

Supplementary Information

1.1 Synthesis of ZIF-8

To synthesize ZIF-8, a meticulous two-step process was meticulously followed¹. In the first step, zinc nitrate hexahydrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) was meticulously dissolved in deionized (DI) water. Simultaneously, a separate solution of 2-methylimidazole (Hmim) was prepared by dissolving it in DI water. The preparation of these solutions was conducted with great care to ensure precision in the ensuing reaction. The two solutions, the zinc nitrate solution and the 2-methylimidazole solution, were then mixed together continuously. The Zn^{2+} :Hmim molar ratio was adjusted to 1:70, a crucial step to achieving the desired ZIF-8 structure. As the zinc nitrate solution was quickly poured into the Hmim solution, the mixture underwent an immediate transformation, turning milky. This visual change signified the initiation of the reaction, and it was an important indicator of the successful synthesis process. Following the mixing, the resulting milky mixture was subjected to a stirring duration of 4 hours. This extended period of agitation allowed for the reactants to fully combine and form the desired ZIF-8 precipitate. The meticulous timing ensured that the reaction reached completion, and the resulting precipitate was ready for further processing. To isolate the ZIF-8 precipitate from the solution, a filtration step was meticulously performed. This step served to separate the ZIF-8 crystals from the liquid components of the mixture. After filtration, the ZIF-8 precipitate was left behind. The final step in this intricate process involved oven drying the ZIF-8 precipitate. This drying process was conducted under hot air to remove any residual moisture, ensuring that the ZIF-8 crystals were in their purest form. The result of this methodical synthesis process was a high-quality ZIF-8 material ready for further applications and analysis.

1.2 Synthesis of Na-A from CFA

The synthesis process began with the combination of CFA and NaOH in a weight ratio of 1:1.5. This mixture was then meticulously ground to achieve a uniform powder consistency. Subsequently, the uniform powder was placed into a furnace, where it underwent fusion at a temperature of 550°C for a duration of six hours. This high-temperature treatment was essential for the desired chemical transformation ^{2,3}.

After the fusion process, the mixture was left to cool off at room temperature. Once cooled, it was carefully transferred into a glass reagent bottle. To this mixture, water was added at a specific ratio: for every 15 grams of the fused mixture, 200 mL of water were added. The addition of water-initiated a thorough stirring process, which was maintained at a rate of 400 rpm for a continuous period of 12 hours. This extended stirring time allowed for proper mixing and chemical interactions. Following the stirring, the mixture was subjected to a filtration step to separate the solid residue from the clear supernatant liquid. The clear liquid obtained after filtration was designated as "solution A" and was preserved for subsequent use.

Parallely, NaOH and NaAlO₂ were combined in a weight ratio of 2:1. This mixture was then dissolved in water through high-speed stirring, which continued for a duration of four hours. The resulting solution was labeled as "solution B." To create the final product, 50 mL of solution A were mixed with 20 mL of solution B. The two solutions were thoroughly stirred together for a period of two hours to ensure complete homogenization. Subsequently, the combined mixture was subjected to hydrothermal treatment, which involved maintaining it at a temperature of 105°C for six hours. This hydrothermal process facilitated further chemical reactions and transformations. Upon completion of the hydrothermal treatment, a white residue was left behind. This residue was carefully separated through filtration and then cleansed with deionized (DI) water to remove any impurities. Finally, the cleaned residue was dried in an oven at 120 °C for a duration of 12 hours. This meticulous drying process ensured the final product's stability and readiness for further applications or analysis.

1.3 Synthesis of composite

The composite material was synthesized using three methods for comparison of physical properties, CO₂ capture and ease of synthesis. The three methods are physical binding, in-situ method and simultaneous synthesis, are presented in figure 1.

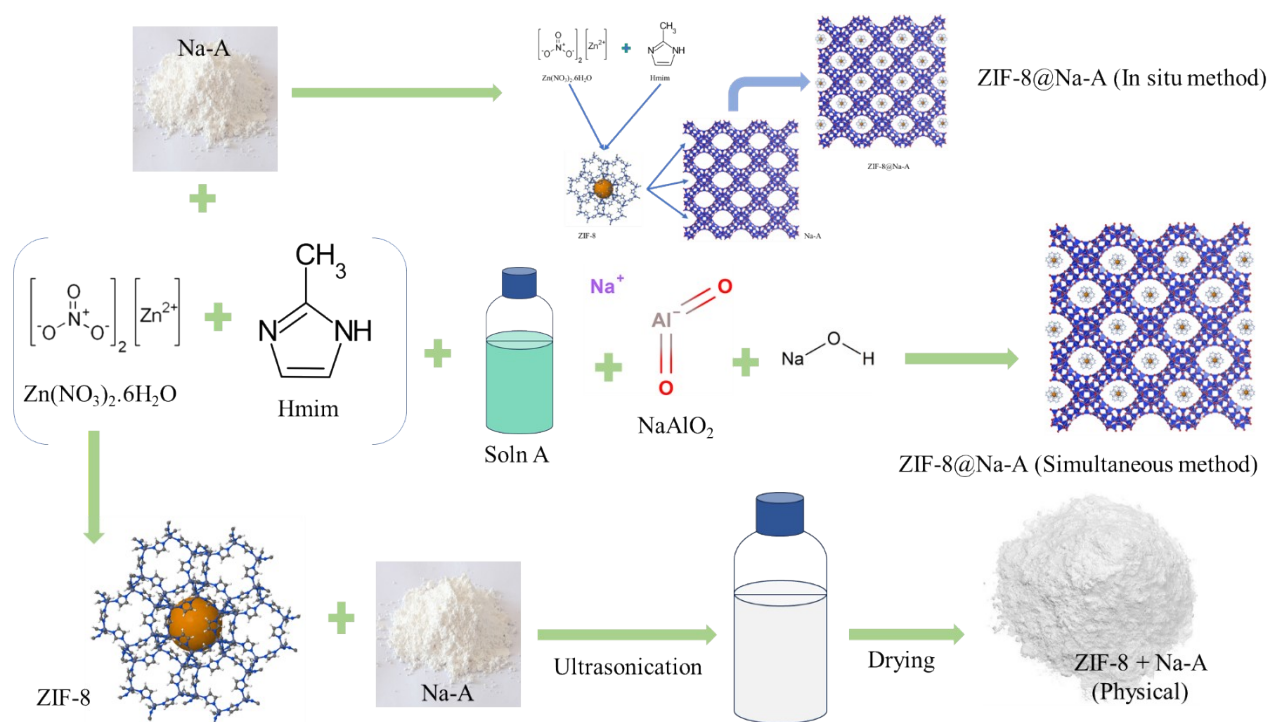


Figure S1: Composite synthesis protocols employed

1.3.1 Synthesis of ZIF-8@Na-A (In situ method)

Calculated weights of Zn (NO₃)₂·6H₂O was mixed in water to form a solution. Simultaneously, calculated weights of Na-A were added to a Hmim solution and stirred for 12 hours. Next, zinc nitrate solution was introduced into the Na-A + Hmim mixture, and the mixture was stirred for a duration of 4 hours. Subsequently, the resulting precipitate was separated by filtration, subjected to a thorough methanol cleaning, and finally, it was dried. The sorbent was designated as ZIF-8@Na-A IM.

1.3.2 Synthesis of ZIF-8@Na-A (Simultaneous method)

In this method, all reactants were added in a predetermined order. In one beaker, soln B was mixed with zinc nitrate solution and stirred for two hours. In another beaker, soln A was mixed with excess Hmim solution and stirred for six hours. Then, both these solutions were mixed with each other quickly and stirred for 12 hours. The resultant precipitate was then filtered, cleaned and dried. The sorbent was designated as ZIF-8@Na-A SM.

1.3.3 Synthesis of ZIF-8@Na-A (Physical mixing)

Both ZIF-8 and Na-A were synthesized independently and were mixed in water. Then, this mixture was treated in a sonicator for uniform dispersion. After sonication for two hours, the mixture was stirred for another two hours, filtered and dried. The sorbent was designated as ZIF-8@Na-A PM.

1.3.4 TEPA loading

In this study TEPA was chosen as the additive to improve the CC uptake of the tested adsorbent. TEPA was loaded onto the pure adsorbent via wetness impregnation method ⁴. Here a calculated amount of TEPA was added to 4 g of the ZIF-8@Na-A 1:5 adsorbent and mixed with 150 mL of water. After properly dispersing the amine throughout the suspension, the water was removed using oven drying at 130 °C. Based on four different weight loadings, the four adsorbents were titled 10%, 20%, 30% and 40% loaded.

Table S1: CO₂ adsorption studies of ZIF-8@Na-A 1:5

Std	Temperature (°C)	Pressure (bar)	Time of Carbonation (min)	CC Uptake (mmol/g)
1	40	1.6	96	5.2
2	85	1.6	96	1.81
3	40	3.4	96	6.63
4	85	3.4	96	3.29
5	40	1.6	200	4.15
6	85	1.6	200	1.83
7	40	3.4	200	7.1
8	85	3.4	200	3.51
9	25	2.5	150	6.3
10	100	2.5	150	2.31
11	62.5	1	150	3.33
12	62.5	4	150	5.8
13	62.5	2.5	60	4.27
14	62.5	2.5	240	4.28
15	62.5	2.5	150	4.32
16	62.5	2.5	150	4.27
17	62.5	2.5	150	4.31
18	62.5	2.5	150	4.56

19	62.5	2.5	150	4.22
20	62.5	2.5	150	4.38

Table S2: Adsorption isotherm modelling fit for Freundlich at 313 K

Adsorbent	Total Pressure (bar)	Freundlich		
		k_F	n	R^2
ZIF-8@Na-A	1	6.48	2.86	0.985

Table S3: Adsorption isotherm modelling fit for Sips and Toth models at 313 K

Adsorbent	Total Pressure (bar)	Sips				Toth			
		k_S	Q_m (mmol/g)	n	R^2	Q_m (mmol/g)	k_T	n	R^2
ZIF-8@Na-A	1	7.51	6.72	1.10	0.999	4.95	7.5638	1.19	0.999

Table S4: Adsorption isotherm modelling fit for Freundlich at 328 K

Adsorbent	Total Pressure (bar)	Freundlich		
		k_F	n	R^2
ZIF-8@Na-A	1	5.56	3.14	0.975

Table S5: Adsorption isotherm modelling fit for Sips and Toth models at 328 K

Adsorbent	Total Pressure (bar)	Sips				Toth			
		k_S	Q_m (mmol/g)	n	R^2	Q_m (mmol/g)	k_T	n	R^2

ZIF-8@Na-A	1	17.12	5.31	1.38	0.999	5.18	14.60	1.65	0.998
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References

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