

## Supporting Information

### Taming the Transition Between ZIF-L-(Zn, Co) and ZIF-(8, 67) Using a Multi-Inlet Vortex Mixer (MIVM)

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## 1 Experimental Section

The concentration of metal ions was kept constant through all the experiments  $[\text{Zn}^{2+}] = [\text{Co}^{2+}] = 0.05 \text{ M}$ .

### 1.1 Transition to ZIF-L

For the transition as function of Zn:Hmim ratio, solutions with  $[\text{Hmim}] = 0.5 \text{ M}$ ,  $0.75 \text{ M}$ ,  $1$ ,  $1.25 \text{ M}$ ,  $1.5 \text{ M}$ ,  $1.75 \text{ M}$  were prepared. As such the Zn:Hmim ratios were as follow: 1:10, 1:15, 1:20, 1:25, 1:30, and 1:35, respectively. The precursor solutions were fed to four syringes (two for metal ions, and two for Hmim) to be pumped into MIVM. The flow rate of all syringes was fixed at  $10 \text{ mL/min}$ .

As for the transition as function of Zn:Hmim flow rate, the organic linker concentration was fixed at  $[\text{Hmim}] = 0.75 \text{ M}$  for a Metal:Hmim ratio of 1:15. Two syringes were filled with  $\text{Zn}^{2+}$  and the other two with Hmim. The flow rate for the Zn ions was fixed at  $10 \text{ mL/min}$  while that for Hmim was varied as follows:  $10 \text{ mL/min}$ ,  $15 \text{ mL/min}$ ,  $20 \text{ mL/min}$ ,  $25 \text{ mL/min}$  and  $30 \text{ mL/min}$ . The same procedure was followed for ZIF-L-Co to ZIF-67 transition. All solutions were prepared using double distilled water. The collected ZIF samples were aged for 12 hours followed by centrifugation at  $5700 \text{ rpm}$  for  $15 \text{ min}$ . The samples

were washed four times with ethanol (EtOH) and freeze-dried overnight in order to remove all traces of methanol and adsorbed water for characterization.

## 1.2 Two-Syringe System

An aqueous solution of 2-Methylimidazole [Hmim = 3.5 M] was prepared in double distilled water, for a Metal:Hmim ratio of 1:70. For bimetallic ZnCo-ZIF-8 three atomic Zn:Co ratios were used as follows: 75:25, 50:50, and 25:75. Each precursor solution is fed into two separate syringes. The synthesis of all ZIFs is done at three flow rates: 6 mL/min, 20 mL/min, and 59 mL/min. The samples are then centrifuged at 5700 rpm for 20 minutes to remove excess solvent, then washed four times in ethanol, and dried overnight for further characterization.

## 1.3 Four-Syringe System

The four syringes system study was done only for ZIF-8. The same concentrations of the precursor solutions were used ( $[\text{Zn}^{2+}] = 0.05 \text{ M}$  and  $[\text{Hmim}] = 3.5 \text{ M}$ ). However, the total number of used syringes was four where each of the metal and organic linker solutions were fed into two syringes. The synthesis of ZIF-8 was done at the following flow rates: 0.625 mL/min, 10 mL/min, 20 mL/min, 30 mL/min, 40 mL/min, 50 mL/min, and 60 mL/min.

Please note that the two-syringe system experiments in section 3.1 are concerned with the transition from ZIF-L to ZIF-8/67. The experiments that used the four-syringe system in section 3.2 dealt with the control the particle size of the ZIF particles.

## 1.4 Batch Synthesis of ZIF-8

The synthesis of ZIF-8 was done by adding 6 mL of Hmim solution ( $[\text{Hmim}] = 3.5 \text{ M}$ ) to a 50 mL beaker containing 6 mL of  $\text{Zn}^{2+}$  solution ( $[\text{Zn}^{2+}] = 0.05 \text{ M}$ ). The reaction was left to proceed for 30 min at a stirring rate of 500 rpm at room temperature.

## 1.5 Sample Calculation of the Reynolds Number $Re$

The Reynolds number ( $Re$ ) was calculated using Eq. (1) in the main manuscript. The dynamic viscosity ( $\mu$ ) of Hmim at a concentration of 3.5 M and 25 °C is measured and found to be equal to 2.6 cP, which is higher than that of the metal precursor solution (0.89 cP). The measured density ( $\rho$ ) of the Hmim solution is 1003 kg/m<sup>3</sup>, slightly higher than that of the metal solution (997 kg/m<sup>3</sup>). The velocity of the streams is calculated at the inlets; the mixing chamber diameter is  $L = 0.6 \text{ cm}$ . The velocity of the inlet streams was calculated using the following equation:

$$V = \frac{Q}{A}$$

where  $V$  is the flow velocity in (m/s),  $Q$  is the flow rate in (l/min),  $A$  is the cross-sectional circular area of the inlet leading to the main mixing chamber (see Fig. 2 in the main manuscript).

Due to the significant difference in viscosity between the solutions of the organic linker precursor (2.6 cP) and the metal ion precursor (0.891 cP),  $Re$  for each solution was calculated at the inlet, and the reported  $Re$  is the sum of the  $Re$ 's for each stream.

Here is a representative calculation of  $Re$  for a two-syringe system operating at 20 ml/min which corresponds to  $Re = 3170$ :

The velocities of both organic linker and metal ion solutions at the inlet have the same value  $V = Q/A = 20 \text{ cm}^3/\text{min}/0.009503 \text{ cm}^2 = 0.35075 \text{ m/s}$ .

$Re$  for the metal ion solution is then calculated as:

$$Re = 997 \text{ kg/m}^3 \times 0.35075 \text{ m/s} \times 0.006 \text{ m}/0.00089 \text{ kg/m.s} = 2358.$$

$Re$  for the organic linker solution is then calculated as:

$$Re = 1003 \text{ kg/m}^3 \times 0.35075 \text{ m/s} \times 0.006/0.0026 \text{ kg/m.s} = 811.7.$$

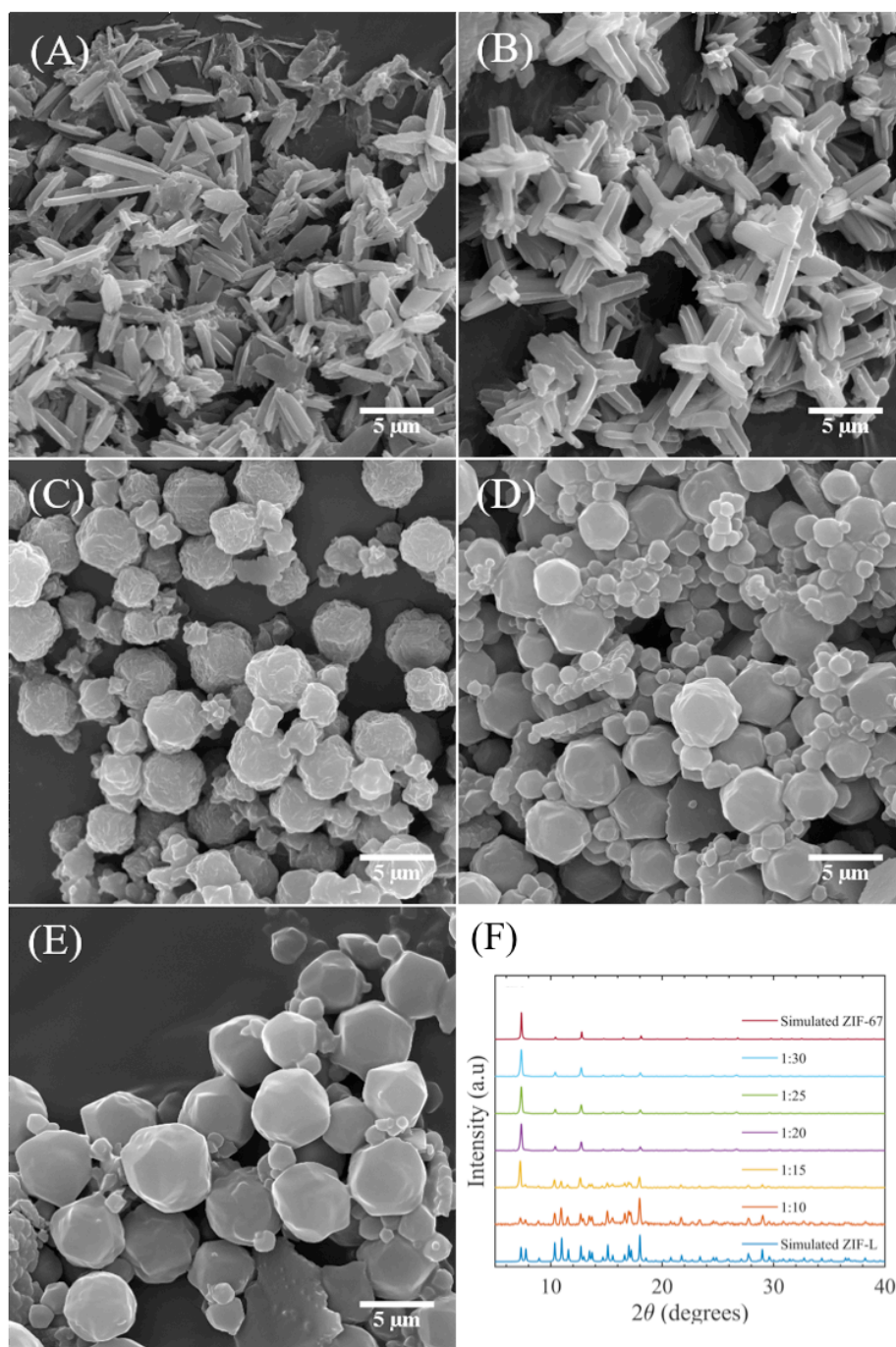
The total reported is then  $Re = 2358 + 811.7 = 3170$ .

Here is a table for the full calculations:

<b>Two-Syringe System</b>			
Flow rate (ml/min)	$Re$ Metal Solution	$Re$ Organic Solution	$Re$ Total
6	707	244	951
20	2358	812	3170
59	6950	2395	9345

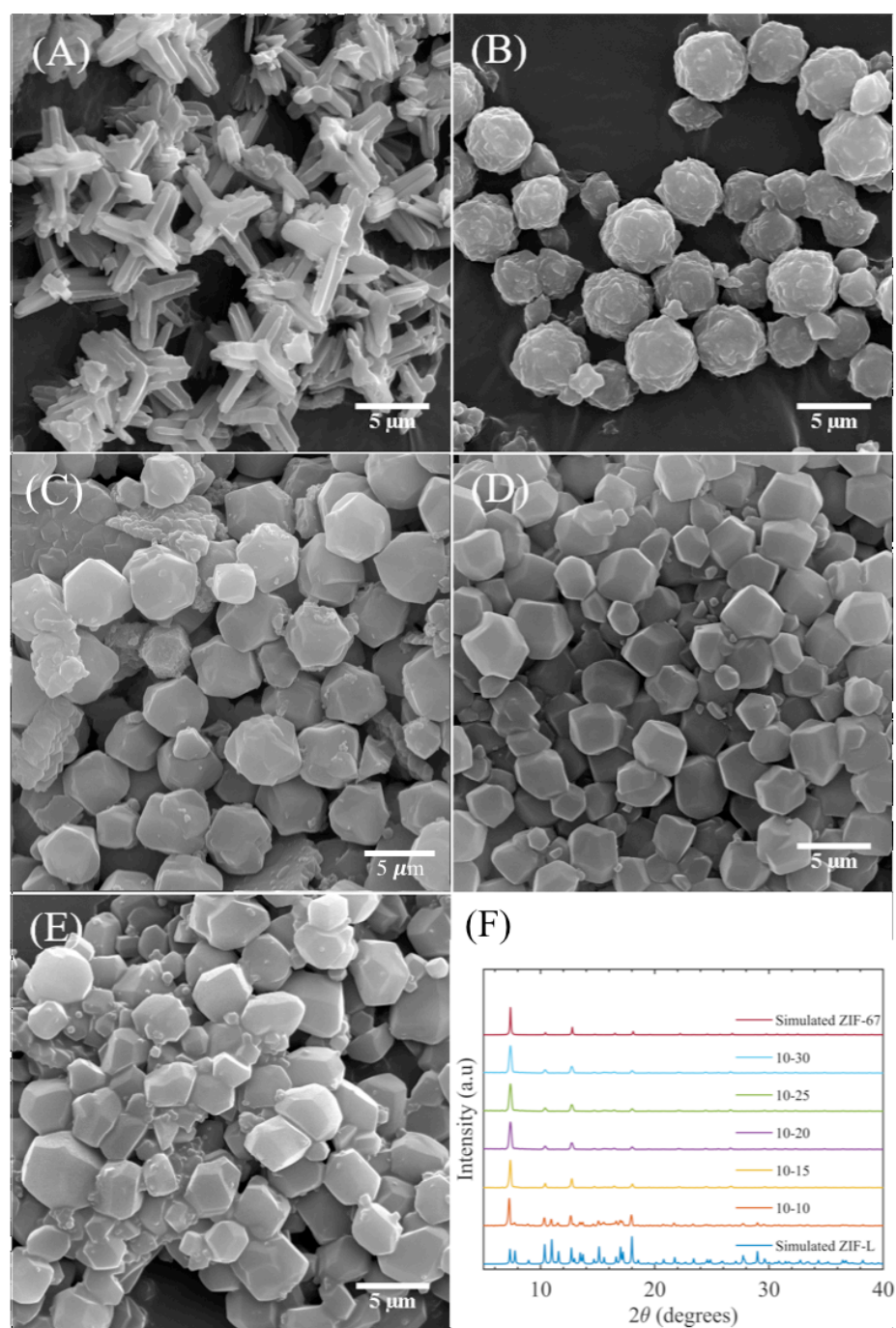
<b>Four-Syringe System</b>			
Flow rate (ml/min)	$Re$ Metal Solution	$Re$ Organic Solution	$Re$ Total
0.625	147.34	50.74	198
10	2353	810	3163
20	4705	1624	6329
30	7057	2430	9487
40	9410	3241	12651
50	11762	4051	15813
60	14142	4870	19012

## 2 X-Ray Diffraction Profiles, Scanning Electron Microscopy, and Particle Size Distributions

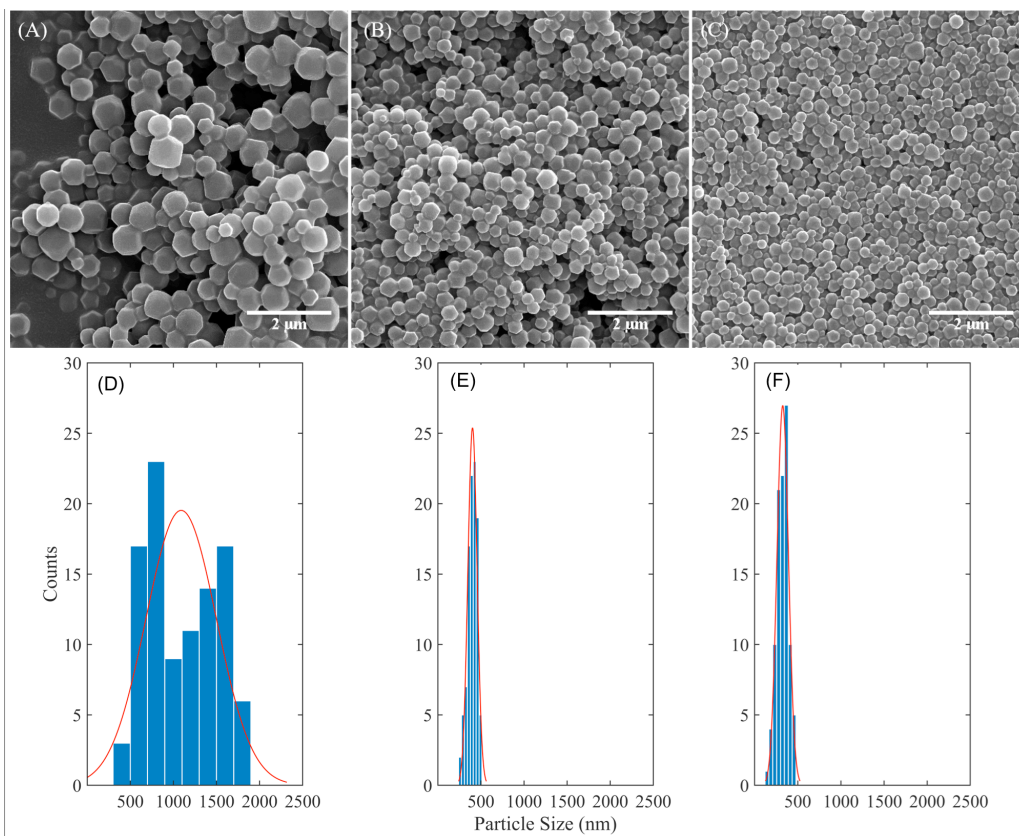


**Fig. S1:** SEM images of ZIF samples at different Co:Hmim ratios: (A) 1:10, (B) 1:15, (C) 1:20, (D) 1:25, and (E) 1:30. And (F) PXRD comparison between as synthesized ZIF samples at different Zn:Hmim ratios and the simulated ZIF-67 and ZIF-L.

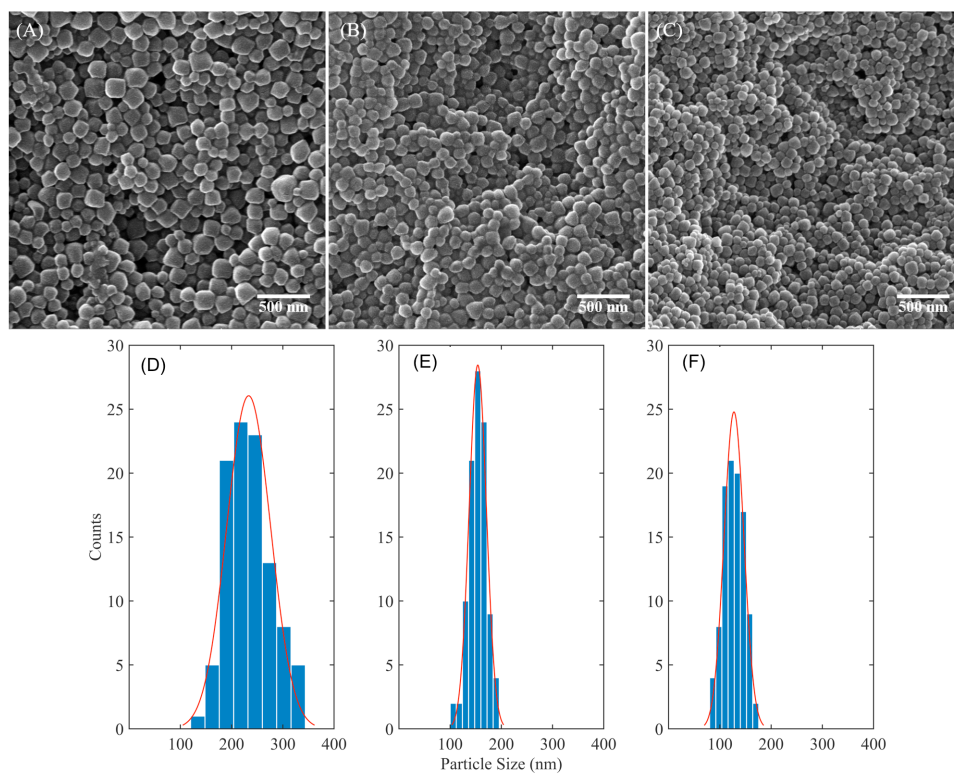




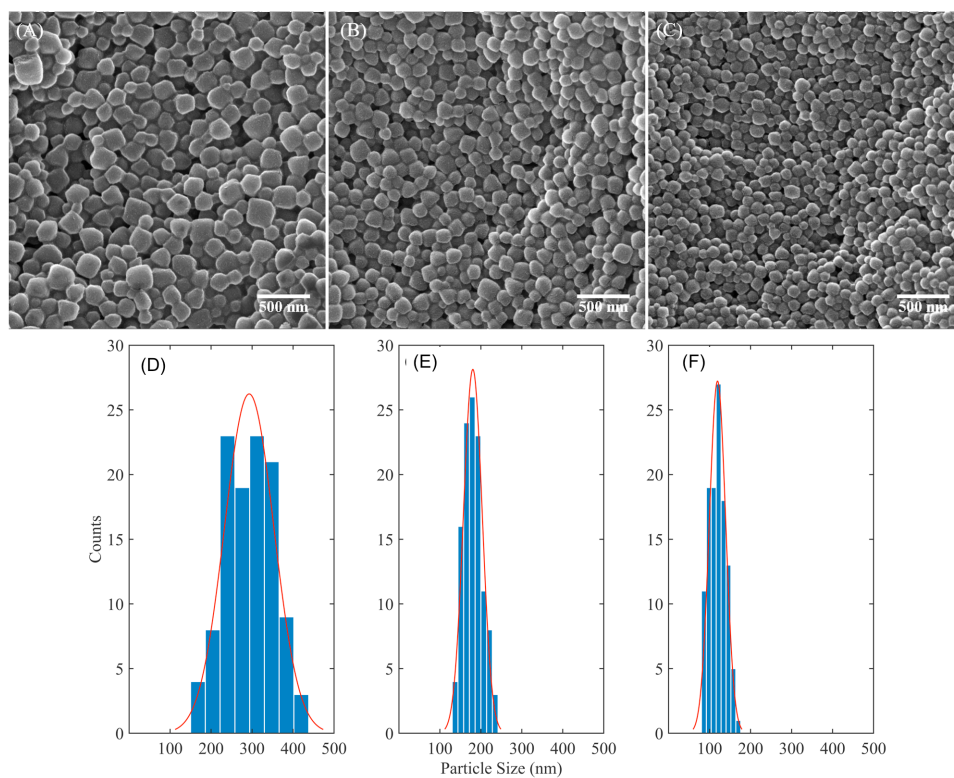
**Fig. S2:** SEM images of ZIF samples at different Co-Hmim flow rates: (A) 10-10 mL/min, (B) 10-15 mL/min, (C) 10-20 mL/min, (D) 10-25 mL/min, and (E) 10-30 mL/min. And (F) PXRD comparison between as synthesized ZIF samples at different Co-Hmim flow rates and the simulated ZIF-67 and ZIF-L.



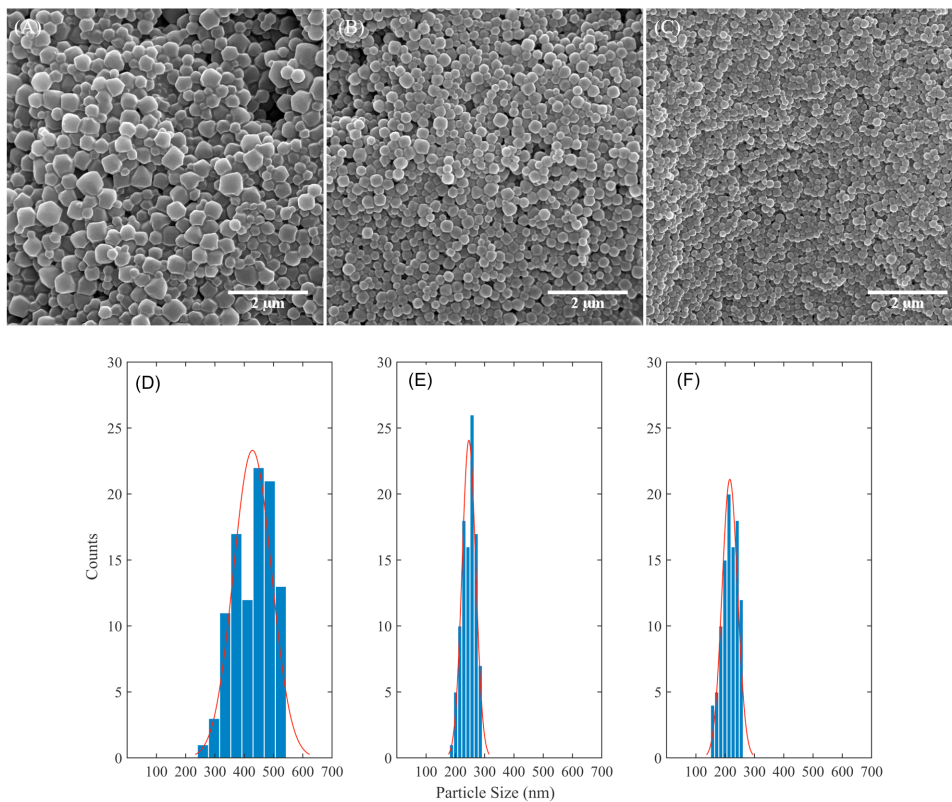
**Fig. S3:** SEM images of as synthesized ZIF-67 at (A)  $Re = 951$ , (B)  $Re = 3170$ , and (C)  $Re = 9345$  showing the decrease in particle size. And the normal distribution of particle size showing a standard deviation of (D) 406 nm at  $Re = 951$ , (E) 53 nm at  $Re = 3170$ , and (F) 65 nm at  $Re = 9345$ .



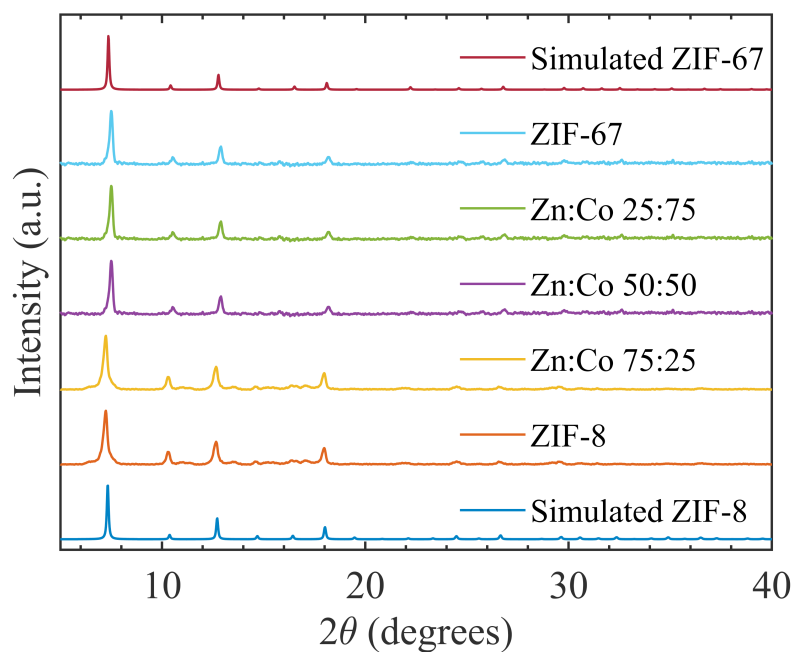
**Fig. S4:** SEM images of as synthesized Zn:Co 75:25 ZIF samples at (A)  $Re = 951$ , (B)  $Re = 3170$ , and (C)  $Re = 9345$ . And the normal distribution of particle size showing a standard deviation of (D) 36 nm, (E) 16 nm, and (F) 18 nm, respectively.



**Fig. S5:** SEM images of as synthesized Zn:Co 50:50 ZIF samples at (A)  $Re = 951$ , (B)  $Re = 3170$ , and (C)  $Re = 9345$ . And the normal distribution of particle size showing a standard deviation of (D) 57 nm, (E) 21 nm, and (F) 18 nm, respectively.



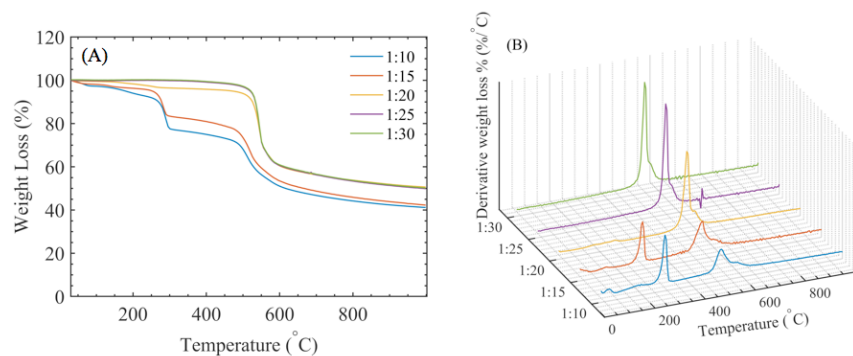
**Fig. S6:** SEM images of as synthesized Zn:Co 25:75 ZIF samples at (A)  $Re = 951$ , (B)  $Re = 3170$ , and (C)  $Re = 9345$ . And the normal distribution of particle size showing a standard deviation of (D) 60 nm, (E) 28 nm, and (F) 28 nm, respectively.



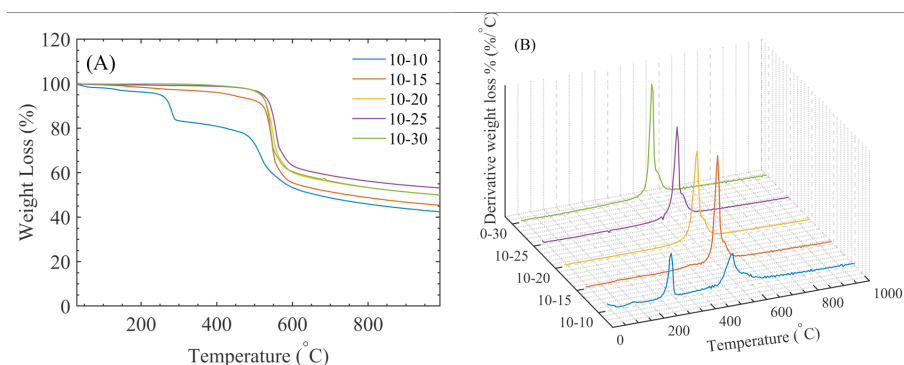
**Fig. S7:** PXRD comparison of the synthesized ZIF samples and the simulated ZIF-8 and ZIF-67, and their mixed metal derivatives at initial Zn:Co ratios of 75:25, 50:50, and 25:75.



### 3 Thermogravimetric Analysis



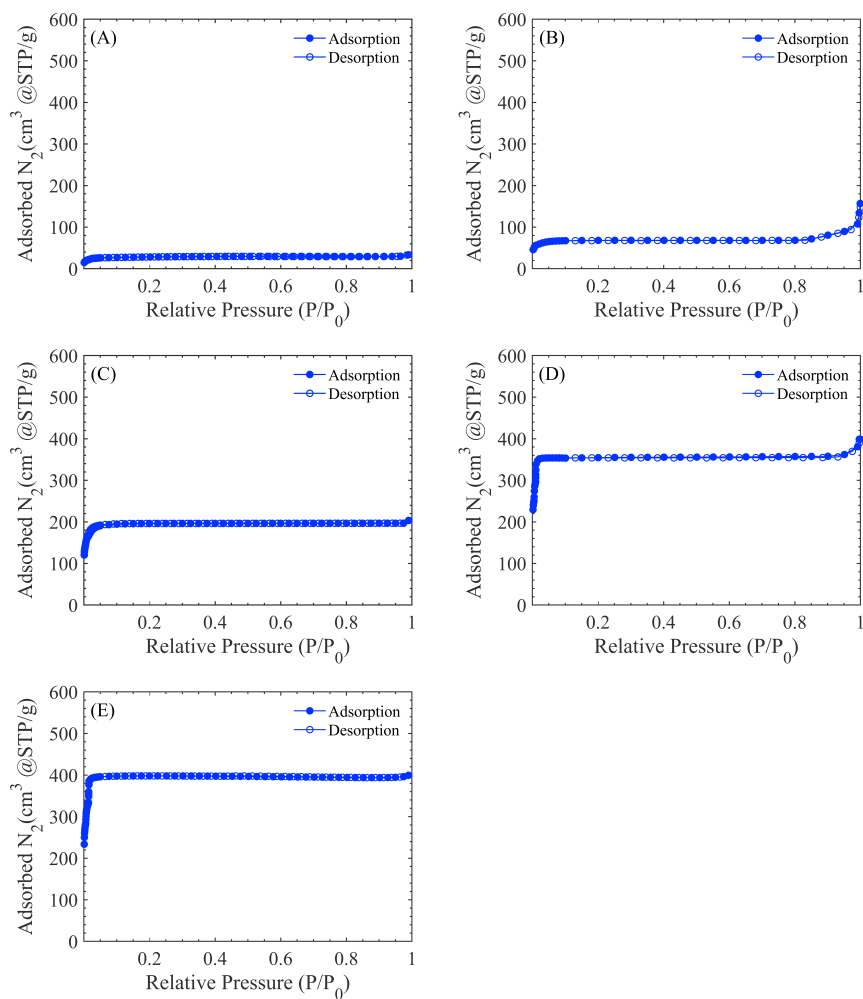
**Fig. S8:** (A) Thermogravimetric curves of ZIF-67 samples at different Co:Hmim ratios, and (B) first derivative of the weight loss as a function of temperature for the thermogravimetric curves of ZIF samples.



**Fig. S9:** (A) Thermogravimetric curves of ZIF samples at different Co-Hmim flow rates (mL/min), and (B) and their first derivatives.



## 4 Nitrogen Adsorption-Desorption Isotherms



**Fig. S10:** Nitrogen adsorption and desorption isotherms at 77 K for the ZIF samples at different Zn-Hmim flow rates: (A) 10-10 mL/min, (B) 10-15 mL/min, (C) 10-25 mL/min, (D) 10-25 mL/min, and (E) 10-30 mL/min.

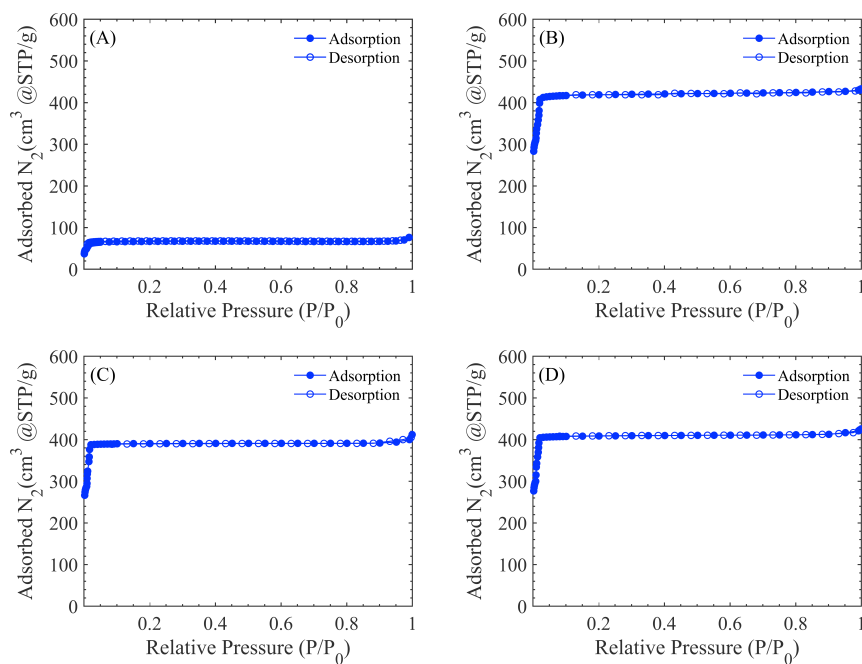
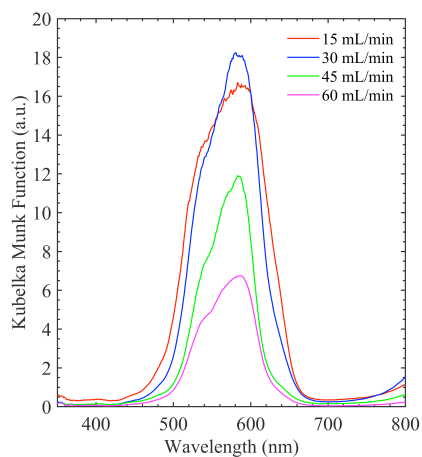
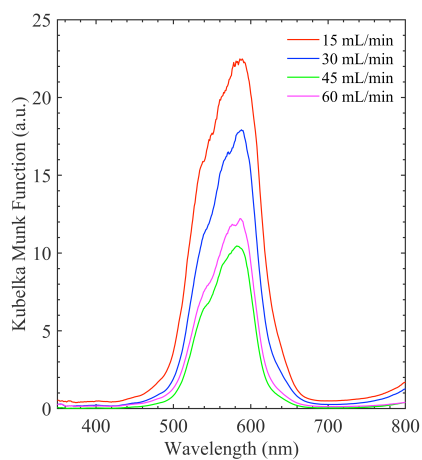


Fig. S11: Nitrogen adsorption and desorption isotherms at 77 K for the ZIF samples at different Co-Hmim flow rates: (A) 10-10 mL/min, (B) 10-15 mL/min, (C) 10-20 mL/min, and (D) 10-25 mL/min.

## 5 Uv-vis Spectra



**Fig. S12:** Visible diffuse reflectance of the ZIF samples as function of the second pump flow rate (mL/min) that contains the ( $\text{Co}^{2+}$ ) solution.



**Fig. S13:** Visible diffuse reflectance of the ZIF samples as function of the second pump flow rate (mL/min) that contains the ( $\text{Zn}^{2+}$ ) solution.

Co-Hmim flow rate (mL/min)	Specific BET surface area (m <sup>2</sup> /g)	Micropore volume (cm <sup>3</sup> /g)
10-10	279	0.097
10-15	1335	0.63
10-20	1242	0.60
10-25	1300	0.62

**Table S1:** Specific BET surface area, micropore volume and micropore area for the as synthesized ZIF-67 samples at different Metal-Hmim flow rates.

Synthesis	Conditions	Particle Size	$S_{BET}$ (m <sup>2</sup> /g)
Solvothermal	+TEA, 120 °C, 24 h	96-985 nm	1340
Microwave	120 °C, 30 min	190-350 nm	1075
Microwave	+F127 surf.,120 °C, 30 min	34-59 nm	1600
Microwave	+P123 surf.,120 °C, 30 min	68-104 nm	1020
Mechanochemical	+NH <sub>4</sub> NO <sub>3</sub> , 45 min, 25 Hz	amorphous	1256
Dry-gel conversion	120 °C, 24 h	agglomerates	1470
Microfluidic	T-mixer, 25 °C	300 nm-900 nm	1700
Microfluidic (ZIF-67)	MIVM, 25 °C	agglomerates	8.49
Microfluidic (This work)	MIVM (2- & 4- syringe), 25 °C	75 nm-1100 nm	1710

**Table S2:** Various synthetic methods to prepare ZIF-8/67 in water.