Electronic Supplementary Information

The coordination chemistry of benzhydrazide with lanthanide(III) ions: hydrothermal in situ ligand formation, structures, magnetic, and photoluminescence sensing properties

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Table S1 Selected bond lengths (Å) and bond angles (°) for **1-5**

	$\mathbf{1}_{\mathbf{Sm}}$	$\mathbf{2}_{\mathbf{Eu}}$	$\mathbf{3}_{\mathbf{Gd}}$	$\mathbf{4}_{\mathbf{Tb}}$	$\mathbf{5}_{\mathbf{Dy}}$
Ln1−O1	2.395(3)	2.380(4)	2.369(3)	2.355(4)	2.343(3)
Ln1−O2	2.420(3)	2.409(4)	2.404(2)	2.392(3)	2.381(3)
Ln1−O3	2.542(7)	2.522(8)	2.508(5)	2.509(7)	2.497(5)
Ln1−N1	2.620(5)	2.601(7)	2.592(4)	2.574(6)	2.559(5)
Ln1−N3	2.644(4)	2.630(6)	2.615(3)	2.595(5)	2.585(4)
O1−Ln1−O1^{i}	103.42(16)	102.78(19)	102.77(12)	102.76(18)	102.63(14)
O1−Ln1−O2	74.46(11)	74.41(14)	74.19(9)	74.10(13)	73.91(10)
O1−Ln1−O2^{i}	71.80(11)	71.43(13)	71.48(9)	71.44(13)	71.35(10)
O1−Ln1−O3	113.12(15	113.55(19)	113.85(12)	113.98(17)	114.16(13)
O1^{i}−Ln1−O3	143.45(15)	143.66(19)	143.36(12)	143.24(17)	143.18(13)
O1−Ln1−N1^{i}	145.64(14)	145.57(17)	145.34(11)	145.13(16)	145.19(13)
O1−Ln1−N1	63.28(15)	63.91(19)	64.00(12)	64.54(18)	64.73(14)
O1−Ln1−N3^{i}	132.94(12)	132.90(15)	132.98(9)	133.39(14)	133.45(10)
O1−Ln1−N3	75.15(14)	75.43(17)	75.60(11)	75.51(16)	75.70(12)
O2^{i}−Ln1−O2	124.17(16)	123.9(2)	123.59(13)	123.34(19)	122.96(15)
O2−Ln1−O3	118.43(16)	118.7(2)	118.85(13)	118.84(19)	119.09(14)
O2^{i}−Ln1−O3	115.31(17)	115.4(2)	115.56(13)	115.83(19)	115.98(15)
O2−Ln1−N1^{i}	71.28(15)	71.24(18)	71.22(11)	71.07(17)	71.30(13)
O2−Ln1−N1	127.22(16)	127.7(2)	127.80(13)	128.42(19)	128.44(15)
O2−Ln1−N3^{i}	141.63(13)	141.46(17)	141.72(11)	141.46(16)	141.59(12)
O2−Ln1−N3	62.42(11)	62.70(14)	62.80(9)	63.24(13)	63.46(10)
O3−Ln1−N1^{i}	85.95(19)	85.5(2)	85.31(15)	84.8(2)	84.56(17)
O3−Ln1−N1	59.02(18)	58.8(2)	59.10(14)	59.1(2)	58.97(16)
O3−Ln1−N3	61.76(16)	61.8(2)	61.83(13)	61.53(19)	61.49(15)
O3−Ln1−N3^{i}	79.13(17)	79.0(2)	78.54(13)	78.45(19)	78.15(15)
N1−Ln1−N1^{i}	144.7(2)	144.1(3)	144.14(19)	143.5(3)	143.2(2)
N1−Ln1−N3	76.82(16)	76.9(2)	77.19(12)	77.24(19)	77.18(15)
N1−Ln1−N3^{i}	91.14(18)	90.8(2)	90.47(14)	90.1(2)	89.97(17)
N3−Ln1−N3^{i}	139.90(19)	139.8(2)	139.40(15)	139.0(2)	138.70(17)

Symmetry codes: (i) $-x + 1, y, -z + 3/2$.

Table S2 Selected bond lengths (Å) and bond angles (°) for **6-13**

	$\mathbf{6}_{\mathbf{Pr}}$	$\mathbf{7}_{\mathbf{Nd}}$	$\mathbf{8}_{\mathbf{Sm}}$	$\mathbf{9}_{\mathbf{Eu}}$	$\mathbf{10}_{\mathbf{Gd}}$	$\mathbf{11}_{\mathbf{Tb}}$	$\mathbf{12}_{\mathbf{Dy}}$	$\mathbf{13}_{\mathbf{Er}}$
Ln1−O1	2.5567(14)	2.5355(16)	2.511(2)	2.4965(13)	2.481(2)	2.470(2)	2.4857(18)	2.440(4)
Ln1−O2	2.3781(16)	2.3607(17)	2.326(2)	2.3124(13)	2.298(2)	2.279(2)	2.2974(19)	2.250(4)
Ln1−O3^{i}	2.4395(15)	2.4227(17)	2.396(2)	2.3825(13)	2.366(2)	2.352(2)	2.3629(18)	2.311(4)
Ln1−O4	2.3531(15)	2.3406(17)	2.312(2)	2.2992(13)	2.288(2)	2.269(2)	2.2837(18)	2.233(4)
Ln1−O5^{ii}	2.4203(16)	2.4088(17)	2.382(2)	2.3682(13)	2.354(2)	2.339(2)	2.3508(19)	2.303(4)
Ln1−O6	2.4085(15)	2.3987(16)	2.381(2)	2.3717(13)	2.362(2)	2.354(2)	2.3604(18)	2.326(4)
Ln1−O8	2.5775(17)	2.5600(19)	2.531(3)	2.5175(15)	2.501(2)	2.490(2)	2.499(2)	2.446(4)
Ln1−N1	2.6887(17)	2.670(2)	2.638(3)	2.6207(16)	2.609(3)	2.592(2)	2.606(2)	2.554(4)
O1−Ln1−O8	136.43(6)	136.11(7)	135.52(9)	135.18(5)	135.00(9)	134.59(9)	134.84(7)	133.79(15)
O1−Ln1−N1	60.09(5)	60.52(6)	61.23(8)	61.53(4)	61.80(7)	62.12(8)	61.82(6)	62.99(13)
O2−Ln1−O1	131.71(5)	132.12(6)	132.63(8)	132.92(5)	133.21(7)	133.38(8)	133.14(7)	134.10(13)
O2−Ln1−O3^{i}	86.11(6)	86.39(6)	86.54(9)	86.96(5)	87.09(8)	87.38(8)	87.17(7)	87.83(14)
O2−Ln1−O5^{ii}	96.49(6)	96.55(6)	96.70(8)	96.62(5)	96.65(8)	96.60(8)	96.66(7)	96.76(14)
O2−Ln1−O6	82.93(6)	82.73(6)	82.12(8)	82.05(5)	81.93(8)	81.75(8)	81.84(7)	81.22(14)
O2−Ln1−O8	74.82(6)	75.05(7)	75.74(9)	75.88(5)	76.06(8)	76.38(9)	76.23(7)	76.96(15)
O2−Ln1−N1	71.64(5)	71.62(6)	71.44(8)	71.43(5)	71.45(8)	71.30(8)	71.36(7)	71.14(14)
O3^{i}−Ln1−O1	77.50(5)	77.37(6)	77.28(8)	77.22(5)	77.20(8)	77.10(8)	77.15(7)	77.03(13)
O3^{i}−Ln1−O8	145.33(6)	145.62(7)	146.04(9)	146.26(5)	146.32(9)	146.66(9)	146.48(7)	147.08(15)
O3^{i}−Ln1−N1	70.65(5)	70.95(6)	71.32(8)	71.60(5)	71.70(8)	72.04(8)	71.73(7)	72.10(14)
O4−Ln1−O1	74.78(5)	74.68(6)	74.49(8)	74.32(5)	74.12(8)	73.90(8)	74.17(7)	73.39(13)
O4−Ln1−O2	152.90(6)	152.69(7)	152.56(9)	152.49(5)	152.44(8)	152.54(8)	152.47(7)	152.39(15)
O4−Ln1−O3^{i}	108.56(5)	107.95(6)	106.91(8)	106.08(5)	105.76(8)	105.02(8)	105.58(7)	103.96(14)
O4−Ln1−O5^{ii}	86.28(6)	86.69(6)	87.60(8)	88.06(5)	88.31(8)	88.68(8)	88.33(7)	89.33(13)
O4−Ln1−O6	78.72(6)	78.51(6)	78.40(8)	78.17(5)	78.11(8)	78.12(8)	78.14(7)	78.08(14)
O4−Ln1−O8	80.71(6)	80.54(7)	80.26(9)	80.30(5)	80.19(8)	80.21(9)	80.11(7)	79.95(14)
O4−Ln1−N1	134.19(6)	134.55(6)	135.16(8)	135.36(5)	135.45(8)	135.58(8)	135.53(7)	136.07(14)
O5^{ii}−Ln1−O1	71.89(5)	72.08(6)	72.26(8)	72.42(5)	72.45(7)	72.61(7)	72.50(7)	72.56(13)
O5^{ii}−Ln1−O3^{i}	140.99(5)	141.13(6)	141.23(8)	141.49(5)	141.46(8)	141.67(8)	141.58(7)	141.61(14)
O5^{ii}−Ln1−O8	71.07(6)	70.85(7)	70.65(9)	70.37(5)	70.46(8)	70.10(9)	70.25(7)	70.05(14)
O5^{ii}−Ln1−N1	73.34(6)	73.33(6)	73.22(8)	73.33(5)	73.27(8)	73.28(8)	73.42(7)	73.49(14)
O6−Ln1−O1	134.39(5)	134.03(5)	133.75(7)	133.53(4)	133.35(7)	133.30(7)	133.48(6)	133.14(12)
O6−Ln1−O3^{i}	76.69(5)	76.33(6)	75.71(8)	75.39(5)	75.26(8)	75.04(8)	75.27(6)	74.16(14)
O6−Ln1−O5^{ii}	142.31(6)	142.54(6)	143.06(8)	143.12(5)	143.28(8)	143.29(8)	143.15(7)	143.62(14)
O6−Ln1−O8	72.49(6)	72.86(6)	73.35(9)	73.64(5)	73.66(8)	73.95(8)	73.71(7)	74.16(14)
O6−Ln1−N1	139.42(6)	139.15(7)	138.43(9)	138.21(5)	138.08(9)	137.88(8)	137.98(7)	137.11(15)
O8−Ln1−N1	127.01(6)	126.95(6)	127.01(9)	127.03(5)	127.18(8)	127.06(8)	127.20(7)	127.44(14)

Symmetry codes: (i) x, $-y + 3/2$, $z - 1/2$; (ii) x, $-y + 3/2$, $z + 1/2$.

Table S3 Selected bond lengths (Å) and bond angles (°) for **14-21**

	14$_{Eu}$	**15**$_{Gd}$	**16**$_{Tb}$	**17**$_{Dy}$	**18**$_{Er}$	**19**$_{Tm}$	**20**$_{Yb}$	**21**$_{Lu}$
Ln1−O1	2.388(3)	2.370(2)	2.349(2)	2.338(2)	2.273(3)	2.298(7)	2.236(10)	2.204(14)
Ln1−O3	2.246(3)	2.23 (3)	2.225(3)	2.209(2)	2.182(3)	2.167(2)	2.152(3)	2.144(3)
Ln1−O5	2.261(3)	2.250(2)	2.239(2)	2.229(2)	2.210(3)	2.200(2)	2.199(3)	2.189(3)
Ln1−O7	2.254(3)	2.232(3)	2.223(3)	2.216(2)	2.193(3)	2.176(2)	2.171(3)	2.165(3)
Ln1−O9	2.305(3)	2.291(2)	2.279(2)	2.266(2)	2.241(3)	2.226(2)	2.213(3)	2.208(3)
Ln1−O11	2.287(3)	2.278(2)	2.262(3)	2.247(2)	2.222(3)	2.207(2)	2.191(3)	2.180(3)
Ln2−O8	2.341(3)	2.329(2)	2.312(2)	2.300(2)	2.271(2)	2.254(2)	2.239(2)	2.231(3)
Ln2−O10	2.280(3)	2.269(2)	2.250(2)	2.240(2)	2.213(3)	2.195(2)	2.184(2)	2.172(3)
Ln2−O12	2.303(2)	2.290(2)	2.277(2)	2.270(2)	2.244(2)	2.231(2)	2.223(2)	2.213(3)
Ln2−O13	2.449(3)	2.440(2)	2.426(2)	2.417(2)	2.391(2)	2.375(2)	2.366(2)	2.356(3)
Ln2−O14	2.527(2)	2.514(2)	2.498(2)	2.4866(19)	2.468(2)	2.4642(19)	2.459(2)	2.452(2)
Ln2−O15	2.378(2)	2.359(2)	2.346(2)	2.3380(19)	2.315(2)	2.306(2)	2.297(2)	2.293(3)
Ln2−O17	2.301(3)	2.295(2)	2.279(2)	2.268(2)	2.245(2)	2.231(2)	2.219(2)	2.210(3)
Ln3−O1^{ii}	2.657(3)	2.659(2)	2.684(3)	2.689(2)	2.882(4)	2.834(13)	3.029(19)	2.170(3)
Ln3−O2^{ii}	2.438(3)	2.428(2)	2.407(2)	2.392(2)	2.331(3)	2.400(9)	2.368(15)	2.239(6)
Ln3−O4^{ii}	2.302(3)	2.291(2)	2.276(2)	2.267(2)	2.241(3)	2.229(2)	2.222(2)	2.215(3)
Ln3−O6^{ii}	2.290(3)	2.284(2)	2.263(2)	2.249(2)	2.214(3)	2.190(2)	2.172(2)	2.158(3)
Ln3−O14	2.403(2)	2.392(2)	2.377(2)	2.3655(19)	2.331(2)	2.310(2)	2.297(2)	2.284(3)
Ln3−O15	2.569(2)	2.555(2)	2.544(2)	2.5312(19)	2.512(2)	2.496(2)	2.492(2)	2.482(2)
Ln3−O16	2.443(3)	2.432(2)	2.412(3)	2.401(2)	2.372(3)	2.361(2)	2.349(3)	2.341(3)
Ln3−O18	2.308(3)	2.297(2)	2.283(2)	2.2713(19)	2.240(2)	2.225(2)	2.212(2)	2.204(2)
O3−Ln1−O1	83.81(11)	84.13(10)	83.62(10)	83.77(9)	84.95(14)	94.9(3)	94.1(2)	92.2(6)
O3−Ln1−O5	97.21(13)	97.57(12)	97.06(12)	97.30(10)	96.36(12)	94.37(10)	94.1(2)	93.32(13)
O3−Ln1−O9	85.32(13)	85.32(11)	85.68(11)	85.87(10)	85.97(12)	86.12(10)	86.52(11)	86.45(13)
O3−Ln1−O11	170.73(12)	170.24(10)	170.37(11)	170.48(9)	171.51(12)	172.89(10)	173.94(11)	174.51(13)
O5−Ln1−O1	77.25(10)	77.40(9)	77.69(9)	77.99(8)	78.93(11)	78.1(2)	79.3(4)	96.1(6)
O5−Ln1−O9	166.58(12)	167.18(10)	168.35(10)	168.78(9)	169.57(11)	171.01(10)	171.83(11)	172.58(13)
O5−Ln1−O11	89.80(12)	89.91(11)	90.27(11)	89.96(10)	90.42(12)	91.47(11)	91.58(12)	91.29(14)
O7−Ln1−O1	161.32(11)	161.41(9)	161.47(10)	161.54(8)	163.80(12)	160.5(3)	162.8(5)	169.51(15)
O7−Ln1−O5	87.37(11)	87.38(10)	87.26(10)	87.12(8)	87.27(10)	87.30(9)	87.33(10)	87.18(12)
O7−Ln1−O9	105.94(11)	105.27(10)	104.20(10)	103.81(8)	102.99(10)	101.68(9)	100.83(10)	100.24(12)
O7−Ln1−O11	86.53(12)	86.67(10)	86.50(11)	86.80(9)	87.00(12)	86.47(10)	86.52(12)	86.70(14)
O9−Ln1−O1	89.97(11)	90.53(9)	91.42(9)	91.72(8)	91.18(11)	93.3(2)	92.7(4)	90.24(15)
O11−Ln1−O1	103.73(10)	103.71(9)	104.13(10)	103.85(8)	101.43(13)	106.6(3)	104.5(5)	93.29(17)
O11−Ln1−O9	89.26(12)	88.76(11)	88.38(11)	88.17(9)	88.28(12)	88.73(11)	89.03(12)	89.36(14)
O8−Ln2−O13	156.08(10)	156.02(8)	155.91(9)	155.68(7)	155.75(9)	156.01(8)	155.91(9)	155.89(10)

O8−Ln2−O14	146.02(9)	146.49(8)	146.82(8)	146.94(7)	147.02(9)	147.21(7)	147.63(8)	147.87(9)
O8−Ln2−O15	82.34(9)	82.55(8)	82.54(8)	82.59(7)	82.44(9)	82.49(7)	82.62(9)	82.59(10)
O10−Ln2−O8	105.19(11)	104.62(9)	104.08(9)	103.86(8)	103.28(10)	103.02(9)	102.61(10)	102.29(11)
O10−Ln2−O12	87.51(11)	87.60(9)	87.56(10)	87.86(8)	88.87(10)	89.55(9)	90.34(10)	91.23(11)
O10−Ln2−O13	88.00(10)	88.16(9)	88.14(9)	88.37(8)	88.36(10)	87.93(9)	87.95(10)	88.03(11)
O10−Ln2−O14	88.05(10)	87.92(9)	87.66(9)	87.56(7)	87.08(9)	86.37(8)	85.78(9)	85.51(10)
O10−Ln2−O15	87.23(10)	86.99(9)	86.85(9)	86.49(8)	86.16(9)	86.20(8)	86.13(9)	86.14(10)
O10−Ln2−O17	168.98(11)	169.14(9)	168.72(9)	168.67(8)	168.50(10)	168.10(8)	168.09(9)	168.06(10)
O12−Ln2−O8	83.71(10)	83.25(9)	82.85(9)	82.60(8)	82.23(10)	81.87(8)	81.47(9)	81.23(11)
O12−Ln2−O13	76.91(9)	77.01(8)	76.93(9)	76.86(7)	76.74(9)	76.87(8)	76.83(9)	76.77(10)
O12−Ln2−O14	128.77(9)	128.92(8)	129.21(8)	129.37(7)	129.79(9)	130.11(8)	130.21(9)	130.21(10)
O12−Ln2−O15	163.24(9)	163.00(8)	162.58(8)	162.39(7)	162.34(9)	162.43(8)	162.52(9)	162.66(10)
O13−Ln2−O14	51.94(8)	52.00(7)	52.39(8)	52.63(6)	53.14(8)	53.32(7)	53.46(8)	53.50(9)
O15−Ln2−O13	118.77(8)	118.89(7)	119.33(8)	119.58(7)	119.98(9)	119.93(7)	120.06(8)	120.20(9)
O15−Ln2−O14	66.91(8)	66.96(7)	67.00(8)	67.02(6)	66.89(8)	66.65(7)	66.63(8)	66.71(9)
O17−Ln2−O8	82.48(11)	82.88(9)	83.35(9)	83.44(8)	84.07(10)	84.54(8)	85.00(9)	85.34(11)
O17−Ln2−O12	101.37(10)	101.24(9)	101.94(9)	101.82(8)	101.00(10)	100.72(9)	99.94(10)	99.08(11)
O17−Ln2−O13	87.69(10)	87.71(9)	88.12(9)	88.12(8)	88.19(10)	88.57(9)	88.55(10)	88.49(11)
O17−Ln2−O14	81.31(9)	81.62(8)	81.64(8)	81.75(7)	82.08(9)	66.65(7)	82.99(8)	83.23(9)
O17−Ln2−O15	85.96(9)	86.22(8)	85.71(9)	85.88(7)	86.07(9)	85.70(8)	85.78(9)	85.76(10)
O2^{ii}−Ln3−O1^{ii}	50.50(9)	50.66(8)	50.48(8)	50.38(7)	47.63(11)	37.5(3)	35.6(3)	53.50(9)
O2^{ii}−Ln3−O15	123.13(9)	123.41(8)	123.73(8)	124.04(7)	125.69(9)	127.4(2)	128.5(2)	128.5(2)
O2^{ii}−Ln3−O16	74.93(9)	75.02(8)	75.14(9)	75.15(8)	75.05(10)	74.3(2)	75.1(2)	75.1(2)
O4^{ii}−Ln3−O1^{ii}	81.14(10)	80.99(8)	80.84(9)	80.73(7)	80.61(9)	78.40(17)	78.6(3)	96.7(8)
O4^{ii}−Ln3−O2^{ii}	85.78(10)	85.67(9)	85.43(9)	85.13(8)	84.62(10)	82.6(3)	82.1(4)	84.5(2)
O4^{ii}−Ln3−O14	80.89(9)	80.78(8)	80.70(8)	80.68(7)	80.78(9)	80.67(8)	80.74(9)	80.48(10)
O4^{ii}−Ln3−O15	145.83(9)	145.69(8)	145.53(8)	145.62(7)	145.76(9)	145.77(7)	145.71(9)	145.59(10)
O4^{ii}−Ln3−O16	160.71(10)	160.68(9)	160.57(9)	160.27(8)	159.64(9)	158.92(8)	158.33(9)	157.94(11)
O4^{ii}−Ln3−O18	87.64(12)	87.76(10)	87.69(10)	87.76(9)	87.56(11)	87.05(9)	84.6(2)	86.13(11)
O6^{ii}−Ln3−O1^{ii}	73.85(9)	73.67(8)	73.12(8)	72.74(7)	69.77(10)	67.19(15)	64.9(2)	96.2(8)
O6^{ii}−Ln3−O2^{ii}	122.11(10)	122.06(9)	121.37(9)	120.97(8)	115.48(12)	113.5(3)	109.0(4)	99.0(2)
O6^{ii}−Ln3−O4^{ii}	101.93(12)	101.97(10)	101.93(10)	101.86(9)	101.16(11)	100.26(9)	99.93(10)	100.01(11)
O6^{ii}−Ln3−O14	77.20(10)	77.36(9)	77.71(9)	77.97(8)	79.46(10)	81.16(9)	82.40(10)	83.29(11)
O6^{ii}−Ln3−O15	79.00(10)	78.91(8)	79.44(9)	79.59(7)	81.08(9)	83.04(8)	84.55(9)	85.45(10)
O6^{ii}−Ln3−O16	88.47(11)	88.13(10)	88.19(10)	88.11(9)	88.71(11)	90.27(10)	91.22(11)	91.61(12)
O6^{ii}−Ln3−O18	155.00(10)	155.07(9)	155.81(9)	156.12(8)	159.00(10)	162.58(8)	165.04(10)	166.57(11)
O14−Ln3−O1^{ii}	141.70(8)	141.48(7)	141.15(8)	140.98(7)	139.88(8)	137.76(14)	137.1(2)	165.0(2)
O14−Ln3−O2^{ii}	158.68(9)	158.50(8)	158.61(9)	158.54(8)	161.00(11)	159.5(3)	160.9(4)	159.0(11)
O14−Ln3−O15	65.87(8)	65.81(7)	65.79(7)	65.89(6)	65.92(8)	66.07(7)	66.07(8)	66.34(9)
O14−Ln3−O16	117.57(8)	117.81(8)	117.97(8)	118.35(7)	118.82(9)	119.25(7)	119.41(9)	119.76(9)

O15−Ln3−O1^{ii}	130.27(8)	130.44(7)	130.80(8)	130.75(6)	130.39(8)	132.23(17)	132.1(3)	134.6(11)
O16−Ln3−O1^{ii}	86.33(8)	86.24(8)	86.42(8)	86.16(7)	86.30(9)	89.15(16)	89.6(2)	81.2(11)
O16−Ln3−O15	51.75(8)	52.05(7)	52.24(8)	52.53(7)	52.94(8)	53.20(7)	53.34(8)	53.42(9)
O18−Ln3−O1^{ii}	130.87(9)	130.97(8)	130.77(8)	130.82(7)	130.93(9)	130.10(15)	129.9(2)	96.7(8)
O18−Ln3−O2^{ii}	81.21(10)	81.14(9)	81.13(9)	81.24(8)	84.11(11)	82.9(2)	85.1(4)	93.4(2)
O18−Ln3−O14	81.68(10)	81.72(8)	82.11(9)	82.19(7)	83.20(9)	84.50(8)	85.38(9)	85.99(10)
O18−Ln3−O15	80.25(9)	80.25(8)	80.21(8)	80.27(7)	81.08(9)	82.13(7)	82.62(9)	82.84(9)
O18−Ln3−O16	89.56(10)	89.76(9)	89.62(10)	89.76(8)	89.34(11)	88.14(9)	87.29(10)	86.69(12)

Symmetry codes: (i) $-x + 3/2, y + 1/2, -z + 3/2$; (ii) $-x + 3/2, y - 1/2, -z + 3/2$.

Table S4 Emission bands, luminescent lifetimes (τ), and CIE color coordinates of the Eu- and Tb-based complexes

	λex (nm)	λem (nm)	τ (μs)	CIE Coordinate
$\mathbf{2_{Eu}}$	290	592, 614, 649, 693	0.18	(0.55, 0.34)
$\mathbf{9_{Eu}}$	290	591, 617, 654, 699	0.40	(0.64, 0.34)
$\mathbf{14_{Eu}}$	290	591, 615, 653, 698	2.27	(0.65, 0.35)
$\mathbf{4_{Tb}}$	300	489, 544, 584, 620	0.57	(0.31, 0.55)
$\mathbf{11_{Tb}}$	300	488, 544, 589, 621	1.01	(0.31, 0.59)
$\mathbf{15_{Tb}}$	300	487, 542, 586, 620	1.43	(0.29, 0.60)

CIE1931 color space chromaticity diagram

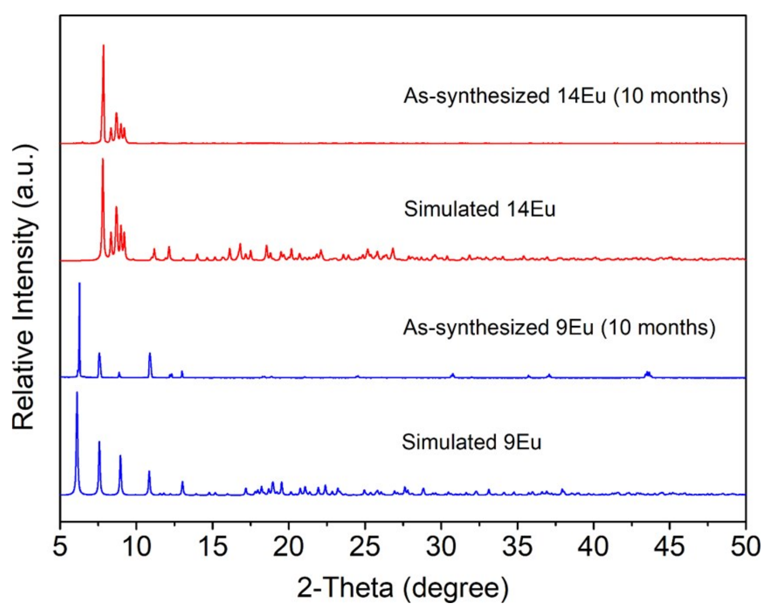
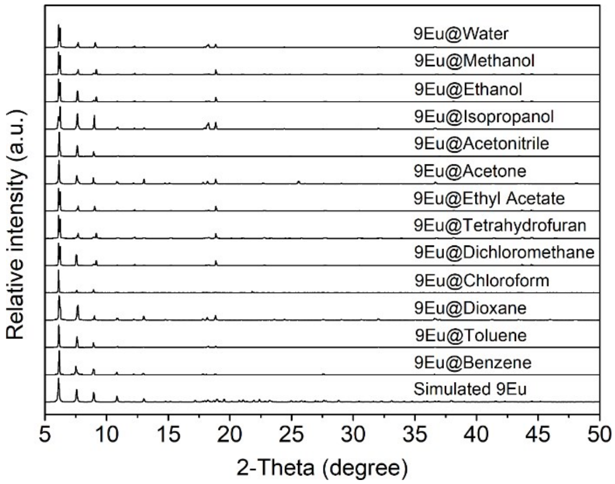
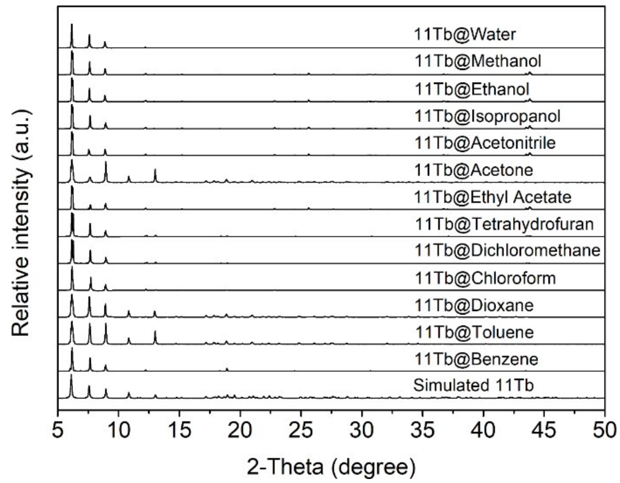


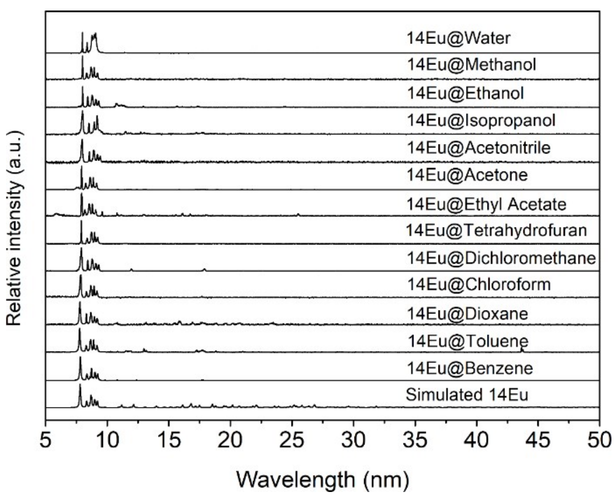
Figure S1. Comparison of the experimental PXRD patterns of the representative sample **9** (series **2Ln**) and **14** (series **3Ln**) after exposure to air for ten months with the simulated diffraction patterns based on the single crystal X-ray structure determinations.



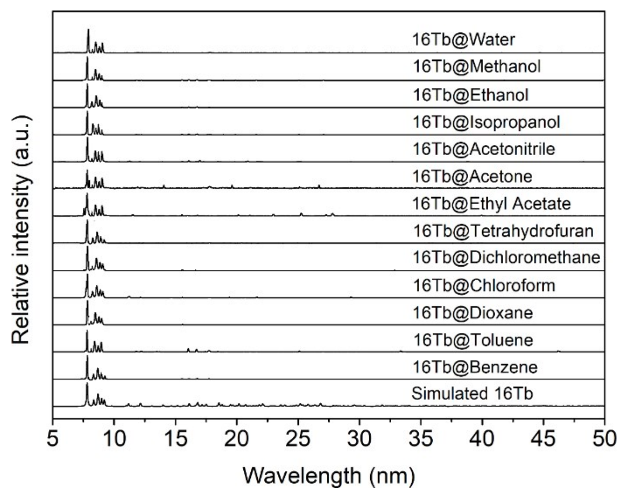
(a)



(b)

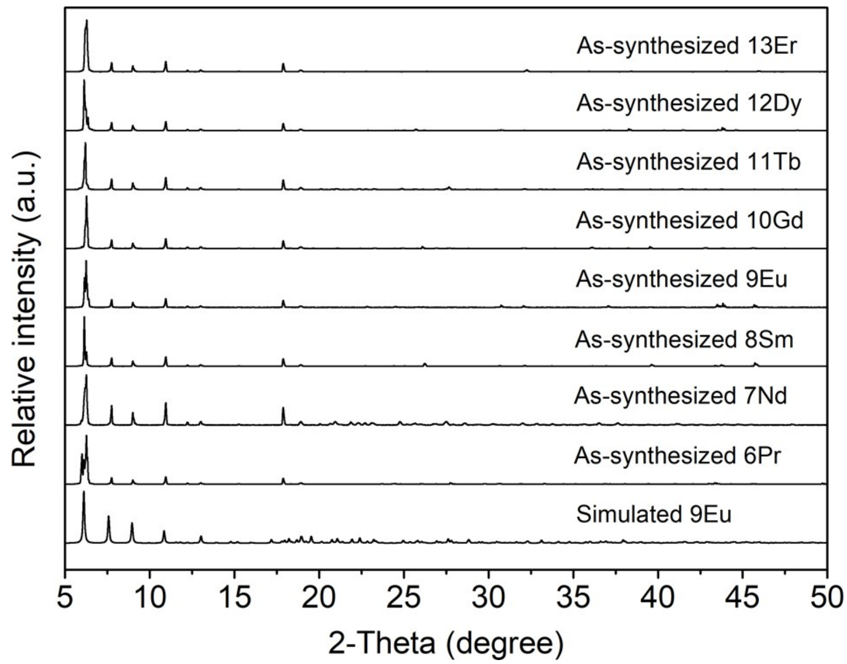


(a)

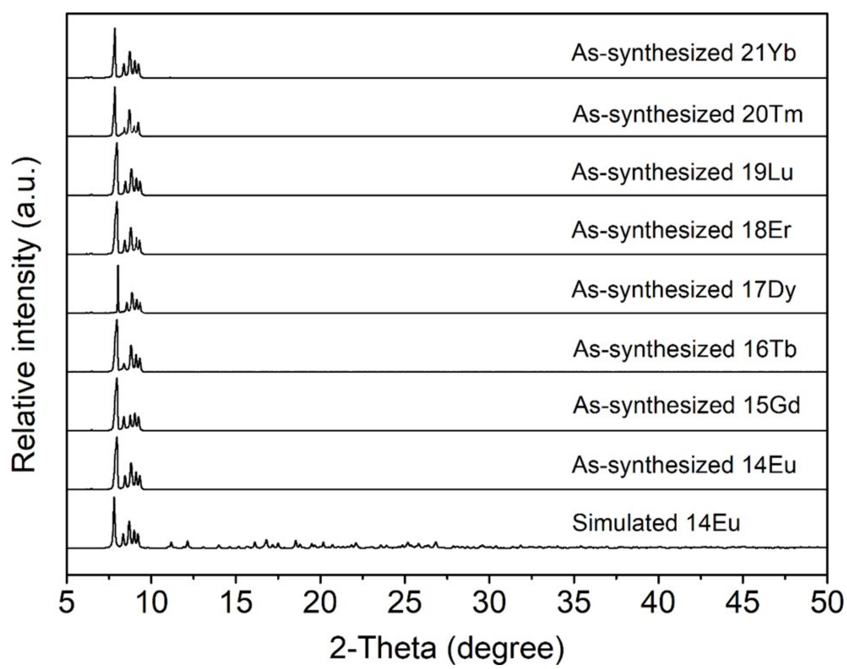


(b)

Figure S2. The PXRD patterns of the representative example complexes **9** and **11** (a, b) and **14** and **16** (c, d) after immersing in various organic solvents for 24 h.

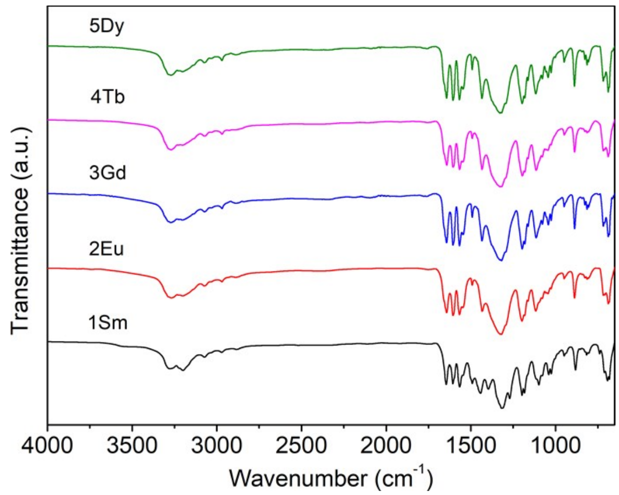


(a)

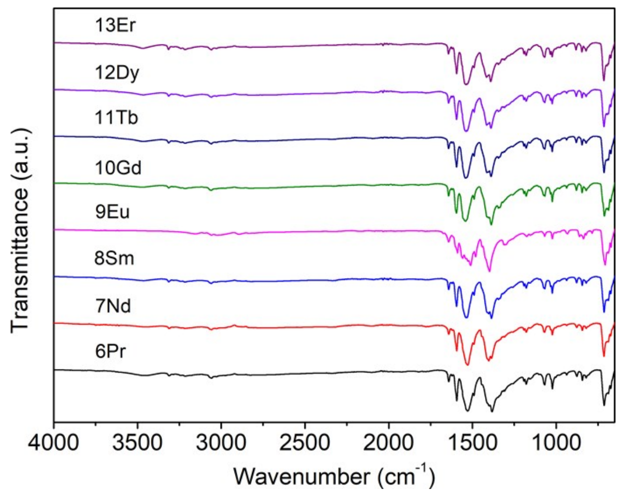


(b)

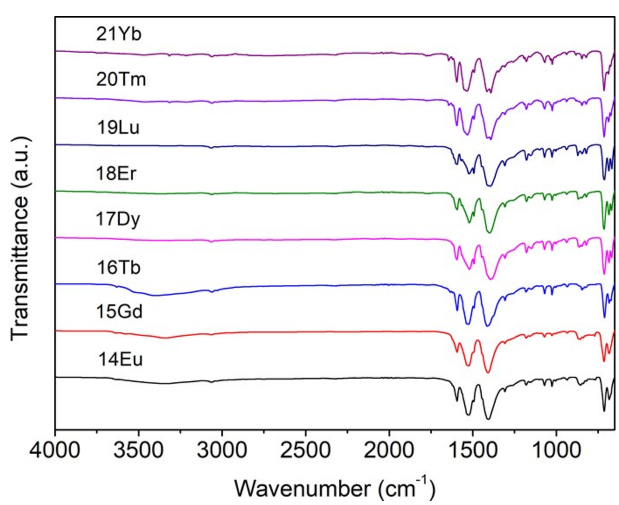
Figure S3. Comparison of the simulated and experimental PXRD patterns of the as-synthesized complexes **6-13** (a) and **14-21** (b).



(a)

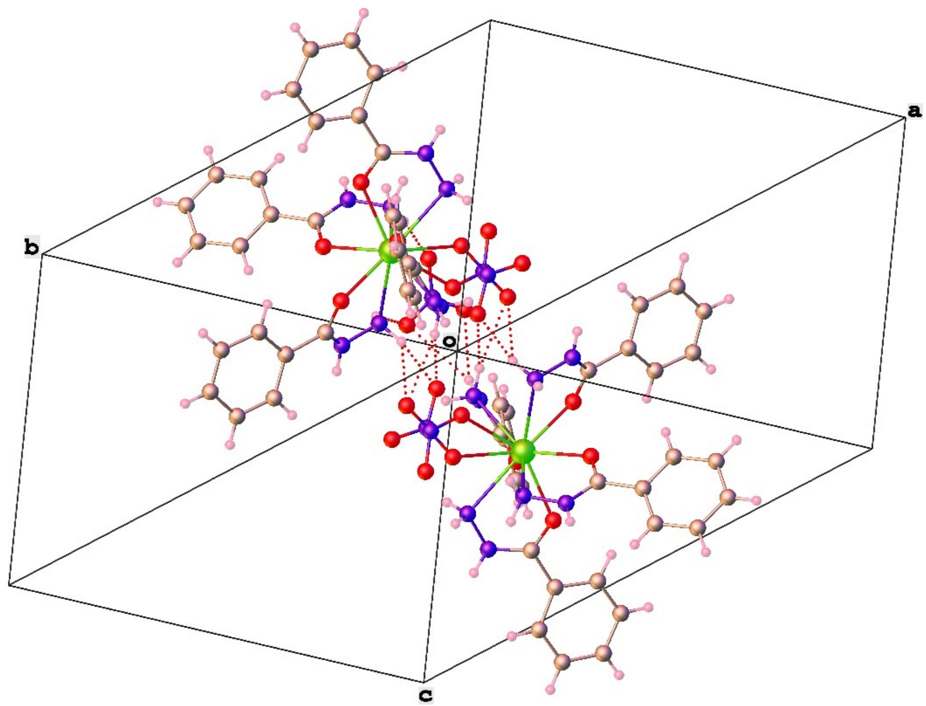


(b)



(c)

Figure S4. IR spectra of complexes **1-5** (a), **6-13** (b), and **14-21** (c).



(a)

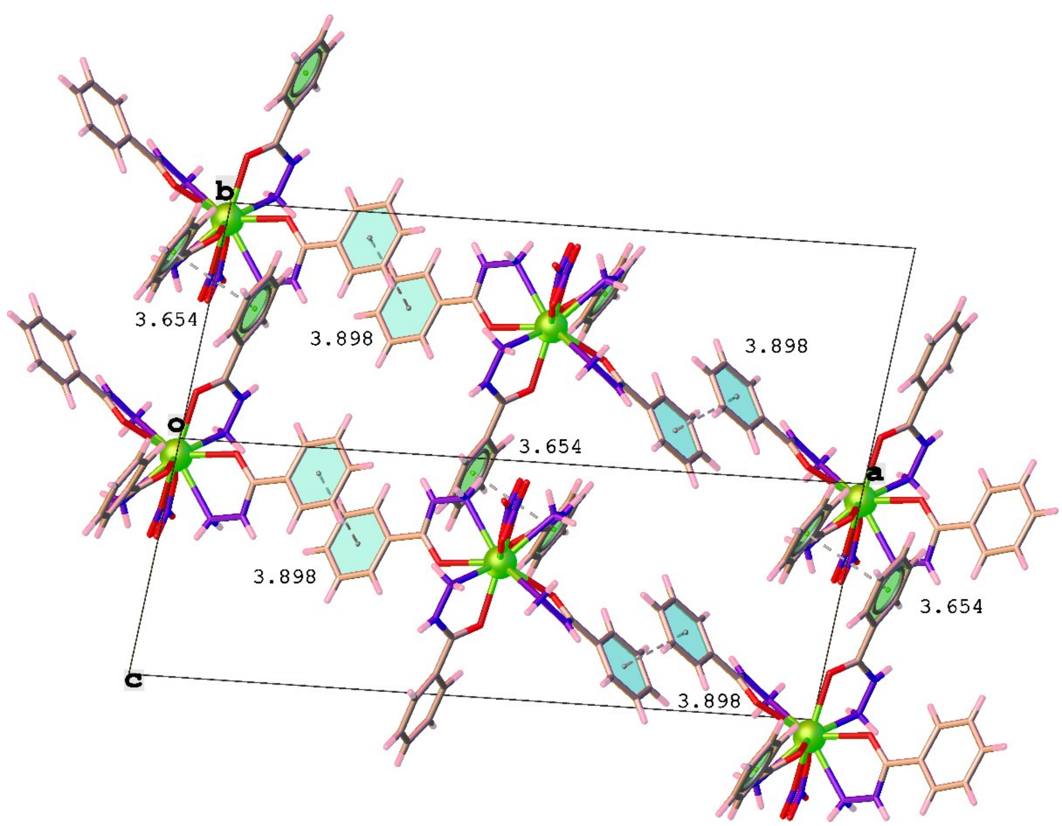
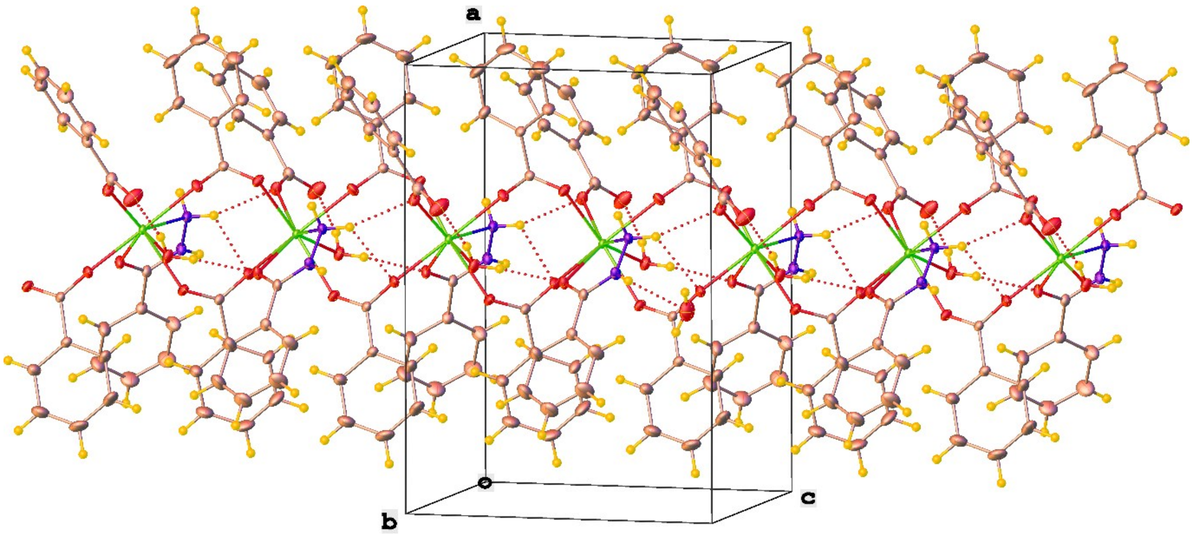


Figure S5. Partial packing diagram of **2**, showing N−H···O hydrogen bonds (a) and π–π interactions (b) (dashed lines).



(a)

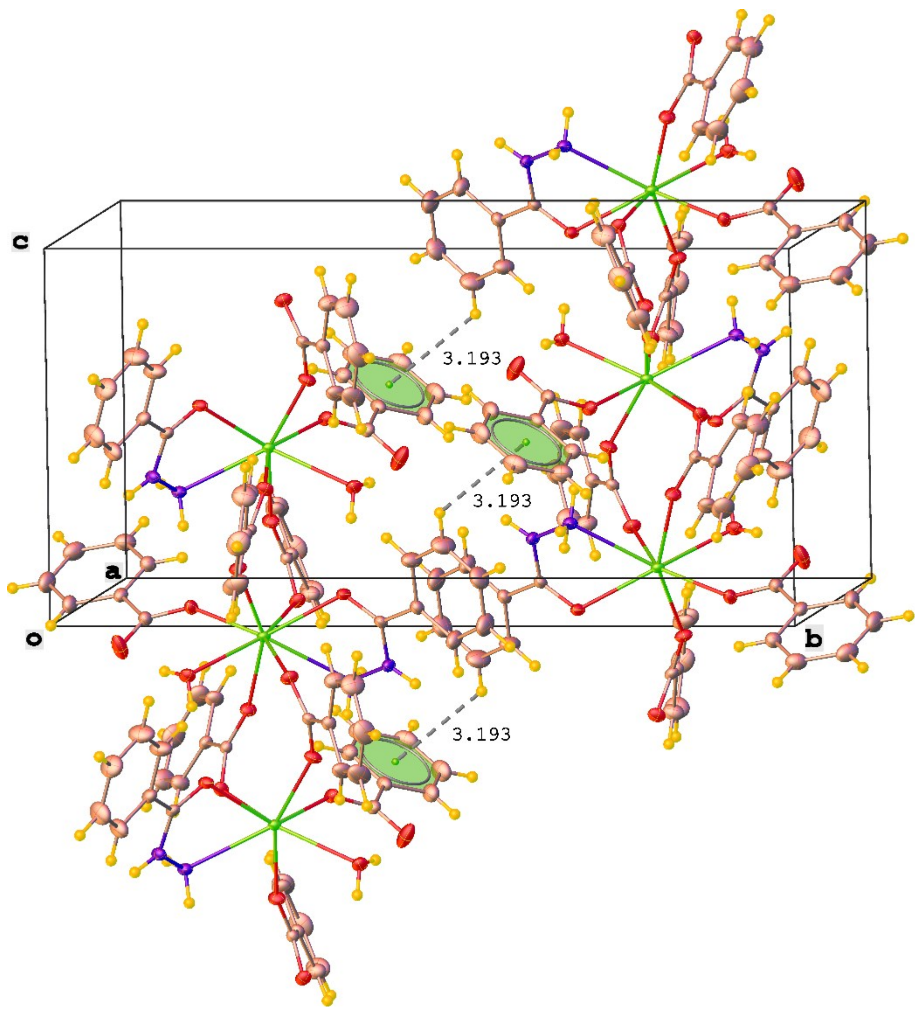
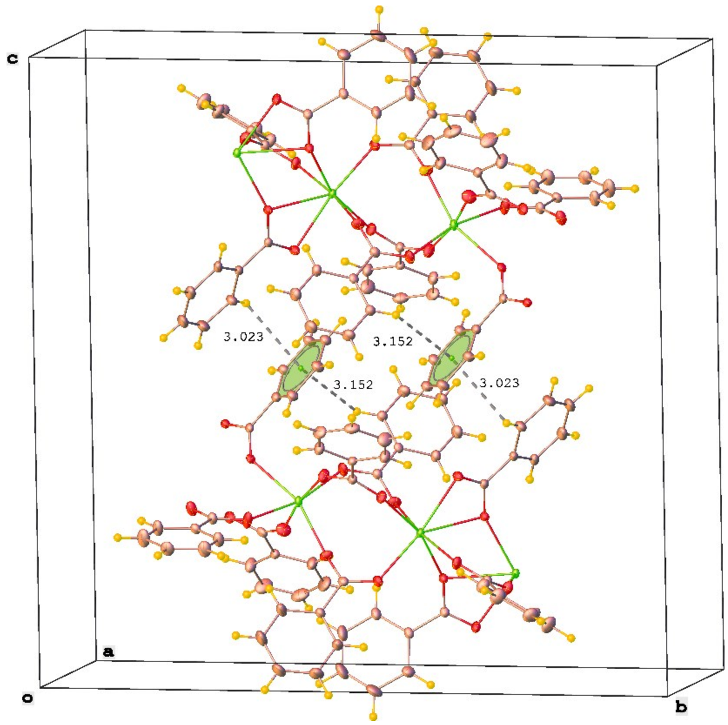


Figure S6. Partial packing diagram of **9**, showing N−H···O hydrogen bonds (a) and C−H···π interactions (b) (dashed lines).



(a)

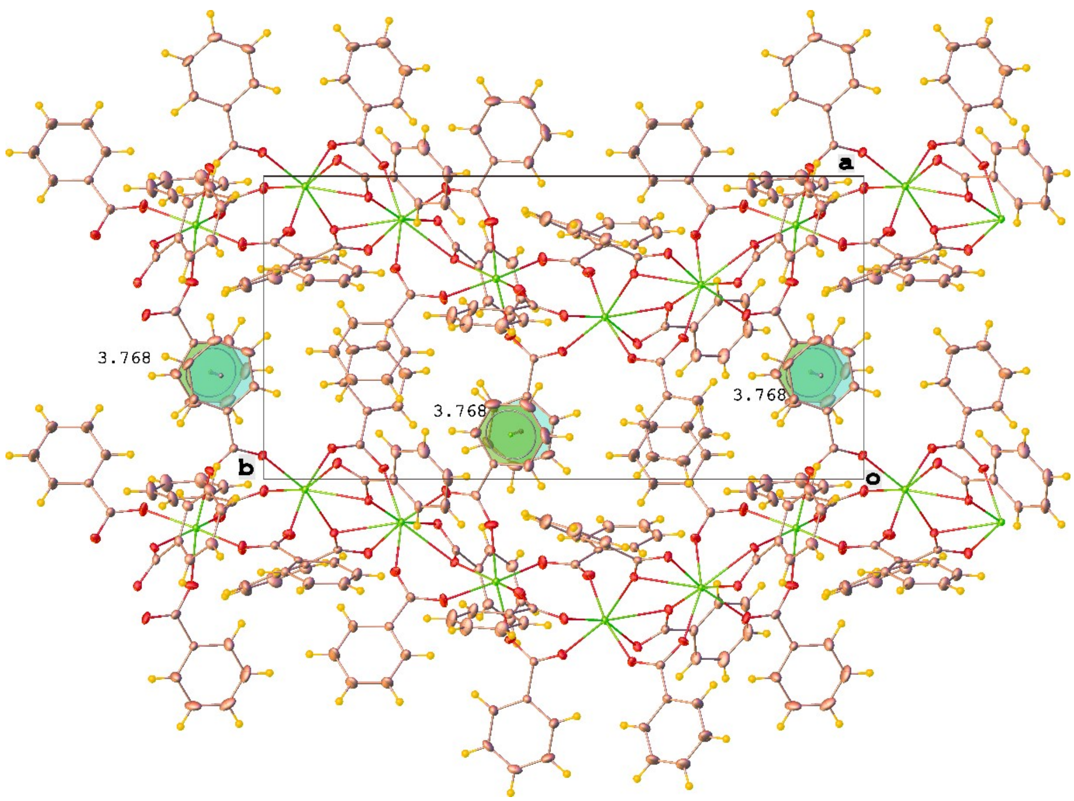


Figure S7. Partial packing diagram of **14**, showing C−H···π (a) and π–π interactions (b) (dashed lines).

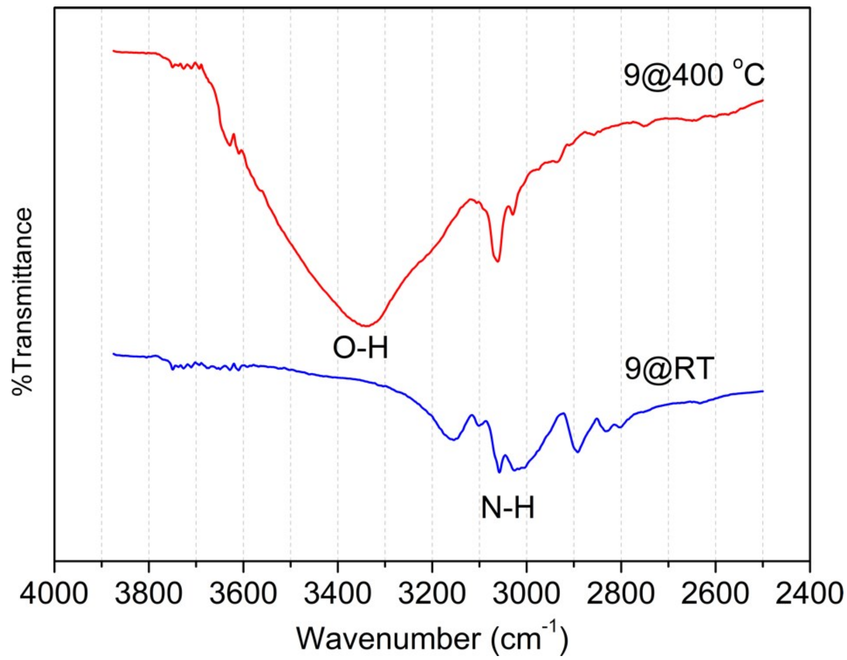
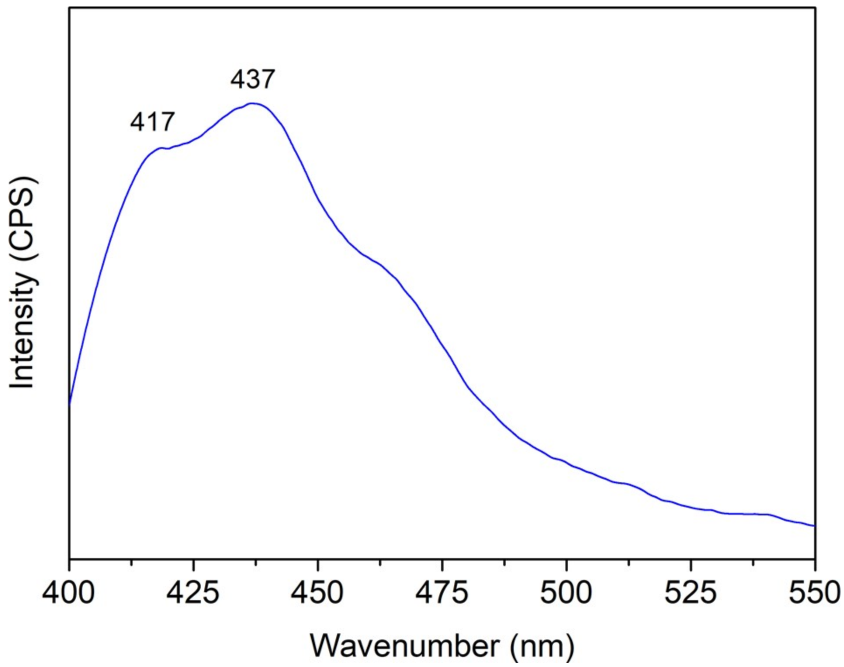
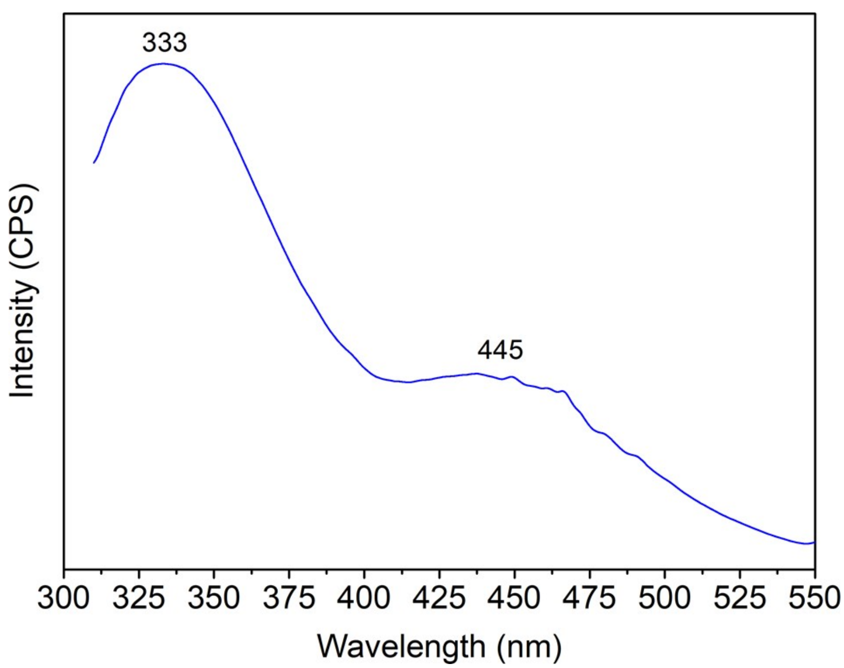


Figure S8. IR spectra of **9** at room temperature and at 400 °C.



(a)



(b)

Figure S9. The solid state PL emission spectra of the free ligands Bzz (a) and Ben (b) when excited at 290 nm.

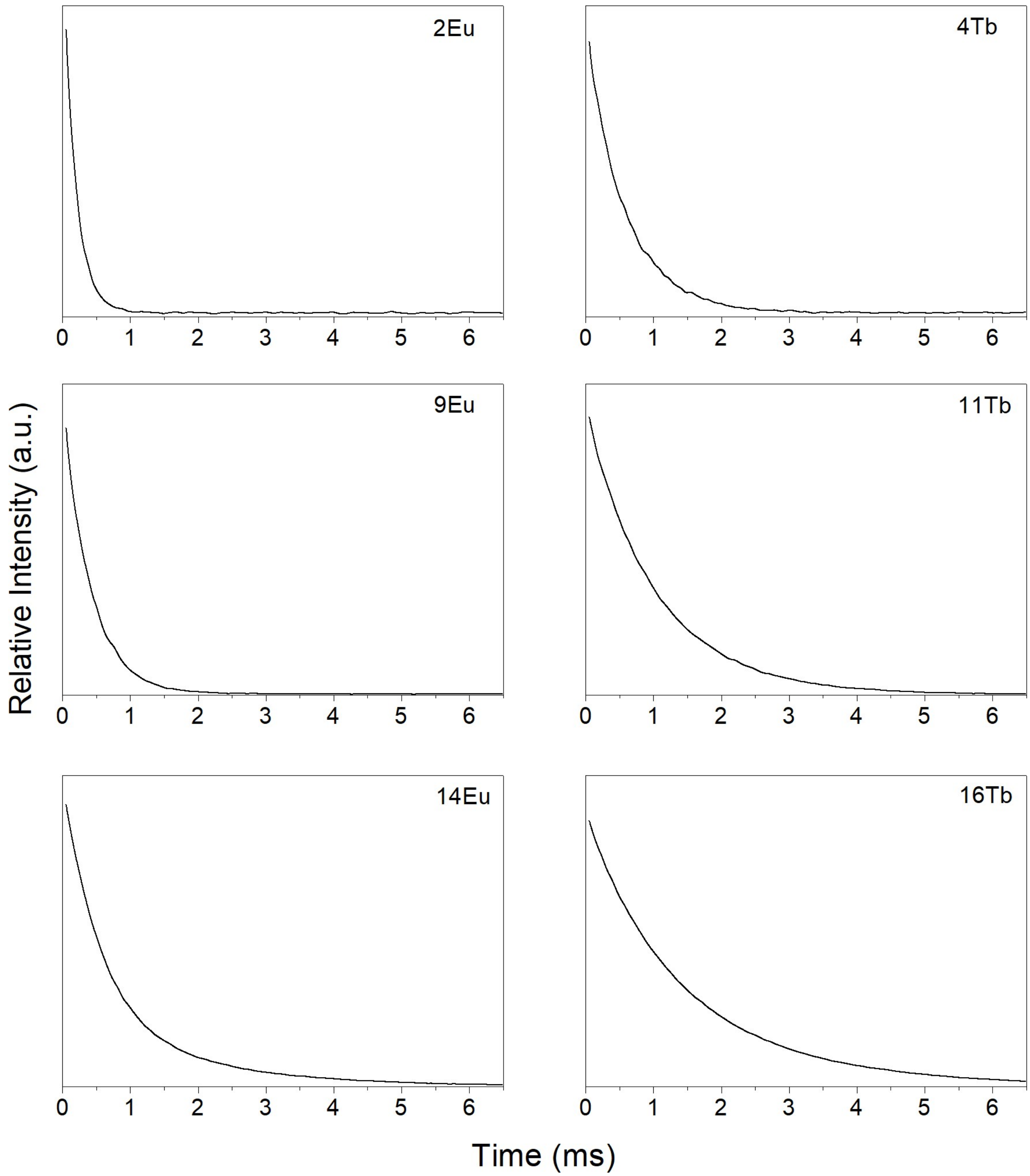


Figure S10. Luminescence decay (τ) curves for **2**, **4**, **9**, **11**, **14**, and **16**.

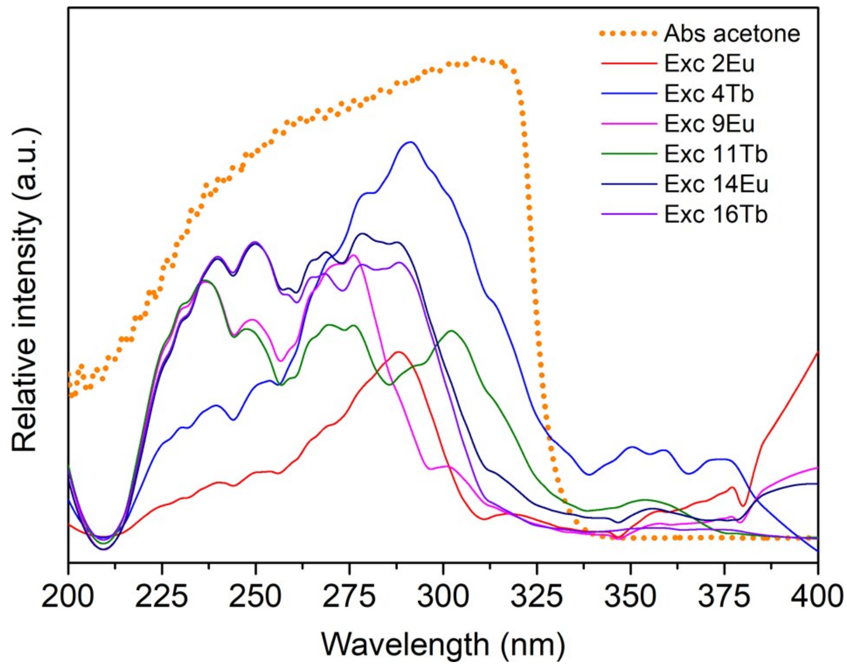


Figure S11. UV-Vis spectra of acetone and excitation spectra of **2**, **4**, **9**, **11**, **14** and **16**.

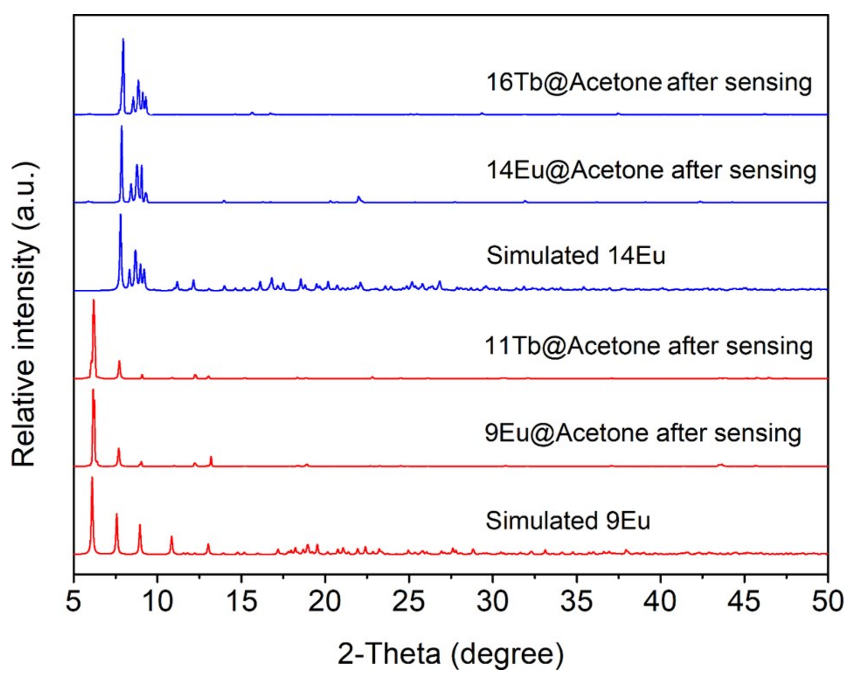


Figure S12. PXRD patterns of **9**, **11**, **14**, and **16**: simulated and after sensing of acetone.

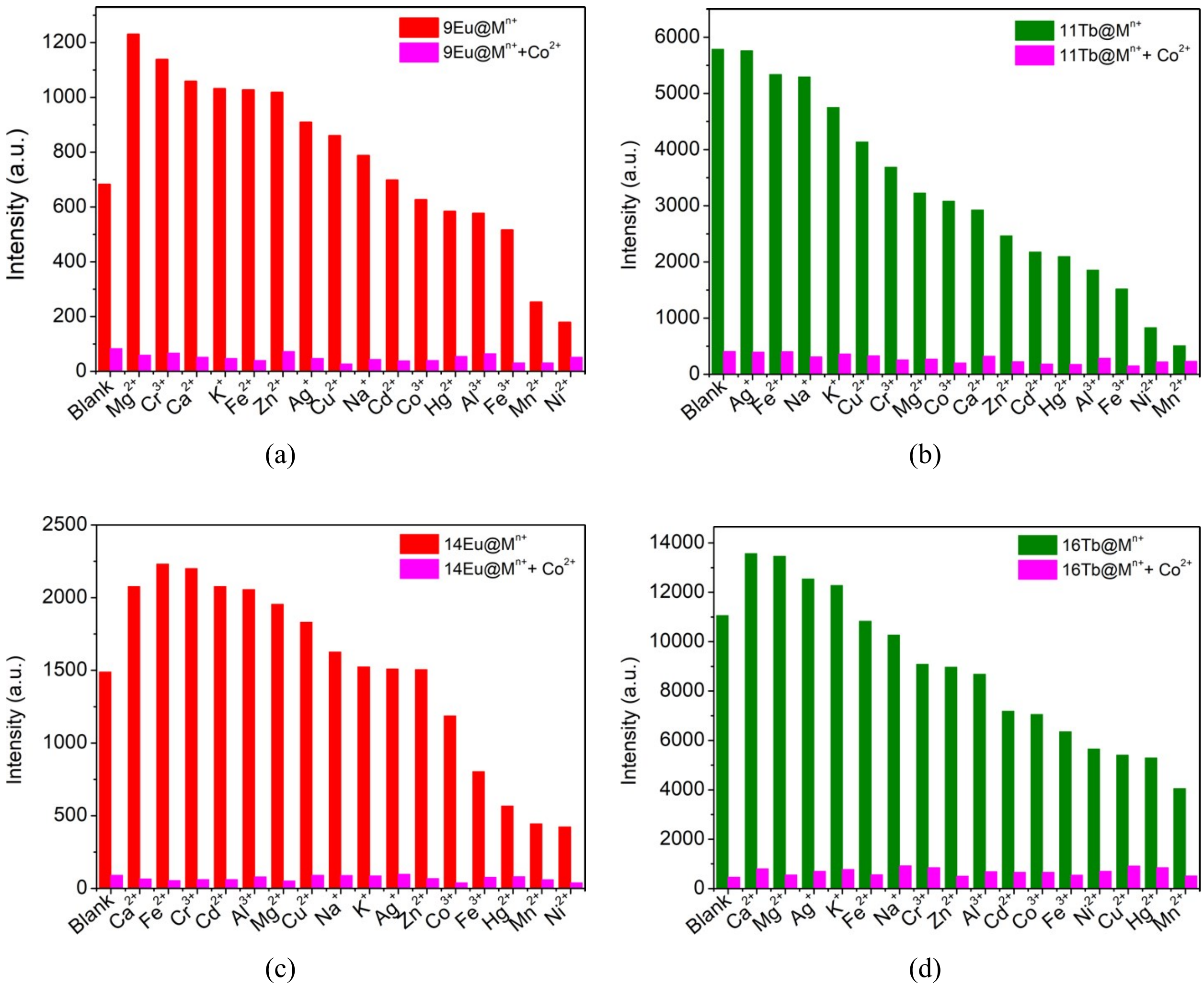
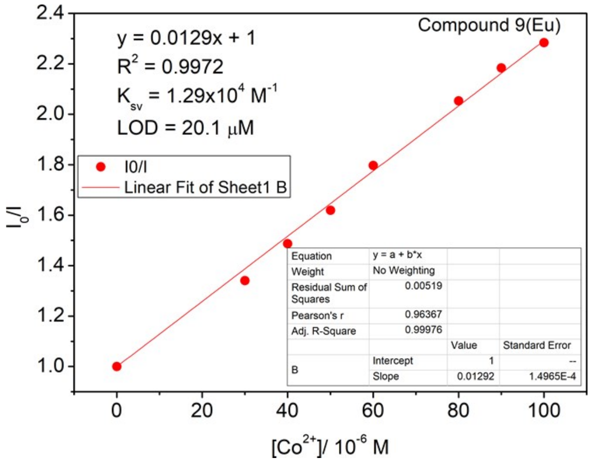
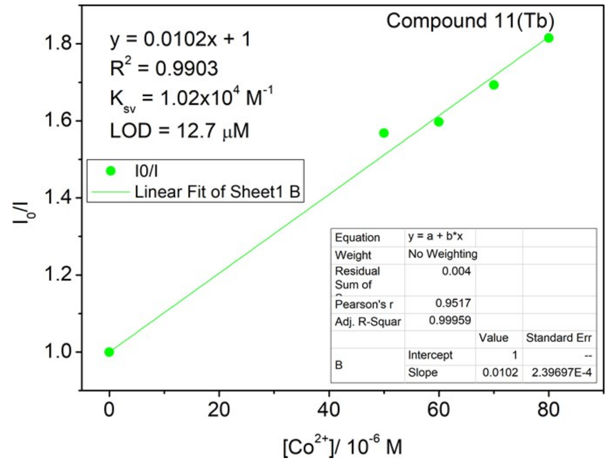


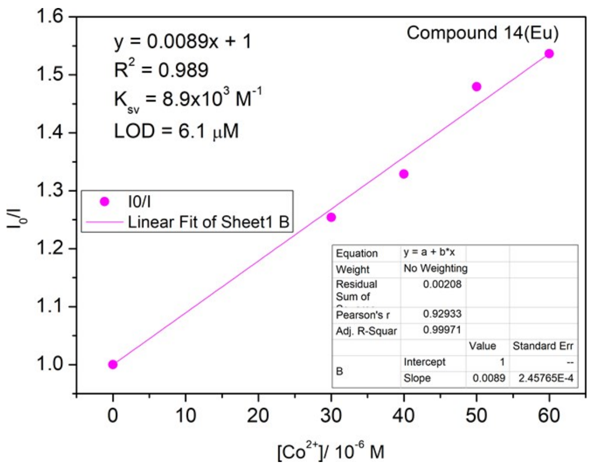
Figure S13. Relative luminescence intensities of **9** (a), **11** (b), **14** (c), and **16** (d) dispersed in the solutions of individual metal ions and the quenched luminescence intensities after the addition of solutions of Co^{2+} in acetonitrile.



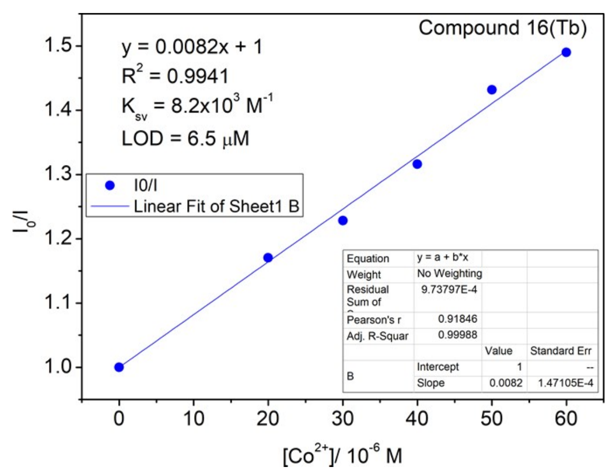
(a)



(b)



(c)



(d)

Figure S14. Plots of I_0/I versus the concentration of Co^{2+} ions plotted according to the Stern–Volmer equation as well as the linear fitting results for **9** (a), **11** (b), **14** (c), and **16** (d).

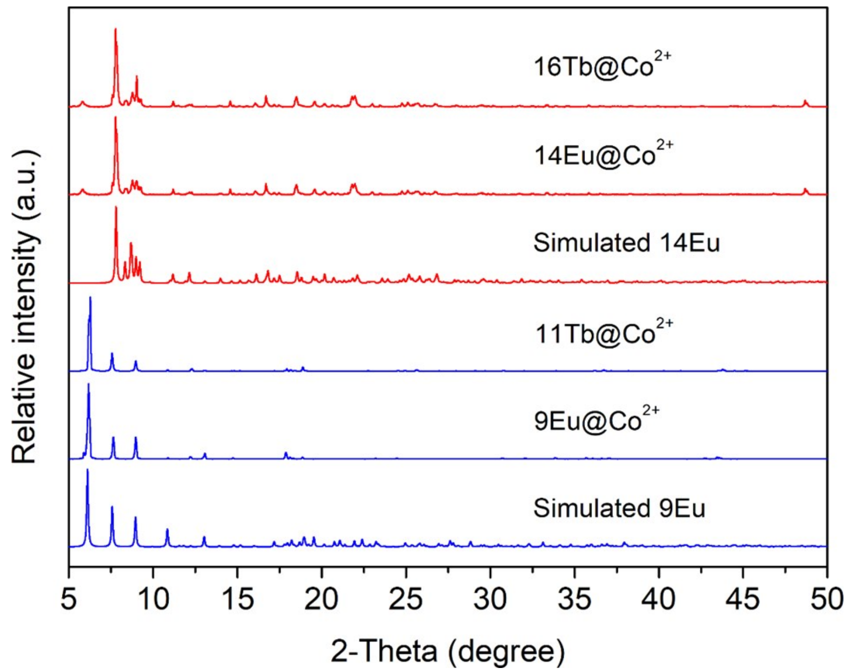
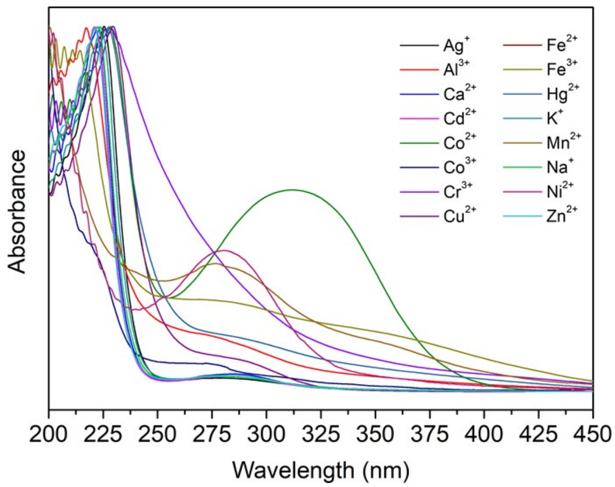
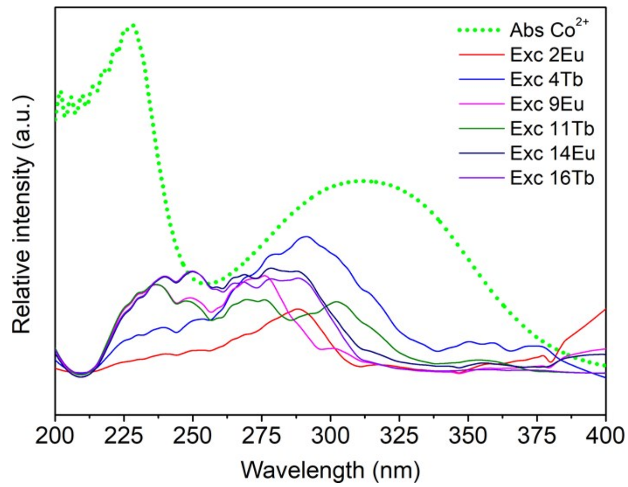


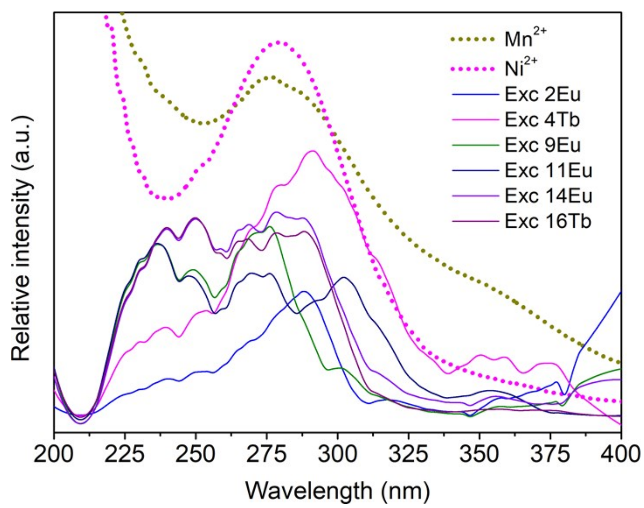
Figure S15. PXRD patterns of **9**, **11**, **14**, and **16**: simulated and after sensing of Co^{2+} ions.



(a)

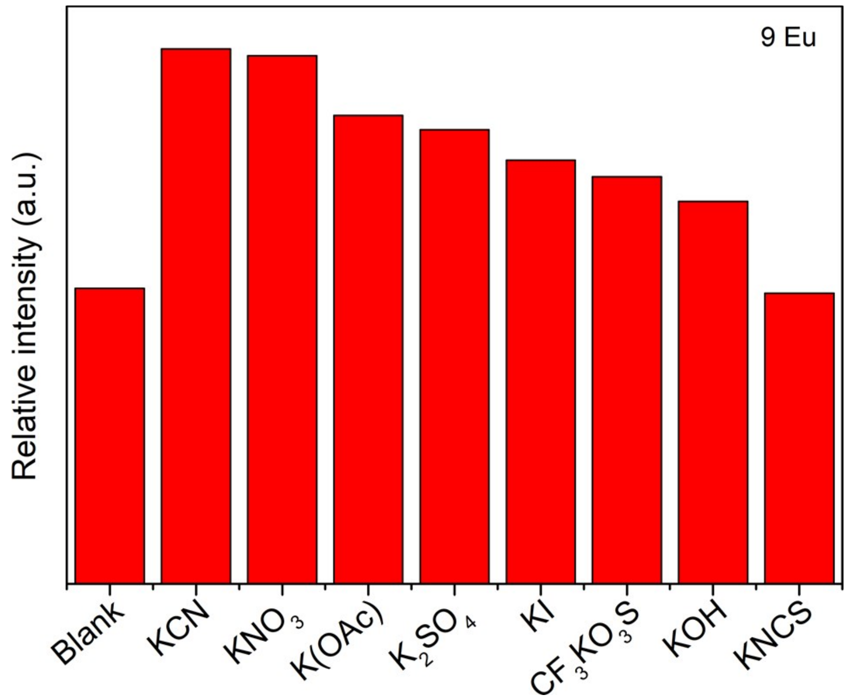


(b)

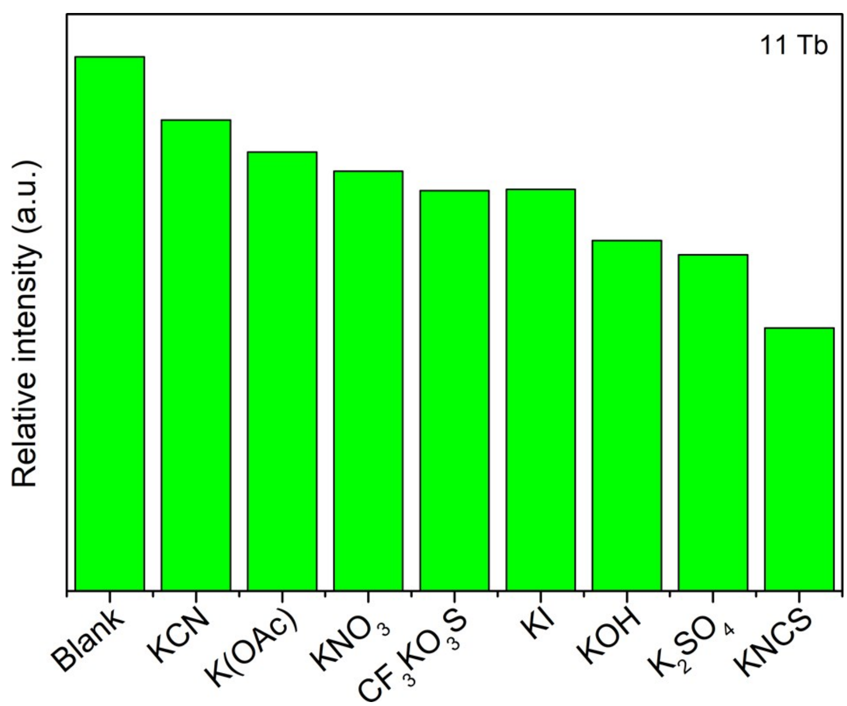


(c)

Figure S16. UV-Vis absorption spectra for metal ions in acetonitrile solutions (a). UV-Vis spectra of Co^{2+} and excitation spectra of **2**, **4**, **9**, **11**, **14**, and **16** (b). UV-Vis spectra of Mn^{2+} and Ni^{2+} and excitation spectra of **2**, **4**, **9**, **11**, **14**, and **16** (c).



(a)



(b)

Figure S17. Emission intensities of the representative complexes **9** (a) and **11** (b) in acetonitrile solutions containing potassium salts at the concentration of 5.0×10^{-4} M.

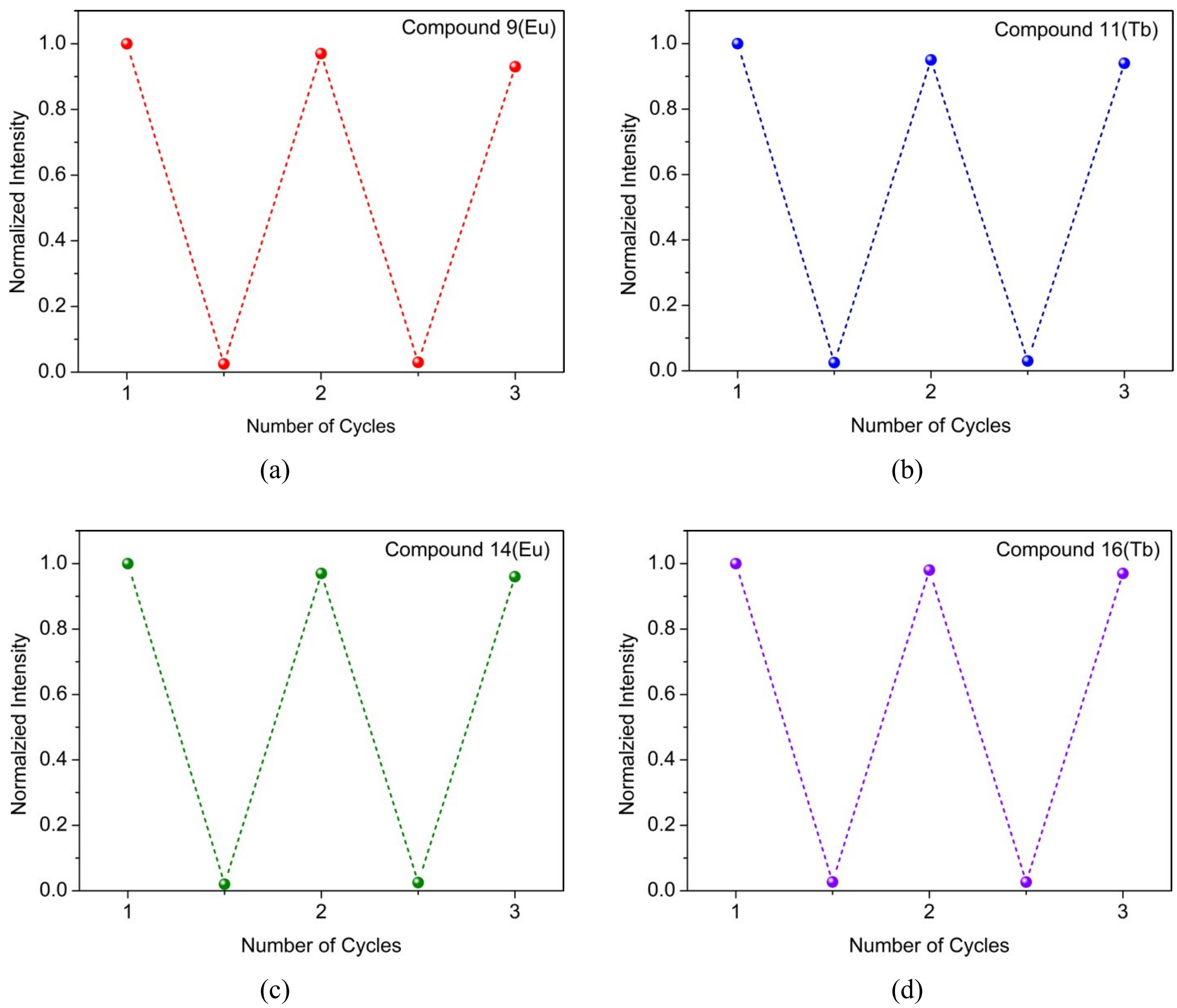


Figure S18. The quenching and recyclability test of **9** (a), **11** (b), **14** (c), and **16** (d), the upper dots represent the initial luminescence intensity and the lower dots represent the intensity upon addition of 5×10^{-4} M of acetonitrile solution of Co^{2+}.

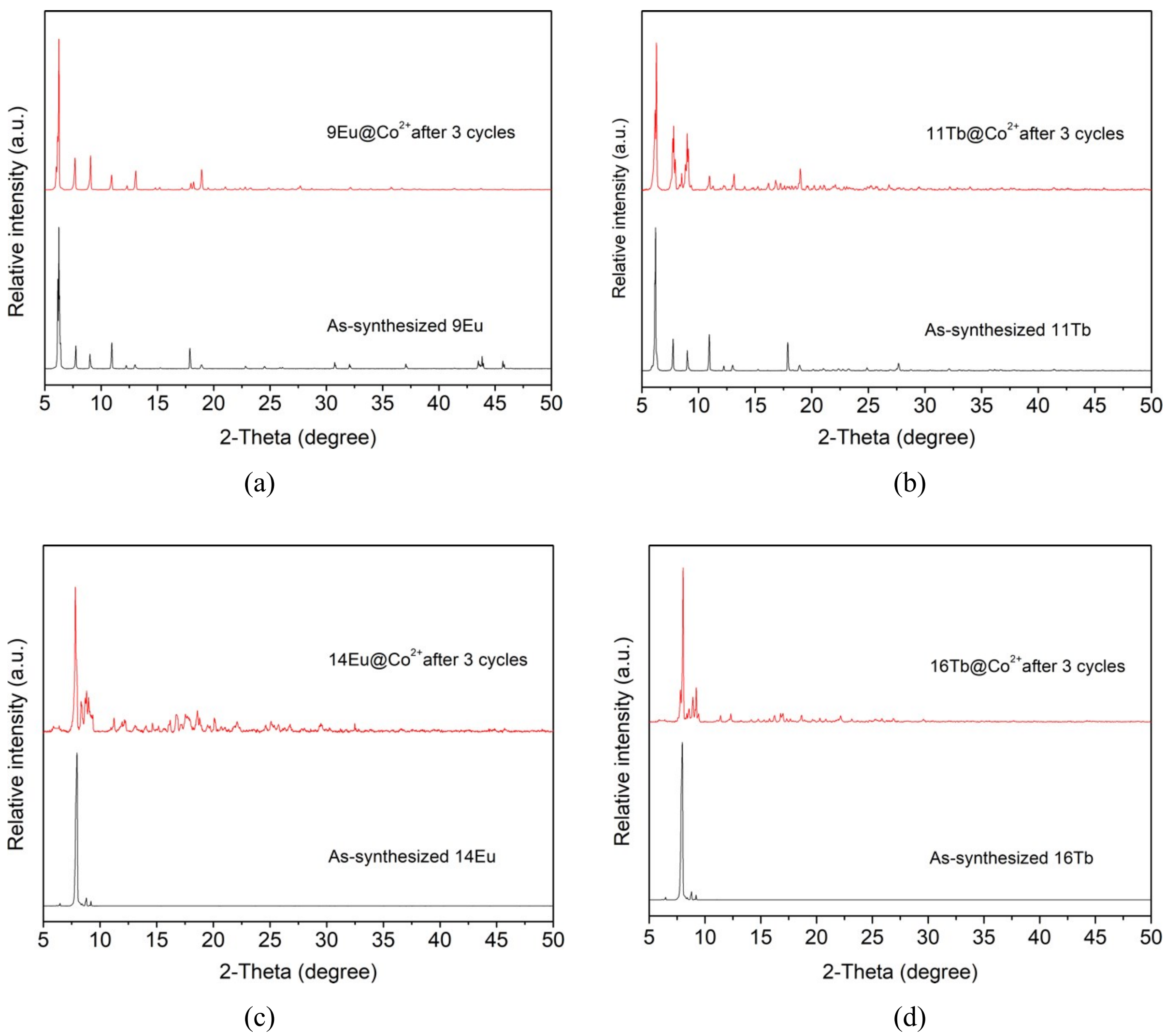
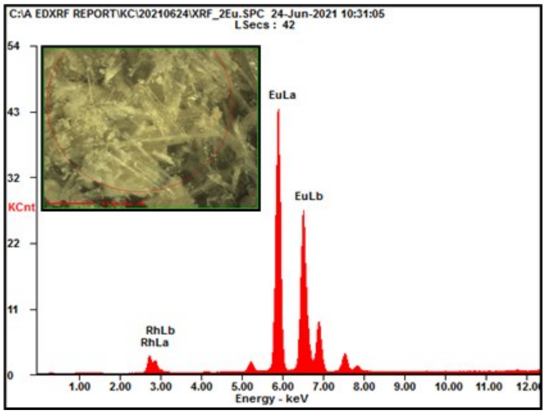
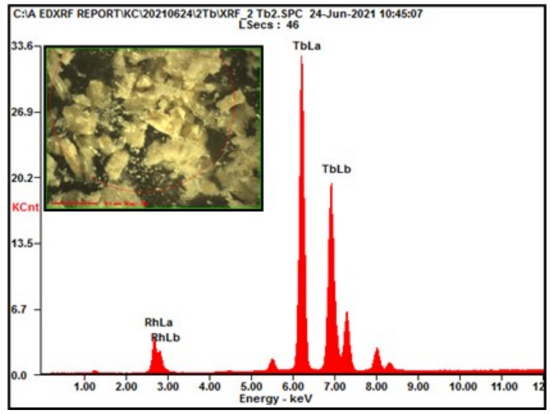


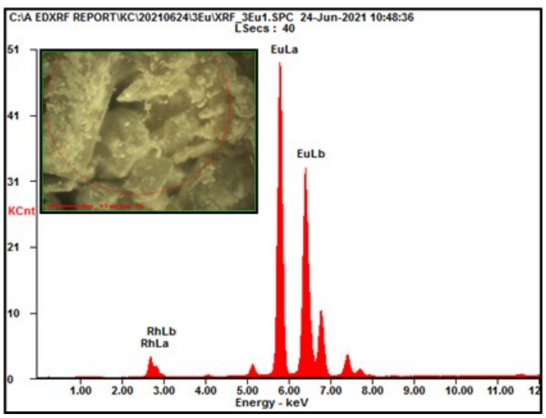
Figure S19. PXRD patterns of **9** (a), **11** (b), **14** (c), and **16** (d), before and after three cycles of sensing tests.



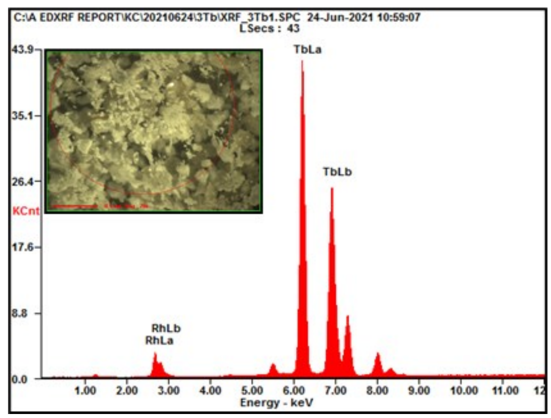
(a)



(b)



(c)



(d)

Figure S20. XRF spectra of **9** (a), **11** (b), **14** (c), and **16** (d) after sensing tests.