Supporting Information

Lanthanide-uranyl Phosphonates Constructed from Diethyl ((phenylsulfonyl)methyl)phosphonate

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Materials and Instruments. The ligand precursor, diethyl ((phenylsulfonyl)methyl)phosphonate (Et_2L), was purchased from Aladin Chemicals. Other chemicals were purchased from commercial sources and used directly. Elemental analyses were performed on a Vario EL III elemental analyzer. Powder X-ray diffraction patterns (Figure S1-S5) were measured on a Bruker D8 Advance diffractometer using CuKα radiation. Compound **4** was soaked in in different solutions of varying pH=2-12 for 6 hours and recovered for the XRD measurements. IR spectra (Figure S6) were recorded in the range of 4000-400 cm^{-1} on a Thermo Fisher Nicolet iS-10 FTIR Spectrometer with KBr pellets. Thermogravimetric analyses (TGA) were carried out on a NETZSCH STA 449C unit at a heating rate of 10 °C/min under nitrogen atmosphere. Fluorescent analyses of compounds **1**, **2**, **3** and **5** were performed on a HITACHI F-7000 FL spectrophotometer. The photophysical properties of compound **4** were measured on FLS-1000 spectrophotometer. The UV-Vis spectra of compounds **1**-**5** were recorded on HITACHI U-3900 spectrophotometer.

Single-Crystal Structure Determination. The diffraction intensity data sets of compounds **1**-**5** were collected on a Bruker SMART APEX II CCD diffractometer (Mo Kα radiation, λ= 0.71073 Å) at room temperature. SAINT was used for integration of intensity of reflections and scaling[1]. Absorption corrections were carried out with the program SADABS[2]. Crystal structures were solved by direct methods using SHELXS[3]. Subsequent difference Fourier analyses and least squares refinement with SHELXL-2013[4] allowed for the location of the atom positions. All non-hydrogen atoms were refined with anisotropic thermal parameters except O1, O2, O6, O7, C9, C10, C11, C15, C16, C17, C18 and C19 in compound **2**, C17, C18, C19 and C20 in compounds **4** and **5**. The crystal structure of compound **3** is a twin. It was refined on the data in HKLF5 format with SHELXL-2013 using TWIN and BASF

commands. The high residue peaks around Sm1 are meaningless. The crystallographic details are summarized in Table S1. The data have been deposited in the Cambridge Crystallographic Data Centre (CCDC), deposition numbers CCDC 2092516-2092520 for compounds **1-5**. These data can be obtained free of charge via www.ccdc.cam.ac.uk/data_request/cif, or by emailing data_request@ccdc.cam.ac.uk, or by contacting The Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033.

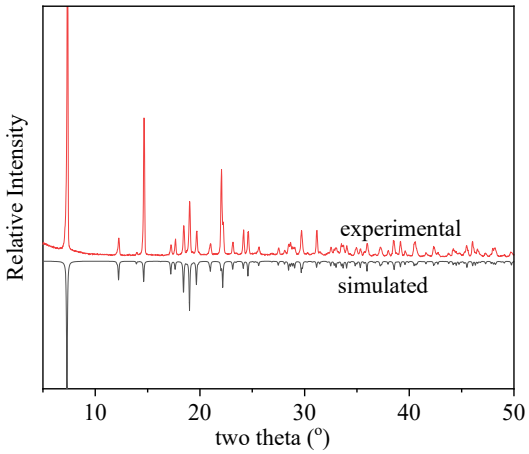


Figure S1 Simulated and experimental XRD patterns of compound **1**.

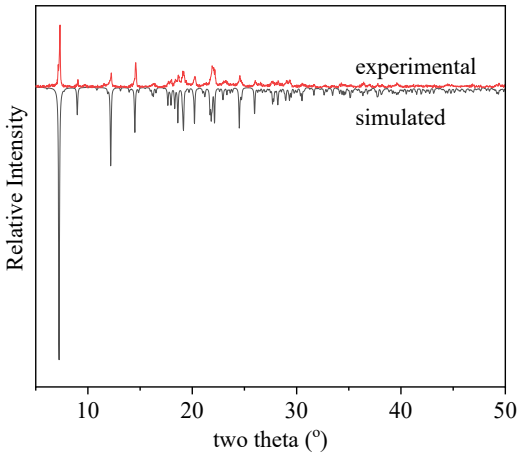


Figure S2 Simulated and experimental XRD patterns of compound **2**.

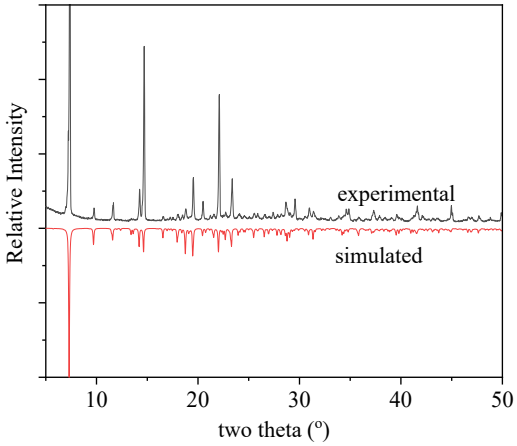


Figure S3 Simulated and experimental XRD patterns of compound **3**.

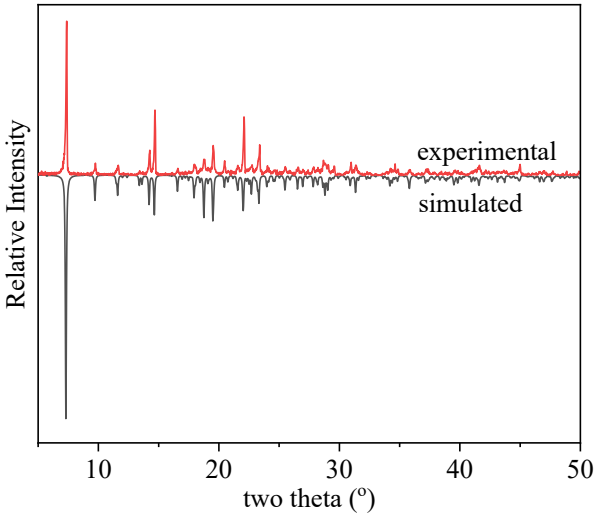


Figure S4 Simulated and experimental XRD patterns of compound **4**.

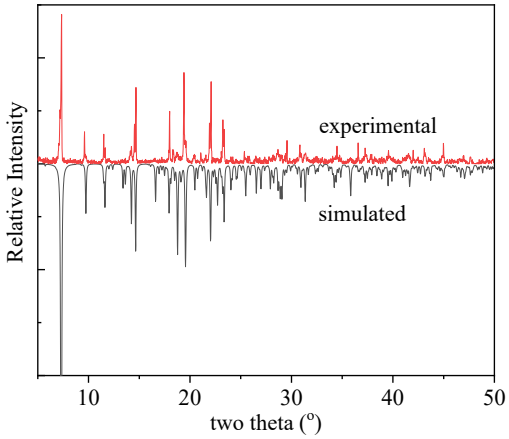


Figure S5 Simulated and experimental XRD patterns of compound **5**.

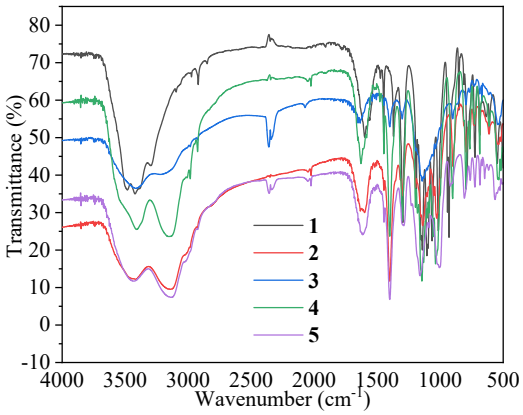
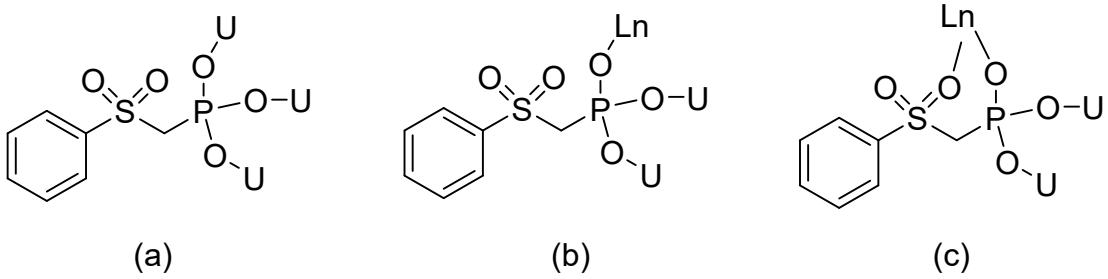


Figure S6 IR spectra of compounds **1**-**5**.



Scheme S1. Coordination modes of the ligand in compounds **1**-**5**.

Table S1. Selected bonds (Å) of compounds **1**-**5**.

Compound **1**			
U(1)-O(7)	1.756(4)	U(1)-O(2)#2	2.356(3)
U(1)-O(6)	1.759(4)	U(1)-O(1W)	2.471(4)
U(1)-O(1)#1	2.320(3)	U(1)-O(2W)	2.493(4)
U(1)-O(3)	2.344(3)		
Compound **2**			
U(1)-O(18)	1.779(10)	U(2)-O(10)#2	2.269(10)
U(1)-O(18)#1	1.779(11)	U(2)-O(9)	2.302(10)
U(1)-O(3)#1	2.262(12)	La(1)-O(8)#2	2.381(10)
U(1)-O(3)	2.262(12)	La(1)-O(4)#3	2.404(11)
U(1)-O(14)#1	2.275(10)	La(1)-O(15)	2.410(11)
U(1)-O(14)	2.275(10)	La(1)-O(4W)	2.515(13)
U(2)-O(17)	1.786(11)	La(1)-O(1W)	2.557(12)
U(2)-O(16)	1.788(11)	La(1)-O(3W)	2.570(12)
U(2)-O(13)	2.243(11)	La(1)-O(12)	2.667(12)
U(2)-O(5)	2.258(10)	La(1)-O(2W)	2.693(12)

Compound **3**			
U(1)-O(16)#1	1.789(7)	U(2)-O(3)#4	2.258(7)
U(1)-O(16)	1.789(7)	U(2)-O(4)	2.296(7)
U(1)-O(9)#1	2.272(7)	Sm(1)-O(10)	2.296(7)
U(1)-O(9)	2.272(7)	Sm(1)-O(5)	2.309(7)
U(1)-O(14)#2	2.285(7)	Sm(1)-O(13)#4	2.341(7)
U(1)-O(14)#3	2.285(7)	Sm(1)-O(2W)	2.420(8)
U(2)-O(18)	1.780(7)	Sm(1)-O(1W)	2.470(8)
U(2)-O(17)	1.788(7)	Sm(1)-O(3W)	2.479(8)
U(2)-O(15)	2.235(8)	Sm(1)-O(12)#4	2.563(8)
U(2)-O(8)#3	2.250(7)	Sm(1)-O(4W)	2.576(7)
Compound **4**			
U(1)-O(16)	1.777(5)	U(2)-O(3)#4	2.259(5)
U(1)-O(16)#1	1.777(5)	U(2)-O(4)	2.297(5)
U(1)-O(9)	2.270(5)	Eu(1)-O(5)	2.290(5)
U(1)-O(9)#1	2.270(5)	Eu(1)-O(10)	2.297(5)
U(1)-O(14)#2	2.286(5)	Eu(1)-O(13)#4	2.335(5)
U(1)-O(14)#3	2.286(5)	Eu(1)-O(2W)	2.394(5)
U(2)-O(18)	1.781(5)	Eu(1)-O(3W)	2.461(5)
U(2)-O(17)	1.786(5)	Eu(1)-O(1W)	2.465(5)
U(2)-O(15)	2.231(5)	Eu(1)-O(12)#4	2.558(5)
U(2)-O(8)#3	2.255(5)	Eu(1)-O(4W)	2.567(5)
Compound **5**			
U(1)-O(16)#1	1.785(6)	U(2)-O(8)#3	2.256(6)
U(1)-O(16)	1.785(6)	U(2)-O(4)	2.295(5)
U(1)-O(9)#1	2.276(5)	Tb(1)-O(5)	2.261(6)
U(1)-O(9)	2.276(5)	Tb(1)-O(10)	2.267(6)
U(1)-O(14)#2	2.279(5)	Tb(1)-O(13)#4	2.304(5)
U(1)-O(14)#3	2.279(5)	Tb(1)-O(2W)	2.366(6)
U(2)-O(17)	1.787(6)	Tb(1)-O(3W)	2.433(6)
U(2)-O(18)	1.788(6)	Tb(1)-O(1W)	2.435(6)
U(2)-O(15)	2.228(6)	Tb(1)-O(12)#4	2.548(6)
U(2)-O(3)#4	2.254(5)	Tb(1)-O(4W)	2.555(6)

Symmetry transformations used to generate equivalent atoms: for **1**: #1 -x+2, -y+1, -z; #2 x, -y+1/2, z+1/2. For **2**: #1 -x, -y, -z+1; #2 -x, -y, -z+2; #3 x+1, y, z. For **3**-**5**: #1 -x+3, -y+1, -z+3; #2 x+1, y, z+1; #3 -x+2, -y+1, -z+2; #4 -x+2, -y+1, -z+1.

Table S2. Selected angels (°) in compounds **1**-**5**.

Compound **1**			
O(7)-U(1)-O(6)	178.79(16)	O(6)-U(1)-O(1W)	93.04(16)
O(7)-U(1)-O(1)#1	91.13(15)	O(1)#1-U(1)-O(1W)	72.24(12)
O(6)-U(1)-O(1)#1	88.74(16)	O(3)-U(1)-O(1W)	151.65(13)

O(7)-U(1)-O(3)	87.93(15)	O(2)#2-U(1)-O(1W)	71.58(12)
O(6)-U(1)-O(3)	90.86(15)	O(7)-U(1)-O(2W)	89.95(16)
O(1)#1-U(1)-O(3)	79.80(12)	O(6)-U(1)-O(2W)	89.56(16)
O(7)-U(1)-O(2)#2	90.93(15)	O(1)#1-U(1)-O(2W)	150.33(12)
O(6)-U(1)-O(2)#2	89.88(15)	O(3)-U(1)-O(2W)	70.61(12)
O(1)#1-U(1)-O(2)#2	143.66(12)	O(2)#2-U(1)-O(2W)	65.94(12)
O(3)-U(1)-O(2)#2	136.54(12)	O(1W)-U(1)-O(2W)	137.43(12)
O(7)-U(1)-O(1W)	88.07(15)		
Compound **2**			
O(18)-U(1)-O(18)#1	180.0(10)	O(10)#2-U(2)-O(9)	92.6(4)
O(18)-U(1)-O(3)#1	89.9(4)	O(8)#2-La(1)-O(4)#3	144.6(4)
O(18)#1-U(1)-O(3)#1	90.1(4)	O(8)#2-La(1)-O(15)	102.0(4)
O(18)-U(1)-O(3)	90.1(4)	O(4)#3-La(1)-O(15)	96.5(4)
O(18)#1-U(1)-O(3)	89.9(4)	O(8)#2-La(1)-O(4W)	83.6(4)
O(3)#1-U(1)-O(3)	180.0(5)	O(4)#3-La(1)-O(4W)	100.9(4)
O(18)-U(1)-O(14)#1	90.3(4)	O(15)-La(1)-O(4W)	140.5(4)
O(18)#1-U(1)-O(14)#1	89.7(4)	O(8)#2-La(1)-O(1W)	77.2(4)
O(3)#1-U(1)-O(14)#1	93.5(4)	O(4)#3-La(1)-O(1W)	78.7(4)
O(3)-U(1)-O(14)#1	86.5(4)	O(15)-La(1)-O(1W)	75.3(4)
O(18)-U(1)-O(14)	89.7(4)	O(4W)-La(1)-O(1W)	142.8(5)
O(18)#1-U(1)-O(14)	90.3(4)	O(8)#2-La(1)-O(3W)	140.0(4)
O(3)#1-U(1)-O(14)	86.5(4)	O(4)#3-La(1)-O(3W)	73.9(4)
O(3)-U(1)-O(14)	93.5(4)	O(15)-La(1)-O(3W)	75.2(4)
O(14)#1-U(1)-O(14)	180.0(4)	O(4W)-La(1)-O(3W)	76.0(5)
O(17)-U(2)-O(16)	179.1(6)	O(1W)-La(1)-O(3W)	136.7(4)
O(17)-U(2)-O(13)	91.4(5)	O(8)#2-La(1)-O(12)	72.9(4)
O(16)-U(2)-O(13)	89.4(5)	O(4)#3-La(1)-O(12)	142.3(4)
O(17)-U(2)-O(5)	90.5(5)	O(15)-La(1)-O(12)	71.2(4)
O(16)-U(2)-O(5)	89.2(5)	O(4W)-La(1)-O(12)	73.4(4)
O(13)-U(2)-O(5)	87.3(4)	O(1W)-La(1)-O(12)	128.5(4)
O(17)-U(2)-O(10)#2	89.1(5)	O(3W)-La(1)-O(12)	68.5(4)
O(16)-U(2)-O(10)#2	91.2(4)	O(8)#2-La(1)-O(2W)	73.1(4)
O(13)-U(2)-O(10)#2	90.3(4)	O(4)#3-La(1)-O(2W)	74.1(5)
O(5)-U(2)-O(10)#2	177.6(4)	O(15)-La(1)-O(2W)	143.9(4)
O(17)-U(2)-O(9)	87.7(5)	O(4W)-La(1)-O(2W)	75.4(5)
O(16)-U(2)-O(9)	91.5(5)	O(1W)-La(1)-O(2W)	68.7(5)
O(13)-U(2)-O(9)	176.8(4)	O(3W)-La(1)-O(2W)	131.5(4)
O(5)-U(2)-O(9)	89.7(4)	O(12)-La(1)-O(2W)	135.7(5)
Compound **3**			
O(16)#1-U(1)-O(16)	180.0	O(3)#4-U(2)-O(4)	94.1(3)
O(16)#1-U(1)-O(9)#1	89.3(3)	O(10)-Sm(1)-O(5)	143.7(3)
O(16)-U(1)-O(9)#1	90.7(3)	O(10)-Sm(1)-O(13)#4	97.9(3)
O(16)#1-U(1)-O(9)	90.7(3)	O(5)-Sm(1)-O(13)#4	100.3(3)

O(16)-U(1)-O(9)	89.3(3)	O(10)-Sm(1)-O(2W)	100.4(3)
O(9)#1-U(1)-O(9)	180.0	O(5)-Sm(1)-O(2W)	85.4(3)
O(16)#1-U(1)-O(14)#2	90.8(3)	O(13)#4-Sm(1)-O(2W)	139.6(3)
O(16)-U(1)-O(14)#2	89.2(3)	O(10)-Sm(1)-O(1W)	74.3(3)
O(9)#1-U(1)-O(14)#2	92.9(3)	O(5)-Sm(1)-O(1W)	140.9(3)
O(9)-U(1)-O(14)#2	87.1(3)	O(13)#4-Sm(1)-O(1W)	75.1(3)
O(16)#1-U(1)-O(14)#3	89.2(3)	O(2W)-Sm(1)-O(1W)	75.7(3)
O(16)-U(1)-O(14)#3	90.8(3)	O(10)-Sm(1)-O(3W)	77.5(3)
O(9)#1-U(1)-O(14)#3	87.1(3)	O(5)-Sm(1)-O(3W)	76.7(3)
O(9)-U(1)-O(14)#3	92.9(3)	O(13)#4-Sm(1)-O(3W)	76.3(3)
O(14)#2-U(1)-O(14)#3	180.0	O(2W)-Sm(1)-O(3W)	142.9(3)
O(18)-U(2)-O(17)	179.1(4)	O(1W)-Sm(1)-O(3W)	136.1(3)
O(18)-U(2)-O(15)	89.6(4)	O(10)-Sm(1)-O(12)#4	143.5(3)
O(17)-U(2)-O(15)	90.8(4)	O(5)-Sm(1)-O(12)#4	72.4(3)
O(18)-U(2)-O(8)#3	91.0(3)	O(13)#4-Sm(1)-O(12)#4	72.5(3)
O(17)-U(2)-O(8)#3	89.8(3)	O(2W)-Sm(1)-O(12)#4	71.3(3)
O(15)-U(2)-O(8)#3	86.2(3)	O(1W)-Sm(1)-O(12)#4	69.3(3)
O(18)-U(2)-O(3)#4	88.9(3)	O(3W)-Sm(1)-O(12)#4	130.6(3)
O(17)-U(2)-O(3)#4	90.3(3)	O(10)-Sm(1)-O(4W)	74.7(3)
O(15)-U(2)-O(3)#4	91.1(3)	O(5)-Sm(1)-O(4W)	72.6(3)
O(8)#3-U(2)-O(3)#4	177.4(3)	O(13)#4-Sm(1)-O(4W)	145.7(3)
O(18)-U(2)-O(4)	87.2(3)	O(2W)-Sm(1)-O(4W)	74.3(3)
O(17)-U(2)-O(4)	92.6(3)	O(1W)-Sm(1)-O(4W)	131.4(2)
O(15)-U(2)-O(4)	173.8(3)	O(3W)-Sm(1)-O(4W)	69.5(3)
O(8)#3-U(2)-O(4)	88.5(3)	O(12)#4-Sm(1)-O(4W)	132.1(3)
Compound **4**			
O(16)-U(1)-O(16)#1	180.0(2)	O(3)#4-U(2)-O(4)	94.17(17)
O(16)-U(1)-O(9)	89.7(2)	O(5)-Eu(1)-O(10)	143.87(19)
O(16)#1-U(1)-O(9)	90.3(2)	O(5)-Eu(1)-O(13)#4	100.00(18)
O(16)-U(1)-O(9)#1	90.3(2)	O(10)-Eu(1)-O(13)#4	98.26(18)
O(16)#1-U(1)-O(9)#1	89.7(2)	O(5)-Eu(1)-O(2W)	85.57(19)
O(9)-U(1)-O(9)#1	180.0	O(10)-Eu(1)-O(2W)	99.99(18)
O(16)-U(1)-O(14)#2	89.2(2)	O(13)#4-Eu(1)-O(2W)	139.81(18)
O(16)#1-U(1)-O(14)#2	90.8(2)	O(5)-Eu(1)-O(3W)	77.45(18)
O(9)-U(1)-O(14)#2	87.23(18)	O(10)-Eu(1)-O(3W)	77.15(18)
O(9)#1-U(1)-O(14)#2	92.77(18)	O(13)#4-Eu(1)-O(3W)	75.92(18)
O(16)-U(1)-O(14)#3	90.8(2)	O(2W)-Eu(1)-O(3W)	143.2(2)
O(16)#1-U(1)-O(14)#3	89.2(2)	O(5)-Eu(1)-O(1W)	140.83(18)
O(9)-U(1)-O(14)#3	92.77(18)	O(10)-Eu(1)-O(1W)	74.20(19)
O(9)#1-U(1)-O(14)#3	87.23(18)	O(13)#4-Eu(1)-O(1W)	75.24(19)
O(14)#2-U(1)-O(14)#3	180.0	O(2W)-Eu(1)-O(1W)	75.79(19)
O(18)-U(2)-O(17)	179.1(2)	O(3W)-Eu(1)-O(1W)	135.41(19)
O(18)-U(2)-O(15)	89.4(2)	O(5)-Eu(1)-O(12)#4	72.17(18)

O(17)-U(2)-O(15)	90.9(2)	O(10)-Eu(1)-O(12)#4	143.50(19)
O(18)-U(2)-O(8)#3	90.9(2)	O(13)#4-Eu(1)-O(12)#4	72.86(17)
O(17)-U(2)-O(8)#3	90.0(2)	O(2W)-Eu(1)-O(12)#4	71.17(17)
O(15)-U(2)-O(8)#3	85.85(19)	O(3W)-Eu(1)-O(12)#4	131.14(18)
O(18)-U(2)-O(3)#4	88.9(2)	O(1W)-Eu(1)-O(12)#4	69.29(18)
O(17)-U(2)-O(3)#4	90.3(2)	O(5)-Eu(1)-O(4W)	72.55(17)
O(15)-U(2)-O(3)#4	91.00(18)	O(10)-Eu(1)-O(4W)	74.80(19)
O(8)#3-U(2)-O(3)#4	176.85(18)	O(13)#4-Eu(1)-O(4W)	145.74(18)
O(18)-U(2)-O(4)	87.4(2)	O(2W)-Eu(1)-O(4W)	74.02(18)
O(17)-U(2)-O(4)	92.3(2)	O(3W)-Eu(1)-O(4W)	69.82(18)
O(15)-U(2)-O(4)	173.88(18)	O(1W)-Eu(1)-O(4W)	131.54(17)
O(8)#3-U(2)-O(4)	88.96(18)	O(12)#4-Eu(1)-O(4W)	131.56(18)
Compound **5**			
O(16)#1-U(1)-O(16)	180.0	O(8)#3-U(2)-O(4)	89.1(2)
O(16)#1-U(1)-O(9)#1	89.7(2)	O(5)-Tb(1)-O(10)	143.8(2)
O(16)-U(1)-O(9)#1	90.3(2)	O(5)-Tb(1)-O(13)#4	100.0(2)
O(16)#1-U(1)-O(9)	90.3(2)	O(10)-Tb(1)-O(13)#4	98.6(2)
O(16)-U(1)-O(9)	89.7(2)	O(5)-Tb(1)-O(2W)	85.5(2)
O(9)#1-U(1)-O(9)	180.0	O(10)-Tb(1)-O(2W)	99.5(2)
O(16)#1-U(1)-O(14)#2	90.8(2)	O(13)#4-Tb(1)-O(2W)	140.1(2)
O(16)-U(1)-O(14)#2	89.2(2)	O(5)-Tb(1)-O(3W)	78.0(2)
O(9)#1-U(1)-O(14)#2	93.0(2)	O(10)-Tb(1)-O(3W)	76.7(2)
O(9)-U(1)-O(14)#2	87.0(2)	O(13)#4-Tb(1)-O(3W)	76.3(2)
O(16)#1-U(1)-O(14)#3	89.2(2)	O(2W)-Tb(1)-O(3W)	142.6(2)
O(16)-U(1)-O(14)#3	90.8(2)	O(5)-Tb(1)-O(1W)	140.8(2)
O(9)#1-U(1)-O(14)#3	87.0(2)	O(10)-Tb(1)-O(1W)	74.1(2)
O(9)-U(1)-O(14)#3	93.0(2)	O(13)#4-Tb(1)-O(1W)	75.3(2)
O(14)#2-U(1)-O(14)#3	180.0	O(2W)-Tb(1)-O(1W)	76.0(2)
O(17)-U(2)-O(18)	179.2(3)	O(3W)-Tb(1)-O(1W)	135.2(2)
O(17)-U(2)-O(15)	90.8(3)	O(5)-Tb(1)-O(12)#4	72.1(2)
O(18)-U(2)-O(15)	89.4(3)	O(10)-Tb(1)-O(12)#4	143.5(2)
O(17)-U(2)-O(3)#4	90.4(2)	O(13)#4-Tb(1)-O(12)#4	73.03(19)
O(18)-U(2)-O(3)#4	88.8(2)	O(2W)-Tb(1)-O(12)#4	71.3(2)
O(15)-U(2)-O(3)#4	90.9(2)	O(3W)-Tb(1)-O(12)#4	132.0(2)
O(17)-U(2)-O(8)#3	89.9(2)	O(1W)-Tb(1)-O(12)#4	69.4(2)
O(18)-U(2)-O(8)#3	90.9(2)	O(5)-Tb(1)-O(4W)	72.7(2)
O(15)-U(2)-O(8)#3	85.8(2)	O(10)-Tb(1)-O(4W)	74.6(2)
O(3)#4-U(2)-O(8)#3	176.7(2)	O(13)#4-Tb(1)-O(4W)	146.3(2)
O(17)-U(2)-O(4)	92.5(2)	O(2W)-Tb(1)-O(4W)	73.1(2)
O(18)-U(2)-O(4)	87.4(2)	O(3W)-Tb(1)-O(4W)	70.0(2)
O(15)-U(2)-O(4)	174.0(2)	O(1W)-Tb(1)-O(4W)	131.0(2)
O(3)#4-U(2)-O(4)	94.1(2)	O(12)#4-Tb(1)-O(4W)	131.1(2)

Symmetry transformations used to generate equivalent atoms: Symmetry

transformations used to generate equivalent atoms:

For **1**: #1 -x, -y, -z+1; #2 -x, -y, -z+2; #3 x+1, y, z; #4 x-1, y, z.

For **2**: #1 -x, -y, -z+1; #2 -x, -y, -z+2; #3 x+1, y, z; #4 x-1, y, z.

For **3**-**5**: #1 -x+3, -y+1, -z+3; #2 x+1, y, z+1; #3 -x+2, -y+1, -z+2; #4 -x+2, -y+1, -z+1; #5 x-1, y, z-1.

Table S3. Analysis of Short Ring-Interactions with Cg-Cg Distances < 6.0 Angstrom and Beta < 60.0Deg.
- Cg(I) = Plane number I (= ring number in () above)
- Alpha = Dihedral Angle between Planes I and J (Deg)
- Beta = Angle Cg(I)-->Cg(J) or Cg(I)-->Me vector and normal to plane I (Deg)
- Gamma = Angle Cg(I)-->Cg(J) vector and normal to plane J (Deg)
- Cg-Cg = Distance between ring Centroids (Ang.)
- CgI_Perp = Perpendicular distance of Cg(I) on ring J (Ang.)
- CgJ_Perp = Perpendicular distance of Cg(J) on ring I (Ang.)
- Slippage = Distance between Cg(I) and Perpendicular Projection of Cg(J) on Ring I (Ang).

Cg(I)···Cg(J)	Cg···Cg	Alpha	Beta	Gamma	CgI_Perp	CgJ_Perp	Slippage
Compound **1**							
Cg(1)···Cg(1)#1	5.1955	0	14.4	14.4	-5.0331	-5.0331	1.289
Compound **2**							
Cg(1)···Cg(2)	5.4817(3)	8	47.8	56.2	3.0467	-3.6838	4.059
Cg(1)···Cg(2)#1	5.6139(4)	8	28.6	21.4	-5.2281	-4.9293	2.686
Cg(1)···Cg(2)#2	5.3763(3)	8	39.8	35.9	4.3545	4.1280	3.444
Cg(1)···Cg(3)#3	5.2145(3)	24	58.2	36.0	4.2210	2.7513	
Cg(2)···Cg(1)	5.4817(3)	8	56.2	47.8	-3.6838	3.0467	4.557
Cg(2)···Cg(1)#1	5.6139(4)	8	21.4	28.6	-4.9293	-5.2281	2.045
Cg(2)···Cg(1)#2	5.3763(3)	8	35.9	39.8	4.1280	4.3545	3.153
Cg(3)···Cg(1)#3	5.2145(3)	24	36.0	58.2	2.7513	4.2210	
Cg(3)···Cg(2)#4	5.2691(3)	32	31.4	63.4	-2.3580	-4.4976	
Cg(3)···Cg(3)#5	4.8984(3)	0	47.9	47.9	-3.2820	-3.2820	3.636
Cg(3)···Cg(3)#6	5.2765(3)	0	51.5	51.5	3.2823	3.2823	4.131
Compound **3**							
Cg(1)···Cg(1)#1	5.4163	16	41.8	31.9	4.5997	-4.0402	
Cg(1)···Cg(1) #2	5.4163	16	31.9	41.8	-4.0402	4.5997	
Cg(1)···Cg(2) #3	5.3388	25	40.9	64.9	-2.2664	-4.0378	
Cg(1)···Cg(3)	5.7577	20	53.6	33.9	4.7778	3.4162	
Cg(2)···Cg(2) #4	5.5861	6	23.1	29.0	4.8856	-5.1377	
Cg(2)···Cg(2) #5	5.5861	6	29.0	23.1	-5.1377	4.8856	
Cg(3)···Cg(1)	5.7577	20	33.9	53.6	3.4162	4.7778	
Cg(3)···Cg(2) #6	4.9652	44	31.1	70.5	1.6544	-4.2508	
Cg(3)···Cg(3) #1	5.0630	14	51.4	49.8	-3.2676	3.1605	

Cg(3)···Cg(3) #2	5.0630	14	49.8	51.4	3.1605	-3.2676	
Compound **4**							
Cg(1)···Cg(1)#1	5.4079(2)	16	41.4	31.6	4.6074	-4.0593	3.573
Cg(1)···Cg(1) #2	5.4079(2)	16	31.6	41.4	-4.0593	4.6074	2.831
Cg(1)···Cg(2) #3	5.3255(2)	25	41.1	64.9	-2.2560	-4.0152	
Cg(1)···Cg(3)	5.7406(2)	20	53.7	33.4	4.7902	3.4013	
Cg(2)···Cg(2) #4	5.5780(2)	6	23.0	28.9	4.8852	-5.1351	2.178
Cg(2)···Cg(2) #5	5.5780(2)	6	28.9	23.0	-5.1351	4.8852	2.692
Cg(3)···Cg(1)	5.7406(2)	20	33.4	53.7	3.4013	4.7902	
Cg(3)···Cg(2) #6	4.9867(2)	45	30.9	70.8	1.6387	-4.2791	
Cg(3)···Cg(3) #1	5.0601(2)	14	51.5	50.0	-3.2528	3.1473	3.962
Cg(3)···Cg(3) #2	5.0601(2)	14	50.0	51.5	3.1473	-3.2528	3.876
Compound **5**							
Cg(1)···Cg(1)#1	5.3897(2)	16	40.5	30.4	4.6510	-4.0973	3.502
Cg(1)···Cg(1) #2	5.3897(2)	16	30.4	40.5	-4.0973	4.6510	2.724
Cg(1)···Cg(2) #3	5.3188(2)	23	42.1	64.8	-2.2673	-3.9447	
Cg(1)···Cg(3)	5.7249(2)	21	54.6	33.6	4.7687	3.3203	
Cg(2)···Cg(2) #4	5.5644(2)	6	23.0	28.8	4.8749	-5.1223	2.174
Cg(2)···Cg(2) #5	5.5644(2)	6	28.8	23.0	-5.1223	4.8749	2.683
Cg(3)···Cg(1)	5.7249(2)	21	33.6	54.6	3.3203	4.7687	
Cg(3)···Cg(2) #6	4.9898(2)	44	31.5	70.9	1.6312	-4.2543	
Cg(3)···Cg(3) #1	5.0502(2)	14	51.2	49.5	-3.2827	3.1674	3.933
Cg(3)···Cg(3) #2	5.0502(2)	14	49.5	51.2	3.1674	-3.2827	3.838

For **2**: #1: = -X, 1-Y, -Z; #2: 1-X, 1-Y, -Z; #3: -X, -Y, 1-Z; #4: -X, -Y, -Z; #5: -1-X, -1-Y, 1-Z; #6: -X, -1-Y, 1-Z.

For **3-5**: #1: X, 1/2-Y, -1/2+Z; #2: X, 1/2-Y, 1/2+Z; #3: 2-X, -Y, 2-Z; #4: X, -1/2-Y, -1/2+Z; #5: X, -1/2-Y, 1/2+Z; #6: 1-X, -Y, 2-Z

For **1**: Cg (1): C(1)-C(2)-C(3)-C(4)-C(5)-C(6)→

For **2**:

Cg (1): C(1)-C(2)-C(3)-C(4)-C(5)-C(6)→

Cg (2): C(8)-C(9)-C(10)-C(11)-C(12)-C(13)→

Cg (3): C(15)-C(16)-C(17)-C(18)-C(19)-C(20)→

For **3-5**:

Cg (1): C(2)-C(3)-C(4)-C(5)-C(6)-C(7)→

Cg (2): C(9)-C(10)-C(11)-C(12)-C(13)-C(14)→

Cg (3): C(16)-C(17)-C(18)-C(19)-C(20)-C(21)→

Table S4. Hydrogen bonds in compounds **1**-**5**.

Donor-H···Acceptor	D-H	H···A	D···A	D-H···A
Compound **1**				
O(1W)-H(1W)···O(2)#1	0.85	2.54	2.8245	100
O(1W)-H(1W)···O(3)#1	0.85	1.93	2.7449	161
O(1W)-H(2W)···O(2)	0.85	2.03	2.7994	149

O(2W)-H(4W)···O(5)#2	0.85	2.08	2.8519	150
C(1)-H(1A)···O(4)	0.93	2.56	2.9140	103
C(2)-H(2A)···O(7)#3	0.93	2.59	3.2596	129
Compound **2**				
O1W-H1W···O2	0.85	2.14	2.7068(2)	124
O1W-H2W···O16#1	0.85	2.08	2.9246(2)	170
O2W-H4W···O1W	0.85	2.49	2.9646(2)	116
O3W-H5W···O5W#2	0.85	1.91	2.7424(2)	167
O3W-H6W···O18#3	0.85	2.42	2.9657(2)	123
O4W-H7W···O17	0.85	2.33	2.7338(2)	109
O5W-H10W···O14#1	0.85	2.50	3.3359(2)	167
O5W-H10W···O15#1	0.85	2.40	2.9027(2)	119
C1-H1A···O2	0.93	2.55	2.8952(2)	103
C4-H4A···O11#4	0.93	2.52	3.2816(2)	139
C7-H7B···O6	0.97	2.21	3.1752(2)	171
C11-H11A···O16#5	0.93	2.51	3.2790(2)	140
C12-H12A···O6	0.93	2.52	2.8945(2)	104
C14-H14B···O11#6	0.97	2.37	3.2388(2)	149
C19-H19A···O11	0.93	2.41	2.8091(2)	106
C21-H21A···O1#3	0.97	2.46	3.2483(2)	139
Compound **3**				
O(1W)-H(1W)···O(5W)	0.85	1.91	2.7144	156
O(1W)-H(2W)···O(16)#1	0.85	2.11	2.9604	174
O(2W)-H(3W)···O(1)	0.85	1.96	2.6818	142
O(2W)-H(4W)···O(18)#1	0.85	1.99	2.6900	139
O(3W)-H(5W)···O(7)#1	0.85	2.37	2.7506	107
O(3W)-H(6W)···O(17)	0.85	2.22	2.9338	141
O(4W)-H(7W)···O(5)	0.85	2.38	2.9003	120
O(4W)-H(8W)···O(2)#2	0.85	2.49	3.2960	159
O(5W)-H(9W)···O(13)#3	0.85	2.14	2.8669	144
O(5W)-H(10W)···O(6)#4	0.85	2.37	3.0019	132
C(1)-H(1A)···O(11)	0.97	2.36	3.2347	150
C(7)-H(7A)···O(1)	0.93	2.53	2.8904	103
C(8)-H(8B)···O(2)#5	0.97	2.33	3.2609	160
C(10)-H(10A)···O(6)	0.93	2.55	2.9116	104
C(13)-H(13A)···O(17)#6	0.93	2.57	3.2848	135
C(15)-H(15B)···O(6)#4	0.97	2.41	3.2187	141
C(19)-H(19A)···O(16)#7	0.93	2.42	3.2909	157
C(21)-H(21A)···O(11)	0.93	2.50	2.8883	105
Compound **4**				
O(1W)-H(1W)···O(10)#1	0.85	1.90	2.7057(1)	157
O(1W)-H(2W)···O(16) #1	0.85	2.13	2.9730(1)	174

O(2W)-H(3W)···O(1)	0.85	1.97	2.6867(1)	142
O(2W)-H(4W)···O(18) #1	0.85	1.99	2.6925(1)	139
O(3W)-H(5W)···O(7) #1	0.85	2.35	2.7279(1)	107
O(3W)-H(6W)···O(17)	0.85	2.25	2.9605(1)	142
O(4W)-H(7W)···O(5)	0.85	2.37	2.8827(1)	119
O(4W)-H(8W)···O(2) #2	0.85	2.50	3.3060(1)	160
O(5W)-H(9W)···O(13) #3	0.85	2.14	2.8706(1)	143
O(5W)-H(10W)···O(1W)	0.85	2.35	2.9871(1)	132
C(1)-H(1A)···O(11)	0.97	2.36	3.2391(1)	150
C(7)-H(7A)···O(1)	0.93	2.53	2.8901(1)	104
C(8)-H(8B)···O(2) #4	0.97	2.33	3.2608(1)	160
C(10)-H(10A)···O(6)	0.93	2.54	2.9150(1)	104
C(13)-H(13A)···O(17) #5	0.93	2.57	3.2968(1)	136
C(15)-H(15B)···O(6) #6	0.97	2.40	3.2134(1)	141
C(19)-H(19A)···O(16) #7	0.93	2.40	3.2927(1)	160
C(21)-H(21A)···O(11)	0.93	2.48	2.8735(1)	105
Compound **5**				
O1W-H1W···O5W	0.85	1.90	2.7018(1)	157
O1W-H2W···O16#1	0.85	2.13	2.9754(1)	175
O2W-H3W···O1	0.85	1.97	2.6910(1)	142
O2W-H3W···O12	0.85	2.58	2.8680(1)	101
O2W-H4W···O18#1	0.85	1.97	2.6744(1)	140
O3W-H5W···O7#1	0.85	2.34	2.7170(1)	107
O3W-H5W···O13	0.85	2.58	2.9292(1)	106
O3W-H6W···O17	0.85	2.26	2.9773(1)	142
O4W-H7W···O5	0.85	2.36	2.8624(1)	119
O4W-H8W···O2#2	0.85	2.49	3.3027(1)	160
O5W-H9W···O13#3	0.85	2.15	2.8766(1)	143
O5W-H10W···O6#4	0.85	2.36	2.9916(1)	132
C1-H1A···O11	0.97	2.35	3.2266(1)	149
C7-H7A···O1	0.93	2.53	2.8983(1)	104
C8-H8B···O2#5	0.97	2.35	3.2706(1)	159
C10-H10A···O6	0.93	2.53	2.9074(1)	104
C13-H13A···O17#6	0.93	2.56	3.2860(1)	136
C15-H15B···O6#4	0.97	2.40	3.2166(1)	141

C19-H19A···O16#7	0.93	2.37	3.2648(1)	160
C21-H21A···O11	0.93	2.50	2.8899(1)	106

Translation of ARU-Code to CIF and Equivalent Position Code
For **1**: #1: -x, -1/2+y, 1/2-z; #2: -x, 1-y, -z; #3: 1+x, y, z;
For **2**: #1: 1+x, y, z; #2: x, 1+y, z; #3: -x, 1-y, -z; #4: -x, -y, -z; #5: -x, -y, 1-z.
For **3-5**: #1: x, y, -1+z; #2: 2-x, -y, 1-z; #3: 1-x, -y, 1-z; #4: 2-x, -y, 2-z; #5: x, -1/2-y, 1/2+z; #6:1-x, -y, 2-z; #7: x, 1/2-y, -1/2+z.

Table S5. Analysis of Y-X···Cg(Pi-Ring) Interactions (X···Cg < 4.0 Ang. - Gamma < 30.0 Deg)

Compound **2**						
Y--X(I)···Cg(J)	X···Cg	X-Perp	Gamma	Y-X···Cg	Y···Cg	Y-X···Pi
S(1) -O(2)···Cg(2)#1	3.8446(2)	3.683	16.67	124.88(1)	4.8239(3)	33.04
S(2) -O(7)···Cg(1)#1	3.7934(2)	3.486	23.23	123.76(1)	4.7211(3)	24.59
Compound **3**						
C(10)-H(10A)→Cg(3)#1	2.98	-2.90	13.82	140	3.7443	36
S(2) -O(7)···Cg(2)#2	3.9241	-3.700	19.45	124	4.8743	45.52
Compound **4**						
S(2) -O(7) ···Cg(2)#2	3.9172(1)	-3.688	19.70	124.18(1)	4.8698(2)	45.78
Compound **5**						
S(2) -O(7)···Cg(2)#2	3.8957(1)	-3.666	19.78	124.60(1)	4.8580(2)	46.16

For **2**: #1= 1-x, 1-y, -z; For **3-5**: #1 = 1-x, -y, 2-z; #2 = x, -1/2-y, 1/2+z.

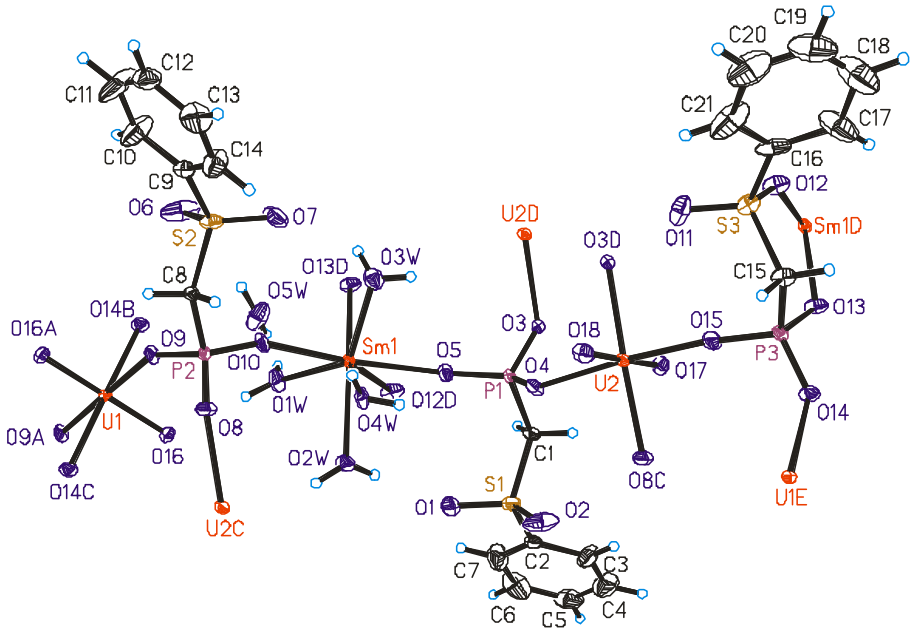


Figure S7 Depiction of the coordination environments of the phosphonate ligand, uranyl and samarium cations in **3** (thermal ellipsoids are given at 30% probability). A: 3-x, 1-y, 3-z; B: 1+x, y, 1+z; C: 2-x, 1-y, 2-z; D: 2-x, 1-y, 1-z; E: -1+x, y, -1+z.

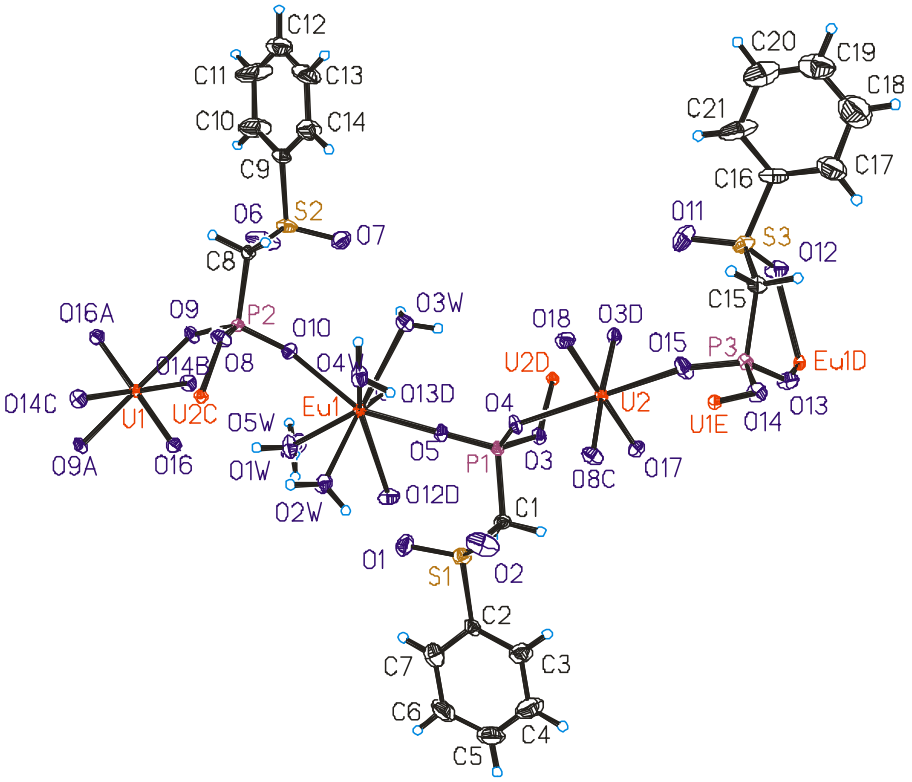


Figure S8 Depiction of the coordination environments of the phosphonate ligand, uranyl and samarium cations in **4** (thermal ellipsoids are given at 30% probability). A: 3-x, 1-y, 3-z; B: 1+x, y, 1+z; C: 2-x, 1-y, 2-z; D: 2-x, 1-y, 1-z; E: -1+x, y, -1+z.

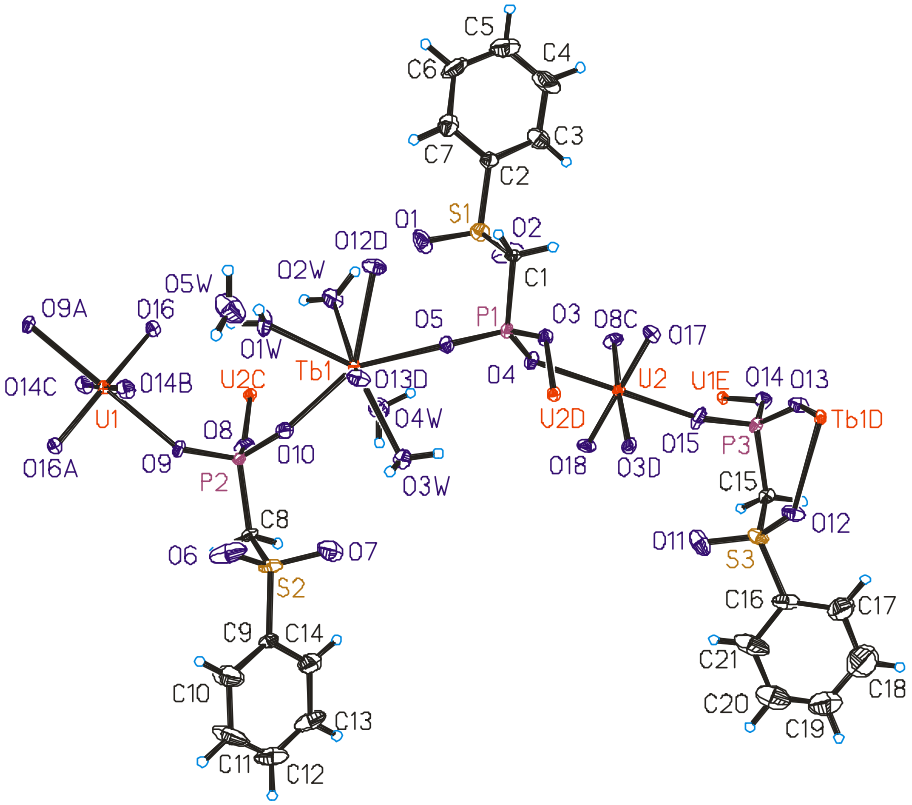
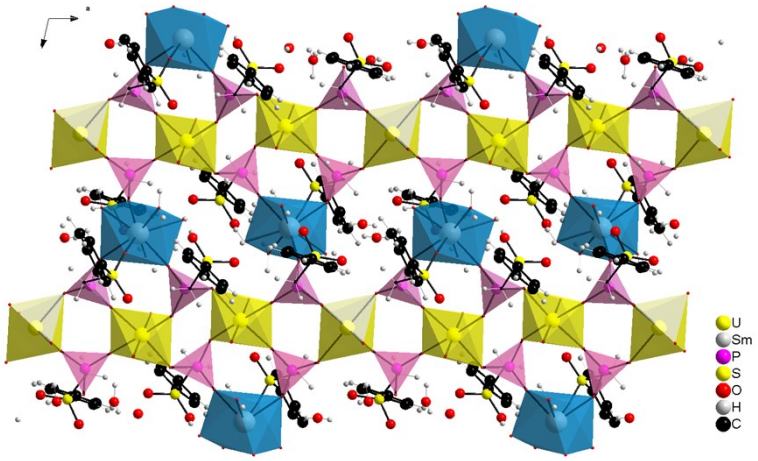
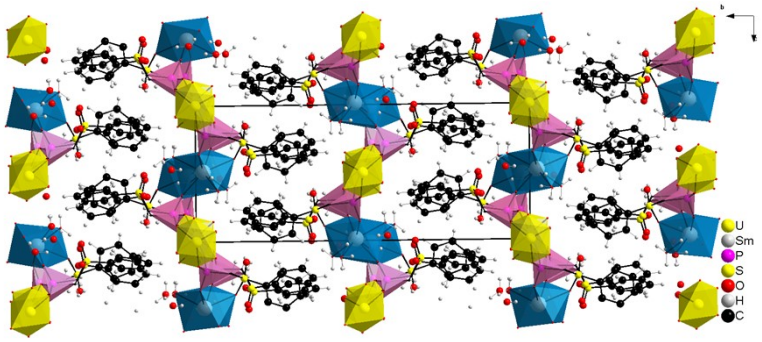


Figure S9 Depiction of the coordination environments of the phosphonate ligand, uranyl and samarium cations in **5** (thermal ellipsoids are given at 30% probability). A: 3-x, 1-y, 3-z; B: 1+x, y, 1+z; C: 2-x, 1-y, 2-z; D: 2-x, 1-y, 1-z; E: -1+x, y, -1+z.

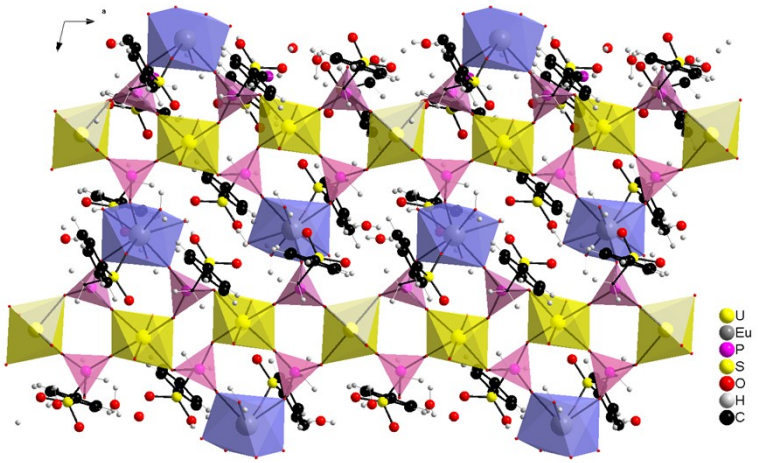


(a)

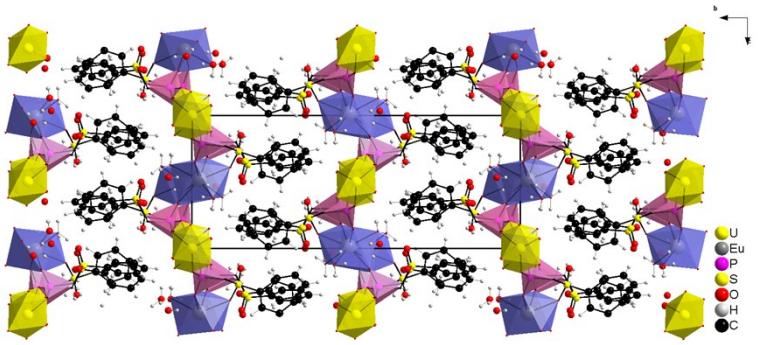


(b)

Figure S10 Two-dimensional layer (a) in *ac*-plane and three-dimensional stacking crystal structure along *a*-direction (b) of compound **3**.

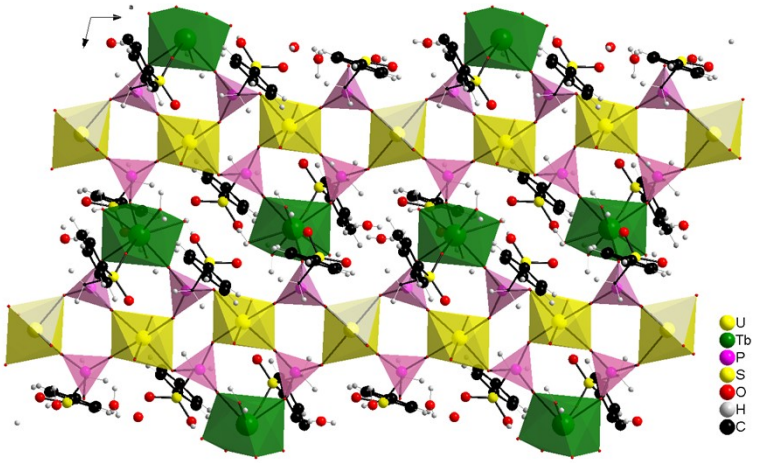


(a)

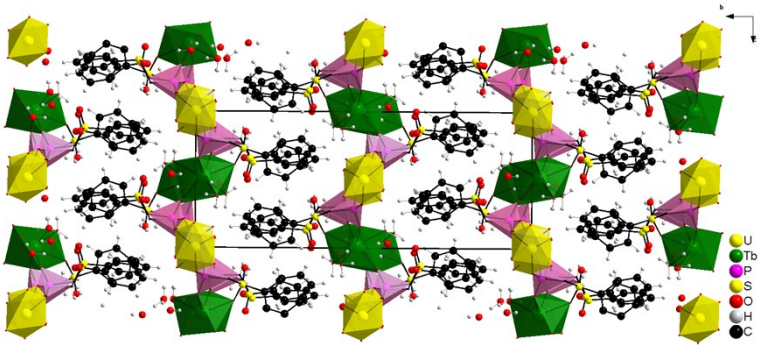


(b)

Figure S11 Two-dimensional layer (a) in *ac*-plane and three-dimensional stacking crystal structure along *a*-direction (b) of compound **4**.



(a)



(b)

Figure S12 Two-dimensional layer (a) in *ac*-plane and three-dimensional stacking crystal structure along *a*-direction (b) of compound **5**.

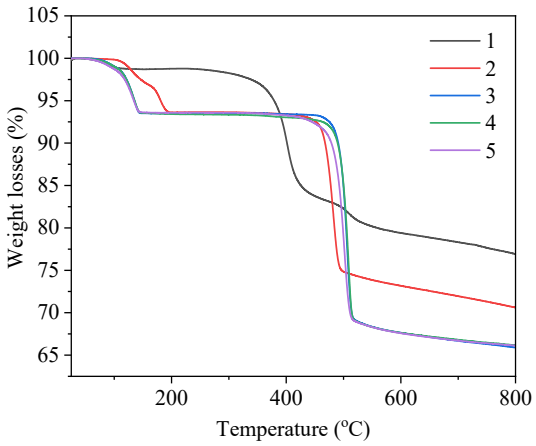


Figure S13 TGA curves of compounds **1-5**.

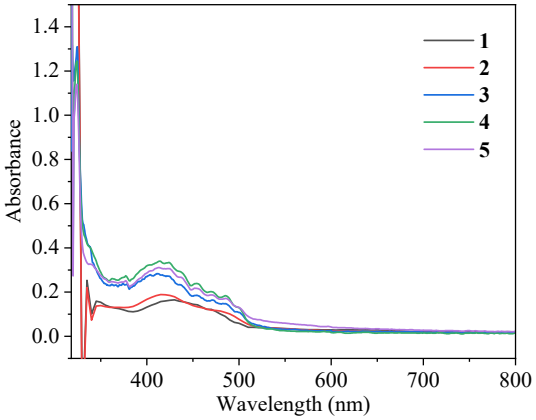


Figure S14 UV-Vis absorption spectra of compounds **1-5**.

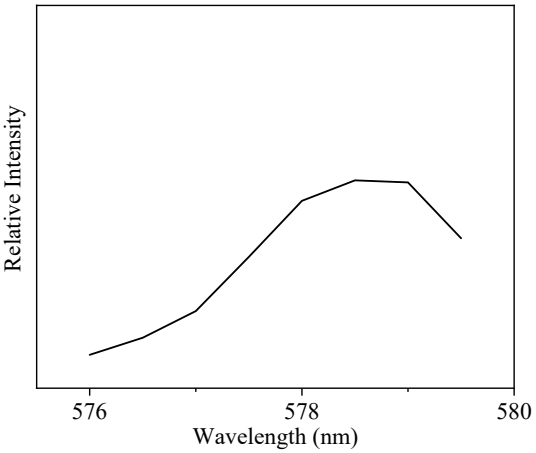


Figure S15 The $^5D_0 \rightarrow {}^7F_0$ transition on the emission spectrum of compound **4**.

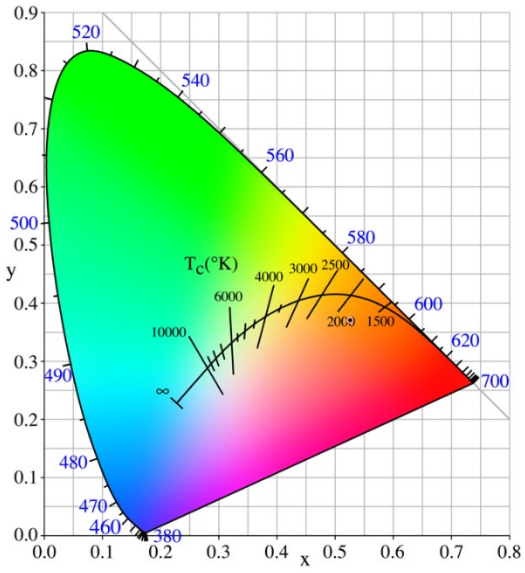


Figure S16 CIE diagram of compound **4** under excitation of 365 nm.

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[4] Sheldrick, G. M. *SHELXL* 2013 (Univ. Göttingen, 2013).