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Table S1. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement coefficients ($\text{\AA}^2 \times 10^3$) for $[\text{HB}(\text{pz})_3]_4\text{V}_4(\mu\text{-C}_6\text{H}_5\text{OPO}_3)_4 \cdot 4\text{CH}_3\text{CN}$.

Atom	x	y	z	U _{eq} *
V(1)	5323(1)	7189(1)	1116(1)	34(1)
V(2)	7569(1)	8839(1)	1767(1)	37(1)
V(3)	8016(1)	5914(1)	2455(1)	38(1)
V(4)	5564(1)	7919(1)	3803(1)	32(1)
P(1)	5428(1)	8773(1)	2166(1)	31(1)
P(2)	7512(1)	7066(1)	1017(1)	36(1)
P(3)	7727(1)	7696(1)	3238(1)	35(1)
P(4)	5800(1)	6330(1)	2711(1)	33(1)
O(1)	5576(2)	6379(2)	1905(2)	39(1)
O(2)	6855(2)	7633(2)	3610(2)	42(1)
O(3)	6554(2)	7260(2)	864(2)	43(1)
O(4)	6766(2)	6080(2)	2819(2)	42(1)
O(5)	7839(2)	7745(2)	1354(2)	41(1)
O(6)	4765(2)	9588(2)	1951(2)	43(1)
O(7)	6291(2)	8917(2)	1825(2)	49(1)
O(8)	7720(2)	8464(2)	2795(2)	43(1)
O(9)	7790(2)	6286(2)	1432(2)	41(1)
O(10)	5354(2)	7058(2)	3157(2)	36(1)
O(11)	5115(2)	8092(2)	1819(2)	38(1)
O(12)	8170(2)	6959(2)	2800(2)	40(1)
O(13)	5370(2)	8734(2)	3009(2)	37(1)
O(14)	5422(2)	5576(2)	3067(2)	39(1)
O(15)	8313(2)	7755(2)	3927(2)	53(1)
O(16)	7969(2)	6932(2)	184(2)	49(1)
B(1)	4010(5)	7019(4)	-176(4)	60(3)
N(1)	5468(3)	6310(2)	271(2)	47(2)
N(2)	4827(3)	6343(3)	-224(2)	55(2)
C(11)	6121(4)	5711(3)	65(3)	60(2)
C(12)	5914(6)	5343(4)	-568(4)	79(3)
C(13)	5095(6)	5761(4)	-723(3)	73(3)
N(3)	3989(3)	7057(2)	1224(2)	44(2)
N(4)	3538(3)	6995(3)	606(3)	56(2)
C(14)	3429(4)	7015(3)	1817(3)	53(2)
C(15)	2613(4)	6928(4)	1588(4)	73(3)
C(16)	2722(4)	6920(4)	830(4)	76(3)
N(5)	4861(3)	8016(2)	269(2)	42(2)
N(6)	4327(3)	7823(3)	-247(2)	53(2)
C(17)	4983(4)	8758(3)	95(3)	52(2)
C(18)	4550(4)	9039(4)	-532(3)	66(3)
C(19)	4142(4)	8437(4)	-724(3)	67(3)
B(2)	8482(4)	10243(4)	1020(4)	55(3)
N(7)	8918(3)	8901(3)	1629(3)	48(2)
N(8)	9188(3)	9533(3)	1276(3)	57(2)
C(21)	9653(4)	8384(4)	1787(4)	69(3)
C(22)	10376(4)	8695(5)	1530(5)	96(4)
C(23)	10061(4)	9417(4)	1227(4)	83(3)
N(9)	7391(3)	10057(2)	2073(2)	45(2)
N(10)	7867(3)	10555(2)	1702(2)	47(2)
C(24)	6890(4)	10499(3)	2615(3)	59(2)

C(25)	7060(5)	11272(3)	2594(4)	73(3)
C(26)	7673(4)	11284(3)	2021(3)	60(2)
N(11)	7557(3)	9271(2)	666(2)	45(2)
N(12)	7941(3)	9906(3)	461(3)	51(2)
C(27)	7208(4)	9050(3)	60(3)	52(2)
C(28)	7360(4)	9523(4)	-542(3)	67(3)
C(29)	7824(4)	10046(4)	-271(3)	66(3)
B(3)	9353(4)	4263(4)	2834(4)	56(3)
N(13)	9376(3)	5553(3)	2137(2)	48(2)
N(14)	9838(3)	4834(3)	2381(3)	54(2)
C(31)	9956(4)	5927(3)	1766(3)	57(2)
C(32)	10798(4)	5457(4)	1760(4)	70(3)
C(33)	10697(4)	4780(4)	2153(4)	70(3)
N(15)	8397(3)	5427(2)	3510(2)	47(2)
N(16)	8968(3)	4708(3)	3564(3)	53(2)
C(34)	8219(4)	5704(4)	4201(3)	59(2)
C(35)	8662(5)	5186(4)	4706(4)	81(3)
C(36)	9129(5)	4565(4)	4277(4)	76(3)
N(17)	7957(3)	4726(2)	2188(2)	46(2)
N(18)	8582(3)	4097(2)	2410(2)	51(2)
C(37)	7341(4)	4411(3)	1894(3)	54(2)
C(38)	7539(4)	3587(3)	1930(3)	69(3)
C(39)	8323(4)	3415(3)	2257(3)	64(2)
B(4)	4678(5)	8217(4)	5486(3)	57(3)
N(19)	5593(3)	7102(2)	4704(2)	41(2)
N(20)	5205(3)	7343(3)	5386(2)	50(2)
C(41)	5941(4)	6330(3)	4767(3)	52(2)
C(42)	5790(5)	6062(4)	5487(3)	71(3)
C(43)	5322(4)	6724(4)	5865(3)	68(3)
N(21)	5775(3)	8727(2)	4611(2)	42(2)
N(22)	5307(3)	8783(3)	5291(2)	48(2)
C(44)	6329(4)	9231(3)	4623(3)	54(2)
C(45)	6224(5)	9616(3)	5307(4)	72(3)
C(46)	5590(5)	9323(4)	5705(3)	69(3)
N(23)	4223(3)	8244(2)	4173(2)	41(2)
N(24)	3963(3)	8337(3)	4912(2)	52(2)
C(47)	3484(4)	8390(3)	3815(3)	54(2)
C(48)	2758(4)	8587(4)	4325(4)	72(3)
C(49)	3087(4)	8542(4)	5005(4)	67(3)
C(101)	3864(4)	9716(3)	2169(3)	54(2)
C(102)	3327(4)	9208(4)	1984(4)	64(3)
C(103)	2448(5)	9377(6)	2207(5)	98(4)
C(104)	2109(7)	10071(8)	2594(5)	132(6)
C(105)	2601(7)	10588(7)	2773(5)	123(5)
C(106)	3507(5)	10403(5)	2556(4)	91(3)
C(201)	8877(4)	6793(3)	25(3)	52(2)
C(202)	9346(4)	7385(4)	124(3)	70(3)
C(203)	10240(5)	7244(5)	-80(4)	98(4)
C(204)	10656(5)	6509(6)	-381(4)	101(4)
C(205)	10173(5)	5934(5)	-484(4)	92(3)
C(206)	9280(5)	6067(4)	-275(3)	73(3)
C(301) #	9227(6)	7747(8)	3798(8)	74(4)
C(302) #	9504(7)	8464(7)	3770(6)	108(5)
C(303) #	10416(9)	8471(9)	3610(9)	152(8)

C(304) #	10992 (8)	7758 (10)	3483 (8)	135 (7)
C(305)	10691 (6)	7083 (7)	3511 (7)	155 (6)
C(306)	9807 (4)	7056 (5)	3614 (5)	101 (4)
C(307) #	9133	7606	3947	127 (39)
C(308) #	9447	8115	4419	92 (13)
C(309) #	10352	8024	4493	164 (24)
C(310) #	10942	7425	4095	107 (16)
C(402)	4238 (4)	5001 (3)	2726 (3)	57 (2)
C(403)	3370 (5)	4930 (4)	2800 (4)	78 (3)
C(404)	2786 (5)	5411 (4)	3287 (4)	76 (3)
C(405)	3076 (4)	5979 (4)	3702 (4)	67 (3)
C(406)	3957 (4)	6054 (3)	3629 (3)	52 (2)
C(401)	4525 (3)	5556 (3)	3136 (3)	39 (2)
N(50)	2108 (9)	3285 (8)	3107 (9)	251 (9)
C(501)	2273 (7)	2930 (6)	3656 (7)	132 (5)
C(502)	2543 (7)	2504 (6)	4300 (5)	131 (5)
N(51)	7659 (10)	2822 (6)	4002 (7)	233 (8)
C(503)	7234 (8)	3437 (6)	4036 (6)	133 (5)
C(504)	6709 (6)	4190 (5)	4044 (6)	127 (5)
N(52)	4732 (12)	2805 (9)	2262 (7)	243 (9)
C(505)	4194 (13)	2720 (8)	1872 (7)	163 (8)
C(506)	3549 (8)	2638 (7)	1336 (7)	158 (7)
N(53)	-111 (9)	1396 (9)	2050 (11)	271 (11)
C(507)	416 (9)	1765 (7)	1920 (9)	153 (7)
C(508)	1068 (7)	2239 (6)	1813 (6)	148 (6)

* Equivalent isotropic U defined as one-third of the trace of the orthogonalized U_{ij} tensor.

Occupancy factors are 0.80(1) for C(301) to C(304), 0.20(1) for C(307) to C(310).

Table S2. Bond lengths (Å) for $[\text{HB}(\text{pz})_3]_4\text{V}_4(\mu\text{-C}_6\text{H}_5\text{OPO}_3)_4\cdot 4\text{CH}_3\text{CN}$.

V(1)-O(1)	1.966 (3)	V(1)-O(3)	1.956 (3)
V(1)-O(11)	1.965 (3)	V(1)-N(1)	2.113 (4)
V(1)-N(3)	2.118 (4)	V(1)-N(5)	2.119 (4)
V(2)-O(5)	1.967 (3)	V(2)-O(7)	1.954 (4)
V(2)-O(8)	1.957 (3)	V(2)-N(7)	2.110 (4)
V(2)-N(9)	2.110 (4)	V(2)-N(11)	2.100 (4)
V(3)-O(4)	1.967 (3)	V(3)-O(9)	1.964 (3)
V(3)-O(12)	1.954 (3)	V(3)-N(13)	2.115 (4)
V(3)-N(15)	2.130 (4)	V(3)-N(17)	2.103 (4)
V(4)-O(2)	1.970 (3)	V(4)-O(10)	1.970 (3)
V(4)-O(13)	1.969 (3)	V(4)-N(19)	2.114 (4)
V(4)-N(21)	2.098 (4)	V(4)-N(23)	2.104 (4)
P(1)-O(6)	1.613 (3)	P(1)-O(7)	1.494 (4)
P(1)-O(11)	1.500 (4)	P(1)-O(13)	1.509 (3)
P(2)-O(3)	1.499 (3)	P(2)-O(5)	1.501 (4)
P(2)-O(9)	1.513 (3)	P(2)-O(16)	1.608 (3)
P(3)-O(2)	1.488 (3)	P(3)-O(8)	1.517 (3)
P(3)-O(12)	1.508 (3)	P(3)-O(15)	1.600 (4)
P(4)-O(1)	1.508 (3)	P(4)-O(4)	1.498 (3)
P(4)-O(10)	1.505 (3)	P(4)-O(14)	1.617 (3)
O(6)-C(101)	1.399 (6)	O(14)-C(401)	1.393 (6)
O(15)-C(301)	1.416 (10)	O(16)-C(201)	1.389 (6)
B(1)-N(2)	1.531 (8)	B(1)-N(4)	1.540 (8)
B(1)-N(6)	1.542 (9)	N(1)-N(2)	1.371 (7)
N(1)-C(11)	1.323 (7)	N(2)-C(13)	1.328 (8)
C(11)-C(12)	1.391 (9)	C(12)-C(13)	1.371 (11)
N(3)-N(4)	1.368 (6)	N(3)-C(14)	1.336 (7)
N(4)-C(16)	1.331 (8)	C(14)-C(15)	1.391 (9)
C(15)-C(16)	1.359 (11)	N(5)-N(6)	1.369 (6)
N(5)-C(17)	1.341 (7)	N(6)-C(19)	1.340 (7)
C(17)-C(18)	1.381 (8)	C(18)-C(19)	1.365 (10)
B(2)-N(8)	1.548 (7)	B(2)-N(10)	1.530 (8)
B(2)-N(12)	1.537 (9)	N(7)-N(8)	1.361 (7)
N(7)-C(21)	1.339 (7)	N(8)-C(23)	1.325 (8)
C(21)-C(22)	1.379 (10)	C(22)-C(23)	1.350 (10)
N(9)-N(10)	1.366 (6)	N(9)-C(24)	1.341 (7)
N(10)-C(26)	1.343 (7)	C(24)-C(25)	1.390 (9)
C(25)-C(26)	1.354 (9)	N(11)-N(12)	1.365 (6)
N(11)-C(27)	1.337 (7)	N(12)-C(29)	1.350 (7)
C(27)-C(28)	1.372 (8)	C(28)-C(29)	1.364 (10)
B(3)-N(14)	1.533 (8)	B(3)-N(16)	1.546 (8)
B(3)-N(18)	1.534 (9)	N(13)-N(14)	1.372 (6)
N(13)-C(31)	1.337 (8)	N(14)-C(33)	1.349 (7)
C(31)-C(32)	1.388 (8)	C(32)-C(33)	1.371 (10)
N(15)-N(16)	1.368 (6)	N(15)-C(34)	1.325 (7)
N(16)-C(36)	1.331 (8)	C(34)-C(35)	1.376 (9)
C(35)-C(36)	1.368 (9)	N(17)-N(18)	1.368 (5)
N(17)-C(37)	1.325 (8)	N(18)-C(39)	1.337 (7)
C(37)-C(38)	1.375 (8)	C(38)-C(39)	1.363 (9)
B(4)-N(20)	1.554 (8)	B(4)-N(22)	1.521 (9)

B(4)-N(24)	1.547	(8)	N(19)-N(20)	1.357	(5)
N(19)-C(41)	1.323	(6)	N(20)-C(43)	1.342	(7)
C(41)-C(42)	1.385	(8)	C(42)-C(43)	1.373	(8)
N(21)-N(22)	1.370	(6)	N(21)-C(44)	1.328	(8)
N(22)-C(46)	1.345	(8)	C(44)-C(45)	1.385	(9)
C(45)-C(46)	1.346	(10)	N(23)-N(24)	1.362	(6)
N(23)-C(47)	1.331	(7)	N(24)-C(49)	1.332	(7)
C(47)-C(48)	1.390	(8)	C(48)-C(49)	1.351	(10)
C(101)-C(102)	1.371	(9)	C(101)-C(106)	1.366	(9)
C(102)-C(103)	1.368	(10)	C(103)-C(104)	1.371	(15)
C(104)-C(105)	1.328	(18)	C(105)-C(106)	1.404	(12)
C(201)-C(202)	1.373	(10)	C(201)-C(206)	1.367	(8)
C(202)-C(203)	1.381	(10)	C(203)-C(204)	1.386	(12)
C(204)-C(205)	1.365	(14)	C(205)-C(206)	1.384	(10)
C(302)-C(303)	1.425	(17)	C(302)-C(301)	1.370	(18)
C(303)-C(304)	1.366	(20)	C(304)-C(305)	1.321	(21)
C(305)-C(306)	1.379	(11)	C(306)-C(301)	1.360	(13)
C(402)-C(403)	1.371	(10)	C(402)-C(401)	1.366	(8)
C(403)-C(404)	1.366	(10)	C(404)-C(405)	1.389	(11)
C(405)-C(406)	1.393	(9)	C(406)-C(401)	1.377	(7)
N(50)-C(501)	1.165	(19)	C(501)-C(502)	1.400	(15)
N(51)-C(503)	1.120	(14)	C(503)-C(504)	1.374	(13)
N(52)-C(505)	1.156	(24)	C(505)-C(506)	1.461	(21)
N(53)-C(507)	1.133	(21)	C(507)-C(508)	1.415	(19)

Table S3. Bond angles (°) for [HB(pz)₃]₄V₄(μ-C₆H₅OPO₃)₄·4CH₃CN.

O(1)-V(1)-O(3)	95.7(1)	O(1)-V(1)-O(11)	93.5(1)
O(3)-V(1)-O(11)	93.9(1)	O(1)-V(1)-N(1)	92.8(1)
O(3)-V(1)-N(1)	87.6(2)	O(11)-V(1)-N(1)	173.4(1)
O(1)-V(1)-N(3)	88.4(2)	O(3)-V(1)-N(3)	171.6(2)
O(11)-V(1)-N(3)	93.2(1)	N(1)-V(1)-N(3)	84.9(2)
O(1)-V(1)-N(5)	171.4(2)	O(3)-V(1)-N(5)	92.6(1)
O(11)-V(1)-N(5)	88.2(1)	N(1)-V(1)-N(5)	85.3(2)
N(3)-V(1)-N(5)	83.1(2)	O(5)-V(2)-O(7)	94.6(1)
O(5)-V(2)-O(8)	93.4(1)	O(7)-V(2)-O(8)	95.3(1)
O(5)-V(2)-N(7)	90.1(2)	O(7)-V(2)-N(7)	172.4(2)
O(8)-V(2)-N(7)	90.4(2)	O(5)-V(2)-N(9)	171.6(2)
O(7)-V(2)-N(9)	90.2(2)	O(8)-V(2)-N(9)	93.0(1)
N(7)-V(2)-N(9)	84.4(2)	O(5)-V(2)-N(11)	87.9(1)
O(7)-V(2)-N(11)	91.2(2)	O(8)-V(2)-N(11)	173.2(2)
N(7)-V(2)-N(11)	83.0(2)	N(9)-V(2)-N(11)	85.1(2)
O(4)-V(3)-O(9)	95.6(1)	O(4)-V(3)-O(12)	94.1(1)
O(9)-V(3)-O(12)	94.1(1)	O(4)-V(3)-N(13)	171.0(2)
O(9)-V(3)-N(13)	90.2(2)	O(12)-V(3)-N(13)	92.3(2)
O(4)-V(3)-N(15)	90.2(1)	O(9)-V(3)-N(15)	173.5(1)
O(12)-V(3)-N(15)	88.4(2)	N(13)-V(3)-N(15)	83.6(2)
O(4)-V(3)-N(17)	88.6(2)	O(9)-V(3)-N(17)	92.3(2)
O(12)-V(3)-N(17)	172.7(2)	N(13)-V(3)-N(17)	84.3(2)
N(15)-V(3)-N(17)	84.8(2)	O(2)-V(4)-O(10)	93.3(1)
O(2)-V(4)-O(13)	95.8(1)	O(10)-V(4)-O(13)	93.2(1)
O(2)-V(4)-N(19)	91.7(1)	O(10)-V(4)-N(19)	87.7(1)
O(13)-V(4)-N(19)	172.3(1)	O(2)-V(4)-N(21)	87.6(1)
O(10)-V(4)-N(21)	172.2(1)	O(13)-V(4)-N(21)	94.4(1)
N(19)-V(4)-N(21)	84.5(2)	O(2)-V(4)-N(23)	171.6(2)
O(10)-V(4)-N(23)	92.9(1)	O(13)-V(4)-N(23)	89.5(1)
N(19)-V(4)-N(23)	82.8(2)	N(21)-V(4)-N(23)	85.6(2)
O(6)-P(1)-O(7)	101.7(2)	O(6)-P(1)-O(11)	107.1(2)
O(7)-P(1)-O(11)	113.9(2)	O(6)-P(1)-O(13)	106.1(2)
O(7)-P(1)-O(13)	114.5(2)	O(11)-P(1)-O(13)	112.5(2)
O(3)-P(2)-O(5)	114.0(2)	O(3)-P(2)-O(9)	114.5(2)
O(5)-P(2)-O(9)	112.0(2)	O(3)-P(2)-O(16)	101.3(2)
O(5)-P(2)-O(16)	107.3(2)	O(9)-P(2)-O(16)	106.6(2)
O(2)-P(3)-O(8)	114.0(2)	O(2)-P(3)-O(12)	113.7(2)
O(8)-P(3)-O(12)	112.6(2)	O(2)-P(3)-O(15)	102.9(2)
O(8)-P(3)-O(15)	105.7(2)	O(12)-P(3)-O(15)	106.9(2)
O(1)-P(4)-O(4)	114.3(2)	O(1)-P(4)-O(10)	113.0(2)
O(4)-P(4)-O(10)	113.7(2)	O(1)-P(4)-O(14)	106.1(2)
O(4)-P(4)-O(14)	101.7(2)	O(10)-P(4)-O(14)	106.8(2)
V(1)-O(1)-P(4)	139.2(2)	V(4)-O(2)-P(3)	155.8(2)
V(1)-O(3)-P(2)	151.2(2)	V(3)-O(4)-P(4)	152.2(2)
V(2)-O(5)-P(2)	148.4(2)	P(1)-O(6)-C(101)	121.3(3)
V(2)-O(7)-P(1)	155.0(2)	V(2)-O(8)-P(3)	138.1(2)
V(3)-O(9)-P(2)	139.1(2)	V(4)-O(10)-P(4)	143.8(2)
V(1)-O(11)-P(1)	149.8(2)	V(3)-O(12)-P(3)	144.0(2)
V(4)-O(13)-P(1)	138.7(2)	P(4)-O(14)-C(401)	122.9(3)
P(3)-O(15)-C(307)	129.2(5)	P(3)-O(15)-C(301)	119.9(6)

C(307)-O(15)-C(301)	115.5(7)	P(2)-O(16)-C(201)	123.4(3)
N(2)-B(1)-N(4)	108.2(5)	N(2)-B(1)-N(6)	107.8(5)
N(4)-B(1)-N(6)	107.5(5)	V(1)-N(1)-N(2)	119.9(3)
V(1)-N(1)-C(11)	132.3(4)	N(2)-N(1)-C(11)	107.6(4)
B(1)-N(2)-N(1)	120.4(4)	B(1)-N(2)-C(13)	130.9(5)
N(1)-N(2)-C(13)	108.4(5)	N(1)-C(11)-C(12)	109.6(6)
C(11)-C(12)-C(13)	104.9(6)	N(2)-C(13)-C(12)	109.5(6)
V(1)-N(3)-N(4)	120.7(3)	V(1)-N(3)-C(14)	132.6(4)
N(4)-N(3)-C(14)	106.7(4)	B(1)-N(4)-N(3)	119.4(5)
B(1)-N(4)-C(16)	132.2(6)	N(3)-N(4)-C(16)	108.4(5)
N(3)-C(14)-C(15)	110.2(5)	C(14)-C(15)-C(16)	104.3(6)
N(4)-C(16)-C(15)	110.5(6)	V(1)-N(5)-N(6)	120.6(3)
V(1)-N(5)-C(17)	133.3(4)	N(6)-N(5)-C(17)	106.1(4)
B(1)-N(6)-N(5)	119.4(4)	B(1)-N(6)-C(19)	131.6(5)
N(5)-N(6)-C(19)	108.9(5)	N(5)-C(17)-C(18)	110.8(5)
C(17)-C(18)-C(19)	104.7(5)	N(6)-C(19)-C(18)	109.5(5)
N(8)-B(2)-N(10)	108.0(5)	N(8)-B(2)-N(12)	107.1(5)
N(10)-B(2)-N(12)	108.3(5)	V(2)-N(7)-N(8)	121.3(3)
V(2)-N(7)-C(21)	132.3(4)	N(8)-N(7)-C(21)	106.2(4)
B(2)-N(8)-N(7)	118.8(4)	B(2)-N(8)-C(23)	131.1(5)
N(7)-N(8)-C(23)	110.1(5)	N(7)-C(21)-C(22)	108.9(6)
C(21)-C(22)-C(23)	106.7(6)	N(8)-C(23)-C(22)	108.1(6)
V(2)-N(9)-N(10)	120.6(3)	V(2)-N(9)-C(24)	133.1(4)
N(10)-N(9)-C(24)	106.3(4)	B(2)-N(10)-N(9)	119.6(4)
B(2)-N(10)-C(26)	130.7(5)	N(9)-N(10)-C(26)	109.6(4)
N(9)-C(24)-C(25)	109.5(5)	C(24)-C(25)-C(26)	106.1(5)
N(10)-C(26)-C(25)	108.5(5)	V(2)-N(11)-N(12)	120.9(3)
V(2)-N(11)-C(27)	132.0(4)	N(12)-N(11)-C(27)	107.0(4)
B(2)-N(12)-N(11)	119.3(4)	B(2)-N(12)-C(29)	132.8(5)
N(11)-N(12)-C(29)	107.4(5)	N(11)-C(27)-C(28)	111.0(5)
C(27)-C(28)-C(29)	104.3(5)	N(12)-C(29)-C(28)	110.4(6)
N(14)-B(3)-N(16)	106.4(5)	N(14)-B(3)-N(18)	110.0(5)
N(16)-B(3)-N(18)	108.1(5)	V(3)-N(13)-N(14)	120.1(3)
V(3)-N(13)-C(31)	132.5(3)	N(14)-N(13)-C(31)	107.2(4)
B(3)-N(14)-N(13)	119.8(4)	B(3)-N(14)-C(33)	132.0(5)
N(13)-N(14)-C(33)	108.2(5)	N(13)-C(31)-C(32)	110.2(5)
C(31)-C(32)-C(33)	105.0(5)	N(14)-C(33)-C(32)	109.4(5)
V(3)-N(15)-N(16)	121.4(3)	V(3)-N(15)-C(34)	132.6(3)
N(16)-N(15)-C(34)	105.9(4)	B(3)-N(16)-N(15)	118.1(4)
B(3)-N(16)-C(36)	132.6(5)	N(15)-N(16)-C(36)	109.1(4)
N(15)-C(34)-C(35)	111.4(5)	C(34)-C(35)-C(36)	104.3(6)
N(16)-C(36)-C(35)	109.3(6)	V(3)-N(17)-N(18)	120.8(3)
V(3)-N(17)-C(37)	132.0(3)	N(18)-N(17)-C(37)	106.6(4)
B(3)-N(18)-N(17)	119.5(4)	B(3)-N(18)-C(39)	131.9(4)
N(17)-N(18)-C(39)	108.2(5)	N(17)-C(37)-C(38)	110.8(5)
C(37)-C(38)-C(39)	104.6(6)	N(18)-C(39)-C(38)	109.7(5)
N(20)-B(4)-N(22)	107.9(5)	N(20)-B(4)-N(24)	106.6(5)
N(22)-B(4)-N(24)	108.7(5)	V(4)-N(19)-N(20)	120.9(3)
V(4)-N(19)-C(41)	132.7(3)	N(20)-N(19)-C(41)	106.4(4)
B(4)-N(20)-N(19)	119.2(4)	B(4)-N(20)-C(43)	130.9(4)
N(19)-N(20)-C(43)	109.8(4)	N(19)-C(41)-C(42)	110.8(5)
C(41)-C(42)-C(43)	104.8(5)	N(20)-C(43)-C(42)	108.1(5)
V(4)-N(21)-N(22)	120.4(3)	V(4)-N(21)-C(44)	132.5(3)
N(22)-N(21)-C(44)	107.1(4)	B(4)-N(22)-N(21)	120.1(4)

B(4)-N(22)-C(46)	131.5(5)	N(21)-N(22)-C(46)	108.1(5)
N(21)-C(44)-C(45)	109.7(5)	C(44)-C(45)-C(46)	105.7(6)
N(22)-C(46)-C(45)	109.4(6)	V(4)-N(23)-N(24)	121.5(3)
V(4)-N(23)-C(47)	132.7(3)	N(24)-N(23)-C(47)	105.8(4)
B(4)-N(24)-N(23)	118.7(4)	B(4)-N(24)-C(49)	131.0(5)
N(23)-N(24)-C(49)	110.2(5)	N(23)-C(47)-C(48)	109.9(5)
C(47)-C(48)-C(49)	105.7(5)	N(24)-C(49)-C(48)	108.3(5)
O(6)-C(101)-C(102)	123.2(5)	O(6)-C(101)-C(106)	117.3(6)
C(102)-C(101)-C(106)	119.4(6)	C(101)-C(102)-C(103)	120.1(6)
C(102)-C(103)-C(104)	119.1(9)	C(103)-C(104)-C(105)	122.9(9)
C(104)-C(105)-C(106)	117.7(9)	C(101)-C(106)-C(105)	120.8(8)
O(16)-C(201)-C(202)	120.8(5)	O(16)-C(201)-C(206)	118.1(6)
C(202)-C(201)-C(206)	121.0(6)	C(201)-C(202)-C(203)	119.5(6)
C(202)-C(203)-C(204)	120.0(8)	C(203)-C(204)-C(205)	119.5(7)
C(204)-C(205)-C(206)	120.9(7)	C(201)-C(206)-C(205)	119.1(7)
C(303)-C(302)-C(301)	119.4(10)	C(302)-C(303)-C(304)	118.9(14)
C(303)-C(304)-C(305)	119.4(12)	C(304)-C(305)-C(306)	123.3(9)
C(305)-C(306)-C(301)	118.3(9)	O(15)-C(301)-C(302)	118.5(9)
O(15)-C(301)-C(306)	121.0(10)	C(302)-C(301)-C(306)	120.0(9)
C(403)-C(402)-C(401)	120.2(5)	C(402)-C(403)-C(404)	120.4(7)
C(403)-C(404)-C(405)	119.5(7)	C(404)-C(405)-C(406)	120.4(5)
C(405)-C(406)-C(401)	118.3(5)	O(14)-C(401)-C(402)	117.9(4)
O(14)-C(401)-C(406)	120.9(5)	C(402)-C(401)-C(406)	121.2(5)
N(50)-C(501)-C(502)	175.3(13)	N(51)-C(503)-C(504)	177.5(13)
N(52)-C(505)-C(506)	175.9(16)	N(53)-C(507)-C(508)	176.0(18)

Table S4. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for
 $[\text{HB}(\text{pz})_3]_4\text{V}_4(\mu\text{-C}_6\text{H}_5\text{OPO}_3)_4 \cdot 4\text{CH}_3\text{CN}$.

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
V(1)	39(1)	34(1)	31(1)	-10(1)	-8(1)	2(1)
V(2)	36(1)	36(1)	40(1)	-12(1)	-1(1)	-1(1)
V(3)	36(1)	34(1)	42(1)	-2(1)	-7(1)	-2(1)
V(4)	37(1)	30(1)	28(1)	-4(1)	-4(1)	1(1)
P(1)	32(1)	30(1)	33(1)	-7(1)	-6(1)	4(1)
P(2)	38(1)	35(1)	34(1)	-8(1)	-1(1)	-2(1)
P(3)	34(1)	37(1)	35(1)	-6(1)	-8(1)	-3(1)
P(4)	36(1)	29(1)	34(1)	-7(1)	-4(1)	2(1)
O(1)	51(2)	33(2)	35(2)	-12(2)	-8(2)	2(1)
O(2)	37(2)	43(2)	43(2)	-5(2)	-2(2)	-1(2)
O(3)	41(2)	48(2)	42(2)	-11(2)	-5(2)	4(2)
O(4)	36(2)	44(2)	46(2)	-6(2)	-4(2)	0(2)
O(5)	46(2)	39(2)	40(2)	-13(2)	-3(2)	-3(2)
O(6)	43(2)	38(2)	49(2)	-4(2)	-7(2)	12(2)
O(7)	39(2)	55(2)	55(2)	-17(2)	-1(2)	3(2)
O(8)	54(2)	35(2)	43(2)	-13(2)	-6(2)	-2(2)
O(9)	48(2)	34(2)	41(2)	-5(2)	-8(2)	-2(2)
O(10)	41(2)	31(2)	35(2)	-6(1)	-5(2)	0(1)
O(11)	43(2)	34(2)	36(2)	-10(2)	-6(2)	-1(1)
O(12)	36(2)	37(2)	47(2)	-5(2)	-6(2)	-5(2)
O(13)	47(2)	30(2)	34(2)	-8(2)	-12(2)	3(1)
O(14)	42(2)	29(2)	46(2)	-8(1)	-3(2)	9(1)
O(15)	43(2)	62(2)	52(2)	0(2)	-18(2)	-12(2)
O(16)	48(2)	62(2)	37(2)	-13(2)	4(2)	-8(2)
B(1)	71(5)	70(5)	50(4)	-31(4)	-29(4)	3(4)
N(1)	62(3)	43(3)	37(2)	-15(2)	-6(2)	-6(2)
N(2)	79(4)	57(3)	36(3)	-29(3)	-12(2)	-8(2)
C(11)	73(4)	49(4)	59(4)	-14(3)	8(3)	-6(3)
C(12)	129(7)	49(4)	58(4)	-26(4)	19(4)	-23(3)
C(13)	115(6)	69(4)	46(4)	-41(4)	-4(4)	-16(3)
N(3)	45(3)	47(3)	45(3)	-16(2)	-12(2)	5(2)
N(4)	49(3)	68(3)	58(3)	-25(2)	-23(2)	8(2)
C(14)	53(4)	50(3)	58(4)	-11(3)	-3(3)	9(3)
C(15)	47(4)	83(5)	89(5)	-20(3)	4(4)	11(4)
C(16)	50(4)	88(5)	100(6)	-33(4)	-28(4)	10(4)
N(5)	47(3)	44(3)	39(2)	-11(2)	-12(2)	4(2)
N(6)	60(3)	61(3)	42(3)	-12(2)	-23(2)	6(2)
C(17)	65(4)	42(3)	48(3)	-10(3)	-4(3)	8(3)
C(18)	90(5)	48(4)	55(4)	-3(3)	-9(4)	16(3)
C(19)	83(5)	69(4)	44(3)	3(4)	-27(3)	15(3)
B(2)	51(4)	49(4)	68(5)	-23(3)	11(4)	6(3)
N(7)	40(3)	43(3)	63(3)	-11(2)	-4(2)	-5(2)
N(8)	36(3)	55(3)	82(4)	-19(2)	10(2)	-5(3)
C(21)	43(4)	55(4)	106(5)	0(3)	-13(4)	-14(4)
C(22)	29(4)	87(6)	168(8)	-4(4)	-7(4)	-13(5)
C(23)	37(4)	77(5)	137(7)	-23(4)	9(4)	-6(5)
N(9)	42(3)	41(2)	52(3)	-11(2)	4(2)	-1(2)

N(10)	48(3)	37(3)	60(3)	-17(2)	2(2)	-2(2)
C(24)	66(4)	54(4)	54(4)	-5(3)	8(3)	-6(3)
C(25)	93(5)	46(4)	75(5)	1(4)	-1(4)	-21(3)
C(26)	73(4)	42(4)	70(4)	-21(3)	-4(3)	-6(3)
N(11)	44(3)	46(3)	46(3)	-14(2)	7(2)	4(2)
N(12)	53(3)	48(3)	52(3)	-16(2)	12(2)	5(2)
C(27)	57(4)	54(3)	47(3)	-10(3)	-6(3)	0(3)
C(28)	82(5)	73(4)	43(4)	-11(4)	-8(3)	6(3)
C(29)	77(5)	63(4)	53(4)	-10(4)	15(3)	20(3)
B(3)	51(4)	40(4)	71(5)	8(3)	-12(4)	2(3)
N(13)	42(3)	40(3)	59(3)	-1(2)	-5(2)	-7(2)
N(14)	44(3)	42(3)	69(3)	7(2)	-10(2)	-7(2)
C(31)	53(4)	54(4)	67(4)	-16(3)	-3(3)	-2(3)
C(32)	37(4)	71(4)	102(5)	-10(3)	5(3)	-11(4)
C(33)	36(4)	67(4)	102(5)	8(3)	-9(3)	-18(4)
N(15)	48(3)	44(3)	47(3)	-4(2)	-10(2)	3(2)
N(16)	54(3)	47(3)	57(3)	-3(2)	-20(2)	11(2)
C(34)	66(4)	61(4)	50(4)	-12(3)	-12(3)	5(3)
C(35)	105(6)	86(5)	48(4)	-9(4)	-18(4)	7(4)
C(36)	80(5)	75(5)	70(5)	-3(4)	-34(4)	26(4)
N(17)	48(3)	35(2)	54(3)	-5(2)	-9(2)	1(2)
N(18)	57(3)	36(2)	55(3)	2(2)	-6(2)	1(2)
C(37)	59(4)	49(4)	57(4)	-13(3)	-10(3)	-6(3)
C(38)	78(5)	48(4)	87(5)	-23(3)	-23(4)	-8(3)
C(39)	85(5)	27(3)	80(4)	-8(3)	-7(4)	-6(3)
B(4)	68(5)	65(5)	33(3)	0(4)	7(3)	0(3)
N(19)	50(3)	40(3)	33(2)	-4(2)	-6(2)	6(2)
N(20)	58(3)	56(3)	32(2)	-5(2)	-2(2)	11(2)
C(41)	62(4)	41(3)	48(3)	0(3)	-6(3)	5(3)
C(42)	98(5)	55(4)	55(4)	-3(4)	-6(4)	28(3)
C(43)	86(5)	76(4)	37(3)	-10(4)	2(3)	19(3)
N(21)	48(3)	38(2)	39(3)	-5(2)	-9(2)	-6(2)
N(22)	58(3)	48(3)	34(2)	3(2)	-8(2)	-11(2)
C(44)	62(4)	39(3)	64(4)	-12(3)	-20(3)	-2(3)
C(45)	90(5)	44(4)	84(5)	-11(4)	-32(4)	-14(3)
C(46)	88(5)	62(4)	50(4)	12(4)	-24(4)	-22(3)
N(23)	41(3)	42(2)	38(2)	-5(2)	-3(2)	-2(2)
N(24)	47(3)	59(3)	46(3)	-3(2)	6(2)	2(2)
C(47)	53(4)	52(3)	61(4)	-14(3)	-20(3)	6(3)
C(48)	36(4)	90(5)	90(5)	-12(3)	0(4)	5(4)
C(49)	50(4)	77(4)	69(4)	-5(3)	13(3)	6(3)
C(101)	57(4)	55(4)	41(3)	14(3)	-11(3)	11(3)
C(102)	47(4)	60(4)	88(5)	-12(3)	-18(3)	23(3)
C(103)	55(5)	118(7)	123(7)	-21(5)	-22(5)	60(6)
C(104)	69(7)	214(14)	82(7)	36(8)	13(5)	39(8)
C(105)	82(7)	184(11)	78(6)	39(7)	-13(5)	-34(6)
C(106)	85(6)	109(6)	71(5)	16(5)	-34(4)	-26(4)
C(201)	49(3)	63(4)	43(3)	-12(3)	4(3)	-4(3)
C(202)	59(4)	82(5)	69(4)	-22(4)	20(3)	-18(3)
C(203)	74(5)	136(7)	90(6)	-41(5)	22(4)	-29(5)
C(204)	57(5)	164(9)	75(5)	-11(5)	12(4)	-25(5)
C(205)	80(6)	105(6)	77(5)	16(5)	1(4)	-23(4)
C(206)	70(5)	73(4)	68(4)	0(4)	17(4)	-15(3)
C(301)	50(6)	79(7)	98(7)	-14(5)	-31(4)	-39(5)

C(302)	81(7)	101(8)	146(11)	-19(6)	-24(7)	-35(7)
C(303)	88(9)	156(13)	236(17)	-73(9)	-29(10)	-39(12)
C(304)	60(7)	182(14)	171(14)	-39(9)	-7(8)	-54(12)
C(305)	53(5)	138(9)	270(14)	11(5)	-54(7)	-81(9)
C(306)	51(5)	94(6)	154(8)	9(4)	-37(5)	-19(5)
C(402)	69(4)	56(4)	52(3)	-23(3)	-3(3)	-3(3)
C(403)	84(5)	90(5)	76(5)	-50(4)	-19(4)	4(4)
C(404)	55(4)	83(5)	97(5)	-29(4)	-16(4)	31(4)
C(405)	51(4)	64(4)	79(5)	-1(3)	4(3)	20(3)
C(406)	51(4)	41(3)	62(4)	-9(3)	-1(3)	9(3)
C(401)	43(3)	36(3)	39(3)	-12(2)	-2(2)	13(2)
N(50)	226(13)	237(14)	313(18)	-62(10)	-162(12)	114(12)
C(501)	130(8)	104(7)	168(11)	-18(6)	-74(8)	20(7)
C(502)	159(9)	124(8)	106(7)	-6(7)	-47(7)	10(6)
N(51)	312(16)	105(7)	249(13)	-1(9)	77(11)	66(9)
C(503)	185(11)	77(6)	135(8)	-37(7)	27(8)	38(6)
C(504)	133(8)	89(7)	159(9)	-21(6)	-12(7)	24(6)
N(52)	369(21)	208(12)	111(9)	62(12)	-70(10)	-54(8)
C(505)	261(19)	124(9)	76(8)	19(11)	29(9)	-11(7)
C(506)	199(13)	167(11)	114(9)	-66(9)	19(9)	4(8)
N(53)	158(12)	199(13)	476(26)	-61(10)	-79(14)	-1(14)
C(507)	115(10)	106(9)	239(14)	-15(7)	-41(10)	-2(9)
C(508)	144(10)	142(10)	151(10)	-10(8)	-15(8)	3(8)

Table S5. Hydrogen atom coordinates for $[\text{HB}(\text{pz})_3]_4\text{V}_4(\mu\text{-C}_6\text{H}_5\text{OPO}_3)_4\cdot 4\text{CH}_3\text{CN}$.

<u>Atom</u>	<u>X</u>	<u>Y</u>	<u>Z</u>	<u>U</u>
H(1B)	3612	6883	-677	44
H(11A)	6655	5552	317	74
H(12A)	6262	4896	-841	74
H(13A)	4763	5652	-1128	74
H(14A)	3569	7038	2328	74
H(15A)	2092	6886	1895	74
H(16A)	2279	6865	497	74
H(17A)	5327	9053	367	74
H(18A)	4537	9545	-781	74
H(19A)	3777	8449	-1136	74
H(2B)	8749	10734	782	44
H(21A)	9670	7881	2041	74
H(22A)	10988	8452	1557	74
H(23A)	10397	9787	1002	74
H(24A)	6482	10304	2962	74
H(25A)	6789	11708	2919	74
H(26A)	7926	11737	1865	74
H(27A)	6893	8618	50	74
H(28A)	7182	9493	-1040	74
H(29A)	8031	10462	-557	74
H(3B)	9784	3742	2964	44
H(31A)	9807	6444	1533	74
H(32A)	11336	5583	1539	74
H(33A)	11161	4332	2246	74
H(34A)	7840	6209	4330	74
H(35A)	8637	5238	5239	74
H(36A)	9518	4104	4462	74
H(37A)	6823	4720	1690	74
H(38A)	7202	3215	1763	74
H(39A)	8648	2888	2360	74
H(4B)	4415	8301	6080	44
H(41A)	6252	6002	4364	44
H(42A)	5979	5531	5682	74
H(43A)	5101	6741	6379	74
H(44A)	6735	9320	4216	74
H(45A)	6541	10008	5468	74
H(46A)	5367	9473	6206	74
H(47A)	3464	8366	3282	74
H(48A)	2149	8723	4214	74
H(49A)	2746	8636	5473	74
H(102)	3567	8735	1694	54
H(103)	2064	9019	2100	118
H(104)	1494	10195	2757	118
H(105)	2351	11075	3040	118
H(106)	3884	10766	2666	54
H(202)	9057	7893	332	79
H(203)	10574	7652	-8	147
H(204)	11279	6406	-512	147
H(205)	10458	5434	-712	147

H(206)	8949	5655	-343	79
H(30A)	9082	8952	3871	74
H(30B)	10608	8975	3610	118
H(30C)	11605	7774	3382	118
H(30D)	11112	6594	3410	118
H(30E)	9615	6552	3614	74
H(40A)	4640	4658	2382	57
H(40B)	3157	4545	2512	91
H(404)	2179	5362	3341	91
H(405)	2669	6321	4043	91
H(406)	4161	6442	3920	91
H(50A)	2037	2397	4592	200
H(50B)	2850	2815	4590	200
H(50C)	2927	2005	4161	200
H(50D)	6631	4397	4541	200
H(50E)	6145	4161	3870	200
H(50F)	6985	4540	3725	200
H(50G)	3096	2395	1577	200
H(50H)	3836	2306	928	200
H(50I)	3291	3158	1153	200
H(50J)	1274	2229	1297	200
H(50K)	1553	2030	2109	200
H(50L)	821	2782	1959	200

Table S6. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement coefficients ($\text{\AA}^2 \times 10^3$) for $(t\text{-Bupz})_5\text{V}_4\text{O}_4(\mu\text{-C}_6\text{H}_5\text{PO}_3)_4 \cdot 4\text{CH}_3\text{CN} \cdot 0.6\text{H}_2\text{O}$.

Atom	X	Y	Z	U _{eq} *
V(1)	7015(2)	5399(1)	1373(1)	35(1)
V(2)	5652(2)	8233(1)	1085(1)	33(1)
V(3)	8838(2)	7013(2)	2100(1)	37(1)
V(4)	5530(2)	7066(2)	3112(1)	36(1)
P(1)	7687(3)	7065(2)	769(2)	35(1)
P(2)	6368(3)	8286(2)	2599(2)	33(1)
P(3)	7845(2)	5571(2)	2543(2)	35(1)
P(4)	4760(3)	6801(2)	1721(2)	35(1)
O(1)	5381(6)	6605(5)	3859(4)	45(2)
O(2)	9983(6)	6401(6)	1998(4)	48(2)
O(3)	6263(5)	7749(5)	3296(4)	37(2)
O(4)	5029(6)	9296(5)	991(4)	46(2)
O(5)	7390(5)	8066(5)	2285(4)	37(2)
O(6)	8334(6)	6165(5)	2633(4)	41(2)
O(7)	8158(5)	5096(5)	1840(4)	41(2)
O(8)	4840(5)	6686(5)	2518(4)	39(2)
O(9)	4660(5)	7742(5)	1412(4)	33(2)
O(10)	6743(5)	6078(5)	2617(4)	39(2)
O(11)	7009(5)	8096(5)	761(4)	33(2)
O(12)	8500(5)	6816(5)	1179(4)	31(2)
O(13)	7450(6)	4797(5)	693(4)	46(2)
O(14)	5607(5)	6028(5)	1296(4)	39(2)
O(15)	6972(6)	6627(5)	1044(4)	41(2)
O(16)	5788(5)	8082(5)	2119(4)	33(2)
N(11)	8989(7)	8202(7)	1593(5)	43(2)
N(12)	8329(7)	8805(7)	1214(5)	42(2)
C(11)	9609(9)	8550(9)	1614(8)	58(2)
C(12)	9323(10)	9372(9)	1249(8)	65(2)
C(13)	8505(9)	9536(8)	983(7)	51(2)
C(14)	7858(10)	10305(10)	566(9)	68(2)
C(15)	7119(12)	10126(11)	257(10)	137(2)
C(16)	8500(11)	10520(10)	-88(9)	115(2)
C(17)	7296(12)	11159(11)	1010(10)	136(2)
N(21)	8853(7)	7366(7)	3141(5)	39(2)
N(22)	8119(7)	7501(7)	3684(6)	51(2)
C(21)	9483(10)	7489(9)	3427(8)	64(2)
C(22)	9180(10)	7693(11)	4130(8)	81(2)
C(23)	8294(10)	7686(9)	4287(8)	56(2)
C(24)	7552(12)	7894(11)	4962(8)	82(2)
C(25)	6738(12)	8827(13)	4895(10)	186(2)
C(26)	7095(14)	7236(14)	5080(10)	191(2)
C(27)	8026(12)	7850(14)	5600(8)	157(2)
N(31)	5627(7)	7926(7)	44(5)	41(2)
N(32)	5734(8)	7111(6)	-140(6)	50(2)
C(31)	5481(9)	8446(9)	-544(7)	47(2)
C(32)	5500(10)	7962(9)	-1103(8)	61(2)
C(33)	5657(10)	7116(9)	-809(8)	62(2)
C(34)	5764(8)	6298(6)	-1130(6)	99(2)
C(35)**	6819(8)	5782(16)	-1447(13)	182(2)

C(35') **	6257(15)	6176(14)	-1897(7)	150(2)
C(36) **	5190(15)	6513(14)	-1743(10)	80(2)
C(36') **	4757(8)	6381(15)	-1141(13)	186(2)
C(37) **	5511(16)	5613(13)	-645(10)	140(2)
C(37') **	6292(14)	5397(10)	-741(9)	85(2)
N(41)	4211(7)	8280(7)	3265(6)	40(2)
N(42)	3648(7)	8679(7)	2758(6)	42(2)
C(41)	3709(10)	8761(9)	3836(7)	59(2)
C(42)	2856(10)	9464(10)	3670(8)	70(2)
C(43)	2811(9)	9407(9)	2960(8)	51(2)
C(44)	2081(10)	9972(9)	2493(9)	66(2)
C(45)	1251(11)	10744(11)	2872(10)	137(2)
C(46)	1736(12)	9376(10)	2206(9)	124(2)
C(47)	2541(12)	10363(11)	1846(9)	128(2)
N(51)	6724(7)	4420(6)	2070(5)	39(2)
N(52)	6067(7)	4624(7)	2672(6)	46(2)
C(51)	7140(10)	3525(9)	2072(8)	58(2)
C(52)	6763(10)	3165(9)	2665(8)	70(2)
C(53)	6094(9)	3872(8)	3059(7)	40(2)
C(54)	5484(10)	3919(9)	3769(8)	60(2)
C(55)	4513(11)	4742(11)	3811(9)	130(2)
C(56)	5976(12)	4064(11)	4329(8)	108(2)
C(57)	5344(14)	3081(11)	3956(10)	187(2)
C(61)	8184(8)	4712(9)	3250(8)	46(2)
C(62)	7964(11)	4955(10)	3949(8)	82(2)
C(63)	8249(11)	4341(12)	4494(9)	98(2)
C(64)	8798(11)	3468(12)	4364(10)	101(2)
C(65)	9072(11)	3161(10)	3674(10)	103(2)
C(66)	8738(10)	3803(10)	3116(8)	69(2)
C(71)	8238(8)	6683(8)	-112(7)	33(2)
C(72)	8096(9)	7288(9)	-704(8)	62(2)
C(73)	8570(12)	6987(11)	-1381(8)	105(2)
C(74)	9141(11)	6097(11)	-1466(8)	89(2)
C(75)	9279(11)	5478(10)	-917(8)	83(2)
C(76)	8823(9)	5783(9)	-236(7)	52(2)
C(81)	5799(9)	9474(8)	2782(7)	39(2)
C(82)	4860(9)	10025(8)	2717(8)	59(2)
C(83)	4394(10)	10935(9)	2879(8)	68(2)
C(84)	4895(11)	11297(9)	3158(8)	78(2)
C(85)	5853(13)	10783(10)	3175(10)	115(2)
C(86)	6311(10)	9876(10)	2984(9)	90(2)
C(91)	3659(8)	6779(7)	1594(7)	34(2)
C(92)	3125(9)	6488(9)	2149(8)	57(2)
C(93)	2262(9)	6472(9)	2009(8)	64(2)
C(94)	1982(10)	6710(9)	1359(9)	72(2)
C(95)	2488(10)	7002(9)	831(9)	82(2)
C(96)	3339(10)	7031(10)	951(9)	77(2)
C(1)	7345(10)	2815(12)	5900(10)	167(2)
C(2)	8284(11)	2165(10)	6063(10)	152(2)
C(3)	9600(12)	5049(13)	6474(12)	251(2)
C(4)	8756(11)	4925(12)	6416(10)	145(2)
N(2)	8113(12)	4804(16)	6340(10)	237(2)
C(5)	9558(10)	2950(10)	267(9)	141(2)
C(6)	9634(12)	2217(10)	783(9)	149(2)

N(3)	9655(14)	1668(11)	1210(10)	221(2)
C(7)	8597(14)	289(16)	3340(11)	341(2)
C(8)	8070(12)	764(15)	3984(10)	202(2)
N(4)	7711(21)	1132(24)	4503(14)	872(2)
N(5)	9028(10)	1616(13)	6155(14)	302(2)
O(1W)	162(18)	557(18)	4793(14)	237(2)

* Equivalent isotropic U defined as one third of the trace of the orthogonalized U_{ij} tensor.

** Site occupation factor for C(35), C(36), and C(37) is 0.451(3); for C(35'), C(36'), and C(37') is 0.549(3).

Table S7. Bond lengths (Å) for $(t\text{-Bupz})_5\text{V}_4\text{O}_4(\mu\text{-C}_6\text{H}_5\text{PO}_3)_4 \cdot 4\text{CH}_3\text{CN} \cdot 0.6\text{H}_2\text{O}$.

V(1)-O(7)	1.970 (9)	V(1)-O(10)	2.614 (9)
V(1)-O(13)	1.578 (9)	V(1)-O(14)	1.989 (8)
V(1)-O(15)	2.010 (9)	V(1)-N(51)	2.117 (11)
V(2)-O(4)	1.578 (8)	V(2)-O(9)	2.012 (9)
V(2)-O(11)	2.006 (8)	V(2)-O(15)	2.514 (7)
V(2)-O(16)	1.997 (8)	V(2)-N(31)	2.111 (12)
V(3)-O(2)	1.596 (7)	V(3)-O(5)	2.136 (7)
V(3)-O(6)	1.981 (10)	V(3)-O(12)	2.018 (9)
V(3)-N(11)	2.161 (12)	V(3)-N(21)	2.135 (11)
V(4)-O(1)	1.565 (9)	V(4)-O(3)	2.008 (11)
V(4)-O(8)	1.999 (10)	V(4)-O(10)	2.014 (7)
V(4)-O(16)	2.474 (8)	V(4)-N(41)	2.129 (8)
P(1)-O(11)	1.541 (7)	P(1)-O(12)	1.482 (9)
P(1)-O(15)	1.560 (11)	P(1)-C(71)	1.787 (12)
P(2)-O(3)	1.534 (9)	P(2)-O(5)	1.487 (9)
P(2)-O(16)	1.548 (11)	P(2)-C(81)	1.785 (12)
P(3)-O(6)	1.514 (12)	P(3)-O(7)	1.517 (9)
P(3)-O(10)	1.523 (8)	P(3)-C(61)	1.800 (14)
P(4)-O(8)	1.530 (9)	P(4)-O(9)	1.535 (9)
P(4)-O(14)	1.534 (7)	P(4)-C(91)	1.787 (15)
N(11)-N(12)	1.324 (13)	N(11)-C(11)	1.326 (22)
N(12)-C(13)	1.351 (19)	C(11)-C(12)	1.363 (19)
C(12)-C(13)	1.359 (23)	C(13)-C(14)	1.470 (18)
C(14)-C(15)	1.527 (29)	C(14)-C(16)	1.563 (23)
C(14)-C(17)	1.532 (22)	N(21)-N(22)	1.352 (14)
N(21)-C(21)	1.298 (22)	N(22)-C(23)	1.322 (21)
C(21)-C(22)	1.366 (21)	C(22)-C(23)	1.360 (24)
C(23)-C(24)	1.524 (20)	C(24)-C(25)	1.499 (21)
C(24)-C(26)	1.508 (34)	C(24)-C(27)	1.510 (26)
N(31)-N(32)	1.345 (17)	N(31)-C(31)	1.318 (16)
N(32)-C(33)	1.301 (20)	C(31)-C(32)	1.384 (22)
C(32)-C(33)	1.364 (20)	C(33)-C(34)	1.462 (20)
C(34)-C(35)	1.515 (16)	C(34)-C(35')	1.515 (18)
C(34)-C(36)	1.515 (25)	C(34)-C(36')	1.515 (21)
C(34)-C(37)	1.515 (26)	C(34)-C(37')	1.515 (17)
N(41)-N(42)	1.335 (15)	N(41)-C(41)	1.323 (16)
N(42)-C(43)	1.343 (13)	C(41)-C(42)	1.377 (17)
C(42)-C(43)	1.383 (23)	C(43)-C(44)	1.478 (20)
C(44)-C(45)	1.484 (18)	C(44)-C(46)	1.491 (28)
C(44)-C(47)	1.533 (24)	N(51)-N(52)	1.354 (13)
N(51)-C(51)	1.311 (16)	N(52)-C(53)	1.350 (17)
C(51)-C(52)	1.373 (21)	C(52)-C(53)	1.353 (16)
C(53)-C(54)	1.495 (19)	C(54)-C(55)	1.508 (18)
C(54)-C(56)	1.514 (26)	C(54)-C(57)	1.476 (27)
C(61)-C(62)	1.373 (22)	C(61)-C(66)	1.373 (18)
C(62)-C(63)	1.349 (23)	C(63)-C(64)	1.322 (23)
C(64)-C(65)	1.388 (26)	C(65)-C(66)	1.402 (22)
C(71)-C(72)	1.395 (18)	C(71)-C(76)	1.364 (16)
C(72)-C(73)	1.389 (20)	C(73)-C(74)	1.337 (22)
C(74)-C(75)	1.347 (22)	C(75)-C(76)	1.392 (19)

C(81)-C(82)	1.353	(17)	C(81)-C(86)	1.357	(25)
C(82)-C(83)	1.378	(19)	C(83)-C(84)	1.366	(28)
C(84)-C(85)	1.351	(23)	C(85)-C(86)	1.389	(21)
C(91)-C(92)	1.398	(19)	C(91)-C(96)	1.359	(22)
C(92)-C(93)	1.425	(23)	C(93)-C(94)	1.347	(24)
C(94)-C(95)	1.342	(23)	C(95)-C(96)	1.404	(26)
C(1)-C(2)	1.439	(20)	C(2)-N(5)	1.136	(20)
C(3)-C(4)	1.438	(31)	C(4)-N(2)	1.135	(32)
C(5)-C(6)	1.438	(23)	C(6)-N(3)	1.135	(25)
C(7)-C(8)	1.439	(28)	C(8)-N(4)	1.136	(34)

Table S8. Bond angles (°) for $(t\text{-Bupz})_5\text{V}_4\text{O}_4(\mu\text{-C}_6\text{H}_5\text{PO}_3)_4 \cdot 4\text{CH}_3\text{CN} \cdot 0.6\text{H}_2\text{O}$.

O(7)-V(1)-O(10)	61.8(3)	O(10)-V(1)-O(13)	164.2(3)
O(10)-V(1)-O(14)	92.4(3)	O(10)-V(1)-O(15)	80.9(3)
O(10)-V(1)-N(51)	78.6(4)	O(7)-V(1)-O(13)	103.5(4)
O(7)-V(1)-O(14)	154.0(4)	O(13)-V(1)-O(14)	102.5(4)
O(7)-V(1)-O(15)	90.8(4)	O(13)-V(1)-O(15)	105.7(4)
O(14)-V(1)-O(15)	83.3(4)	O(7)-V(1)-N(51)	88.2(4)
O(13)-V(1)-N(51)	96.6(4)	O(14)-V(1)-N(51)	87.7(4)
O(15)-V(1)-N(51)	157.2(4)	O(4)-V(2)-O(9)	104.2(4)
O(4)-V(2)-O(11)	102.3(4)	O(9)-V(2)-O(11)	153.4(3)
O(4)-V(2)-O(16)	102.4(4)	O(9)-V(2)-O(16)	83.2(4)
O(11)-V(2)-O(16)	93.9(4)	O(4)-V(2)-N(31)	95.6(4)
O(9)-V(2)-N(31)	86.4(4)	O(11)-V(2)-N(31)	88.2(4)
O(16)-V(2)-N(31)	160.9(4)	O(4)-V(2)-O(15)	165.6(4)
O(9)-V(2)-O(15)	89.7(3)	O(11)-V(2)-O(15)	63.7(3)
O(15)-V(2)-O(16)	83.1(3)	O(15)-V(2)-N(31)	80.9(3)
O(2)-V(3)-O(5)	167.1(5)	O(5)-V(3)-O(6)	87.3(3)
O(2)-V(3)-O(12)	101.3(4)	O(5)-V(3)-O(12)	86.6(3)
O(6)-V(3)-O(12)	91.5(4)	O(2)-V(3)-N(11)	92.6(4)
O(5)-V(3)-N(11)	77.3(3)	O(6)-V(3)-N(11)	164.6(3)
O(12)-V(3)-N(11)	88.2(4)	O(2)-V(3)-N(21)	91.3(4)
O(5)-V(3)-N(21)	81.4(3)	O(6)-V(3)-N(21)	83.7(4)
O(12)-V(3)-N(21)	167.2(3)	N(11)-V(3)-N(21)	93.3(4)
O(1)-V(4)-O(3)	102.3(5)	O(1)-V(4)-O(8)	103.1(5)
O(3)-V(4)-O(8)	154.4(3)	O(1)-V(4)-O(10)	102.6(3)
O(3)-V(4)-O(10)	91.7(4)	O(8)-V(4)-O(10)	84.8(4)
O(1)-V(4)-O(16)	165.1(5)	O(3)-V(4)-O(16)	64.3(3)
O(8)-V(4)-O(16)	90.2(3)	O(10)-V(4)-O(16)	85.1(3)
O(16)-V(4)-N(41)	77.8(3)	O(1)-V(4)-N(41)	96.3(4)
O(3)-V(4)-N(41)	89.8(4)	O(8)-V(4)-N(41)	85.4(4)
O(10)-V(4)-N(41)	160.2(4)	O(11)-P(1)-O(12)	113.3(5)
O(11)-P(1)-O(15)	102.8(4)	O(12)-P(1)-O(15)	116.3(5)
O(11)-P(1)-C(71)	111.2(5)	O(12)-P(1)-C(71)	105.8(5)
O(15)-P(1)-C(71)	107.4(6)	O(3)-P(2)-O(5)	114.1(4)
O(3)-P(2)-O(16)	103.4(6)	O(5)-P(2)-O(16)	114.6(5)
O(3)-P(2)-C(81)	108.3(5)	O(5)-P(2)-C(81)	106.5(6)
O(16)-P(2)-C(81)	109.8(6)	O(6)-P(3)-O(7)	115.6(5)
O(6)-P(3)-O(10)	113.9(5)	O(7)-P(3)-O(10)	105.3(5)
O(6)-P(3)-C(61)	104.0(6)	O(7)-P(3)-C(61)	108.4(6)
O(10)-P(3)-C(61)	109.5(5)	O(8)-P(4)-O(9)	112.2(5)
O(8)-P(4)-O(14)	111.6(4)	O(9)-P(4)-O(14)	111.4(5)
O(8)-P(4)-C(91)	109.3(6)	O(9)-P(4)-C(91)	104.0(5)
O(14)-P(4)-C(91)	107.9(6)	V(4)-O(3)-P(2)	105.7(5)
V(3)-O(5)-P(2)	145.6(6)	V(3)-O(6)-P(3)	142.4(5)
V(1)-O(7)-P(3)	109.9(4)	V(4)-O(8)-P(4)	132.9(6)
V(2)-O(9)-P(4)	131.6(4)	V(1)-O(10)-P(3)	83.2(3)
V(4)-O(10)-P(3)	146.5(7)	V(2)-O(11)-P(1)	107.3(5)
V(3)-O(12)-P(1)	143.6(4)	V(1)-O(14)-P(4)	131.5(6)
V(1)-O(15)-P(1)	139.1(4)	V(2)-O(15)-P(1)	86.2(4)
V(2)-O(16)-P(2)	136.8(6)	V(4)-O(16)-P(2)	86.5(4)
V(3)-N(11)-N(12)	121.0(10)	V(3)-N(11)-C(11)	133.3(9)
N(12)-N(11)-C(11)	105.5(11)	N(11)-N(12)-C(13)	112.8(12)

N(11)-C(11)-C(12)	109.6(13)	C(11)-C(12)-C(13)	108.0(15)
N(12)-C(13)-C(12)	104.1(11)	N(12)-C(13)-C(14)	122.6(15)
C(12)-C(13)-C(14)	133.2(15)	C(13)-C(14)-C(15)	115.3(14)
C(13)-C(14)-C(16)	107.9(11)	C(15)-C(14)-C(16)	105.8(14)
C(13)-C(14)-C(17)	111.3(14)	C(15)-C(14)-C(17)	107.5(12)
C(16)-C(14)-C(17)	108.7(14)	V(3)-N(21)-N(22)	123.2(10)
V(3)-N(21)-C(21)	133.4(9)	N(22)-N(21)-C(21)	103.5(11)
N(21)-N(22)-C(23)	113.0(13)	N(21)-C(21)-C(22)	112.5(14)
C(21)-C(22)-C(23)	105.6(16)	N(22)-C(23)-C(22)	105.5(12)
N(22)-C(23)-C(24)	122.0(15)	C(22)-C(23)-C(24)	132.4(16)
C(23)-C(24)-C(25)	110.2(13)	C(23)-C(24)-C(26)	111.1(15)
C(25)-C(24)-C(26)	106.2(15)	C(23)-C(24)-C(27)	110.2(14)
C(25)-C(24)-C(27)	109.1(15)	C(26)-C(24)-C(27)	110.0(15)
V(2)-N(31)-N(32)	124.3(8)	V(2)-N(31)-C(31)	130.5(10)
N(32)-N(31)-C(31)	105.2(11)	N(31)-N(32)-C(33)	111.7(11)
N(31)-C(31)-C(32)	110.6(13)	C(31)-C(32)-C(33)	104.4(13)
N(32)-C(33)-C(32)	108.1(14)	N(32)-C(33)-C(34)	121.3(12)
C(32)-C(33)-C(34)	130.6(14)	C(33)-C(34)-C(35)	108.0(15)
C(33)-C(34)-C(35')	114.7(14)	C(33)-C(34)-C(36)	113.1(11)
C(35)-C(34)-C(36)	105.8(13)	C(33)-C(34)-C(36')	107.8(12)
C(33)-C(34)-C(37)	117.6(12)	C(33)-C(34)-C(37')	116.2(13)
V(4)-N(41)-N(42)	123.2(8)	V(4)-N(41)-C(41)	132.4(9)
N(42)-N(41)-C(41)	104.1(9)	N(41)-N(42)-C(43)	115.3(11)
N(41)-C(41)-C(42)	110.2(13)	C(41)-C(42)-C(43)	108.0(11)
N(42)-C(43)-C(42)	102.4(11)	N(42)-C(43)-C(44)	125.4(13)
C(42)-C(43)-C(44)	132.2(11)	C(43)-C(44)-C(45)	112.2(14)
C(43)-C(44)-C(46)	108.7(12)	C(45)-C(44)-C(46)	110.5(14)
C(43)-C(44)-C(47)	110.5(13)	C(45)-C(44)-C(47)	108.4(12)
C(46)-C(44)-C(47)	106.4(15)	V(1)-N(51)-N(52)	124.4(7)
V(1)-N(51)-C(51)	131.0(8)	N(52)-N(51)-C(51)	104.4(10)
N(51)-N(52)-C(53)	112.5(9)	N(51)-C(51)-C(52)	110.9(11)
C(51)-C(52)-C(53)	107.5(12)	N(52)-C(53)-C(52)	104.6(11)
N(52)-C(53)-C(54)	122.7(10)	C(52)-C(53)-C(54)	132.7(12)
C(53)-C(54)-C(55)	112.1(11)	C(53)-C(54)-C(56)	108.5(14)
C(55)-C(54)-C(56)	104.3(13)	C(53)-C(54)-C(57)	112.2(13)
C(55)-C(54)-C(57)	109.9(15)	C(56)-C(54)-C(57)	109.5(14)
P(3)-C(61)-C(62)	120.6(10)	P(3)-C(61)-C(66)	122.1(11)
C(62)-C(61)-C(66)	117.1(13)	C(61)-C(62)-C(63)	122.8(14)
C(62)-C(63)-C(64)	120.0(17)	C(63)-C(64)-C(65)	121.2(16)
C(64)-C(65)-C(66)	117.9(13)	C(61)-C(66)-C(65)	120.8(14)
P(1)-C(71)-C(72)	121.8(9)	P(1)-C(71)-C(76)	121.3(9)
C(72)-C(71)-C(76)	116.9(11)	C(71)-C(72)-C(73)	121.1(12)
C(72)-C(73)-C(74)	119.1(14)	C(73)-C(74)-C(75)	122.4(14)
C(74)-C(75)-C(76)	118.5(13)	C(71)-C(76)-C(75)	122.0(12)
P(2)-C(81)-C(82)	123.2(13)	P(2)-C(81)-C(86)	120.5(9)
C(82)-C(81)-C(86)	116.3(12)	C(81)-C(82)-C(83)	124.3(16)
C(82)-C(83)-C(84)	118.1(13)	C(83)-C(84)-C(85)	118.4(14)
C(84)-C(85)-C(86)	121.7(20)	C(81)-C(86)-C(85)	120.5(14)
P(4)-C(91)-C(92)	120.7(11)	P(4)-C(91)-C(96)	120.1(11)
C(92)-C(91)-C(96)	119.2(14)	C(91)-C(92)-C(93)	117.9(13)
C(92)-C(93)-C(94)	121.0(14)	C(93)-C(94)-C(95)	120.9(17)
C(94)-C(95)-C(96)	119.6(16)	C(91)-C(96)-C(95)	121.4(15)
C(1)-C(2)-N(5)	175.3(22)	C(3)-C(4)-N(2)	176.7(19)
C(5)-C(6)-N(3)	176.7(15)	C(7)-C(8)-N(4)	175.5(29)

Table S9. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for
 $(t\text{-Bupz})_5\text{V}_4\text{O}_4(\mu\text{-C}_6\text{H}_5\text{PO}_3)_4 \cdot 4\text{CH}_3\text{CN} \cdot 0.6\text{H}_2\text{O}$.

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
V(1)	35(1)	29(1)	43(1)	-15(1)	-1(1)	-6(1)
V(2)	33(1)	27(1)	40(1)	-15(1)	1(1)	-6(1)
V(3)	32(1)	39(1)	46(1)	-22(1)	-2(1)	-3(1)
V(4)	35(1)	37(1)	38(1)	-21(1)	4(1)	-5(1)
P(1)	32(2)	36(2)	38(2)	-18(1)	3(2)	-5(2)
P(2)	33(2)	33(2)	38(2)	-20(1)	2(2)	-9(2)
P(3)	30(2)	34(2)	41(2)	-16(1)	-3(2)	1(2)
P(4)	36(2)	30(2)	46(2)	-20(1)	-3(2)	-6(2)
O(1)	57(3)	45(3)	43(3)	-31(2)	-4(2)	-2(2)
O(2)	33(2)	60(3)	52(3)	-20(2)	-14(2)	5(3)
O(3)	43(2)	51(3)	32(3)	-32(2)	-4(2)	-9(2)
O(4)	42(3)	39(3)	55(3)	-15(2)	1(2)	-11(2)
O(5)	40(2)	37(2)	37(3)	-23(2)	3(2)	-5(2)
O(6)	53(3)	50(3)	33(3)	-36(2)	-2(2)	-1(2)
O(7)	36(2)	43(2)	47(3)	-21(2)	7(2)	-16(2)
O(8)	35(2)	41(3)	39(3)	-18(2)	1(2)	-3(2)
O(9)	20(2)	34(2)	49(3)	-17(2)	1(2)	-3(2)
O(10)	37(2)	35(2)	49(3)	-21(2)	4(2)	-8(2)
O(11)	29(2)	25(2)	47(3)	-15(2)	5(2)	-7(2)
O(12)	21(2)	33(2)	38(3)	-12(2)	2(2)	-1(2)
O(13)	48(3)	38(3)	49(3)	-18(2)	0(2)	-8(2)
O(14)	33(2)	42(2)	47(3)	-20(2)	-4(2)	-9(2)
O(15)	45(2)	42(2)	42(3)	-31(2)	4(2)	2(2)
O(16)	30(2)	40(2)	39(3)	-23(2)	-3(2)	-4(2)
N(11)	32(3)	47(3)	49(3)	-15(2)	-6(3)	-1(3)
N(12)	24(3)	48(3)	61(3)	-20(2)	-15(3)	1(3)
C(11)	31(3)	69(3)	91(3)	-37(3)	-11(3)	1(3)
C(12)	60(3)	51(3)	103(3)	-41(3)	-24(3)	12(3)
C(13)	54(3)	32(3)	65(3)	-26(3)	3(3)	13(3)
C(14)	59(3)	55(3)	86(3)	-20(3)	-29(3)	32(3)
C(15)	143(3)	84(3)	198(3)	-55(3)	-90(3)	77(3)
C(16)	120(3)	82(3)	118(3)	-36(3)	-10(3)	46(3)
C(17)	92(3)	118(3)	167(3)	-1(3)	-50(3)	-36(3)
N(21)	39(3)	43(3)	43(3)	-19(2)	-13(3)	-12(3)
N(22)	47(3)	74(3)	44(3)	-38(2)	7(3)	-15(3)
C(21)	65(3)	81(3)	66(3)	-41(3)	-24(3)	-12(3)
C(22)	68(3)	141(3)	61(3)	-59(3)	-23(3)	-28(3)
C(23)	54(3)	76(3)	50(3)	-37(3)	-10(3)	-8(3)
C(24)	103(3)	122(3)	44(3)	-65(3)	-1(3)	-41(3)
C(25)	117(3)	197(3)	119(3)	12(3)	64(3)	-4(3)
C(26)	260(3)	298(3)	86(3)	-199(3)	76(3)	-98(3)
C(27)	120(3)	309(3)	59(3)	-110(3)	26(3)	-71(3)
N(31)	45(3)	52(3)	40(3)	-33(2)	0(3)	-18(3)
N(32)	88(3)	22(3)	46(3)	-24(2)	-22(3)	-1(3)
C(31)	60(3)	44(3)	33(3)	-21(3)	-4(3)	2(3)
C(32)	80(3)	38(3)	43(3)	-4(3)	-10(3)	-6(3)
C(33)	94(3)	47(3)	61(3)	-36(3)	-25(3)	-12(3)
C(34)	108(3)	52(3)	134(3)	-15(3)	-39(3)	-40(3)
C(35)	193(3)	183(3)	187(3)	-89(3)	-37(3)	-20(3)

C(35')	221(3)	95(3)	147(3)	-62(3)	-57(3)	-45(3)
C(36)	155(3)	35(3)	70(3)	-35(3)	-95(3)	21(3)
C(36')	201(3)	198(3)	194(3)	-102(3)	-60(3)	-32(3)
C(37)	224(3)	90(3)	156(3)	-96(3)	-54(3)	-36(3)
C(37')	127(3)	56(3)	69(3)	-26(3)	-35(3)	-20(3)
N(41)	36(3)	45(3)	41(3)	-22(2)	4(3)	-5(3)
N(42)	28(3)	40(3)	54(3)	-10(2)	-3(3)	-15(3)
C(41)	63(3)	64(3)	42(3)	-26(3)	23(3)	-28(3)
C(42)	45(3)	67(3)	77(3)	-7(3)	11(3)	-35(3)
C(43)	29(3)	44(3)	72(3)	-8(3)	0(3)	-15(3)
C(44)	45(3)	46(3)	97(3)	-13(3)	-1(3)	-10(3)
C(45)	69(3)	116(3)	149(3)	38(3)	-17(3)	-48(3)
C(46)	139(3)	83(3)	141(3)	-21(3)	-86(3)	18(3)
C(47)	109(3)	116(3)	129(3)	-31(3)	-27(3)	47(3)
N(51)	44(3)	33(3)	47(3)	-23(2)	-6(3)	-5(3)
N(52)	53(3)	30(3)	52(3)	-21(2)	7(3)	-3(3)
C(51)	66(3)	46(3)	57(3)	-25(3)	14(3)	-18(3)
C(52)	93(3)	27(3)	79(3)	-27(3)	14(3)	-3(3)
C(53)	46(3)	24(3)	59(3)	-21(2)	-13(3)	1(3)
C(54)	75(3)	56(3)	59(3)	-43(3)	10(3)	-12(3)
C(55)	101(3)	162(3)	106(3)	-59(3)	35(3)	5(3)
C(56)	144(3)	145(3)	42(3)	-72(3)	-5(3)	-3(3)
C(57)	326(3)	152(3)	137(3)	-197(3)	123(3)	-63(3)
C(61)	23(3)	54(3)	70(3)	-25(2)	-3(3)	-10(3)
C(62)	126(3)	75(3)	62(3)	-62(3)	-19(3)	19(3)
C(63)	102(3)	124(3)	73(3)	-55(3)	0(3)	-11(3)
C(64)	89(3)	120(3)	86(3)	-44(3)	-32(3)	59(3)
C(65)	93(3)	44(3)	147(3)	-12(3)	-29(3)	42(3)
C(66)	62(3)	63(3)	61(3)	-6(3)	-11(3)	-7(3)
C(71)	26(3)	29(3)	40(3)	-13(2)	4(3)	-1(3)
C(72)	60(3)	46(3)	58(3)	-8(3)	10(3)	-10(3)
C(73)	144(3)	94(3)	34(3)	-33(3)	40(3)	-7(3)
C(74)	83(3)	120(3)	40(3)	-24(3)	11(3)	-33(3)
C(75)	117(3)	62(3)	50(3)	-26(3)	19(3)	-30(3)
C(76)	56(3)	39(3)	46(3)	-7(3)	-2(3)	-9(3)
C(81)	40(3)	38(3)	51(3)	-27(2)	-6(3)	-5(3)
C(82)	49(3)	39(3)	85(3)	-19(3)	15(3)	-25(3)
C(83)	65(3)	53(3)	77(3)	-20(3)	9(3)	-20(3)
C(84)	101(3)	34(3)	70(3)	-4(3)	-1(3)	-24(3)
C(85)	147(3)	68(3)	172(3)	-63(3)	-63(3)	-40(3)
C(86)	54(3)	59(3)	165(3)	-9(3)	-52(3)	-44(3)
C(91)	42(3)	15(3)	49(3)	-10(2)	-11(3)	-12(3)
C(92)	46(3)	69(3)	69(3)	-38(3)	13(3)	-26(3)
C(93)	40(3)	64(3)	87(3)	-28(3)	13(3)	-11(3)
C(94)	50(3)	55(3)	121(3)	-29(3)	-24(3)	-4(3)
C(95)	83(3)	74(3)	121(3)	-57(3)	-56(3)	36(3)
C(96)	71(3)	80(3)	101(3)	-52(3)	-24(3)	14(3)
C(1)	194(3)	166(3)	115(3)	-78(3)	31(3)	11(3)
C(2)	166(3)	115(3)	204(3)	-101(3)	-10(3)	32(3)
C(3)	320(3)	163(3)	372(3)	-147(3)	-268(3)	128(3)
C(4)	150(3)	133(3)	149(3)	-50(3)	-63(3)	23(3)
N(2)	214(3)	424(3)	126(3)	-186(3)	-14(3)	-28(3)
C(5)	37(3)	138(3)	223(3)	-10(3)	-7(3)	-50(3)
C(6)	94(3)	87(3)	254(3)	-18(3)	-29(3)	-42(3)

N(3)	203(3)	156(3)	325(3)	-103(3)	-21(3)	-5(3)
C(7)	309(3)	363(3)	503(3)	-247(3)	-232(3)	102(3)
C(8)	244(3)	186(3)	183(3)	-72(3)	-124(3)	27(3)
N(4)	912(3)	859(3)	839(3)	-403(3)	-77(3)	4(3)
N(5)	118(3)	297(3)	519(3)	-128(3)	-104(3)	136(3)
O(1W)	202(3)	331(3)	144(3)	-81(3)	-9(3)	-54(3)

Table S10. Hydrogen atom coordinates for $(t\text{-Bupz})_5\text{V}_4\text{O}_4(\mu\text{-C}_6\text{H}_5\text{PO}_3)_4 \cdot 4\text{CH}_3\text{CN} \cdot 0.6\text{H}_2\text{O}$.

<u>Atom</u>	<u>X</u>	<u>Y</u>	<u>Z</u>	<u>U</u>
H(12B)	7824	8729	1118	80
H(11A)	10170	8267	1853	80
H(12A)	9639	9769	1182	80
H(15A)	6696	10005	642	80
H(15B)	6741	10639	-30	80
H(15C)	7466	9600	-32	80
H(16A)	8966	10659	78	80
H(16B)	8832	9989	-373	80
H(16C)	8107	11029	-371	80
H(17A)	6890	11043	1411	80
H(17B)	7751	11315	1175	80
H(17C)	6902	11657	720	80
H(22B)	7571	7464	3639	80
H(21A)	10091	7444	3166	80
H(22A)	9518	7805	4455	80
H(25A)	6410	8876	4501	80
H(25B)	6282	8969	5327	80
H(25C)	7027	9248	4817	80
H(26A)	6768	7281	4685	80
H(26B)	7603	6629	5117	80
H(26C)	6641	7367	5512	80
H(27A)	7572	7978	6032	80
H(27B)	8535	7244	5635	80
H(27C)	8299	8283	5527	80
H(32B)	5852	6624	165	80
H(31A)	5390	9069	-572	80
H(32A)	5419	8176	-1587	80
H(35A)**	7240	5638	-1094	80
H(35B)**	6959	5236	-1679	80
H(35C)**	6920	6216	-1794	80
H(35D)**	6910	6102	-1928	80
H(35E)**	6266	5624	-2032	80
H(35F)**	5917	6672	-2212	80
H(36A)**	4514	6774	-1547	80
H(36B)**	5315	6938	-2087	80
H(36C)**	5354	5958	-1972	80
H(36D)**	4408	6923	-1414	80
H(36E)**	4789	5858	-1350	80
H(36F)**	4427	6427	-660	80
H(37A)**	4856	5832	-404	80
H(37B)**	5643	5123	-944	80
H(37C)**	5951	5395	-300	80
H(37D)**	6945	5295	-721	80
H(37E)**	5940	5458	-267	80
H(37F)**	6302	4889	-957	80
H(42B)	3817	8477	2311	80
H(41A)	3916	8635	4297	80
H(42A)	2366	9909	3989	80
H(45A)	953	10500	3271	80
H(45B)	1494	11122	3040	80

H(45C)	780	11102	2564	80
H(46A)	1434	9122	2594	80
H(46B)	1274	9732	1891	80
H(46C)	2276	8892	1949	80
H(47A)	3067	9871	1586	80
H(47B)	2069	10721	1538	80
H(47C)	2783	10741	2015	80
H(52B)	5669	5199	2805	80
H(51A)	7650	3174	1711	80
H(52A)	6907	2533	2774	80
H(55A)	4151	4680	3479	80
H(55B)	4148	4813	4281	80
H(55C)	4635	5270	3676	80
H(56A)	6590	3543	4345	80
H(56B)	6080	4600	4188	80
H(56C)	5594	4143	4793	80
H(57A)	5006	3011	3609	80
H(57B)	5974	2573	3940	80
H(57C)	4982	3099	4423	80
H(62A)	7619	5594	4046	80
H(63A)	8044	4522	4981	80
H(64A)	9019	3025	4745	80
H(65A)	9496	2533	3573	80
H(66A)	8881	3616	2633	80
H(72A)	7646	7916	-635	80
H(73A)	8494	7411	-1783	80
H(74A)	9457	5878	-1933	80
H(75A)	9707	4843	-985	80
H(76A)	8893	5349	158	80
H(82A)	4510	9754	2539	80
H(83A)	3736	11311	2803	80
H(84A)	4586	11907	3330	80
H(85A)	6224	11050	3330	80
H(86A)	6994	9520	2991	80
H(92A)	3331	6315	2610	80
H(93A)	1888	6277	2387	80
H(94A)	1402	6687	1275	80
H(95A)	2282	7175	369	80
H(96A)	3713	7227	573	80
H(1A)	7152	3372	6135	80
H(1B)	6884	2570	6068	80
H(1C)	7369	2938	5395	80
H(3A)	9538	5298	6923	80
H(3B)	10164	4473	6425	80
H(3C)	9665	5464	6093	80
H(5A)	9910	2688	-181	80
H(5B)	9837	3309	416	80
H(5C)	8892	3331	213	80
H(7A)	8200	144	3091	80
H(7B)	8815	697	3047	80
H(7C)	9148	-262	3446	80

** Site occupation factors for H35(A, B, C), H36(A, B, C), and H37(A, B, C) is 0.451(3); for H35(D, E, F), H36(D, E, F), and H37(D, E, F) is 0.549(3).

Table S11. Summary of Data Collection and Structure Determination Parameters for $\text{I} \cdot 4\text{CH}_3\text{CN}$.^a

Compound	$[\text{HB}(\text{pz})_3]_4\text{V}_4(\text{C}_6\text{H}_5\text{OPO}_3)_4 \cdot 4\text{CH}_3\text{CN}$
Formula	$\text{C}_{68}\text{H}_{72}\text{N}_{28}\text{O}_{16}\text{B}_4\text{P}_4\text{V}_4$
fw	1908.4
Color, habit	Green, block
Cryst size, mm	0.5 x 0.5 x 0.7
Crystal system	Triclinic
Space group	$\text{P}\bar{1}$
a , Å	15.495(3)
b , Å	17.000(3)
c , Å	17.949(4)
α , °	89.17(3)
β , °	86.00(3)
γ , °	78.60(3)
V , Å ³	4623.5(16)
Z	2
ρ_{calcd} , g cm ⁻³	1.371
$F(000)$	1952
Temp, K	298
Radiation (λ , Å)	Mo K α (0.71073)
μ , cm ⁻¹	5.35
Scan type	θ -2 θ
Data collection	3.5°–45°
Scan speed, deg min ⁻¹ in ω	Variable; 3.0 to 15.0
Scan range, deg in ω	1.20 + K α separation
Total reflections	13654
Unique reflections	12039
Reflections with $ F \geq 2\sigma(F)$	9058
Data to param ratio	8.1:1
Min/max transmission	0.7550, 0.8135
gof	1.45
R	0.0478
R_w	0.0655
Max diff peak, e/Å ³	0.86
Largest Δ/σ	0.014

^a Quantity minimized = $\sum w(F_o - F_c)^2$ where $w^{-1} = \sigma^2(F) + 0.0008F^2$, $R = \sum |F_o - F_c| / \sum F_o$ and $R_w = (\sum (w[F_o - F_c]^2 / \sum (wF_o^2)))^{1/2}$.

Table S12. Summary of Data Collection and Structure Determination Parameters for IV.^a

Compound	(<i>t</i> -Bupz) ₅ V ₄ O ₄ (C ₆ H ₅ PO ₃) ₄ ·4CH ₃ CN
Formula	C ₆₇ H ₉₂ N ₁₄ O _{18.6} P ₄ V ₄
fw	1686.8
Color, habit	Green, plate
Cryst size, mm	1.0 x 0.7 x 0.05
Crystal system	Triclinic
Space group	P $\bar{1}$
<i>a</i> , Å	15.541(3)
<i>b</i> , Å	16.340(2)
<i>c</i> , Å	19.069(5)
α , deg	83.58(2)
β , deg	79.67(2)
γ , deg	63.68(1)
<i>V</i> , Å ³	4267(2)
<i>Z</i>	2
ρ_{calcd} , g cm ⁻³	1.313
<i>F</i> (000)	1753.6
Temp, K	298
Radiation (λ , Å)	Mo K α (0.71073)
μ , cm ⁻¹	5.66
Scan type	θ -2 θ
Data collection	3.5°–45°
Scan speed, deg min ⁻¹ in ω	20.0
Scan range, deg in ω	1.20
Total reflections	12009
Unique reflections	11149
Reflections with $ F \geq 2\sigma(F)$	5004
Data to param ratio	5.1:1
Min/max transmission	0.814, 0.994
gof	1.49
<i>R</i>	0.0794
<i>R</i> _w	0.0789
Max diff peak, e/Å ³	0.70
Largest Δ/σ	0.514

^a Quantity minimized = $\sum w(F_o - F_c)^2$ where $w^{-1} = \sigma^2(F) + 0.0008F^2$, $R = \sum |F_o - F_c| / \sum F_o$ and $R_w = (\sum (w[F_o - F_c]^2 / \sum (wF_o^2)))^{1/2}$.

Figure 1s. (Top) Stereoview of the cluster framework of I-4CH₃CN. Non-carbon atoms are depicted as open circles and labels for the vanadium and phosphorus atoms are shown. One orientation of a disordered phenyl ring and all hydrogen atoms have been omitted to improve clarity. (Bottom) Stereoview of the cluster framework of IV. Non-carbon atoms are depicted as open circles. One orientation of a disordered *t*-butyl group and all hydrogen atoms are omitted to improve clarity. Weak axial V...O interactions are not depicted

