

Supporting Information for:

## **A Mixed Heterobimetallic Y/Eu-MOF for the Cyanosilylation and Hydroboration of Carbonyls**

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## 1. General experimental information

**Elemental analyses** (EA) of synthesized novel catalyst compound **Y/Eu-MOF** performed on an Elementar vario EL cube in the CHN mode.

**IR (ATR) spectra** of 3-amino-4-hydroxybenzoic acid ligand and compound **Y/Eu-MOF** collected in the region 400–4000  $\text{cm}^{-1}$  on a Nicolet 6700 FTIR (Fourier transform infrared) spectrophotometer (Thermo Fisher Scientific, TX, USA) KBr pellets.

**NMR measurements:** NMR spectra were measured in a Bruker Avance III 300 spectrometer equipped with a direct double SmartProbe BBFO  $^1\text{H}/\text{BB}(^{19}\text{F})$  probe. Chemical shifts are reported in parts per million (ppm) relative to residual solvent peak ( $\text{CDCl}_3$ ,  $^1\text{H}$ : 7.26 ppm;  $^{13}\text{C}$ : 77.16 ppm). Coupling constants are reported in Hertz. Multiplicity is reported with the usual abbreviations (s: singlet, bs: broad singlet, d: doublet, dd: doublet of doublets, ddd: doublet of doublet of doublets, t: triplet, td: triplet of doublets, q: quartet, dq: doublet of quartet, p: pentet, sex: sextet, hept: heptet, m: multiplet).

**ICP-AES** analysis of catalyst **1Y-Eu** was conducted in Horiba Yobin Yvon Activa atomic emission spectrometer equipped with a glass and Teflon nebulizer system, which enables samples from acidic digestion to be determined using HF. This equipment also provides the option of coupling a hydride generation system which enables elements such as As, Hg and Sb, etc. to be determined, in very low levels of concentration (ppb). The equipment is controlled by a control computer with Activa Analyst 5.4 software, which enables it to interact with the equipment at all times. It permits sequential multi-elemental analysis and also enables numerous analytical requirements to be met due to the large linear interval that characterizes this technique, which in turn facilitates the analysis of majority and minority elements.

**SEM-EDX**, Scanning Electron Microscopy (SEM Leo 1430 VP) with microanalysis of elements (EDX): Scanning electron microscopy, SEM Leo 1430VP, linked to a system for the microanalysis of elements via energy-dispersive X-ray spectroscopy, Inca 350, version 17 (hereinafter, SEM-EDX). Scanning electron microscopy enables the identification of elements with low atomic numbers, including carbon.

**X-ray Diffraction Data Collection and Structure Determination.** Single-crystal diffraction data were collected at 100(2) K on a Bruker X8 APEX II and Bruker D8 Venture with a Photon detector equipped with graphite monochromated MoK $\alpha$  radiation ( $\lambda = 0.71073$  Å). The data reduction was performed with the APEX2 software[1] and corrected for absorption using SADABS.[2] These structures were solved by direct methods using the SHELXT program[3] and refined by full-matrix least-squares of F2 including all reflections with SHELXL-2018/3 program.[4] All calculations for these structures were performed using the WINGX crystallographic software package.[5]

**Powder X-ray diffractions (XRPD)** patterns were collected on a Philips X'PERT powder diffractometer with Cu K $\alpha$  radiation ( $\lambda = 1.5418$  Å) over the range of  $5 < 2\theta < 50^\circ$  with a step size of  $0.02^\circ$  and an acquisition time of 2.5 s per step at 25 °C. Indexation of the diffraction profiles were carried out using the FULLPROF program,[6] on the basis of the space group and cell parameters found for isostructural compounds by single crystal X-ray diffraction.

**Electrophoretic mobility and  $\zeta$ -potential** measurements were made in a ZetaSizer Nano-Z ZEN2600 instrument, DTS1070 cell type and laser wavelength of 632.8 nm. Samples were prepared at 0.05% w/w, dispersing 2.5 mg of the powder and making up to weight with distilled water. Two vials per measuring point were prepared, adjusting the pH manually under magnetic stirring with NaOH and HCl solutions. The dispersions were sonicated after preparation for 1 min to tear the aggregates. Subsequently, they were shaken and left for sedimentation for another 5 min. After this time, the supernatant was extracted, and the deposit was placed in the stove at 70°C to estimate the representative amount of sample studied. Afterwards, the pH of the supernatants was checked, and the conductivity was set to 330  $\mu$ S/cm with NaCl 0.1 M. Samples were shaken vigorously prior to their insertion into

the measuring devices. Equilibration time was set to 120 s. Average results obtained from 5 measurements.

**Optic micrograph analysis** was carried out with a Nikon Eclipse Ti-E motorized inverted microscope equipped with objectives: Plan Fluor 10x, Plan apochromatic 20x, 40x, 60x oil, with a transmission detector.

## 2. General procedures

**General procedure for the scale-up synthesis of Y/Eu-MOF**  
 $\{[Y_{3.5}Eu_{1.5}L_6(OH)_3(H_2O)_3] \cdot 12DMF\}_n$ : 3-amino-4-hydroxybenzoic acid (0.2 g, 1.2 mmol) was weighted and dissolved in 1.2/3 mL DMF/H<sub>2</sub>O solvent-mixture containing 200  $\mu$ l of Et<sub>3</sub>N (1.44 mmol). To this solution, a metal mixture composed by 0.124 g (0.289 mmol) of Eu(NO<sub>3</sub>)<sub>3</sub>·5H<sub>2</sub>O and 0.221 g (0.578mmol) of Y(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O previously dissolved in 1.8 mL water, was added dropwise keeping magnetic agitation. A brownish precipitate stars appearing while metal solution is added, nonetheless it ends up redissolving to give rise to a solution. After heating in the oven at 100 °C for 2 hours, single crystalline product was obtained (83 mg) of heterobimetallic **Y/Eu-MOF**. PXRD allowed us confirming the purity of the sample.

**Quantitative NMR acquisition parameters.** <sup>1</sup>H NMR determination of product conversion was carried out by comparing signals arising from both CH of aldehyde or CH<sub>3</sub> of ketone **1** and products **2**, **3** or **4**. The standard acquisition parameters were one-dimensional pulse sequence which includes a 30° flip angle (Bruker zg30), recycle time (D1 = 30 s), time domain (TD = 27k), number of scans (NS = 32), acquisition time (AQ = 2.05 s), transmitter (frequency) offset (O1P = 6.0 ppm), and spectral width (SW = 22.0 ppm).

### 3. Chemical characterization of compounds

#### Elemental analysis

**Table S1.** Elemental analysis of compounds **Y/Eu-MOF**.

Compound	Formula	Molecular weight	Calc.	Found.
<b>1Y-Eu</b>	$C_{78}H_{115}N_{18}O_{36}Y_{3.5}Eu_{1.5}$	2419.98	C: 38.71; H: 4.79; N: 10.42; O: 23.80; Y: 12.86; Eu: 9.42	C: 38.76; H: 4.83; N: 10.44; O: 23.81; Y: 12.88; Eu: 9.46

#### Determination of the metal content by ICP-AES

**Table S2.** ICP-AES results of compound **Y/Eu-MOF**.

Compound	Y content in $mgL^{-1}$	Eu content in $mgL^{-1}$	Y to Eu relationship
<b>1Y-Eu</b>	1672 (18.8 mM)	1268 (8.34 mM)	$\pm 2.25 \sim 2.33$

## 4. Crystallographic data

**Table S3.** Crystallographic data and structure refinement details of compound **Y/Eu-MOF**.

Compound	Y/Eu-MOF
Formula	C <sub>78</sub> H <sub>115</sub> N <sub>18</sub> O <sub>36</sub> Y <sub>3.5</sub> Eu <sub>1.5</sub>
$M_r$	2419.98
Crystal system	hexagonal
Space group (no.)	$P6_3/m$ (176)
a(Å)	15.8510(3)
b(Å)	15.8510(3)
c(Å)	16.8255(7)
$\alpha(^{\circ})$	90
$\beta(^{\circ})$	90
$\gamma(^{\circ})$	120
V(Å <sup>3</sup> )	3661.1(2)
Z	2
$\rho_{\text{calc}}/\text{cm}^3$	2.193
$\mu/\text{mm}^{-1}$	4.130
F(000)	2454.0
Crystal size/mm <sup>3</sup>	0.658 × 0.167 × 0.103
Radiation	MoK $\alpha$ ( $\lambda$ = 0.71073)
2 $\Theta$ range for data collection/ $^{\circ}$	5.14 to 54.952
Index ranges	-18 ≤ h ≤ 20, -20 ≤ k ≤ 20, -21 ≤ l ≤ 21
Reflections collected	72657
Independent reflections	2898 [ $R_{\text{int}}$ = 0.0886, $R_{\text{sigma}}$ = 0.0238]
Data/restraints/parameters	2898/0/123
Goodness-of-fit on F <sup>2</sup>	1.127
Final R indexes [ $I \geq 2\sigma(I)$ ]	$R_1$ = 0.0276, $wR_2$ = 0.0889
Final R indexes [all data]	$R_1$ = 0.0341, $wR_2$ = 0.0922
Largest diff. peak/hole / e Å <sup>-3</sup>	0.75/-0.52



## 5. Selected bond lengths and angles data

**Table S4.** Selected bond lengths (Å) and angles (°) for compound **Y/Eu-MOF**.

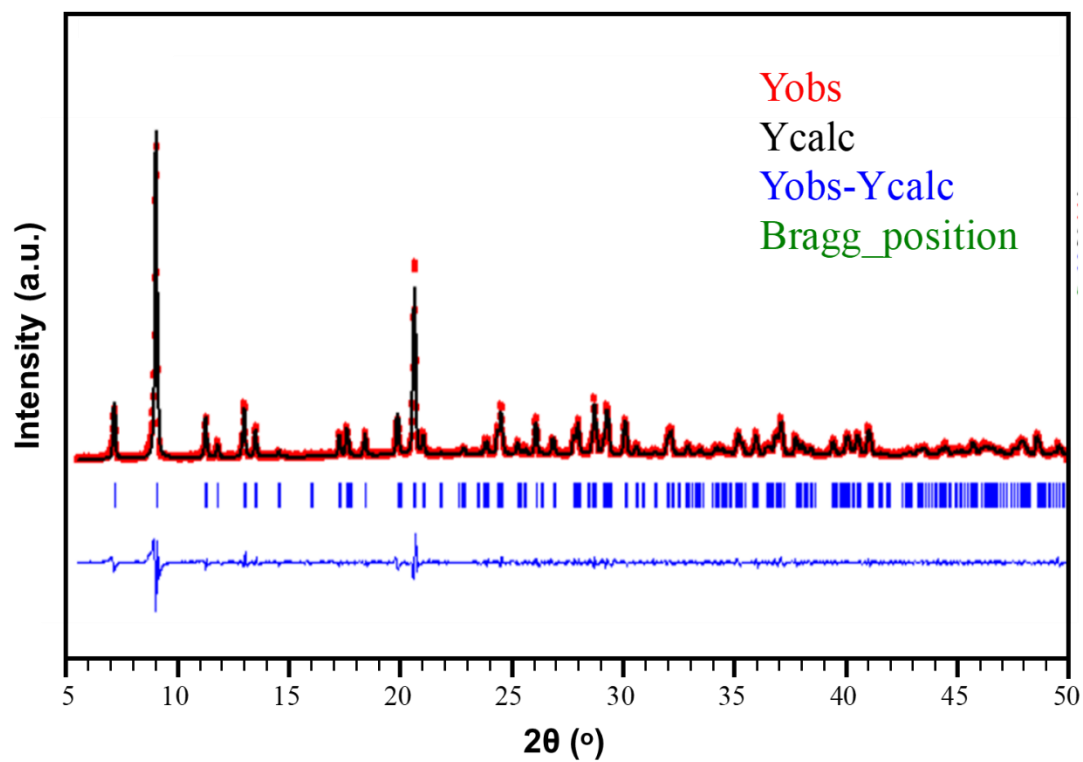
Atom	Atom	Length/Å
Eu1	Eu1 <sup>2</sup>	3.5335(7)
Eu1	Y2	3.9093(3)
Eu1	Y2 <sup>3</sup>	3.9093(3)
Eu2	Y1	3.9093(3)
N1	Eu1	2.507(3)
N1	Y1	2.507(3)
O1	Eu2	2.341(2)
O1	Y1	2.512(2)
O1	Eu1	2.512(2)
O1	Y2	2.341(2)
O2	Y2 <sup>1</sup>	2.419(2)
O3	Y2 <sup>1</sup>	2.428(2)
O4	Eu1	2.3830(19)
O4	Eu1 <sup>2</sup>	2.3831(19)
O4	Y2	2.321(3)
O4	Eu2	2.321(3)
O4	Y1	2.3830(19)
O5	Y2	2.332(3)
O5	Eu2	2.332(3)
<sup>1</sup> +y, -x+y, 1-z; <sup>2</sup> +x, +y, 1/2-z; <sup>3</sup> 1-y, +x-y, +z		

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
Eu1 <sup>4</sup>	Eu1	Y2	63.133(5)	O1 <sup>3</sup>	Eu1	Y2 <sup>2</sup>	135.14(5)	O4	Eu2	Y1	34.30(4)
Eu2	O1	Y1	107.28(8)	O1 <sup>3</sup>	Y1	Eu2	97.73(5)	O4	Y1	N1 <sup>3</sup>	89.48(8)
Eu2	O4	Y1	112.41(8)	O1 <sup>4</sup>	Y2	O1	101.22(10)	O4	Y1	N1 <sup>2</sup>	142.66(9)
N1	Eu1	N1 <sup>3</sup>	81.02(10)	O1 <sup>4</sup>	Y2	O2 <sup>5</sup>	145.35(7)	O4	Y1	N1	133.23(9)
N1	Eu1	O1	65.68(8)	O1 <sup>4</sup>	Y2	O2 <sup>6</sup>	82.25(7)	O4	Y1	O1 <sup>3</sup>	74.26(8)
N1	Eu1	O1 <sup>2</sup>	68.90(8)	O1 <sup>4</sup>	Y2	O3 <sup>6</sup>	87.22(7)	O4	Y1	O1	68.16(8)
N1	Eu1	O1 <sup>3</sup>	137.57(8)	O1 <sup>4</sup>	Y2	O3 <sup>5</sup>	159.46(7)	O4	Y1	O1 <sup>2</sup>	132.92(7)
N1	Eu1	Eu1 <sup>4</sup>	131.40(7)	O1 <sup>4</sup>	Y2	Eu1 <sup>4</sup>	37.84(5)	O4	Y1	Eu2	33.29(6)
N1	Eu1	Y2 <sup>2</sup>	77.87(6)	O1 <sup>4</sup>	Y2	Eu1	84.77(5)	O4 <sup>2</sup>	Eu1	N1 <sup>2</sup>	133.23(9)
N1	Eu1	Y2	100.22(6)	O1 <sup>4</sup>	Eu2	O1	101.22(10)	O4 <sup>2</sup>	Eu1	N1 <sup>3</sup>	142.66(9)
N1	Y1	N1 <sup>3</sup>	81.02(10)	O1 <sup>4</sup>	Eu2	O2 <sup>6</sup>	82.25(7)	O4 <sup>2</sup>	Eu1	N1	89.48(8)
N1	Y1	O1	65.68(8)	O1 <sup>4</sup>	Eu2	O2 <sup>5</sup>	145.35(7)	O4 <sup>2</sup>	Eu1	O1 <sup>2</sup>	68.16(8)
N1	Y1	O1 <sup>2</sup>	68.90(8)	O1 <sup>4</sup>	Eu2	O3 <sup>5</sup>	159.46(7)	O4 <sup>2</sup>	Eu1	O1 <sup>3</sup>	132.92(7)
N1	Y1	O1 <sup>3</sup>	137.57(8)	O1 <sup>4</sup>	Eu2	O3 <sup>6</sup>	87.22(7)	O4 <sup>2</sup>	Eu1	O1	74.26(8)
N1	Y1	Eu2	100.22(6)	O1 <sup>4</sup>	Eu2	Y1	84.77(5)	O4 <sup>2</sup>	Eu1	O4	71.06(8)
N1 <sup>2</sup>	Eu1	N1 <sup>3</sup>	81.02(10)	O2 <sup>5</sup>	Y2	O2 <sup>6</sup>	76.55(11)	O4 <sup>2</sup>	Eu1	O4 <sup>3</sup>	71.06(8)
N1 <sup>2</sup>	Eu1	N1	81.02(10)	O2 <sup>5</sup>	Y2	O3 <sup>5</sup>	53.83(7)	O4 <sup>2</sup>	Eu1	Eu1 <sup>4</sup>	42.15(5)
N1 <sup>2</sup>	Eu1	O1 <sup>3</sup>	68.90(8)	O2 <sup>5</sup>	Y2	O3 <sup>6</sup>	101.49(8)	O4 <sup>2</sup>	Eu1	Y2	68.37(7)
N1 <sup>2</sup>	Eu1	O1	137.58(8)	O2 <sup>5</sup>	Y2	Eu1 <sup>4</sup>	109.74(5)	O4 <sup>2</sup>	Eu1	Y2 <sup>2</sup>	33.29(6)
N1 <sup>2</sup>	Eu1	O1 <sup>2</sup>	65.68(8)	O2 <sup>5</sup>	Y2	Eu1	77.16(5)	O4 <sup>2</sup>	Y1	N1	89.48(8)
N1 <sup>2</sup>	Eu1	Eu1 <sup>4</sup>	131.40(7)	O2 <sup>5</sup>	Eu2	O2 <sup>6</sup>	76.55(11)	O4 <sup>2</sup>	Y1	N1 <sup>3</sup>	142.66(9)
N1 <sup>2</sup>	Eu1	Y2 <sup>2</sup>	100.22(6)	O2 <sup>5</sup>	Eu2	O3 <sup>5</sup>	53.83(7)	O4 <sup>2</sup>	Y1	N1 <sup>2</sup>	133.23(9)
N1 <sup>2</sup>	Eu1	Y2	158.35(6)	O2 <sup>5</sup>	Eu2	O3 <sup>6</sup>	101.49(8)	O4 <sup>2</sup>	Y1	O1	74.26(8)
N1 <sup>2</sup>	Y1	N1 <sup>3</sup>	81.02(10)	O2 <sup>5</sup>	Eu2	Y1	77.16(5)	O4 <sup>2</sup>	Y1	O1 <sup>2</sup>	68.16(8)

N1 <sup>2</sup>	Y1	N1	81.02(10)	O2 <sup>6</sup>	Y2	O3 <sup>6</sup>	53.83(7)	O4 <sup>2</sup>	Y1	O1 <sup>3</sup>	132.92(7)
N1 <sup>2</sup>	Y1	O1 <sup>3</sup>	68.90(8)	O2 <sup>6</sup>	Y2	O3 <sup>5</sup>	101.49(8)	O4 <sup>2</sup>	Y1	O4 <sup>3</sup>	71.06(8)
N1 <sup>2</sup>	Y1	O1 <sup>2</sup>	65.68(8)	O2 <sup>6</sup>	Y2	Eu1 <sup>4</sup>	77.16(5)	O4 <sup>2</sup>	Y1	O4	71.06(8)
N1 <sup>2</sup>	Y1	O1	137.58(8)	O2 <sup>6</sup>	Y2	Eu1	109.74(5)	O4 <sup>2</sup>	Y1	Eu2	68.37(7)
N1 <sup>2</sup>	Y1	Eu2	158.35(6)	O2 <sup>6</sup>	Eu2	O3 <sup>5</sup>	101.49(8)	O4 <sup>3</sup>	Eu1	N1 <sup>3</sup>	133.23(9)
N1 <sup>3</sup>	Eu1	O1 <sup>3</sup>	65.68(8)	O2 <sup>6</sup>	Eu2	O3 <sup>6</sup>	53.83(7)	O4 <sup>3</sup>	Eu1	N1	142.66(9)
N1 <sup>3</sup>	Eu1	O1	68.90(8)	O2 <sup>6</sup>	Eu2	Y1	109.74(5)	O4 <sup>3</sup>	Eu1	N1 <sup>2</sup>	89.48(9)
N1 <sup>3</sup>	Eu1	O1 <sup>2</sup>	137.58(8)	O3 <sup>5</sup>	Y2	Eu1	112.25(5)	O4 <sup>3</sup>	Eu1	O1 <sup>2</sup>	74.26(8)
N1 <sup>3</sup>	Eu1	Eu1 <sup>4</sup>	131.40(7)	O3 <sup>5</sup>	Y2	Eu1 <sup>4</sup>	162.69(5)	O4 <sup>3</sup>	Eu1	O1	132.92(7)
N1 <sup>3</sup>	Eu1	Y2	77.86(6)	O3 <sup>5</sup>	Eu2	Y1	112.25(5)	O4 <sup>3</sup>	Eu1	O1 <sup>3</sup>	68.16(8)
N1 <sup>3</sup>	Eu1	Y2 <sup>2</sup>	158.35(6)	O3 <sup>6</sup>	Y2	O3 <sup>5</sup>	79.17(11)	O4 <sup>3</sup>	Eu1	O4	71.07(8)
N1 <sup>3</sup>	Y1	O1	68.90(8)	O3 <sup>6</sup>	Y2	Eu1	162.69(5)	O4 <sup>3</sup>	Eu1	Eu1 <sup>4</sup>	42.15(5)
N1 <sup>3</sup>	Y1	O1 <sup>3</sup>	65.68(8)	O3 <sup>6</sup>	Y2	Eu1 <sup>4</sup>	112.25(5)	O4 <sup>3</sup>	Eu1	Y2 <sup>2</sup>	68.37(7)
N1 <sup>3</sup>	Y1	O1 <sup>2</sup>	137.58(8)	O3 <sup>6</sup>	Eu2	O3 <sup>5</sup>	79.17(11)	O4 <sup>3</sup>	Eu1	Y2	101.50(6)
N1 <sup>3</sup>	Y1	Eu2	77.86(6)	O3 <sup>6</sup>	Eu2	Y1	162.69(5)	O4 <sup>3</sup>	Y1	N1 <sup>2</sup>	89.48(9)
O1	Eu1	O1 <sup>3</sup>	119.971(3)	O4	Eu1	N1 <sup>3</sup>	89.48(8)	O4 <sup>3</sup>	Y1	N1	142.66(9)
O1	Eu1	Eu1 <sup>4</sup>	90.97(5)	O4	Eu1	N1	133.23(9)	O4 <sup>3</sup>	Y1	N1 <sup>3</sup>	133.23(9)
O1	Eu1	Y2 <sup>2</sup>	97.73(5)	O4	Eu1	N1 <sup>2</sup>	142.66(9)	O4 <sup>3</sup>	Y1	O1 <sup>3</sup>	68.16(8)
O1	Eu1	Y2	34.88(5)	O4	Eu1	O1	68.16(8)	O4 <sup>3</sup>	Y1	O1	132.92(7)
O1	Y2	O2 <sup>6</sup>	145.35(7)	O4	Eu1	O1 <sup>2</sup>	132.92(7)	O4 <sup>3</sup>	Y1	O1 <sup>2</sup>	74.26(8)
O1	Y2	O2 <sup>5</sup>	82.25(7)	O4	Eu1	O1 <sup>3</sup>	74.26(8)	O4 <sup>3</sup>	Y1	O4	71.07(8)
O1	Y2	O3 <sup>6</sup>	159.46(7)	O4	Eu1	Eu1 <sup>4</sup>	42.15(5)	O4 <sup>3</sup>	Y1	Eu2	101.50(6)
O1	Y2	O3 <sup>5</sup>	87.22(7)	O4	Eu1	Y2	33.29(6)	O5	Y2	O1 <sup>4</sup>	79.84(7)
O1	Y2	Eu1	37.84(5)	O4	Eu1	Y2 <sup>2</sup>	101.50(6)	O5	Y2	O1	79.84(7)
O1	Y2	Eu1 <sup>4</sup>	84.77(5)	O4	Y2	O1	72.14(7)	O5	Y2	O2 <sup>6</sup>	134.13(7)

O1	Eu2	O2 <sup>5</sup>	82.25(7)	O4	Y2	O1 <sup>4</sup>	72.14(7)	O5	Y2	O2 <sup>5</sup>	134.13(7)
O1	Eu2	O2 <sup>6</sup>	145.35(7)	O4	Y2	O2 <sup>5</sup>	76.39(8)	O5	Y2	O3 <sup>6</sup>	83.34(9)
O1	Eu2	O3 <sup>6</sup>	159.46(7)	O4	Y2	O2 <sup>6</sup>	76.39(8)	O5	Y2	O3 <sup>5</sup>	83.34(9)
O1	Eu2	O3 <sup>5</sup>	87.22(7)	O4	Y2	O3 <sup>6</sup>	128.40(7)	O5	Y2	Eu1	110.21(8)
O1	Eu2	Y1	37.84(5)	O4	Y2	O3 <sup>5</sup>	128.40(7)	O5	Y2	Eu1 <sup>4</sup>	110.21(8)
O1	Y1	O1 <sup>3</sup>	119.971(3)	O4	Y2	O5	134.95(11)	O5	Eu2	O1 <sup>4</sup>	79.84(7)
O1	Y1	Eu2	34.88(5)	O4	Y2	Eu1 <sup>4</sup>	34.30(4)	O5	Eu2	O1	79.84(7)
O1 <sup>2</sup>	Eu1	O1	119.972(3)	O4	Y2	Eu1	34.30(4)	O5	Eu2	O2 <sup>6</sup>	134.13(7)
O1 <sup>2</sup>	Eu1	O1 <sup>3</sup>	119.971(3)	O4	Eu2	O1 <sup>4</sup>	72.14(7)	O5	Eu2	O2 <sup>5</sup>	134.13(7)
O1 <sup>2</sup>	Eu1	Eu1 <sup>4</sup>	90.97(5)	O4	Eu2	O1	72.14(7)	O5	Eu2	O3 <sup>6</sup>	83.34(9)
O1 <sup>2</sup>	Eu1	Y2 <sup>2</sup>	34.88(5)	O4	Eu2	O2 <sup>6</sup>	76.39(8)	O5	Eu2	O3 <sup>5</sup>	83.34(9)
O1 <sup>2</sup>	Eu1	Y2	135.14(5)	O4	Eu2	O2 <sup>5</sup>	76.39(8)	O5	Eu2	Y1	110.21(8)
O1 <sup>2</sup>	Y1	O1 <sup>3</sup>	119.971(3)	O4	Eu2	O3 <sup>5</sup>	128.40(7)	Y2	O1	Eu1	107.28(8)
O1 <sup>2</sup>	Y1	O1	119.972(3)	O4	Eu2	O3 <sup>6</sup>	128.40(7)	Y2	O4	Eu1	112.41(8)
O1 <sup>2</sup>	Y1	Eu2	135.14(5)	O4	Eu2	O5	134.95(11)	Y2 <sup>2</sup>	Eu1	Y2	101.165(7)
O1 <sup>3</sup>	Eu1	Eu1 <sup>4</sup>	90.97(5)								
O1 <sup>3</sup>	Eu1	Y2	97.73(5)								
<hr/>											
<sup>1</sup> +y,-x+y,1-z; <sup>2</sup> 1-y,+x-y,+z; <sup>3</sup> 1+y-x,1-x,+z; <sup>4</sup> +x,+y,1/2-z; <sup>5</sup> -y+x,+x,1-z; <sup>6</sup> -y+x,+x,-1/2+z <hr/>											

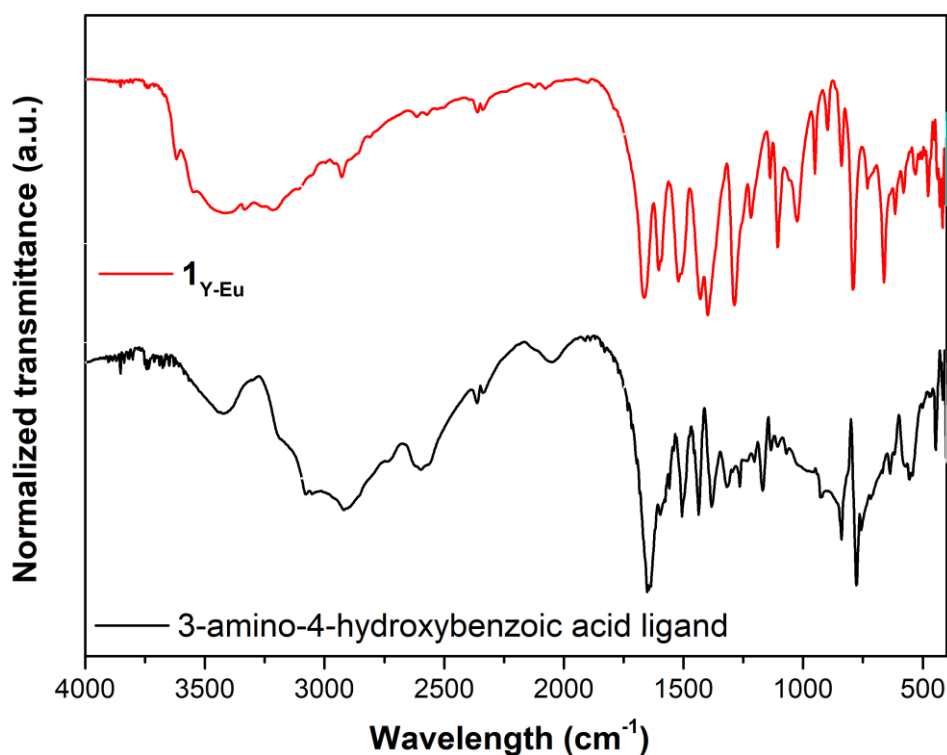
## 6. Powder X-ray diffraction analysis



**Figure S1.** Figure of the pattern matching analysis and experimental <sup>PXRD</sup> for **Y/Eu-MOF**.

## 7. FTIR analysis of Ligand and Catalyst

FTIR spectrum of **Y/Eu-MOF** display a narrow peak at around  $3617\text{ cm}^{-1}$  attributed to the N–H stretching vibration of the amine group of 3-amino-4-hydroxybenzoate ligand. Also, an intense and broad band attributable to O–H bond of free ligand along with a set of weak bands between  $3325$  and  $2917\text{ cm}^{-1}$  corresponding to the C–H vibrations of the aromatic ring of the 3-amino-4-hydroxybenzoate ligand are visible in the spectrum. Peaks in  $1683$ – $1408\text{ cm}^{-1}$  region are associated with asymmetric stretching vibrations of the carboxylate groups and the aromatic C–C and C–N bonds. The group of signals at lower range,  $1390$ – $1263\text{ cm}^{-1}$ , can be linked to symmetric stretching vibrations of the carboxylate groups. Remaining bands found at lower frequencies originated by distortions in the aromatic and carboxylate group of the ligand. Note that vibrational bands M–O and M–N bonds are observed below  $660\text{ cm}^{-1}$ .



**Figure S2.** Figure of the infrared spectra of the ligand and **Y/Eu-MOF**.

## 8. Continuous Shape Measurements

**Table S5.** Table of the continuous Shape Measurements for the  $\text{MN}_3\text{O}_6$  coordination environment.

EP-9	1 D9h	Enneagon
OPY-9	2 C8v	Octagonal pyramid
HBPY-9	3 D7h	Heptagonal bipyramid
JTC-9	4 C3v	Johnson triangular cupola J3
JCCU-9	5 C4v	Capped cube J8
CCU-9	6 C4v	Spherical-relaxed capped cube
JCSAPR-9	7 C4v	Capped square antiprism J10
CSAPR-9	8 C4v	Spherical capped square antiprism
JTCTPR-9	9 D3h	Tricapped trigonal prism J51
TCTPR-9	10 D3h	Spherical tricapped trigonal prism
JTDIC-9	11 C3v	Tridiminished icosahedron J63
HH-9	12 C2v	Hula-hoop
MFF-9	13 Cs	Muffin

Complex	JCSAPR-9	CSAPR-9	JTCTPR-9	TCTPR-9	MFF-9
<b>M1</b>	2.342	1.370	1.958	<b>0.666</b>	2.050

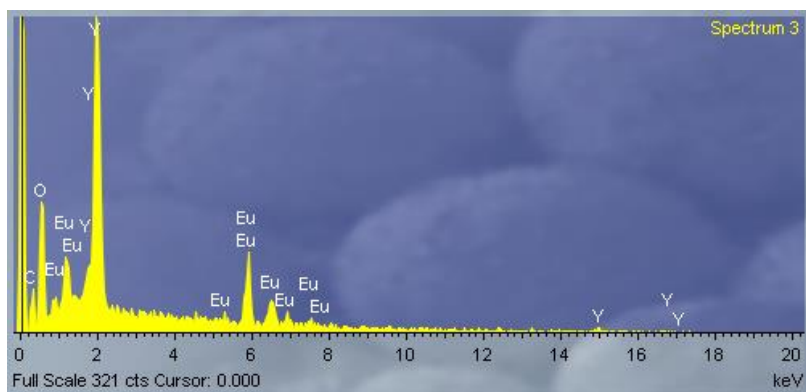
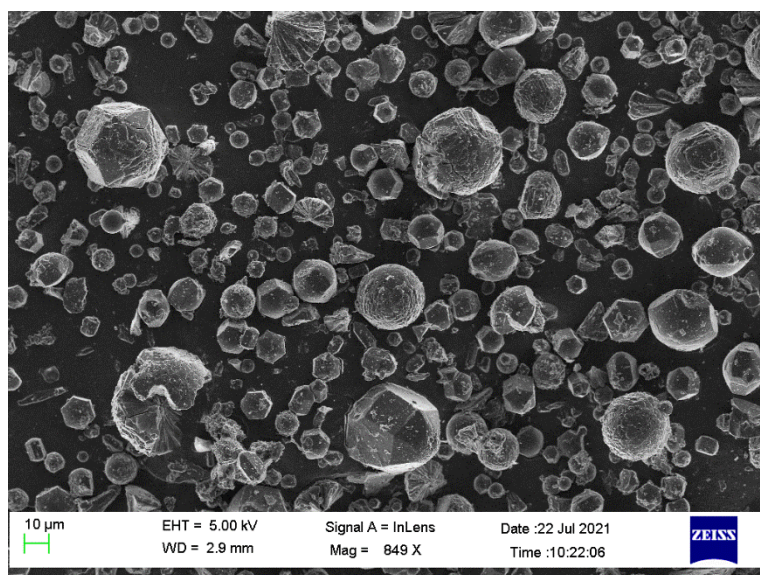
**Table S6.** Table of the continuous Shape Measurements for the MO<sub>8</sub> coordination environment.

OP-8	1 D8h	Octagon
HPY-8	2 C7v	Heptagonal pyramid
HBPY-8	3 D6h	Hexagonal bipyramid
CU-8	4 Oh	Cube
SAPR-8	5 D4d	Square antiprism
TDD-8	6 D2d	Triangular dodecahedron
JGBF-8	7 D2d	Johnson - Gyrobifastigium (J26)
JETBPY-8	8 D3h	Johnson - Elongated triangular bipyramid (J14)
JBTP-8	9 C2v	Johnson - Biaugmented trigonal prism (J50)
BTPR-8	10 C2v	Biaugmented trigonal prism
JSD-8	11 D2d	Snub disphenoid (J84)
TT-8	12 Td	Triakis tetrahedron
ETBPY-8	13 D3h	Elongated trigonal bipyramid

Complex	SAPR-8	TDD-8	JBTPR-8	BTPR-8	JSD-8
<b>M2</b>	2.703	<b>2.693</b>	3.668	3.234	5.526



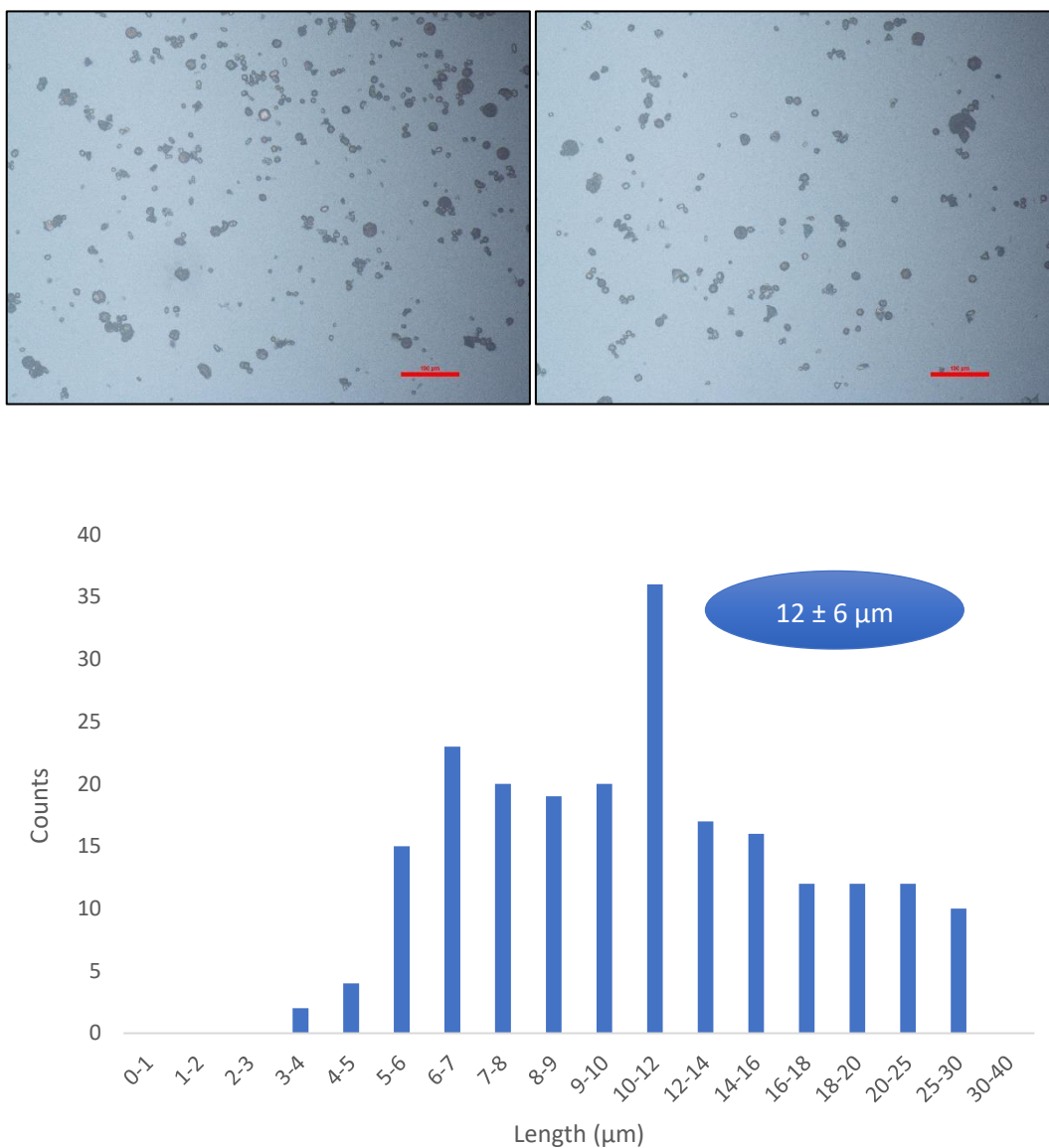
## 9. Scanning Electron Microscopy



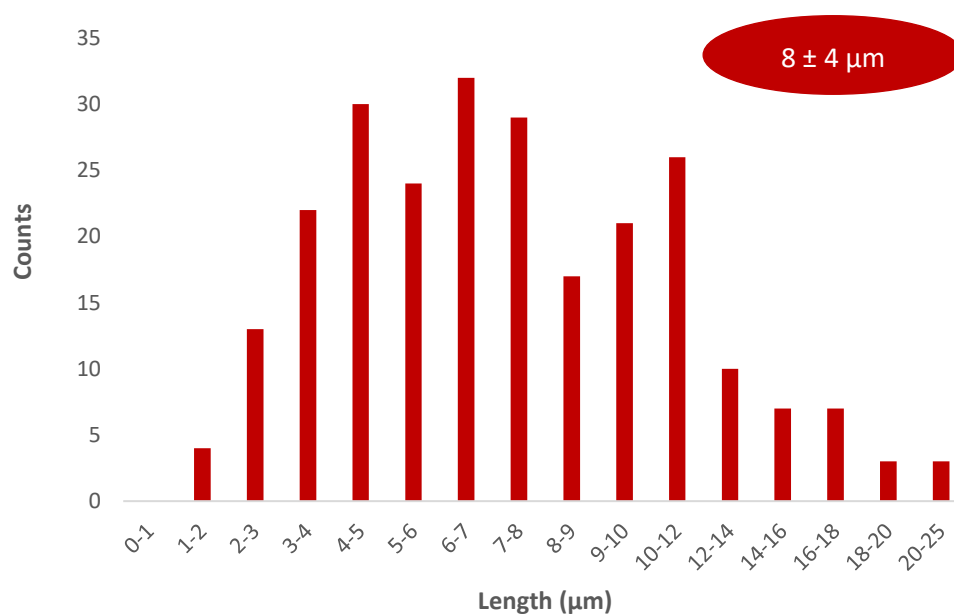
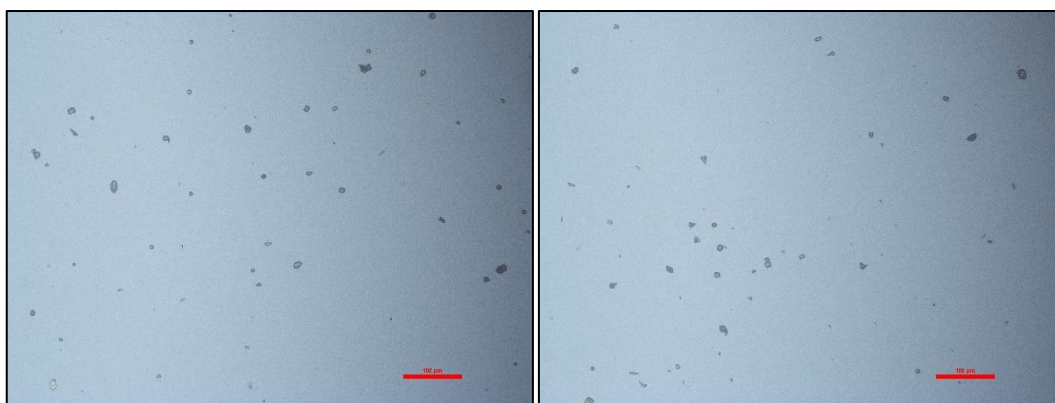
Spectrum	Weight%	Weight%	Weight%	Weight% Total	Atomic%	Atomic%	Atomic%
	O	Y	Eu		O	Y	Eu
	K_SERIES	L_SERIES	L_SERIES		K_SERIES	L_SERIES	L_SERIES
<b>Spectrum 3</b>	35.707	41.004	21.526	98.237	78.733	16.27	4.997

**Figure S3.** SEM and EDS mapping of bulk material of **Y/Eu-MOF**.

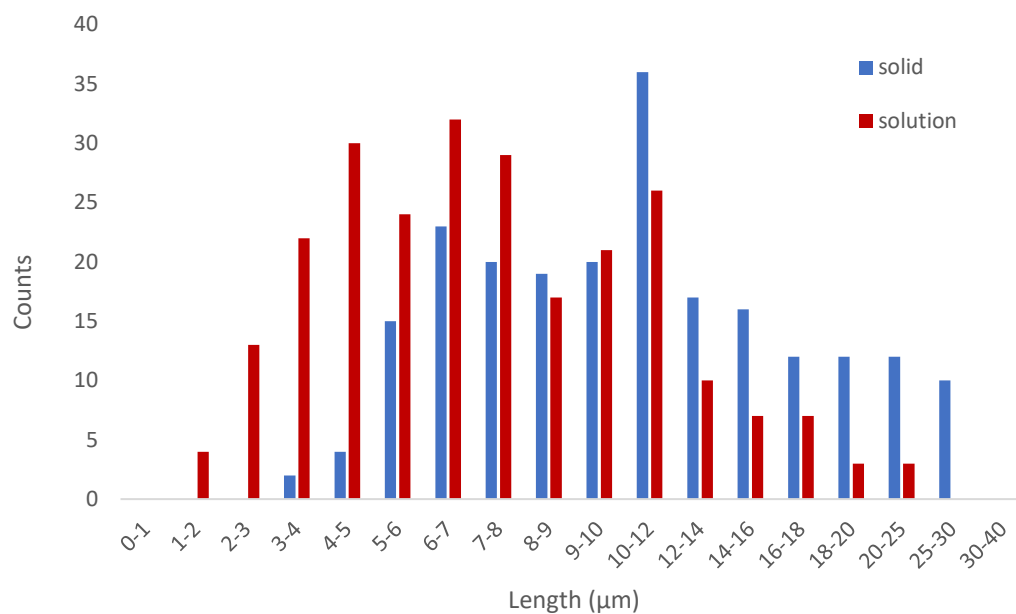
## 10. Particle size distribution of catalyst



**Figure S4.** Images and particle size distribution (an overall of 250 particles) in the deposited fraction of **Y/Eu-MOF** catalyst non-suspended in water (about a 68 % of the total amount), determined from optical microscope images.

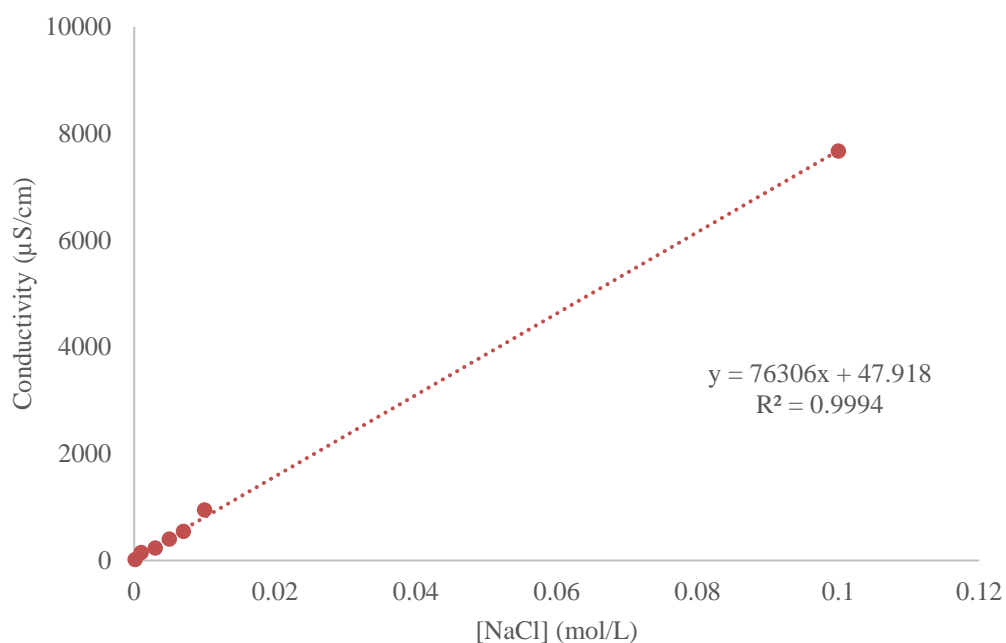


**Figure S5.** Images and particle size distribution (an overall of 250 particles) of **Y/Eu-MOF** crystals in the fraction steadily suspended in water (about a 32 % of the total amount), determined from optical microscope images.



**Figure S6.** Comparison of the particle size distribution of **Y/Eu-MOF** in the fraction steadily suspended in water and the non-suspended, determined from optical microscope images.

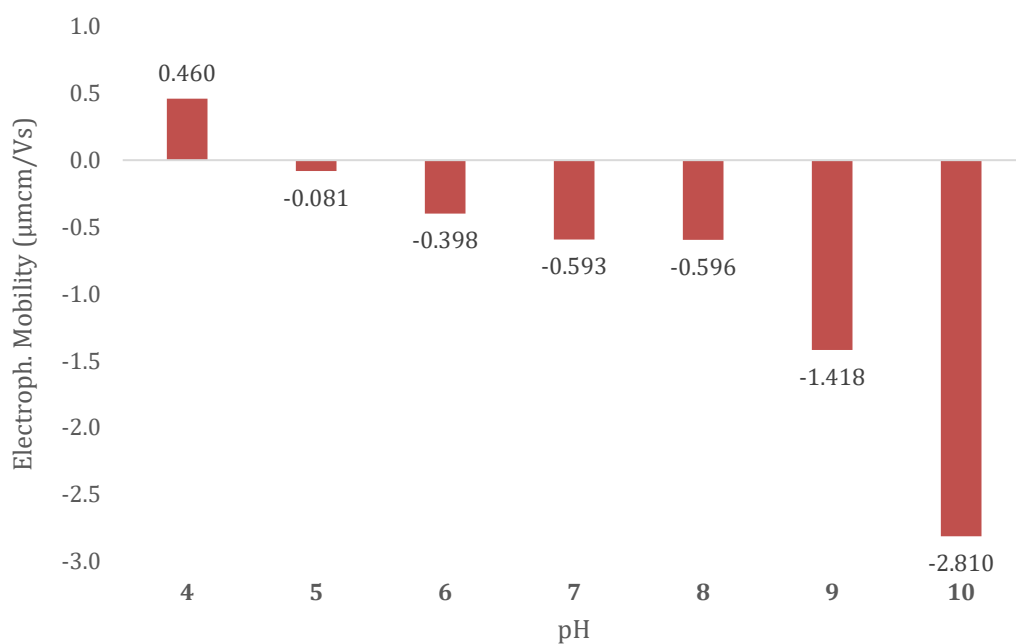
## 11. Study of $\zeta$ -potential at different pH values



**Figure S7.** Calibration line of Conductivity ( $\mu\text{S/cm}$ ) vs [NaCl] (mol/L).



**Figure S8.**  $\zeta$ -potential (mV) dependence with the pH of the **Y/Eu-MOF**. All the measurements were performed with constant conductivity of  $330 \mu\text{S/cm}$ .



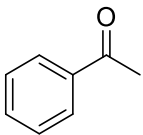
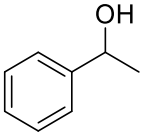
**Figure S9.** Electrophoretic mobility ( $\mu\text{m}\cdot\text{cm}/\text{V}\cdot\text{s}$ ) dependence with the pH of the **Y/Eu-MOF**. All the measurements were performed with constant conductivity of  $330\ \mu\text{S}/\text{cm}$ .

**Table S7.** Electrophoretic mobility and  $\zeta$ -potential dependence with the pH of the **Y/Eu-MOFs** particles dispersed in water. Conductivity fixed at  $330\ \mu\text{S}/\text{cm}$ .

Y/Eu-MOF		
pH	Electroph. Mob. ( $\mu\text{mcm}/\text{Vs}$ )	$\zeta$ -potential (mV)
4	$0.460 \pm 0.032$	$5.9 \pm 0.4$
5	$-0.081 \pm 0.025$	$-1.0 \pm 0.3$
6	$-0.398 \pm 0.041$	$-5.1 \pm 0.5$
7	$-0.593 \pm 0.049$	$6.7 \pm 0.6$
8	$-0.596 \pm 0.056$	$-8.5 \pm 0.4$
9	$-1.418 \pm 0.026$	$18.1 \pm 0.3$
10	$-2.252 \pm 0.035$	$-35.8 \pm 0.5$

## 12. Optimization of the hydroboration reaction conditions.

**Table S8.** Optimization of the reaction conditions in the hydroboration reaction.<sup>a</sup>

<div style="display: flex; align-items: center; justify-content: space-around;"> <div style="text-align: center;">  </div> <div>+</div> <div style="text-align: center;">             HBPIn         </div> <div style="text-align: center;"> <math>\xrightarrow[\text{2) Et}_2\text{O (0.5 mL), NaOH (0.5 mL), RT, 24h}]{\text{1) Y/Eu-MOF (x mol\%), Solvent, N}_2 \text{ atm, RT, 24h}}</math> </div> <div style="text-align: center;">    <b>4a</b> </div> </div>			
Entry	Y/Eu-MOF (x mol%)	Solvent	Conversion (%) <sup>b</sup>
1	0.35	-	71
2	0.35	PhMe	13
3	0.35	THF	25
4	0.35	DCM	9
5	0.35	CH <sub>3</sub> CN	4
6	0.5	-	95
7	0.75	-	95
8	1.1	-	92

<sup>a</sup> Reaction carried out using acetophenone (28  $\mu$ L, 0.25 mmol), HBPIn (40  $\mu$ L, 0.275 mmol) in 0.5 mL of the corresponding solvent at room temperature and under nitrogen inert atmosphere during 24h. <sup>b</sup> Conversions (relative to acetophenone) determined by <sup>1</sup>H NMR of the reaction crude.

### 13. Characterization Data of Products

**2-Phenyl-2-((trimethylsilyl)oxy)acetonitrile (2a).** This product has been previously reported.<sup>7</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.50–7.35 (m, 5H, ArH), 5.50 (s, 1H, CHCN), 0.23 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 136.2 (C<sub>ipso</sub>), 129.3 (ArCH), 128.9 (ArCH), 126.3 (ArCH), 119.1 (CN), 63.6 (CH), -0.29 (TMS) ppm.

**2-(4-Methoxyphenyl)-2-((trimethylsilyl)oxy)acetonitrile (2b).** This product has been previously reported.<sup>8</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.38 (d, *J* = 8.6 Hz, 2H, ArH), 6.92 (d, *J* = 8.6 Hz, 2H, ArH), 5.43 (s, 1H, CHCN), 3.82 (s, 3H, OMe), -0.21 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 160.3 (C<sub>ipso</sub>), 128.3 (C<sub>ipso</sub>), 127.9 (ArCH), 119.3 (CN), 114.2 (ArCH), 63.3 (CH), 55.3 (OCH<sub>3</sub>), -0.24 (TMS) ppm.

**2-(4-Chlorophenyl)-2-((trimethylsilyl)oxy)acetonitrile (2c).** This product has been previously reported.<sup>9</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.45–7.40 (m, 5H, ArH), 5.49 (s, 1H, CHCN), 0.26 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 135.3 (C<sub>ipso</sub>), 134.8 (C<sub>ipso</sub>), 129.1 (ArCH), 127.7 (ArCH), 118.8 (CN), 63.0 (CH), -0.30 (TMS) ppm.

**2-(Pyridin-2-yl)-2-((trimethylsilyl)oxy)acetonitrile (2d).** This product has been previously reported.<sup>10</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 8.60–8.55 (m, 1H, ArH), 7.79 (dt, *J* = 7.7, 1.7 Hz, 1H, ArH), 7.59 (d, *J* = 7.7 Hz, 1H, ArH), 7.35–7.25 (m, 1H, ArH), 5.58 (s, 1H, CHCN), 0.26 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 155.4 (C<sub>ipso</sub>), 149.3 (ArCH), 137.5 (ArCH), 124.0 (ArCH), 120.5 (ArCH), 118.6 (CN), 65.1 (CH), -0.37 (TMS) ppm.

**2-((Trimethylsilyl)oxy)butanenitrile (2e).** This product has been previously reported.<sup>11</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 4.34 (t, *J* = 6.3 Hz, 1H, CH), 1.85–1.75 (m, 2H, CH<sub>2</sub>), 1.04 (t, *J* = 7.4 Hz, 3H, CH<sub>3</sub>), 0.21 (s, 9H, CH<sub>3</sub> x 3) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 119.9 (CN), 62.7 (CH), 29.6 (CH<sub>2</sub>), 8.9 (CH<sub>3</sub>), 0.4 (TMS) ppm.

**2-Phenyl-2-((trimethylsilyl)oxy)propanenitrile (3a).** This product has been previously reported.<sup>12</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.60–7.50 (m, 2H, ArH), 7.45–7.30 (m, 3H, ArH), 1.86 (s, 3H, CH<sub>3</sub>), 0.18 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 142.0 (C<sub>ipso</sub>), 128.68 (ArCH), 128.66 (ArCH), 124.6 (ArCH), 121.6 (CN), 71.6 (C), 33.5 (CH<sub>3</sub>), 1.03 (TMS) ppm.

**2-(4-Methoxyphenyl)-2-((trimethylsilyl)oxy)propanenitrile (3b).** This product has been previously reported.<sup>13</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.46 (d, *J* = 8.7 Hz, 2H, ArH), 6.91 (d, *J* = 8.7 Hz, 2H, ArH), 3.82 (s, 3H, OMe), 1.85 (s, 3H, CH<sub>3</sub>), 0.16 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 159.7 (C<sub>ipso</sub>), 133.9 (C<sub>ipso</sub>), 126.0 (ArCH), 121.7 (CN), 113.8 (ArCH), 71.2 (C), 55.2 (OCH<sub>3</sub>), 33.3 (CH<sub>3</sub>), 1.00 (TMS) ppm.

**2-(4-Chlorophenyl)-2-((trimethylsilyl)oxy)propanenitrile (3c).** This product has been previously reported.<sup>13</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.48 (d, *J* = 8.5 Hz, 2H, ArH), 7.37 (d, *J* = 8.5 Hz, 2H, ArH), 1.83 (s, 3H, CH<sub>3</sub>), 0.19 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48



MHz, CDCl<sub>3</sub>):  $\delta$  140.6 (*C*<sub>ipso</sub>), 134.5 (*C*<sub>ipso</sub>), 128.8 (ArCH), 126.0 (ArCH), 121.2 (CN), 71.0 (C), 33.4 (CH<sub>3</sub>), 1.00 (TMS) ppm.

**2-(2,4-Difluorophenyl)-2-((trimethylsilyl)oxy)propanenitrile (3d).** This product has been previously reported.<sup>15</sup> <sup>1</sup>H NMR (500.13 MHz, CDCl<sub>3</sub>):  $\delta$  7.56 (td, *J* = 8.8, 6.4 Hz, 1H, ArH), 6.95–6.90 (m, 1H, ArH), 6.86 (ddd, *J* = 11.1, 8.8, 2.5 Hz, 1H, ArH), 1.92 (s, 3H, CH<sub>3</sub>), 0.27 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (125.77 MHz, CDCl<sub>3</sub>):  $\delta$  163.2 (dd, *J* = 250.8, 12.0 Hz, *C*<sub>ipso</sub>-F), 159.4 (dd, *J* = 252.5, 12.0 Hz, *C*<sub>ipso</sub>-F), 127.8 (dd, *J* = 9.7, 4.3 Hz, ArCH), 125.0 (dd, *J* = 11.2, 3.9 Hz, *C*<sub>ipso</sub>-F), 120.4 (CN), 111.2 (d, *J* = 21.2 Hz, ArCH), 104.9 (t, *J* = 25.6 Hz, ArCH), 68.0 (d, *J* = 2.0 Hz, C), 30.8 (d, *J* = 2.9 Hz, CH<sub>3</sub>), 1.08 (TMS) ppm.

**2-(Pyridin-2-yl)-2-((trimethylsilyl)oxy)propanenitrile (3e).** This product has been previously reported.<sup>14</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>):  $\delta$  8.62 (d, *J* = 4.7 Hz, 1H, ArH), 7.77 (t, *J* = 7.8 Hz, 1H, ArH), 7.60 (d, *J* = 7.8 Hz, 1H, ArH), 7.30–7.25 (m, 1H, ArH), 1.93 (s, 3H, CH<sub>3</sub>), 0.26 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>):  $\delta$  160.0 (*C*<sub>ipso</sub>), 149.0 (ArCH), 137.2 (ArCH), 123.4 (ArCH), 121.3 (CN), 118.9 (ArCH), 72.9 (C), 31.2 (CH<sub>3</sub>), 1.06 (TMS) ppm.

**2-Methyl-2-((trimethylsilyl)oxy)butanenitrile (3f).** This product has been previously reported.<sup>13</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>):  $\delta$  1.85–1.65 (m, 2H, CH<sub>2</sub>), 1.55 (s, 3H, CH<sub>3</sub>), 1.04 (t, *J* = 7.4 Hz, 3H, CH<sub>3</sub>CH<sub>2</sub>), 0.23 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>):  $\delta$  121.9 (CN), 70.2 (C), 36.4 (CH<sub>2</sub>), 28.4 (CH<sub>3</sub>), 8.6 (CH<sub>3</sub>), 1.21 (TMS) ppm.

**1-((Trimethylsilyl)oxy)cyclohexane-1-carbonitrile (3g).** This product has been previously reported.<sup>13</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>):  $\delta$  2.10–2.00 (m, 2H, CH<sub>2</sub>), 1.75–1.70 (m, 2H, CH<sub>2</sub>), 1.70–1.45 (m, 6H, CH<sub>2</sub> x 3), 1.30–1.20 (m, 2H, CH<sub>2</sub>), 0.23 (s, 9H, TMS) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>):  $\delta$  121.9 (CN), 70.6 (C), 39.3 (CH<sub>2</sub>), 24.5 (CH<sub>2</sub>), 22.6 (CH<sub>2</sub>), 1.38 (TMS) ppm.

**1-Phenylethan-1-ol (4a).** This product has been previously reported.<sup>15</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>):  $\delta$  7.30–7.20 (m, 5H, ArH), 4.82 (q, *J* = 6.5 Hz, 1H, CH), 1.83 (br s, 1H, OH), 1.42 (d, *J* = 6.5 Hz, 3H, CH<sub>3</sub>) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>):  $\delta$  145.8 (*C*<sub>ipso</sub>), 128.5 (ArCH), 127.5 (ArCH), 125.3 (ArCH), 70.4 (CH), 25.1 (CH<sub>3</sub>) ppm.

**1-(4-Isobutylphenyl)ethan-1-ol (4b).** This product has been previously reported.<sup>16</sup> <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  7.27 (d, *J* = 8.0 Hz, 2H, ArH), 7.12 (d, *J* = 8.0 Hz, 2H, ArH), 4.86 (q, *J* = 6.4 Hz, 1H, CH), 2.46 (d, *J* = 7.2 Hz, 2H, CH<sub>2</sub>), 2.12 (br s, 1H, OH), 2.00–1.75 (m, 1H, CH), 1.48 (d, *J* = 6.5 Hz, 3H, CH<sub>3</sub>CHOH), 0.90 (d, *J* = 6.4 Hz, 6H, CH<sub>3</sub> x 2). <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>):  $\delta$  143.0, 141.0, 129.2, 125.2, 70.3, 45.1, 30.2, 25.0, 22.4 ppm.

**1-(4-Methoxyphenyl)ethan-1-ol (4c).** This product has been previously reported.<sup>15</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>):  $\delta$  7.30 (d, *J* = 8.6 Hz, 2H, ArH), 6.88 (d, *J* = 8.6 Hz, 2H, ArH), 7.85 (q, *J* = 6.5 Hz, 1H, CH), 3.8 (s, 3H, MeO), 1.48 (d, *J* = 6.5 Hz, 3H, CH<sub>3</sub>) ppm.

<sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 159.0 (*C<sub>ipso</sub>*), 138.0 (*C<sub>ipso</sub>*), 130.6 (ArCH), 113.6 (ArCH), 70.0 (CH), 55.2 (OMe), 25.0 (CH<sub>3</sub>) ppm.

**1-(4-Chlorophenyl)ethan-1-ol (4d).** This product has been previously reported.<sup>15</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.30–7.25 (m, 5H, ArH), 4.88 (q, *J* = 6.5 Hz, 1H, CH), 1.85 (br s, 1H, OH), 1.47 (d, *J* = 6.5 Hz, 3H, CH<sub>3</sub>) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 144.2 (*C<sub>ipso</sub>*), 133.0 (*C<sub>ipso</sub>*), 128.6 (ArCH), 126.8 (ArCH), 69.7 (CH), 25.2 (CH<sub>3</sub>) ppm.

**1-(3-Chlorophenyl)ethan-1-ol (4e).** This product has been previously reported.<sup>17</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.40–7.35 (m, 1H, ArH), 7.30–7.25 (m, 3H, ArH), 4.91 (q, *J* = 6.4 Hz, 1H, CH), 1.83 (br s, 1H, OH), 1.51 (d, *J* = 6.4 Hz, 3H, CH<sub>3</sub>) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 147.8 (*C<sub>ipso</sub>*), 134.4 (*C<sub>ipso</sub>*), 129.8 (ArCH), 127.6 (ArCH), 125.6 (ArCH), 123.5 (ArCH), 69.8 (CH), 25.3 (CH<sub>3</sub>) ppm.

**1-(2-Chlorophenyl)ethan-1-ol (4f).** This product has been previously reported.<sup>17</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.61 (d, *J* = 7.7 Hz, 1H, ArH), 7.35–7.30 (m, 2H, ArH), 7.25–7.20 (m, 1H, ArH), 5.31 (q, *J* = 6.4 Hz, 1H, CH), 2.2 (br s, 1H, OH), 1.51 (d, *J* = 6.4 Hz, 3H, CH<sub>3</sub>) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 143.0 (*C<sub>ipso</sub>*), 131.6 (*C<sub>ipso</sub>*), 129.4 (ArCH), 128.3 (ArCH), 127.2 (ArCH), 126.4 (ArCH), 66.9 (CH), 23.5 (CH<sub>3</sub>) ppm.

**1-(2,4-Difluorophenyl)ethan-1-ol (4g).** This product has been previously reported.<sup>18</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.55–7.45 (m, 1H, ArH), 6.95–6.85 (m, 1H, ArH), 6.85–6.75 (m, 1H, ArH), 5.25–5.10 (m, 1H, CH), 1.52 (d, *J* = 6.5 Hz, 3H, CH<sub>3</sub>) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 162.1 (dd, *J* = 247.8, 12.2 Hz), 159.6 (dd, *J* = 248.1, 11.9 Hz), 128.6 (dd, *J* = 13.7, 3.8 Hz), 127.5 (dd, *J* = 9.5, 6.5 Hz), 111.2 (dd, *J* = 20.9, 3.5 Hz), 103.6 (t, *J* = 25.4 Hz), 63.9 (d, *J* = 2.5 Hz), 24.0 ppm. <sup>19</sup>F-NMR (CDCl<sub>3</sub>, 282.37 MHz): δ -112.1 (d, *J* = 6.6 Hz), -116.1 (d, *J* = 7.1 Hz) ppm.

**2,2,2-Trifluoro-1-(4-fluorophenyl)ethan-1-ol (4h):** This product has been previously reported.<sup>19</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 7.55–7.45 (m, 2H, ArH), 7.15–7.10 (m, 2H, ArH), 5.15–5.00 (m, 1H, CH), 2.78 (br s, 1H, OH) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 163.4 (d, <sup>1</sup>*J*<sub>C-F</sub> = 248.4 Hz), 129.8 (*C<sub>ipso</sub>*), 129.3 (d, <sup>3</sup>*J*<sub>C-F</sub> = 8.3 Hz), 124.1 (q, <sup>1</sup>*J*<sub>C-F</sub> = 282.2 Hz), 115.6 (d, <sup>2</sup>*J*<sub>C-F</sub> = 21.5 Hz), 72.1 (q, <sup>2</sup>*J*<sub>C-F</sub> = 32.1 Hz) ppm. <sup>19</sup>F-NMR (CDCl<sub>3</sub>, 282.5 MHz): δ -78.6 (d, *J* = 6.1 Hz, 3F), -111.9 (m, 1F) ppm.

**1-(Pyridin-2-yl)ethan-1-ol (4i).** This product has been previously reported.<sup>20</sup> <sup>1</sup>H NMR (300.13 MHz, CDCl<sub>3</sub>): δ 8.55 (d, *J* = 4.8 Hz, 1H, ArH), 7.75–7.65 (m, 1H, ArH), 7.30 (d, *J* = 8.1 Hz, 1H, ArH), 7.30–7.20 (m, 1H, ArH), 4.91 (q, *J* = 6.5 Hz, 1H, CH), 4.35 (br s, 1H, OH), 1.52 (d, *J* = 6.5 Hz, 3H, CH<sub>3</sub>) ppm. <sup>13</sup>C NMR (75.48 MHz, CDCl<sub>3</sub>): δ 163.0 (*C<sub>ipso</sub>*), 148.1 (ArCH), 136.8 (ArCH), 122.2 (ArCH), 119.8 (ArCH), 68.8 (CH), 24.25 (CH<sub>3</sub>) ppm.

**Diphenylmethanol (4j):** This product has been previously reported.<sup>16</sup> <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.45–7.25 (m, 10 H, Ar-H), 5.87 (s, 1 H, CH), 2.52 (s, 1 H, OH) ppm.

$^{13}\text{C}$  NMR (75.48 MHz,  $\text{CDCl}_3$ ):  $\delta$  143.8 ( $\text{C}_{\text{ipso}}$ ), 128.5 (ArCH), 127.6 (ArCH), 126.5 (ArCH), 76.2 (CH) ppm.

**(*E*)-1,3-Diphenylprop-2-en-1-ol (4k):** This product has been previously reported.<sup>21</sup>  $^1\text{H}$  NMR (300.13 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.55–7.20 (m, 10H, ArH), 6.73 (d,  $J$  = 15.9 Hz, 1H,  $\text{CH-Ph}$ ), 6.42 (dd,  $J$  = 15.9, 6.5 Hz, 1H,  $\text{CH=CH-Ph}$ ), 5.42 (d,  $J$  = 6.5 Hz, 1H,  $\text{CH-OH}$ ), 2.19 (br s, 1H, OH) ppm.  $^{13}\text{C}$  NMR (75.48 MHz,  $\text{CDCl}_3$ ):  $\delta$  124.7 ( $\text{C}_{\text{ipso}}$ ), 136.5 ( $\text{C}_{\text{ipso}}$ ), 131.5, 130.5, 128.6 (ArCH), 128.5 (ArCH), 127.76, 127.74 (ArCH), 126.6 (ArCH), 126.3, 75.1 (CH) ppm.

**Butan-2-ol (4m).** This product has been previously reported.<sup>22</sup>  $^1\text{H}$  NMR (300.13 MHz,  $\text{CDCl}_3$ ):  $\delta$  3.75–3.70 (m, 1H, CH), 1.67 (br s, 1H, OH), 1.50–1.45 (m, 2H,  $\text{CH}_2$ ), 1.18 (d,  $J$  = 6.2 Hz, 3H,  $\text{CH}_3\text{-CH}$ ), 0.93 (t,  $J$  = 7.5 Hz, 3H,  $\text{CH}_3\text{-CH}_2$ ) ppm.  $^{13}\text{C}$  NMR (75.48 MHz,  $\text{CDCl}_3$ ):  $\delta$  69.4 (CH), 32.0 ( $\text{CH}_2$ ), 22.8 ( $\text{CH}_3$ ), 9.9 ( $\text{CH}_3$ ) ppm.

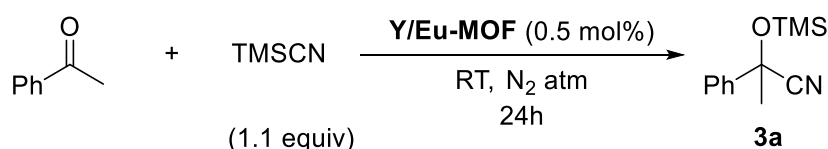
**Cyclohexanol (4n):** This product has been previously reported.<sup>23,24</sup>  $^1\text{H}$  NMR (300.13 MHz,  $\text{CDCl}_3$ ):  $\delta$  3.70–3.55 (m, 1H, CH), 2.05 (br s, 1H, OH), 2.00–1.85 (m, 2H,  $\text{CH}_2$ ), 1.80–1.65 (m, 2H,  $\text{CH}_2$ ), 1.60–1.45 (m, 2H,  $\text{CH}_2$ ), 1.40–1.20 (m, 4H,  $\text{CH}_2 \times 2$ ) ppm.  $^{13}\text{C}$  NMR (75.48 MHz,  $\text{CDCl}_3$ ):  $\delta$  70.3 (CH), 35.5 ( $\text{CH}_2$ ), 25.4 ( $\text{CH}_2$ ), 24.1 ( $\text{CH}_2$ ) ppm.

**1,2,3,4-Tetrahydronaphthalen-1-ol (4o):** This product has been previously reported.<sup>16</sup>  $^1\text{H}$  NMR (300.13 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.50–7.40 (m, 1 H, ArH), 7.30–7.05 (m, 3 H, ArH), 4.85–4.75 (m, 1 H,  $\text{CH-OH}$ ), 2.90–2.70 (m, 2 H,  $\text{CH}_2$ ), 2.10–1.70 (m, 5 H,  $\text{CH}_2 \times 2$ , CH) ppm.  $^{13}\text{C}$  NMR (75.48 MHz,  $\text{CDCl}_3$ ):  $\delta$  138.8 ( $\text{C}_{\text{ipso}}$ ), 137.1 ( $\text{C}_{\text{ipso}}$ ), 129.0 (ArCH), 128.7 (ArCH), 127.6 (ArCH), 126.2 (ArCH), 68.1 (CH), 32.3 ( $\text{CH}_2$ ), 29.2 ( $\text{CH}_2$ ), 18.8 ( $\text{CH}_2$ ) ppm.

**(1*r*,3*r*,5*r*,7*r*)-Adamantan-2-ol (4p):** This product has been previously reported.  $^1\text{H}$  NMR (300.13 MHz,  $\text{CDCl}_3$ ):  $\delta$  3.95–3.85 (m, 1H, CH), 2.50–1.50 (m, 15H) ppm.  $^{13}\text{C}$  NMR (75.48 MHz,  $\text{CDCl}_3$ ):  $\delta$  74.5 (CH), 37.5 ( $\text{CH}_2$ ), 36.2 ( $\text{CH}_2$ ), 34.5 (CH), 31.0 ( $\text{CH}_2$ ), 27.5 (CH), 27.0 (CH) ppm.

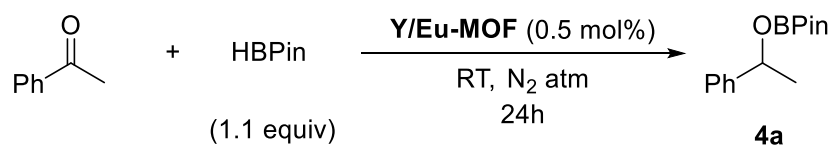
## 14. Catalyst Recyclability

**Recyclability of the catalyst in the cyanosilylation reaction:** In a 1 mL vial with a septum screw capped equipped with a stirring bar, the catalysts **Y/Eu-MOF** (6.4 mg, 0.5 mol%) were weighed. Then, the corresponding amount of acetophenone (56  $\mu$ L, 0.5 mmol) followed by TMSCN (68  $\mu$ L, 0.55 mmol, 1.1 equiv.) were added and the reaction was stirred under inert N<sub>2</sub> atmosphere at room temperature overnight. After this time, 1.5 mL of DCM was added to the reaction mixture and transferred to a centrifuge tube for the separation of the catalyst. The centrifugation of the mixture was carried out at 12300 rpm during 5 min. After that, the solution was eliminated, and the catalyst was washed with DCM (2 x 1.5 mL). Later on, the catalyst was dried under vacuum and reused in the next cycle of the reaction with the same reaction conditions previously described.

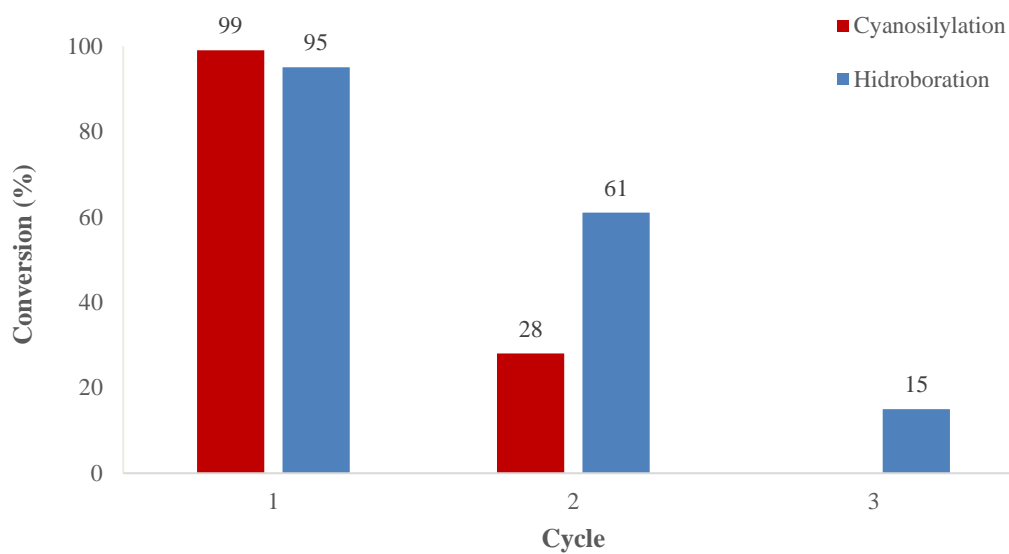


**Scheme S1.** Reaction conditions used for the study of recyclability of **Y/Eu-MOF** catalysts in the cyanosilylation reaction.

**Recyclability of the catalyst in the hydroboration reaction:** In a 1 mL vial with a septum screw capped equipped with a stirring bar, the catalysts **Y/Eu-MOF** (9.2 mg, 0.5 mol%) were weighed. Then, the corresponding amount of acetophenone (84  $\mu$ L, 0.75 mmol) followed by HBPIn (120  $\mu$ L, 0.825 mmol, 1.1 equiv.) were added and the reaction was stirred under inert N<sub>2</sub> atmosphere at room temperature overnight. After this time, 1.5 mL of DCM was added to the reaction mixture and transferred to a centrifuge tube for the separation of the catalyst. The centrifugation of the mixture was carried out at 12300 rpm during 5 min. After that, the solution was eliminated, and the catalyst was washed with DCM (2 x 1.5 mL). Later on, the catalyst was dried under vacuum and reused in the next cycle of the reaction with the same reaction conditions previously described.



**Scheme S2.** Reaction conditions used for the study of recyclability of **Y/Eu-MOF** catalysts in the hydroboration reaction.



**Figure S10.** Study of the recyclability of **Y/Eu-MOF** (0.5 mol%) catalyst on the cyanosilylation and hydroboration reaction of acetophenone as carbonyl substrate.

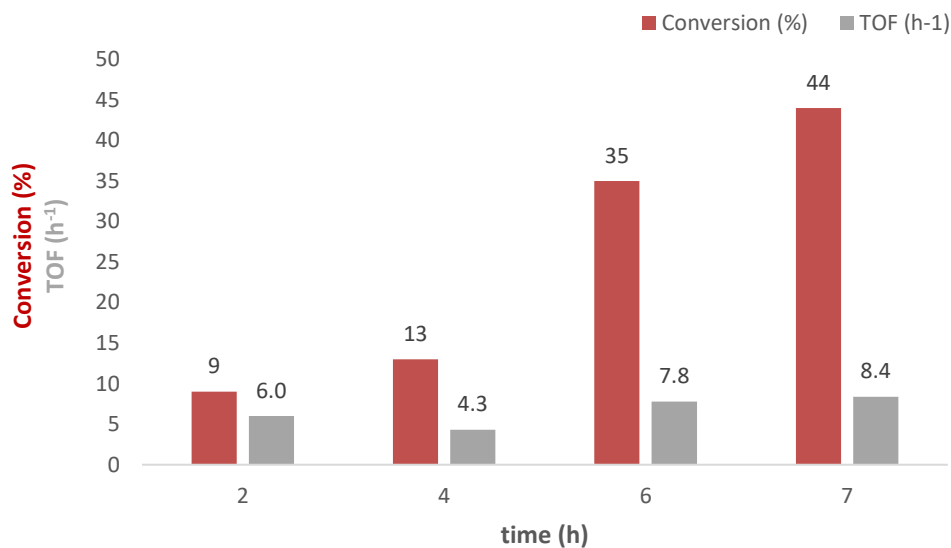
## 15. Leaching test

**Leaching test:** after the first and second reaction of the recyclability test was complete in the hydroboration reaction, the reaction was centrifuged and the supernatant was filtered through a plug of celite and dried under vacuum. Later, 1-(pyridin-2-yl)ethan-1-one (86  $\mu$ L, 0.75 mmol) and HBPIn (120  $\mu$ L, 0.825 mmol, 1.1 equiv.) were added to the crude of the corresponding reaction cycle and the reaction was stirred under inert  $N_2$  atmosphere at room temperature during 24 h. After that time, an aliquot was analysed by  $^1H$  NMR obtaining in 38 % and 13 % of product **4j**, corroborating that the leaching of Y or Eu take place.

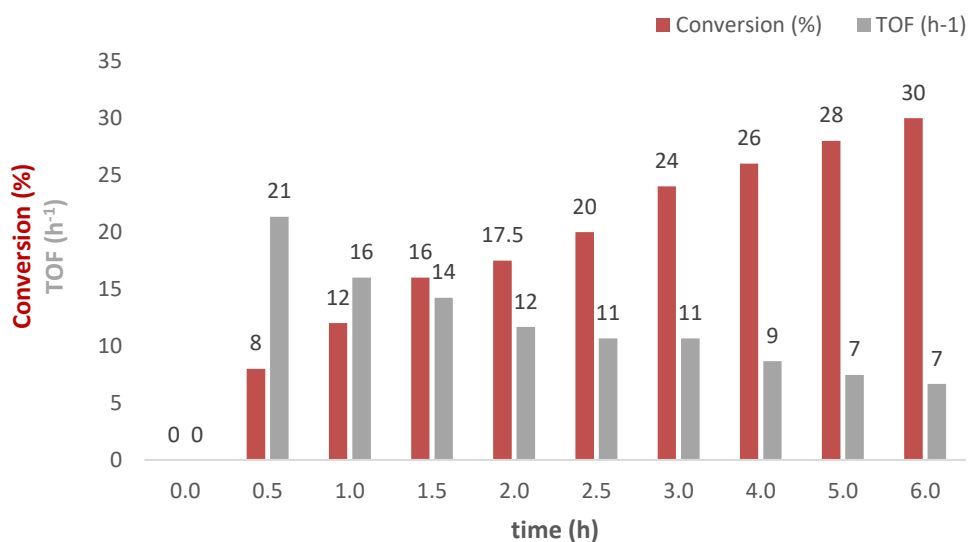


**Scheme S3.** Leaching test carried out after the first and second cycle.

## 16. TOF of Y/Eu-MOF



**Figure S11.** Analysis of the TOF (h<sup>-1</sup>) obtained in the cyanosilylation reaction of acetophenone at different times of reaction with **Y/Eu-MOF** (0.5 mol%) with the optimized reaction conditions.



**Figure S12.** Analysis of the TOF (h<sup>-1</sup>) obtained in the hydroboration reaction of acetophenone at different times of reaction with **Y/Eu-MOF** (0.5 mol%) with the optimized reaction conditions.

## 17. Green chemistry metrics

**Table S9.** Green metrics calculated for **Y/Eu-MOF** catalyst.

<b>Sample</b>	<b>AE (Atom Economy) (%)</b>	<b>MI (Mass Intensity)</b>	<b>RME (Reaction Mass Efficiency)</b>	<b>CE (Carbon Efficiency)</b>
<b>Cyanosilylation</b>	100	1.127	95.7	96.8
<b>Hydroboration</b>	49.1	2.406	46.8	52.1



## 18. Results obtained with different lanthanides MOF catalysts.

**Table S10.** Catalytic cyanosilylation of benzaldehyde performances of Ln-MOFs reported in the literature.

MOF	mol% MOF	mol% RE	Recyclability	Conversion	Conditions	Ratio <sup>[a]</sup>	TOF h <sup>-1</sup>	Ref.
Eu <sub>2</sub> (MELL)(H <sub>2</sub> O) <sub>6</sub> <sup>[b]</sup>	10	20	5 cycles	>99% (3h)	RT, MeCN	1:2	3.2 h <sup>-1</sup>	[7]
[Sm/Dy/Yb(3,5-DSB)(Phen)(H <sub>2</sub> O)]·H <sub>2</sub> O <sup>[c]</sup>	2	2	4 cycles	>99% (3h) (Sm) 70%, 3h (Dy) 50%, 3h (Yb)	40 °C,	1:1.5	6-79 h <sup>-1</sup>	[8–10]
Sm/Eu/Gd/Tb/Eu-Gd/Eu-Tb-psa <sup>[d]</sup>	5	5	3 cycles	70%, <2h (Eu) 80%, <2h (Sm) 87%, <2h (Eu-Gd) 74%, <2h (Tb) 94%, <2h (Eu-Tb)	RT, DCM	1:1.5	84-112 h <sup>-1</sup>	[11]
Nd/Eu/Sm/Ho/Yb/Er-dms <sup>[e]</sup>	5	10	3 cycles	85%, <1h (Nd) 96%, <1h (Eu) 92%, <2h (Sm) 79%, <2h (Ho) 98%, <2h (Yb) 93%, <2h (Er)	RT, DCM	1:1.5	159-234 h <sup>-1</sup>	[12]
Nd, Ho, Er, Yb-btc <sup>[f]</sup>	4.5	4.5	5 cycles	>99%, 2h	RT, DCM	1:2	1-11 h <sup>-1</sup>	[13]
Tm(BDC) <sub>1.5</sub> (DMF)-(H <sub>2</sub> O) <sup>[g]</sup>	2	2	<sup>[h]</sup>	57%, 5h	RT	1:1	<sup>[h]</sup>	[14]
La/Ce/Nd/Sm/Dy(L)(NO <sub>3</sub> )(DMF) <sub>2</sub> ] <sub>n</sub> ·n(DMF) <sup>[i]</sup>	3	3	5 cycles	93%, 2h (La) 94%, 2h (Ce) 91%, 2h (Nd) 89%, 2h (Sm) 90%, 2h (Dy)	RT	1:4	2-16 h <sup>-1</sup>	[15]
Tb-TCA <sup>[j]</sup>	2	2	<sup>[h]</sup>	78%, 4h	RT, DCM <sup>[k]</sup>	1:2.4	9.8 h <sup>-1</sup>	[16]

[Sm(L-H <sub>2</sub> )(R-L-H <sub>3</sub> )(H <sub>2</sub> O) <sub>4</sub> ].nH <sub>2</sub> O <sup>[l]</sup>	10	10	[h]	69%, 16h	RT, DCM	1:2	0.4 h <sup>-1</sup>	[17]
[Yb <sub>2</sub> (L) <sub>2</sub> (H <sub>2</sub> O) <sub>3</sub> ].2H <sub>2</sub> O <sup>[m]</sup>	1.4	2.8	5 cycles	>99%, 24h	RT	1:2	5 h <sup>-1</sup>	[18]
[Pr(L <sup>OMe</sup> )(H <sub>2</sub> O) <sub>4</sub> ].2.5DMA.3 H <sub>2</sub> O <sup>[n]</sup>	1.9	1.9	2 cycles	99%, 14h	RT	1:2	3.8 h <sup>-1</sup>	[19]
[Sm(H <sub>2</sub> O) <sub>5</sub> ][Sm(H <sub>2</sub> O) <sub>7</sub> ][Co <sub>2</sub> Mo <sub>10</sub> H <sub>4</sub> O <sub>38</sub> ].6H <sub>2</sub> O	2	4	3 cycles	98%, 5h	RT	1:3	9.8 h <sup>-1</sup>	[20]
[La/Ce/Nd(H <sub>2</sub> O) <sub>5</sub> ] <sub>2</sub> Mo <sub>6</sub> V <sub>2</sub> O <sub>26</sub> .8H <sub>2</sub> O	1	2	3 cycles	94%, 5h (La) 90%, 5h (Ce) 96%, 5h (Nd)	RT	1:3	10.4 h <sup>-1</sup> 10.0 h <sup>-1</sup> 13.7 h <sup>-1</sup>	[21]
Our <b>Y-MOF</b> catalyst	0.5	2.5	7 cycles	>99%, 5h	RT	1:1	106 h <sup>-1</sup>	[22]
Our <b>GR-MOF-6</b> catalyst	0.5	0.5	7 cycles	>99%, 7.5h	RT	1:1	108 h <sup>-1</sup>	[23]
Our <b>Eu-MOF</b> catalyst	0.5	2.5	7 cycles	>99%, 30 min	RT	1:1	1301 h <sup>-1</sup>	
Our <b>Y/Eu-MOF</b> catalyst	0.35	1.2Y/0.5Eu	2 cycles	>99%, 24h	RT	1:1	8.4 h <sup>-1</sup>	

<sup>[a]</sup> Ratio between benzaldehyde **1a** and TMSCN; <sup>[b]</sup> MELL = mellitic acid; <sup>[c]</sup> 3,5-DSB = 3,5-disulfobenzoate, Phen = 1,10-phenanthroline; <sup>[d]</sup> psa = 2-phenylsuccinate; <sup>[e]</sup> dms = 2,3-dimethylsuccinate; <sup>[f]</sup> btc = 1,3,5-benzenetricarboxylate; <sup>[g]</sup> BDC = 1,4-benzenedicarboxylate; <sup>[h]</sup> Not given; <sup>[i]</sup> L = 5-[2-{2,4,6-trioxotetrahydropyrimidin-5(2*H*)-ylidene}hydrazinyl]isophthalate; <sup>[j]</sup> TCA = tricarboxytriphenylamine; <sup>[k]</sup> The aldehyde employed is 2-nitrobenzaldehyde. <sup>[l]</sup> L-H<sub>4</sub> = 2,2'-diethoxy-1,1'-binaphthalene-6,6'-bisphosphonic acid. <sup>[m]</sup> L = 4,4',4''-((2,4,6-trimethylbenzene-1,3,5-triyl)tris(methylene))tribenzoate; <sup>[n]</sup> L<sup>OMe</sup> = 3,3'-((2,3,6,7-tetramethoxyanthracene-9,10-diyl)bis(4,1-phenylene))diacrylate.

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