Supporting Information

Padmini Ramaswamy[a], Nayana N Hegde[a], Ramanath Prabhu[a], V.M.Vidya[a],

Ayan Datta[b] and Srinivasan Natarajan[a]*

[a] Prof. Dr. S. Natarajan, Framework solids Laboratory, Solid State and Structural Chemistry Unit, Indian Institute of Science, Bangalore-560012, India.
Fax: (+91) 80-2360-1310; E-mail: snatarajan@sscu.iisc.ernet.in

[b] School of Chemistry, Indian Institute of Science
Education and Research Thiruvananthapuram, CET Campus,
Thiruvananthapuram – 695016, Kerala (India).

Table S1. Important observed hydrogen bond interactions in the compounds **I – VIII.**

D – H … A	**D – H (Å)**	**H … A (Å)**	**D … A (Å)**	**D – H … A (°)**
		Compound I		
O(100)--H(101) ..O(7)	0.85	1.93	2.720(4)	154
O(100)--H(102) ..O(10)	0.84	2.13	2.935(4)	159
O(200)--H(202) ..O(7)	0.85	2.09	2.877(5)	153
C(10) --H(10A) ..O(6)	0.97	1.88	2.825(4)	164
C(10) --H(10B) ..O(8)	0.97	1.78	2.721(4)	162
C(12) --H(12A) ..O(9)	0.97	1.91	2.852(4)	163
C(12) --H(12B) ..O(100)	0.97	1.73	2.697(4)	171
C(14) --H(14A) ..O(7)	0.97	1.76	2.729(4)	177
C(14) --H(14B) ..O(10)	0.97	1.95	2.831(4)	150
		Compound IV		
C(10) --H(10A) ..O(13)	0.97	1.80	2.752(7)	166
C(10) --H(10B) ..O(16)	0.97	1.82	2.790(7)	175
C(11) --H(11B) ..O(2)	0.97	2.48	3.413(7)	162
C(12) --H(12A) ..O(3)	0.97	1.93	2.885(6)	169
C(12) --H(12B) ..O(4)	0.97	1.89	2.800(6)	155
C(15) --H(15A) ..O(100)	0.97	1.86	2.770(8)	154
C(15) --H(15B) ..O(17)	0.97	2.07	2.948(7)	149
C(16) --H(16A) ..O(5)	0.97	2.02	2.920(6)	154
C(16) --H(16B) ..O(13)	0.97	1.95	2.833(6)	150
O(200)--H(202) ..O(14)	0.85(11)	2.02(13)	2.843(10)	164(13)
O(300)--H(301) ..O(13)	0.84(9)	2.03(8)	2.797(8)	152(10)
O(300)--H(302) ..O(17)	0.84(9)	1.92(8)	2.764(8)	174(10)
		Compound V		
N(1) --H(1B) ..O(6)	0.90	2.42	3.24(2)	151

N(2) --H(2B) ..O(10)	0.90	1.92	2.79(2)	160
N(3) --H(3A) ..O(14)	0.90	1.89	2.73(2)	154
C(2) --H(2C) ..O(13)	0.95	1.79	2.70(2)	157
C(5) --H(5B) ..O(7)	0.97	2.46	3.33(2)	149
		Compound VII		
N(1) --H(1A) ..O(100)	0.90	1.79	2.679(4)	167
O(100)--H(101) ..O(2)	0.85	2.11	2.858(4)	148
O(100)--H(102) ..O(9)	0.84	1.99	2.796(4)	160
C(11) --H(11A) ..O(5)	0.91	2.34	3.251(5)	156

Table S2. Elemental Analyses for compounds **I** – **VIII**.

Compound	**Calculated (%)**	**Found (%)**
$[C_4N_2H_{12}]_3[In_2(HPO_3)_2(C_2O_4)_4].4H_2O$, **I**	C 22.27	C 22.78
	H 4.27	H 4.05
	N 7.79	N 7.86
$[In_2(HPO_3)_2(C_2O_4)(C_5H_5N)_2]$, **II**	C 22.65	C 22.34
	H 1.89	H 1.54
	N 4.41	N 4.45
$[C_4N_2H_{12}]_2[In_2(HPO_3)_3(C_2O_4)_2].3H_2O$, **IV**	C 16.45	C 16.90
	H 3.77	H 3.26
	N 6.39	N 6.32
$[C_4N_2H_{12}]_3[In_4(HPO_3)_6(C_2O_4)_3]$,**V**	C 14.72	C 14.35
	H 2.86	H 2.24
	N 5.73	N 5.02
$[C_4N_2H_{14}][In_4(H_2O)(HPO_3)_5(C_2O_4)_2].2H_2O$, **VI**	C 8.14	C 8.12
	H 1.95	H 1.23
	N 2.37	N 2.25
$[C_4N_2H_{12}]_{0.5}[In(SO_4)(C_2O_4)].H_2O$, **VII**	C 13.30	C 13.05
	H 2.22	H 2.17
	N 3.88	N 3.58
$[C_4N_2H_{12}]_{0.5}[In(HPO_4)(C_2O_4)].H_2O$, **VIII**	C 13.30	C 13.13
	H 2.49	H 2.09
	N 3.88	N 3.72

Table S3. Selected bond distances and angles for the compounds **I - VIII.**

Compound I

Bond	**Distance (Å)**	**Bond**	**Distance (Å)**
In(1)-O(1)	2.091(2)	O(9)-C(2)	1.233(4)
In(1)-O(2)	2.103(2)	O(10)-C(4)	1.229(4)
In(1)-O(3)	2.136(2)	O(11)-C(3)	1.207(4)
In(1)-O(4)	2.151(2)	C(1)-C(2)	1.558(4)
In(1)-O(5)	2.184(2)	C(3)-C(4)	1.564(5)
In(1)-O(6)	2.188(2)	C(14)-C(15)	1.484(5)
P(1)-O(7)	1.507(3)	C(14)-N(3)	1.485(5)
P(1)-O(1)#1	1.520(3)	C(10)-N(1)	1.468(5)
P(1)-O(2)	1.520(2)	C(10)-C(11)	1.486(5)
O(3)-C(4)	1.274(4)	C(12)-N(2)	1.475(4)
O(4)-C(2)	1.276(4)	C(12)-C(13)	1.482(5)
O(5)-C(1)	1.269(4)	N(2)-C(15)	1.508(5)
O(6)-C(3)	1.287(4)	N(3)-C(13)	1.507(6)
O(8)-C(1)	1.238(4)	N(1)-C(11)#2	1.508(5)
Angle	**Amplitude(˚)**	**Angle**	**Amplitude(˚)**
O(1)-In(1)-O(2)	96.19(11)	C(3)-O(6)-In(1)	114.4(2)
O(1)-In(1)-O(3)	95.68(10)	O(8)-C(1)-O(5)	124.6(3)
O(2)-In(1)-O(3)	89.56(9)	O(8)-C(1)-C(2)	119.0(3)
O(1)-In(1)-O(4)	94.52(10)	O(5)-C(1)-C(2)	116.4(3)
O(2)-In(1)-O(4)	97.80(10)	O(9)-C(2)-O(4)	124.4(3)
O(3)-In(1)-O(4)	166.71(10)	O(9)-C(2)-C(1)	119.1(3)
O(1)-In(1)-O(5)	170.57(10)	O(4)-C(2)-C(1)	116.5(3)
O(2)-In(1)-O(5)	88.75(10)	O(11)-C(3)-O(6)	126.0(3)
O(3)-In(1)-O(5)	92.37(9)	O(11)-C(3)-C(4)	119.3(3)
O(4)-In(1)-O(5)	76.82(9)	O(6)-C(3)-C(4)	114.6(3)
O(1)-In(1)-O(6)	88.06(11)	O(10)-C(4)-O(3)	124.6(3)
O(2)-In(1)-O(6)	165.82(10)	O(10)-C(4)-C(3)	118.6(3)
O(3)-In(1)-O(6)	76.53(9)	O(3)-C(4)-C(3)	116.8(3)
O(4)-In(1)-O(6)	95.34(9)	C(15)-C(14)-N(3)	112.2(3)
O(5)-In(1)-O(6)	89.05(10)	N(1)-C(10)-C(11)	110.7(3)
O(7)-P(1)-O(1)#1	112.02(16)	N(2)-C(12)-C(13)	111.4(3)
O(7)-P(1)-O(2)	112.03(15)	C(12)-N(2)-C(15)	110.3(3)
O(1)#1-P(1)-O(2)	112.88(14)	C(10)-C(11)-N(1)#2	110.6(3)
P(1)#1-O(1)-In(1)	134.87(15)	C(14)-N(3)-C(13)	111.3(3)

P(1)-O(2)-In(1)	134.33(15)	C(12)-C(13)-N(3)	110.4(3)
C(4)-O(3)-In(1)	115.9(2)	C(10)-N(1)-C(11)#2	110.7(3)
C(2)-O(4)-In(1)	114.66(19)	C(14)-C(15)-N(2)	110.1(3)
C(1)-O(5)-In(1)	113.7(2)		

Compound II

Bond	**Distance (Å)**	**Bond**	**Distance (Å)**
In(1)-O(1)	2.055(5)	O(4)-C(10)	1.257(7)
In(1)-O(2)	2.071(4)	O(5)-C(10)#4	1.249(7)
In(1)-O(3)	2.084(4)	N(1)-C(5)	1.321(8)
In(1)-O(4)	2.196(4)	N(1)-C(1)	1.332(8)
In(1)-O(5)	2.215(4)	C(10)-C(10)#4	1.577(10)
In(1)-N(1)	2.233(5)	C(5)-C(4)	1.386(10)
P(1)-O(3)#1	1.486(5)	C(1)-C(2)	1.379(11)
P(1)-O(1)#2	1.494(5)	C(2)-C(3)	1.362(13)
P(1)-O(2)	1.497(5)	C(3)-C(4)	1.359(13)
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)-In(1)-O(2)	89.6(2)	O(1)#2-P(1)-O(2)	106.1(4)
O(1)-In(1)-O(3)	172.6(2)	P(1)#3-O(1)-In(1)	149.7(4)
O(2)-In(1)-O(3)	93.4(2)	P(1)-O(2)-In(1)	135.5(3)
O(1)-In(1)-O(4)	90.3(2)	P(1)#1-O(3)-In(1)	138.1(3)
O(2)-In(1)-O(4)	96.57(19)	C(10)-O(4)-In(1)	115.0(3)
O(3)-In(1)-O(4)	95.98(19)	C(10)#4-O(5)-In(1)	114.0(3)
O(1)-In(1)-O(5)	87.2(2)	C(5)-N(1)-C(1)	118.3(6)
O(2)-In(1)-O(5)	171.97(18)	C(5)-N(1)-In(1)	121.0(4)
O(3)-In(1)-O(5)	90.6(2)	C(1)-N(1)-In(1)	120.6(4)
O(4)-In(1)-O(5)	76.09(14)	O(5)#4-C(10)-O(4)	125.8(5)
O(1)-In(1)-N(1)	85.8(2)	O(5)#4-C(10)-C(10)#4	117.7(6)
O(2)-In(1)-N(1)	98.4(2)	O(4)-C(10)-C(10)#4	116.5(6)
O(3)-In(1)-N(1)	87.15(18)	N(1)-C(5)-C(4)	122.6(7)
O(4)-In(1)-N(1)	164.54(16)	N(1)-C(1)-C(2)	121.8(8)
O(5)-In(1)-N(1)	88.76(17)	C(3)-C(2)-C(1)	119.7(8)
O(3)#1-P(1)-O(1)#2	111.7(3)	C(4)-C(3)-C(2)	118.8(8)
O(3)#1-P(1)-O(2)	112.0(4)	C(3)-C(4)-C(5)	118.8(8)

Compound III

Bond	**Distance (Å)**	**Bond**	**Distance (Å)**
In(1)-O(1)	2.070(3)	O(4)-C(10)	1.249(5)
In(1)-O(2)	2.086(3)	O(5)-C(10)#5	1.251(5)
In(1)-O(3)	2.102(3)	N(1)-C(1)	1.331(6)
In(1)-O(4)	2.202(3)	N(1)-C(2)	1.332(6)
In(1)-O(5)	2.226(3)	C(1)-C(3)	1.379(7)
In(1)-N(1)	2.231(3)	C(2)-C(5)	1.378(7)
P(1)-O(2)	1.493(3)	C(3)-C(4)	1.365(9)
P(1)-O(1)#1	1.500(3)	C(4)-C(5)	1.356(8)
P(1)-O(3)#2	1.502(3)	C(10)-C(10)#5	1.576(7)
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)-In(1)-O(2)	94.97(12)	O(1)#1-P(1)-O(3)#2	114.10(18)
O(1)-In(1)-O(3)	92.87(12)	P(1)#3-O(1)-In(1)	139.12(18)
O(2)-In(1)-O(3)	170.17(11)	P(1)-O(2)-In(1)	146.98(19)
O(1)-In(1)-O(4)	97.63(11)	P(1)#4-O(3)-In(1)	134.10(17)
O(2)-In(1)-O(4)	92.60(11)	C(10)-O(4)-In(1)	115.3(2)
O(3)-In(1)-O(4)	92.22(10)	C(10)#5-O(5)-In(1)	114.7(2)
O(1)-In(1)-O(5)	172.99(10)	C(1)-N(1)-C(2)	117.8(4)
O(2)-In(1)-O(5)	87.16(12)	C(1)-N(1)-In(1)	121.6(3)
O(3)-In(1)-O(5)	85.76(11)	C(2)-N(1)-In(1)	120.6(3)
O(4)-In(1)-O(5)	75.58(10)	N(1)-C(1)-C(3)	121.5(6)
O(1)-In(1)-N(1)	96.91(12)	N(1)-C(2)-C(5)	122.7(5)
O(2)-In(1)-N(1)	85.66(12)	C(4)-C(3)-C(1)	120.7(6)
O(3)-In(1)-N(1)	87.53(11)	C(5)-C(4)-C(3)	117.6(6)
O(4)-In(1)-N(1)	165.46(11)	C(4)-C(5)-C(2)	119.7(6)
O(5)-In(1)-N(1)	89.90(11)	O(4)-C(10)-O(5)#5	125.9(4)
O(2)-P(1)-O(1)#1	115.11(18)	O(4)-C(10)-C(10)#5	117.3(4)
O(2)-P(1)-O(3)#2	109.92(18)	O(5)#5-C(10)-C(10)#5	116.8(4)

Compound IV

Bond	Distance (Å)	Bond	Distance (Å)
In(1)-O(1)	2.115(3)	O(6)-C(3)	1.286(7)
In(1)-O(2)	2.139(3)	O(11)-C(2)	1.265(6)
In(1)-O(3)	2.145(3)	O(12)-C(1)	1.266(6)
In(1)-O(4)	2.146(3)	O(14)-C(3)	1.219(7)
In(1)-O(5)	2.150(3)	O(15)-C(4)	1.218(7)
In(1)-O(6)	2.152(4)	O(16)-C(2)	1.236(6)
In(2)-O(7)	2.091(4)	O(17)-C(1)	1.227(7)
In(2)-O(8)	2.097(3)	C(1)-C(2)	1.553(7)
In(2)-O(9)	2.105(3)	C(3)-C(4)	1.558(8)
In(2)-O(10)	2.118(3)	C(10)-C(11)	1.477(8)
In(2)-O(11)	2.188(4)	C(10)-N(4)	1.484(8)
In(2)-O(12)	2.219(4)	C(11)-N(3)	1.516(8)
P(1)-O(2)	1.508(4)	C(12)-C(13)	1.484(7)
P(1)-O(10)	1.521(4)	C(12)-N(3)	1.493(8)
P(1)-O(3)#1	1.539(3)	C(13)-N(4)	1.506(7)
P(2)-O(8)#2	1.520(4)	C(14)-C(15)	1.471(8)
P(2)-O(1)#2	1.524(3)	C(14)-N(1)	1.513(9)
P(2)-O(5)	1.531(3)	C(15)-N(2)	1.487(7)
P(3)-O(9)#3	1.516(4)	C(16)-C(17)	1.486(7)
P(3)-O(13)	1.517(4)	C(16)-N(1)	1.491(7)
P(3)-O(7)	1.523(4)	C(17)-N(2)	1.510(8)
O(4)-C(4)	1.290(6)		
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)-In(1)-O(2)	90.60(13)	O(9)#3-P(3)-O(7)	113.6(2)
O(1)-In(1)-O(3)	91.84(13)	O(13)-P(3)-O(7)	110.1(2)
O(2)-In(1)-O(3)	88.99(13)	P(2)#2-O(1)-In(1)	129.39(19)
O(1)-In(1)-O(4)	98.02(13)	P(1)-O(2)-In(1)	134.4(2)
O(2)-In(1)-O(4)	98.54(14)	P(1)#1-O(3)-In(1)	127.96(19)
O(3)-In(1)-O(4)	167.49(14)	C(4)-O(4)-In(1)	115.6(3)
O(1)-In(1)-O(5)	90.94(13)	P(2)-O(5)-In(1)	130.5(2)
O(2)-In(1)-O(5)	169.84(13)	C(3)-O(6)-In(1)	115.0(3)
O(3)-In(1)-O(5)	80.92(13)	P(3)-O(7)-In(2)	134.5(2)
O(4)-In(1)-O(5)	91.20(15)	P(2)#2-O(8)-In(2)	139.3(2)
O(1)-In(1)-O(6)	173.95(13)	P(3)#3-O(9)-In(2)	138.2(3)

O(2)-In(1)-O(6)	86.99(14)	P(1)-O(10)-In(2)	134.4(2)
O(3)-In(1)-O(6)	93.66(14)	C(2)-O(11)-In(2)	116.8(3)
O(4)-In(1)-O(6)	76.88(14)	C(1)-O(12)-In(2)	115.7(3)
O(5)-In(1)-O(6)	92.42(14)	O(17)-C(1)-O(12)	126.3(5)
O(7)-In(2)-O(8)	107.75(16)	O(17)-C(1)-C(2)	117.8(5)
O(7)-In(2)-O(9)	92.20(15)	O(12)-C(1)-C(2)	115.8(4)
O(8)-In(2)-O(9)	85.70(14)	O(16)-C(2)-O(11)	125.9(5)
O(7)-In(2)-O(10)	89.71(15)	O(16)-C(2)-C(1)	117.9(5)
O(8)-In(2)-O(10)	91.89(14)	O(11)-C(2)-C(1)	116.2(4)
O(9)-In(2)-O(10)	177.29(15)	O(14)-C(3)-O(6)	124.6(6)
O(7)-In(2)-O(11)	90.23(15)	O(14)-C(3)-C(4)	119.3(5)
O(8)-In(2)-O(11)	161.91(14)	O(6)-C(3)-C(4)	116.1(5)
O(9)-In(2)-O(11)	95.76(15)	O(15)-C(4)-O(4)	124.3(5)
O(10)-In(2)-O(11)	86.14(14)	O(15)-C(4)-C(3)	120.5(5)
O(7)-In(2)-O(12)	164.58(15)	C(11)-C(10)-N(4)	111.1(4)
O(8)-In(2)-O(12)	87.63(14)	C(10)-C(11)-N(3)	109.9(5)
O(9)-In(2)-O(12)	87.86(16)	C(13)-C(12)-N(3)	112.2(4)
O(10)-In(2)-O(12)	90.81(15)	C(12)-C(13)-N(4)	110.2(5)
O(11)-In(2)-O(12)	74.43(13)	C(15)-C(14)-N(1)	111.4(5)
O(2)-P(1)-O(10)	113.4(2)	C(14)-C(15)-N(2)	111.1(4)
O(2)-P(1)-O(3)#1	111.9(2)	C(17)-C(16)-N(1)	111.8(4)
O(10)-P(1)-O(3)#1	108.2(2)	C(16)-C(17)-N(2)	110.9(5)
O(8)#2-P(2)-O(1)#2	112.7(2)	C(16)-N(1)-C(14)	110.8(5)
O(8)#2-P(2)-O(5)	110.0(2)	C(15)-N(2)-C(17)	110.8(5)
O(1)#2-P(2)-O(5)	110.9(2)	C(12)-N(3)-C(11)	111.4(5)
O(9)#3-P(3)-O(13)	112.02(19)	C(10)-N(4)-C(13)	109.4(5)

Compound V

Bond	Distance (Å)	Bond	Distance (Å)
In(1)-O(2)	2.081(11)	O(5)-C(15)	1.225(18)
In(1)-O(1)	2.089(12)	O(6)-C(15)#4	1.261(18)
In(1)-O(3)	2.134(11)	O(7)-In(2)#5	2.097(11)
In(1)-O(4)	2.139(13)	O(8)-In(2)#3	2.103(11)
In(1)-O(6)	2.222(10)	O(10)-C(4)	1.29(2)
In(1)-O(5)	2.271(12)	O(11)-C(1)	1.29(2)
In(2)-O(7)#1	2.097(11)	O(14)-C(1)	1.20(2)
In(2)-O(8)#2	2.103(11)	O(15)-C(4)	1.22(2)
In(2)-O(12)	2.103(12)	N(2)-C(5)	1.46(2)
In(2)-O(9)	2.144(14)	N(2)-C(6)	1.49(2)
In(2)-O(11)	2.203(12)	N(3)-C(8)	1.45(2)
In(2)-O(10)	2.215(12)	N(3)-C(7)	1.49(3)
P(1)-O(4)	1.499(12)	C(2)-C(3)	1.45(3)
P(1)-O(8)	1.502(12)	C(2)-N(1)	1.47(3)
P(1)-O(9)	1.520(14)	C(3)-N(1)#6	1.54(3)
P(2)-O(12)	1.511(12)	C(1)-C(4)	1.56(2)
P(2)-O(3)#2	1.517(12)	N(1)-C(3)#6	1.54(3)
P(2)-O(2)	1.527(11)	C(6)-C(7)	1.52(3)
P(3)-O(13)	1.491(12)	C(8)-C(5)	1.47(2)
P(3)-O(7)	1.538(12)	C(15)-O(6)#4	1.261(18)
P(3)-O(1)	1.540(12)	C(15)-C(15)#4	1.51(3)
O(3)-P(2)#3	1.517(12)		
Angle	**Amplitude(˚)**	**Angle**	**Amplitude(˚)**
O(2)-In(1)-O(1)	93.1(5)	O(12)-P(2)-O(2)	111.4(7)
O(2)-In(1)-O(3)	171.8(5)	O(3)#2-P(2)-O(2)	112.5(7)
O(1)-In(1)-O(3)	83.6(5)	O(13)-P(3)-O(7)	116.0(7)
O(2)-In(1)-O(4)	93.6(5)	O(13)-P(3)-O(1)	112.0(7)
O(1)-In(1)-O(4)	87.2(4)	O(7)-P(3)-O(1)	107.4(7)
O(3)-In(1)-O(4)	93.8(6)	P(3)-O(1)-In(1)	128.3(7)
O(2)-In(1)-O(6)	92.8(5)	P(2)-O(2)-In(1)	134.0(6)
O(1)-In(1)-O(6)	172.7(4)	P(2)#3-O(3)-In(1)	137.8(8)
O(3)-In(1)-O(6)	90.0(4)	P(1)-O(4)-In(1)	135.1(9)
O(4)-In(1)-O(6)	96.8(4)	C(15)-O(5)-In(1)	113.2(10)
O(2)-In(1)-O(5)	87.2(5)	C(15)#4-O(6)-In(1)	115.9(9)

O(1)-In(1)-O(5)	102.2(4)	P(3)-O(7)-In(2)#5	134.0(7)
O(3)-In(1)-O(5)	86.1(5)	P(1)-O(8)-In(2)#3	140.9(8)
O(4)-In(1)-O(5)	170.6(4)	P(1)-O(9)-In(2)	132.7(9)
O(6)-In(1)-O(5)	73.8(4)	C(4)-O(10)-In(2)	116.4(11)
O(7)#1-In(2)-O(8)#2	87.5(5)	C(1)-O(11)-In(2)	114.5(11)
O(7)#1-In(2)-O(12)	96.2(4)	P(2)-O(12)-In(2)	136.4(8)
O(8)#2-In(2)-O(12)	98.0(5)	C(5)-N(2)-C(6)	113.6(14)
O(7)#1-In(2)-O(9)	88.4(5)	C(8)-N(3)-C(7)	111.8(14)
O(8)#2-In(2)-O(9)	170.5(5)	C(3)-C(2)-N(1)	112.8(16)
O(12)-In(2)-O(9)	91.0(5)	C(2)-C(3)-N(1)#6	109.2(15)
O(7)#1-In(2)-O(11)	90.1(4)	O(14)-C(1)-O(11)	126.3(16)
O(8)#2-In(2)-O(11)	90.9(5)	O(14)-C(1)-C(4)	117.6(15)
O(12)-In(2)-O(11)	169.3(5)	O(11)-C(1)-C(4)	116.1(17)
O(9)-In(2)-O(11)	80.5(5)	C(2)-N(1)-C(3)#6	107.9(15)
O(7)#1-In(2)-O(10)	164.3(4)	O(15)-C(4)-O(10)	125.4(18)
O(8)#2-In(2)-O(10)	89.1(5)	O(15)-C(4)-C(1)	119.9(17)
O(12)-In(2)-O(10)	99.4(5)	O(10)-C(4)-C(1)	114.7(15)
O(9)-In(2)-O(10)	92.5(5)	N(2)-C(6)-C(7)	111.0(15)
O(11)-In(2)-O(10)	74.7(4)	N(3)-C(7)-C(6)	108.3(15)
O(4)-P(1)-O(8)	118.8(9)	N(2)-C(5)-C(8)	111.7(15)
O(4)-P(1)-O(9)	112.1(9)	O(5)-C(15)-O(6)#4	123.0(14)
O(8)-P(1)-O(9)	107.0(7)	O(5)-C(15)-C(15)#4	120.4(17)
O(12)-P(2)-O(3)#2	115.0(8)	O(6)#4-C(15)-C(15)#4	116.3(16)

Compound VI

Bond	Distance (Å)	Bond	Distance (Å)
In(1)-O(1)	2.080(5)	P(2)-O(10)	1.512(5)
In(1)-O(2)	2.113(5)	P(2)-O(14)	1.519(6)
In(1)-O(3)	2.125(5)	P(2)-O(3)	1.533(5)
In(1)-O(4)	2.129(5)	P(3)-O(4)#5	1.512(5)
In(1)-O(5)	2.201(5)	P(3)-O(9)	1.518(5)
In(1)-O(6)	2.211(5)	P(3)-O(2)	1.522(5)
In(2)-O(7)	2.089(5)	P(4)-O(1)	1.502(6)
In(2)-O(10)#1	2.107(5)	P(4)-O(16)	1.504(6)
In(2)-O(8)	2.107(6)	P(4)-O(7)	1.518(6)
In(2)-O(9)#1	2.111(5)	P(5)-O(8)	1.435(6)
In(2)-O(11)#2	2.195(5)	P(5)-O(20)#2	1.446(6)
In(2)-O(12)#2	2.231(5)	P(5)-O(13)#1	1.503(6)
In(3)-O(13)	2.083(5)	O(5)-C(3)	1.250(9)
In(3)-O(14)	2.083(6)	O(6)-C(4)	1.253(8)
In(3)-O(15)	2.103(6)	O(10)-In(2)#3	2.107(5)
In(3)-O(16)#3	2.106(6)	O(11)-C(3)	1.255(8)
In(3)-O(17)	2.202(5)	O(12)-C(4)	1.243(8)
In(3)-O(18)	2.214(5)	O(17)-C(2)	1.264(9)
In(4)-O(19)	2.065(6)	O(18)-C(1)	1.246(9)
In(4)-O(20)	2.077(5)	O(23)-C(1)#1	1.256(9)
In(4)-O(21)	2.101(5)	O(24)-C(2)#1	1.242(9)
In(4)-O(22)	2.167(6)	C(1)-C(2)	1.548(10)
In(4)-O(23)	2.197(5)	C(3)-C(4)	1.544(10)
In(4)-O(24)	2.218(5)	C(11)-N(1)	1.53(2)
P(1)-O(21)#4	1.512(6)	C(11)-C(11)#7	1.57(3)
P(1)-O(19)	1.513(6)	C(12)-C(14)	1.96(5)
P(1)-O(15)	1.513(6)	C(13)-C(13)#8	1.49(2)
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)-In(1)-O(2)	103.5(2)	O(19)-In(4)-O(24)	170.9(2)
O(1)-In(1)-O(3)	95.7(2)	O(20)-In(4)-O(24)	94.0(2)
O(2)-In(1)-O(3)	91.0(2)	O(21)-In(4)-O(24)	88.1(2)
O(1)-In(1)-O(4)	86.0(2)	O(22)-In(4)-O(24)	82.2(2)
O(2)-In(1)-O(4)	89.90(19)	O(23)-In(4)-O(24)	75.71(19)
O(3)-In(1)-O(4)	177.9(2)	O(21)#4-P(1)-O(19)	113.0(3)

O(1)-In(1)-O(5)	167.3(2)	O(21)#4-P(1)-O(15)	110.4(3)
O(2)-In(1)-O(5)	87.88(19)	O(19)-P(1)-O(15)	113.4(3)
O(3)-In(1)-O(5)	89.5(2)	O(10)-P(2)-O(14)	113.6(3)
O(4)-In(1)-O(5)	88.6(2)	O(10)-P(2)-O(3)	113.3(3)
O(1)-In(1)-O(6)	93.3(2)	O(14)-P(2)-O(3)	109.8(3)
O(2)-In(1)-O(6)	163.08(19)	O(4)#5-P(3)-O(9)	110.4(3)
O(3)-In(1)-O(6)	88.90(19)	O(4)#5-P(3)-O(2)	111.7(3)
O(4)-In(1)-O(6)	89.67(19)	O(9)-P(3)-O(2)	112.6(3)
O(5)-In(1)-O(6)	75.20(18)	O(1)-P(4)-O(16)	114.0(3)
O(7)-In(2)-O(10)#1	106.5(2)	O(1)-P(4)-O(7)	108.1(3)
O(7)-In(2)-O(8)	94.9(3)	O(16)-P(4)-O(7)	114.6(3)
O(10)#1-In(2)-O(8)	91.1(3)	O(8)-P(5)-O(20)#2	121.3(4)
O(7)-In(2)-O(9)#1	89.3(2)	O(8)-P(5)-O(13)#1	118.2(4)
O(10)#1-In(2)-O(9)#1	91.3(2)	O(20)#2-P(5)-O(13)#1	112.0(3)
O(8)-In(2)-O(9)#1	174.3(2)	P(4)-O(1)-In(1)	141.7(4)
O(7)-In(2)-O(11)#2	89.0(2)	P(3)-O(2)-In(1)	131.2(3)
O(10)#1-In(2)-O(11)#2	164.4(2)	P(2)-O(3)-In(1)	131.1(3)
O(8)-In(2)-O(11)#2	85.3(2)	P(3)#5-O(4)-In(1)	132.9(3)
O(9)#1-In(2)-O(11)#2	91.08(19)	C(3)-O(5)-In(1)	114.9(5)
O(7)-In(2)-O(12)#2	163.6(2)	C(4)-O(6)-In(1)	115.1(4)
O(10)#1-In(2)-O(12)#2	89.66(19)	P(4)-O(7)-In(2)	136.8(3)
O(8)-In(2)-O(12)#2	87.3(2)	P(5)-O(8)-In(2)	155.7(4)
O(9)#1-In(2)-O(12)#2	87.6(2)	P(3)-O(9)-In(2)#3	133.7(3)
O(11)#2-In(2)-O(12)#2	74.99(18)	P(2)-O(10)-In(2)#3	131.9(3)
O(13)-In(3)-O(14)	97.4(2)	C(3)-O(11)-In(2)#6	115.1(4)
O(13)-In(3)-O(15)	84.7(2)	C(4)-O(12)-In(2)#6	114.8(4)
O(14)-In(3)-O(15)	93.2(3)	P(5)#3-O(13)-In(3)	130.7(4)
O(13)-In(3)-O(16)#3	93.3(2)	P(2)-O(14)-In(3)	133.8(4)
O(14)-In(3)-O(16)#3	93.3(2)	P(1)-O(15)-In(3)	142.7(4)
O(15)-In(3)-O(16)#3	173.4(2)	P(4)-O(16)-In(3)#1	141.0(4)
O(13)-In(3)-O(17)	167.0(2)	C(2)-O(17)-In(3)	114.7(4)
O(14)-In(3)-O(17)	93.5(2)	C(1)-O(18)-In(3)	114.6(4)
O(15)-In(3)-O(17)	87.6(2)	P(1)-O(19)-In(4)	137.9(3)
O(16)#3-In(3)-O(17)	93.2(2)	P(5)#6-O(20)-In(4)	138.6(4)
O(13)-In(3)-O(18)	94.1(2)	P(1)#4-O(21)-In(4)	138.6(3)
O(14)-In(3)-O(18)	168.1(2)	C(1)#1-O(23)-In(4)	114.3(4)
O(15)-In(3)-O(18)	91.2(2)	C(2)#1-O(24)-In(4)	114.0(5)

O(16)#3-In(3)-O(18)	82.7(2)	O(18)-C(1)-O(23)#3	125.1(6)
O(17)-In(3)-O(18)	75.57(19)	O(18)-C(1)-C(2)	117.7(6)
O(19)-In(4)-O(20)	94.6(2)	O(23)#3-C(1)-C(2)	117.2(6)
O(19)-In(4)-O(21)	95.6(2)	O(24)#3-C(2)-O(17)	125.0(7)
O(20)-In(4)-O(21)	86.3(2)	O(24)#3-C(2)-C(1)	118.0(6)
O(19)-In(4)-O(22)	94.4(3)	O(17)-C(2)-C(1)	117.0(6)
O(20)-In(4)-O(22)	92.3(2)	O(5)-C(3)-O(11)	124.4(7)
O(21)-In(4)-O(22)	170.0(3)	O(5)-C(3)-C(4)	117.8(6)
O(19)-In(4)-O(23)	96.1(2)	O(11)-C(3)-C(4)	117.7(6)
O(20)-In(4)-O(23)	168.1(2)	O(12)-C(4)-O(6)	126.2(6)
O(21)-In(4)-O(23)	87.4(2)	O(12)-C(4)-C(3)	117.0(6)
O(22)-In(4)-O(23)	92.1(2)	O(6)-C(4)-C(3)	116.7(6)

Compound VII

Bond	Distance (Å)	Bond	Distance (Å)
In(1)-O(1)	2.143(3)	O(1)-S(1)#3	1.497(3)
In(1)-O(2)	2.145(2)	O(4)-C(2)	1.258(4)
In(1)-O(3)	2.219(2)	O(4)-In(1)#4	2.230(2)
In(1)-O(4)#1	2.230(2)	O(5)-C(2)	1.242(4)
In(1)-O(5)	2.236(2)	O(6)-C(1)	1.249(4)
In(1)-O(6)#1	2.238(2)	O(6)-In(1)#4	2.238(2)
In(1)-O(7)	2.275(2)	O(7)-C(1)	1.252(4)
S(1)-O(8)	1.450(3)	C(1)-C(2)	1.537(5)
S(1)-O(9)	1.460(3)	N(1)-C(10)	1.474(5)
S(1)-O(1)#2	1.497(3)	N(1)-C(11)	1.492(5)
S(1)-O(2)	1.499(3)	C(10)-C(11)#5	1.502(6)
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)-In(1)-O(2)	167.45(9)	O(8)-S(1)-O(9)	111.67(16)
O(1)-In(1)-O(3)	108.69(10)	O(8)-S(1)-O(1)#2	110.18(16)
O(2)-In(1)-O(3)	80.13(10)	O(9)-S(1)-O(1)#2	110.37(15)
O(1)-In(1)-O(4)#1	85.36(9)	O(8)-S(1)-O(2)	111.05(16)
O(2)-In(1)-O(4)#1	106.23(9)	O(9)-S(1)-O(2)	108.75(15)
O(3)-In(1)-O(4)#1	71.70(9)	O(1)#2-S(1)-O(2)	104.59(15)
O(1)-In(1)-O(5)	85.37(10)	S(1)#3-O(1)-In(1)	130.05(16)
O(2)-In(1)-O(5)	82.32(10)	S(1)-O(2)-In(1)	129.63(15)
O(3)-In(1)-O(5)	142.82(9)	C(2)-O(4)-In(1)#4	115.7(2)
O(4)#1-In(1)-O(5)	145.15(9)	C(2)-O(5)-In(1)	117.3(2)
O(1)-In(1)-O(6)#1	87.86(10)	C(1)-O(6)-In(1)#4	115.7(2)
O(2)-In(1)-O(6)#1	90.77(10)	C(1)-O(7)-In(1)	116.5(2)
O(3)-In(1)-O(6)#1	139.96(9)	O(6)-C(1)-O(7)	126.7(3)
O(4)#1-In(1)-O(6)#1	73.75(8)	O(6)-C(1)-C(2)	117.2(3)
O(5)-In(1)-O(6)#1	72.39(9)	O(7)-C(1)-C(2)	116.1(3)
O(1)-In(1)-O(7)	84.63(9)	O(5)-C(2)-O(4)	125.2(3)
O(2)-In(1)-O(7)	89.28(9)	O(5)-C(2)-C(1)	117.7(3)
O(3)-In(1)-O(7)	74.86(9)	O(4)-C(2)-C(1)	117.1(3)
O(4)#1-In(1)-O(7)	139.70(10)	C(10)-N(1)-C(11)	111.3(3)
O(5)-In(1)-O(7)	72.40(9)	N(1)-C(10)-C(11)#5	111.5(4)
O(6)#1-In(1)-O(7)	144.45(9)	N(1)-C(11)-C(10)#5	110.4(3)

Compound VIII

Bond	Distance (Å)	Bond	Distance (Å)

In(1)-O(1)	2.136(3)	O(5)-C(11)	1.242(4)
In(1)-O(2)	2.145(3)	O(6)-C(10)#3	1.243(5)
In(1)-O(3)	2.217(3)	O(7)-C(11)#3	1.262(4)
In(1)-O(4)	2.235(3)	C(10)-O(6)#4	1.243(5)
In(1)-O(6)	2.237(3)	C(10)-C(11)	1.541(5)
In(1)-O(5)	2.240(3)	C(11)-O(7)#4	1.262(4)
In(1)-O(7)	2.276(3)	C(2)-N(1)#2	1.500(6)
P(1)-O(8)	1.449(3)	C(2)-C(1)#5	1.503(6)
P(1)-O(9)	1.456(3)	C(1)-C(2)#6	1.503(6)
P(1)-O(2)#1	1.494(3)	C(1)-N(1)#7	1.530(7)
P(1)-O(1)	1.509(3)	N(1)-C(2)#1	1.500(6)
O(2)-P(1)#2	1.494(3)	N(1)-C(1)#8	1.530(7)
O(4)-C(10)	1.251(4)		
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)-In(1)-O(2)	167.32(11)	O(5)-In(1)-O(7)	144.64(9)
O(1)-In(1)-O(3)	80.28(11)	O(8)-P(1)-O(9)	112.10(18)
O(2)-In(1)-O(3)	108.63(11)	O(8)-P(1)-O(2)#1	110.23(19)
O(1)-In(1)-O(4)	106.34(11)	O(9)-P(1)-O(2)#1	110.45(17)
O(2)-In(1)-O(4)	85.36(11)	O(8)-P(1)-O(1)	110.93(19)
O(3)-In(1)-O(4)	71.78(10)	O(9)-P(1)-O(1)	108.49(17)
O(1)-In(1)-O(6)	82.06(11)	O(2)#1-P(1)-O(1)	104.36(16)
O(2)-In(1)-O(6)	85.51(11)	P(1)-O(1)-In(1)	129.57(17)
O(3)-In(1)-O(6)	142.69(10)	C(10)-O(4)-In(1)	115.9(2)
O(4)-In(1)-O(6)	145.18(10)	C(11)-O(5)-In(1)	115.7(2)
O(1)-In(1)-O(5)	90.73(11)	C(10)#3-O(6)-In(1)	117.6(2)
O(2)-In(1)-O(5)	87.90(11)	C(11)#3-O(7)-In(1)	116.6(2)
O(3)-In(1)-O(5)	139.89(10)	O(6)#4-C(10)-O(4)	125.5(3)
O(4)-In(1)-O(5)	73.57(9)	O(6)#4-C(10)-C(11)	117.6(3)
O(6)-In(1)-O(5)	72.58(10)	O(4)-C(10)-C(11)	116.9(3)
O(1)-In(1)-O(7)	89.27(11)	O(5)-C(11)-O(7)#4	126.8(3)
O(2)-In(1)-O(7)	84.60(11)	O(7)#4-C(11)-C(10)	115.7(3)
O(3)-In(1)-O(7)	74.77(10)	N(1)#2-C(2)-C(1)#5	111.1(4)
O(4)-In(1)-O(7)	139.70(10)	C(2)#6-C(1)-N(1)#7	110.1(4)
O(6)-In(1)-O(7)	72.41(9)	C(2)#1-N(1)-C(1)#8	110.8(4)

Symmetry transformations used to generate equivalent atoms:

For **I**: #1 -x,-y,-z+1; #2 -x+2,-y+1,-z+2

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

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

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

For **V** : #1 x-1,y-1,z; #2 x-1,y,z ; #3 x+1,y,z; #4 -x+1,-y,-z+1 ; #5 x+1,y+1,z; #6 -x,-y+1,-z+1

For **VI** : #1 x+1,y,z; #2 x+1,y+1,z; #3 x-1,y,z ; #4 -x,-y+1,-z+2; #5 -x,-y+1,-z+1; #6 x-1,y-1,z

#7 -x+1,-y+2,-z+1; #8 -x+1,-y+2,-z

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

For **VIII**: #1 x-1,y,z #2 x+1,y,z #3 -x+1,y-1/2,-z+1/2 #4 -x+1,y+1/2,-z+1/2 #5 x+1,-y+1/2,z+1/2

#6 x-1,-y+1/2,z-1/2 ; #7 -x,y+1/2,-z+1/2; #8 -x,y-1/2,-z+1/2

Table S4. Selected bond distances and angles for the compound **T1.**

Compound I

Bond	**Distance (Å)**	**Bond**	**Distance (Å)**
In(1)-O(1)#1	2.141(7)	In(3)-O(8)	2.255(8)
In(1)-O(1)	2.141(7)	P(1)-O(7)	1.520(8)
In(1)-O(1)#2	2.141(7)	P(1)-O(1)	1.524(8)
In(1)-O(1)#3	2.142(7)	P(1)-O(2)	1.531(8)
In(1)-O(1)#4	2.141(7)	P(2)-O(4)	1.516(8)
In(1)-O(1)#5	2.141(7)	P(2)-O(5)	1.518(8)
In(2)-O(2)#6	2.126(7)	P(2)-O(6)	1.539(9)
In(2)-O(2)#7	2.126(7)	P(3)-O(3)#9	1.506(8)
In(2)-O(2)#4	2.126(7)	P(3)-O(3)	1.506(8)
In(2)-O(2)#8	2.126(7)	P(3)-O(3)#10	1.506(8)
In(2)-O(2)#1	2.126(7)	O(4)-In(3)#10	2.105(8)
In(2)-O(2)	2.126(7)	O(5)-In(3)#5	2.120(8)
In(3)-O(3)	2.097(9)	C(1)-N(1)#11	1.69(6)
In(3)-O(4)#9	2.105(8)	C(1)-N(1)#3	1.71(6)
In(3)-O(5)#3	2.120(8)	N(1)-C(1)#12	1.69(6)
In(3)-O(6)	2.121(8)	N(1)-C(1)#5	1.71(6)
In(3)-O(7)	2.126(8)		
Angle	**Amplitude(°)**	**Angle**	**Amplitude(°)**
O(1)#1-In(1)-O(1)	92.6(3)	O(3)-In(3)-O(5)#3	87.9(3)
O(1)#1-In(1)-O(1)#2	87.4(3)	O(4)#9-In(3)-O(5)#3	93.9(3)
O(1)-In(1)-O(1)#2	180.0(5)	O(3)-In(3)-O(6)	94.2(4)
O(1)#1-In(1)-O(1)#3	180.0(5)	O(4)#9-In(3)-O(6)	90.1(3)
O(1)-In(1)-O(1)#3	87.4(3)	O(5)#3-In(3)-O(6)	175.3(3)
O(1)#2-In(1)-O(1)#3	92.6(3)	O(3)-In(3)-O(7)	92.0(3)
O(1)#1-In(1)-O(1)#4	92.6(3)	O(4)#9-In(3)-O(7)	169.1(4)
O(1)-In(1)-O(1)#4	92.6(3)	O(5)#3-In(3)-O(7)	92.8(4)
O(1)#2-In(1)-O(1)#4	87.4(3)	O(6)-In(3)-O(7)	82.9(3)
O(1)#3-In(1)-O(1)#4	87.4(3)	O(3)-In(3)-O(8)	178.4(3)
O(1)#1-In(1)-O(1)#5	87.4(3)	O(4)#9-In(3)-O(8)	84.5(3)
O(1)-In(1)-O(1)#5	87.4(3)	O(5)#3-In(3)-O(8)	92.9(3)
O(1)#2-In(1)-O(1)#5	92.6(3)	O(6)-In(3)-O(8)	84.9(3)
O(1)#3-In(1)-O(1)#5	92.6(3)	O(7)-In(3)-O(8)	86.6(3)
O(1)#4-In(1)-O(1)#5	180.0(8)	O(7)-P(1)-O(1)	112.5(5)
O(2)#6-In(2)-O(2)#7	90.5(3)	O(7)-P(1)-O(2)	107.6(5)

O(2)#6-In(2)-O(2)#4	176.5(5)	O(1)-P(1)-O(2)	114.9(4)
O(2)#7-In(2)-O(2)#4	92.1(4)	O(4)-P(2)-O(5)	112.4(5)
O(2)#6-In(2)-O(2)#8	90.5(3)	O(4)-P(2)-O(6)	112.6(5)
O(2)#7-In(2)-O(2)#8	90.5(3)	O(5)-P(2)-O(6)	110.9(5)
O(2)#4-In(2)-O(2)#8	87.1(4)	O(3)#9-P(3)-O(3)	113.4(4)
O(2)#6-In(2)-O(2)#1	92.1(4)	O(3)#9-P(3)-O(3)#10	113.4(4)
O(2)#7-In(2)-O(2)#1	87.1(4)	O(3)-P(3)-O(3)#10	113.4(4)
O(2)#4-In(2)-O(2)#1	90.5(3)	P(1)-O(1)-In(1)	138.3(5)
O(2)#8-In(2)-O(2)#1	176.5(5)	P(1)-O(2)-In(2)	132.5(5)
O(2)#6-In(2)-O(2)	87.1(4)	P(3)-O(3)-In(3)	148.7(6)
O(2)#7-In(2)-O(2)	176.5(5)	P(2)-O(4)-In(3)#10	135.4(5)
O(2)#4-In(2)-O(2)	90.5(3)	P(2)-O(5)-In(3)#5	135.9(5)
O(2)#8-In(2)-O(2)	92.1(4)	P(2)-O(6)-In(3)	131.7(5)
O(2)#1-In(2)-O(2)	90.5(3)	P(1)-O(7)-In(3)	137.1(5)
O(3)-In(3)-O(4)#9	96.9(4)		

Symmetry transformations used to generate equivalent atoms:

#1 -y,x-y,z #2 -x,-y,-z #3 y,-x+y,-z #4 -x+y,-x,z #5 x-y,x,-z #6 y,x,-z+1/2 #7 -x,-x+y,-z+1/2

#8 x-y,-y,-z+1/2 #9 -y+1,x-y,z #10 -x+y+1,-x+1,z #11 -y,-x,z+1/2 #12 -y,-x,z-1/2

Table S5. Crystal data and structure refinement parameters for compound **T1**, $[C_4N_2H_{12}][In_4(H_2O_3)(HPO_3)_7]$

	T1
Empirical formula	$CH_{4.33}In_{1.33}N_0O_8P_{2.33}$
Formula weight	369.73
Crystal system	Trigonal
Space group	*P*-3*c*1 (no.165)
Crystal size (mm)	0.16 × 0.14 × 0.08
a (Å)	13.8684(6)
b(Å)	13.8684(6)
c(Å)	18.6368(16)
α(°)	90.0
β(°)	90.0
γ(°)	120.0
Volume ($Å^3$)	3104.2(3)
Z	12
Temperature(K)	293
ρ_{calcd} (gcm^{-3})	2.373
μ (mm^{-1})	3.385
Wavelength (Å)	0.71073
θ range (deg)	1.70 to 28.05
Reflection collected	23980
Unique reflections	2449
Number of parameters	134
Goodness of fit	1.162
R index [I>2σ(I)]	R_1 = 0.0672,
	wR_2 = 0.1516
R (all data)	R_1 = 0.0815,
	wR_2 = 0.1600

$R_1 = \Sigma|| F_o | - | F_c || / \Sigma | F_o |$; $wR_2 = \{\Sigma [w (F_o^2 - F_c^2)]/\Sigma [w (F_o^2)^2]\}^{1/2}$. $w = 1/[\rho^2(F_o)^2+(aP)^2+bP]$. $P = [\max (F_o, O)+2(F_c)^2]/3$, where a = 0.0728and b = 41.822 .

CHN analysis: %C 4.13(calcd.) 4.02(obsd); %N 2.41(calcd) 2.13(obsd); %H 2.15(calcd) 2.02(obsd.)

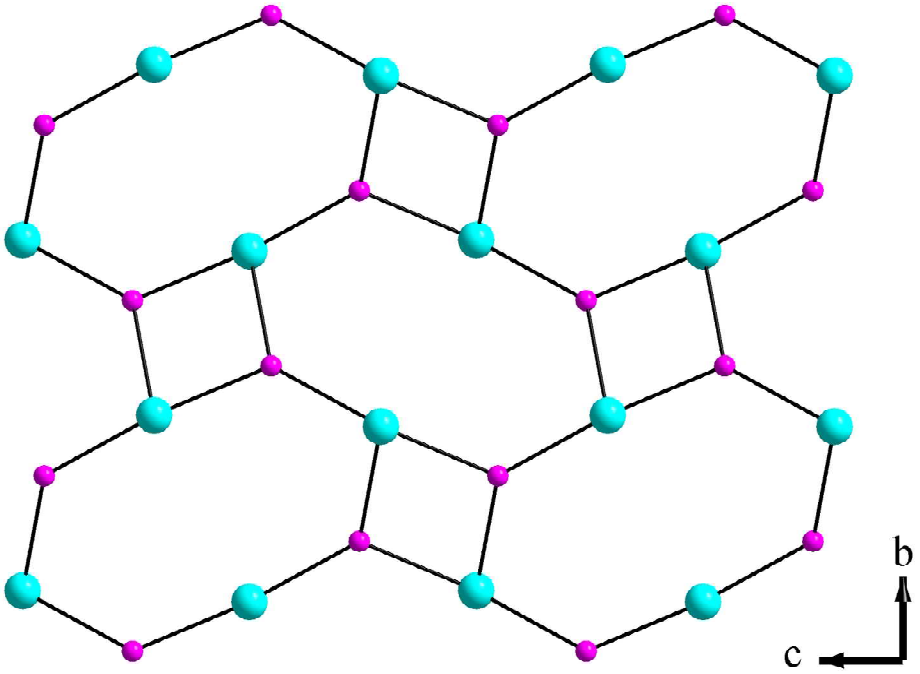
Table S6. Phosphorous-oxygen bond lengths and the calculated bond strengths (S) at phosphorous and at oxygen in the compounds **I – VIII**.

Bond	**Bond length (Å)**	**S(P^{3+}) (v.u)**	**S(O^{2-}) (v.u)**
		Compound I	
P(1) – O(1)	1.519	1.301	1.717
P(1) – O(2)	1.520	1.299	2.118
P(1) – O(7)	1.507	1.117	1.117
			$\Sigma S(O^{2-}) = 4.952$
		Compound II	
P(1) – O(1)	1.494	1.394	2.055
P(1) – O(2)	1.497	1.383	2.017
P(1) – O(3)	1.486	1.426	2.037
			$\Sigma S(O^{2-}) = 6.109$
		Compound IV	
P(1) – O(2)	1.511	1.508	1.869
P(1) – O(3)	1.514	1.539	1.754
P(1) – O(10)	1.521	1.521	1.854
			$\Sigma S(O^{2-}) = 5.477$
P(2) – O(8)	1.519	1.301	1.892
P(2) – O(1)	1.525	1.284	1.847
P(2) – O(5)	1.531	1.262	1.774
			$\Sigma S(O^{2-}) = 5.512$
P(3) – O(9)	1.516	1.315	1.893
P(3) – O(13)	1.517	1.312	1.312
P(3) – O(7)	1.523	1.288	1.888
			$\Sigma S(O^{2-}) = 5.093$
		Compound V	
P(1) – O(4)	1.499	1.376	2.165
P(1) – O(8)	1.502	1.364	2.182
P(1) – O(9)	1.520	1.298	1.818
			$\Sigma S(O^{2-}) = 6.165$
P(2) – O(12)	1.511	1.332	1.912
P(2) – O(3)	1.517	1.309	1.839
P(2) – O(2)	1.527	1.277	1.829

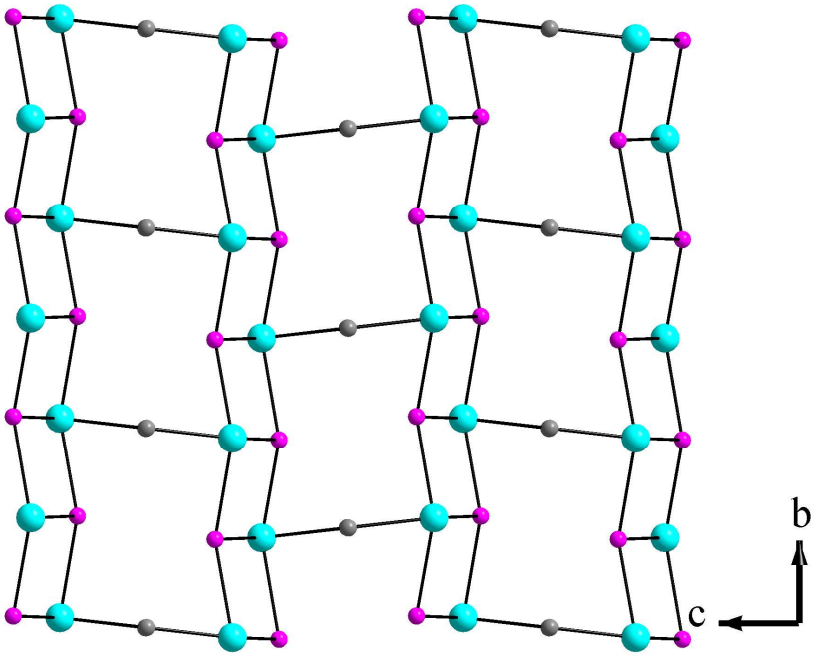
			$\Sigma S(O^{2-}) = 5.644$
P(3) – O(13)	1.491	1.406	1.406
P(3) – O(7)	1.538	1.239	3.915
P(3) – O(1)	1.540	1.231	1.847
			$\Sigma S(O^{2-}) = 7.168$
		Compound VI	
P(1) – O(21)	1.513	1.326	1.9106
P(1) – O(19)	1.513	1.326	1.9684
P(1) – O(15)	1.513	1.326	1.9054
			$\Sigma S(O^{2-}) = 5.784$
P(2) – O(10)	1.512	1.327	1.902
P(2) – O(14)	1.519	1.303	1.915
P(2) – O(3)	1.533	1.255	1.802
			$\Sigma S(O^{2-}) = 5.619$
P(3) – O(10)	1.512	1.327	1.868
P(3) – O(14)	1.519	1.305	1.873
P(3) – O(3)	1.522	1.292	1.857
			$\Sigma S(O^{2-}) = 5.598$
P(4) – O(10)	1.502	1.363	1.981
P(4) – O(14)	1.504	1.358	1.934
P(4) – O(3)	1.518	1.308	1.911
			$\Sigma S(O^{2-}) = 5.826$
P(5) – O(10)	1.435	1.635	2.209
P(5) – O(14)	1.446	1.587	2.199
P(5) – O(3)	1.503	1.359	2.199
			$\Sigma S(O^{2-}) = 6.607$
		Compound VIII	
P(1) – O(8)	1.449	1.573	1.573
P(1) – O(9)	1.456	1.545	1.545
P(1) – O(2)	1.449	1.395	1.928
P(1) – O(1)	1.509	1.339	1.871
			$\Sigma S(O^{2-}) = 6.917$

Table S7. Assignment of the observed NMR peaks for the compounds **I – VIII** using the bond strength values.

Compound	NMR peak position (ppm)	Bond strength values	Assignment
I	0.157	4.952	P(1)
II	-9.140	6.109	P(1)
IV	1.842	5.512	P(2)
	5.251	5.477	P(1)
	6.481	5.093	P(3)
V	-6.419	7.168	P(3)
	-3.981	6.165	P(1)
	0.833	5.644	P(2)
VI	-18.871	6.607	P(5)
	-17.422	5.826	P(4)
	6.173	5.784	P(1)
	7.144	5.619	P(2)
	8.110	5.598	P(3)
VIII	-20.914	6.917	P(1)



(a)



(b)

Fig. S1. Figures showing the T-atom connectivity in (a) compound II and (b) compound III.

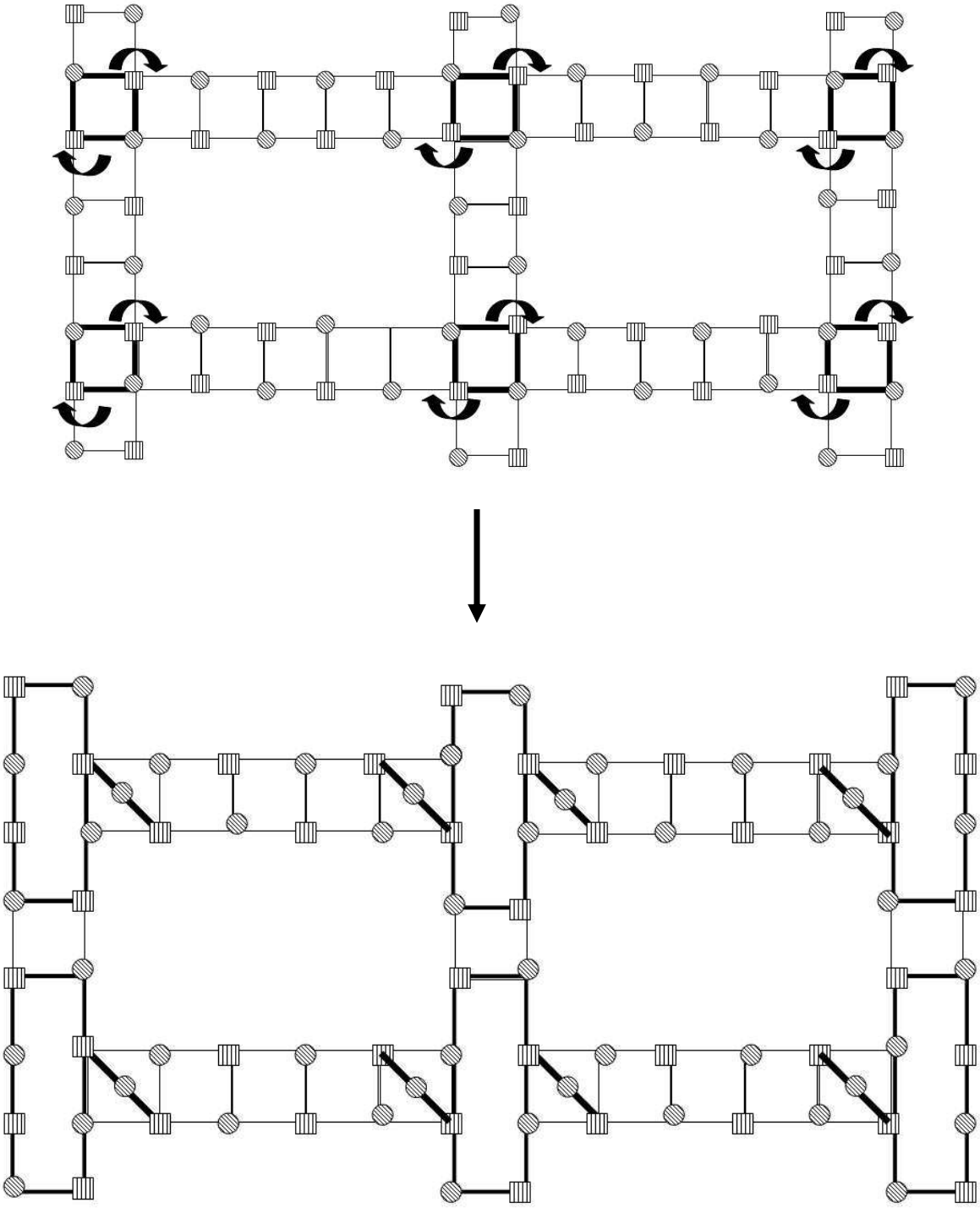


Fig. S2. Schematic visualization of the formation of compound VI

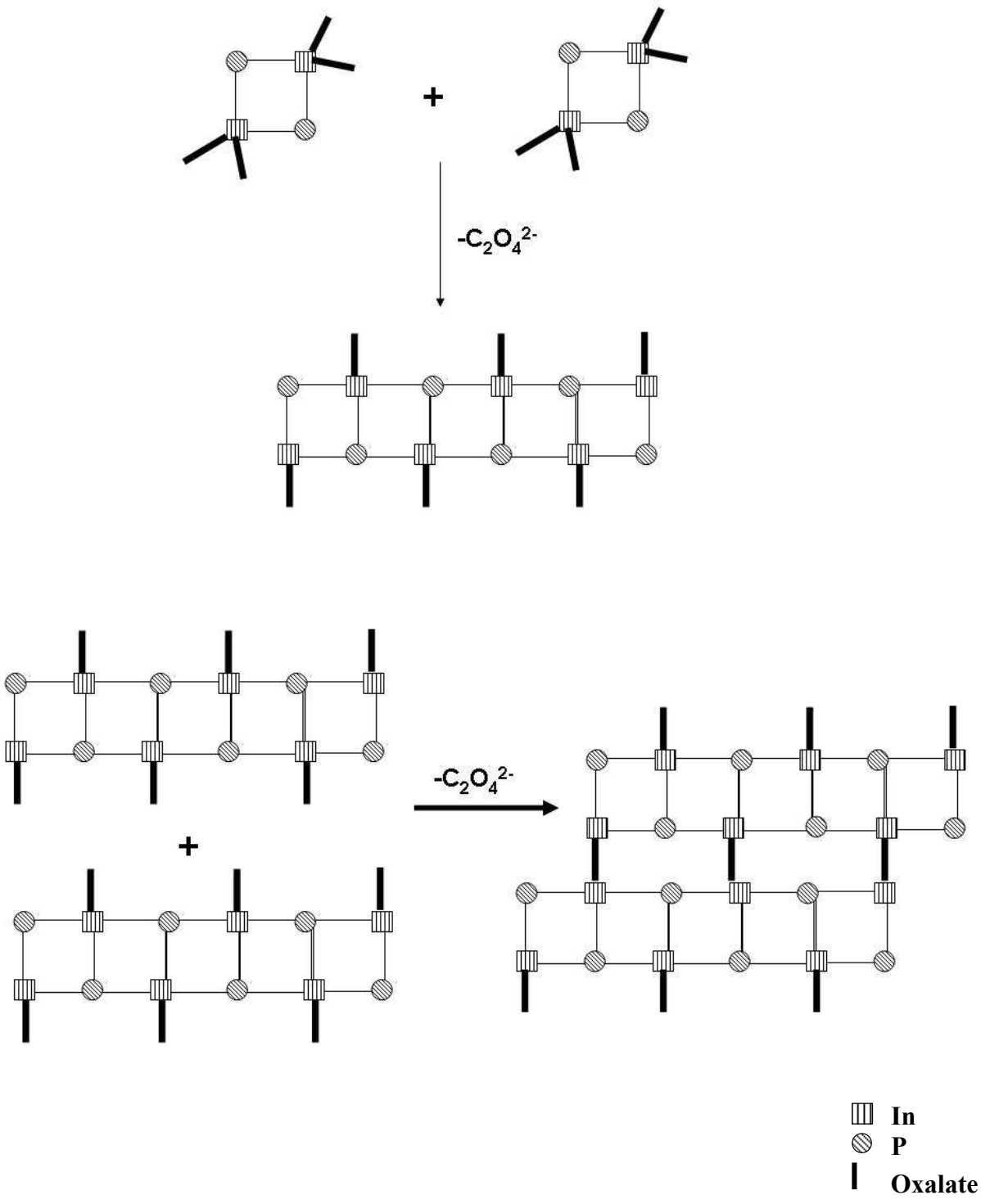


Fig. S3. Schematic pathway for the formation of compound III from the zero-dimensional compound I

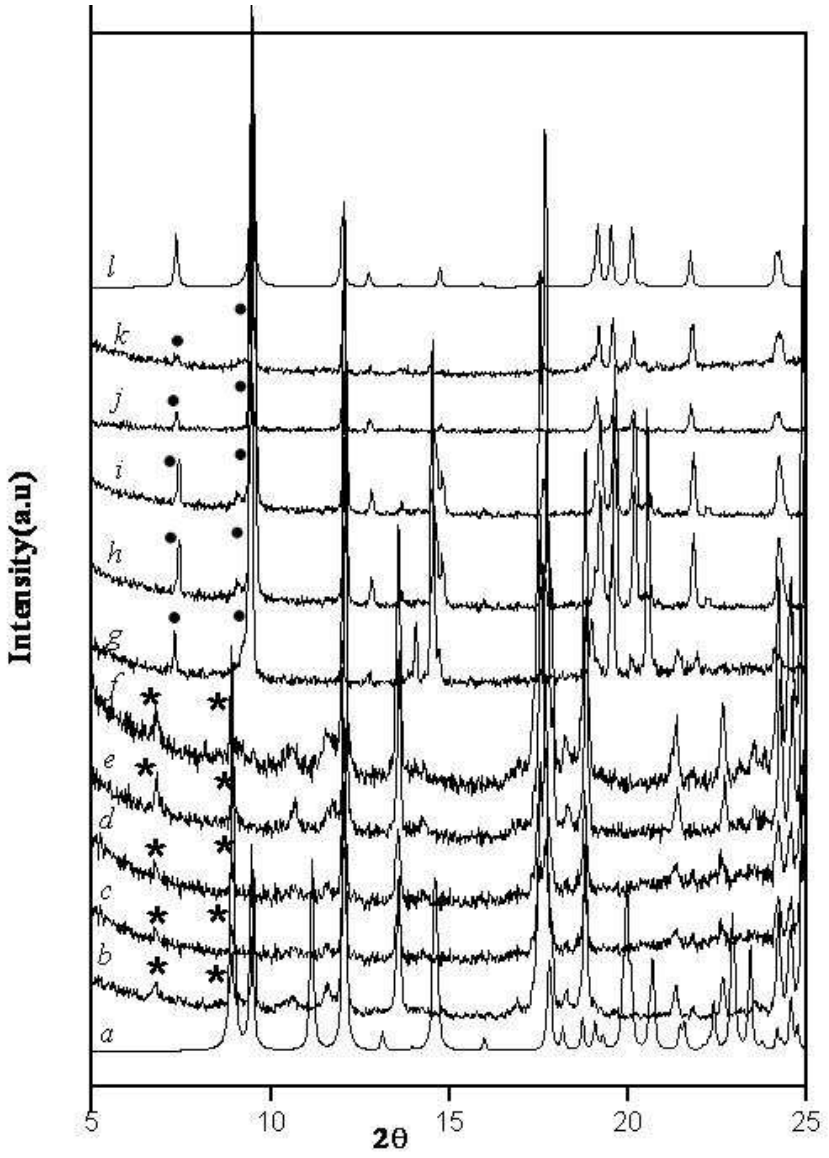
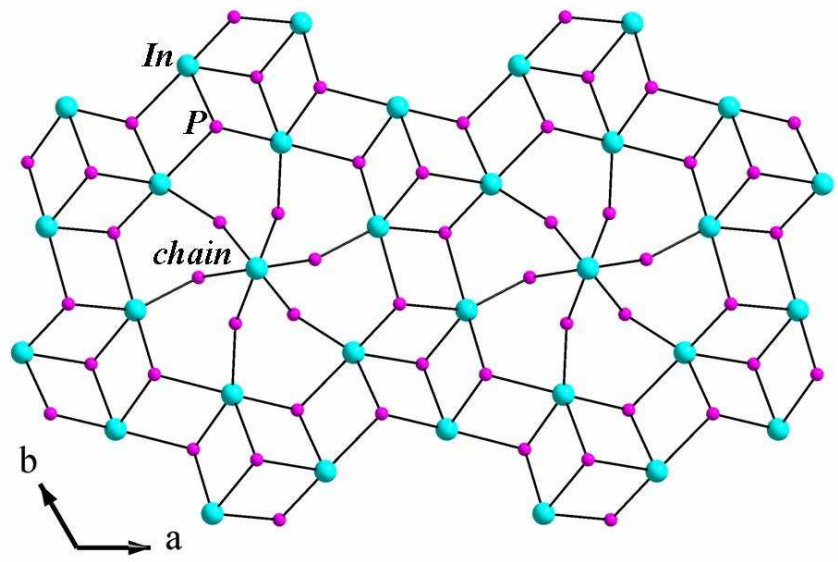
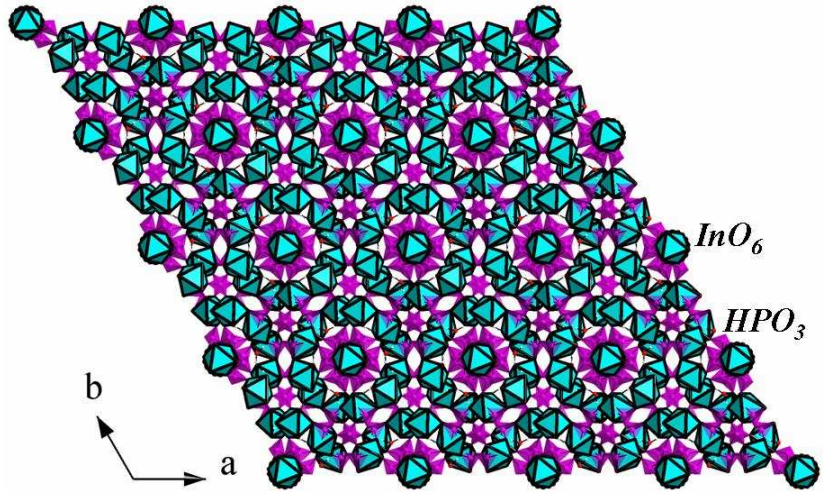


Fig.S4.Time dependent powder X-ray diffraction patterns of the product of the transformation reaction of the zero-dimensional molecular compound $[C_4N_2H_{12}]_3[In_2(HPO_3)_2(C_2O_4)_4].4H_2O$, I to the three-dimensional indium phosphite, $[C_4N_2H_{12}][In_4(H_2O)_3(HPO_3)_7]$, T1. (a) simulated pattern of I; Experimental pattern of I + phosphorous acid: (b) after 1 hour, (c) after 3 hours, (d) after 5 hours, (e) after 6 hours, (f) after 12 hours, (g) after 18 hours, (h) after 24 hours, (i) after 36 hours (j) after 42 hours (k) after 48 hours (l) simulated pattern of compound T1.



(a)



(b)

Fig. S5. (a) The T-atom connectivity between the layer and chain in T1;
(b) Polyhedral view of the three-dimensional structure along the *c*-axis.

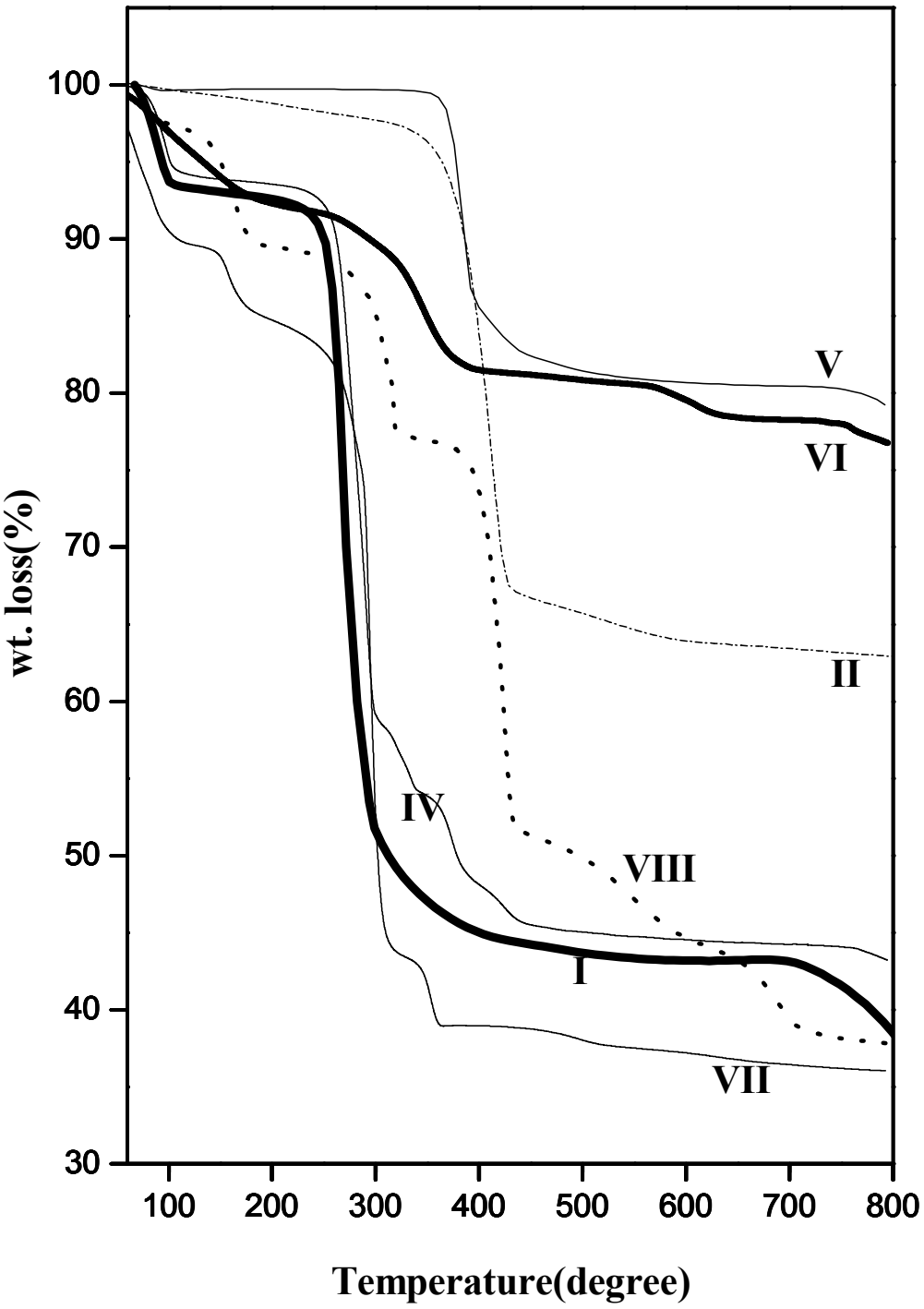


Fig. S6. Thermogravimetric analysis curves of all the pure compounds

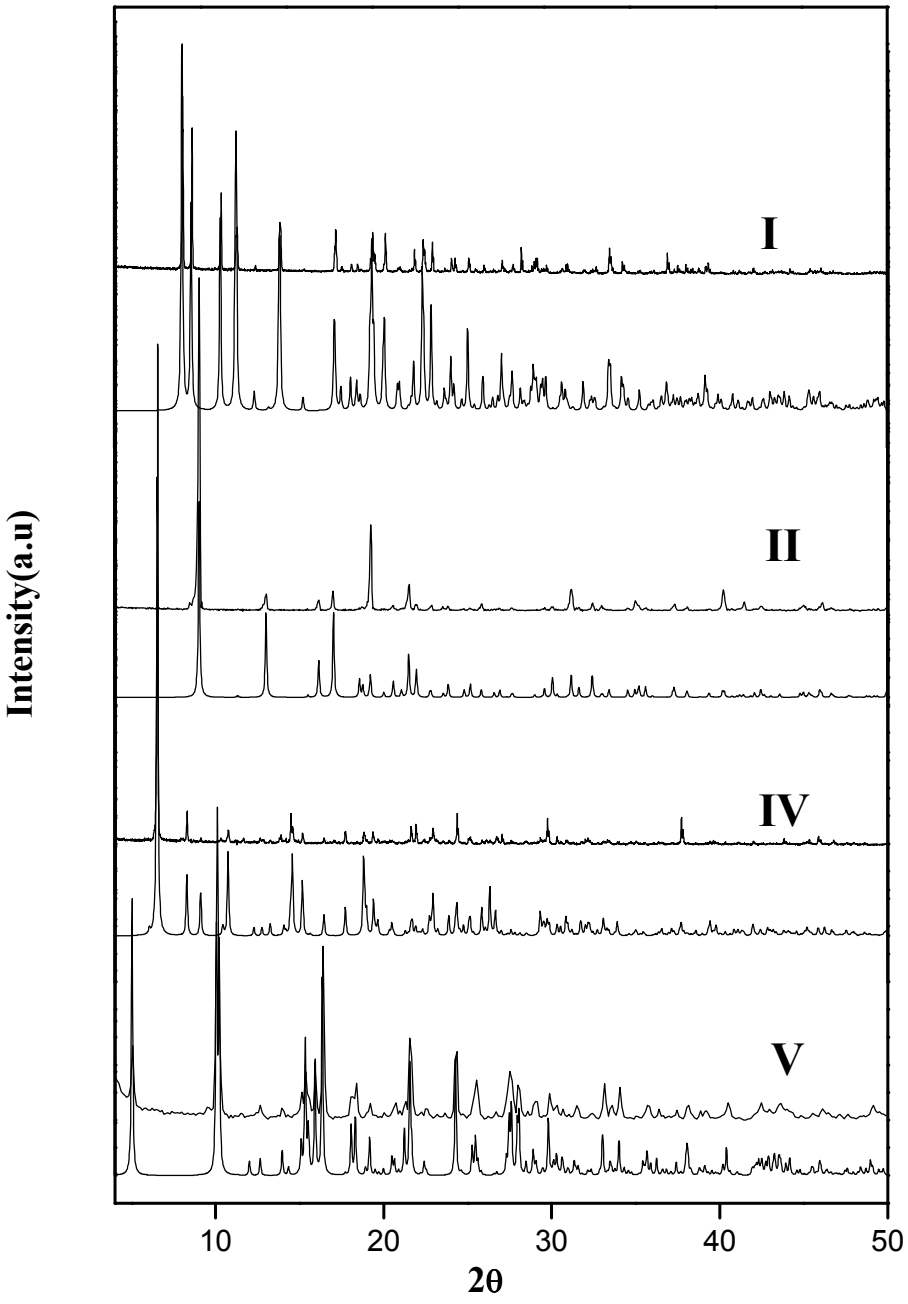


Fig.S7(a). Simulated and experimental XRD patterns of all the pure compounds I-V

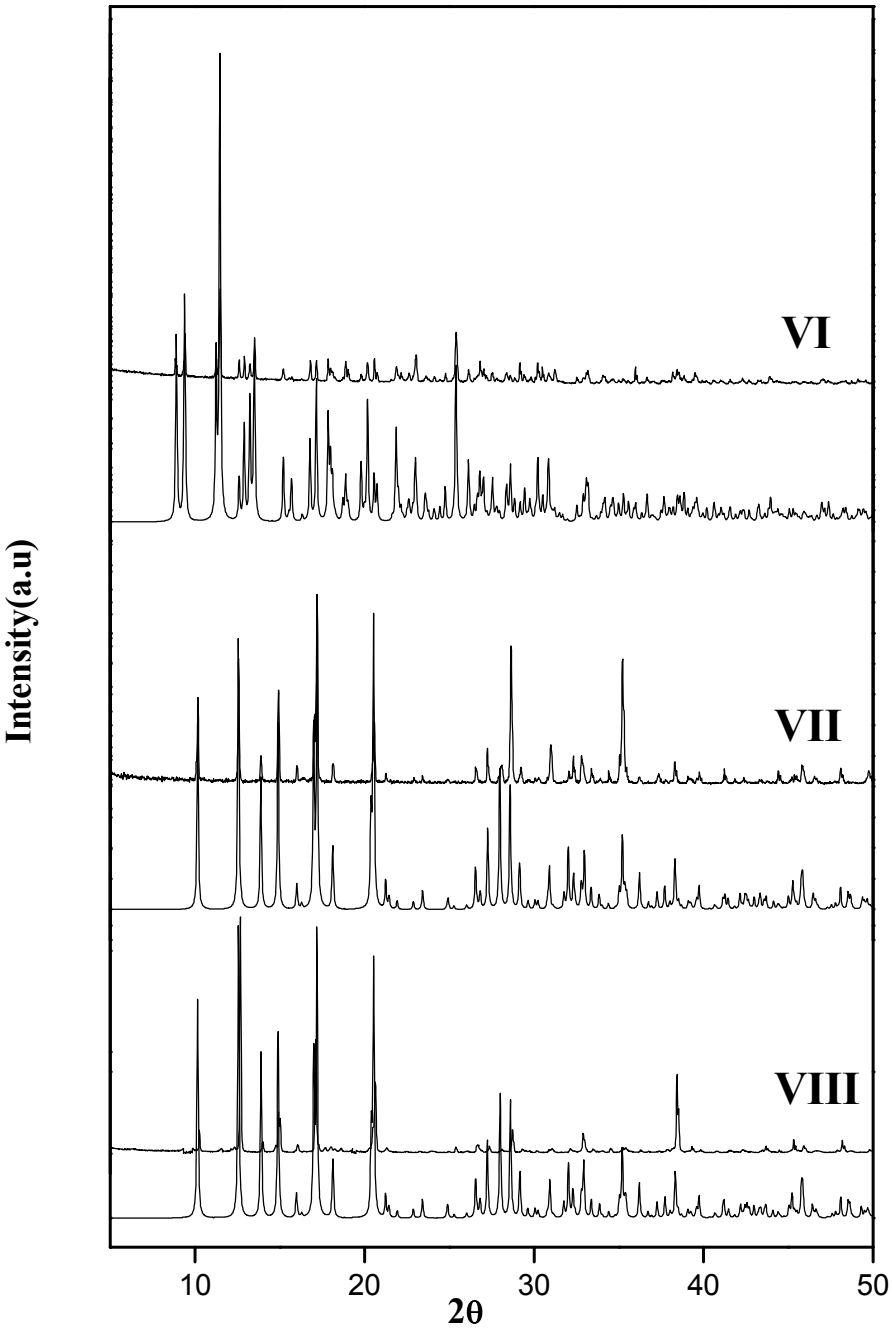


Fig.S7(b). Simulated and experimental XRD patterns of all the pure compounds VI-VIII

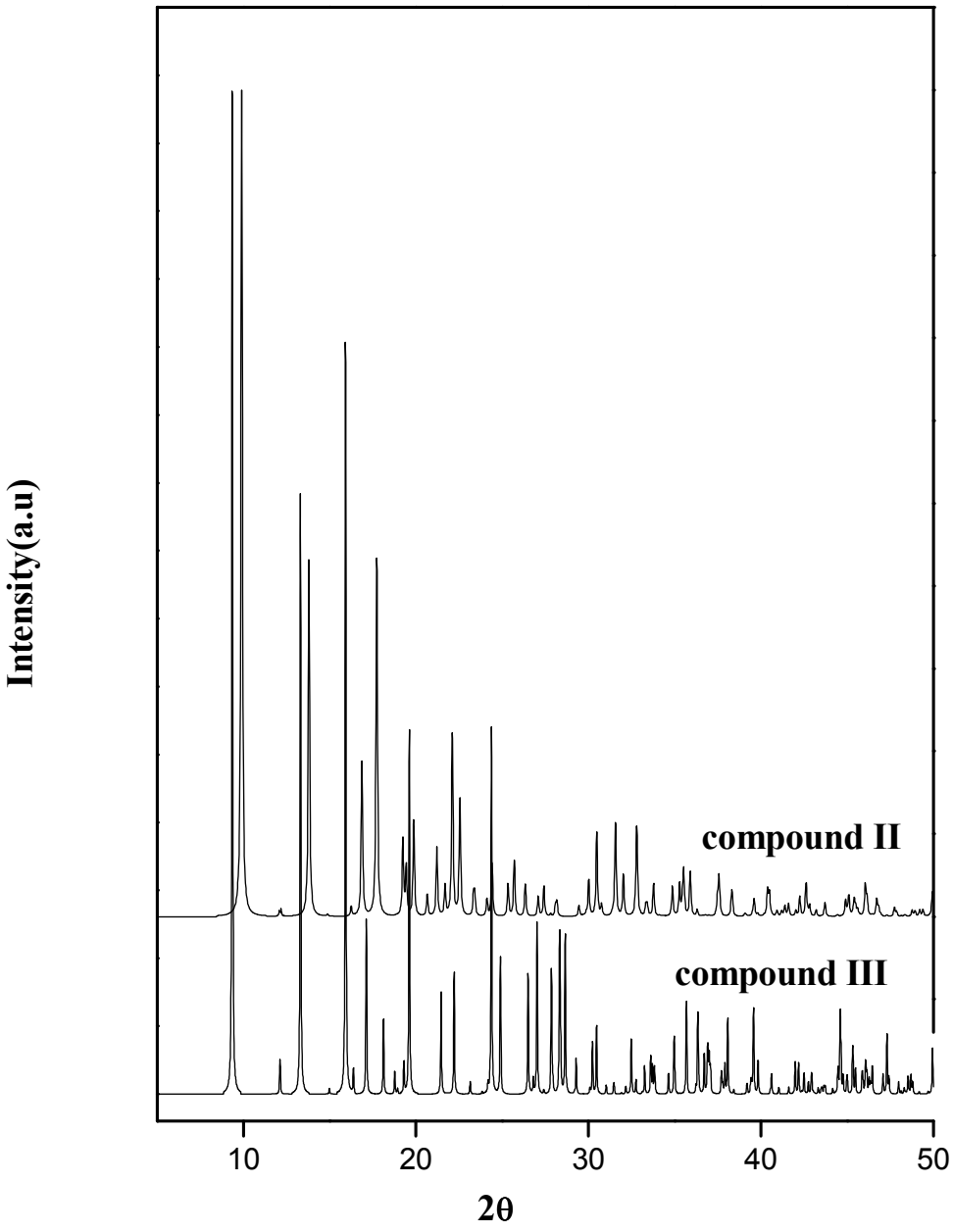


Fig.S7(c). Simulated XRD patterns of the polymorphic phases II and III.

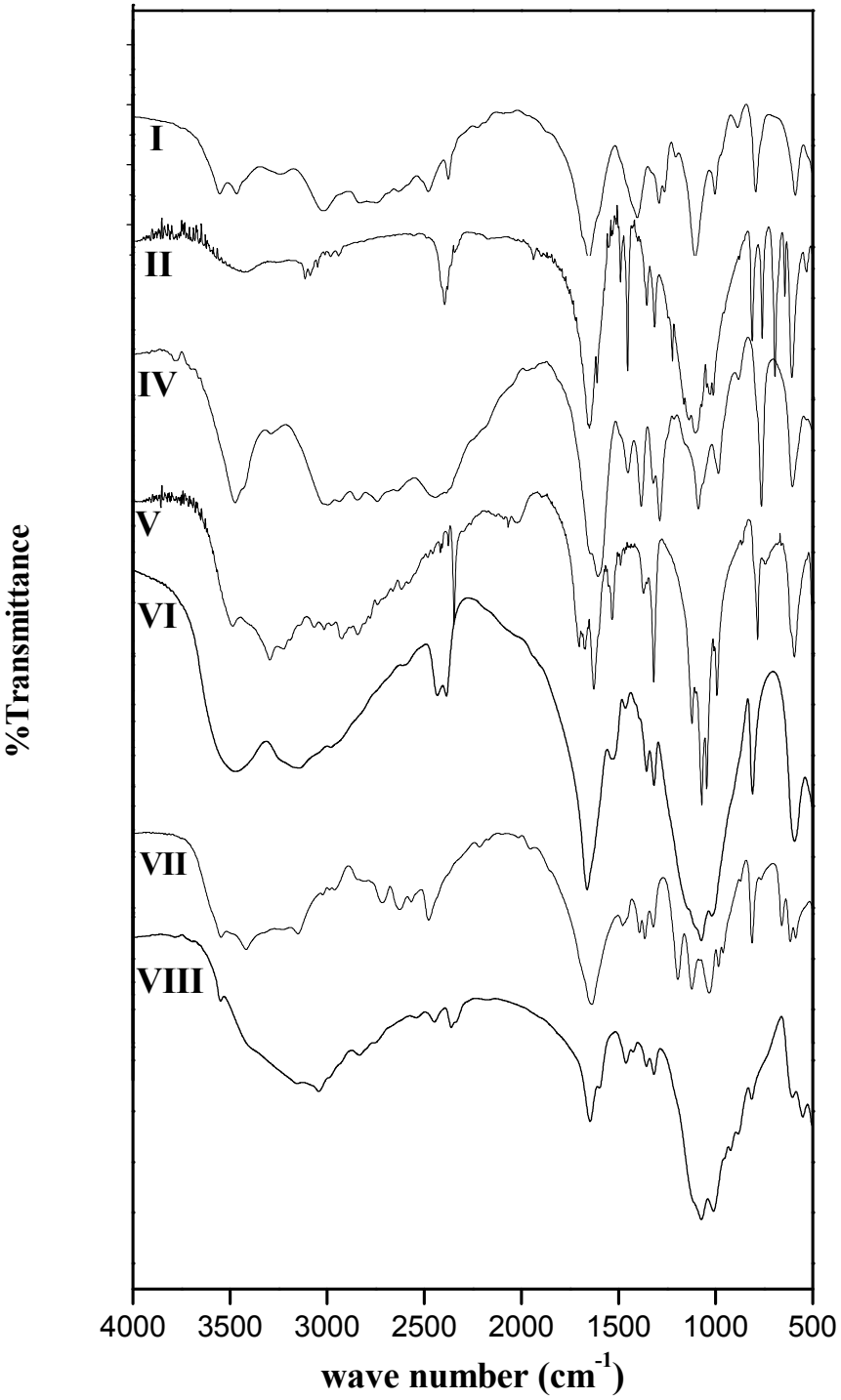
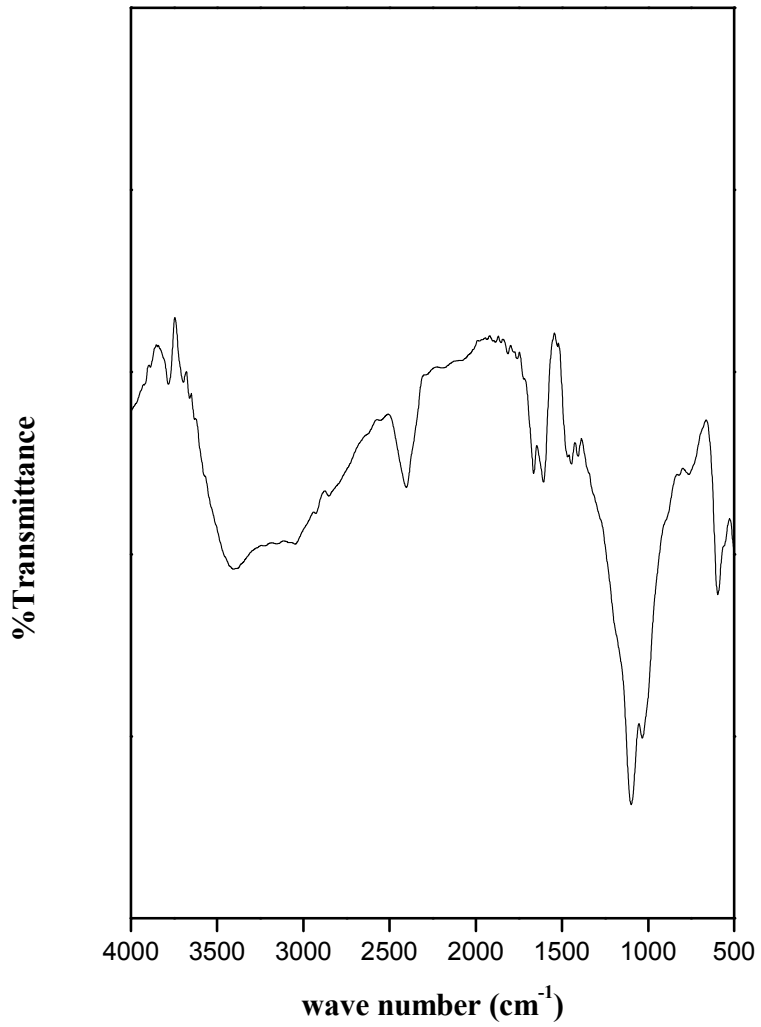
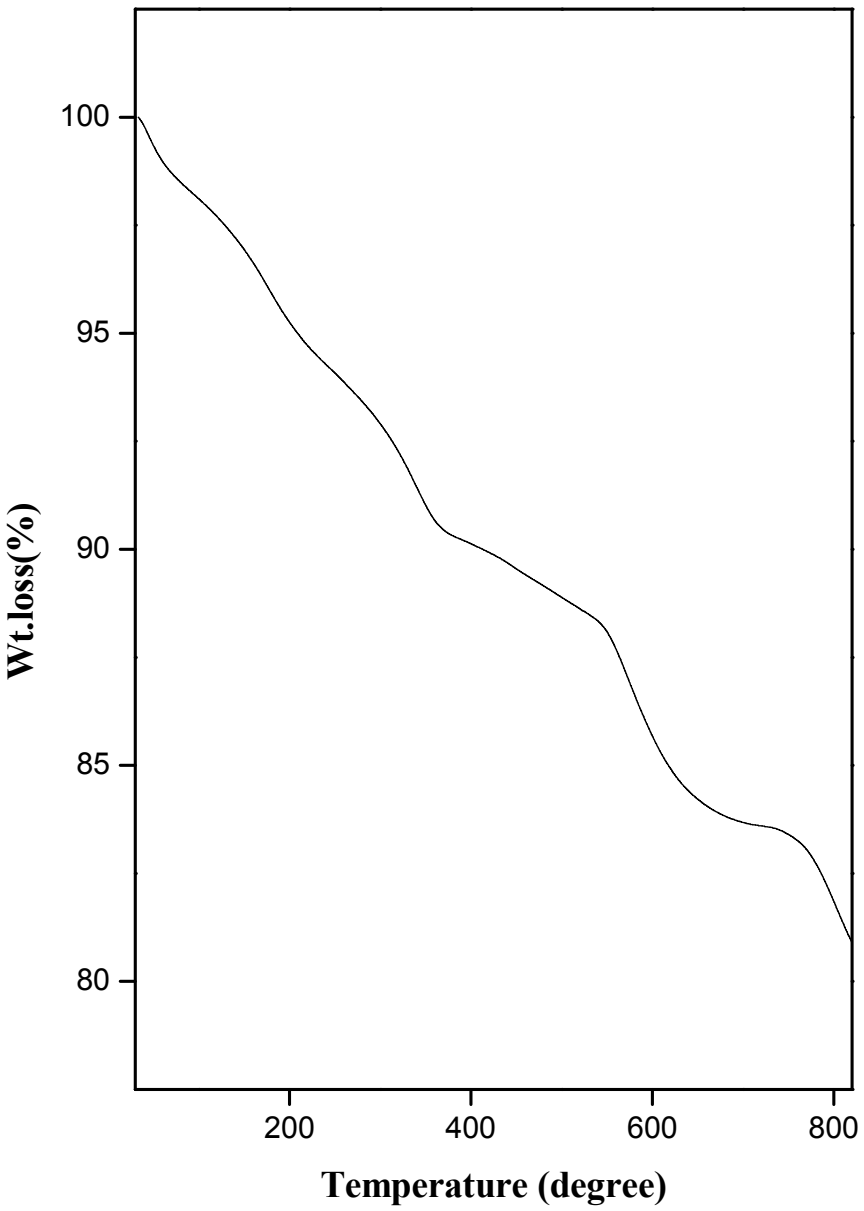


Fig. S8. Infra-red spectra of all the pure compounds



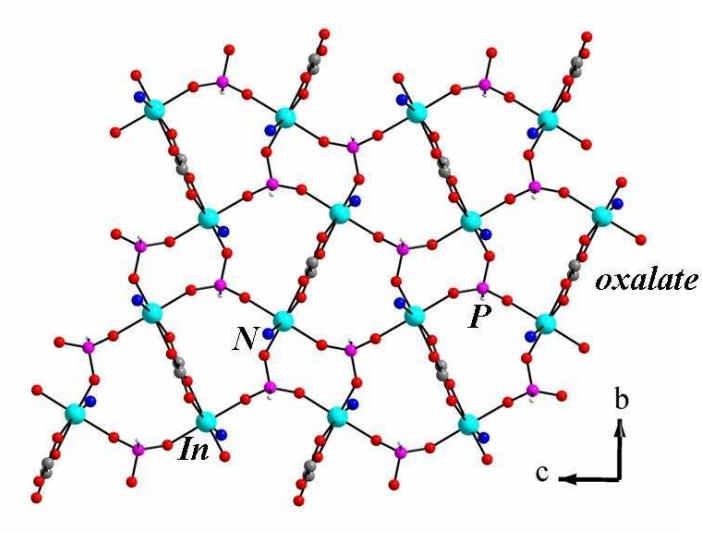
δas(P-H): 1087 cm^{-1}; γ(P-H): 2397 cm^{-1}; δas(PO_3):477 cm^{-1};
γ_s(PO_3):1020 cm^{-1}; γ_{as}(PO_3):1107 cm^{-1}; γ(N-H):3415 cm^{-1};
γas(C-H):3037 cm^{-1}; γ_s(C-H):2843 cm^{-1};γ_s(H_2O):3776cm^{-1};

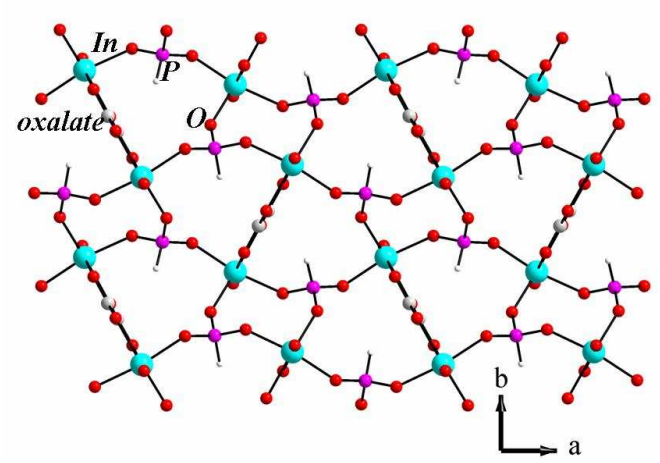
Fig. S9. Infra-red spectrum of the compound T1



Calculated weight loss due to decomposition of amine and loss of water: 12.23%; calculated weight gain due to oxidation of P^{III} to P^{V}: 9.6%. Total weight loss: 12.23 + 9.6 = 21.83%
Observed weight loss: 19.67%
Final decomposed product: $InPO_4$ (JCPDS: 08-0052)

Fig. S10. TGA curve for the compound T1





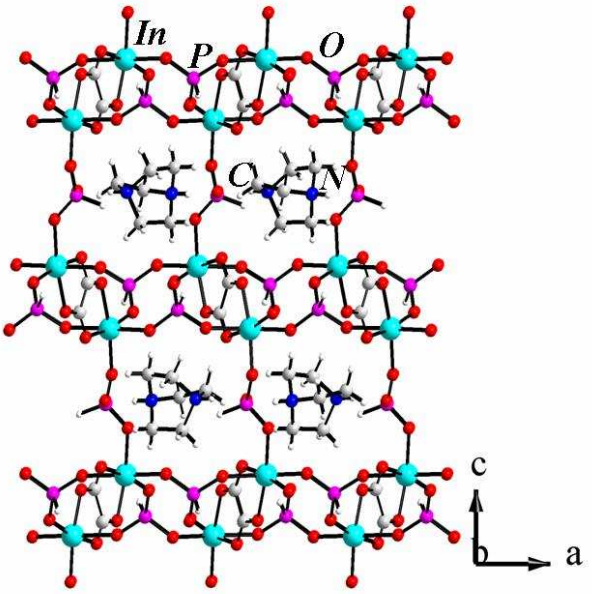
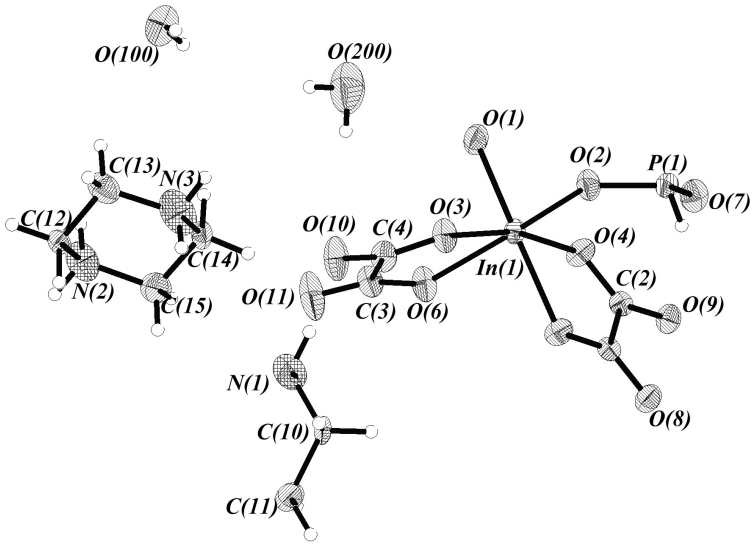
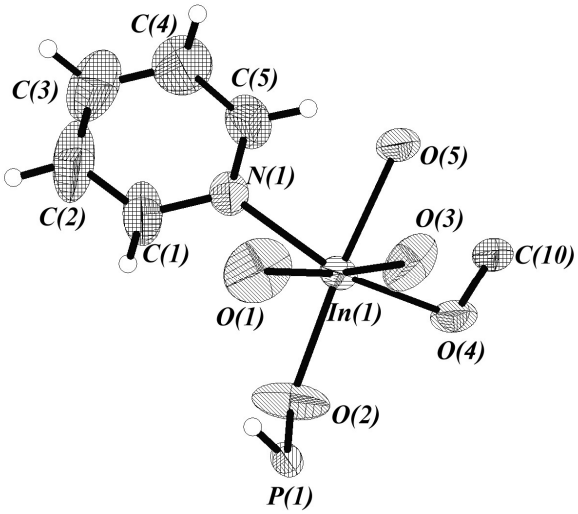


Fig. S11. The layer structures in (a) $[In_2(HPO_3)_2(C_2O_4)(C_5H_5N)_2]$, II, and (b) $[C_6H_{14}N_2][In_2(HPO_3)_3(C_2O_4)]$ (Ref.23);

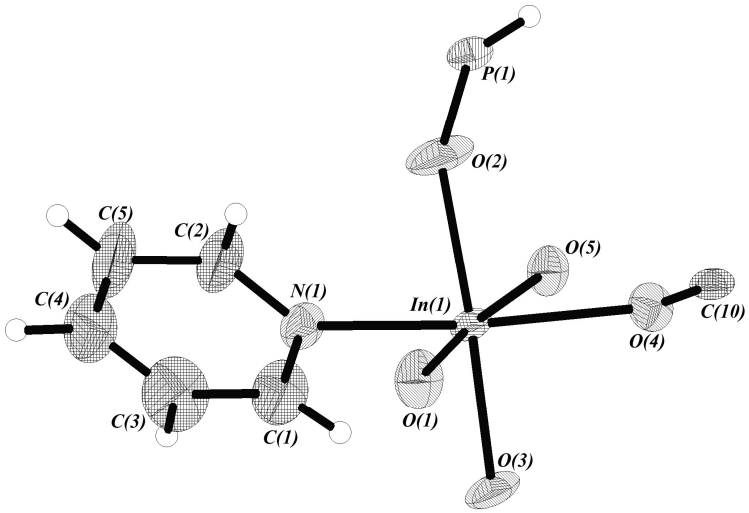
(c) The three-dimensional structure of $[C_6H_{14}N_2][In_2(HPO_3)_3(C_2O_4)]$(Ref.23)



(a) Compound I

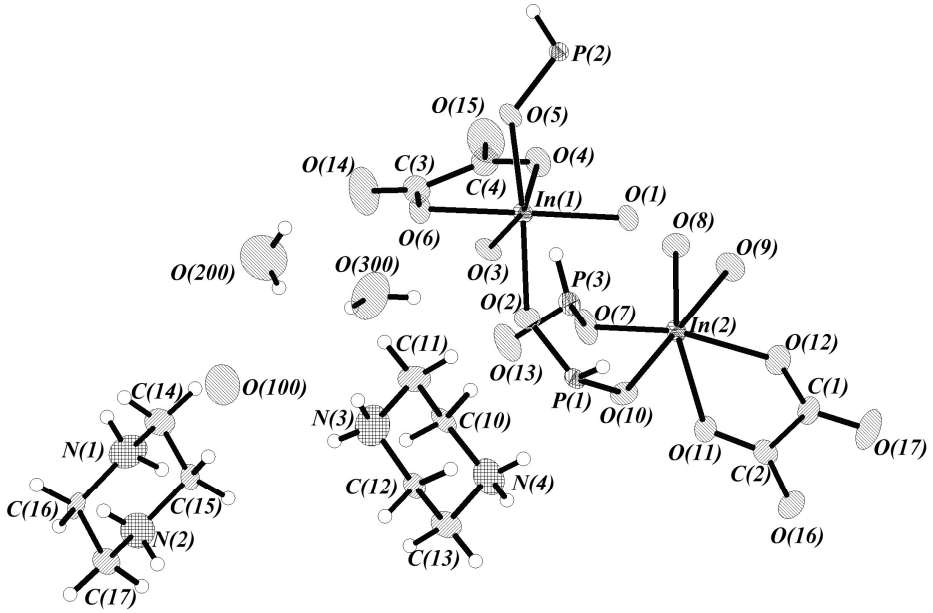


(a) Compound II

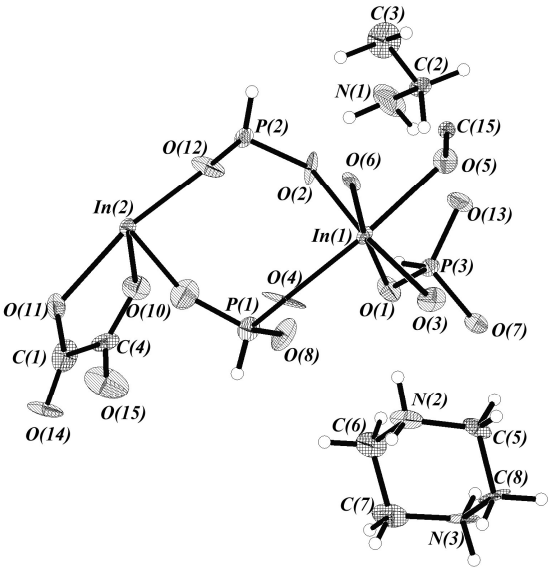


(a) Compound III

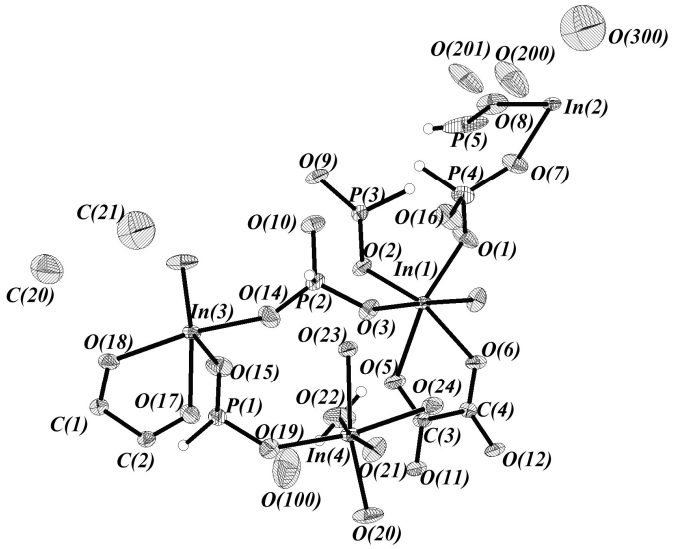
Fig. S12. ORTEP plots for compounds I-III



(a) Compound IV

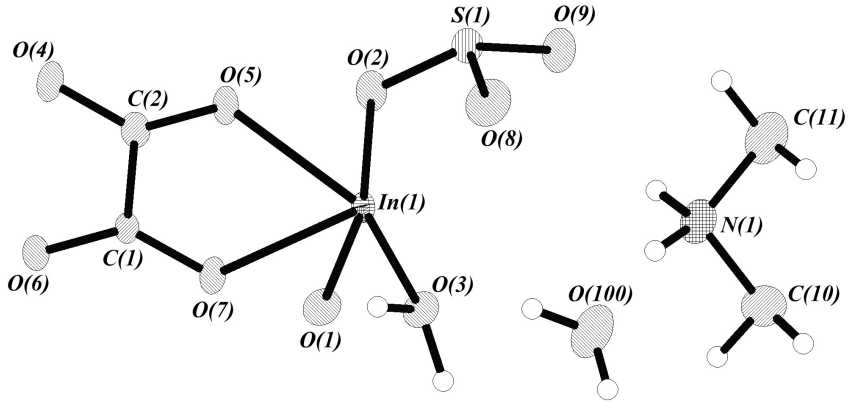


(b) Compound V

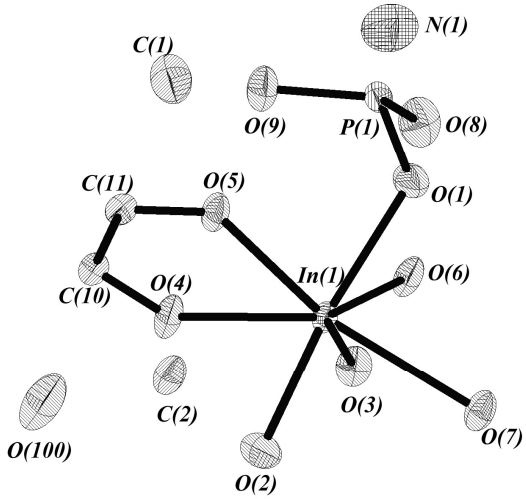


(b) Compound VI

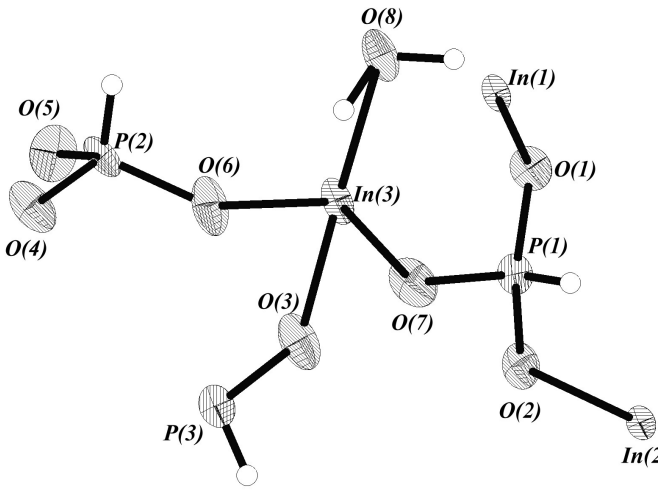
Fig. S13. ORTEP plots for compounds IV-VI



(a) Compound VII



(b) Compound VIII



(c) Compound T1

Fig. S14. ORTEP plots for compounds VII-VIII, T1.

According to Boltzman distribution, population of any species at a given temperature is proportional to exp(-E/kT), where:

E is the energy of the species,

k is the Boltzman constant and

T is the temperature in Kelvin.

So, N(I)=w*exp(-E(pr475h)/kT).....(1)

where w is just the proportionality const.

and,N(II)=w*exp(-E(pr475)/kT) .....(2)

where w is just the proportionality const.

Here, N(I) and N(II) are the populations

of the two clusters.

Taking ratio of (1) and (2),

N(I)/N(II)=exp[-(E(I)-E(II))/kT)]

Substituting, T=453 K, k=0.0000735 eV/mol and

(E(I)-E(II)=-0.88 eV/mol, we get the

number as 3.0088*10^11 .