

## **Supplementary Information**

# **Fabrication of $[\text{Cu}_2(\text{bdc})_2(\text{bpy})]_n$ thin films using coordination modulation-assisted layer-by-layer growth**

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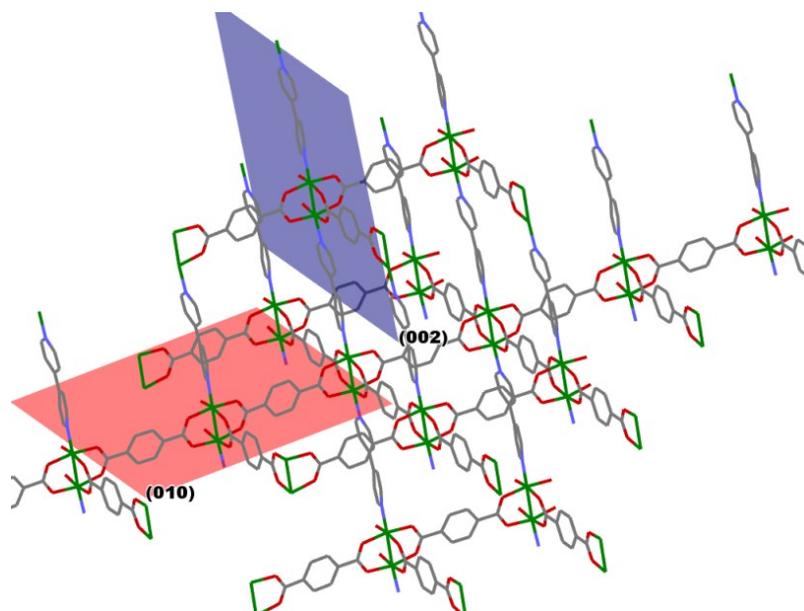


Figure S1. Structure of  $[\text{Cu}_2(\text{bdc})_2(\text{bpy})]_n$  showing (010) and (002) planes (based on the single-crystal structural assignment). Color code: green: copper; red: oxygen; blue: nitrogen; black: carbon. Hydrogen atoms were omitted for clarity.

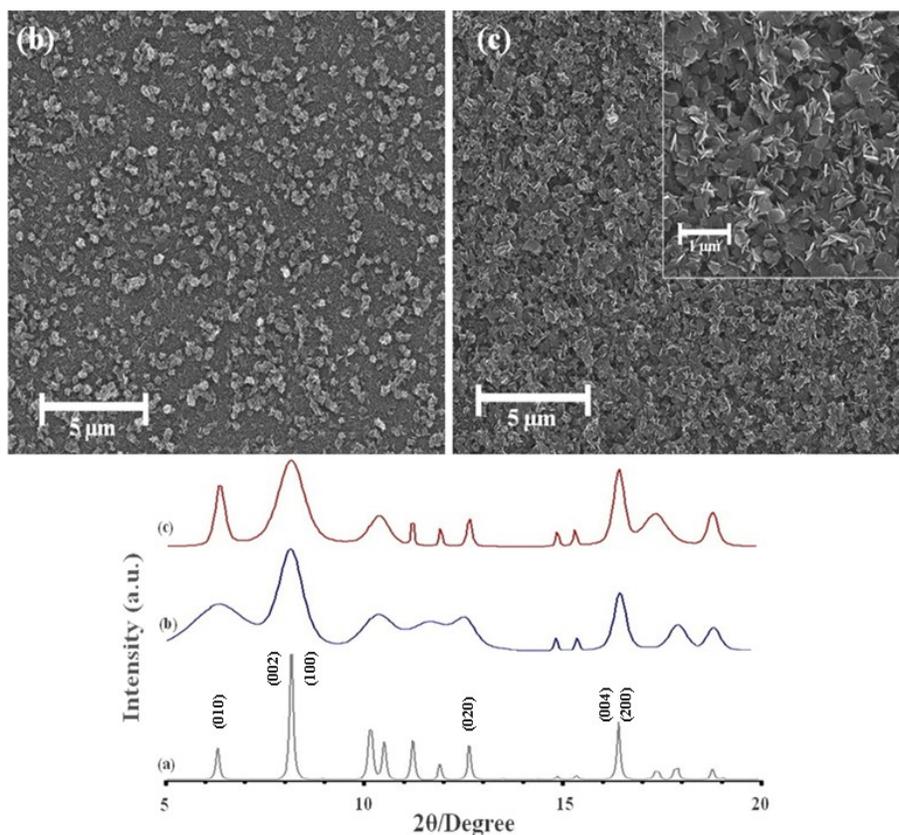


Figure S2. SEM images and XRD patterns of  $[\text{Cu}_2(\text{bdc})_2(\text{bpy})]_n$  thin films obtained through a two-step (b) and three-step deposition procedure (c). Simulated powder pattern profile is also depicted for the sake of comparison (a).

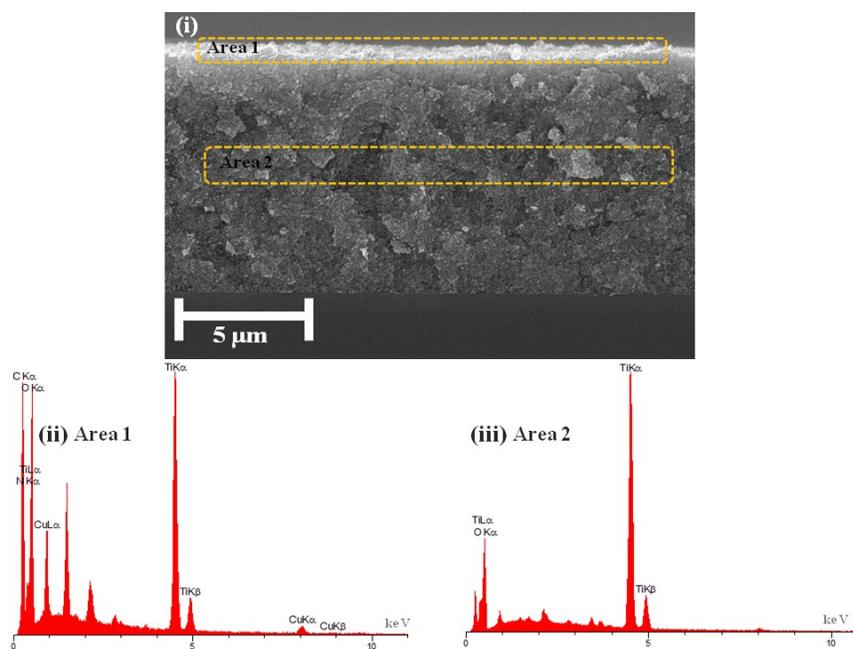


Figure S3. Cross-sectional SEM view of the film prepared through a three-step deposition procedure (i), and EDX data showing the semi-quantitative composition of the film at various area (top and bottom) of the cress section.

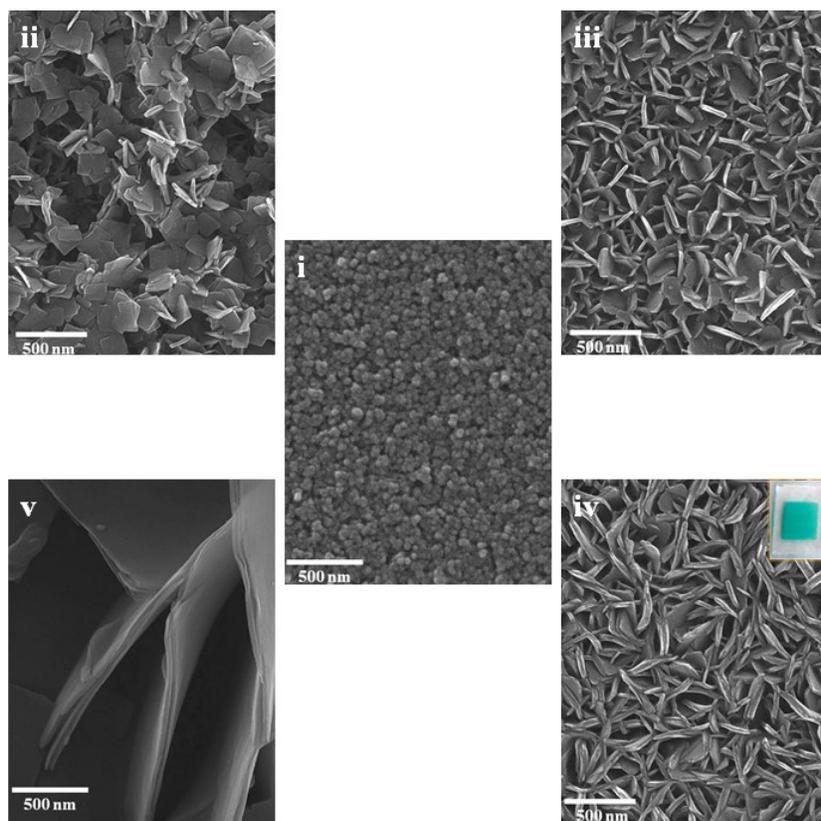


Figure S4. Top view SEM images of: (i) mesoporous  $\text{TiO}_2$  support and  $[\text{Cu}_2(\text{bdc})_2(\text{bpy})]_n$  thin films prepared (ii) without modulator, (iii) with 35  $\mu\text{l}$ , (iv) 70  $\mu\text{l}$  and (v) 140  $\mu\text{l}$  of modulator (added to metal solution). Inset of (iv) shows corresponding digital image.

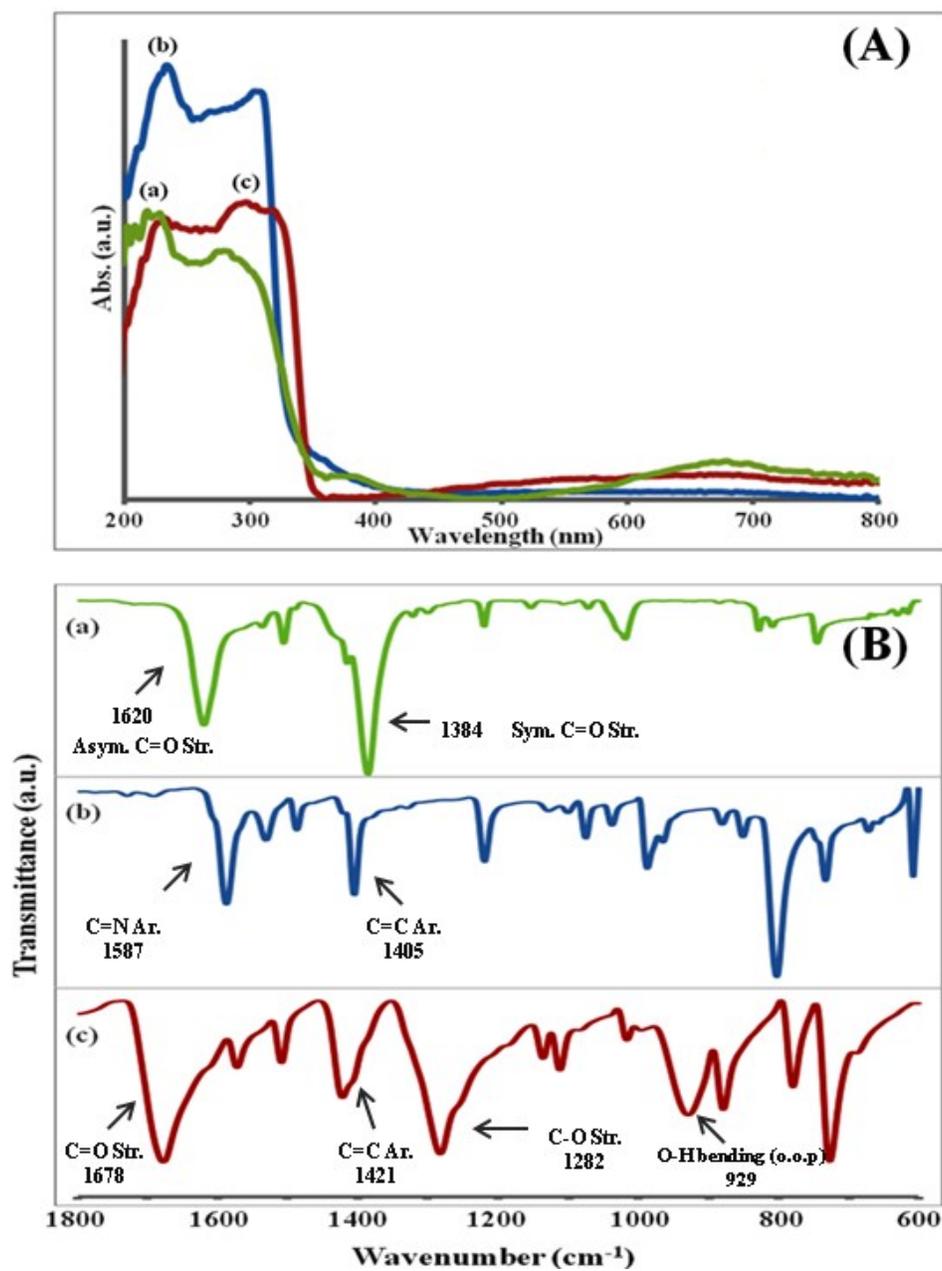


Figure S5. DRS and ATR-FTIR spectra of  $[\text{Cu}_2(\text{bdc})_2(\text{bpy})]_n$  thin film (obtained after the addition of 70  $\mu\text{l}$  HOAC) (a), bpy (b) and bdc (c) ligands.

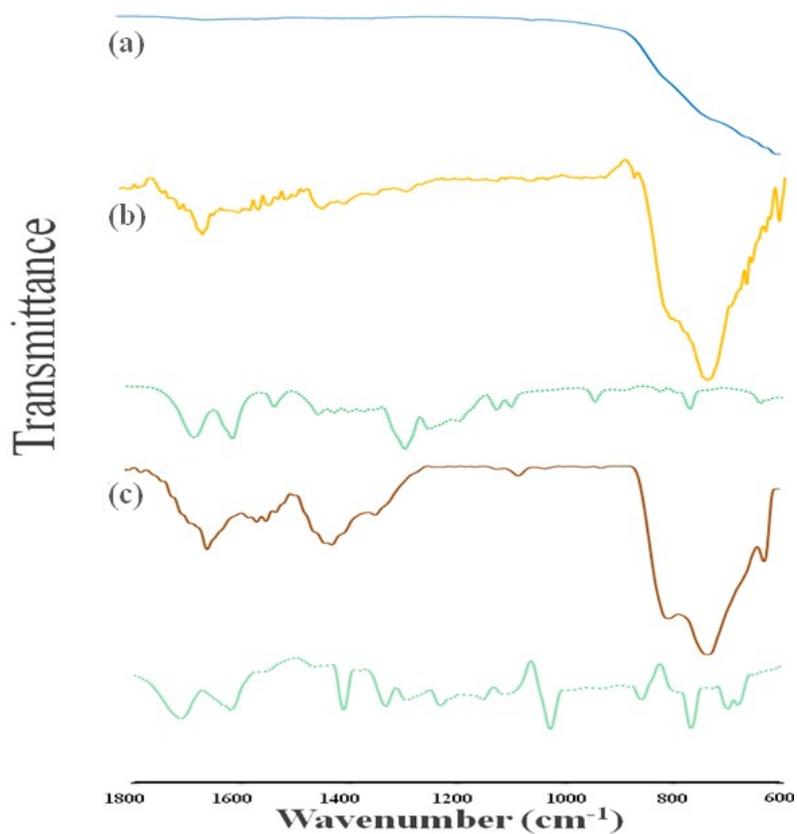


Figure S6. ATR-FTIR spectra of TiO<sub>2</sub> film (a), DHBA- (b) and INA-treated TiO<sub>2</sub> film (c). (Green dotted lines represent free DHBA and INA spectra).

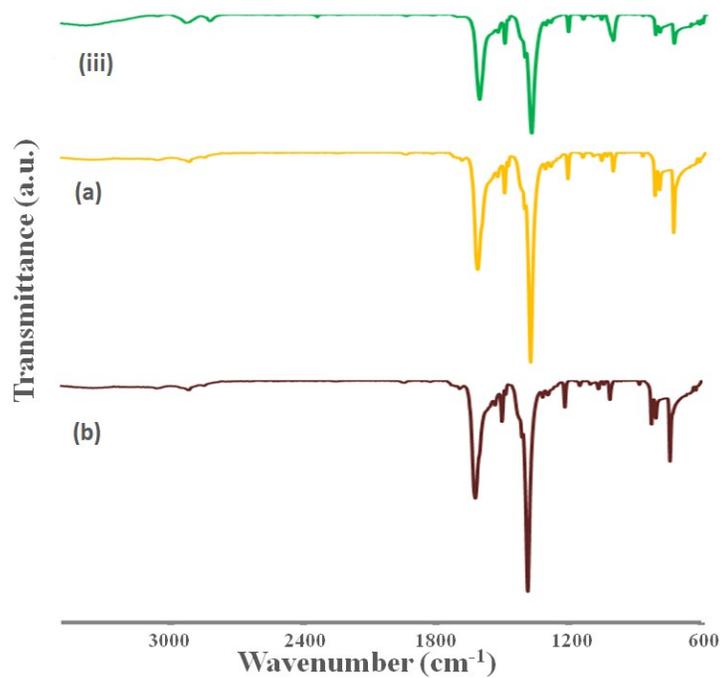


Figure S7. ATR-FTIR spectra of [Cu<sub>2</sub>(bdc)<sub>2</sub>(bpy)]<sub>n</sub> thin films on bare (iii), DHBA- (a) and INA-treated TiO<sub>2</sub> surface (b).

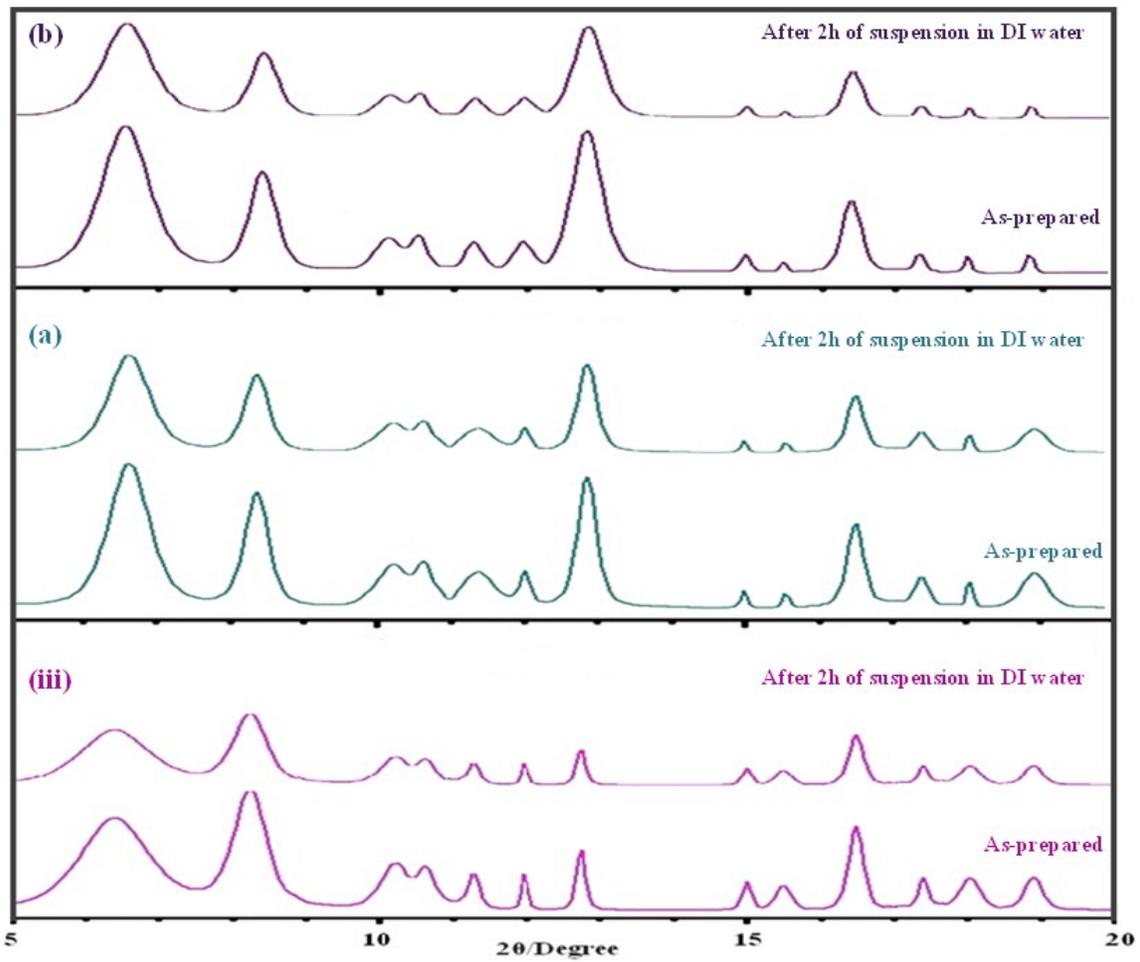


Figure S8. In-plane XRD patterns of  $[\text{Cu}_2(\text{bdc})_2(\text{bpy})]_n$  thin films grown on bare (iii), DHBA- (a) and INA-treated  $\text{TiO}_2$  surfaces (b) after 120 min. of suspension in deionized water.