

## Electronic Supplementary Information (ESI)

### Liquid-Phase Epitaxial Growth of Multiple MOF Thin Films with Highly Lattice Mismatch

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## **Materials and instrumentations**

All of the chemicals were used after purchasing without further purification. Out-of-plane XRD measurements was carried out on Bruker D8 advance equipped with a Si-strip detector (PSD Lynxeye (C)) using Cu  $K_{\alpha 1}$  radiation. In-plane XRD measurements were carried out using Bruker D8 Discover. IR spectra were recorded by using a using a FTIR spectrometer (Bruker VERTEX 80v). Scanning electron microscope (SEM) images for the morphology of thin films were recorded using a Philips XL30 (FEI Co., Eindhoven, NL) field emission gun environmental scanning electron microscope (FEG-ESEM). A Q-Sense E4 QCM was used to study the vapor adsorption of SURMOFs.

## **Preparation of MUD SAMs based substrates**

Self-assembled monolayers (SAMs) were fabricated by immersing 150 nm Au/2 nm Ti/evaporated on Si wafers or commercially available Au substrates in ethanolic solutions of MUD (1mmol/L) (MUD=11-mercapto-1-undecanol). 1mM/L ethanolic solutions of MUD ethanolic solutions was used for SAMs preparation by 72 h immersion and then rinsed with ethanol and dried under nitrogen flux to obtain -OH group functionalized Au substrate.

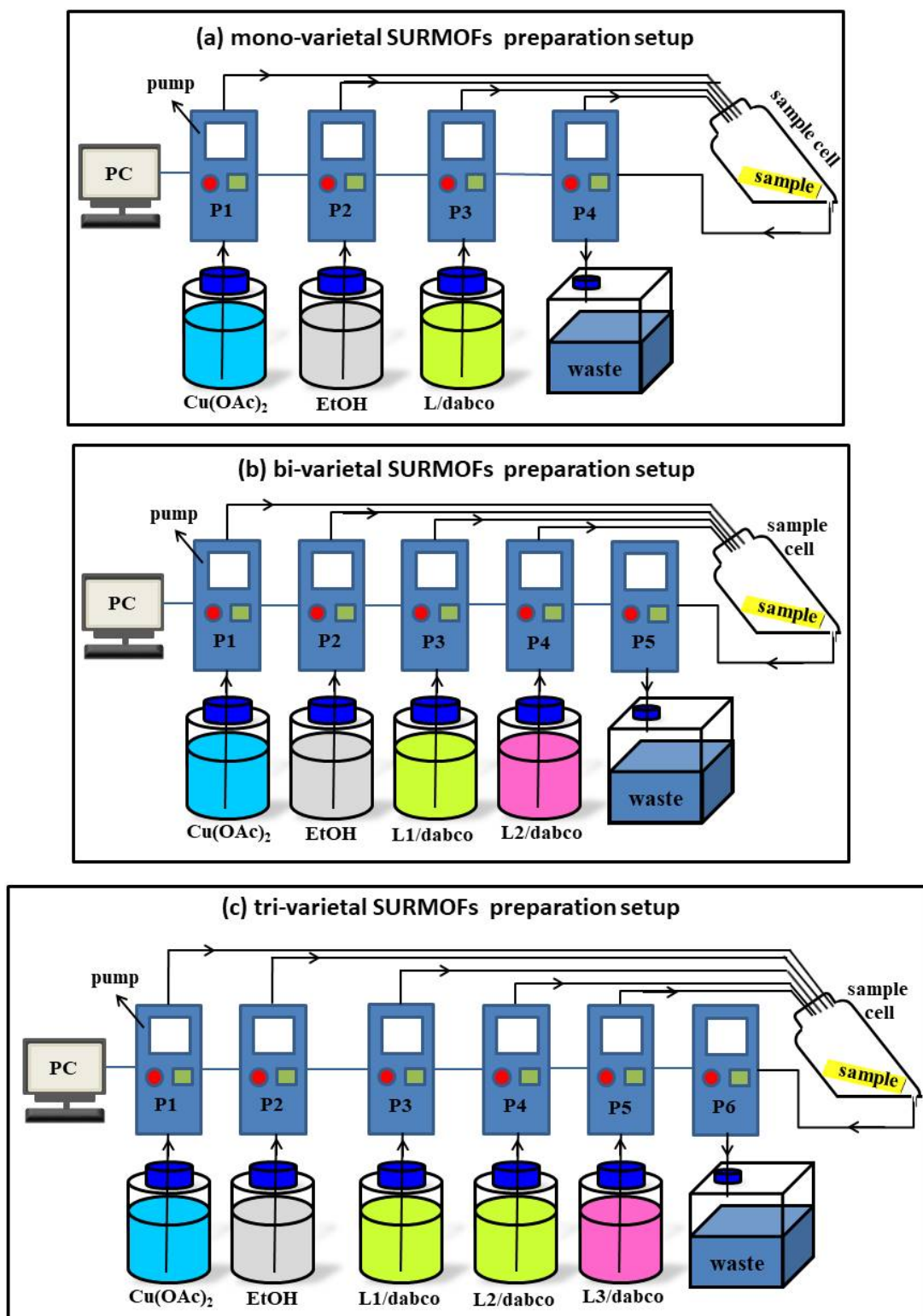
## **Fabrication of SURMOFs on substrate surfaces**

The SURMOFs used in the present work were grown using the layer by layer autoarm pump method. The -OH group functionalized Au substrates was put in the sample cell, and then subsequently filled with  $\text{Cu}(\text{OAc})_2$  and  $\text{H}_2\text{L}/\text{dabco}$  ( $\text{L} = \text{bdc}$ ,  $\text{bpc}$  and  $\text{tpdc}$ ) ethanolic solutions in the sample cell for 30 min at 50 °C. There was 2 min ethanol washing in each

step. Then 30 repeated cycles of preparation process resulted in the formation of homogeneous SURMOFs **SURMOF-a, -b, -c**.

For further prepare bi-varietal SURMOFs, the pre-prepared mono-varietal SURMOF was used for growth substrate, then this layer by layer pump process allows us to separately control the  $\text{Cu}(\text{OAc})_2$  and organic linker L2 deposition on the mono-varietal SURMOF to form bi-varietal SURMOF.

To prepare tri-varietal SURMOFs, the pre-prepared bi-varietal SURMOF was used for growth substrate, then this layer by layer pump process allows us to separately control the  $\text{Cu}(\text{OAc})_2$  and organic linker L3 deposition on the bi-varietal SURMOF to form tri-varietal SURMOF.



**Scheme S1.** The preparation setups for mono- varietal (a), bi-varietal (b) and tri-varietal) SURMOFs prepared by LEP layer by layer pump methods.

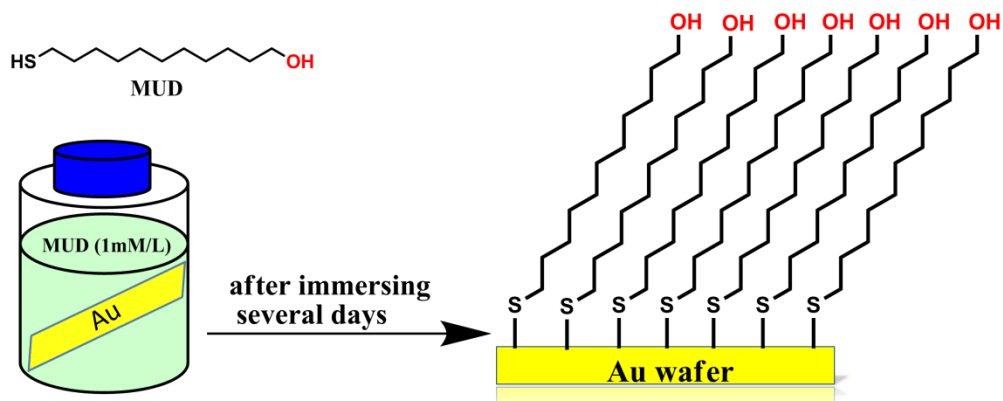


Figure S1. The preparation process of MUD SAMs.

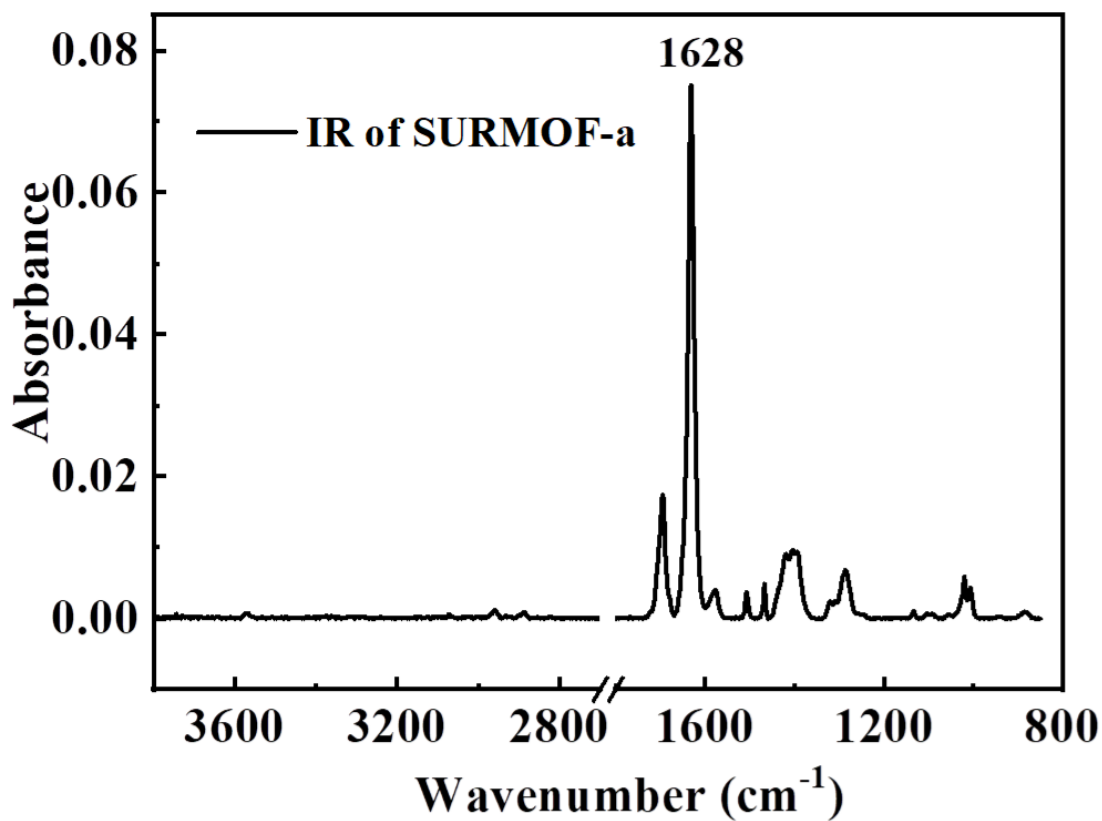


Figure S2. IR spectra of SURMOF-a.

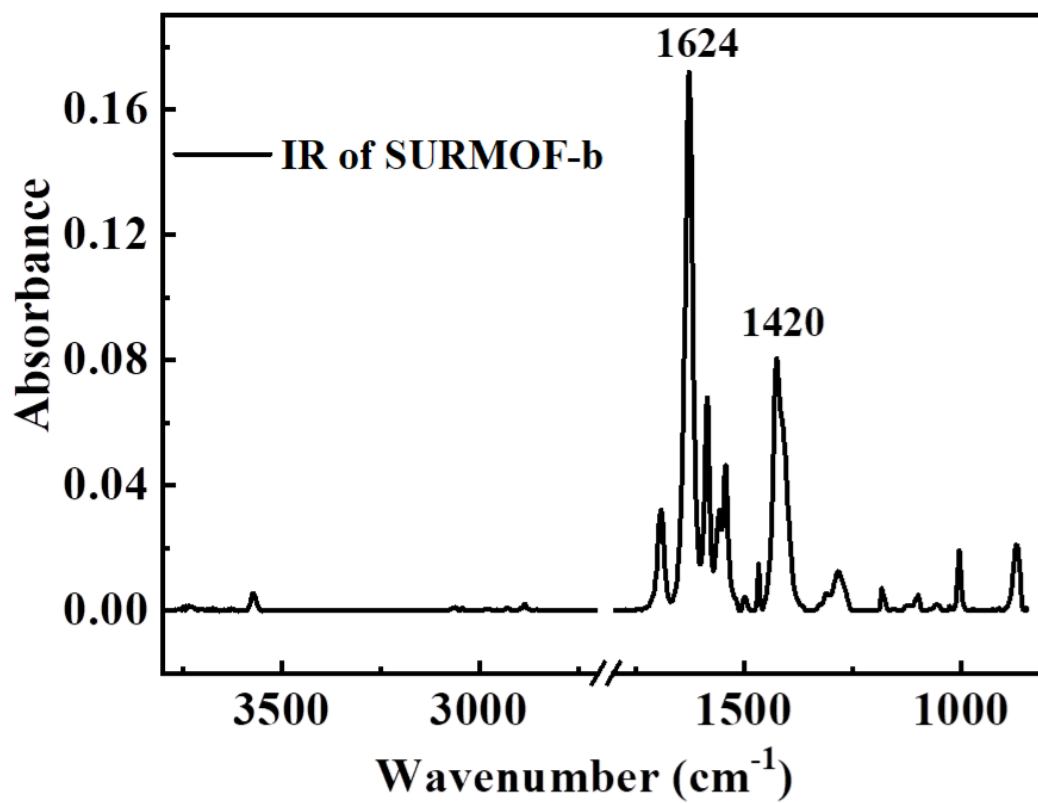


Figure S3. IR spectra of SURMOF-b.

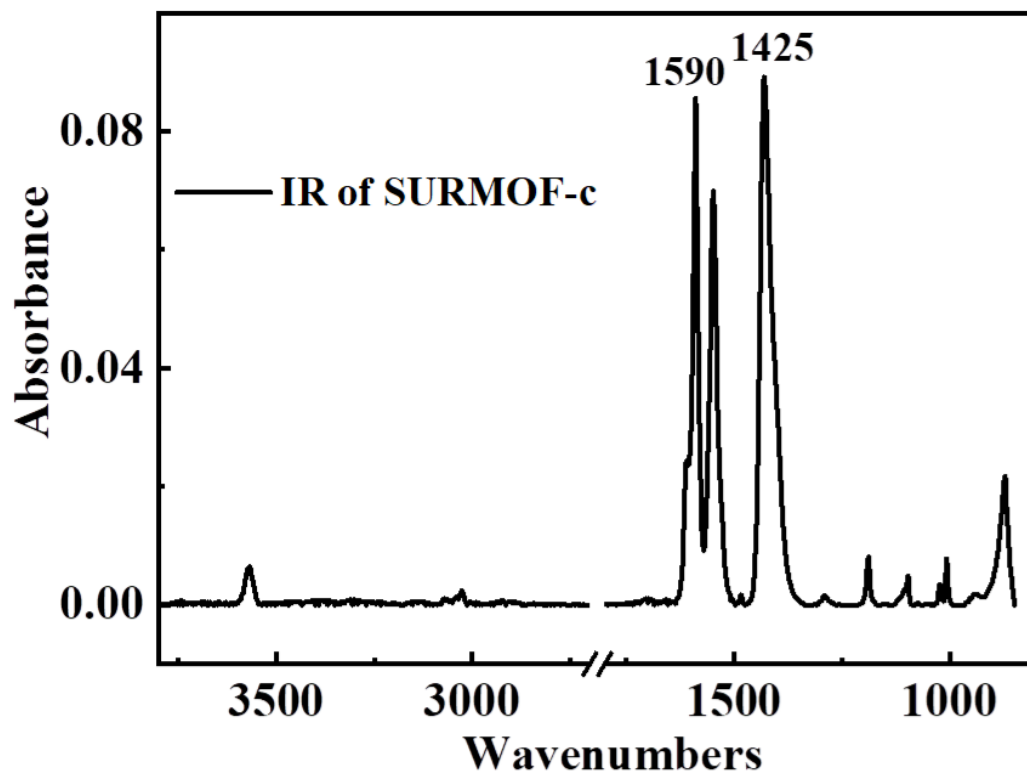
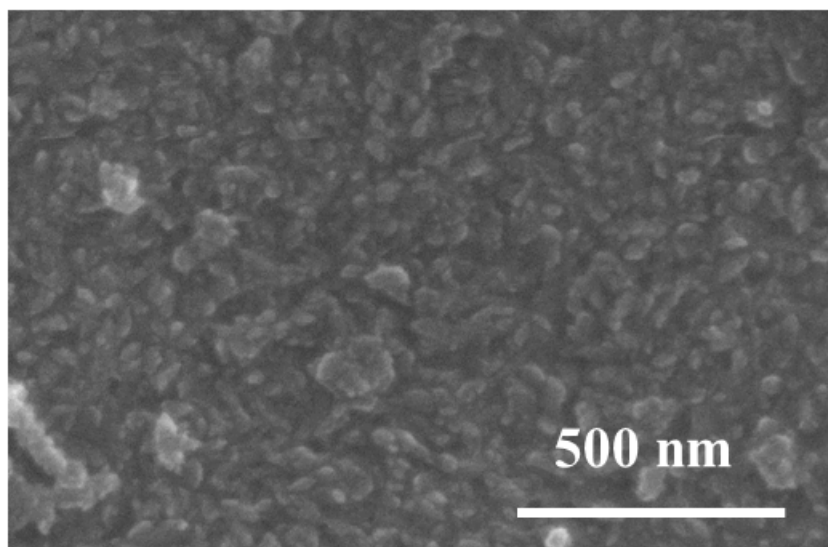
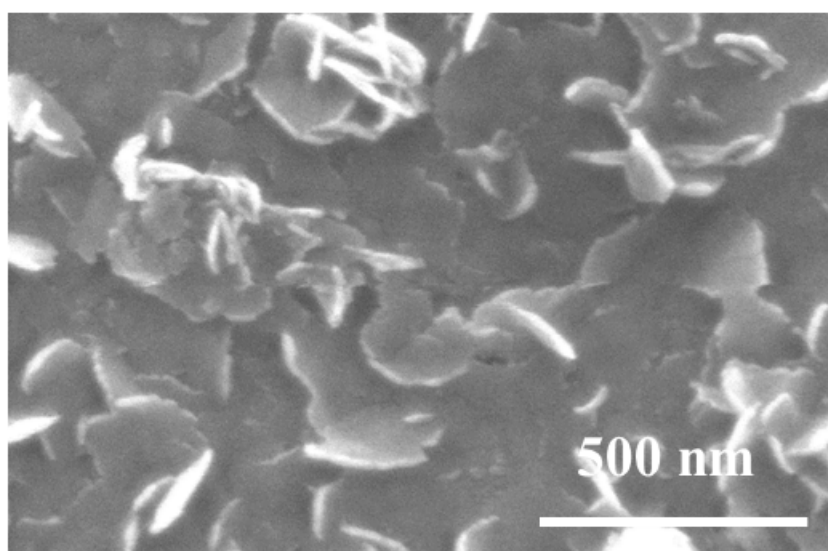


Figure S4. IR spectra of SURMOF-c.

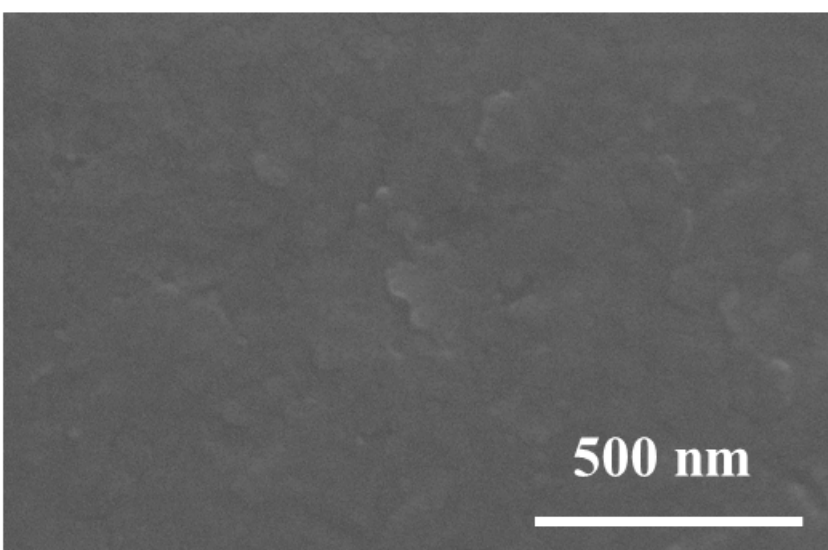
**(a)**



**(b)**

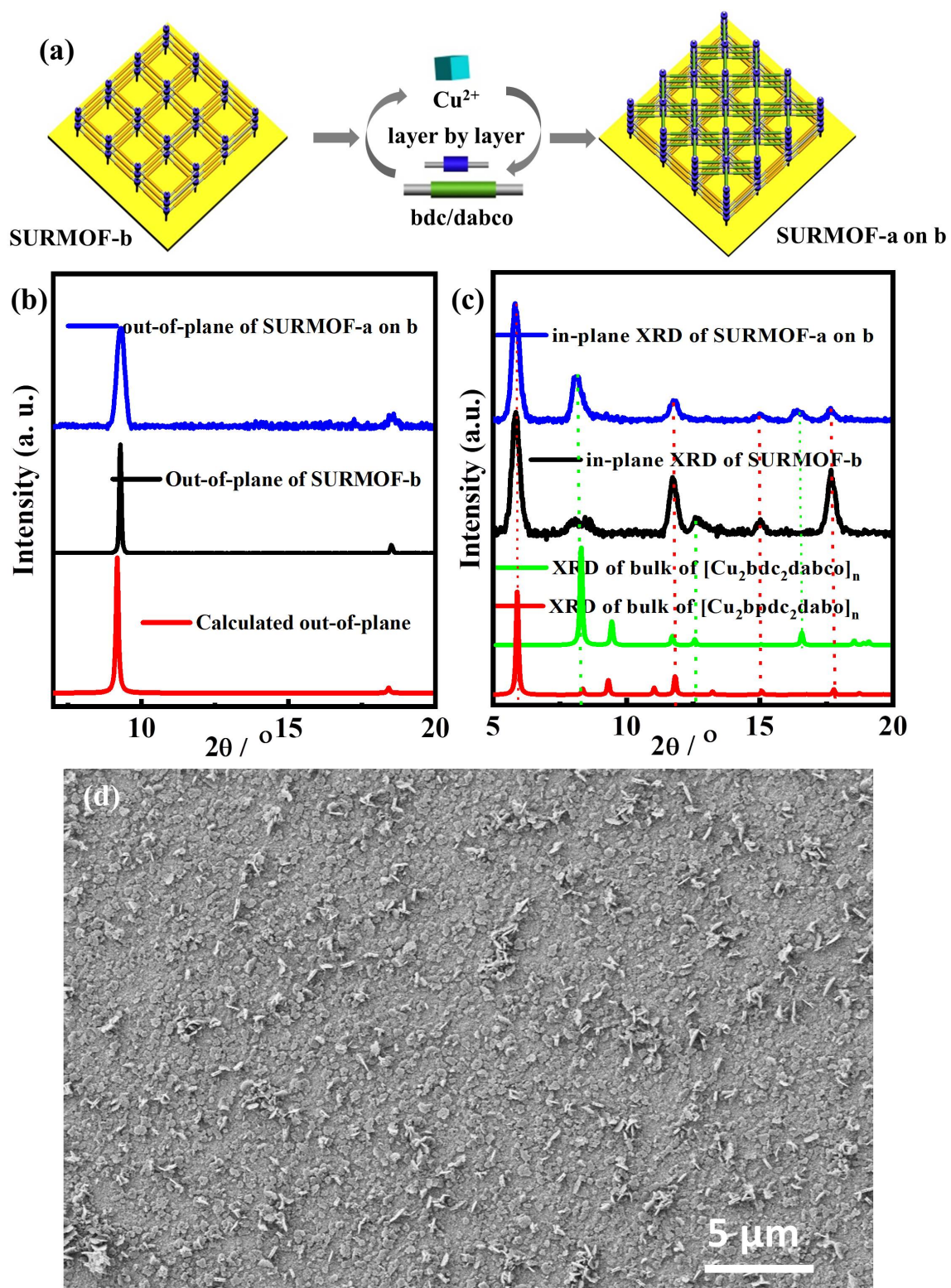


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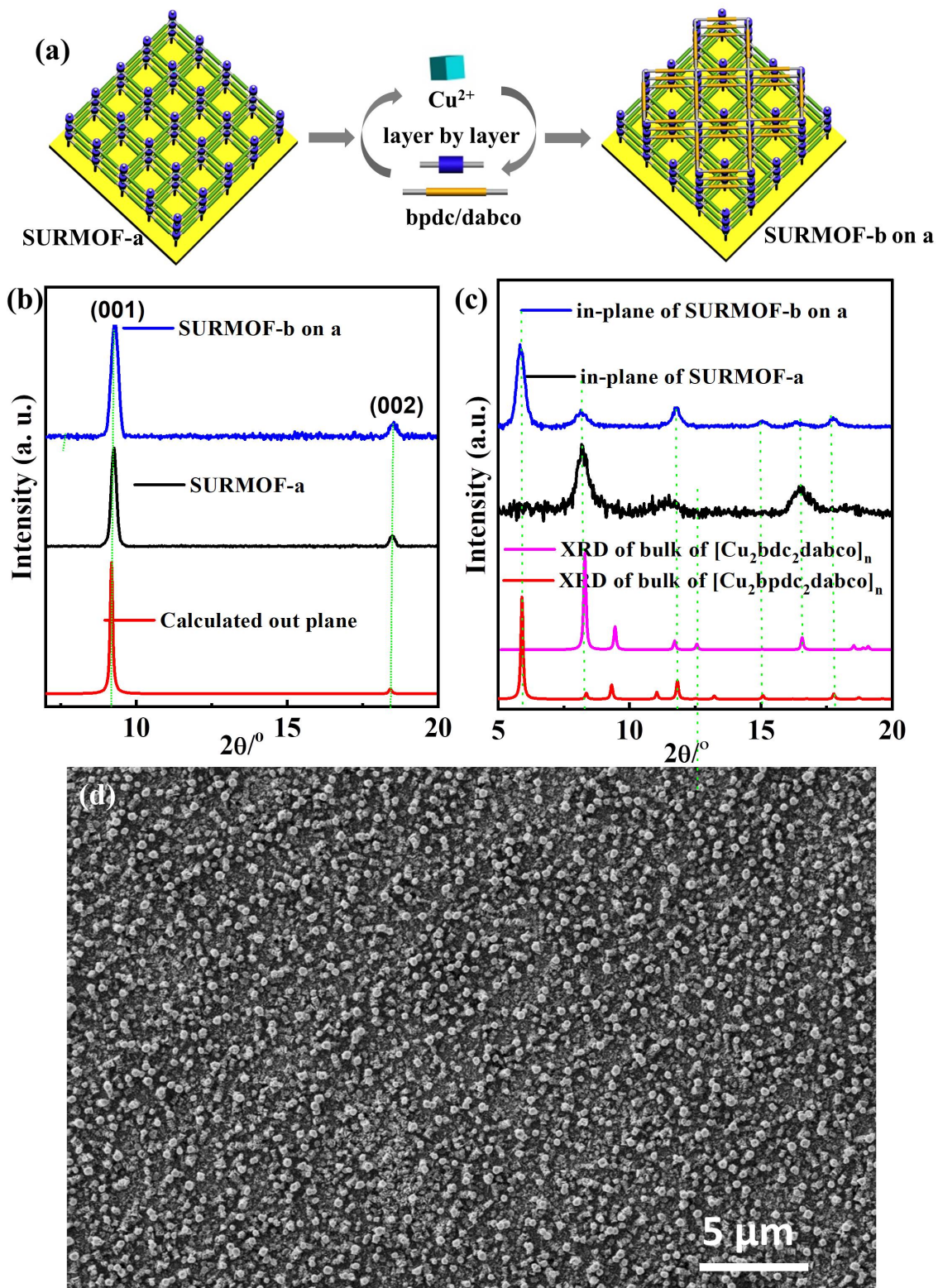


**Figure S5.** SEM images of SURMOF-a (a), SURMOF-b (b), SURMOF-c (c).

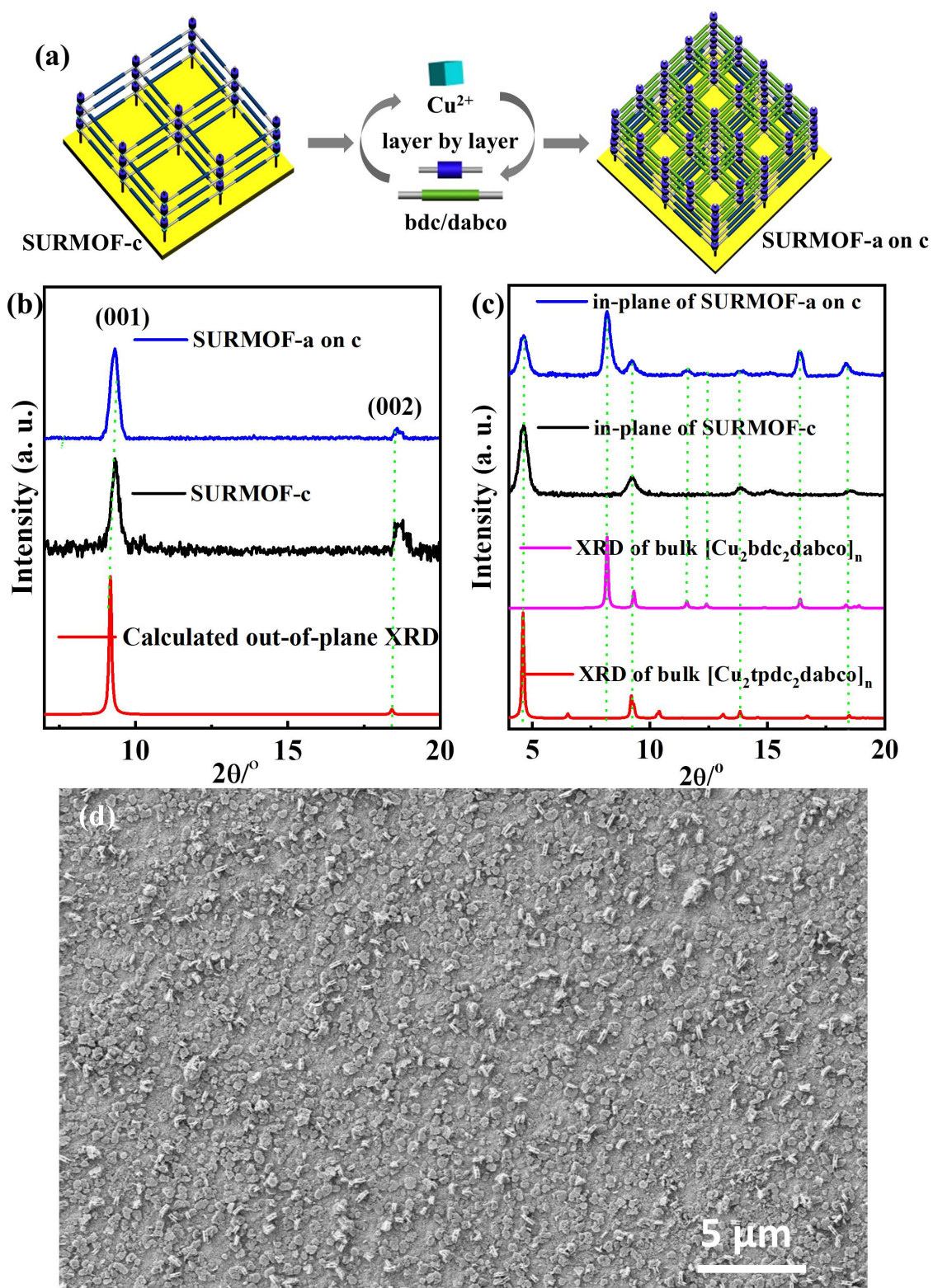




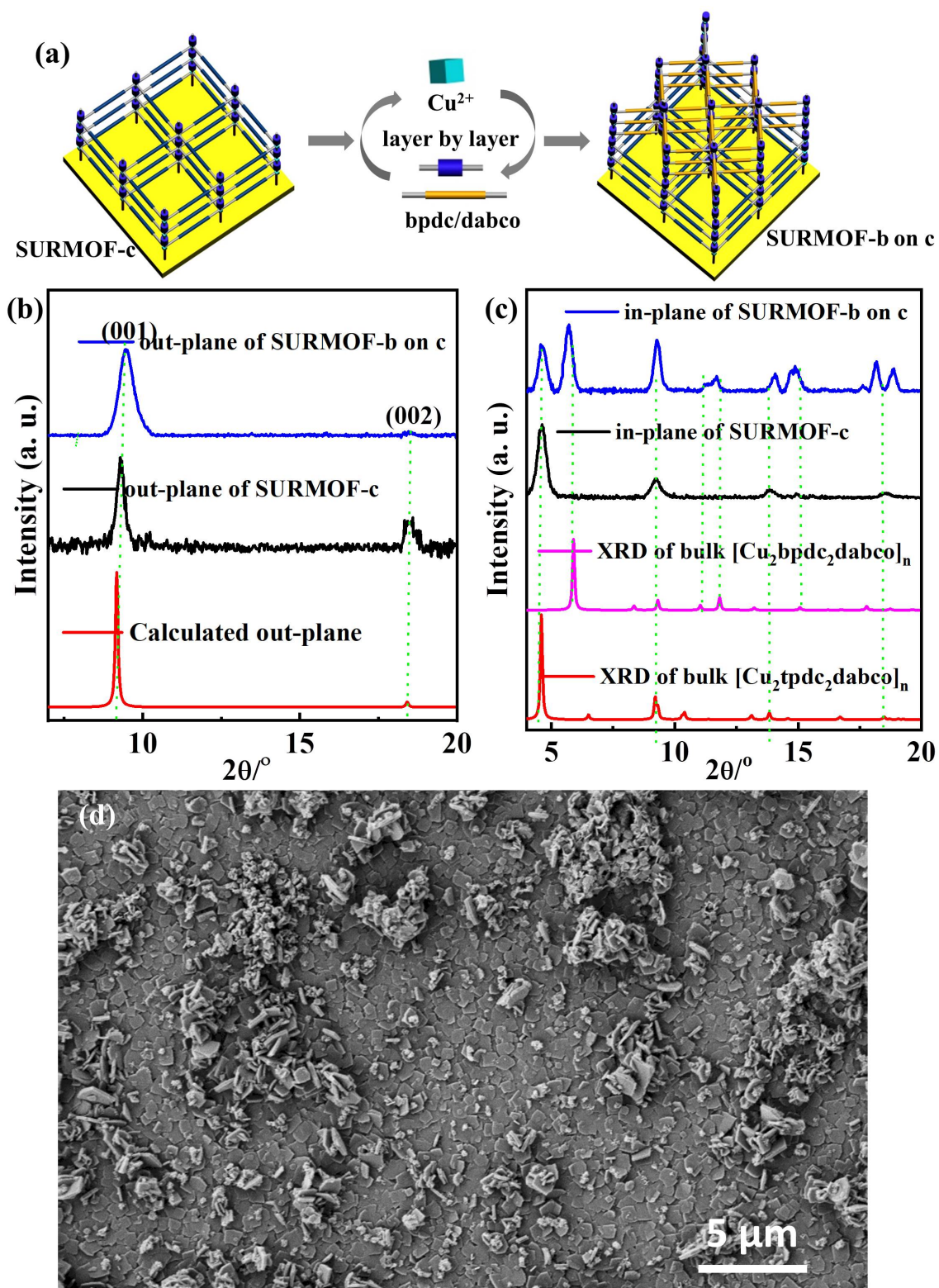
**Figure S6.** (a) Preparation process of SURMOF-a on b; out-of-plane (b) and in-plane XRD patterns (c) and as well as SEM image (d) of SURMOF-a on b.



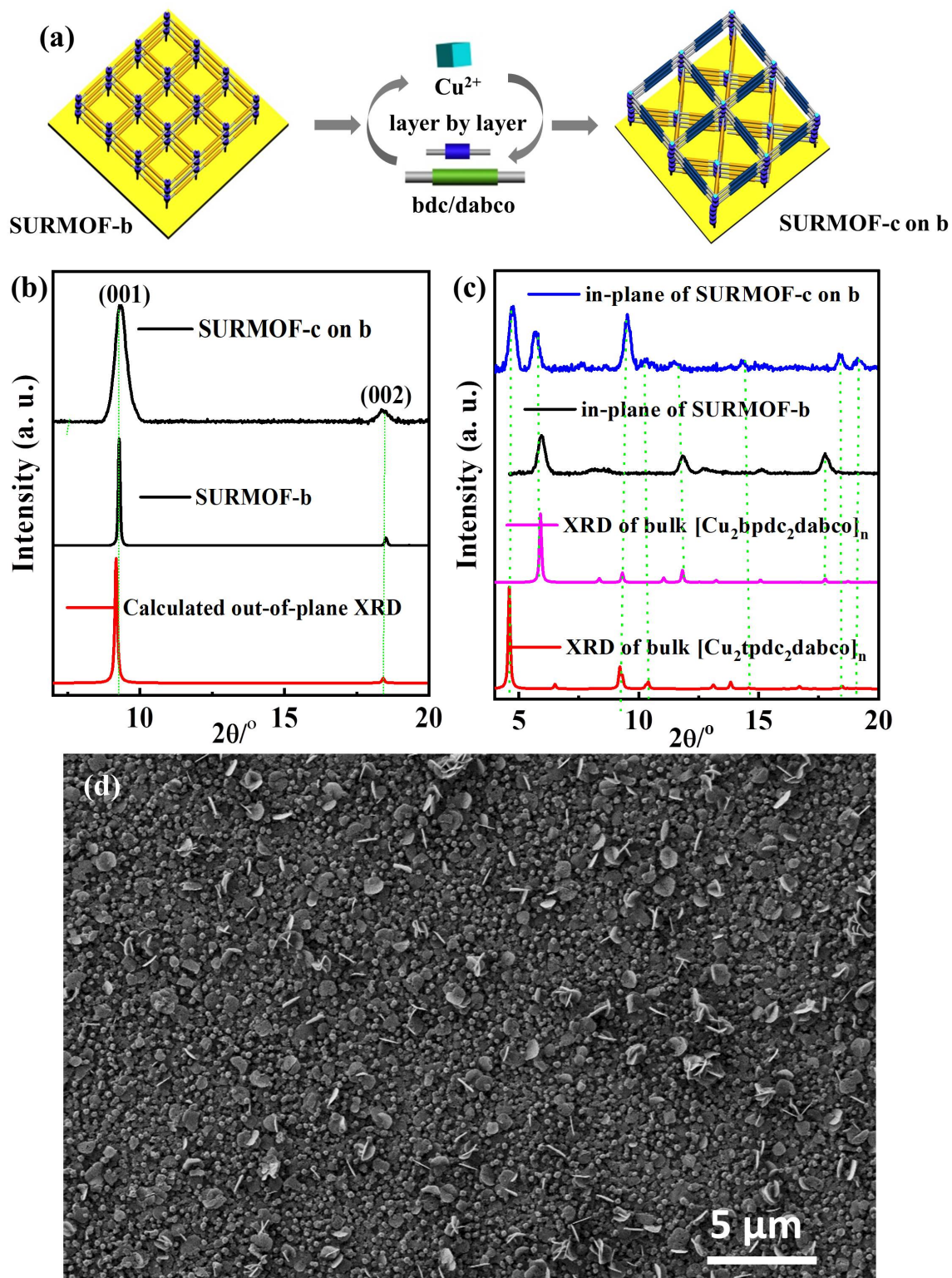
**Figure S7.** (a) Preparation process of SURMOF-b on a; out-of-plane (b) and in-plane XRD patterns (c) and as well as SEM image (d) of SURMOF-b on a.



**Figure S8.** (a) Preparation process of SURMOF-a on c; out-of-plane (b) and in-plane XRD patterns (c) and as well as SEM image (d) of SURMOF-a on c.



**Figure S9.** (a) Preparation process of SURMOF-b on c; out-of-plane (b) and in-plane XRD patterns (c) and as well as SEM image (d) of SURMOF-b on c.



**Figure S10.** (a) Preparation process of SURMOF-c on b; out-of-plane (b) and in-plane XRD patterns (c) and as well as SEM image (d) of SURMOF-c on b.

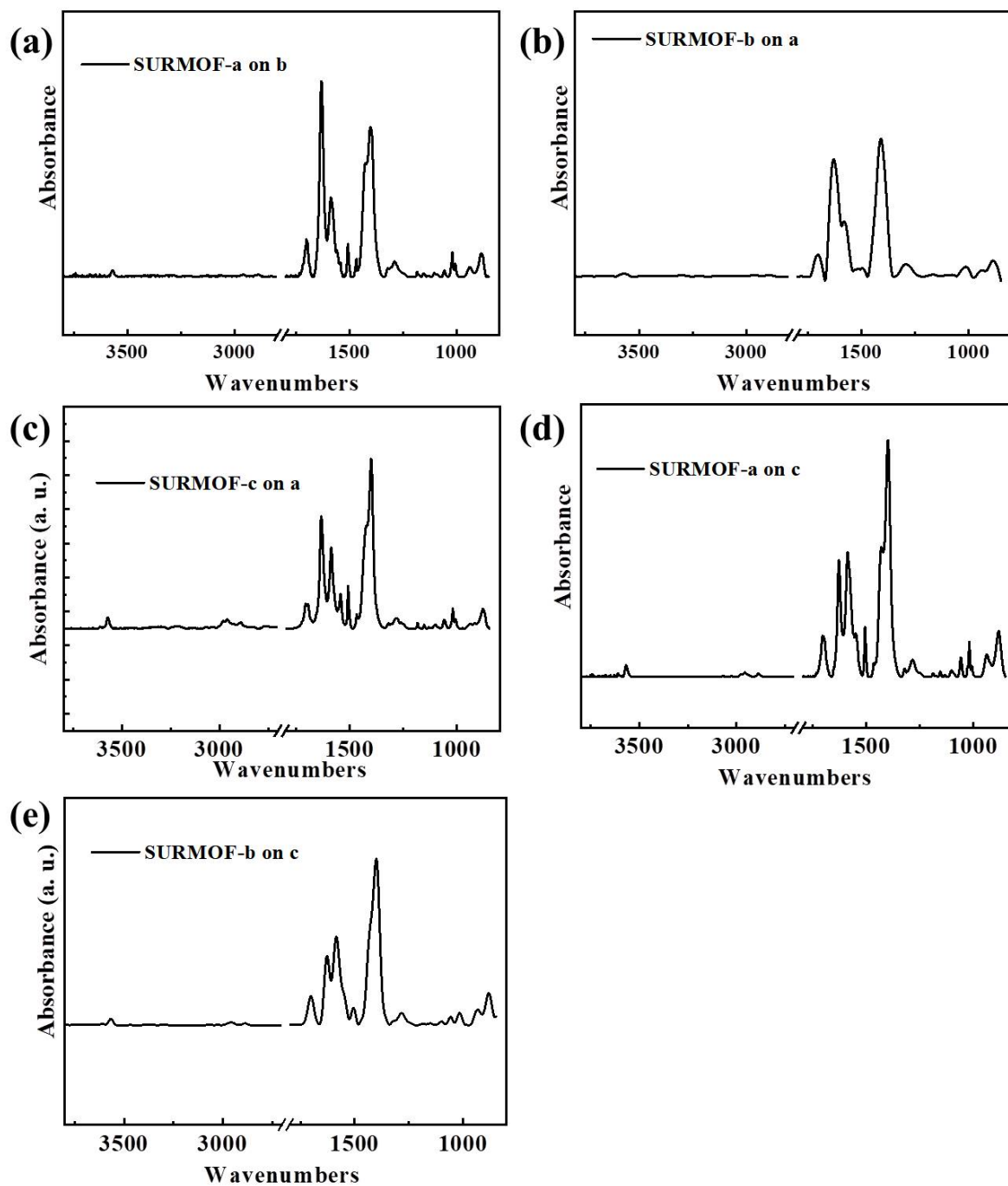
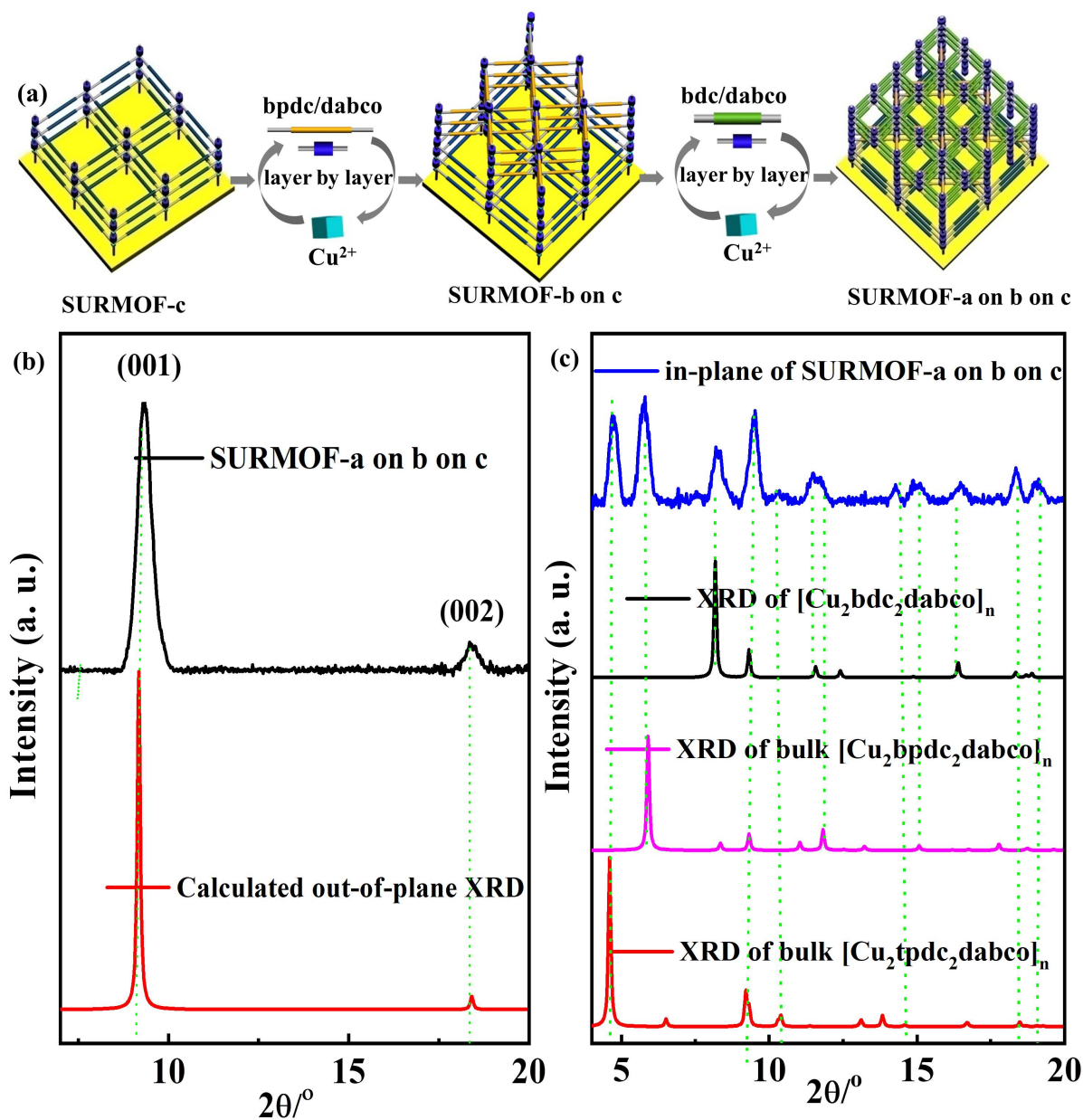
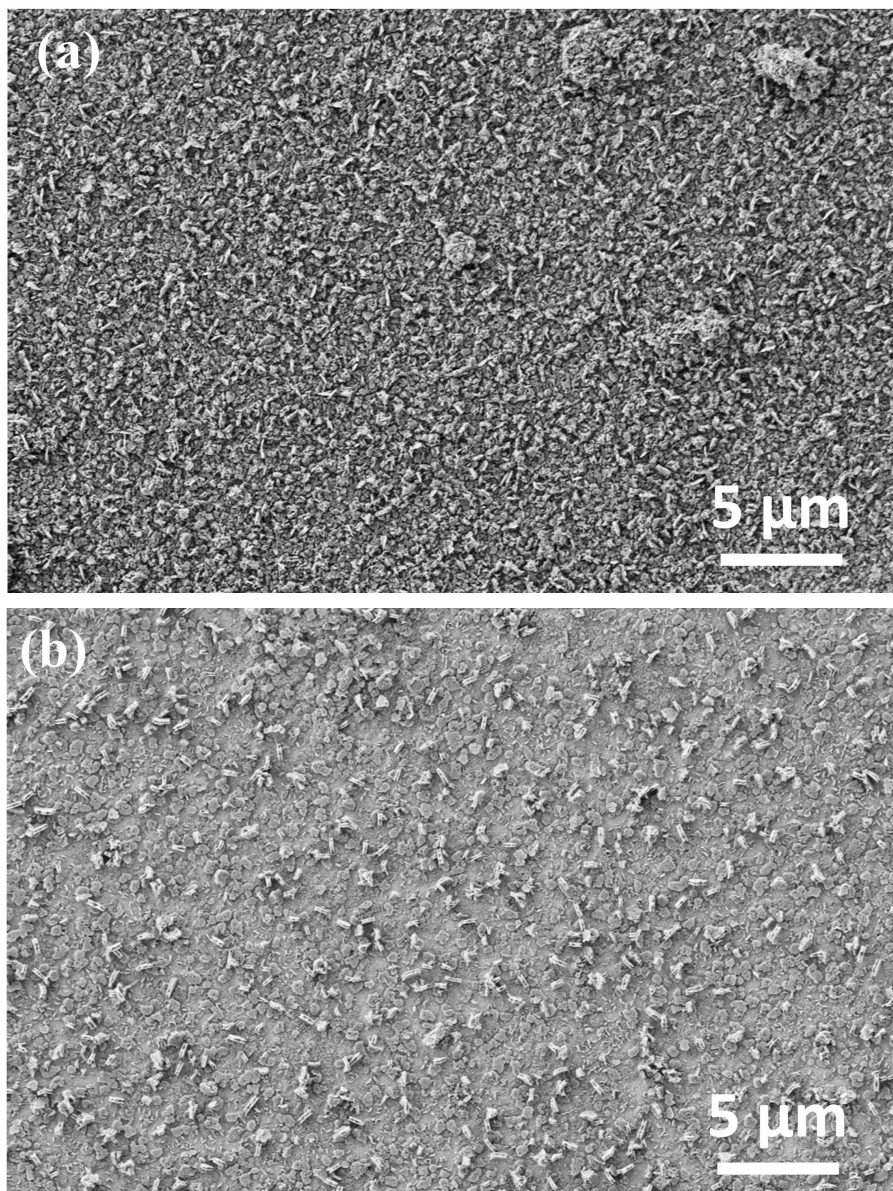


Figure S11. IR spectra of SURMOF-a on b, SURMOF-a on b, SURMOF-c on a, SURMOF-a on c, SURMOF-b on c, respectively.



**Figure S12.** (a) Preparation process of SURMOF-a on b on c; out-of-plane (b) and in-plane XRD patterns (c) of SURMOF-a on b on c.



**Figure S13.** SEM images of SURMOF-c on b on a (a) and SURMOF-a on b on c (b).



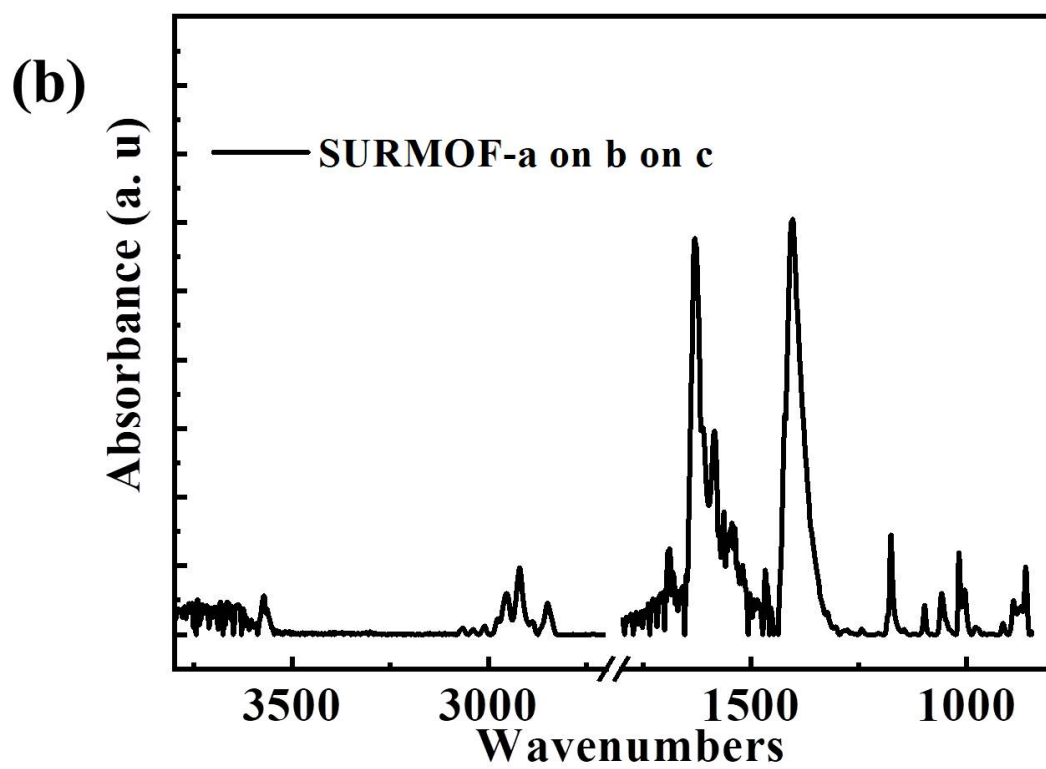
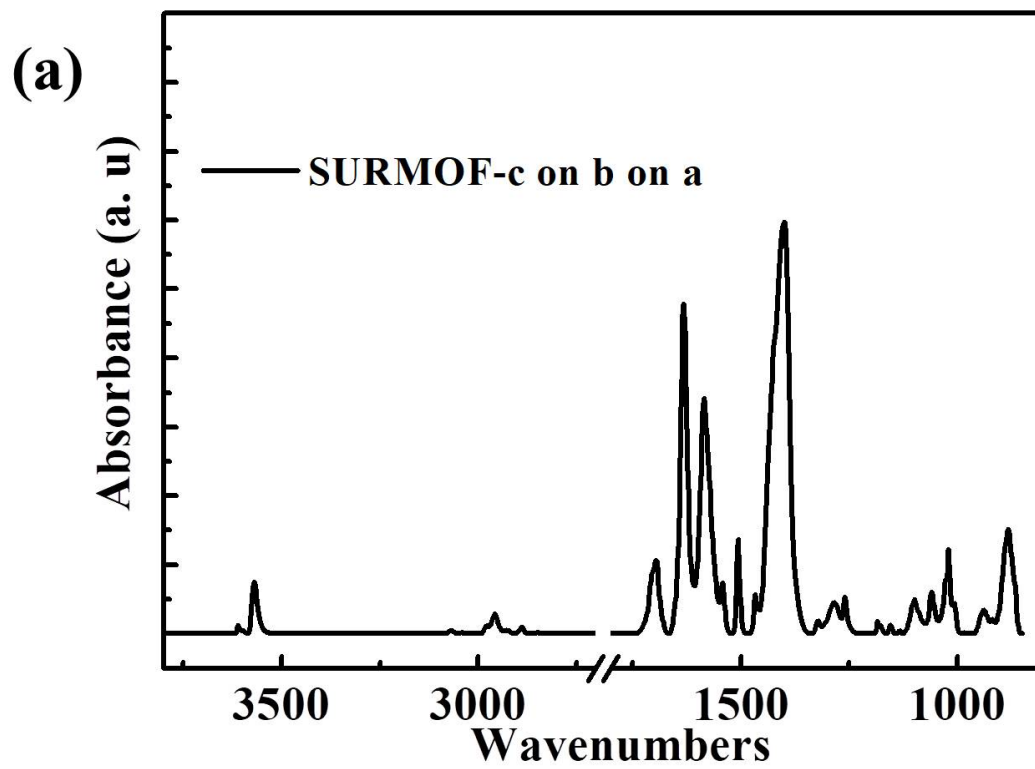
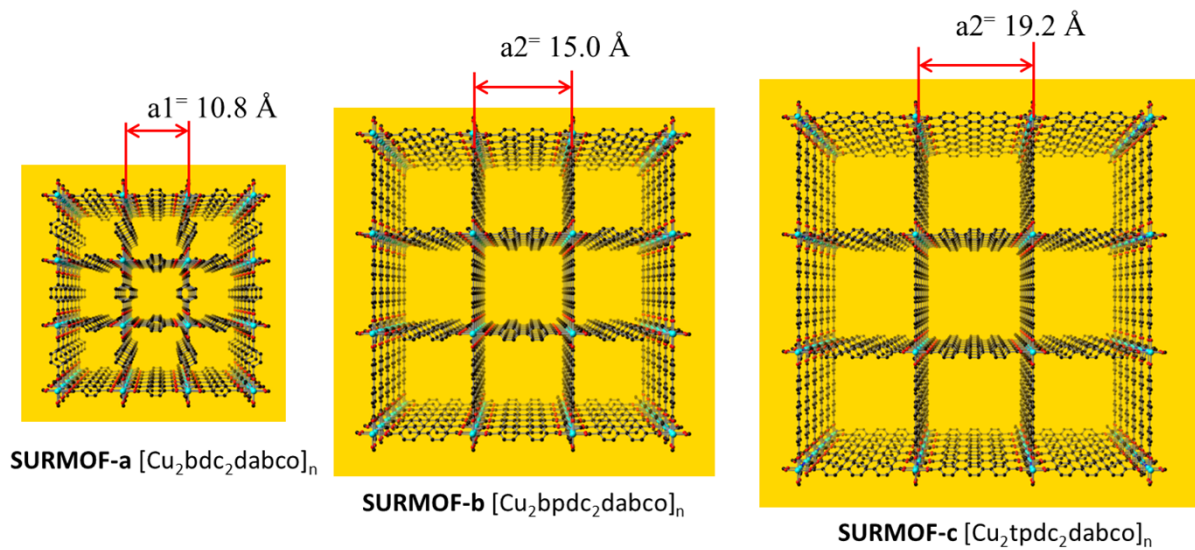


Figure S14. IR spectra of SURMOF-c on b on a (a) and SURMOF-a on b on c (b).



$$a_2 \approx \sqrt{2}a_1 \quad a_3 \approx \sqrt{2}a_2$$

**Figure S15.** The lattice constant mismatch analysis between **SURMOF-a** and **SURMOF-b**  $[(a_2 - a_1)/a_1]$  and between **SURMOF-a** and **SURMOF-c**  $[(a_3 - a_1)/a_1]$ .