Supporting Information

A frontier Zn- and N-rich complex grafted onto reduced graphene oxide for electrocatalyst of dye-sensitized solar cells

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Table 1S. The crystallographic parameters of macrocyclic zinc complex, $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$.

Empirical formula	$C_{10}H_{20.5}B_{1.5}F_6N_8O_{0.5}Zn$	
Formula weight	456.43	
Temperature	150(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 25.4632(6) Å	α= 90°
	b = 10.9818(3) Å	β= 127.9356(8)°
	c = 15.7522(4) Å	γ = 90°
Volume	3474.09(16) $Å^3$	
Z	8	
Density (calculated)	1.745 mg/m^3	
Absorption coefficient	1.492 mm^{-1}	
F(000)	1856	
Crystal size	0.128 x 0.088 x 0.057 mm^3	
Theta range for data collection	2.028 to 27.492°	
Index ranges	$-32 \leq h \leq 32$, $-14 \leq k \leq 14$, $-19 \leq l \leq 20$	
Reflections collected	13171	
Independent reflections	3965 [R(int) = 0.0185]	
Completeness to theta = 25.242°	99.8 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7456 and 0.7108	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	3965 / 20 / 299	
Goodness-of-fit on F^2	1.076	
Final R indices [I>2sigma(I)]	R_1 = 0.0233, wR_2 = 0.0557	
R indices (all data)	R_1 = 0.0295, wR_2 = 0.0606	
Largest diff. peak and hole	0.374 and -0.310 $e \cdot Å^{-3}$	

Table 2S. Selected bond lengths and angles of $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$.

bond lengths (Å)		bond angles (°)	
Zn-O(1)	1.9342(5)	Zn-O(1)-ZnA	143.89(9)
O(1)-ZnA	1.9342(5)	O(1)-Zn-N(4)	115.23(4)
Zn-N(1)	2.1117(14)	O(1)-Zn-N(8)	111.36(4)
Zn-N(4)	2.0938(13)	O(1)-Zn-N(5)	119.21(6)
Zn-N(5)	2.1060(13)	O(1)-Zn-N(1)	112.64(6)
Zn-N(8)	2.0994(13)	N(4)-Zn-N(8)	133.27(5)
N(1)-C(6)	1.293(2)	N(5)-Zn-N(1)	128.15(5)
N(4)-C(2)	1.283(2)	N(4)-Zn-N(5)	76.27(5)
N(5)-C(3)	1.290(2)	N(8)-Zn-N(5)	84.60(5)
N(8)-C(5)	1.288(2)	N(4)-Zn-N(1)	83.12(5)
N(1)-N(2)	1.366(2)	N(8)-Zn-N(1)	76.00(5)
N(3)-N(4)	1.3921(19)	N(1)-N(2)-C(1)	114.76(14)
N(5)-N(6)	1.3726(19)	N(4)-N(3)-C(1)	110.84(12)
N(7)-N(8)	1.3827(19)	N(5)-N(6)-C(4)	113.41(13)
N(2)-C(1)	1.452(2)	N(8)-N(7)-C(4)	112.31(13)
N(3)-C(1)	1.458(2)	N(2)-C(1)-N(3)	115.21(14)
N(6)-C(4)	1.455(2)	N(6)-C(4)-N(7)	117.05(14)
N(7)-C(4)	1.460(2)	N(1)-C(6)-C(5)	114.59(14)
C(2)-C(3)	1.495(2)	N(4)-C(2)-C(3)	115.15(14)
C(5)-C(6)	1.491(2)	N(5)-C(3)-C(2)	115.04(14)
C(2)-C(7)	1.492(2)	N(8)-C(5)-C(6)	115.89(14)
C(3)-C(8)	1.497(2)	N(2)-N(1)-Zn	119.69(11)
C(5)-C(9)	1.492(2)	N(3)-N(4)-Zn	122.26(10)
C(6)-C(10)	1.495(2)	N(6)-N(5)-Zn	121.32(10)
		N(7)-N(8)-Zn	121.99(10)

Symmetry transformations used to generate equivalent atoms: -x+1,y,-z+1/2

Table 3S. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Zn	5825(1)	4897(1)	3826(1)	14(1)
O(1)	5000	5443(1)	2500	15(1)
N(1)	5802(1)	3037(1)	4142(1)	17(1)
N(2)	5869(1)	2167(1)	3595(1)	27(1)
N(3)	6455(1)	3309(1)	3062(1)	19(1)
N(4)	6552(1)	4308(1)	3689(1)	16(1)
N(5)	6607(1)	6151(1)	4771(1)	15(1)
N(6)	6580(1)	7033(1)	5360(1)	19(1)
N(7)	5817(1)	5989(1)	5609(1)	18(1)
N(8)	5771(1)	4919(1)	5104(1)	16(1)
C(1)	5863(1)	2628(2)	2725(1)	20(1)
C(2)	7120(1)	4840(2)	4296(1)	16(1)
C(3)	7166(1)	5839(2)	4984(1)	17(1)
C(4)	5956(1)	7040(2)	5208(1)	19(1)
C(5)	5798(1)	3879(2)	5506(1)	17(1)
C(6)	5790(1)	2790(2)	4933(1)	18(1)
C(7)	7712(1)	4461(2)	4379(2)	24(1)
C(8)	7833(1)	6332(2)	5896(1)	23(1)
C(9)	5875(1)	3755(2)	6520(1)	23(1)
C(10)	5811(1)	1535(2)	5320(2)	30(1)
B(1)	7406(1)	8846(2)	7865(2)	23(1)
F(1)	7622(1)	7919(1)	7540(1)	34(1)
F(2)	7747(1)	9909(1)	8030(1)	29(1)
F(3)	6726(1)	9014(1)	7090(1)	38(1)
F(4)	7531(1)	8487(1)	8826(1)	34(1)
B(2)	5070(5)	-884(4)	2494(7)	39(2)
F(5)	4874(3)	-2059(2)	2559(5)	54(1)
F(6)	5162(5)	-190(6)	3257(6)	43(1)
F(7)	4505(10)	-400(20)	1559(14)	91(3)
F(8)	5367(8)	-325(13)	2105(13)	88(2)
F(6')	5654(4)	-471(8)	3500(5)	43(1)
F(7')	4460(5)	-316(11)	1996(10)	91(3)
F(8')	5227(4)	-936(9)	1804(7)	88(2)

Table 4S. Bond lengths [Å] and angles [°] of all data for $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$.

Zn-O(1)	1.9342(5)	B(2)-F(7)	1.385(10)
Zn-N(4)	2.0938(13)	B(2)-F(7')	1.385(9)
Zn-N(8)	2.0994(13)	B(2)-F(5)	1.410(5)
Zn-N(5)	2.1060(13)	B(2)-F(6')	1.425(7)
Zn-N(1)	2.1117(14)	F(5)-F(5)#1	0.769(7)
O(1)-Zn#1	1.9342(5)	F(5)-B(2)#1	1.306(5)
N(1)-C(6)	1.293(2)	F(5)-F(8')#1	1.707(9)
N(1)-N(2)	1.366(2)	F(6)-F(7)#1	0.74(2)
N(2)-C(1)	1.452(2)	F(6)-F(8)#1	1.103(15)
N(3)-N(4)	1.3921(19)	F(6)-B(2)#1	1.212(11)
N(3)-C(1)	1.458(2)	F(7)-F(6)#1	0.74(2)
N(4)-C(2)	1.283(2)	F(7)-B(2)#1	1.303(19)
N(5)-C(3)	1.290(2)	F(8)-F(6)#1	1.103(15)
N(5)-N(6)	1.3726(19)	F(8)-B(2)#1	1.697(13)
N(6)-C(4)	1.455(2)	F(6')-F(7')#1	0.667(13)
N(7)-N(8)	1.3827(19)	F(6')-B(2)#1	1.583(15)
N(7)-C(4)	1.460(2)	F(7')-F(6')#1	0.667(13)
N(8)-C(5)	1.288(2)	F(7')-B(2)#1	1.133(13)
C(2)-C(7)	1.492(2)	F(7')-F(8')#1	1.679(12)
C(2)-C(3)	1.495(2)	F(8')-F(7')#1	1.680(12)
C(3)-C(8)	1.497(2)	F(8')-B(2)#1	1.683(10)
C(5)-C(6)	1.491(2)	F(8')-F(5)#1	1.707(9)
C(5)-C(9)	1.492(2)		
C(6)-C(10)	1.495(2)	O(1)-Zn-N(4)	115.23(4)
B(1)-F(2)	1.382(2)	O(1)-Zn-N(8)	111.36(4)
B(1)-F(3)	1.387(2)	N(4)-Zn-N(8)	133.27(5)
B(1)-F(1)	1.395(2)	O(1)-Zn-N(5)	119.21(6)
B(1)-F(4)	1.399(2)	N(4)-Zn-N(5)	76.27(5)
B(2)-B(2)#1	0.371(15)	N(8)-Zn-N(5)	84.60(5)
B(2)-F(7')#1	1.133(13)	O(1)-Zn-N(1)	112.64(6)
B(2)-F(6)#1	1.212(10)	N(4)-Zn-N(1)	83.12(5)
B(2)-F(7)#1	1.303(19)	N(8)-Zn-N(1)	76.00(5)
B(2)-F(5)#1	1.306(5)	N(5)-Zn-N(1)	128.15(5)
B(2)-F(6)	1.318(7)	Zn-O(1)-Zn#1	143.89(9)
B(2)-F(8')	1.370(8)	C(6)-N(1)-N(2)	123.21(15)
B(2)-F(8)	1.375(9)	C(6)-N(1)-Zn	116.73(11)
		N(2)-N(1)-Zn	119.69(11)

N(1)-N(2)-C(1)	114.76(14)	F(7')#1-B(2)-F(5)#1	118.8(10)
N(4)-N(3)-C(1)	110.84(12)	F(6)#1-B(2)-F(5)#1	124.5(8)
C(2)-N(4)-N(3)	120.99(14)	F(7)#1-B(2)-F(5)#1	115.6(15)
C(2)-N(4)-Zn	116.41(11)	B(2)#1-B(2)-F(6)	66(2)
N(3)-N(4)-Zn	122.26(10)	F(6)#1-B(2)-F(6)	103.4(8)
C(3)-N(5)-N(6)	121.15(14)	F(7)#1-B(2)-F(6)	32.9(10)
C(3)-N(5)-Zn	115.00(11)	F(5)#1-B(2)-F(6)	131.6(7)
N(6)-N(5)-Zn	121.32(10)	B(2)#1-B(2)-F(8')	144(3)
N(5)-N(6)-C(4)	113.41(13)	F(7')#1-B(2)-F(8')	83.7(8)
N(8)-N(7)-C(4)	112.31(13)	F(5)#1-B(2)-F(8')	79.2(6)
C(5)-N(8)-N(7)	120.66(14)	B(2)#1-B(2)-F(8)	147(2)
C(5)-N(8)-Zn	116.50(11)	F(6)#1-B(2)-F(8)	50.0(8)
N(7)-N(8)-Zn	121.99(10)	F(7)#1-B(2)-F(8)	91.7(12)
N(2)-C(1)-N(3)	115.21(14)	F(5)#1-B(2)-F(8)	107.6(8)
N(4)-C(2)-C(7)	123.62(15)	F(6)-B(2)-F(8)	108.0(7)
N(4)-C(2)-C(3)	115.15(14)	B(2)#1-B(2)-F(7)	70(3)
C(7)-C(2)-C(3)	121.13(15)	F(6)#1-B(2)-F(7)	32.4(10)
N(5)-C(3)-C(2)	115.04(14)	F(7)#1-B(2)-F(7)	130(2)
N(5)-C(3)-C(8)	124.95(15)	F(5)#1-B(2)-F(7)	113.3(13)
C(2)-C(3)-C(8)	119.84(14)	F(6)-B(2)-F(7)	103.4(12)
N(6)-C(4)-N(7)	117.05(14)	F(8)-B(2)-F(7)	81.4(13)
N(8)-C(5)-C(6)	115.89(14)	B(2)#1-B(2)-F(7')	41.1(19)
N(8)-C(5)-C(9)	122.70(15)	F(7')#1-B(2)-F(7')	118.7(12)
C(6)-C(5)-C(9)	121.30(15)	F(5)#1-B(2)-F(7')	122.5(9)
N(1)-C(6)-C(5)	114.59(14)	F(8')-B(2)-F(7')	110.0(9)
N(1)-C(6)-C(10)	124.74(16)	B(2)#1-B(2)-F(5)	66.3(3)
C(5)-C(6)-C(10)	120.56(15)	F(7')#1-B(2)-F(5)	136.0(10)
F(2)-B(1)-F(3)	110.68(16)	F(6)#1-B(2)-F(5)	131.6(8)
F(2)-B(1)-F(1)	110.00(15)	F(7)#1-B(2)-F(5)	112.0(13)
F(3)-B(1)-F(1)	109.54(15)	F(5)#1-B(2)-F(5)	32.6(3)
F(2)-B(1)-F(4)	109.64(15)	F(6)-B(2)-F(5)	109.6(6)
F(3)-B(1)-F(4)	108.68(15)	F(8')-B(2)-F(5)	108.0(6)
F(1)-B(1)-F(4)	108.25(16)	F(8)-B(2)-F(5)	139.2(8)
B(2)#1-B(2)-F(7')#1	126(2)	F(7)-B(2)-F(5)	104.3(13)
B(2)#1-B(2)-F(6)#1	98(2)	F(7')-B(2)-F(5)	97.4(7)
B(2)#1-B(2)-F(7)#1	95(3)	B(2)#1-B(2)-F(6')	109(3)
F(6)#1-B(2)-F(7)#1	115.0(13)	F(7')#1-B(2)-F(6')	27.3(7)
B(2)#1-B(2)-F(5)#1	98.6(4)	F(5)#1-B(2)-F(6')	107.8(7)

F(8')-B(2)-F(6')	106.4(7)	B(2)-F(8)-B(2)#1	6.9(5)
F(7')-B(2)-F(6')	121.7(8)	F(7')#1-F(6')-B(2)	51.1(10)
F(5)-B(2)-F(6')	112.6(7)	F(7')#1-F(6')-B(2)#1	60.7(11)
F(5)#1-F(5)-B(2)#1	81.1(4)	B(2)-F(6')-B(2)#1	12.8(5)
F(5)#1-F(5)-B(2)	66.2(3)	F(6')#1-F(7')-B(2)#1	101.5(13)
B(2)#1-F(5)-B(2)	15.1(6)	F(6')#1-F(7')-B(2)	94.4(15)
F(5)#1-F(5)-F(8')#1	126.2(6)	B(2)#1-F(7')-B(2)	12.4(6)
B(2)#1-F(5)-F(8')#1	52.0(4)	F(6')#1-F(7')-F(8')#1	141.1(16)
B(2)-F(5)-F(8')#1	64.5(4)	B(2)#1-F(7')-F(8')#1	54.2(6)
F(7)#1-F(6)-F(8)#1	154(2)	B(2)-F(7')-F(8')#1	65.8(6)
F(7)#1-F(6)-B(2)#1	86.7(12)	B(2)-F(8')-F(7')#1	42.1(5)
F(8)#1-F(6)-B(2)#1	72.7(8)	B(2)-F(8')-B(2)#1	7.5(7)
F(7)#1-F(6)-B(2)	72.4(14)	F(7')#1-F(8')-B(2)#1	48.7(4)
F(8)#1-F(6)-B(2)	88.6(9)	B(2)-F(8')-F(5)#1	48.7(3)
B(2)#1-F(6)-B(2)	16.2(7)	F(7')#1-F(8')-F(5)#1	76.7(5)
F(6)#1-F(7)-B(2)#1	74.7(13)	B(2)#1-F(8')-F(5)#1	49.1(3)
F(6)#1-F(7)-B(2)	60.9(10)		
B(2)#1-F(7)-B(2)	15.5(7)	Symmetry transformations used to generate	
F(6)#1-F(8)-B(2)	57.3(7)	equivalent atoms:	
F(6)#1-F(8)-B(2)#1	50.9(5)	#1 -x+1,y,-z+1/2	

Table 5S. Anisotropic displacement parameters ($Å^2x10^3$) for $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$. The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2}U^{11} + ... + 2 h k a^* b^* U^{12}]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Zn	13(1)	16(1)	11(1)	-1(1)	6(1)	0(1)
O(1)	14(1)	13(1)	12(1)	0	5(1)	0
N(1)	19(1)	16(1)	16(1)	-2(1)	11(1)	-1(1)
N(2)	47(1)	15(1)	31(1)	-4(1)	30(1)	-3(1)
N(3)	17(1)	21(1)	17(1)	-2(1)	10(1)	3(1)
N(4)	18(1)	18(1)	12(1)	1(1)	9(1)	2(1)
N(5)	17(1)	14(1)	13(1)	0(1)	9(1)	-1(1)
N(6)	18(1)	18(1)	16(1)	-5(1)	9(1)	-3(1)
N(7)	20(1)	22(1)	14(1)	-4(1)	10(1)	0(1)
N(8)	14(1)	20(1)	12(1)	-3(1)	7(1)	-1(1)
C(1)	21(1)	21(1)	16(1)	-4(1)	10(1)	0(1)
C(2)	16(1)	19(1)	14(1)	5(1)	9(1)	2(1)
C(3)	17(1)	18(1)	14(1)	3(1)	9(1)	-2(1)
C(4)	20(1)	18(1)	18(1)	-2(1)	10(1)	2(1)
C(5)	13(1)	24(1)	14(1)	1(1)	8(1)	-1(1)
C(6)	16(1)	19(1)	18(1)	1(1)	10(1)	-1(1)
C(7)	18(1)	30(1)	25(1)	-1(1)	14(1)	0(1)
C(8)	17(1)	26(1)	21(1)	-1(1)	9(1)	-5(1)
C(9)	26(1)	30(1)	18(1)	2(1)	16(1)	1(1)
C(10)	44(1)	22(1)	34(1)	3(1)	30(1)	-2(1)
B(1)	22(1)	24(1)	20(1)	-5(1)	12(1)	-5(1)
F(1)	34(1)	35(1)	32(1)	-14(1)	19(1)	-3(1)
F(2)	31(1)	27(1)	25(1)	-3(1)	16(1)	-10(1)
F(3)	22(1)	41(1)	34(1)	-8(1)	10(1)	-4(1)
F(4)	49(1)	29(1)	27(1)	-6(1)	25(1)	-13(1)
B(2)	36(4)	25(2)	57(3)	-11(4)	28(3)	-8(3)
F(5)	84(4)	14(1)	39(2)	2(1)	24(2)	-2(1)
F(6)	55(3)	29(2)	29(2)	-17(2)	17(2)	-10(2)
F(7)	87(5)	61(4)	139(7)	46(5)	77(5)	30(3)
F(8)	94(4)	110(6)	93(4)	-30(4)	73(4)	-20(4)
F(6')	55(3)	29(2)	29(2)	-17(2)	17(2)	-10(2)
F(7')	87(5)	61(4)	139(7)	46(5)	77(5)	30(3)
F(8')	94(4)	110(6)	93(4)	-30(4)	73(4)	-20(4)

Table 6S. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for {[Zn($C_{10}H_{20}N_8$)]$_2$(OH)}(BF_4)$_3$.

	x	y	z	U(eq)
H(1)	5000	6189(9)	2500	32(9)
H(2)	5759(11)	1470(20)	3590(18)	32(6)
H(3)	6796(10)	2880(18)	3352(16)	18(5)
H(6)	6930(10)	7065(18)	6022(17)	19(5)
H(7)	6071(10)	5937(18)	6284(18)	21(5)
H(1A)	5470	3162	2264	24
H(1B)	5809	1931	2278	24
H(4A)	5588	7118	4428	23
H(4B)	5948	7777	5562	23
H(7C)	7950	3811	4915	36
H(7D)	8010	5160	4597	36
H(7E)	7567	4162	3678	36
H(8A)	7774	7081	6166	35
H(8B)	8093	6510	5642	35
H(8C)	8070	5728	6476	35
H(9A)	6337	3928	7138	35
H(9B)	5760	2924	6577	35
H(9C)	5578	4333	6512	35
H(10A)	5731	937	4790	45
H(10B)	5466	1455	5414	45
H(10C)	6249	1389	6009	45

Table 7S. Hydrogen bonds for $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$ [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
O(1)-H(1)...F(5)#2	0.820(10)	1.962(10)	2.771(3)	168.70(12)
O(1)-H(1)...F(5)#3	0.820(10)	1.962(10)	2.771(3)	168.70(12)
N(2)-H(2)...F(6)	0.81(2)	2.22(3)	3.008(8)	163(2)
N(2)-H(2)...F(7)#1	0.81(2)	2.13(3)	2.93(2)	173(2)
N(2)-H(2)...F(6')	0.81(2)	2.14(3)	2.935(9)	165(2)
N(2)-H(2)...F(7')#1	0.81(2)	2.09(3)	2.836(10)	152(2)
N(3)-H(3)...F(4)#4	0.84(2)	2.14(2)	2.9648(18)	167.9(19)
N(6)-H(6)...F(1)	0.86(2)	2.13(2)	2.9287(19)	155.0(18)
N(7)-H(7)...N(3)#5	0.84(2)	2.47(2)	3.232(2)	150.6(18)
C(1)-H(1A)...N(1)#1	0.99	2.57	3.390(2)	140.5
C(4)-H(4A)...F(5)#2	0.99	2.49	3.442(6)	160.8
C(4)-H(4B)...F(3)	0.99	2.39	3.191(2)	137.8
C(7)-H(7C)...N(2)#6	0.98	2.65	3.501(3)	145.2
C(7)-H(7E)...F(2)#7	0.98	2.51	3.285(2)	136.4
C(8)-H(8A)...F(1)	0.98	2.59	3.428(2)	143.5
C(9)-H(9A)...F(1)#8	0.98	2.62	3.273(2)	124.4
C(10)-H(10A)...F(6)	0.98	2.27	3.205(6)	158.9
C(10)-H(10A)...F(7)#1	0.98	2.34	3.314(19)	176.3
C(10)-H(10A)...F(6')	0.98	2.46	3.437(7)	173.1

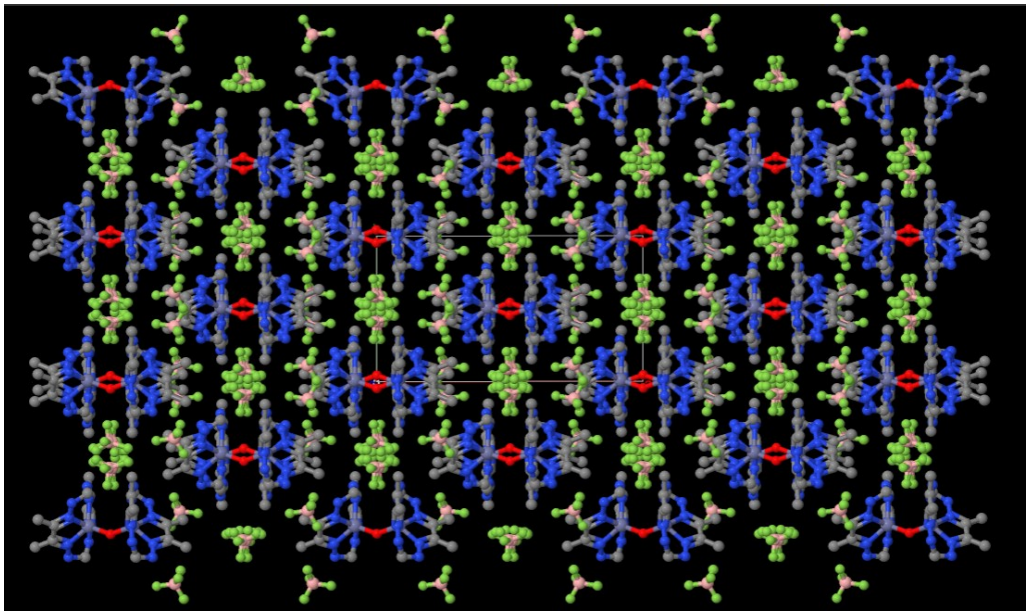
Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,-z+1/2 #2 x,y+1,z #3 -x+1,y+1,-z+1/2

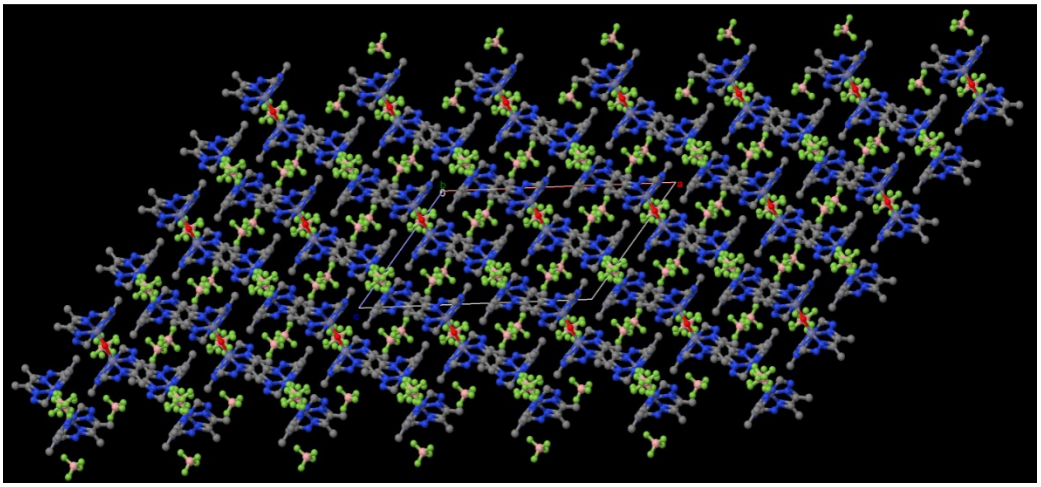
#4 x,-y+1,z-1/2 #5 x,-y+1,z+1/2 #6 -x+3/2,-y+1/2,-z+1

#7 -x+3/2,-y+3/2,-z+1 #8 -x+3/2,y-1/2,-z+3/2

(a)



(b)



(c)

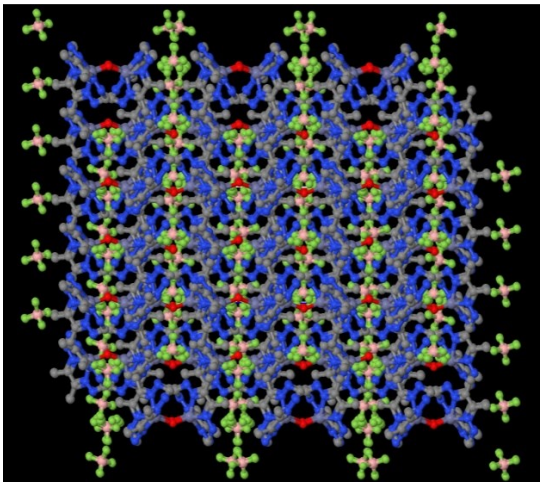


Figure 1S. Crystal packing diagrams of $\{[Zn(C_{10}H_{20}N_8)]_2(OH)\}(BF_4)_3$ viewed

perpendicular to (a) ab plane, (b) ac plane, and (c) bc plane.