# Supporting Information

# Microporous Hexanuclear Ln(III) Cluster-based Metal-Organic Frameworks: Color Tunability for Barcode Application and Selective Removal of Methylene Blue

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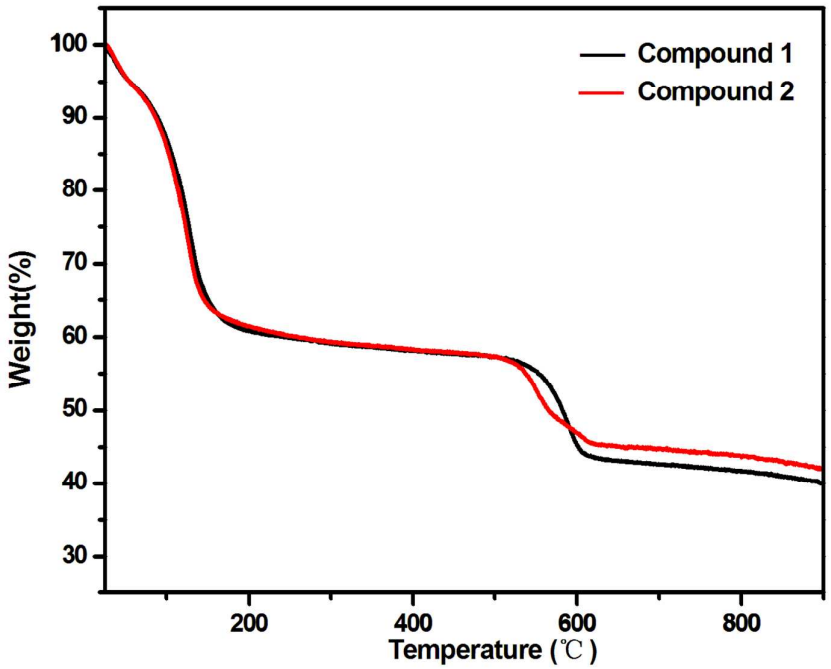


**Figure S1** The thermal stability of **1** and **2** was examined by thermogravimetric analysis (TGA) in the temperature range of 25-900 $^{o}$C with a heating rate of 5 $^{o}$C min$^{-1}$ under a $N_2$ atmosphere.

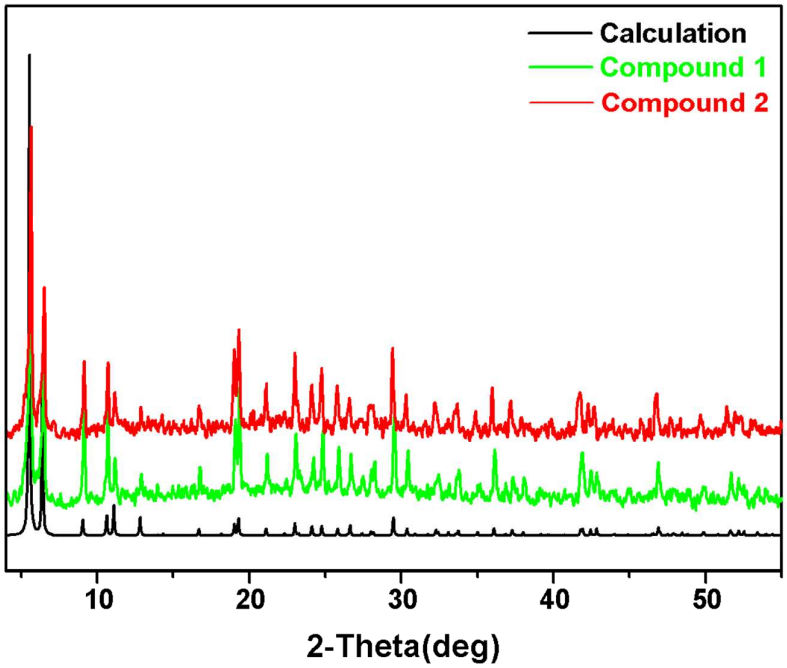


**Figure S2** The measured PXRD patterns of complexes **1**, **2** in comparison to the simulated pattern of single-crystal data.

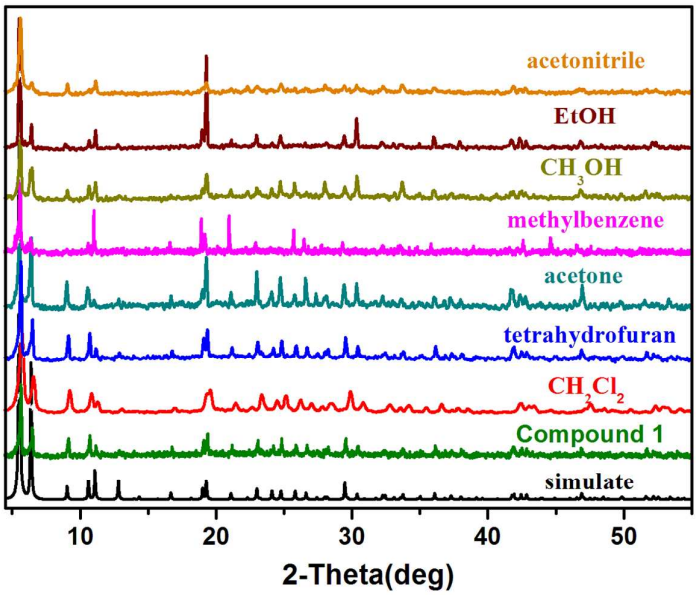


**Figure S3** The measured PXRD patterns of solvent-stable to the compound **1** in comparison to the simulated pattern of single-crystal data.

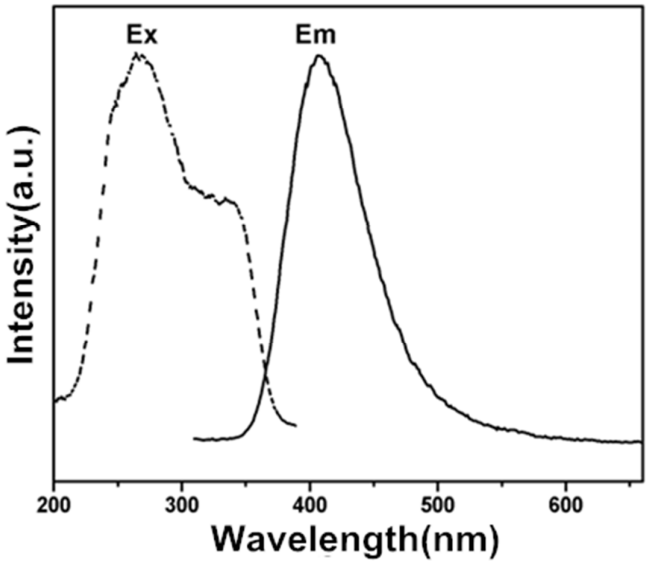


**Figure S4** Excitation and emission spectra of the $H_2$BPDC.

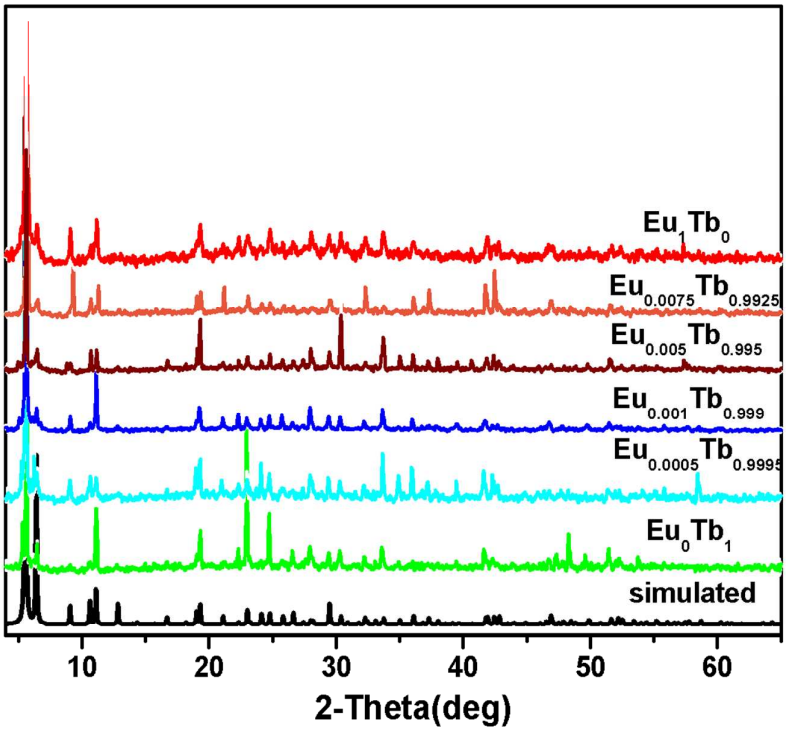


**Figure S5** The measured PXRD patterns of bimetallic $Eu_xTb_{1-x}$ series of complexes **1**, **2**, **3-6** in comparison to the simulated pattern of single-crystal **1**.

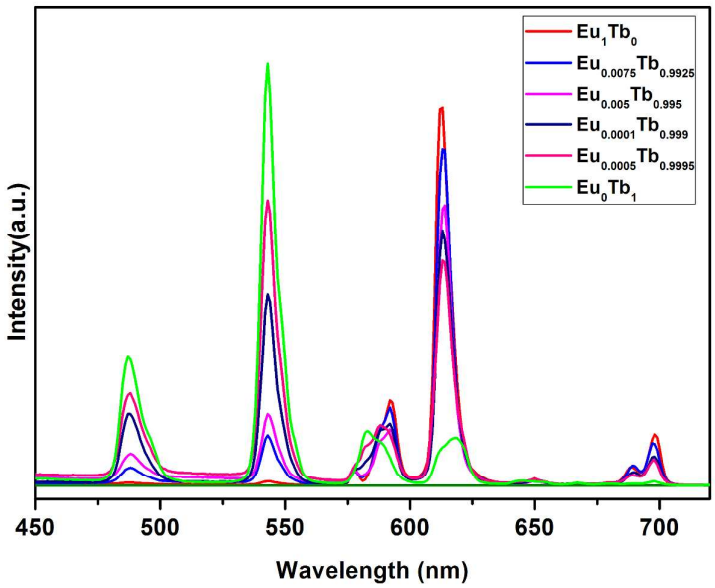


**Figure S6** Solid-state emission spectra of **1** and $Eu_xTb_{1-x}$-MOF excited with 300 nm UV light.

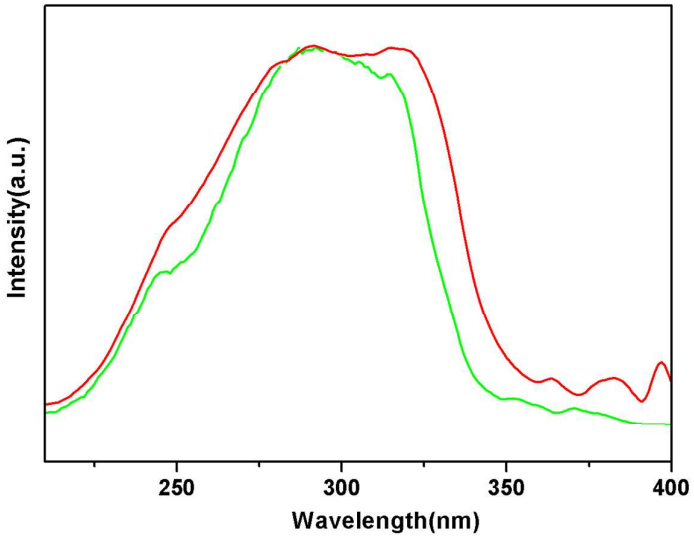


**Figure S7** Solid-state excitation spectra at room temperature.Represented by **$Eu_{0.0075}Tb_{0.9925}$-6** when monitored at respective $Eu^{3+}$ and $Tb^{3+}$ characteristic emission peaks.

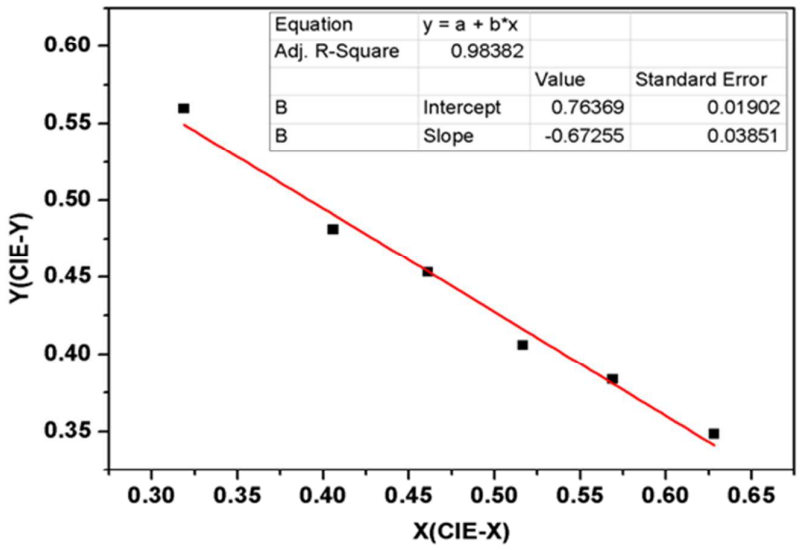


**Figure S8** Coordinates of mixed Ln(III)-based MOFs, **3**, **4**, **5** and **6** in comparison to compound **1** and **2** when excited at 300 nm, showing variation depending on $Tb^{3+}/Eu^{3+}$ atomic ratio. By changing the content of $Tb^{3+}$ and $Eu^{3+}$, the emission color of these materials can be fine-tuned in the RG spectra region.

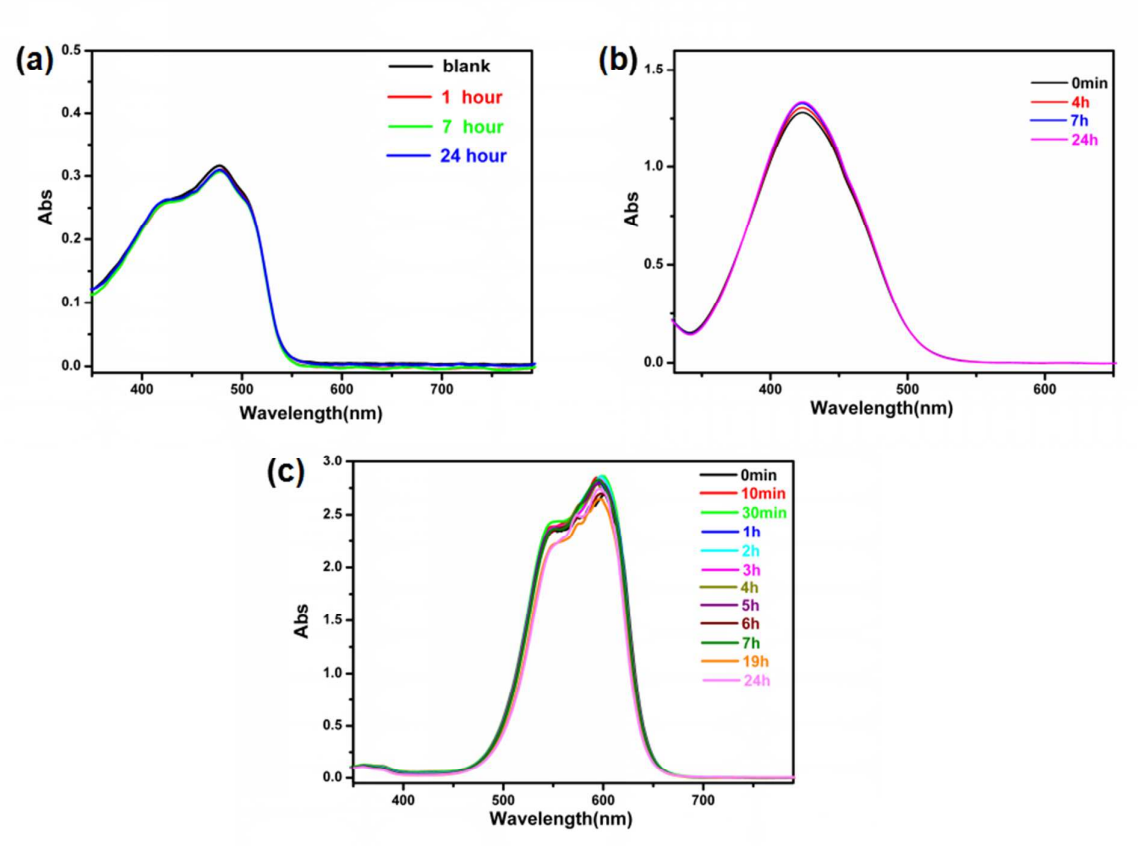


**Fig. S9** UV-vis spectra of DMF solution of $MO^-$, $SD^0$ and $CV^+$ in the presence of Ln(III)-based MOF **1**.

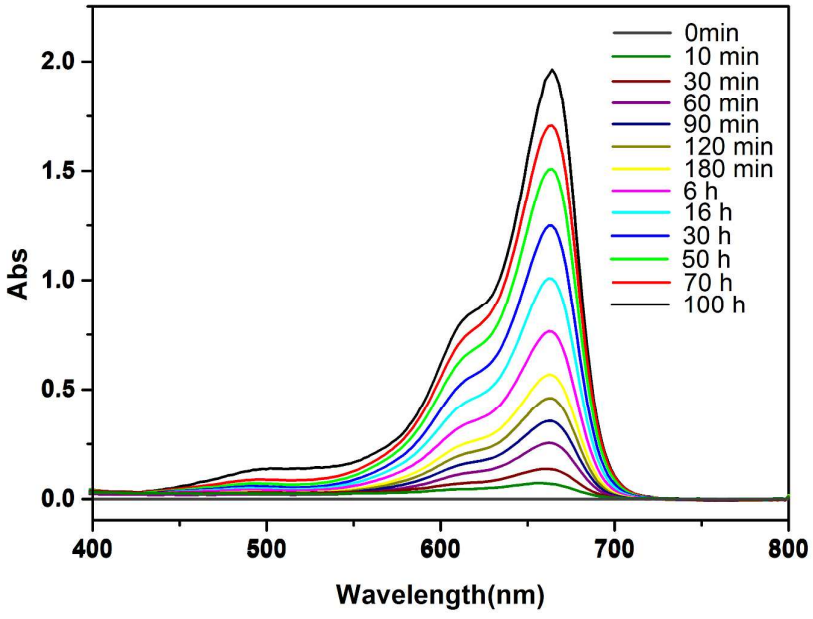

**Fig. S10** UV-vis spectra of $MB^+$ release from $MB^+$@**1** in the DMF solution.

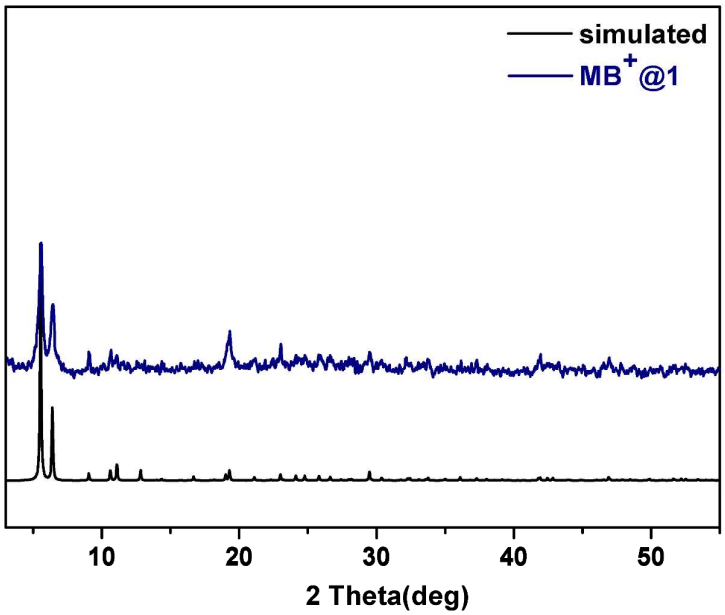


**Fig. S11** Powder XRD patterns for simulated **1** and $MB^+$@**1** after $MB^+$ release.

**Table S1** Crystal data and structure refinement for **1** and **2**

| Compounds | **1** | **2** |
|---|---|---|
| Empirical formula | C42 H28 O16 Tb3 | C42 H28 O16 Eu3 |
| Formula weight | 1265.40 | 1244.52 |
| Temperature | 100(2) K | 100(2) K |
| Wavelength | 0.71073 A | 1.54184 A |
| Crystal system | Cubic | Cubic |
| Space group | *F*m-3m | *F*m-3m |
| Unit cell dimensions | a = 27.5460(11) Å α = 90°.<br>b = 27.5460(11) Å β = 90°.<br>c = 27.5460(11) Å γ = 90°. | a = 27.5460(11) Å α = 90°.<br>b = 27.5460(11) Å β = 90°.<br>c = 27.5460(11) Å γ = 90°. |
| Volume | 20901(3) $Å^3$ | 20901(3) $Å^3$ |
| Z | 8 | 8 |
| Density (calculated) | 0.804 $Mg/m^3$ | 0.791 $Mg/m^3$ |
| Absorption coefficient | 2.037 $mm^{-1}$ | 12.962 $mm^{-1}$ |

| | | |
|---|---|---|
| F(000) | 4824 | 4776 |
| Crystal size | 0.150 x 0.140 x 0.130 $mm^3$ | 0.150 x 0.130 x 0.100 $mm^3$ |
| Theta range for data collection | 2.958 to 28.272°. | 4.540 to 73.938°. |
| Index ranges | -36<=h<=36, -36<=k<=36, -36<=l<=36 | -33<=h<=32, -14<=k<=23, -30<=l<=22 |
| Reflections collected | 87687 | 5684 |
| Independent reflections | 1343 [R(int) = 0.0669] | 1087 [R(int) = 0.0298] |
| Completeness to theta = 25.03° | 99.5 % | 99.1 % |
| Absorption correction | Semi-empirical from equivalents | Semi-empirical from equivalents |
| Max. and min. transmission | 0.7457 and 0.6544 | 1.00000 and 0.72208 |
| Refinement method | Full-matrix least-squares on $F^2$ | Full-matrix least-squares on $F^2$ |
| Data / restraints / parameters | 1343 / 27 / 37 | 1087 / 45 / 43 |
| Goodness-of-fit on F2 | 1.132 | 1.088 |
| Final R indices [I>2sigma(I)][a] | R1 = 0.0655, wR2 = 0.1652 | R1 = 0.0432, wR2 = 0.1180 |
| R indices (all data) | R1 = 0.0881, wR2 = 0.1970 | R1 = 0.0471, wR2 = 0.1218 |
| Largest diff. peak and hole | 3.348 and -2.414 $e.A^{-3}$ | 1.951 and -0.950 $e.A^{-3}$ |

[a] $R_1 = \sum||F_o|-|F_c||/\sum|F_o|$; $wR_2 = \sum[w(F_o^2-F_c^2)^2]/\sum[w(F_o^2)^2]^{1/2}$

**Table S2** The $^5D_4$ of $Tb^{3+}$ and $^5D_0$ of $Eu^{3+}$ lifetimes for Ln(III)-based MOFs **1**, **2**, **3**-**6**. The decay curves are monitored at 547 nm and 617 nm and excited at 300 nm.

| Mixed-$Tb_xEu_{1-x}$-MOFs | The mole ratio of Eu (based on RE) | |
|---|---|---|
| | $^5D_4$ of $Tb^{3+}$/μs | $^5D_0$ of $Eu^{3+}$/μs |
| 1 | 144.179 | — |
| 2 | — | 781.5773 |
| 3 | 38.4892 | 917.135 |
| 4 | 31.0129 | 945.127 |
| 5 | 25.5415 | 972.682 |
| 6 | 19.9444 | 975.511 |

**Table S3** The ICP-OES analysis results of mixed $Eu_xTb_{1-x}$-MOFs **3-6**.

| Mixed-$Tb_xEu_{1-x}$-MOFs | The mole ratio of Eu (based on Ln(III)) | | Molecular formula |
|---|---|---|---|
| | Theoretical | Measured | |
| 3 | 0.0005 | 0.0005 | $Eu_{0.0005}Tb_{0.9995}C_{88}H_{80}N_2O_{18}$ |
| 4 | 0.001 | 0.0009 | $Eu_{0.0009}Tb_{0.9991}C_{88}H_{80}N_2O_{18}$ |
| 5 | 0.005 | 0.0053 | $Eu_{0.0053}Tb_{0.9967}C_{88}H_{80}N_2O_{18}$ |
| 6 | 0.0075 | 0.0076 | $Eu_{0.0076}Tb_{1.9924}C_{88}H_{80}N_2O_{18}$ |

**Table S4** The chromaticity coordinates of the six binary $Eu_xTb_{1-x}$ complexes when excitedwith 300 nm.

| Mixed-$Tb_xEu_{1-x}$-MOFs(Samples) | CIE(X, Y ) |
|---|---|
| 1 | 0.3187, 0.5598 |
| 3 | 0.4058, 0.481 |
| 4 | 0.4612, 0.4531 |
| 5 | 0.5164,0.406 |
| 6 | 0.569, 0.3838 |
| 2 | 0.6282, 0.3485 |