Controllable and efficient synthesis of two-dimensional metal-organic framework nanosheets for heterogeneous catalysis

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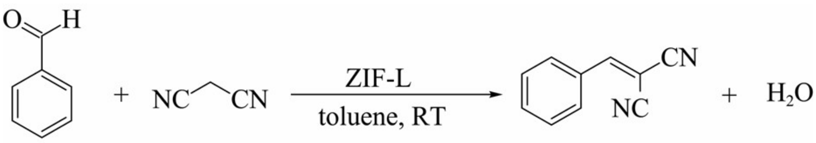
1. Catalytic Knoevenagel condensation

The conversion of Knoevenagel condensation reaction of benzaldehyde with malononitrile using the ZIF-L catalyst is based on Equation S1 and calculated by Equation S2.[1]



(S1)

$$\text{Conversion of benzaldehyde} = \frac{n\ \text{(initial benzaldehyde)} - n\ \text{(residual benzaldehyde)}}{n\ \text{(initial benzaldehyde)}} \times 100\% \tag{S2}$$

2. Catalytic glycolysis of PET

The conversion of glycolysis reaction of PET and the yield of monomer BHET are calculated by Equation S3 and S4, respectively.[2]

$$\text{Conversion of PET} = \frac{m\ \text{(initial PET)} - m\ \text{(residual PET)}}{m\ \text{(initial PET)}} \times 100\% \tag{S3}$$

$$\text{Yield of BHET} = \frac{\text{conversion of PET(\%)} \times \text{selectivity of BHET(\%)}}{100\%} \tag{S4}$$

The pseudo-first-order kinetics are calculated as follow (Equation S5-S7), where C_t and C_0 refer to solid contents of PET at different times; X refers to PET conversion; k is the apparent rate constant; E_a refers to the apparent activation energy. A, R, and T refer to the pre-exponential factor, gas constant ($8.314\ J \cdot k^{-1} \cdot mol^{-1}$), and reaction temperature in Kelvin, respectively.[3]

$$\ln(C_t / C_0) = \ln(1 - X) \tag{S5}$$

$$\ln(1 - X) = -kt \tag{S6}$$

$$\ln k = -\frac{E_a}{RT} + \ln A \tag{S7}$$

3. Preparation of 2D MOFs using RPB

High-gravity level (HGL) is represented by Equation S7, where ω is the angular speed of the RPB, r is the geometric average radius of the packing, and g is the acceleration of gravity (10 m/s^2).[4]

$$HGL = \frac{w^2 r}{g} \tag{S8}$$

Table S1. Comparison of several reported catalysts for Knoevenagel reaction.

catalyst	reaction condition	catalyst amount	benzaldehyde conversion (%)	reference
ZIF-L-RPB nanosheets	**25 °C, 2 h**	**16 mg**	**99.3**	**This work**
HP-ZIF-8	25 °C, 3 h	20 mg	100	1
ZIF-67	40 °C, 8h	25 mg	87	5
hydrotalcite	40 °C, 8 h	25 mg	75	5
ZIF-8	30 °C, 6 h	100 mg	76.5	6
DES functionalized MIL-101	70 °C, 1 h	5 wt %	100	7
SOM-ZIF-8	25 °C, 2 h	6.6 mg	100	8

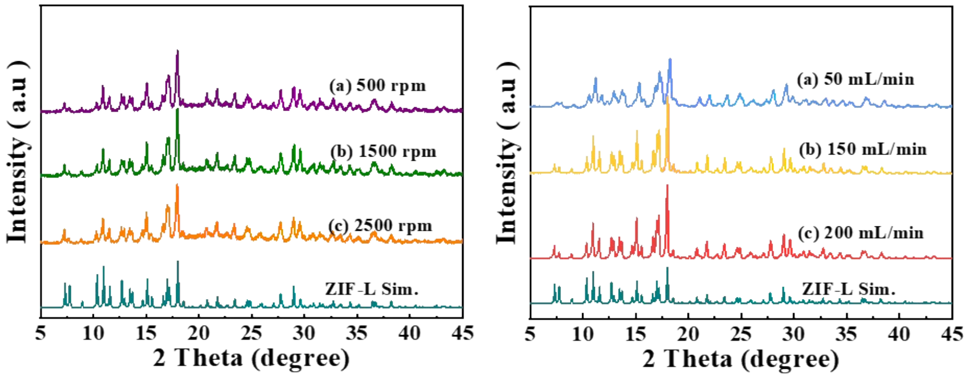


Figure S1. XRD patterns of ZIF-L nanosheets obtained at different (a) RPB rotating speeds and (b) feed rates.

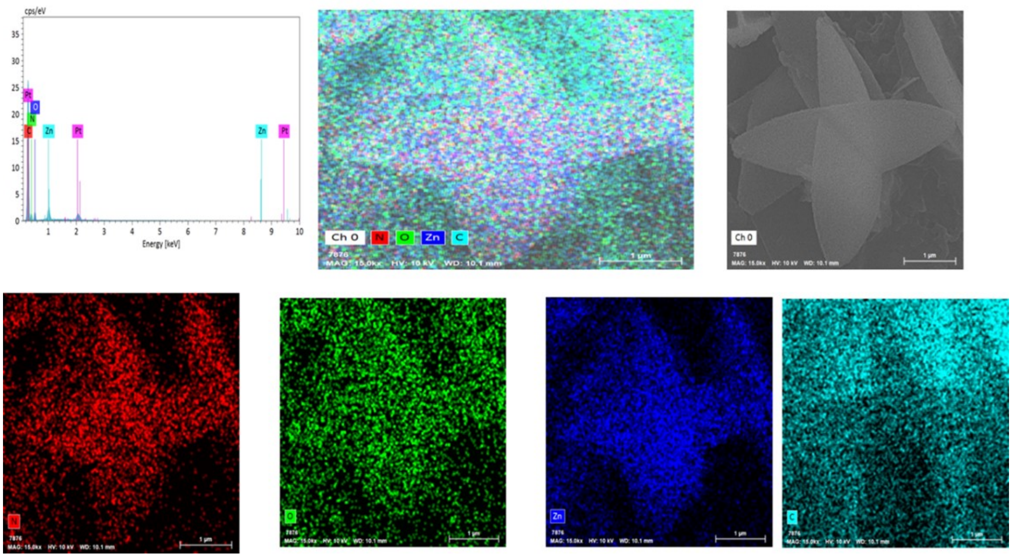


Figure S2. EDS images of ZIF-L-RPB nanosheets.

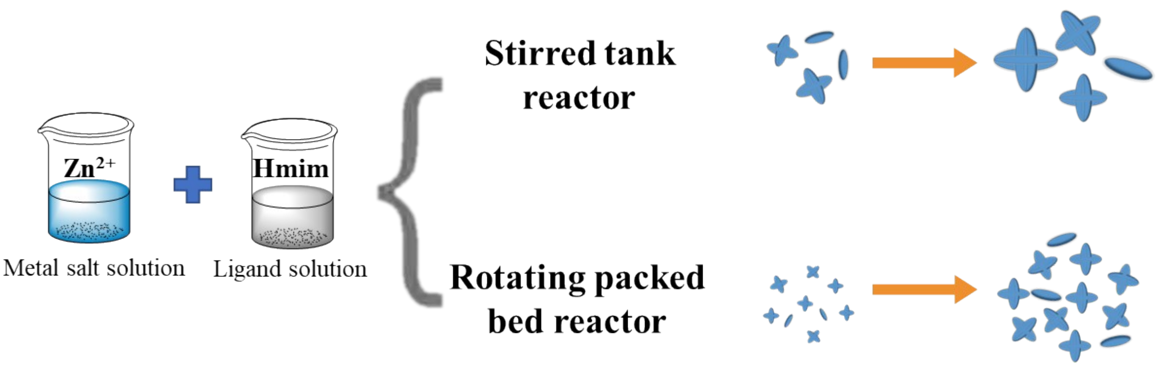


Figure S3. Illustration of the growth process of ZIF-L-STR and ZIF-L-RPB.

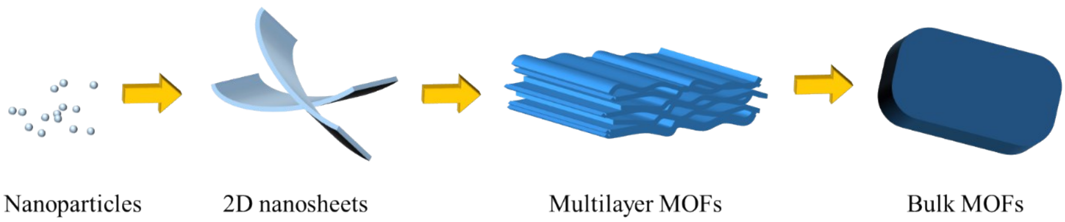


Figure S4. Growth process of 2D MOFs crystals.

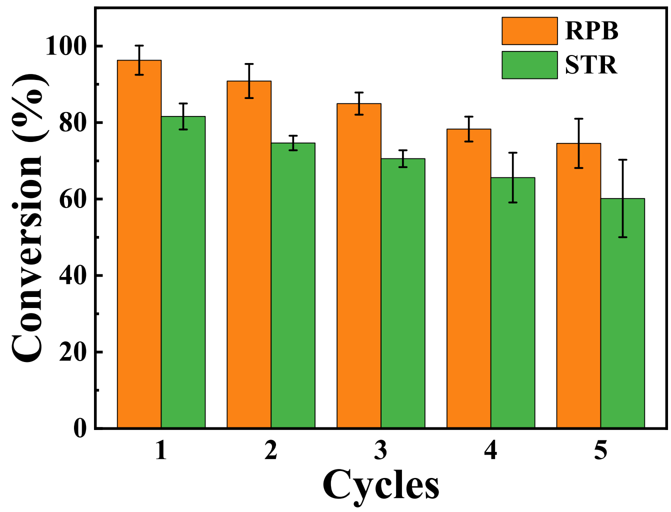


Figure S5. The reusability capacities of ZIF-L-STR and ZIF-L-RPB for the Knoevenagel condensation reaction. (the catalyst amount of 3.5 mol%, molar ratio of benzaldehyde to malononitrile of 1:1.9, 25 °C, 2 h)

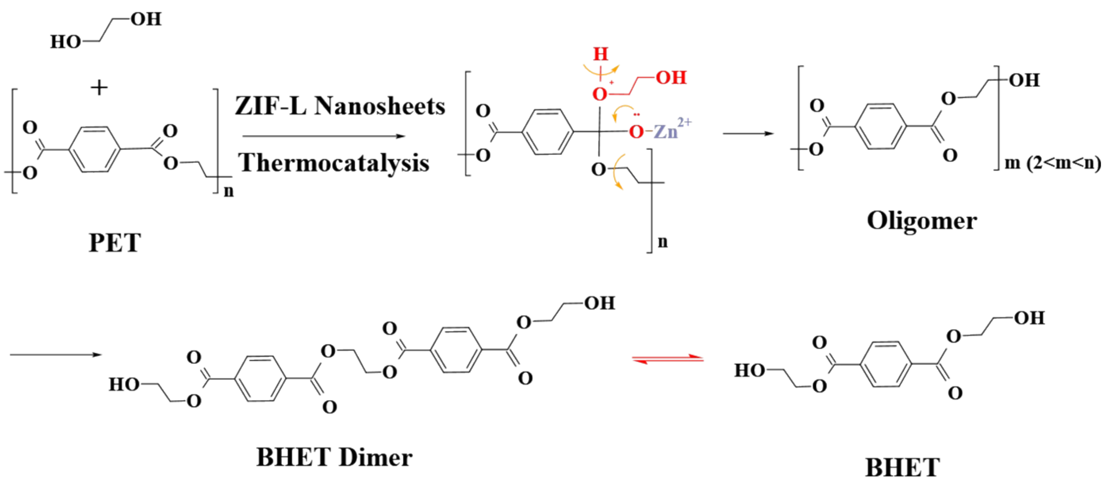


Figure S6. The schematic diagram of PET glycolysis over ZIF-L catalysts.

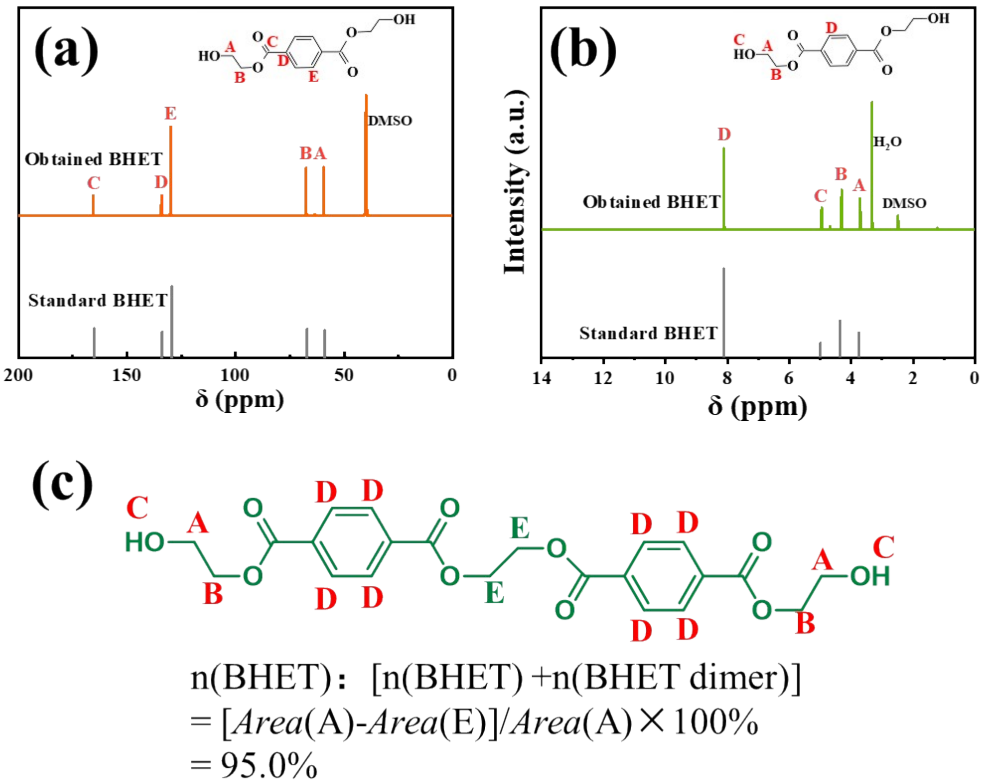


Figure S7. (a) ^{1}H NMR and (b) ^{13}C NMR spectra of the obtained BHET. (c) The ratio of BHET to BHET dimer in the product calculated by ^{1}H NMR spectrum.

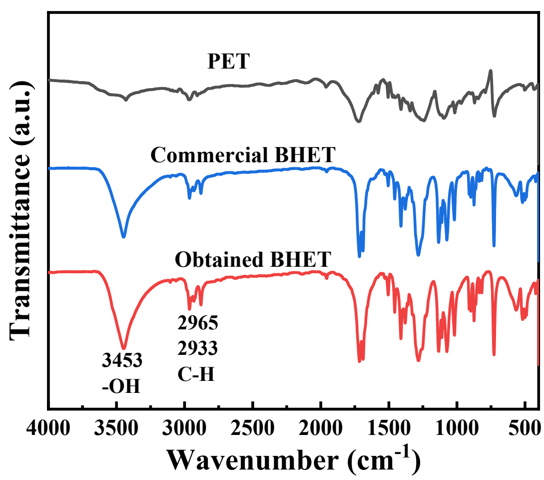


Figure S8. FTIR spectra of PET, commercial BHET, and BHET product obtained from the glycolysis reaction.

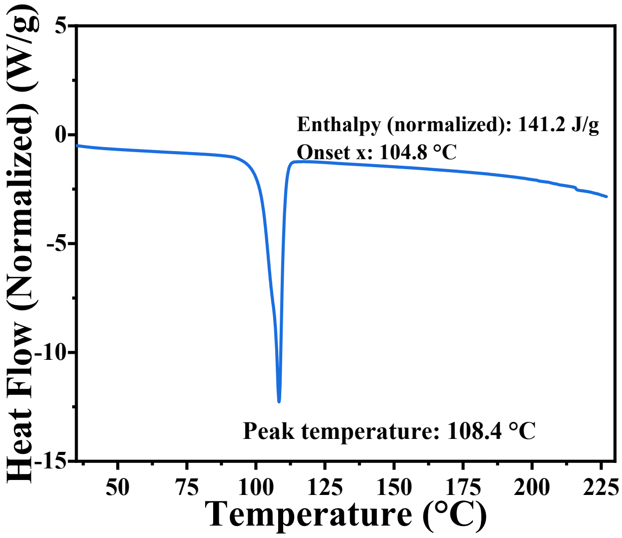


Figure S9. DSC curve of BHET produced from the glycolysis reaction.

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