Chemicals and Reagents

Cobalt (II) nitrate hexahydrate ($Co\ NO_3 \cdot 6H_2O$) and Nickel (II) nitrate hexahydrate ($Ni\ NO_3 \cdot 6H_2O$) was purchased from Sigma-Aldrich (China). 4,4′-Oxybis (benzoic acid) **(**98 %) by Aladdin. The deionized water used in our experiments was obtained from the Milli-Q System. Other analytical grade solvents including DMF, Ethanol, Methanol were supplied by Beijing Chemical Reagent Company (China). All the chemicals were used without further purification.

General Characterization

Scanning electron microscopy (SEM) measurement was performed on a Hitachi SU8200 scanning electron microscope at 6.0 kV. Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) imaging and High angle annular dark field scanning transmission electron microscopy (HAADF-STEM) imaging were carried out using Tecnai G_2 F20 S-TWIN at 200 kV. The thickness of ultrathin sheets was measured by atomic force microscopy (AFM) by using a Bruker multimode-8 scanning probe microscope. Powder X-ray diffraction (XRD) patterns were recorded on D/MAX-TTRIII (CBO) with Cu Kα radiation (λ = 1.542 Å) operating at 40 kV and 300 mA. The specific surface area, pore volume and pore size of catalysts were measured using a BET (Brunauer-Emmett-Teller) instrument (ASAP 2420-4, Micromeritics).

Synthesis of $Co(Hoba)_2 \cdot 2H_2O$ at different temperature.

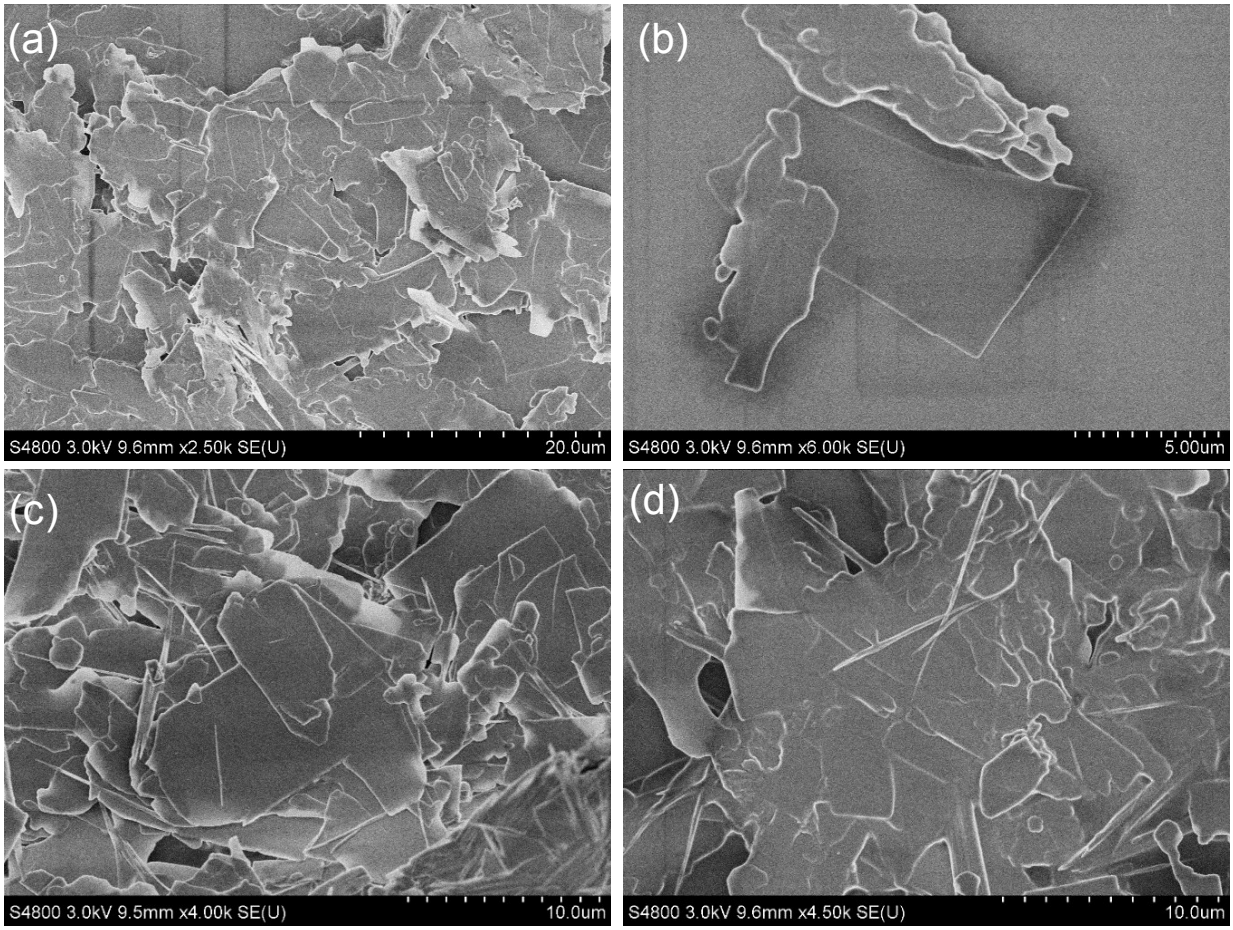


Fig. S1: SEM image shows $Co(Hoba)_2 \cdot 2H_2O$ at different temperature. (a) 120 ℃, (b) 150 ℃, (c) 160 ℃, (d) 180 ℃.

$Co(NO_3)_2 \cdot 6H_2O$ (0.4 mmol) and H_2oba (0.8 mmol) were mixed in 5 mL of distilled water, 20µl of TEA were added into the solution to adjust the pH value to ~7.00. Then the solution was kept in a 20 mL Teflon-lined stainless-steel autoclave heated at 120 or 150 or 180℃ for 12h, followed by cooling to room temperature. The products were centrifuged and washed with ethanol several times and finally dried at room temperature to collect the pink colored MOF with a 60% yield. From this different temperature screening analysis, we have found 120 ℃ is better for this MOF synthesis.

Synthesis of $Co(Hoba)_2 \cdot 2H_2O$ at different Time variation.

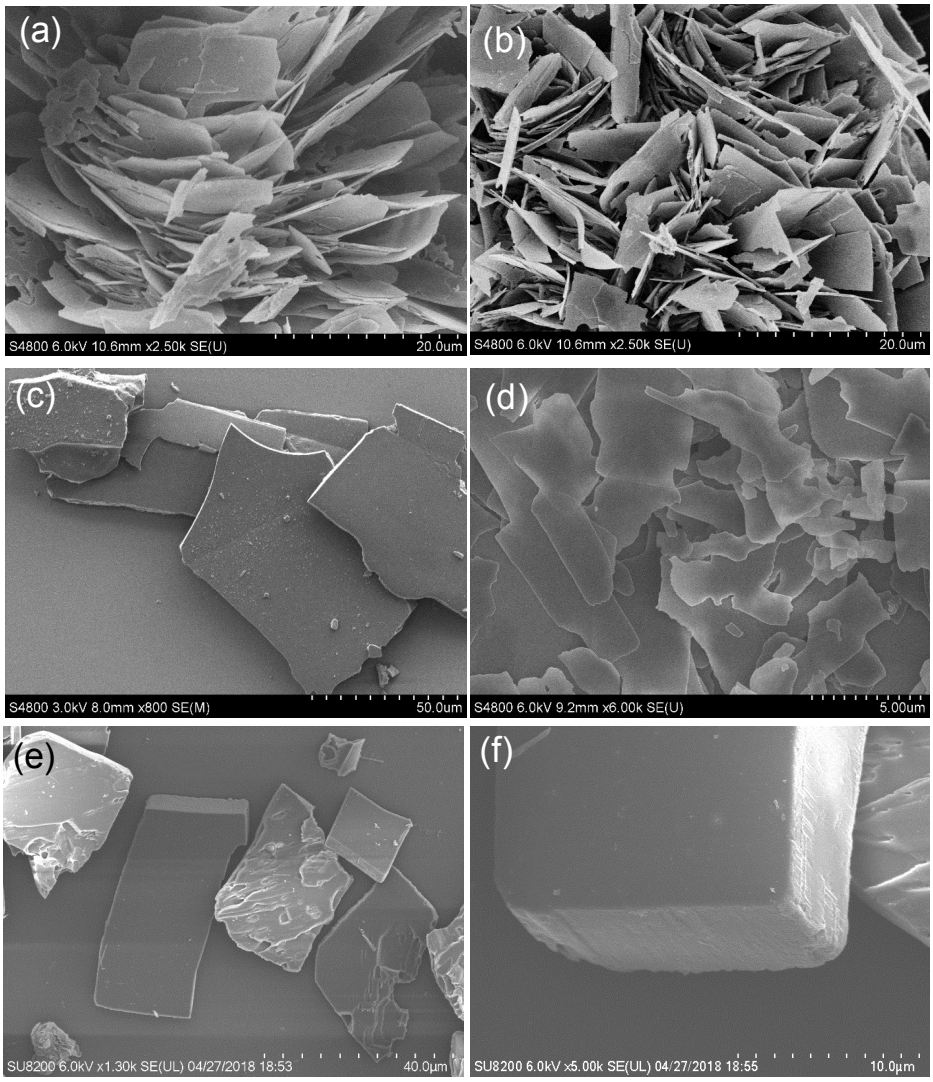


Fig. S2: SEM image shows $Co(Hoba)_2 \cdot 2H_2O$ at different time variation. (a) 6h, (b) 12h, (c) 24h, (d) 48h, (e) 62h, (f) 72h.

$Co(NO_3)_2 \cdot 6H_2O$ (0.4 mmol) and H_2oba (0.8 mmol) were mixed in 5 mL of distilled water, 20µl of TEA were added into the solution to adjust the pH value to ~7.00. Then the solution was kept in a 20 mL Teflon-lined stainless-steel autoclave heated at 120℃ for 6, 12, 24, 48, 62, 72h, respectively, followed by cooling to room temperature. The products were centrifuged and washed with ethanol several times and finally dried at room temperature to collect the pink colored MOF with a 60% yield. From this different time screening analysis, we have found 6h to 12h is the ideal time for this MOF nanosheet synthesis.

Synthesis of $Co(Hoba)_2 \cdot 2H_2O$ at various solvents.

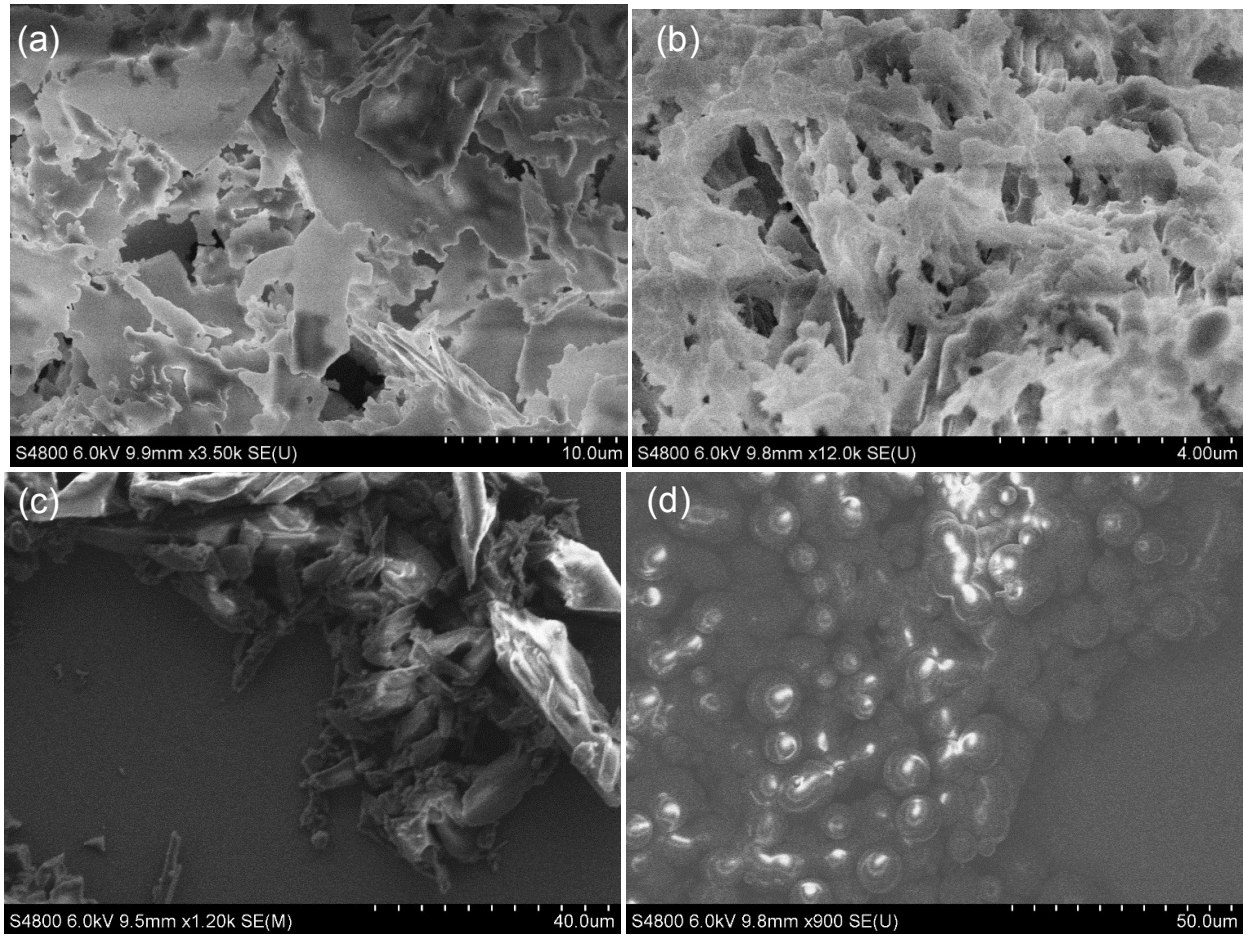


Fig. S3: SEM image shows $Co(Hoba)_2 \cdot 2H_2O$ at various solvents. (a) Ethanol, (b) Methanol, (c) DMF/H_2O, (d) DMF.

$Co(NO_3)_2 \cdot 6H_2O$ (0.4 mmol) and H_2oba (0.8 mmol) were mixed in 5 mL of distilled water, or water/DMF or Ethanol or Methanol as a solvent. 20µl of TEA were added into the solution to adjust the pH value to ~7.00. Then the solution was kept in a 20 mL Teflon-lined stainless-steel autoclave heated at 120℃ for 12h, followed by cooling to room temperature. The products were centrifuged and washed with ethanol several times and finally dried at room temperature to collect the pink colored MOF with a 60% yield. From this different solvent screening analysis, we have found that water is an ideal solvent to produce nanosheet for this MOF synthesis.

Synthesis of $Co(Hoba)_2 \cdot 2H_2O$ without base (TEA).

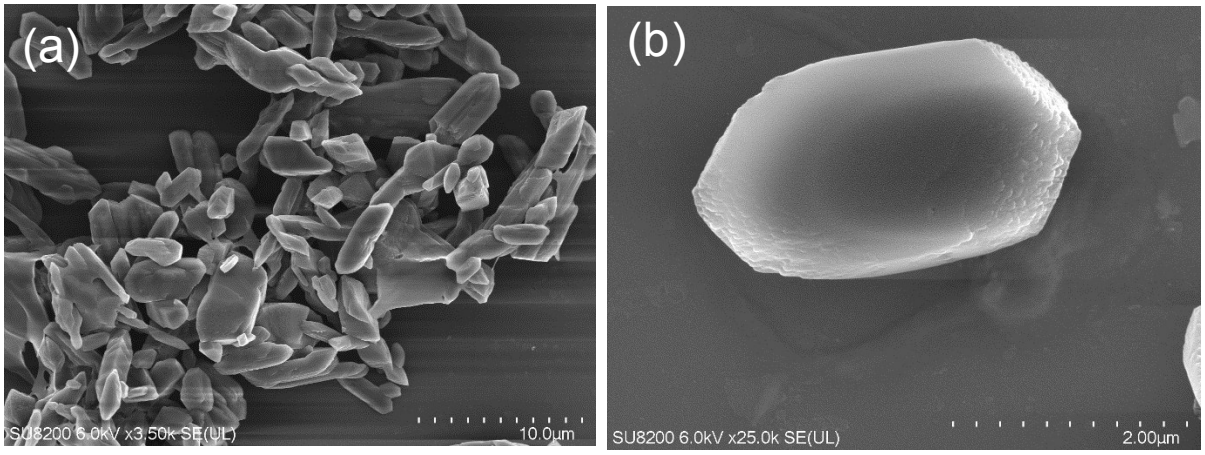


Fig. S4: SEM image (a, b) shows $Co(Hoba)_2 \cdot 2H_2O$ at without base.

$Co(NO_3)_2 \cdot 6H_2O$ (0.4 mmol) and H_2oba (0.8 mmol) were mixed in 5 mL of distilled water, as a solvent .without base Triethylamine. Then the solution was kept in a 20 mL Teflon-lined stainless-steel autoclave heated at 120℃ for 12h, followed by cooling to room temperature. The products were centrifuged and washed with ethanol several times and finally dried at room temperature to collect the white colored reactant only. From this screening analysis, we have found that the base is playing a vital role in the synthesis of MOF formation.

Synthesis of Ni $(Hoba)_2 \cdot 2H_2O$ without base (TEA).

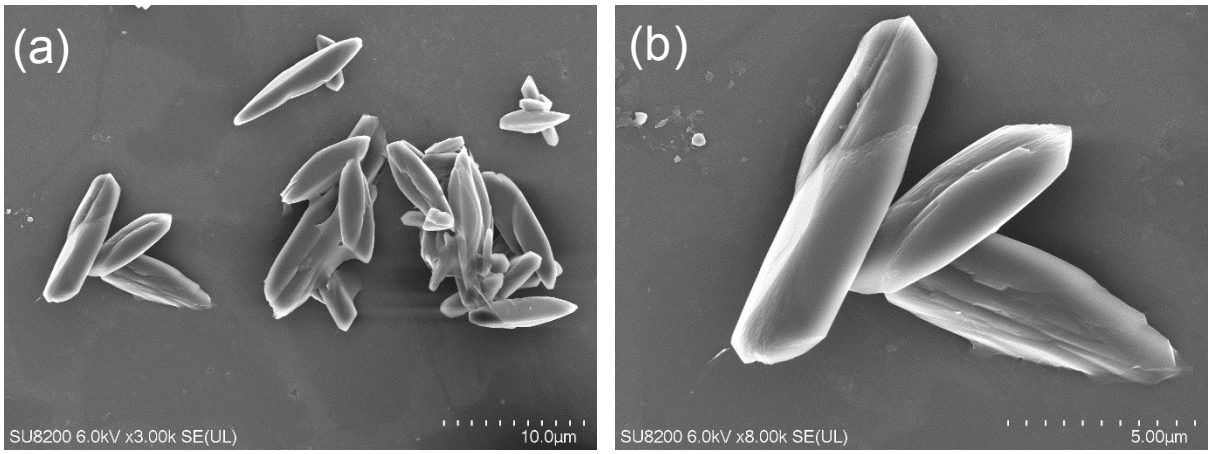


Fig. S5: SEM image (a, b) shows $Ni(Hoba)_2 \cdot 2H_2O$ at without base.

$Ni(NO_3)_2 \cdot 6H_2O$ (0.4 mmol) and H_2oba (0.8 mmol) were mixed in 5 mL of distilled water, as a solvent .without base Triethylamine. Then the solution was kept in a 20 mL Teflon-lined stainless-

steel autoclave heated at 120℃ for 12h, followed by cooling to room temperature. The products were centrifuged and washed with ethanol several times and finally dried at room temperature to collect the white colored reactant only. From this screening analysis, we have found that the base is playing a vital role in the synthesis of MOF formation.

Synthesis of Ni $(Hoba)_2 \cdot 2H_2O$ at different Mole ratio of Precursor's

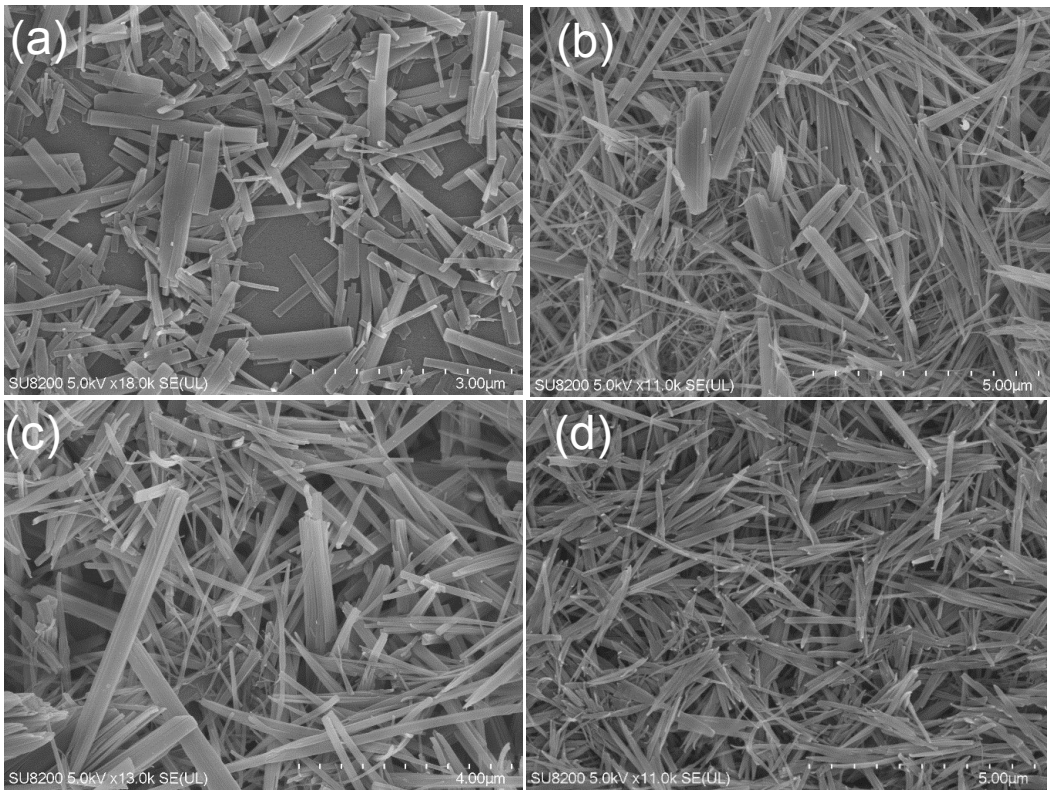


Fig. S6: SEM image shows $Ni(Hoba)_2 \cdot 2H_2O$ at a different Mole ratio of precursors. (a) 1:1, (b) 1:2, (c) 1:3, (d) 1:4 mole ratio.

$Ni(NO_3)_2 \cdot 6H_2O$ and H_2oba with different ratio of 1:1 or 1:2 or 1:3 or 1:4 respectively, were mixed in 5 mL of distilled water, 20µl of TEA were added into the solution to adjust the pH value to ~7.00. The solution was kept in a 20 mL Teflon-lined stainless-steel autoclave heated at 120 ℃ for 12h, followed by cooling to room temperature. Then washed with ethanol several times and collected the light green colored MOF with a 56 % yield.

Synthesis of Ni $(Hoba)_2 \cdot 2H_2O$ at the variation of Time.

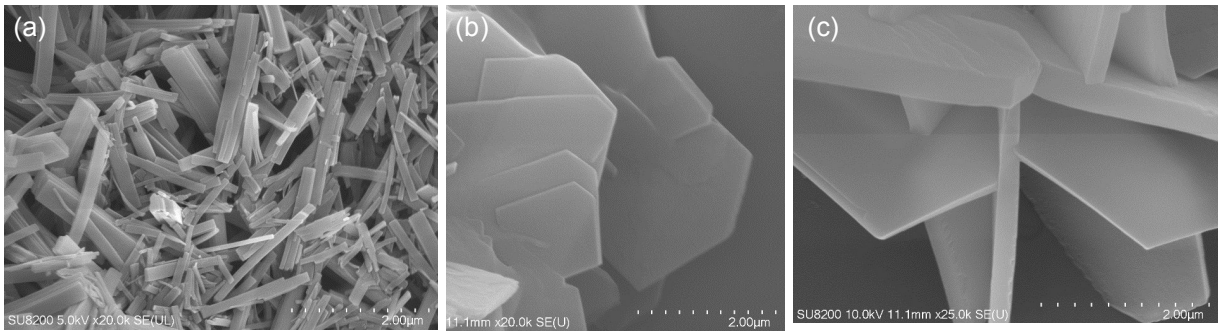
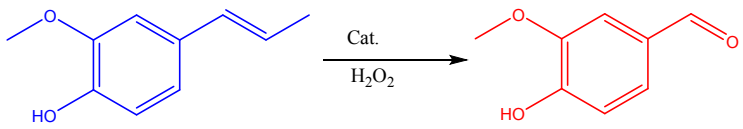


Fig. S7: SEM image shows $Ni(Hoba)_2 \cdot 2H_2O$ at different time variation. (a) 24h, (b) 36h, (c) 48h.

$Ni(NO_3)_2 \cdot 6H_2O$ (0.4 mmol) and H_2oba (0.8 mmol) were mixed in 5 mL of distilled water, 20µl of TEA were added into the solution to adjust the pH value to ~7.00. Then the solution was kept in a 20 mL Teflon-lined stainless-steel autoclave heated at 120℃ for 24, 36, 48h, respectively, followed by cooling to room temperature. The products were centrifuged and washed with ethanol several times and finally dried at room temperature the light green colored MOF with 56 % yield. From this different time screening analysis, we have found 12 to 24h is the ideal time for this MOF nanosheet synthesis.

Experimental Results



Synthesis of Vanillin by various oxidants

Table 1: The synthesis of vanillin using various oxidant.

No	Oxidant	conversion **(%)**
1	Air	NR
2	H_2O_2	23
3	TBHP	NR

Reaction condition: 0.5ml Isoeugenol, cat: Co/ Ni – Hoba, 20mg, oxidant 2ml, rt, 24h.

Synthesis of Vanillin by various solvents

Table 2: The synthesis of vanillin using various solvents.

No	Solvent	Conversion **(%)**	Selectivity **(%)**
1	Neat	1% ≤	98
2	Acetonitrile	23 %	92
3	Methanol	17%	88
4	DMF	18 %	87

Reaction condition: 0.5ml Isoeugenol, cat: Ni – Hoba, 20mg, H_2O_2 - 2ml, rt, 24h.