0.42357	2.13	-0.01	-3.15
O	0.24955	0.49611	0.05680			
O	0.74798	0.09959	0.06127			
O	0.24868	0.03084	0.05784			
O	0.74880	0.50412	0.04837			
O	0.24991	0.50006	0.12109			
O	0.75162	0.99100	0.12124			
O	0.49432	0.74764	0.11478			
O	0.00314	0.25190	0.11380			
O	0.49658	0.25127	0.11400			
O	0.00652	0.74789	0.11472			
O	0.25126	0.99883	0.12222			
O	0.75002	0.50045	0.11681			
O	0.25008	0.49799	0.18692			
O	0.74993	0.99962	0.18663			
O	0.99828	0.24801	0.18376			
O	0.50185	0.74900	0.18457			
O	0.99839	0.74899	0.18456			

O	0.50183	0.24807	0.18381
O	0.24995	0.99944	0.18753
O	0.75012	0.49637	0.18492
O	0.25005	0.49957	0.25353
O	0.75004	0.99964	0.25380
O	0.50037	0.74886	0.25294
O	0.99966	0.24990	0.25270
O	0.50043	0.24990	0.25272
O	0.99971	0.74884	0.25293
O	0.25001	0.99971	0.25355
O	0.75006	0.49969	0.25288
O	0.25002	0.50049	0.32083
O	0.75003	0.00043	0.32050
O	0.99966	0.25121	0.32109
O	0.50038	0.75018	0.32132
O	0.99967	0.75018	0.32130
O	0.50041	0.25122	0.32110
O	0.25006	0.00037	0.32014
O	0.75001	0.50037	0.32074
O	0.25001	0.50215	0.38741
O	0.75013	0.00056	0.38756
O	0.50179	0.75211	0.39024
O	0.99819	0.25104	0.38947
O	0.50173	0.25104	0.38948
O	0.99824	0.75205	0.39021
O	0.25010	0.00063	0.38626
O	0.74995	0.50363	0.38871
O	0.25008	0.50024	0.45300
O	0.74853	0.00864	0.45273
O	0.00574	0.25224	0.45925
O	0.49675	0.74815	0.46018
O	0.00350	0.74882	0.46001
O	0.49368	0.25214	0.45934
O	0.24906	0.00164	0.45182
O	0.75009	0.49962	0.45704
O	0.25035	0.50271	0.51722
O	0.75177	0.90118	0.51278
O	0.25120	0.96722	0.51618
O	0.75096	0.49581	0.52558
O	0.89715	0.75235	0.04922
O	0.90143	0.24884	0.52484
O	0.59813	0.75104	0.04916
O	0.60232	0.24777	0.52487
C	0.74813	0.67479	0.04759
C	0.75147	0.32508	0.52642

Table S14 - *The final minimised structure of the {100}-Redu-Flat-CO_3 || configuration IIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.02078	0.00938	0.08546	4.27	1.70	-1.31
Pu A	0.48516	0.49846	0.08749	1.44	3.47	1.11
Pu A	0.99800	0.00475	0.22109	2.18	3.11	0.07
Pu A	0.50097	0.50580	0.22064	2.16	3.13	0.13
Pu A	0.99853	0.00633	0.35651	2.15	3.13	0.12
Pu A	0.50174	0.50703	0.35628	2.18	3.11	0.07
Pu A	0.01811	0.01635	0.49013	-2.28	3.05	0.29
Pu A	0.47839	0.50928	0.49201	1.86	2.96	3.29
Pu B	0.02489	0.49633	0.08567	-0.54	-2.63	3.97
Pu B	0.48885	0.01248	0.08663	1.86	-3.35	0.53
Pu B	0.99838	0.50582	0.22089	2.15	-3.13	0.12
Pu B	0.50135	0.00471	0.22082	2.19	-3.10	0.06
Pu B	0.99893	0.50703	0.35627	2.19	-3.10	0.07
Pu B	0.50214	0.00633	0.35652	2.16	-3.12	0.13
Pu B	0.02365	0.50932	0.49202	1.86	-2.96	3.29
Pu B	0.48394	0.01636	0.49013	-2.28	-3.05	0.29
Pu C	0.24794	0.75561	0.15389	-2.27	-0.02	-3.05
Pu C	0.75089	0.25488	0.15224	-2.19	0.02	-3.11
Pu C	0.25007	0.75615	0.28865	-2.23	-0.01	-3.08
Pu C	0.74982	0.25599	0.28861	-2.23	0.01	-3.08
Pu C	0.25070	0.75453	0.42319	-2.21	0.00	-3.09
Pu C	0.75074	0.26143	0.42530	-2.14	0.00	-3.14
Pu D	0.24845	0.25274	0.15528	2.19	-0.02	-3.11
Pu D	0.75125	0.75581	0.15091	2.06	-0.03	-3.20
Pu D	0.25007	0.25588	0.28864	2.10	0.01	-3.17
Pu D	0.74983	0.75592	0.28862	2.08	-0.01	-3.18
Pu D	0.25070	0.25646	0.42208	2.12	0.00	-3.15
Pu D	0.75076	0.75697	0.42610	2.16	0.00	-3.13
O	0.27467	0.53401	0.05709			
O	0.74488	0.02241	0.04852			
O	0.27331	0.00867	0.05694			
O	0.25654	0.50250	0.12342			
O	0.74241	0.00887	0.11925			
O	0.49971	0.75617	0.11471			
O	0.00721	0.25460	0.11934			
O	0.49075	0.25464	0.11873			
O	0.00067	0.75308	0.11683			
O	0.25734	0.00415	0.12381			
O	0.74137	0.50272	0.11643			
O	0.24883	0.50543	0.18890			
O	0.75043	0.00388	0.18672			
O	0.99642	0.25479	0.18725			
O	0.50080	0.75501	0.18524			
O	0.99831	0.75527	0.18598			
O	0.50257	0.25481	0.18683			

O	0.24873	0.00362	0.18904
O	0.75049	0.50631	0.18557
O	0.25015	0.50613	0.25511
O	0.74934	0.00548	0.25512
O	0.50066	0.75536	0.25430
O	0.99911	0.25574	0.25480
O	0.50054	0.25568	0.25451
O	0.99898	0.75533	0.25466
O	0.25014	0.00544	0.25508
O	0.74938	0.50599	0.25437
O	0.25021	0.50656	0.32224
O	0.75015	0.00596	0.32274
O	0.99925	0.25609	0.32267
O	0.50100	0.75668	0.32268
O	0.99915	0.75669	0.32276
O	0.50091	0.25610	0.32261
O	0.25021	0.00605	0.32223
O	0.75014	0.50639	0.32207
O	0.25049	0.50724	0.38837
O	0.75047	0.00700	0.39132
O	0.50127	0.75621	0.39142
O	0.99792	0.25780	0.39039
O	0.50326	0.25778	0.39035
O	0.99992	0.75620	0.39147
O	0.25049	0.00532	0.38853
O	0.75045	0.50896	0.39044
O	0.25096	0.50170	0.45358
O	0.75094	0.01381	0.45969
O	0.00860	0.25046	0.45851
O	0.50085	0.76648	0.46086
O	0.00102	0.76650	0.46088
O	0.49317	0.25047	0.45849
O	0.25097	0.00402	0.45412
O	0.75099	0.50912	0.45859
O	0.25106	0.61797	0.51771
O	0.75114	0.01725	0.52707
O	0.25109	0.06245	0.51764
O	0.89932	0.27107	0.04868
O	0.89909	0.26802	0.52676
O	0.60507	0.27653	0.05209
O	0.60320	0.26802	0.52673
C	0.75118	0.19282	0.04870
C	0.75114	0.18908	0.52804

Table S15 - *The final minimised structure of the {100}-Redu-Flat-CO3 // configuration IIb using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.02047	0.00060	0.08286	1.63	2.33	3.88
Pu A	0.47860	0.49427	0.08604	1.25	3.63	0.05
Pu A	0.99837	0.00515	0.21951	2.18	3.11	0.07
Pu A	0.50158	0.50621	0.21891	2.13	3.14	0.12
Pu A	0.99849	0.00314	0.35458	2.15	3.13	0.12
Pu A	0.50149	0.50394	0.35481	2.17	3.12	0.06
Pu A	0.02519	0.00466	0.49142	2.36	-1.64	-3.85
Pu A	0.47116	0.49741	0.49044	2.54	2.20	3.42
Pu B	0.02172	0.49427	0.08603	1.25	-3.63	0.05
Pu B	0.47985	0.00060	0.08286	1.63	-2.33	3.88
Pu B	0.99874	0.50620	0.21891	2.13	-3.14	0.12
Pu B	0.50196	0.00515	0.21951	2.18	-3.11	0.07
Pu B	0.99899	0.50394	0.35482	2.18	-3.11	0.06
Pu B	0.50175	0.00295	0.35462	2.17	-3.12	0.13
Pu B	0.01037	0.49220	0.48741	-1.37	-3.52	1.05
Pu B	0.49171	0.00888	0.48802	-1.25	-3.59	0.70
Pu C	0.25015	0.76164	0.15247	-2.24	0.00	-3.07
Pu C	0.75015	0.25155	0.15011	-2.09	0.00	-3.18
Pu C	0.25018	0.75420	0.28697	-2.24	0.00	-3.07
Pu C	0.75016	0.25496	0.28694	-2.25	0.00	-3.06
Pu C	0.25002	0.75357	0.42167	-2.29	0.05	-3.03
Pu C	0.75011	0.25196	0.42329	-2.21	-0.04	-3.10
Pu D	0.25015	0.25490	0.15388	2.08	0.00	-3.18
Pu D	0.75016	0.75852	0.14933	2.07	0.00	-3.19
Pu D	0.25021	0.25484	0.28695	2.08	0.00	-3.18
Pu D	0.75019	0.75416	0.28698	2.07	0.00	-3.19
Pu D	0.25023	0.25099	0.42039	2.19	0.00	-3.11
Pu D	0.75064	0.75356	0.42449	2.08	-0.04	-3.18
O	0.25017	0.43284	0.05868			
O	0.25016	0.87250	0.06015			
O	0.25015	0.51036	0.12171			
O	0.75018	0.00686	0.11798			
O	0.50131	0.74808	0.11550			
O	0.00780	0.26360	0.11748			
O	0.49251	0.26360	0.11748			
O	0.99900	0.74807	0.11550			
O	0.25017	0.01274	0.12250			
O	0.75015	0.50133	0.11132			
O	0.25015	0.50767	0.18706			
O	0.75015	0.00487	0.18520			
O	0.99691	0.25490	0.18539			
O	0.50079	0.75714	0.18455			
O	0.99953	0.75714	0.18455			
O	0.50338	0.25491	0.18539			
O	0.25016	0.00653	0.18761			

O	0.75016	0.50629	0.18280
O	0.25019	0.50532	0.25326
O	0.75019	0.00479	0.25353
O	0.50117	0.75459	0.25303
O	0.99939	0.25598	0.25302
O	0.50098	0.25597	0.25303
O	0.99920	0.75457	0.25303
O	0.25017	0.00459	0.25348
O	0.75016	0.50538	0.25240
O	0.25011	0.50439	0.32069
O	0.75000	0.00393	0.32080
O	0.99943	0.25404	0.32094
O	0.50107	0.75342	0.32119
O	0.99931	0.75360	0.32117
O	0.50097	0.25414	0.32096
O	0.25036	0.00387	0.32032
O	0.75034	0.50431	0.32063
O	0.25077	0.50385	0.38690
O	0.75079	0.00178	0.38906
O	0.50146	0.75358	0.39000
O	0.99705	0.25345	0.38853
O	0.50322	0.25252	0.38853
O	0.99896	0.75299	0.38995
O	0.24950	0.00179	0.38640
O	0.74979	0.50449	0.38950
O	0.24324	0.50115	0.45223
O	0.74447	0.00613	0.45635
O	0.00800	0.24918	0.45657
O	0.49984	0.75227	0.45974
O	0.00096	0.75230	0.45966
O	0.49181	0.25503	0.45655
O	0.25678	0.00309	0.45184
O	0.75568	0.49996	0.45872
O	0.21824	0.51619	0.51871
O	0.27993	0.98273	0.51856
O	0.74832	0.01683	0.52698
O	0.75016	0.03202	0.04523
O	0.89447	0.27003	0.52509
O	0.89750	0.28302	0.04912
O	0.59957	0.26625	0.52458
O	0.60286	0.28304	0.04912
C	0.74783	0.18732	0.52662
C	0.75017	0.20081	0.04706

Table S16 - *The final minimised structure of the {100}-Redu-Flat-CO3 // configuration IIc using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.99857	0.00630	0.08539	1.63	2.33	3.88
Pu A	0.45880	0.49950	0.08355	1.25	3.63	0.05
Pu A	0.98034	0.99597	0.21897	2.18	3.11	0.07
Pu A	0.48364	0.49669	0.21923	2.13	3.14	0.12
Pu A	0.98146	0.99539	0.35472	2.15	3.13	0.12
Pu A	0.48467	0.49610	0.35446	2.17	3.12	0.06
Pu A	0.00140	0.00403	0.48828	2.36	-1.64	-3.85
Pu A	0.46172	0.49690	0.49021	2.54	2.20	3.42
Pu B	0.00442	0.49946	0.08356	1.25	-3.63	0.05
Pu B	0.46415	0.00631	0.08538	1.63	-2.33	3.88
Pu B	0.98064	0.49669	0.21923	2.13	-3.14	0.12
Pu B	0.48395	0.99597	0.21897	2.18	-3.11	0.07
Pu B	0.98185	0.49612	0.35446	2.18	-3.11	0.06
Pu B	0.48505	0.99539	0.35472	2.17	-3.12	0.13
Pu B	0.00683	0.49677	0.49022	-1.37	-3.52	1.05
Pu B	0.46731	0.00420	0.48828	-1.25	-3.59	0.70
Pu C	0.23179	0.74418	0.15234	-2.24	0.00	-3.07
Pu C	0.73173	0.25136	0.15017	-2.09	0.00	-3.18
Pu C	0.23266	0.74638	0.28684	-2.24	0.00	-3.07
Pu C	0.73265	0.24577	0.28685	-2.25	0.00	-3.06
Pu C	0.23382	0.74268	0.42137	-2.29	0.05	-3.03
Pu C	0.73391	0.24972	0.42348	-2.21	-0.04	-3.10
Pu D	0.23176	0.24603	0.15347	2.08	0.00	-3.18
Pu D	0.73176	0.74651	0.14940	2.07	0.00	-3.19
Pu D	0.23266	0.24568	0.28684	2.08	0.00	-3.18
Pu D	0.73266	0.74614	0.28685	2.07	0.00	-3.19
Pu D	0.23387	0.24476	0.42026	2.19	0.00	-3.11
Pu D	0.73390	0.74509	0.42429	2.08	-0.04	-3.18
O	0.23156	0.61115	0.05812			
O	0.73134	0.00746	0.04850			
O	0.23129	0.05445	0.05791			
O	0.23157	0.49118	0.12202			
O	0.73152	0.00353	0.11598			
O	0.48169	0.75638	0.11471			
O	0.98939	0.24033	0.11703			
O	0.47378	0.24031	0.11702			
O	0.98160	0.75634	0.11473			
O	0.23149	0.99357	0.12128			
O	0.73156	0.49865	0.11667			
O	0.23198	0.49655	0.18735			
O	0.73196	0.99625	0.18451			
O	0.97909	0.24727	0.18514			
O	0.48253	0.74572	0.18407			
O	0.98128	0.74572	0.18408			
O	0.48465	0.24728	0.18514			

O	0.23199	0.99474	0.18672
O	0.73196	0.49838	0.18471
O	0.23231	0.49643	0.25351
O	0.73230	0.99585	0.25312
O	0.48335	0.74656	0.25276
O	0.98172	0.24590	0.25286
O	0.48322	0.24591	0.25285
O	0.98159	0.74656	0.25276
O	0.23232	0.99590	0.25300
O	0.73230	0.49624	0.25297
O	0.23299	0.49613	0.32046
O	0.73300	0.99556	0.32092
O	0.98213	0.24544	0.32083
O	0.48372	0.74618	0.32096
O	0.98203	0.74616	0.32096
O	0.48360	0.24541	0.32083
O	0.23296	0.99565	0.32036
O	0.73297	0.49594	0.32038
O	0.23344	0.49571	0.38658
O	0.73347	0.99543	0.38950
O	0.48433	0.74494	0.38966
O	0.98102	0.24652	0.38856
O	0.48638	0.24652	0.38855
O	0.98298	0.74497	0.38966
O	0.23345	0.99378	0.38668
O	0.73349	0.49748	0.38867
O	0.23419	0.48987	0.45178
O	0.73419	0.00178	0.45785
O	0.99190	0.23836	0.45667
O	0.48398	0.75437	0.45910
O	0.98434	0.75431	0.45910
O	0.47640	0.23850	0.45666
O	0.23422	0.99217	0.45228
O	0.73421	0.49707	0.45676
O	0.23423	0.60332	0.51613
O	0.73427	0.00442	0.52526
O	0.23440	0.04854	0.51588
O	0.87908	0.25837	0.04885
O	0.88270	0.25493	0.52491
O	0.58316	0.25810	0.04879
O	0.58681	0.25551	0.52487
C	0.73121	0.17934	0.04754
C	0.73457	0.17622	0.52620

Table S17 - *The final minimised structure of the {100}-Redu-Flat-CO3 // configuration IId using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.02007	0.01500	0.08348	2.20	-0.47	3.03
Pu A	0.50622	0.50033	0.08556	3.83	-2.80	0.71
Pu A	0.01490	0.99889	0.21956	2.19	3.11	0.07
Pu A	0.51602	0.49915	0.21900	2.16	3.12	0.09
Pu A	0.01482	0.00117	0.35448	2.19	3.11	0.07
Pu A	0.51600	0.50088	0.35503	2.17	3.12	0.10
Pu A	0.02027	0.98494	0.49080	2.17	0.62	3.03
Pu A	0.50596	0.49902	0.48859	3.87	2.76	0.66
Pu B	0.02516	0.50063	0.08557	-3.81	-2.84	-0.66
Pu B	0.51133	0.01491	0.08350	2.48	0.24	2.84
Pu B	0.01533	0.49916	0.21899	2.16	-3.12	0.09
Pu B	0.51646	0.99889	0.21956	2.19	-3.11	0.07
Pu B	0.01543	0.50087	0.35501	2.17	-3.12	0.10
Pu B	0.51656	0.00120	0.35450	2.19	-3.11	0.07
Pu B	0.02471	0.50040	0.48843	3.86	-2.75	0.75
Pu B	0.51187	0.98465	0.49002	2.77	2.57	-0.46
Pu C	0.26566	0.75116	0.15196	-2.15	-0.03	-3.14
Pu C	0.76559	0.24700	0.15141	-2.21	0.00	-3.10
Pu C	0.26569	0.74968	0.28701	-2.22	0.00	-3.09
Pu C	0.76567	0.25054	0.28702	-2.22	0.00	-3.08
Pu C	0.26559	0.75504	0.42189	-2.20	0.03	-3.10
Pu C	0.76603	0.24951	0.42347	-2.23	-0.01	-3.08
Pu D	0.26560	0.24511	0.15221	2.21	-0.04	-3.09
Pu D	0.76564	0.75052	0.15058	2.05	0.00	-3.21
Pu D	0.26569	0.25038	0.28703	2.09	0.00	-3.17
Pu D	0.76567	0.74949	0.28700	2.09	0.00	-3.17
Pu D	0.26584	0.24929	0.42210	2.06	0.01	-3.21
Pu D	0.76579	0.75272	0.42251	2.22	0.02	-3.09
O	0.11592	0.77624	0.04791			
O	0.76559	0.08851	0.05987			
O	0.26538	0.13540	0.06439			
O	0.26559	0.50012	0.11801			
O	0.76543	0.99703	0.12102			
O	0.51661	0.75410	0.11487			
O	0.01526	0.24912	0.11758			
O	0.51583	0.24931	0.11759			
O	0.01471	0.75435	0.11493			
O	0.26593	0.99335	0.12140			
O	0.76573	0.49979	0.11495			
O	0.26569	0.49764	0.18563			
O	0.76569	0.99882	0.18665			
O	0.01595	0.24832	0.18527			
O	0.51556	0.74879	0.18452			
O	0.01572	0.74873	0.18453			
O	0.51531	0.24828	0.18527			

O	0.26564	0.99873	0.18675
O	0.76567	0.49840	0.18452
O	0.26573	0.49973	0.25319
O	0.76572	0.99961	0.25376
O	0.51594	0.74865	0.25289
O	0.01542	0.25004	0.25314
O	0.51598	0.25006	0.25313
O	0.01547	0.74868	0.25288
O	0.26574	0.99954	0.25346
O	0.76574	0.49966	0.25273
O	0.26577	0.50029	0.32120
O	0.76564	0.00044	0.32056
O	0.01544	0.25131	0.32113
O	0.51592	0.75002	0.32090
O	0.01545	0.75003	0.32087
O	0.51593	0.25136	0.32115
O	0.26580	0.00047	0.32035
O	0.76569	0.50038	0.32092
O	0.26574	0.50255	0.38879
O	0.76592	0.00141	0.38756
O	0.51528	0.75192	0.38876
O	0.01593	0.25141	0.38949
O	0.51566	0.25126	0.38950
O	0.01600	0.75162	0.38871
O	0.26542	0.00117	0.38723
O	0.76575	0.50155	0.38907
O	0.26555	0.49955	0.45628
O	0.76411	0.00242	0.45303
O	0.01467	0.24563	0.45907
O	0.51531	0.74964	0.45641
O	0.01519	0.75168	0.45626
O	0.51774	0.24661	0.45905
O	0.26849	0.00748	0.45287
O	0.76562	0.50084	0.45879
O	0.11228	0.22488	0.52630
O	0.76516	0.91306	0.51430
O	0.26261	0.86224	0.50947
O	0.26592	0.52911	0.04331
O	0.41106	0.22140	0.52582
O	0.41511	0.77676	0.04786
O	0.26474	0.47066	0.53067
C	0.26568	0.69921	0.04520
C	0.26288	0.30059	0.52878

Table S18 - *The final minimised structure of the {100}-Redu-CO_3 ⊥ configuration IIIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.99888	-0.00003	0.08402	3.56	1.38	0.00
Pu A	0.48281	0.49997	0.08539	3.49	-3.27	-0.10
Pu A	0.99259	0.00041	0.21945	2.18	3.11	0.08
Pu A	0.49367	0.50042	0.21911	2.16	3.13	0.08
Pu A	0.99256	0.00030	0.35460	2.19	3.11	0.08
Pu A	0.49365	0.50027	0.35491	2.18	3.11	0.08
Pu A	0.99869	0.00004	0.49014	3.57	-1.37	-0.04
Pu A	0.48285	0.50021	0.48902	0.93	4.69	-0.15
Pu B	0.00461	0.50049	0.08494	-2.60	-1.47	-3.74
Pu B	0.49786	0.99928	0.08369	3.62	1.25	-0.05
Pu B	0.99329	0.50038	0.21909	2.17	-3.12	0.08
Pu B	0.49436	0.00039	0.21945	2.18	-3.11	0.08
Pu B	0.99343	0.50027	0.35496	2.17	-3.12	0.08
Pu B	0.49454	0.00029	0.35457	2.18	-3.11	0.08
Pu B	0.00407	0.50023	0.48890	3.83	-2.88	0.11
Pu B	0.49720	0.00002	0.49016	3.60	-1.28	0.03
Pu C	0.24296	0.75301	0.15199	-2.31	-0.02	-3.02
Pu C	0.74297	0.25056	0.15098	-2.18	0.01	-3.12
Pu C	0.24355	0.75027	0.28702	-2.23	0.00	-3.08
Pu C	0.74358	0.25074	0.28701	-2.23	0.00	-3.08
Pu C	0.24318	0.75256	0.42206	-2.18	0.00	-3.12
Pu C	0.74297	0.25027	0.42302	-2.33	0.05	-3.01
Pu D	0.24298	0.24802	0.15200	2.26	-0.05	-3.06
Pu D	0.74309	0.75013	0.15104	2.12	0.01	-3.16
Pu D	0.24355	0.25043	0.28702	2.08	0.00	-3.18
Pu D	0.74358	0.74996	0.28701	2.08	0.00	-3.18
Pu D	0.24325	0.24806	0.42207	2.16	-0.02	-3.14
Pu D	0.74293	0.75009	0.42298	2.22	-0.03	-3.09
O	0.74831	0.99693	0.05780			
O	0.24759	0.99356	0.05808			
O	0.23637	0.50030	0.12018			
O	0.74419	0.00025	0.12187			
O	0.49162	0.75750	0.11461			
O	0.99401	0.24641	0.11387			
O	0.49161	0.24256	0.11441			
O	0.99422	0.75475	0.11392			
O	0.23973	0.00076	0.12230			
O	0.74445	0.50014	0.11747			
O	0.24367	0.50054	0.18662			
O	0.74340	0.00037	0.18702			
O	0.99272	0.25044	0.18408			
O	0.49377	0.75013	0.18420			
O	0.99277	0.75036	0.18410			
O	0.49379	0.25071	0.18416			
O	0.24369	0.00037	0.18725			

O	0.74342	0.50036	0.18541
O	0.24345	0.50040	0.25350
O	0.74367	0.00042	0.25385
O	0.49377	0.75006	0.25284
O	0.99327	0.25085	0.25279
O	0.49379	0.25078	0.25284
O	0.99329	0.74998	0.25279
O	0.24344	0.00041	0.25351
O	0.74364	0.50040	0.25298
O	0.24346	0.50030	0.32089
O	0.74370	0.00032	0.32049
O	0.99330	0.25066	0.32127
O	0.49380	0.74993	0.32119
O	0.99332	0.74990	0.32127
O	0.49382	0.25064	0.32120
O	0.24349	0.00030	0.32020
O	0.74373	0.50030	0.32063
O	0.24382	0.50036	0.38776
O	0.74349	0.00030	0.38723
O	0.49381	0.75002	0.38983
O	0.99288	0.25042	0.39001
O	0.49385	0.25055	0.38985
O	0.99290	0.75016	0.38998
O	0.24375	0.00028	0.38657
O	0.74342	0.50023	0.38820
O	0.23685	0.50035	0.45404
O	0.74369	0.00016	0.45218
O	0.99413	0.24546	0.46025
O	0.49167	0.75728	0.45944
O	0.99419	0.75502	0.46017
O	0.49171	0.24313	0.45950
O	0.24035	0.00047	0.45174
O	0.74479	0.50026	0.45630
O	0.74759	0.99991	0.51619
O	0.24744	0.99834	0.51601
O	0.22736	0.49799	0.52542
O	0.22770	0.49283	0.04866
O	0.49869	0.50055	0.54544
O	0.49973	0.49968	0.02885
O	0.26883	0.50020	0.58205
O	0.27049	0.50013	0.99210
C	0.33256	0.49967	0.55452
C	0.33428	0.49788	0.01962

Table S19 - *The final minimised structure of the {100}-Redu-CO_2 ⊥ configuration IVa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00242	0.00000	0.08450	3.78	0.64	-0.02
Pu A	0.47893	0.49968	0.08415	-2.27	1.22	-4.04
Pu A	0.99845	0.00004	0.21929	2.18	3.11	0.08
Pu A	0.50038	0.50009	0.21921	2.16	3.12	0.08
Pu A	0.99832	0.00019	0.35470	2.19	3.11	0.08
Pu A	0.50027	0.50017	0.35477	2.18	3.11	0.08
Pu A	0.00145	0.99996	0.48954	3.85	0.13	0.01
Pu A	0.47891	0.50052	0.48983	3.40	2.12	-2.62
Pu B	0.02068	0.49956	0.08421	-2.74	-1.56	-3.62
Pu B	0.49902	0.00010	0.08450	3.83	0.19	-0.11
Pu B	0.99974	0.50009	0.21921	2.16	-3.13	0.08
Pu B	0.50169	0.00004	0.21929	2.18	-3.12	0.07
Pu B	0.99982	0.50020	0.35478	2.18	-3.11	0.08
Pu B	0.50175	0.00022	0.35469	2.18	-3.11	0.08
Pu B	0.02026	0.50056	0.48988	4.26	-2.00	0.91
Pu B	0.49796	0.00024	0.48947	3.84	0.08	-0.07
Pu C	0.25004	0.75226	0.15222	-2.31	-0.02	-3.02
Pu C	0.74998	0.25272	0.15065	-2.26	0.00	-3.06
Pu C	0.25004	0.75043	0.28698	-2.23	0.00	-3.08
Pu C	0.75005	0.25043	0.28699	-2.23	0.00	-3.08
Pu C	0.25013	0.75228	0.42169	-2.22	0.00	-3.09
Pu C	0.75003	0.25293	0.42338	-2.32	0.00	-3.01
Pu D	0.25007	0.24799	0.15234	2.22	0.00	-3.09
Pu D	0.75002	0.74695	0.15059	2.16	0.00	-3.13
Pu D	0.25004	0.24984	0.28699	2.08	0.00	-3.18
Pu D	0.75005	0.74985	0.28699	2.08	0.00	-3.18
Pu D	0.25004	0.24839	0.42172	2.19	-0.01	-3.12
Pu D	0.74997	0.74771	0.42331	2.23	-0.01	-3.09
O	0.24981	0.49170	0.05237			
O	0.75034	0.99904	0.05670			
O	0.25043	0.99379	0.05745			
O	0.24996	0.49976	0.12148			
O	0.74985	0.99975	0.12111			
O	0.49725	0.75487	0.11382			
O	0.00278	0.24513	0.11410			
O	0.49726	0.24518	0.11414			
O	0.00281	0.75496	0.11384			
O	0.24977	0.00045	0.12185			
O	0.75014	0.49917	0.11793			
O	0.25007	0.50025	0.18709			
O	0.75010	0.99971	0.18666			
O	0.99926	0.25044	0.18411			
O	0.50083	0.74955	0.18393			
O	0.99930	0.74955	0.18394			
O	0.50087	0.25044	0.18412			
O	0.25011	0.99984	0.18709			

O	0.75005	0.50008	0.18561
O	0.25005	0.50014	0.25363
O	0.75007	0.00016	0.25375
O	0.50047	0.74999	0.25274
O	0.99962	0.25026	0.25280
O	0.50049	0.25026	0.25280
O	0.99963	0.74999	0.25274
O	0.25006	0.00013	0.25335
O	0.75006	0.50014	0.25308
O	0.25002	0.50015	0.32069
O	0.75006	0.00013	0.32058
O	0.99961	0.25025	0.32126
O	0.50045	0.75005	0.32120
O	0.99963	0.75005	0.32122
O	0.50047	0.25025	0.32124
O	0.25001	0.00012	0.32026
O	0.75006	0.50014	0.32051
O	0.25005	0.50028	0.38719
O	0.75001	0.00034	0.38762
O	0.50080	0.74979	0.38991
O	0.99929	0.25069	0.39005
O	0.50077	0.25071	0.38999
O	0.99934	0.74982	0.38995
O	0.25007	0.00033	0.38660
O	0.75006	0.50022	0.38801
O	0.25015	0.50051	0.45262
O	0.75032	0.00045	0.45297
O	0.00259	0.24542	0.46023
O	0.49746	0.75581	0.45986
O	0.00290	0.75530	0.45998
O	0.49732	0.24528	0.46000
O	0.24983	0.00052	0.45203
O	0.74977	0.50048	0.45593
O	0.24979	0.49973	0.52171
O	0.74953	0.99878	0.51732
O	0.24999	0.99959	0.51655
O	0.75080	0.50061	0.54183
O	0.74815	0.50670	0.03255
O	0.74984	0.49951	0.60055
O	0.74964	0.49735	0.97385
C	0.75041	0.50007	0.57148
C	0.74885	0.50205	0.00291

Section S5: Surface Structure and Magnetisation - {110} Adsorption

Table S20 - *The final minimised structure of the {110}-Stoich-CO_3 ∠ configuration Va using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.99703	0.49825	0.06487	-1.99	-3.32	-0.04
Pu A	0.50143	0.48527	0.06251	-1.40	-3.51	0.56
Pu A	0.99608	0.99891	0.17554	-2.19	-3.10	-0.01
Pu A	0.49740	0.99827	0.17462	-2.19	-3.11	0.02
Pu A	0.99617	0.49809	0.28814	-2.19	-3.10	-0.01
Pu A	0.49740	0.49855	0.28912	-2.21	-3.09	0.04
Pu A	0.99726	0.99865	0.39885	-1.90	-3.37	0.08
Pu A	0.50168	0.01143	0.40120	-1.80	-3.32	0.62
Pu B	0.24263	0.74846	0.11664	2.42	0.00	2.94
Pu B	0.74512	0.74837	0.11459	2.04	0.00	3.20
Pu B	0.24705	0.24845	0.23186	2.23	0.00	3.07
Pu B	0.74516	0.24839	0.23190	2.20	0.00	3.09
Pu B	0.24276	0.74847	0.34733	2.26	-0.01	3.04
Pu B	0.74521	0.74840	0.34910	2.10	0.00	3.17
Pu B	0.24728	0.24843	0.11482	2.10	0.00	-3.17
Pu B	0.74936	0.24841	0.11534	2.22	0.00	-3.08
Pu C	0.24787	0.74844	0.23190	2.21	0.00	-3.09
Pu C	0.74617	0.74839	0.23185	2.22	0.00	-3.08
Pu C	0.24760	0.24845	0.34882	2.22	0.00	-3.09
Pu C	0.74960	0.24843	0.34851	2.26	0.00	-3.05
Pu C	0.99703	0.99862	0.06488	-1.97	3.34	-0.04
Pu C	0.50146	0.01155	0.06251	-1.32	3.53	0.62
Pu D	0.99609	0.49793	0.17554	-2.19	3.10	0.00
Pu D	0.49740	0.49854	0.17462	-2.19	3.11	0.02
Pu D	0.99616	0.99875	0.28813	-2.20	3.10	-0.01
Pu D	0.49741	0.99831	0.28912	-2.21	3.09	0.04
Pu D	0.99727	0.49818	0.39886	-1.94	3.35	0.09
Pu D	0.50163	0.48560	0.40119	-1.79	3.32	0.62
O	0.11374	0.74843	0.06298			
O	0.60474	0.74820	0.06450			
O	0.36427	0.74897	0.05270			
O	0.87698	0.74840	0.06069			
O	0.11695	0.24842	0.06156			
O	0.62236	0.24849	0.05945			
O	0.38196	0.24831	0.06241			
O	0.88160	0.24841	0.06163			
O	0.12229	0.49653	0.11813			
O	0.62491	0.49241	0.11797			
O	0.12230	0.00033	0.11812			
O	0.62494	0.00434	0.11794			
O	0.37346	0.50838	0.11554			
O	0.87422	0.49996	0.11768			
O	0.37351	0.98853	0.11558			
O	0.87423	0.99681	0.11768			

O	0.12180	0.24841	0.17496
O	0.62104	0.24835	0.17514
O	0.37159	0.24848	0.17293
O	0.87183	0.24841	0.17445
O	0.12211	0.74843	0.17397
O	0.62061	0.74842	0.17391
O	0.37044	0.74839	0.17469
O	0.87152	0.74839	0.17484
O	0.12222	0.99653	0.23184
O	0.62118	0.99612	0.23188
O	0.12225	0.50032	0.23184
O	0.62118	0.50063	0.23187
O	0.37215	0.00101	0.23187
O	0.87113	0.99953	0.23187
O	0.37215	0.49587	0.23187
O	0.87115	0.49722	0.23187
O	0.12208	0.74844	0.28960
O	0.62077	0.74843	0.28994
O	0.37044	0.74840	0.28902
O	0.87164	0.74838	0.28884
O	0.12184	0.24840	0.28875
O	0.62087	0.24837	0.28853
O	0.37156	0.24850	0.29079
O	0.87176	0.24842	0.28930
O	0.12236	0.49739	0.34561
O	0.62490	0.49228	0.34581
O	0.12237	0.99947	0.34561
O	0.62494	0.00456	0.34583
O	0.37354	0.50815	0.34812
O	0.87433	0.49955	0.34608
O	0.37359	0.98883	0.34809
O	0.87434	0.99722	0.34608
O	0.11601	0.24844	0.40182
O	0.62185	0.24855	0.40430
O	0.38132	0.24842	0.40135
O	0.88068	0.24837	0.40242
O	0.11562	0.74842	0.40122
O	0.60658	0.74832	0.39946
O	0.36692	0.74891	0.41067
O	0.87859	0.74839	0.40275
O	0.43247	0.89705	0.99969
O	0.43204	0.89732	0.46404
O	0.43128	0.59904	0.99980
O	0.43118	0.59925	0.46397
C	0.40705	0.74835	0.01462
C	0.40757	0.74848	0.44907

Table S21 - *The final minimised structure of the {110}-Stoich-CO_3 ∠ configuration Vb using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00431	0.49992	0.06487	-2.00	-3.32	-0.03
Pu A	0.50886	0.48694	0.06251	-0.71	-3.69	0.68
Pu A	0.00322	0.00061	0.17553	-2.20	-3.10	-0.01
Pu A	0.50450	0.99996	0.17461	-2.19	-3.11	0.02
Pu A	0.00325	0.49981	0.28814	-2.20	-3.10	-0.01
Pu A	0.50445	0.50022	0.28914	-2.21	-3.09	0.04
Pu A	0.00438	0.00038	0.39886	-1.92	-3.36	0.08
Pu A	0.50895	0.01295	0.40122	-1.79	-3.32	0.63
Pu B	0.24986	0.75005	0.11660	2.43	0.00	2.93
Pu B	0.75238	0.75019	0.11463	2.04	0.02	3.20
Pu B	0.25410	0.25008	0.23187	2.24	0.00	3.07
Pu B	0.75220	0.25014	0.23190	2.21	0.00	3.09
Pu B	0.24990	0.75007	0.34738	2.26	0.00	3.03
Pu B	0.75235	0.75014	0.34906	2.10	0.00	3.17
Pu B	0.25453	0.25003	0.11477	2.10	0.02	-3.17
Pu B	0.75661	0.25013	0.11536	2.22	0.00	-3.08
Pu C	0.25495	0.75009	0.23190	2.22	0.00	-3.09
Pu C	0.75322	0.75014	0.23185	2.22	0.00	-3.08
Pu C	0.25472	0.25008	0.34888	2.23	0.00	-3.08
Pu C	0.75676	0.25011	0.34849	2.26	0.00	-3.05
Pu C	0.00431	0.00028	0.06486	-2.00	3.31	-0.03
Pu C	0.50900	0.01325	0.06247	-1.61	3.42	0.53
Pu D	0.00322	0.49963	0.17553	-2.20	3.10	-0.01
Pu D	0.50450	0.50025	0.17462	-2.19	3.11	0.02
Pu D	0.00326	0.00046	0.28814	-2.20	3.10	-0.01
Pu D	0.50445	0.99997	0.28914	-2.22	3.09	0.04
Pu D	0.00438	0.49992	0.39886	-1.92	3.36	0.08
Pu D	0.50901	0.48702	0.40122	-1.80	3.31	0.64
O	0.12119	0.75012	0.06296			
O	0.61230	0.75039	0.06457			
O	0.37214	0.74912	0.05228			
O	0.88439	0.75012	0.06071			
O	0.12429	0.25005	0.06151			
O	0.62983	0.24998	0.05947			
O	0.38936	0.25023	0.06238			
O	0.88896	0.25016	0.06166			
O	0.12951	0.49809	0.11810			
O	0.63214	0.49416	0.11797			
O	0.12950	0.00205	0.11812			
O	0.63215	0.00618	0.11801			
O	0.38067	0.51016	0.11553			
O	0.88145	0.50174	0.11768			
O	0.38066	0.98985	0.11544			
O	0.88145	0.99856	0.11768			
O	0.12889	0.25011	0.17497			

O	0.62809	0.25017	0.17516
O	0.37866	0.24999	0.17289
O	0.87893	0.25013	0.17446
O	0.12929	0.75008	0.17391
O	0.62769	0.75009	0.17391
O	0.37754	0.75013	0.17469
O	0.87865	0.75015	0.17486
O	0.12929	0.99809	0.23184
O	0.62818	0.99781	0.23187
O	0.12929	0.50211	0.23184
O	0.62817	0.50246	0.23189
O	0.37917	0.00276	0.23188
O	0.87819	0.00136	0.23187
O	0.37917	0.49739	0.23187
O	0.87818	0.49895	0.23187
O	0.12919	0.75012	0.28965
O	0.62779	0.75011	0.28994
O	0.37748	0.75007	0.28902
O	0.87871	0.75016	0.28882
O	0.12887	0.25012	0.28874
O	0.62785	0.25011	0.28850
O	0.37855	0.25007	0.29086
O	0.87880	0.25015	0.28930
O	0.12949	0.49898	0.34563
O	0.63207	0.49385	0.34579
O	0.12947	0.00124	0.34563
O	0.63203	0.00636	0.34579
O	0.38067	0.50995	0.34818
O	0.88149	0.50137	0.34608
O	0.38064	0.99013	0.34819
O	0.88147	0.99894	0.34608
O	0.12317	0.25012	0.40187
O	0.62913	0.24999	0.40429
O	0.38856	0.25003	0.40137
O	0.88785	0.25019	0.40239
O	0.12286	0.75016	0.40124
O	0.61381	0.75011	0.39935
O	0.37438	0.74976	0.41110
O	0.88580	0.75017	0.40275
O	0.44123	0.89899	0.99961
O	0.44105	0.89898	0.46417
O	0.44219	0.60106	0.99950
O	0.44141	0.60097	0.46419
C	0.41633	0.74973	0.01437
C	0.41657	0.74990	0.44931

Table S22 - *The final minimised structure of the {110}-Redu-CO_3 ∠ configuration VIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00347	0.50604	0.06344	-1.77	-3.33	-0.75
Pu A	0.51181	0.50512	0.06146	-0.29	-0.36	-4.79
Pu A	0.00487	0.99297	0.17510	-2.22	-3.08	-0.01
Pu A	0.50845	0.99348	0.17493	-2.21	-3.09	-0.08
Pu A	0.00413	0.49617	0.28931	-2.22	-3.08	-0.01
Pu A	0.50892	0.49587	0.28929	-2.18	-3.11	-0.05
Pu A	0.00466	0.98458	0.39877	1.67	-3.36	-0.86
Pu A	0.51279	0.98751	0.40233	0.12	0.23	4.80
Pu B	0.26149	0.74421	0.11420	2.49	0.01	2.86
Pu B	0.75283	0.74442	0.11484	2.38	0.00	2.95
Pu B	0.25719	0.24483	0.23136	2.10	0.00	3.17
Pu B	0.75604	0.24531	0.23196	2.26	-0.01	3.06
Pu B	0.26097	0.74361	0.34946	2.33	0.11	3.01
Pu B	0.75339	0.74506	0.34953	2.38	-0.01	2.96
Pu B	0.25884	0.24318	0.12010	2.14	0.09	-3.13
Pu B	0.75335	0.24552	0.11393	2.21	0.00	-3.09
Pu C	0.25719	0.74365	0.23189	2.26	-0.01	-3.05
Pu C	0.75583	0.74438	0.23194	2.21	-0.01	-3.09
Pu C	0.25777	0.24161	0.34156	1.89	-0.20	-3.27
Pu C	0.75407	0.24738	0.35004	2.35	-0.04	-2.98
Pu C	0.00246	0.98330	0.06421	-1.80	3.30	-0.84
Pu C	0.51233	0.98588	0.06222	-0.31	0.38	-4.78
Pu D	0.00500	0.49592	0.17503	-2.21	3.09	-0.01
Pu D	0.50831	0.49574	0.17470	-2.22	3.08	-0.07
Pu D	0.00416	0.99193	0.28837	-2.24	3.06	-0.09
Pu D	0.50937	0.99392	0.28911	-2.19	3.10	-0.04
Pu D	0.00569	0.50904	0.40191	2.29	3.04	-0.38
Pu D	0.51201	0.50782	0.40293	-0.46	0.00	-4.78
O	0.12017	0.74617	0.06161			
O	0.63411	0.74599	0.06007			
O	0.37855	0.74620	0.06209			
O	0.88650	0.74530	0.05815			
O	0.14941	0.24681	0.05135			
O	0.62744	0.24531	0.06199			
O	0.90577	0.24442	0.06192			
O	0.12945	0.48409	0.11517			
O	0.63462	0.49709	0.11895			
O	0.12910	0.00461	0.11568			
O	0.63492	0.99304	0.11916			
O	0.38029	0.48339	0.11739			
O	0.88359	0.49999	0.11625			
O	0.38057	0.00501	0.11790			
O	0.88322	0.98950	0.11665			
O	0.13216	0.24421	0.17481			
O	0.63125	0.24503	0.17457			

O	0.38115	0.24478	0.17555
O	0.88242	0.24515	0.17379
O	0.13135	0.74438	0.17321
O	0.63122	0.74473	0.17524
O	0.38220	0.74407	0.17348
O	0.88151	0.74415	0.17472
O	0.13049	0.99255	0.23187
O	0.63230	0.99564	0.23212
O	0.13041	0.49553	0.23164
O	0.63213	0.49429	0.23205
O	0.38092	0.99143	0.23185
O	0.88130	0.99572	0.23156
O	0.38104	0.49732	0.23194
O	0.88208	0.49374	0.23203
O	0.13187	0.74054	0.29037
O	0.63177	0.74519	0.28893
O	0.38271	0.74506	0.29070
O	0.88192	0.74501	0.28889
O	0.13246	0.24632	0.28837
O	0.63208	0.24562	0.28953
O	0.38236	0.24430	0.28712
O	0.88227	0.24480	0.29010
O	0.13007	0.48198	0.34971
O	0.63451	0.49800	0.34511
O	0.12797	0.00074	0.34538
O	0.63417	0.99463	0.34511
O	0.38032	0.47830	0.34687
O	0.88291	0.50093	0.34747
O	0.38061	0.01049	0.34690
O	0.88181	0.99112	0.34692
O	0.17210	0.19512	0.40847
O	0.62678	0.24662	0.40216
O	0.91101	0.24689	0.40246
O	0.12156	0.75172	0.40107
O	0.63450	0.74999	0.40434
O	0.37999	0.74803	0.40122
O	0.88762	0.74524	0.40598
O	0.17718	0.42808	0.44874
O	0.24989	0.38254	0.00076
O	0.35456	0.29061	0.42974
O	0.35494	0.24612	0.05064
C	0.23621	0.30825	0.43091
C	0.25281	0.29436	0.03150

Table S23 - *The final minimised structure of the {110}-Redu-CO_3 ∠ configuration VIb using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.99882	0.51668	0.06463	-1.87	-3.38	0.17
Pu A	0.49997	0.54908	0.06185	1.41	-3.10	-1.76
Pu A	0.99881	0.01359	0.17597	-2.24	-3.07	0.09
Pu A	0.49753	0.01629	0.17332	-2.23	-3.08	-0.10
Pu A	0.99870	0.51846	0.28790	-2.22	-3.08	0.05
Pu A	0.49785	0.51517	0.28962	-2.22	-3.09	-0.04
Pu A	0.99839	0.01608	0.39940	-1.87	-3.39	-0.01
Pu A	0.50210	0.99085	0.40399	0.66	-3.86	-2.75
Pu B	0.24683	0.76723	0.11411	2.32	-0.05	3.01
Pu B	0.75181	0.76726	0.11554	2.19	0.01	3.10
Pu B	0.25043	0.26556	0.23188	2.09	0.00	3.17
Pu B	0.74612	0.26573	0.23204	2.21	0.00	3.09
Pu B	0.24647	0.76702	0.34970	2.12	0.10	3.15
Pu B	0.75166	0.76727	0.34804	2.33	-0.01	3.01
Pu B	0.23960	0.26453	0.11840	2.78	0.09	-3.91
Pu B	0.75454	0.26585	0.11780	2.59	-0.01	-2.80
Pu C	0.25026	0.76607	0.23188	2.25	0.00	-3.06
Pu C	0.74574	0.76665	0.23188	2.22	0.00	-3.08
Pu C	0.23959	0.26451	0.34541	-2.77	-0.10	-3.92
Pu C	0.75378	0.26600	0.34665	2.34	0.10	-2.97
Pu C	0.99858	0.01629	0.06445	-1.88	3.38	0.14
Pu C	0.50223	0.99143	0.05941	-3.25	2.57	-2.40
Pu D	0.99886	0.51891	0.17595	-2.23	3.07	0.09
Pu D	0.49791	0.51501	0.17406	-2.23	3.08	-0.10
Pu D	0.99865	0.01402	0.28790	-2.22	3.08	0.04
Pu D	0.49748	0.01623	0.29029	-2.22	3.08	-0.03
Pu D	0.99862	0.51685	0.39922	-1.86	3.39	-0.01
Pu D	0.50018	0.54913	0.40183	-1.47	3.11	-1.69
O	0.11594	0.76706	0.05999			
O	0.62567	0.76001	0.05977			
O	0.37155	0.75856	0.05776			
O	0.88096	0.76684	0.06180			
O	0.11654	0.26642	0.05883			
O	0.59437	0.27966	0.04749			
O	0.87567	0.26648	0.06286			
O	0.11796	0.52448	0.11785			
O	0.62558	0.50807	0.11535			
O	0.11725	0.00784	0.11783			
O	0.62843	0.02502	0.11535			
O	0.37622	0.50997	0.11410			
O	0.87437	0.51808	0.11838			
O	0.37394	0.01900	0.11467			
O	0.87508	0.01499	0.11836			
O	0.11939	0.26606	0.17625			
O	0.62419	0.26635	0.17438			

O	0.37704	0.26630	0.17622
O	0.87173	0.26627	0.17424
O	0.12425	0.76636	0.17518
O	0.62316	0.76582	0.17297
O	0.37356	0.76457	0.17237
O	0.87272	0.76650	0.17516
O	0.12463	0.01488	0.23194
O	0.62188	0.01467	0.23189
O	0.12467	0.51717	0.23194
O	0.62191	0.51754	0.23189
O	0.37479	0.01082	0.23187
O	0.87179	0.01706	0.23189
O	0.37481	0.52039	0.23187
O	0.87176	0.51549	0.23188
O	0.12425	0.76637	0.28871
O	0.62308	0.76596	0.29069
O	0.37363	0.76449	0.29142
O	0.87257	0.76645	0.28869
O	0.11954	0.26606	0.28774
O	0.62430	0.26642	0.28952
O	0.37718	0.26621	0.28747
O	0.87178	0.26627	0.28945
O	0.11775	0.52508	0.34602
O	0.62545	0.50930	0.34826
O	0.11702	0.00715	0.34605
O	0.62817	0.02433	0.34828
O	0.37608	0.50974	0.34971
O	0.87417	0.51674	0.34551
O	0.37360	0.01892	0.34915
O	0.87479	0.01634	0.34553
O	0.11553	0.26636	0.40472
O	0.59630	0.27952	0.41509
O	0.87446	0.26653	0.40153
O	0.11651	0.76700	0.40418
O	0.62596	0.76059	0.40389
O	0.37150	0.75798	0.40617
O	0.88169	0.76675	0.40159
O	0.51139	0.42487	0.46614
O	0.50630	0.42624	0.99722
O	0.53564	0.12980	0.46965
O	0.52910	0.13082	0.99344
C	0.54884	0.27644	0.45335
C	0.54398	0.27744	0.00967

Table S24 - *The final minimised structure of the {110}-Redu-CO_3 ∠ configuration VIc using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00957	0.50123	0.06471	-1.89	-3.38	-0.07
Pu A	0.52068	0.50518	0.05928	-3.30	-3.47	-0.30
Pu A	0.00254	0.99961	0.17549	-2.21	-3.09	0.05
Pu A	0.50645	0.00106	0.17361	-2.22	-3.08	-0.10
Pu A	0.00293	0.50295	0.28759	-2.23	-3.08	-0.02
Pu A	0.50483	0.50054	0.28912	-2.20	-3.10	-0.11
Pu A	0.00714	0.99886	0.39897	-1.86	-3.37	-0.32
Pu A	0.51764	0.99522	0.40446	-0.92	-3.51	-3.14
Pu B	0.25476	0.75091	0.11549	2.43	0.00	2.94
Pu B	0.75776	0.75063	0.11464	2.25	-0.03	3.05
Pu B	0.25587	0.25056	0.23153	2.16	0.00	3.13
Pu B	0.75210	0.25061	0.23162	2.22	0.00	3.08
Pu B	0.25361	0.75127	0.34906	3.67	0.10	3.11
Pu B	0.75821	0.75246	0.34751	2.24	0.01	3.07
Pu B	0.24976	0.25106	0.11894	2.24	-0.15	-3.05
Pu B	0.75528	0.25051	0.11429	2.18	0.00	-3.11
Pu C	0.25533	0.75099	0.23177	2.19	0.00	-3.11
Pu C	0.75099	0.75108	0.23156	2.21	0.00	-3.09
Pu C	0.24551	0.24960	0.34467	2.65	-0.06	-2.73
Pu C	0.75696	0.25021	0.34825	2.24	0.03	-3.06
Pu C	0.00961	0.00037	0.06470	-1.89	3.37	-0.07
Pu C	0.51993	0.99462	0.05946	-1.14	3.07	-3.52
Pu D	0.00251	0.50199	0.17555	-2.20	3.09	0.05
Pu D	0.50634	0.50054	0.17338	-2.23	3.08	-0.10
Pu D	0.00295	0.99867	0.28753	-2.23	3.08	-0.02
Pu D	0.50503	0.00113	0.29011	-2.21	3.09	-0.09
Pu D	0.00696	0.50309	0.39880	-1.83	3.39	-0.33
Pu D	0.52254	0.50673	0.40191	-3.36	1.70	0.73
O	0.12560	0.75086	0.06235			
O	0.63145	0.75001	0.06064			
O	0.36952	0.75021	0.05522			
O	0.89226	0.75080	0.06066			
O	0.13924	0.25099	0.06227			
O	0.62442	0.25024	0.06080			
O	0.89627	0.25075	0.06073			
O	0.13061	0.50287	0.11868			
O	0.63812	0.50179	0.11916			
O	0.13059	0.99899	0.11866			
O	0.63817	0.99917	0.11920			
O	0.38091	0.48926	0.11305			
O	0.88538	0.50000	0.11700			
O	0.38057	0.01325	0.11291			
O	0.88540	0.00140	0.11697			
O	0.12848	0.25085	0.17441			
O	0.62793	0.25068	0.17428			

O	0.37843	0.25055	0.17441
O	0.87868	0.25077	0.17457
O	0.13029	0.75079	0.17445
O	0.62764	0.75072	0.17502
O	0.37939	0.75124	0.17169
O	0.87890	0.75081	0.17447
O	0.12958	0.00287	0.23125
O	0.62714	0.00210	0.23214
O	0.12957	0.49864	0.23133
O	0.62715	0.49960	0.23198
O	0.37978	0.99849	0.23177
O	0.87710	0.00012	0.23150
O	0.37967	0.50307	0.23140
O	0.87705	0.50161	0.23157
O	0.12674	0.75082	0.28683
O	0.62896	0.75110	0.28924
O	0.38311	0.74982	0.29029
O	0.87822	0.75108	0.28872
O	0.12848	0.25051	0.28838
O	0.62791	0.25094	0.28966
O	0.37752	0.25137	0.28879
O	0.87791	0.25079	0.28893
O	0.12552	0.49895	0.34499
O	0.63504	0.50167	0.34531
O	0.12489	0.00196	0.34498
O	0.63649	0.00142	0.34605
O	0.38569	0.47658	0.35290
O	0.88161	0.50019	0.34577
O	0.38222	0.02269	0.35245
O	0.88229	0.00231	0.34575
O	0.13573	0.25056	0.40181
O	0.61431	0.26010	0.40415
O	0.89044	0.25096	0.40230
O	0.11903	0.75090	0.40539
O	0.62781	0.74354	0.40250
O	0.37607	0.73833	0.41203
O	0.88686	0.75170	0.40138
O	0.40275	0.90006	0.99889
O	0.41918	0.88285	0.46850
O	0.40361	0.60125	0.99880
O	0.43594	0.58495	0.46488
C	0.38781	0.75047	0.01445
C	0.40669	0.73707	0.45154

Table S25 - *The final minimised structure of the {110}-Redu-CO_3 ∠ configuration VId using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.99897	0.49869	0.06462	-1.85	-3.39	0.18
Pu A	0.51339	0.50906	0.06236	-2.36	-0.02	-3.01
Pu A	0.99701	0.99949	0.17579	-2.23	-3.07	0.05
Pu A	0.49824	0.99913	0.17361	-2.21	-3.09	-0.05
Pu A	0.99733	0.49989	0.28796	-2.21	-3.09	0.01
Pu A	0.49807	0.49917	0.28930	-2.20	-3.10	0.04
Pu A	0.99897	0.00074	0.39933	-1.87	-3.39	0.14
Pu A	0.51045	0.99322	0.40412	-4.49	0.76	-1.49
Pu B	0.24334	0.75030	0.11552	2.43	-0.01	2.94
Pu B	0.75263	0.75147	0.11583	2.28	-0.02	3.03
Pu B	0.24969	0.24877	0.23185	2.10	0.00	3.17
Pu B	0.74570	0.24918	0.23190	2.23	0.00	3.07
Pu B	0.24371	0.75033	0.34852	2.19	0.04	3.10
Pu B	0.75261	0.75139	0.34769	2.23	0.02	3.07
Pu B	0.24141	0.24823	0.11804	2.96	0.06	-3.79
Pu B	0.74879	0.24891	0.11565	2.27	0.00	-3.06
Pu C	0.24875	0.74955	0.23186	2.25	0.00	-3.06
Pu C	0.74525	0.75016	0.23183	2.20	0.00	-3.10
Pu C	0.24189	0.24787	0.34569	-2.85	-0.04	-3.87
Pu C	0.74907	0.24927	0.34817	2.28	0.04	-3.03
Pu C	0.99910	0.00096	0.06439	-1.87	3.38	0.17
Pu C	0.50894	0.99216	0.05970	-1.88	3.14	-3.11
Pu D	0.99697	0.49984	0.17578	-2.22	3.08	0.05
Pu D	0.49803	0.49912	0.17438	-2.23	3.08	-0.02
Pu D	0.99728	0.99939	0.28795	-2.22	3.09	0.01
Pu D	0.49811	0.99907	0.29011	-2.23	3.08	0.08
Pu D	0.99895	0.49882	0.39911	-1.84	3.40	0.16
Pu D	0.51325	0.50944	0.40143	-3.36	1.75	0.67
O	0.11721	0.74993	0.06108			
O	0.62094	0.74293	0.06074			
O	0.36498	0.74064	0.05278			
O	0.88119	0.75051	0.06141			
O	0.11775	0.24979	0.05908			
O	0.60013	0.25910	0.06027			
O	0.87773	0.24970	0.06152			
O	0.11928	0.50613	0.11813			
O	0.62888	0.50130	0.11814			
O	0.11853	0.99277	0.11810			
O	0.63023	0.99939	0.11768			
O	0.37908	0.49910	0.11226			
O	0.87507	0.49908	0.11778			
O	0.37573	0.99636	0.11264			
O	0.87578	0.00118	0.11775			
O	0.11967	0.24940	0.17606			
O	0.62251	0.24975	0.17396			

O	0.37627	0.24942	0.17562
O	0.87086	0.24964	0.17444
O	0.12387	0.74970	0.17458
O	0.62190	0.74945	0.17456
O	0.37156	0.74815	0.17236
O	0.87269	0.75004	0.17482
O	0.12339	0.99803	0.23184
O	0.62099	0.00032	0.23186
O	0.12340	0.50074	0.23184
O	0.62098	0.49890	0.23188
O	0.37361	0.99435	0.23188
O	0.87103	0.99968	0.23187
O	0.37345	0.50352	0.23185
O	0.87095	0.49980	0.23187
O	0.12389	0.74974	0.28902
O	0.62203	0.74975	0.28926
O	0.37162	0.74783	0.29144
O	0.87273	0.74993	0.28893
O	0.12002	0.24933	0.28783
O	0.62259	0.24968	0.28976
O	0.37647	0.24936	0.28796
O	0.87112	0.24965	0.28926
O	0.11949	0.50713	0.34566
O	0.62895	0.50145	0.34552
O	0.11866	0.99161	0.34570
O	0.63042	0.99969	0.34619
O	0.37912	0.49810	0.35148
O	0.87526	0.49839	0.34594
O	0.37611	0.99610	0.35105
O	0.87590	0.00185	0.34595
O	0.11666	0.24974	0.40434
O	0.59956	0.25957	0.40355
O	0.87638	0.24975	0.40255
O	0.11805	0.74994	0.40314
O	0.62117	0.74272	0.40292
O	0.36527	0.73923	0.41080
O	0.88209	0.75045	0.40196
O	0.40589	0.88163	0.99566
O	0.40566	0.88259	0.46769
O	0.42051	0.58370	0.99998
O	0.42079	0.58469	0.46394
C	0.39303	0.73693	0.01291
C	0.39300	0.73725	0.45067

Table S26 - *The final minimised structure of the {110}-Redu-CO_3 ⊥ configuration VIIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00184	0.50902	0.06374	-1.79	-3.32	-0.75
Pu A	0.51147	0.50684	0.06157	-0.84	0.00	4.73
Pu A	0.00436	0.99635	0.17515	-2.22	-3.08	-0.01
Pu A	0.50752	0.99669	0.17477	-2.22	-3.09	-0.07
Pu A	0.00449	0.49917	0.28880	-2.22	-3.08	-0.01
Pu A	0.50754	0.49836	0.28887	-2.19	-3.11	-0.04
Pu A	0.00179	0.98627	0.39966	1.80	-3.30	-0.81
Pu A	0.51176	0.98852	0.40167	0.94	-0.85	-4.64
Pu B	0.26082	0.74733	0.11409	2.44	0.01	2.91
Pu B	0.75188	0.74756	0.11494	2.43	-0.05	2.92
Pu B	0.25641	0.24770	0.23190	2.14	0.00	3.14
Pu B	0.75526	0.24778	0.23182	2.24	0.00	3.07
Pu B	0.26052	0.74748	0.34947	2.36	-0.01	2.98
Pu B	0.75179	0.74763	0.34888	2.40	0.00	2.94
Pu B	0.25801	0.24709	0.11997	2.15	0.05	-3.13
Pu B	0.75249	0.24779	0.11398	2.23	0.00	-3.07
Pu C	0.25665	0.74752	0.23186	2.26	0.00	-3.05
Pu C	0.75523	0.74747	0.23184	2.21	0.00	-3.09
Pu C	0.25829	0.24720	0.34388	2.12	-0.02	-3.16
Pu C	0.75272	0.24788	0.34948	2.33	0.00	-3.01
Pu C	0.00161	0.98639	0.06411	-1.82	3.29	-0.81
Pu C	0.51163	0.98797	0.06207	-1.02	0.84	-4.63
Pu D	0.00436	0.49893	0.17504	-2.22	3.08	-0.01
Pu D	0.50724	0.49849	0.17466	-2.22	3.09	-0.03
Pu D	0.00445	0.99616	0.28872	-2.22	3.08	-0.01
Pu D	0.50762	0.99683	0.28881	-2.19	3.11	-0.04
Pu D	0.00211	0.50900	0.39994	1.77	3.33	-0.77
Pu D	0.51187	0.50721	0.40195	1.00	0.95	-4.61
O	0.11935	0.74826	0.06137			
O	0.63331	0.74816	0.06021			
O	0.37746	0.74789	0.06171			
O	0.88553	0.74835	0.05826			
O	0.14889	0.25111	0.05088			
O	0.62677	0.24745	0.06202			
O	0.35427	0.24852	0.05150			
O	0.90493	0.24741	0.06210			
O	0.12815	0.48747	0.11530			
O	0.63346	0.49946	0.11887			
O	0.12833	0.00789	0.11549			
O	0.63421	0.99576	0.11914			
O	0.37919	0.48737	0.11759			
O	0.88240	0.50293	0.11646			
O	0.37971	0.00767	0.11769			
O	0.88258	0.99231	0.11664			
O	0.13140	0.24772	0.17502			

O	0.63043	0.24787	0.17446
O	0.38014	0.24738	0.17556
O	0.88169	0.24770	0.17374
O	0.13067	0.74753	0.17319
O	0.63040	0.74731	0.17509
O	0.38132	0.74766	0.17356
O	0.88092	0.74756	0.17476
O	0.12995	0.99590	0.23194
O	0.63180	0.99808	0.23179
O	0.12988	0.49932	0.23194
O	0.63164	0.49711	0.23181
O	0.38011	0.99551	0.23184
O	0.88147	0.99890	0.23186
O	0.38007	0.49957	0.23180
O	0.88150	0.49634	0.23186
O	0.13121	0.74764	0.29070
O	0.63057	0.74760	0.28861
O	0.38130	0.74739	0.29014
O	0.88155	0.74758	0.28877
O	0.13178	0.24766	0.28905
O	0.63067	0.24768	0.28898
O	0.38068	0.24766	0.28801
O	0.88158	0.24774	0.29009
O	0.12853	0.48864	0.34838
O	0.63414	0.49923	0.34457
O	0.12851	0.00679	0.34827
O	0.63422	0.99627	0.34449
O	0.37973	0.48645	0.34622
O	0.88277	0.50348	0.34723
O	0.37980	0.00837	0.34600
O	0.88273	0.99187	0.34708
O	0.15011	0.25033	0.41199
O	0.62661	0.24774	0.40146
O	0.35540	0.24787	0.41312
O	0.90541	0.24744	0.40151
O	0.11949	0.74809	0.40254
O	0.63368	0.74814	0.40347
O	0.37806	0.74797	0.40179
O	0.88559	0.74813	0.40543
O	0.25365	0.32818	0.99471
O	0.24988	0.30885	0.47026
C	0.25350	0.27758	0.02948
C	0.25318	0.27027	0.43473

Table S27 - *The final minimised structure of the {110}-Stoich-CO_2 || configuration VIIIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.00000	0.49990	0.06440	-2.00	-3.31	0.09
Pu A	0.49991	0.50019	0.06439	-2.04	-3.28	-0.15
Pu A	0.99958	0.00001	0.17539	-2.20	-3.10	0.00
Pu A	0.49989	0.99997	0.17537	-2.20	-3.10	0.00
Pu A	0.99976	0.49997	0.28848	-2.20	-3.10	0.00
Pu A	0.49950	0.49998	0.28848	-2.20	-3.10	0.00
Pu A	0.99944	0.99990	0.39879	-1.86	-3.39	-0.02
Pu A	0.49970	0.00002	0.39879	-1.86	-3.39	0.02
Pu B	0.24982	0.74998	0.11480	2.19	0.01	3.10
Pu B	0.74991	0.74963	0.11455	2.19	0.01	3.10
Pu B	0.24968	0.25014	0.23189	2.24	0.00	3.07
Pu B	0.74967	0.25019	0.23183	2.24	0.00	3.07
Pu B	0.24947	0.74953	0.34918	2.19	-0.01	3.10
Pu B	0.74954	0.75002	0.34892	2.19	0.00	3.11
Pu B	0.24972	0.24994	0.11479	2.19	0.00	-3.11
Pu B	0.74970	0.25043	0.11455	2.19	0.00	-3.11
Pu C	0.24969	0.74977	0.23190	2.24	0.00	-3.07
Pu C	0.74966	0.74982	0.23183	2.24	0.00	-3.07
Pu C	0.24968	0.25034	0.34919	2.19	-0.01	-3.10
Pu C	0.74964	0.24997	0.34892	2.19	0.00	-3.11
Pu C	0.99987	-0.00004	0.06495	-1.86	3.39	-0.02
Pu C	0.49963	0.00009	0.06492	-1.85	3.39	0.01
Pu D	0.99959	0.49998	0.17525	-2.20	3.10	0.00
Pu D	0.49989	0.49999	0.17523	-2.20	3.10	0.00
Pu D	0.99976	0.00000	0.28835	-2.20	3.10	0.00
Pu D	0.49950	0.99995	0.28834	-2.20	3.10	0.00
Pu D	0.99946	0.49982	0.39934	-2.02	3.29	0.12
Pu D	0.49963	0.50011	0.39934	-2.02	3.29	-0.13
O	0.11781	0.75112	0.06109			
O	0.61689	0.75003	0.06096			
O	0.38001	0.75066	0.06055			
O	0.88124	0.74989	0.06062			
O	0.11927	0.24900	0.06070			
O	0.61826	0.24999	0.06066			
O	0.38148	0.24908	0.06092			
O	0.88263	0.25022	0.06093			
O	0.12438	0.49964	0.11724			
O	0.62479	0.49981	0.11740			
O	0.12438	0.00033	0.11762			
O	0.62447	0.00029	0.11742			
O	0.37540	0.50027	0.11722			
O	0.87522	0.50027	0.11733			
O	0.37524	0.99960	0.11755			
O	0.87508	0.99977	0.11743			
O	0.12474	0.25022	0.17437			
O	0.62483	0.24997	0.17430			

O	0.37475	0.25016	0.17434
O	0.87466	0.25001	0.17431
O	0.12466	0.74971	0.17435
O	0.62475	0.75009	0.17431
O	0.37468	0.74977	0.17436
O	0.87459	0.74998	0.17430
O	0.12479	-0.00010	0.23185
O	0.62480	0.00001	0.23188
O	0.12472	0.49999	0.23190
O	0.62473	0.50002	0.23184
O	0.37456	0.00000	0.23184
O	0.87454	0.00001	0.23187
O	0.37465	0.49991	0.23187
O	0.87463	0.50001	0.23184
O	0.12475	0.74996	0.28943
O	0.62469	0.74977	0.28936
O	0.37462	0.74999	0.28940
O	0.87467	0.74978	0.28937
O	0.12466	0.24990	0.28940
O	0.62460	0.25021	0.28936
O	0.37453	0.24994	0.28943
O	0.87458	0.25022	0.28936
O	0.12432	0.50013	0.34638
O	0.62408	0.50029	0.34650
O	0.12428	0.99970	0.34631
O	0.62417	0.99963	0.34614
O	0.37480	0.49974	0.34636
O	0.87507	0.49971	0.34650
O	0.37490	0.00016	0.34629
O	0.87502	0.00035	0.34613
O	0.11668	0.25005	0.40281
O	0.61775	0.24895	0.40272
O	0.38106	0.24997	0.40309
O	0.87994	0.24916	0.40309
O	0.11807	0.74986	0.40310
O	0.61922	0.75077	0.40311
O	0.38244	0.74986	0.40280
O	0.88142	0.75107	0.40271
O	0.63980	0.49822	0.47492
O	0.15183	0.50185	0.98911
O	0.85640	0.50159	0.47485
O	0.36843	0.49936	0.98877
C	0.74811	0.49990	0.47403
C	0.26002	0.50063	0.98982

Table S28 *- The final minimised structure of the {110}-Reduced-CO_2 ‖ configuration IXa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.98681	0.50078	0.06459	-1.89	-3.36	-0.19
Pu A	0.49467	0.51678	0.06155	-1.52	-2.11	-4.04
Pu A	0.97946	0.99787	0.17575	-2.22	-3.08	0.04
Pu A	0.48164	0.99975	0.17420	-2.22	-3.08	-0.10
Pu A	0.98012	0.50101	0.28822	-2.22	-3.09	0.02
Pu A	0.48013	0.49980	0.28864	-2.21	-3.10	0.01
Pu A	0.98257	0.00054	0.39908	-1.74	-3.46	-0.04
Pu A	0.49080	0.98151	0.40157	1.36	-1.94	-4.18
Pu B	0.23713	0.74946	0.11297	2.45	0.00	2.90
Pu B	0.73651	0.74966	0.11503	2.28	0.00	3.04
Pu B	0.23168	0.24942	0.23227	2.08	0.00	3.18
Pu B	0.72944	0.24997	0.23170	2.25	0.00	3.06
Pu B	0.23160	0.74875	0.35012	2.12	0.13	3.15
Pu B	0.73661	0.75165	0.34776	2.32	-0.03	3.01
Pu B	0.22504	0.25004	0.11975	2.22	-0.21	-3.05
Pu B	0.72950	0.24981	0.11374	2.18	0.00	-3.11
Pu C	0.23029	0.74958	0.23155	2.25	0.01	-3.06
Pu C	0.72785	0.75012	0.23173	2.22	0.00	-3.08
Pu C	0.22353	0.24803	0.34532	-2.71	-0.10	-3.96
Pu C	0.72948	0.25104	0.34870	2.18	0.06	-3.11
Pu C	0.98657	0.99920	0.06512	-1.70	3.47	-0.03
Pu C	0.49478	0.98291	0.06215	-1.22	2.06	-4.17
Pu D	0.97950	0.50170	0.17566	-2.21	3.09	0.04
Pu D	0.48169	0.49963	0.17387	-2.23	3.08	-0.10
Pu D	0.98003	0.99897	0.28809	-2.22	3.09	0.02
Pu D	0.48014	0.99927	0.28951	-2.22	3.09	0.01
Pu D	0.98254	0.50012	0.39956	-1.88	3.37	0.12
Pu D	0.49022	0.52097	0.39973	-2.10	2.97	-1.35
O	0.10285	0.74980	0.06014			
O	0.61784	0.74985	0.05865			
O	0.35933	0.74985	0.06046			
O	0.87013	0.75069	0.06111			
O	0.11702	0.24971	0.06132			
O	0.59372	0.24941	0.06087			
O	0.87089	0.24902	0.06024			
O	0.10675	0.50260	0.11835			
O	0.61214	0.50502	0.11828			
O	0.10642	0.99687	0.11837			
O	0.61219	0.99440	0.11851			
O	0.35694	0.48008	0.11445			
O	0.86048	0.49766	0.11681			
O	0.35683	0.01912	0.11459			
O	0.86030	0.00196	0.11714			
O	0.10392	0.24963	0.17490			
O	0.60446	0.24982	0.17342			
O	0.35431	0.24947	0.17582			

O	0.85435	0.25008	0.17456
O	0.10498	0.74980	0.17519
O	0.60510	0.74975	0.17531
O	0.35700	0.74967	0.17129
O	0.85520	0.74972	0.17444
O	0.10574	0.00099	0.23174
O	0.60425	0.00279	0.23201
O	0.10568	0.49823	0.23178
O	0.60438	0.49725	0.23180
O	0.35603	0.99361	0.23176
O	0.85420	0.99873	0.23186
O	0.35591	0.50509	0.23142
O	0.85427	0.50145	0.23189
O	0.10625	0.74971	0.28845
O	0.60528	0.75060	0.28896
O	0.35523	0.74840	0.29136
O	0.85564	0.75017	0.28938
O	0.10166	0.24958	0.28815
O	0.60528	0.25020	0.29052
O	0.35861	0.24925	0.28709
O	0.85294	0.25062	0.28947
O	0.10170	0.50865	0.34621
O	0.60905	0.50667	0.34595
O	0.10108	0.98972	0.34608
O	0.61157	0.99720	0.34674
O	0.36072	0.49145	0.34915
O	0.85684	0.49777	0.34648
O	0.35807	0.00202	0.34951
O	0.85731	0.00457	0.34609
O	0.10066	0.25012	0.40542
O	0.57056	0.26269	0.40359
O	0.85720	0.25032	0.40327
O	0.10262	0.75047	0.40493
O	0.61387	0.74212	0.40508
O	0.36268	0.73620	0.40554
O	0.86726	0.75254	0.40179
O	0.86132	0.50414	0.98797
O	0.84105	0.51390	0.47544
O	0.64502	0.49528	0.98455
O	0.62474	0.50042	0.47775
C	0.75288	0.49987	0.98710
C	0.73283	0.50747	0.47573

Section S6: Surface Structure and Magnetisation - {111} Adsorption

Table S29 - *The final minimised structure of the {111}-Stoich-CO_3 ∠ configuration Xa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.67422	0.65867	0.08555	-3.08	-2.09	-0.85
Pu A	0.33768	0.33193	0.17404	-3.10	-1.82	-1.24
Pu A	0.00341	0.00048	0.26655	-3.10	-1.80	-1.25
Pu A	0.66918	0.66899	0.35905	-3.10	-1.82	-1.23
Pu A	0.33271	0.34208	0.44755	-3.08	-2.09	-0.87
Pu B	0.67784	0.17096	0.08213	0.29	3.55	-1.36
Pu B	0.33588	0.83571	0.17464	0.00	3.59	-1.25
Pu B	0.00342	0.50048	0.26655	-0.01	3.59	-1.25
Pu B	0.67097	0.16517	0.35845	0.00	3.59	-1.25
Pu B	0.32900	0.82989	0.45096	0.26	3.54	-1.38
Pu C	0.16955	0.66727	0.08377	3.09	-2.03	-0.96
Pu C	0.83724	0.33258	0.17548	3.09	-1.80	-1.28
Pu C	0.50342	0.00048	0.26655	3.10	-1.80	-1.25
Pu C	0.16961	0.66830	0.35761	3.09	-1.80	-1.28
Pu C	0.83733	0.33356	0.44933	3.08	-2.05	-0.96
Pu D	0.15914	0.17096	0.08223	-0.11	-0.14	3.79
Pu D	0.83561	0.83561	0.17465	0.00	0.01	3.80
Pu D	0.50342	0.50047	0.26655	-0.03	0.01	3.80
Pu D	0.17123	0.16529	0.35844	0.01	0.01	3.80
Pu D	0.84778	0.82981	0.45089	0.03	-0.19	3.79
O	0.00630	0.99498	0.06243			
O	0.50306	0.50905	0.06468			
O	0.51298	0.98091	0.05534			
O	0.99616	0.50900	0.06458			
O	0.33907	0.84339	0.10713			
O	0.83874	0.33003	0.10697			
O	0.82607	0.84269	0.10693			
O	0.34481	0.31786	0.10473			
O	0.67216	0.66319	0.15197			
O	0.16642	0.17023	0.15222			
O	0.17137	0.66464	0.14982			
O	0.67063	0.16994	0.15209			
O	0.00405	0.50269	0.19776			
O	0.50391	0.99948	0.19730			
O	0.50079	0.50242	0.19775			
O	0.00497	0.99719	0.19774			
O	0.33718	0.33297	0.24339			
O	0.83609	0.83444	0.24366			
O	0.83699	0.33344	0.24374			
O	0.33676	0.83441	0.24370			
O	0.67005	0.16652	0.28940			
O	0.16982	0.66750	0.28937			
O	0.17075	0.16652	0.28943			

O	0.66966	0.66801	0.28971
O	0.00184	0.00376	0.33536
O	0.50607	0.49847	0.33534
O	0.50290	0.00144	0.33580
O	0.00280	0.49820	0.33534
O	0.33621	0.83093	0.38099
O	0.83545	0.33625	0.38328
O	0.84047	0.83063	0.38086
O	0.33472	0.33767	0.38113
O	0.66204	0.68313	0.42837
O	0.18084	0.15808	0.42616
O	0.16808	0.67081	0.42614
O	0.66776	0.15735	0.42597
O	0.01079	0.49167	0.46850
O	0.49382	0.02002	0.47785
O	0.50389	0.49172	0.46838
O	0.00055	0.00583	0.47067
O	0.56137	0.17954	0.01449
O	0.44538	0.82087	0.51856
O	0.26366	0.17656	0.01461
O	0.74337	0.82336	0.51846
C	0.44382	0.11672	0.02438
C	0.56305	0.88366	0.50872

Table S30 - *The final minimised structure of the {111}-Stoich-CO_3 ⊥ configuration XIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.66645	0.65742	0.08315	-3.09	-2.15	-0.69
Pu A	0.33200	0.33086	0.17299	-3.09	-1.82	-1.26
Pu A	0.99772	0.99862	0.26548	-3.10	-1.80	-1.25
Pu A	0.66499	0.66576	0.35697	-3.08	-1.81	-1.30
Pu A	0.32861	0.33957	0.45024	-2.76	-2.19	-1.44
Pu B	0.66743	0.16994	0.08024	0.50	3.48	-1.47
Pu B	0.33063	0.83294	0.17351	0.00	3.58	-1.27
Pu B	0.99792	0.49854	0.26501	0.00	3.59	-1.25
Pu B	0.66361	0.16521	0.35752	0.05	3.58	-1.28
Pu B	0.32935	0.82603	0.44729	0.32	3.75	-0.69
Pu C	0.16310	0.66662	0.08227	3.18	-1.94	-0.85
Pu C	0.82991	0.33122	0.17406	3.08	-1.80	-1.31
Pu C	0.49784	0.99874	0.26525	3.11	-1.79	-1.26
Pu C	0.16581	0.66334	0.35643	3.10	-1.77	-1.31
Pu C	0.83248	0.33208	0.44819	3.27	-1.77	-0.87
Pu D	0.15476	0.16942	0.08300	0.31	-0.18	3.77
Pu D	0.82929	0.83384	0.17304	0.00	-0.01	3.80
Pu D	0.49782	0.49840	0.26523	-0.02	0.01	3.80
Pu D	0.16637	0.16542	0.35745	0.00	0.00	3.80
Pu D	0.84114	0.82619	0.44748	-3.00	1.89	1.40
O	0.00593	0.99511	0.06248			
O	0.49353	0.50750	0.06276			
O	0.48769	0.99029	0.05374			
O	0.99652	0.49847	0.06156			
O	0.33627	0.83932	0.10627			
O	0.83019	0.33136	0.10577			
O	0.81914	0.83838	0.10437			
O	0.33633	0.32019	0.10457			
O	0.66666	0.66191	0.15030			
O	0.16042	0.16814	0.15063			
O	0.16335	0.66500	0.14853			
O	0.66512	0.16626	0.15121			
O	0.99821	0.50014	0.19639			
O	0.49699	0.99773	0.19606			
O	0.49475	0.50101	0.19642			
O	0.99974	0.99568	0.19645			
O	0.33190	0.33108	0.24207			
O	0.83046	0.83305	0.24219			
O	0.83108	0.33108	0.24239			
O	0.33085	0.83241	0.24262			
O	0.66366	0.16529	0.28841			
O	0.16465	0.66427	0.28808			
O	0.16523	0.16567	0.28832			
O	0.66485	0.66539	0.28786			

O	0.99735	0.00051	0.33409
O	0.50095	0.49807	0.33406
O	0.49890	0.99681	0.33444
O	0.99586	0.49777	0.33406
O	0.32894	0.83095	0.38015
O	0.83233	0.33052	0.38192
O	0.83542	0.83073	0.38006
O	0.33074	0.33353	0.37922
O	0.65931	0.67722	0.42419
O	0.17644	0.16005	0.42610
O	0.16585	0.66349	0.42467
O	0.65904	0.15957	0.42592
O	0.98983	0.50363	0.46800
O	0.50849	0.97987	0.47657
O	0.50220	0.50346	0.46777
O	0.99910	0.99688	0.46883
O	0.47838	0.95749	0.98704
O	0.52043	0.93711	0.54332
O	0.62417	0.12566	0.01480
O	0.37479	0.25130	0.51567
C	0.53116	0.02468	0.01378
C	0.46707	0.05735	0.51662

Table S31 - *The final minimised structure of the {111}-Redu-CO_3 ∠ configuration XIIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.68058	0.67064	0.08471	-3.07	-1.79	-3.22
Pu A	0.33790	0.33253	0.17388	-3.07	-1.80	-1.32
Pu A	0.00344	0.00055	0.26660	-3.09	-1.78	-1.31
Pu A	0.66901	0.66854	0.35934	-3.07	-1.80	-1.32
Pu A	0.32634	0.33048	0.44850	-3.07	-1.80	-3.22
Pu B	0.69028	0.15117	0.08521	-3.56	-0.57	-1.31
Pu B	0.33610	0.83396	0.17361	0.00	3.58	-1.27
Pu B	0.00344	0.50053	0.26660	-0.01	3.55	-1.34
Pu B	0.67082	0.16709	0.35960	0.00	3.58	-1.27
Pu B	0.31666	0.84991	0.44800	-3.56	-0.57	-1.31
Pu C	0.15677	0.66873	0.08316	3.29	-1.59	-3.12
Pu C	0.83549	0.33478	0.17537	3.07	-1.78	-1.35
Pu C	0.50345	0.00054	0.26661	3.09	-1.78	-1.31
Pu C	0.17139	0.66631	0.35783	3.07	-1.78	-1.35
Pu C	0.85018	0.33231	0.45005	3.30	-1.60	-3.11
Pu D	0.15410	0.17219	0.08121	-0.86	-0.06	3.69
Pu D	0.83791	0.83224	0.17627	0.04	-0.04	3.80
Pu D	0.50345	0.50054	0.26661	-0.03	0.02	3.80
Pu D	0.16900	0.16883	0.35695	0.04	-0.04	3.80
Pu D	0.85285	0.82888	0.45201	-0.68	-0.15	3.72
O	0.99454	0.01094	0.05996			
O	0.51991	0.99883	0.05086			
O	0.00372	0.48952	0.06298			
O	0.34030	0.83518	0.10403			
O	0.81677	0.33860	0.10548			
O	0.82974	0.84075	0.10764			
O	0.35958	0.31986	0.10472			
O	0.67291	0.66628	0.15326			
O	0.16298	0.17186	0.14962			
O	0.16855	0.66548	0.15175			
O	0.67575	0.16295	0.15368			
O	0.00598	0.49786	0.19820			
O	0.49971	0.00163	0.19743			
O	0.50161	0.50132	0.19737			
O	0.00689	0.99932	0.19804			
O	0.33751	0.33346	0.24352			
O	0.83633	0.83408	0.24395			
O	0.83623	0.33395	0.24370			
O	0.33731	0.83345	0.24344			
O	0.66961	0.16763	0.28977			
O	0.17064	0.66715	0.28950			
O	0.17053	0.16698	0.28926			
O	0.66937	0.66760	0.28969			
O	0.00002	0.00176	0.33518			

O	0.50525	0.49975	0.33584
O	0.50719	0.99947	0.33579
O	0.00093	0.50320	0.33501
O	0.33119	0.83812	0.37954
O	0.83835	0.33559	0.38146
O	0.84390	0.82921	0.38357
O	0.33400	0.33479	0.37996
O	0.64735	0.68116	0.42849
O	0.17719	0.16032	0.42557
O	0.19004	0.66257	0.42772
O	0.66668	0.16587	0.42920
O	0.00319	0.51157	0.47024
O	0.48699	0.00229	0.48233
O	0.01245	0.99011	0.47327
O	0.51525	0.26802	0.02421
O	0.49172	0.73315	0.50903
O	0.26248	0.19867	0.01207
O	0.74440	0.80268	0.52115
C	0.42888	0.16077	0.02595
C	0.57802	0.84046	0.50727

Table S32 *- The final minimised structure of the {111}-Redu-CO_3 ∠ configuration XIIb using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.67283	0.64605	0.08277	-2.86	-2.49	-0.50
Pu A	0.33339	0.32535	0.17501	-3.10	-1.82	-1.24
Pu A	0.99650	0.99218	0.26528	-3.10	-1.79	-1.27
Pu A	0.66497	0.65939	0.35866	-3.09	-1.81	-1.28
Pu A	0.31104	0.33141	0.44672	-1.30	3.37	-1.30
Pu B	0.65425	0.16979	0.07970	-3.59	2.06	2.43
Pu B	0.32831	0.82776	0.17321	0.00	3.63	-1.14
Pu B	0.99524	0.49475	0.26515	0.00	3.58	-1.28
Pu B	0.66322	0.15965	0.35821	0.04	3.55	-1.34
Pu B	0.32036	0.84154	0.44726	-0.20	3.37	-3.41
Pu C	0.17492	0.63960	0.08256	0.20	-3.49	3.29
Pu C	0.83052	0.32337	0.17405	3.11	-1.82	-1.21
Pu C	0.49942	0.99075	0.26546	3.10	-1.79	-1.27
Pu C	0.16520	0.65981	0.35663	3.08	-1.77	-1.35
Pu C	0.84440	0.31507	0.44892	3.08	-2.09	-3.04
Pu D	0.16300	0.17282	0.08362	-0.43	0.41	3.78
Pu D	0.83059	0.82787	0.17227	-0.03	0.01	3.80
Pu D	0.49821	0.49313	0.26580	-0.01	0.03	3.80
Pu D	0.16294	0.15971	0.35555	0.06	-0.02	3.80
Pu D	0.84711	0.81598	0.45088	-3.12	1.95	0.99
O	0.50848	0.50617	0.06047			
O	0.50602	0.96403	0.05319			
O	0.98756	0.48834	0.06109			
O	0.30703	0.85225	0.10403			
O	0.84864	0.29963	0.10443			
O	0.82942	0.83333	0.10395			
O	0.33660	0.31022	0.10433			
O	0.66280	0.65350	0.14849			
O	0.16383	0.16574	0.15234			
O	0.17010	0.65150	0.15080			
O	0.66017	0.16394	0.15225			
O	0.99223	0.49731	0.19674			
O	0.50068	0.98624	0.19633			
O	0.49791	0.49622	0.19735			
O	0.99866	0.98967	0.19594			
O	0.33099	0.32490	0.24245			
O	0.83070	0.82754	0.24231			
O	0.83193	0.32368	0.24250			
O	0.32883	0.82771	0.24249			
O	0.66238	0.16050	0.28861			
O	0.16518	0.65800	0.28829			
O	0.16466	0.15977	0.28806			
O	0.66345	0.65928	0.28845			
O	0.99467	0.99279	0.33366			

O	0.49969	0.49257	0.33484
O	0.50215	0.98956	0.33463
O	0.99292	0.49651	0.33397
O	0.32763	0.82898	0.37871
O	0.83294	0.32298	0.38028
O	0.83792	0.82465	0.38260
O	0.32523	0.32817	0.37821
O	0.66064	0.66446	0.42799
O	0.17155	0.16039	0.42437
O	0.18415	0.64539	0.42648
O	0.64174	0.16927	0.42725
O	0.00638	0.49592	0.47201
O	0.48274	0.00854	0.48105
O	0.99763	0.98284	0.46908
O	0.53561	0.15971	0.00832
O	0.73814	0.95263	0.52031
O	0.24517	0.14311	0.01384
O	0.48635	0.27427	0.50749
C	0.42762	0.09243	0.02125
C	0.57279	0.08046	0.50605

Table S33 - *The final minimised structure of the {111}-Stoich-CO_2 ∠ configuration XIIIa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.66696	0.66638	0.08252	-3.22	-1.83	-0.94
Pu A	0.33294	0.33379	0.17334	-3.09	-1.78	-1.30
Pu A	0.00011	0.99990	0.26521	-3.10	-1.79	-1.26
Pu A	0.66659	0.66686	0.35711	-3.09	-1.79	-1.29
Pu A	0.33356	0.33280	0.44778	-3.20	-1.88	-0.90
Pu B	0.66682	0.16648	0.08269	-0.01	3.71	-0.92
Pu B	0.33315	0.83331	0.17334	0.00	3.57	-1.30
Pu B	0.00009	0.49993	0.26531	0.00	3.58	-1.26
Pu B	0.66702	0.16604	0.35699	0.00	3.57	-1.29
Pu B	0.33409	0.83205	0.44849	0.00	3.69	-1.01
Pu C	0.16553	0.66828	0.08193	3.18	-1.82	-1.10
Pu C	0.83347	0.33346	0.17348	3.09	-1.78	-1.31
Pu C	0.50001	0.99994	0.26516	3.10	-1.79	-1.27
Pu C	0.16667	0.66685	0.35712	3.09	-1.78	-1.29
Pu C	0.83389	0.33279	0.44779	3.20	-1.89	-0.89
Pu D	0.16660	0.16686	0.08253	-0.01	0.04	3.79
Pu D	0.83365	0.83311	0.17340	0.00	0.00	3.80
Pu D	0.49999	0.49999	0.26525	0.00	0.00	3.80
Pu D	0.16707	0.16601	0.35707	0.00	0.00	3.80
Pu D	0.83333	0.83346	0.44813	-0.01	-0.09	3.79
O	0.99812	0.00360	0.06088			
O	0.50187	0.49764	0.05983			
O	0.50000	0.00014	0.06032			
O	0.00057	0.49760	0.05954			
O	0.33368	0.83240	0.10545			
O	0.83316	0.33494	0.10596			
O	0.83431	0.83166	0.10533			
O	0.33204	0.33484	0.10560			
O	0.66631	0.66755	0.14995			
O	0.16707	0.16625	0.15018			
O	0.16618	0.66740	0.14993			
O	0.66677	0.16612	0.14988			
O	0.99986	0.50006	0.19629			
O	0.50009	0.00021	0.19633			
O	0.50019	0.49980	0.19627			
O	0.99965	0.00035	0.19634			
O	0.33308	0.33363	0.24234			
O	0.83360	0.83305	0.24229			
O	0.83337	0.33348	0.24241			
O	0.33336	0.83312	0.24228			
O	0.66681	0.16634	0.28806			
O	0.16662	0.66686	0.28820			
O	0.16691	0.16627	0.28815			
O	0.66657	0.66690	0.28817			

O	0.00001	0.00026	0.33418
O	0.50030	0.49951	0.33415
O	0.49980	0.00027	0.33416
O	0.00041	0.49930	0.33415
O	0.33369	0.83281	0.38052
O	0.83352	0.33357	0.38060
O	0.83378	0.83250	0.38021
O	0.33307	0.33355	0.38060
O	0.66620	0.66864	0.42500
O	0.16771	0.16477	0.42501
O	0.16532	0.66871	0.42499
O	0.66830	0.16373	0.42453
O	0.00021	0.49989	0.47013
O	0.49954	0.00233	0.47092
O	0.50192	0.49625	0.46927
O	0.99846	0.00229	0.47096
O	0.16610	0.15843	0.55938
O	0.33371	0.83200	0.53270
O	0.16641	0.66761	0.99776
O	0.33224	0.34238	0.97053
C	0.25000	0.50378	0.98439
C	0.24921	0.99643	0.54579

Table S34 - *The final minimised structure of the {111}-Redu-CO_2 ∠ configuration XIVa using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.64715	0.66366	0.08072	-3.25	-1.91	-0.63
Pu A	0.31244	0.33341	0.17529	-3.07	-1.81	-1.32
Pu A	0.97734	0.99781	0.26511	-3.10	-1.79	-1.28
Pu A	0.64327	0.66556	0.35760	-3.05	-1.74	-1.44
Pu A	0.30097	0.32332	0.44842	-2.86	-1.95	-3.32
Pu B	0.62493	0.17471	0.08183	-2.89	1.90	3.32
Pu B	0.31004	0.83223	0.17298	0.04	3.60	-1.22
Pu B	0.97605	0.50097	0.26516	0.00	3.57	-1.29
Pu B	0.64280	0.16757	0.35745	-0.01	3.53	-1.39
Pu B	0.30245	0.84858	0.44796	0.37	3.32	-3.44
Pu C	0.15287	0.64931	0.08230	0.31	-3.40	3.37
Pu C	0.81163	0.33003	0.17309	3.13	-1.81	-1.18
Pu C	0.48007	0.99674	0.26499	3.09	-1.79	-1.29
Pu C	0.14508	0.66520	0.35808	3.08	-1.76	-1.35
Pu C	0.82947	0.32443	0.44603	2.62	-0.97	-2.60
Pu D	0.15241	0.17507	0.08455	-2.46	2.69	1.23
Pu D	0.81179	0.83254	0.17214	-0.04	0.00	3.80
Pu D	0.47828	0.49947	0.26554	0.03	0.01	3.80
Pu D	0.14659	0.16445	0.35505	-0.01	0.00	3.80
Pu D	0.81061	0.83645	0.44967	0.32	-3.63	1.12
O	0.48716	0.51209	0.05694			
O	0.46573	0.99125	0.05600			
O	0.97449	0.48666	0.05773			
O	0.29022	0.85083	0.10482			
O	0.83060	0.30538	0.10369			
O	0.81279	0.83705	0.10304			
O	0.30970	0.32926	0.10633			
O	0.64452	0.66095	0.14734			
O	0.14730	0.16980	0.15187			
O	0.14726	0.66085	0.15151			
O	0.63997	0.16865	0.15143			
O	0.97249	0.50219	0.19652			
O	0.48154	0.99284	0.19628			
O	0.48017	0.50101	0.19688			
O	0.97786	0.99808	0.19565			
O	0.31117	0.33172	0.24253			
O	0.81193	0.83289	0.24204			
O	0.81232	0.33024	0.24220			
O	0.30964	0.83311	0.24228			
O	0.64332	0.16699	0.28827			
O	0.14597	0.66411	0.28846			
O	0.14482	0.16583	0.28774			
O	0.64416	0.66474	0.28806			
O	0.97546	0.99644	0.33383			

O	0.47837	0.49916	0.33478
O	0.48278	0.99594	0.33363
O	0.97471	0.50415	0.33411
O	0.30910	0.83631	0.37873
O	0.81826	0.32760	0.37898
O	0.80691	0.83640	0.38295
O	0.31003	0.32946	0.37883
O	0.64153	0.64787	0.42563
O	0.14170	0.16798	0.42386
O	0.15121	0.66066	0.42730
O	0.63775	0.19010	0.42688
O	0.95836	0.50533	0.47408
O	0.50060	0.98519	0.47378
O	0.96269	0.01192	0.47259
O	0.72128	0.71119	0.54939
O	0.56939	0.57049	0.51042
O	0.14150	0.94885	0.02037
O	0.43299	0.81128	0.98114
C	0.29590	0.88001	0.00256
C	0.65054	0.64270	0.52852

Table S35 - *The final minimised structure of the {111}-Redu-CO_2 ∠ configuration XIVb using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.67849	0.67811	0.08391	-0.99	-0.56	-3.64
Pu A	0.33335	0.33327	0.17246	-3.08	-1.78	-1.32
Pu A	0.00002	0.99998	0.26523	-3.07	-1.77	-1.35
Pu A	0.66651	0.66654	0.35801	-3.09	-1.75	-1.35
Pu A	0.32135	0.32207	0.44655	-0.94	-0.60	-3.65
Pu B	0.67420	0.15145	0.08241	-0.11	3.62	-3.14
Pu B	0.33424	0.83135	0.17293	0.00	3.54	-1.38
Pu B	-0.00001	0.50000	0.26524	0.00	3.56	-1.33
Pu B	0.66578	0.16875	0.35748	0.00	3.54	-1.38
Pu B	0.32571	0.84857	0.44808	-0.10	3.57	-3.20
Pu C	0.15177	0.67380	0.08241	3.09	-1.92	-3.13
Pu C	0.83136	0.33418	0.17293	3.06	-1.77	-1.38
Pu C	0.50002	-0.00003	0.26523	3.08	-1.78	-1.33
Pu C	0.16891	0.66573	0.35750	3.06	-1.75	-1.40
Pu C	0.84857	0.32621	0.44806	3.04	-1.91	-3.18
Pu D	0.16612	0.16538	0.08055	-0.07	-0.56	3.74
Pu D	0.83463	0.83451	0.17522	0.02	0.01	3.80
Pu D	0.50001	0.50000	0.26520	-0.01	-0.01	3.80
Pu D	0.16546	0.16546	0.35524	0.02	0.01	3.80
Pu D	0.83322	0.83581	0.44963	-0.22	-3.66	1.07
O	0.00998	0.00955	0.05818			
O	0.49413	0.99211	0.05627			
O	0.99231	0.49399	0.05634			
O	0.35625	0.80815	0.10356			
O	0.80804	0.35612	0.10357			
O	0.83148	0.83123	0.10640			
O	0.33480	0.33443	0.10425			
O	0.67056	0.67047	0.15143			
O	0.16306	0.16311	0.14768			
O	0.16306	0.67010	0.15155			
O	0.67023	0.16287	0.15157			
O	0.00229	0.49596	0.19641			
O	0.49598	0.00222	0.19641			
O	0.50012	0.50008	0.19576			
O	0.00198	0.00187	0.19685			
O	0.33374	0.33370	0.24213			
O	0.83329	0.83328	0.24270			
O	0.83234	0.33405	0.24216			
O	0.33408	0.83232	0.24216			
O	0.66594	0.16770	0.28829			
O	0.16772	0.66593	0.28829			
O	0.16673	0.16670	0.28774			
O	0.66625	0.66624	0.28834			
O	0.99799	0.99809	0.33359			

O	0.49994	0.49991	0.33466
O	0.50422	0.99768	0.33405
O	0.99765	0.50417	0.33407
O	0.32978	0.83712	0.37883
O	0.83690	0.32981	0.37888
O	0.83698	0.83704	0.38287
O	0.32971	0.32958	0.37899
O	0.66485	0.66530	0.42618
O	0.16872	0.16860	0.42399
O	0.19225	0.64388	0.42683
O	0.64440	0.19070	0.42694
O	0.00804	0.50579	0.47394
O	0.50598	0.00769	0.47416
O	0.99034	0.99053	0.47227
O	0.26703	0.26386	0.97789
O	0.58703	0.59053	0.51328
O	0.41234	0.41017	0.01710
O	0.73260	0.73628	0.55257
C	0.33755	0.33473	0.99821
C	0.66194	0.66575	0.53221

Table S36 - *The final minimised structure of the {111}-Redu-CO_2 ∠ configuration XIVc using PBEsol + U. The ionic positions are given as fractional coordinates and the magnetic vectors are presented.*

Atom	Coordinates (Direct)			Magnetic Vector (μ_B)		
	X	Y	Z	X	Y	Z
Pu A	0.67692	0.67595	0.08447	-2.41	-1.37	-2.61
Pu A	0.33344	0.33343	0.17237	-3.09	-1.75	-1.34
Pu A	0.00000	1.00000	0.26522	-3.07	-1.77	-1.34
Pu A	0.66656	0.66657	0.35808	-3.09	-1.75	-1.34
Pu A	0.32308	0.32405	0.44597	-2.41	-1.37	-2.61
Pu B	0.67507	0.14902	0.08242	-0.26	3.37	-3.40
Pu B	0.33358	0.83154	0.17295	0.00	3.53	-1.39
Pu B	0.00000	0.50000	0.26522	0.00	3.55	-1.33
Pu B	0.66642	0.16846	0.35750	0.00	3.53	-1.39
Pu B	0.32493	0.85098	0.44803	-0.26	3.37	-3.40
Pu C	0.14916	0.67458	0.08239	2.81	-1.93	-3.37
Pu C	0.83144	0.33361	0.17295	3.06	-1.74	-1.42
Pu C	0.50000	1.00000	0.26522	3.08	-1.78	-1.33
Pu C	0.16856	0.66639	0.35750	3.06	-1.74	-1.42
Pu C	0.85084	0.32542	0.44805	2.81	-1.93	-3.37
Pu D	0.16699	0.16448	0.08052	-0.29	-3.64	1.10
Pu D	0.83437	0.83432	0.17524	0.01	0.00	3.80
Pu D	0.50000	0.50000	0.26522	-0.01	-0.01	3.80
Pu D	0.16563	0.16568	0.35521	0.01	0.00	3.80
Pu D	0.83301	0.83552	0.44993	-0.29	-3.64	1.10
O	0.01024	0.01023	0.05695			
O	0.52114	0.51717	0.02269			
O	0.49674	0.98681	0.05680			
O	0.98668	0.49659	0.05689			
O	0.35130	0.81118	0.10393			
O	0.81000	0.35166	0.10407			
O	0.83216	0.83194	0.10677			
O	0.33595	0.33560	0.10325			
O	0.67007	0.67021	0.15148			
O	0.16409	0.16396	0.14745			
O	0.16319	0.66887	0.15162			
O	0.66891	0.16294	0.15167			
O	0.00203	0.49568	0.19641			
O	0.49566	0.00200	0.19642			
O	0.50011	0.50011	0.19577			
O	0.00198	0.00191	0.19674			
O	0.33374	0.33375	0.24201			
O	0.83329	0.83330	0.24272			
O	0.83237	0.33388	0.24221			
O	0.33388	0.83237	0.24220			
O	0.66612	0.16763	0.28825			
O	0.16763	0.66612	0.28824			
O	0.16671	0.16670	0.28773			
O	0.66626	0.66625	0.28843			

O	0.99802	0.99809	0.33370
O	0.49989	0.49989	0.33468
O	0.50434	0.99800	0.33403
O	0.99797	0.50432	0.33404
O	0.33109	0.83706	0.37878
O	0.83681	0.33113	0.37883
O	0.83591	0.83604	0.38299
O	0.32993	0.32979	0.37897
O	0.66405	0.66440	0.42719
O	0.16784	0.16806	0.42367
O	0.19000	0.64834	0.42637
O	0.64870	0.18882	0.42651
O	0.01332	0.50341	0.47356
O	0.50326	0.01319	0.47365
O	0.47886	0.48283	0.50776
O	0.98976	0.98977	0.47350
O	0.44104	0.45165	0.57537
O	0.55896	0.54835	0.95507
C	0.45932	0.46670	0.54180
C	0.54068	0.53330	0.98864